



3 4456 0348305 9

ORNL-6596

oml

**OAK RIDGE
NATIONAL
LABORATORY**

MARTIN MARIETTA

**Chemical Technology Division
Progress Report
for the Period July 1, 1988,
to September 30, 1989**

OAK RIDGE NATIONAL LABORATORY

CENTRAL RESEARCH LIBRARY

CIRCULATION SECTION

4500N ROOM 175

LIBRARY LOAN COPY

DO NOT TRANSFER TO ANOTHER PERSON

If you wish someone else to see this report, send in name with report and the library will arrange a loan.

OPERATED BY
MARTIN MARIETTA ENERGY SYSTEMS, INC.
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

This report has been reproduced directly from the best available copy.

Available to DOE and DOE contractors from the Office of Scientific and Technical Information, P.O. Box 62, Oak Ridge, TN 37831, prices available from (615) 576-8401, PTS 826-8401.

Available to the public from the National Technical Information Service, U.S. Department of Commerce, 5285 Port Royal Rd., Springfield, VA 22161.

NTIS price codes—Printed Copy, $\frac{A}{12}$ Microfiche A01

This report was prepared as an outcome of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply an endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

**Chemical Technology Division
Progress Report
for the Period July 1, 1988, to September 30, 1989**

R. K. Genung, Division Director

J. R. Hightower, Associate Director, Radiochemical Processing Programs

J. T. Bell, Section Head, Chemical Development

E. D. Collins, Section Head, Process Development

B. D. Patton, Section Head, Isotopes Section

A. G. Croff, Associate Director, Waste and Environmental Programs

R. L. Jolley, Manager, Waste Management Technology Center

C. H. Brown, Section Head, Engineering Development

J. M. Begovich, Section Head, Engineering Coordination and Analysis

C. D. Scott, Senior Corporate Fellow, Energy Research Programs

E. K. Johnson, Section Head, Resource Systems Management Section

V. C. A. Vaughen, Director, Chemical Technology Division

Office of Operational Safety and Readiness

J. S. Watson, Manager, Special Separations Program

J. W. Snider, Senior Adviser, AVLIS Program Support

Date of Issue — March 1990

**OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37831
operated by
MARTIN MARIETTA ENERGY SYSTEMS, INC.
for the
Department of Energy
under contract DE-AC05-84OR21400**



3 4456 0348305 9

Reports previously issued in this series:

ORNL-2392 Period Ending August 31, 1957
ORNL-2576 Period Ending August 31, 1958
ORNL-2788 Period Ending August 31, 1959
ORNL-2993 Period Ending August 31, 1960
ORNL-3153 Period Ending May 31, 1961
ORNL-3314 Period Ending May 31, 1962
ORNL-3452 Period Ending May 31, 1963
ORNL-3627 Period Ending May 31, 1964
ORNL-3830 Period Ending May 31, 1965
ORNL-3945 Period Ending May 31, 1966
ORNL-4145 Period Ending May 31, 1967
ORNL-4272 Period Ending May 31, 1968
ORNL-4422 Period Ending May 31, 1969
ORNL-4572 Period Ending May 31, 1970
ORNL-4682 Period Ending March 31, 1971
ORNL-4794 Period Ending March 31, 1972
ORNL-4883 Period Ending March 31, 1973
ORNL-4966 Period Ending March 31, 1974
ORNL-5050 Period Ending March 31, 1975
ORNL-5172 Period Ending March 31, 1976
ORNL-5295 Period Ending March 31, 1977
ORNL-5383 Period Ending March 31, 1978
ORNL-5542 Period Ending March 31, 1979
ORNL-5757 Period April 1, 1979, to March 31, 1981
ORNL-5933 Period April 1, 1981, to March 31, 1983
ORNL-6153 Period April 1, 1983, to March 31, 1985
ORNL-6343 Period April 1, 1985, to March 31, 1986
ORNL-6490 Period January 1, 1987, to June 30, 1988

Contents

Acronyms and Initialisms	v
Abstract	xi
1. Overview	1
2. Waste Management and Environmental Programs	3
2.1 DOE Office of Civilian Radioactive Waste Management Programs	4
2.2 Waste Management Technology	20
2.3 Advanced Reactor Systems	57
2.4 Integrated Data Base Program	61
3. Waste Management Technology Center	69
3.1 Technical Support to Programs	69
3.2 Technology Development	72
3.3 Demonstrations	77
3.4 Information Exchange	81
4. Radiochemical and Isotope Programs	87
4.1 Radiochemical Engineering Development Center (REDC) Programs	87
4.2 HTGR Fission Product Transport	91
4.3 ²³³ U Program	91
4.4 Purex Flowsheet Development	92
4.5 AVLIS Processing	92
4.6 RSI Cesium Capsule Recovery	92
4.7 Metallurgical Examination Hot Cells	95
4.8 Isotopes Research Materials Laboratory	97
4.9 Isotope Enrichment	99
4.10 Radioisotope Processing	103
4.11 Packaging and Shipping	104
4.12 Isotope Distribution Office	105
5. Basic Science and Technology	109
5.1 Biotechnology Research	109
5.2 Chemistry Research	120
5.3 Engineering Research	124
5.4 Materials Research	139
5.5 Cold Fusion	143
6. NRC and EPRI Severe Accident Research Programs	153
6.1 Fission Product Release from LWR Fuel	153
6.2 Containment Fission Product Behavior	154
6.3 ACE "Phase-B" Iodine Chemistry Research	156
6.4 LACE Aerosol Code-Comparison Studies	157
7. Office of Safety and Operational Readiness	159
7.1 Health, Safety, and Environmental Protection	159
7.2 Quality Assurance	160
7.3 Training	160
7.4 Operational Readiness	161
7.5 OSOR Management and Information Systems	162

8. Administrative Resources and Facilities	163
8.1 Facilities	163
8.2 Capital Projects	163
8.3 Engineering Design and Facilities Development	165
Administrative Summary	167
Publications and Oral Presentations	169
Patents	233
Consultants	235
Advisory Committee	237
Chemical Technology Division Staffing Level and Financial Summary, FY 1989	239
Chemical Technology Division Organizational Chart	241

Acronyms and Initialisms

ACE	advanced containment experiment
ACS	American Chemical Society
AFB	Air Force base
ALARA	as low as reasonably achievable
ALMR	advanced liquid-metal reactor
ANL	Argonne National Laboratory
ANS	Advanced Neutron Source
ASTM	American Society for Testing Materials
AVLIS	Atomic Vapor Laser Isotope Separation
BEHP	bis(ethylhexyl)phthalate
BES	U.S. Department of Energy Office of Basic Energy Sciences
BNL	Brookhaven National Laboratory
BSR	Bulk Shielding Reactor
BV	Bethel Valley
BWR	boiling-water reactor
C&TD	Computing and Telecommunications Division
CAC	continuous annular chromatography
CAT	collection and transfer
CBH	cellobiohydrolase
CCIX	continuous countercurrent ion exchange
CDB	Characteristics Data Base
CESAR	Center for Engineering Systems Advanced Research
CH	contact handled
CH TRU	contact-handled transuranic (waste)
Chem Tech	Chemical Technology Division
CNSI	Chem-Nuclear Systems, Inc.
CO	capsule overpack
CPCF	Central Pollution Control Facility
CPR	cardiopulmonary resuscitation
CSAUP	Comprehensive (and continuing) Self-Assessment and Upgrade Program
CWMO	Central Waste Management Office
D&D	decontamination and decommissioning
dc	direct current
DCE	dichloroethylene
DCG	Derived Concentration Guide
DLWR	developmental light-water reactor
DOD	U.S. Department of Defense
DOE	U.S. Department of Energy
DOE/CH	U.S. Department of Energy/Chicago Operations
DOE/HQ	U.S. Department of Energy/Headquarters
DOE/OCRWM	U.S. Department of Energy/Office of Civilian Radioactive Waste Management
DOE/ORO	U.S. Department of Energy/Oak Ridge Operations
DOT	U.S. Department of Transportation
E&HP	Environment and Health Protection
EAFTF	Environmentally Acceptable Fire-Training Facility
EC&A	Engineering Coordination and Analysis
ECAMP	Environmental Compliance Assessment Program
EDR	electric dispersion reactor
EDS	energy-dispersive X-ray spectroscopy
Energy Systems	Martin Marietta Energy Systems, Inc.

EP	extraction procedure
EPA	U.S. Environmental Protection Agency
EPC	emulsion-phase contactor
EPRI	Electric Power Research Institute
ES&H	U.S. Department of Energy Office of Environment, Safety, and Health
ESA	Environmental and Safety Activities
ESCA	electron spectroscopy for chemical analysis
ESD	Environmental Sciences Division
ESTSC	Energy Systems Transportation Safety Committee
F&M	Finance and Materials
FBR	fluidized-bed reactor
FICA	Facility Interface Capability Assessment
FIU	Florida International University
FPDL	Fission Products Development Laboratory
FRG	Federal Republic of Germany
FSAR	final safety analysis report
FSM	Facility Safety Manager
FTIR	Fourier transform infrared (spectrometer)
FWMS	federal waste management system
FY	fiscal year
GC	gas chromatography
GE	General Electric Corporation
GET	general employee training
GPHS-RTG	general purpose heat source-radioactive thermoelectric generator
GPP	general plant projects
HAZWDDD	hazardous waste development, demonstration, and disposal
HAZWRAP	Hazardous Waste Remedial Action Program
HBCU/MEI	historically black colleges and universities/minority educational institution
HEPA	high efficiency particulate air
HFIR	High Flux Isotope Reactor
HGDP	Hanford Grout Disposal Program
HGMS	high-gradient magnetic field separations
HLW	high-level waste
HMSA	hybrid mean spherical approximation
HQ	headquarters
HSRD	Health and Safety Research Division
HTGR	high-temperature gas-cooled reactor
HVAC	heating, ventilating, and air conditioning
HWTP	Hazardous Waste Technology Program
IA	isotopes area
IAEA	International Atomic Energy Agency
ID	inside diameter
IDB	Integrated Data Base
IEC	Issues Evaluation Committee
IEF	Isotope Enrichment Facility
INEL	Idaho National Engineering Laboratory
IPNS	intense pulsed neutron source
IRML	Isotopes Research Materials Laboratory
IT	International Technology
ITE	in-tank evaporation
ITOM	Installation Transportation Operations Manager
ITP	International Technology Programs
ITSM	Installation Transportation Safety Manager
LACE	light-water reactor aerosol containment experiments

LANL	Los Alamos National Laboratory
LLW	liquid low-level waste
LLW	low-level waste
LLWDDD	low-level waste disposal development and demonstration
LWR	light-water reactor
MCC	Materials Characterization Center
MCS	monitoring and control stations
MEHC	Metallurgical Examination Hot Cells
MEI	minority educational institution
MFE	Magnetic Fusion Energy
MRF	Metal Recovery Facility
MRS	Monitored Retrievable Storage
MSD	Metropolitan Sewer District
MSR	molten salt reactor
MTIHM	metric tons of initial heavy metal
MTU	metric tons of uranium
MV	Melton Valley
MVST	Melton Valley storage tanks
NAC	Nuclear Assurance Corporation
NEESA	Naval Energy and Environmental Support Activity
NFA	nonfuel assembly
NOS	Naval Ordnance Station
NRC	U.S. Nuclear Regulatory Commission
NRWTP	Nonradiological Wastewater Treatment Plant
NWPA	Nuclear Waste Policy Act
OCRWM	Office of Civilian Radioactive Waste Management
OD	outside diameter
OFF	oldest fuel first
ORAU	Oak Ridge Associated Universities
ORGDP	Oak Ridge Gaseous Diffusion Plant
ORNL	Oak Ridge National Laboratory
ORO	Oak Ridge Operations
ORR	Oak Ridge Reservation
OSOR	Office of Safety and Operational Readiness
OSR	Operational Safety Requirement
P/T	partitioning/transmutation
P&E	Plant and Equipment
PATS	Packaging and Transportation Safety Program
PC	personal computer
PCB	polychlorinated biphenyl
PCRv	Prestressed Concrete Reactor Vessel
PDMS	Post Defueling Monitored Storage
PIP	Performance Improvement Process
PIUS/BWR	process inherent ultimate safety/boiling-water reactor
PNL	Pacific Northwest Laboratory
POC	performance objectives and criteria
PSP	Plasma Separation Process
PVC	polyvinyl chloride
PWR	pressurized-water reactor
PWS	process waste system
PWTP	Process Waste Treatment Plant
PY	Percus-Yevick (theory)
Q&TS	Quality and Technical Services Division
QA	quality assurance

QAP	Quality Assurance Plan
R&D	research and development
RAFB	Robins Air Force Base
RAS	rotary air stripper
RCRA	Resource Conservation and Recovery Act
RD&D	research, development, and demonstration
RDDT&E	Research, Development, Demonstration, Testing, and Evaluation
RDF	Radiochemical Development Facility
RDT	reactor drain tank
REDC	Radiochemical Engineering Development Center
RERTR	Reduced Enrichment Research Test Reactor
RFP	request for proposal
RH	remote handled
RH TRU	remote-handled transuranic (waste)
RHNC	reference hypernetted chain (theory)
RO	reverse osmosis
RRD	Research Reactors Division
RSI	Radiation Sterilizers, Inc.
RTF	Radioiodine Test Facility
RWMD	Reservation Waste Management Division
S&M	surveillance and maintenance
SANS	small-angle neutron scattering
SAR	safety analysis report
SARP	Safety Analysis Reports for Packaging
SAS	Statistical Analysis System
SAW	surface acoustic wave
SAXS	small-angle X-ray scattering
SDI	Strategic Defense Initiative
SEM	scanning electron microscopy
SFD	spent fuel disassembly
SFMP	Surplus Facilities Management Program
SFWG	Spent Fuel Working Group
SIMS	Systems Integration Modeling System
SNF	spent nuclear fuel
SOLMOD	systems integration operations/logistics model
SRL	Savannah River Laboratory
SSC	structures, systems, and components
SST	safe secure trailer
SWIMS	solid waste information management system
TASC	Training Accreditation Steering Committee
TCB	trichlorobenzene
TCCG	Technical Code Coordinating Group
TCE	trichloroethylene
TEC	total estimated cost
TGF	Transportable Grout Facility
TOS	transportation operations system
TRU	transuranic (waste)
TSA	technical safety appraisal
TSCA	Toxic Substances Control Act
TSLCC	Total System Life Cycle Cost
TTG	Transportation Technologies Group
UTF	uranium task force
VOC	volatile organic compound
VOST	volatile organic sampling train

WBS	work breakdown structure
WCA	Weeks-Chandler-Anderson (repulsive reference potential)
WDF	Waste Handling Packaging Plant Development Facility
WESF	Waste Encapsulation Storage Facility
WFO	Work for Others
WHC	Westinghouse Hanford Company
WHPP	Waste Handling and Packaging Plant
WIPP	Waste Isolation Pilot Plant
WIT	waste immobilization technology
WM&ECT	waste management and environmental control technologies
WMPO	Waste Management Program Office
WMTC	Waste Management Technology Center
WMTSG	Waste Management Technology Support Group
WNRE	Whiteshell Nuclear Research Establishment
WRAP	Waste Remedial Action Program
WSA	Waste Stream Analysis
WSUFMP	Weldon Spring Uranium Feed Materials Plant
XRD	X-ray diffraction

Abstract

This progress report summarizes the research and development efforts conducted in the Chemical Technology Division (Chem Tech) during the period July 1, 1988, through September 30, 1989. The following major areas are covered: waste management and environmental programs, the Waste Management Technology Center, radiochemical and isotope programs, basic science and technology, Nuclear Regulatory Commission and Electric Power Research Institute severe accident research programs, the Office of Safety and Operational Readiness, and administrative resources and facilities.

1. Overview

The Chemical Technology Division (Chem Tech) has outstanding facilities and personnel capabilities in chemical engineering and chemistry, as well as significant competence in several other engineering and scientific disciplines. It has major roles in a broad range of Oak Ridge National Laboratory (ORNL) programs for which chemical processing is a critical element. These programs are designed to explore selected aspects of nuclear and nonnuclear energy technologies and to develop advanced processes and systems to meet national priorities. Research and development (R&D) activities include basic and applied research and engineering, engineering development and assessments, and the operation of various pilot plants and specialized facilities. Most of the research programs are funded by the U.S. Department of Energy (DOE), although some important projects are carried out for other governmental agencies, and limited support is derived from industrial sources.

The major objectives of Chem Tech are:

- to perform basic R&D related to chemical separations and biotechnology;
- to perform chemical processing R&D related to nuclear fuel cycle processes;
- to perform R&D related to advanced waste management and environmental control technologies;
- to provide process chemistry and chemical engineering support to other divisions and programs within ORNL, other parts of Martin Marietta Energy System, Inc. (Energy Systems), and other DOE facilities;
- to produce, separate, package, and distribute transuranium elements and other radioactive and stable isotopes for use in research and in applications for medicine and industry;
- to perform R&D related to advanced isotope separation processes and isotope usage; and
- to enhance the transfer of new technology developed within Chem Tech and elsewhere to U.S. industries, thus improving the technical competence and the industrial competitiveness of the nation.

Chem Tech's basic research programs in photo-biochemistry, enzyme kinetics, and fixation of enzymes on substrates continue to be internationally respected. Our activities in the development of bio-

processing applications for fossil energy have continued to be reviewed favorably by DOE and will be expanding further in FY 1990. Collaborative programs with the ORNL Environmental Sciences Division and with the University of Tennessee on in situ biodegradation of organic pollutants in the earth have expanded over the past year. As usual, our basic programs in biotechnology and chemical separations continue to attract industrial attention, and several licensing activities have been pursued. A small effort to study cold fusion phenomena was initiated during this period.

Chem Tech is deeply involved in nearly all aspects of Energy Systems' current waste management activities as well as in planning for future waste operations that will be acceptable to state and federal regulatory bodies. Working with groups at various other DOE and Department of Defense facilities, we are also developing and demonstrating in the field a variety of advanced waste management and environmental control technologies. Chem Tech is involved in DOE's work on permanent federal waste repositories for commercial radioactive wastes, providing support in systems integration, project management, waste packaging, and transportation. We continue to maintain DOE's national data base on present and projected radioactive waste in the United States, and have a significant role in the development of data bases encompassing waste generated on all five Energy Systems sites.

Radiochemical processing programs within Chem Tech emphasize operation of the Radiochemical Engineering Development Center (REDC) for DOE's Office of Basic Energy Sciences (BES) for the isolation, purification, and packaging of Bk, Cf, Es, and Fm for research uses. The REDC was created during FY 1988 and utilizes facilities that historically have been referred to as the Transuranium Processing Plant and the Thorium-Uranium Recycle Facility. In addition to these BES activities, the REDC initiated a long-term program for the recovery of isotopes of americium for other DOE sponsors during FY 1989. We continued to conduct hot cell programs for the Nuclear Regulatory Commission on the transport and deposition of fission products under hypothetical nuclear reactor accident conditions, but expect this work to continue its recent decline. Chem Tech is also engaged in supporting activities for ORNL's High

Temperature Gas-Cooled Reactor and Consolidated Fuel Reprocessing programs and for the Energy Systems Atomic Vapor Laser Isotope Separation (AVLIS) Program.

During this period, Chem Tech expended considerable effort on the management of the isotope production and distribution activities that were assigned to the division in June 1988. The inherent problems deriving from the antiquated facilities used in this program increased in importance during this period as both internal and external audits cast light on them. In the absence of funding to address such issues, many radioisotope activities will probably be phased out in the coming year. Activities related to stable isotopes have likewise been impacted, but are expected to continue into the future.

The Chem Tech Office of Safety and Operational Readiness (OSOR) was created in 1989 to integrate the functions of quality assurance, safety, radiation control, environmental protection, hazards communication, training, crisis management, operational readiness, and the division's self-assessment activities. The integration of these functions will enable Chem Tech to meet the greatly increased formality and documentation requirements in these areas. Under the guidance of the Division Director, the OSOR also serves as the interface between Chem Tech and the safety and operational readiness functions of ORNL, Martin Marietta Energy Systems, Inc., and DOE.

2. Waste Management and Environmental Programs

During this report period, Chem Tech's involvement in waste management and environmental control technologies (WM&ECT) continued, with major expansion in areas related to DOE site waste management. In most areas, Chem Tech's involvement is a combination of (1) performing the technical activities required by a particular program and (2) program management, often involving other ORNL divisions and the private sector. Chem Tech is involved in the following principal WM&ECT areas: DOE Office of Civilian Radioactive Waste Management (DOE/OCRWM), DOE site-related waste technology, DOE generic waste technology, Work for Others (WFO) waste technology, advanced reactor systems, and the Waste Management Technology Center.

First, the tasks supported by DOE/OCRWM remain the single largest WM&ECT program in Chem Tech. The most significant areas of DOE/OCRWM involvement for Chem Tech are as follows:

1. **Transportation:** Providing technical support related to the transportation of highly radioactive spent fuel, including hardware development, transportation operations planning and support, and transportation systems studies;
2. **Systems Integration:** Studies and analyses to ensure that the various parts of the DOE/OCRWM system (generators, storage, repository, transportation) will fit together and work in a near-optimal way; and
3. **Integrated Data Base:** DOE's reference source of data on radioactive wastes. This is jointly supported by various parts of DOE; DOE/OCRWM is the largest sponsor.

Because the number of repository sites being evaluated has been narrowed to only one (Yucca Mountain at the Nevada Test Site), Chem Tech's involvement in repository-related activities has declined substantially. This is expected to remain the case unless DOE/OCRWM begins examining alternative disposal technologies.

Second, Chem Tech is involved in a broad range of WM&ECT technology activities related to hazardous materials on the Oak Ridge Reservation (ORR). The sponsors for this work are internal in that overall programmatic planning and direction is provided by other Energy Systems organizations, who interact with the DOE sponsors. Because of the heightened interest in WM&ECT at DOE sites, work in this area has increased substantially, and this trend is expected to continue. Major areas of Chem Tech involvement include the following:

1. **Biotechnology:** Development of in process and in situ WM&ECT for remediation and treatment of hazardous materials ranging from heavy metals to organic chemicals;
2. **Waste Processing Design:** Planning and design support for many of the new WM&ECT facilities that are being built on the ORR;
3. **Liquid and Gaseous Waste Treatment Processes:** Undertaking activities ranging from a systems analysis of the ORNL low-level waste management system to developing processes for treating process wastewaters;
4. **Waste Handling and Packaging Plant research and development (R&D):** Process research and development for a major new ORNL facility to treat and package transuranic wastes for eventual disposal in the Waste Isolation Pilot Plant;
5. **Decontamination and Decommissioning (D&D) Technology:** Planning and technology development activities related to the D&D of ORNL facilities and the gaseous diffusion plant buildings; and
6. **Transportation Support:** Providing technical support to multiple parts of Energy Systems concerning the oversight of package certification and transportation compliance.

Third, in addition to the waste technology related to the ORR, Chem Tech has a significant amount of other DOE-supported WM&ECT activities. These are diverse and range from continuing technical support in the cleanup of the Three Mile Island Unit II reactor to the development of expert systems for

use in WM&ECT applications to study partitioning-transmutation as a waste management option. Many of these represent new initiative areas, and substantial growth is possible in the future.

Fourth, WFO waste technology is a growing area. Many government organizations other than DOE have WM&ECT problems that are very similar to those being addressed by DOE, except that they do not involve radioactive constituents. These organizations are not generally strong in areas related to WM&ECT, nor do they desire to become so. Chem Tech is pursuing the opportunity to develop technology to solve the problems of these organizations. The predominant sponsoring organizations are the various services making up the Department of Defense (DOD), especially the Air Force. This work not only provides interesting and relevant work for Chem Tech, but benefits DOE because solutions developed for DOD are often directly applicable to DOE problems.

The work ranges from developing air-stripping and soil-venting technologies to remediate contaminated sites to conducting environmental surveys to identify problem areas. This work is expected to continue and increase because of the generally heightened interest in waste management on government sites and the impetus for collaborative interagency ventures to efficiently utilize government resources.

Fifth, Chem Tech is involved in the development of two very different advanced reactor systems. The first is the Advanced Neutron Source (ANS), which is a follow-on to the existing High Flux Isotope Reactor (HFIR) at ORNL. The primary purpose of this reactor is to provide a state-of-the-art facility housing an intense neutron source for use in investigating the structure of materials and in producing radioisotopes. Our involvement is related to the design of the high-power density core of the ANS as it relates to heat transfer considerations.

A second Chem Tech involvement in advanced reactor systems concerns a fledgling task to develop passively safe light-water reactors (LWRs). The results to date are most promising, and it is anticipated that this work will lead to a proof of principle within the next few years.

Sixth, the Waste Management Technology Center (WMTC) (see Sect. 3 for details) is an organization centered in Chem Tech, but involving many members of other ORNL divisions and subcontractors. Its primary purpose is to provide a technical resource to Energy Systems' efforts to develop and implement appropriate and cost-effective WM&ECT solutions. Its activities range from providing technical support and management capabilities to major Energy Systems programs (e.g., program to establish new low-level waste disposal facilities) to investigating waste immobilization technologies (especially grout-based) to conducting demonstrations of new private sector technologies.

2.1 DOE Office of Civilian Radioactive Waste Management Programs

DOE/OCRWM was established by the Nuclear Waste Policy Act (NWPA) of 1982.¹ In this act, DOE is directed to take title to the spent fuel and other high-level wastes (HLWs) from civilian and defense programs and to manage them safely. This broad mandate incorporates all aspects between picking up the spent fuel and HLWs at the reactors (or other sites) and final closure of the geologic repositories where the wastes will be emplaced for ultimate disposal. In other words, DOE is charged with putting into place the system to fulfill its mandate and then acting to discharge its legal obligations.

Chem Tech is assisting DOE in this program with a series of related tasks that include collecting the basic data necessary for the design and licensing of the various facilities, studying the options for packaging and transporting the wastes and spent fuels, determining the development needs of the program, and ensuring that all of the facilities and their system interfaces match properly. Much of this work is carried out by subcontractors who are specialists in their fields.

In the following subsection on transportation programs, summaries of our work in the areas of transportation operations, logistics and modeling, transportation capabilities near commercial power reactors, and shipping cask testing are presented. The subsection on systems integration programs includes an assessment of existing reactors and facilities to ensure that no interfaces will be mismatched, the acceptance criteria that will be used by a national repository, the characteristics of

all the wastes and spent fuel that DOE will need to handle, the logistics required to accomplish this task, an assessment and qualification of computer codes used in the design and operation of facilities, an analysis of the potential impacts of the Resource Conservation and Recovery Act (RCRA) on a repository,² and a history of the path taken to site a national geologic repository.

2.1.1 Transportation Programs

During the reporting period, the Civilian Radioactive Waste Management Transportation Program within Chem Tech focused primarily on providing detailed support to DOE/Chicago (DOE/CH) in preparing for the eventual transport of spent nuclear fuel (SNF) and HLW within the civilian Federal Waste Management System (FWMS). This support focused on three areas: operations support, which provided planning and assessment for future transportation operations within the FWMS; logistics and modeling support, which provided detailed routing and shipment modeling capabilities to systems studies performed within Chem Tech and by other DOE and DOE laboratory organizations; and near-site transportation infrastructure support, which resulted in the initiation of an assessment of the transportation-related aspects of the area immediately surrounding each reactor site to be served by the FWMS. In addition, Chem Tech provided limited support to DOE/Idaho in reviewing early aspects of five cask designs being prepared by five contractors throughout the United States.

The DOE/OCRWM originally assigned Oak Ridge Operations (DOE/ORO) the lead responsibility for the development of an operational transportation system to support its transportation-related activities. ORO, in turn, relied upon Chem Tech to take the lead in the development of this capability. In June 1988, DOE/Headquarters (HQ) expressed its intent to consolidate the primary responsibility for transportation operations with its other civilian transportation program elements then located at DOE/CH. This transition of the operations effort was made effective October 1988. Chem Tech's role in the operations aspects of the civilian transportation program has remained similar to that under DOE/ORO.

Transportation Operations Support (TOS)

Eventually, the FWMS must have a TOS capable of transporting large quantities of SNF and HLW from originating sites, such as reactors and DOE/HLW generating sites, to receiving sites such as a Monitored Retrievable Storage (MRS) facility or a geologic repository. The system must be capable of adapting to the various requirements imposed by the almost 100 different sites to be served and by the contractual arrangements that have been established between DOE and the commercial reactor operators. The system, as currently envisioned, must be capable of providing these services with very limited fixed guidance on what materials are to be shipped and where they are to be shipped from. The only basis upon which the system can currently be developed is that a specified total tonnage of wastes (SNF and HLW) must be transported from reactor sites on an annual basis; even the year in which the FWMS is to become operational is currently undefined. In order to support DOE/CH in understanding what will be required to start up and bring a TOS to full operation, Chem Tech has performed a series of studies that

1. look at developing a Systems Description and a definition of the functions that must be performed by a TOS;
2. provide a detailed analysis of the impacts on the TOS of the "Standard Contract" that has been agreed to between DOE and the commercial reactor operators;
3. develop a concept, cost, and schedule for a Cask Maintenance Facility to support the TOS;
4. define the capability of the existing, commercially available casks to transport spent fuel as the FWMS begins operation; and
5. define the amounts of spent fuel that have been transported within the United States during the past 25 years to serve as a basis for comparison for what will be required in the future.

The results of all of these studies, and additional smaller activities undertaken during FY 1989, are aimed at assisting DOE in preparing short- and long-term plans for the development of a TOS to serve the civilian program.

The transportation functions for the FWMS were initially defined in a document completed in

FY 1988, which included a detailed presentation of the functional relationships using functional flow block diagrams.³ During FY 1989, these diagrams and the functions associated with them were reviewed in detail, modified as needed, and then used to prepare detailed descriptions of each function. As this work proceeded, a number of issues that need to be resolved through study or DOE decisions were identified, and we began to develop proposed methods for issue resolution. Finally, in conjunction with the development of the descriptions of the functions, a detailed systems description of the TOS was developed. At the first level, five subsystems were identified:

1. planning and control,
2. carriage,
3. field operations,
4. servicing and maintenance, and
5. transportation casks.

Each of these subsystems was then described in terms of the next lower level of subsystem elements. For example, the field operations subsystem is described as follows:

3. field operations subsystem,
 - 3.1 facility interface equipment,
 - 3.2 emergency response operations support,
 - 3.3 waste acceptance operations,
 - 3.4 waste generator technical support, and
 - 3.5 waste generator facility interface information.

The contents of each of these subelements were then described. The document providing the revised functional flow block diagrams, the detailed descriptions of the functions, and the TOS systems description was submitted in draft form to DOE/CH in June 1989 and is under review by DOE.

A series of white papers assessing the impacts of the "Standard Contract" on transportation operations and defining methods for resolving these impacts was developed during FY 1989. As a result, an 11-step process that addresses the Standard Contract issues from an upper, integrated level was developed. This process will provide an overview perspective of operating TOS using different interpretations of the contract. Work has been initiated using these 11 steps:

1. identifying operational assumptions,
2. identifying allocation options,
3. forecasting expected fuel quantities by site,
4. developing fuel selection rules,
5. evaluating expected fuel characteristics,
6. identifying and characterizing site/facility constraints,

7. identifying and characterizing near-site infrastructure constraints,
8. estimating shipment capabilities,
9. identifying alternative transit options,
10. identifying feasible operating scenarios, and
11. iterating analysis based upon outside reviews.

In addition, a method for defining site-specific servicing problems and developing site-specific servicing plans was outlined. Both of these processes were submitted to DOE/CH and are under review by DOE.

The Cask Maintenance Facility feasibility study was completed, and a draft document was submitted to DOE/CH in February 1989. This document is under review by DOE. The results of the study indicate that a Cask Maintenance Facility to support the FWMS fleet in full operation is feasible and would not require any significant technology development. The design and construction of the facility are estimated to take 110 months from the start of conceptual design to the start of operations if the project is pursued as a government-owned, contractor-operated major system acquisition by the OCRWM. The cost of constructing a stand-alone facility is estimated to be \$83 million in constant FY 1989 dollars. This includes \$8 million for preliminary (expense-funded) project activities and \$75 million for the capital cost of the facility. Escalated over the project cycle, assuming the start of operations would be first quarter of FY 2003, the total cost becomes \$143 million.

The capabilities of the existing, commercially available cask fleet and its current certification status were assessed and documented in draft reports that were submitted to DOE/CH; these reports are under review by DOE. The results of these studies indicate that the existing fleet and additional casks that are under development privately and can be expected to be added to the fleet will only provide a small portion of the capability needed to serve the FWMS in its early years. For example, during the first year of FWMS operation, 1200 metric tons of uranium (MTU) of SNF is projected to be shipped; however, the existing cask fleet is expected to only be able to ship between 60 and 600 MTU, depending upon the assumed mix of casks available, the reactors and receiving sites served, and the efficiencies attained in operations. Within a few years, the FWMS is expected to be shipping an equivalent of between 6300 and 3400 MTU/year, depending upon whether the system does or does not include an MRS facility, respectively. Thus, the existing cask fleet,

even with the most optimistic assumptions, can only serve a small fraction of the needs of the FWMS.

As an additional basis of comparison for what must be accomplished in the United States before an effective TOS can be implemented to serve the FWMS, a report was developed that documents all commercial and research reactor SNF shipments within the United States over the past 25 years. The document was submitted to DOE/CH and is under review. Results indicate that only a small fraction of the SNF that will need to be transported in the future has been moved in the past. For example, from 1964 through August 1989, 1861 MTUs of commercial SNF have been transported in the United States. This is significantly less than the annual amount that will be transported when the FWMS attains steady-state operations. The amount of research reactor fuel moved is a small increment of the above when viewed in terms of MTUs as compared to the commercial shipments. For example, from 1983 through August 1989, only 20 MTUs of research reactor fuel were shipped in (and into) the United States.

Transportation Logistics and Modeling

The Transportation Logistics and Modeling Analysis project has primary responsibility for the support of DOE/OCRWM transportation projects managed by the Chicago Transportation Program Office. The two areas of support are routing analysis and logistics support.

Routing Analysis

The major emphasis of the routing analysis tasks has been applying the HIGHWAY and INTERLINE routing models to support OCRWM activities. The routing models were used to support specific requests from the MRS Commission and the Repository Project Office for routing studies. The results of the following activities have been reported to DOE in a series of letter reports.

The MRS Commission requested a study to determine a possible MRS location that would minimize the transportation effort needed to move SNF from the reactors to a repository. The assumptions for this study included:

1. SNF would be accepted on an oldest-fuel-first basis;
2. all from-reactor shipments would be sent to the MRS;
3. reactors with rail service would make all fuel shipments in three-car dedicated trains;
4. intact SNF assemblies would be transported from the MRS to the repository in five-car dedicated trains; and
5. a single repository would be located at Yucca Mountain, Nevada.

The site that would minimize the total shipment miles required to transport all of the SNF expected to be emplaced in the first repository is located in the southern part of West Virginia. Part of the study included generating detailed highway and rail routes from the reactors to the potential West Virginia site and the most probable rail routes between there and the repository.

Several additional requests for routing analysis were also received from the MRS commission. One request involved estimating rail routes for transporting SNF that avoid urbanized areas having a population exceeding 500,000 people. It was noted that several of the routes still passed through some of the large urbanized areas because no other option is available. For example, routes from the San Onofre site in southern California can either pass through San Diego and then through a part of Mexico or pass through the Los Angeles area. Another study included analyzing the rail infrastructure in the vicinity of reactors that do not currently have rail service to determine the feasibility of constructing a rail spur to these sites. For a variety of reasons, 22 reactor sites do not have a rail spur. If the appropriate right-of-way can be acquired, it appears that a rail spur could be added to nine of these sites. In addition, the study was expanded to identify reactors that are likely to lose rail service because of rail line abandonments in the foreseeable future.

A rail-routing study was performed for the Repository Project Office analyzing the impact of using alternative rail access points for transporting radioactive materials to the Yucca Mountain site. Currently, the nearest rail line to the Yucca Mountain repository site is in Las Vegas, Nevada, and a rail spur will have to be constructed to the repository site. A number of potential rail spurs were included in the study. Rail spurs originating in the northern part of the state were of particular interest. These spurs would be longer than a spur originating in the Las Vegas area but would have the advantage of passing through areas of lower

population density and also provide access to two major rail carriers rather than a single carrier at Las Vegas.

Logistics Support

The major activity in the logistics support area was the revision of the Preliminary Cask Fleet Projections report, which is currently in draft form. This report was modified and expanded to include the 1989 Total System Life Cycle Cost transportation assumptions. A number of scenarios were investigated in which the total cask fleet varied from 74 to 132 casks. Cask loading and unloading times have a strong influence on the cask fleet numbers, and a small change (on the order of 1 or 2 d) will produce a significant change in the projected cask fleet sizes. The use of dedicated trains to transport SNF from reactors to an MRS or a repository increased the cask fleet by ~5%, as shown in Table 2.1. Cask fleet projections were not as sensitive to other transportation parameters, such as average transport speed or cask utilization, that were investigated in this study.

Near-Site Transportation Infrastructure

DOE has decided to evaluate, for each reactor site, the capabilities of the existing near-site transportation networks (road, rail, and barge) to accommodate SNF shipments and to generally assess the potential to upgrade these capabilities, should that appear desirable. The capability of road, rail, and barge systems to accommodate legal, overweight, or oversize radioactive waste shipments is being assessed. Site-specific trends or future plans for railroad and barge facilities will be documented.

Martin Marietta Energy Systems, Inc., has entered into a 2-year contract, starting in June 1989, with Nuclear Assurance Corporation (NAC) to assess the near-site transportation networks within ~25 miles of each reactor site. Plans call for visits to each reactor site, the collection of significant amounts of transportation-related data, and an assessment by NAC, based upon the data, of the best method of transporting SNF and other highly radioactive wastes from the site.

Table 2.1. Summary of cask fleet projections for different scenario assumptions

Scenario	Maximum number of casks required		
	Truck	Rail ^b	Total
1. MRS ^a and all railroad casks transported as general freight	18	56	74
2. MRS and only loaded rail casks transported in four-car-unit trains	18	61	79
3. MRS and all rail casks transported in four-car-unit trains	18	61	79
4. MRS and all rail casks transported annually in a single-unit train from each site	18	85	103
5. No MRS and all rail casks transported as general freight	35	45	80
6. No MRS and all rail casks transported in four-car-unit trains	35	48	83
7. No MRS; all spent fuel shipments transported in truck casks	63	20	83
8. No MRS; all spent fuel shipments in truck casks; extended loading/unloading times	112	20	132

^aMonitored retrievable storage.

^bIncludes 20 rail casks for transporting HLW.

The first 3 months of the contract were spent developing (1) site visit protocols, (2) details of the data base (which will act as the repository for the data that is collected), (3) arrangements for the training of personnel, (4) acceptable methods for contacting the utilities concerning impending visits, (5) the methodology for contacting state and local people, and (6) the tentative schedule for the site visits. Actual site visits are scheduled to start in October 1989.

Cask Testing

The Y-12 Plant has embarked on the development of a new series of packages for shipping classified materials. Conclusions of the safety analysis reports (SARs), which analyze and evaluate the behavior of the packages under a variety of normal and accident conditions, will be based on a combination of analysis and full-scale package testing. As a result, the Transportation Technologies staff was asked to subject the prototype Y-12 packages to the drop tests specified in the federal regulations.

Under the Chemical Technology Division Quality Assurance (QA) requirements for drop testing,⁴ a drop test plan was developed for each prototype package. The plan, based on an overall plan developed by Y-12, described the specific drop tests to be carried out on that package, the data that were to be collected, a safety and operations check-list, and the applicable QA topics that were invoked.

The series of packages that were tested have been designated as follows: DT-19, DT-20, DT-21, DT-22, and DT-23. At least one model of each package was dropped 9 m (30 ft) onto a solid, unyielding surface and dropped onto a 15-cm (6-in.)-diam steel punch from 82 cm (40 in.). These packages were returned to Y-12 and subjected to the high-temperature thermal test and final evaluation.

Additional prototype packages were subjected to normal handling (drop) tests that are specified in the federal regulations. These packages were also returned to Y-12 for evaluation of results.

2.1.2 Systems Integration Programs

A significant activity in the development of the DOE FWMS is integrating its components, which include handling, packaging, transportation, storage, and final disposal of SNF and HLW. The objective of the DOE Waste Systems Integration program is to ensure that the components of FWMS will constitute an efficient and safe system that operates as a unified entity. A major factor in accomplishing this objective involves collection of data and development of information and capabilities used by all program elements.

In support of this objective, Chem Tech manages the ORNL Systems Integration program, providing OCRWM information and technology with which to integrate the FWMS. Typically, the Systems Integration activities have been focused on the development of special data bases such as the Facilities Interface Capability Assessment, Waste Acceptance Criteria, and Waste Characteristics; the development of system models such as Waste Stream Analysis and Systems Integration Operations/Logistics Model (SOLMOD); and the performance of specific system studies.

Facility Interface Capability Assessment (FICA)

The interface between the nuclear reactor waste generator, the waste transportation system, and the waste repository can have a significant impact on the FWMS performance since a common item, the shipping casks, must be handled by all of them. In an effort to understand and characterize these interfaces, a subcontract was initiated in 1986 with NAC to perform a two-phase FICA. The study involves evaluating each utility's capability to handle, store, and ship SNF and determine where facility upgrades could benefit the FWMS. Phase I has been completed. Phase II, which involves a visit to each reactor site to verify information obtained in the Phase I literature search and to obtain

missing information, was initiated in FY 1988. The site visits continued through this reporting period, and seven sites remain to be visited. The evaluations of the sites visited through September 30, 1989, are presented in a FICA Interim Topical Report.⁵ Two meetings to assess the progress of the site visits were held with the Utility Working Group. The Working Group has provided valuable support in establishing lines of communication with the utilities and in facilitating the site visits; their interaction will continue.

Waste Acceptance Criteria

The NWPA requires DOE to accept SNF and HLW from civilian nuclear reactor plants and to manage it in a manner leading to its eventual disposal in a geologic repository.¹ The possibility that the SNF and HLW may be handled, treated, or packaged to different degrees by various waste generators requires the development of waste acceptance criteria that will result in the optimization of the FWMS performance. Criteria for acceptance, delayed acceptance, or nonacceptance are being developed in the following areas:

1. failed fuel,
2. consolidated fuel,
3. non-fuel-bearing wastes from consolidation and other sources,
4. fuel in metal storage casks,
5. fuel in multielement storage canisters,
6. inspection and testing requirements for wastes,
7. minimum acceptance canister criteria, and
8. defense and commercial high-level waste packages.

This work is being performed by E. R. Johnson Associates, Inc., under subcontract to ORNL.

Draft criteria reports were prepared and submitted to ORNL on failed fuel, consolidated fuel, transportable storage casks, non-fuel-bearing components, fuel in multielement storage canisters, and fuel in metal storage casks. The draft reports are undergoing peer review by a selected group of experts. Following a peer review group meeting, the reports will be revised to include peer comments and will be issued in final form early in FY 1990. The two remaining draft reports, "Inspection and Testing Requirements for Wastes" and "Defense and Commercial High-level Waste Packages," are scheduled to be completed by the end of 1989.

A companion report, "The Incentives and Feasibility for Direct Measurement of Spent Nuclear Fuel Characteristics in the Federal Waste Management System," was completed and given a wide distribution.⁶

Waste Characteristics Data Base (CDB)

Introduction

The OCRWM is responsible for the SNF and HLW that will eventually be disposed of in a geologic repository. The two major sources of these materials are commercial LWR SNF and immobilized HLW from DOE nuclear materials production plants. Other wastes that may require long-term isolation include non-LWR spent fuel and miscellaneous sources such as activated metals. Detailed characterizations, which include physical, chemical, and radiological properties, are required for all of these materials. Radiological properties must take into account decay as a function of time. In addition, the present inventories and projected quantities of the various wastes are needed. This information has been assembled in a CDB, which provides data in four formats: hard-copy standard reports, menu-driven personal computer (PC) data bases, program-level PC data bases, and mainframe computer files.

This data base is an integral part of the systems integration approach being used by OCRWM. Specifically, the data base provides a standard set of self-consistent data for use by the various areas of responsibility within OCRWM, including systems integration and waste stream analysis, storage, transportation, and geologic disposal. The data will be used for design studies, trade-off studies, and system optimization by OCRWM and their supporting contractors. The Waste CDB provides primary input to OCRWM's Systems Integration Modeling System (SIMS) (Fig. 2.1).⁷

Background

The CDB is organized by four major waste stream categories: LWR spent fuel, immobilized HLW, non-LWR spent fuels, and miscellaneous wastes. These four categories are divided into eight types (Fig. 2.2); LWR spent fuel, the largest single

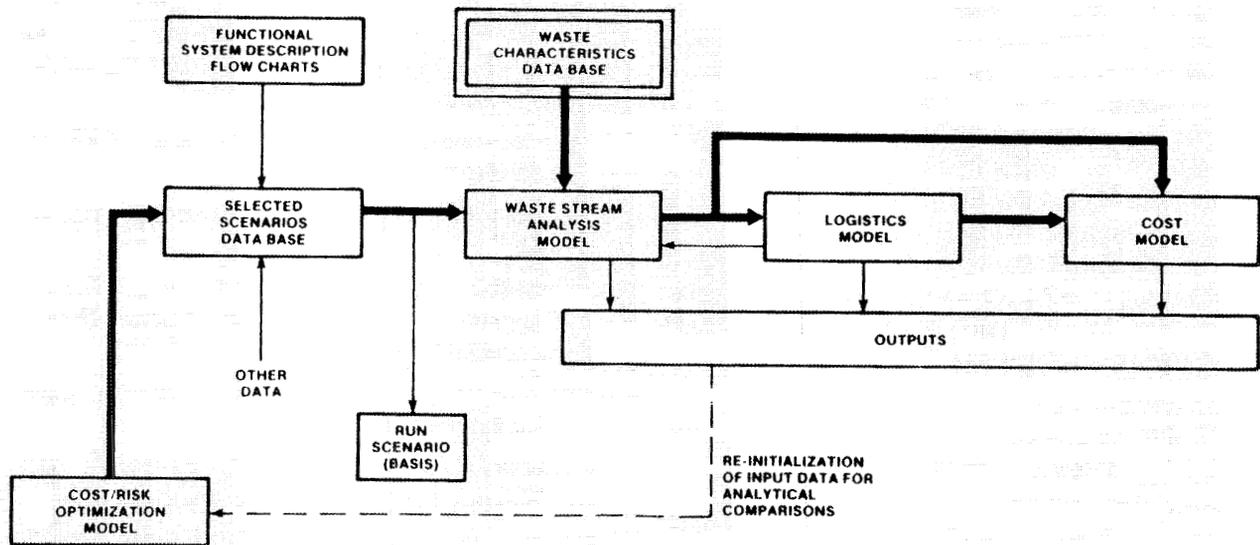


Fig. 2.1. Waste Characteristics Data Base role in Systems Integration Modeling System.

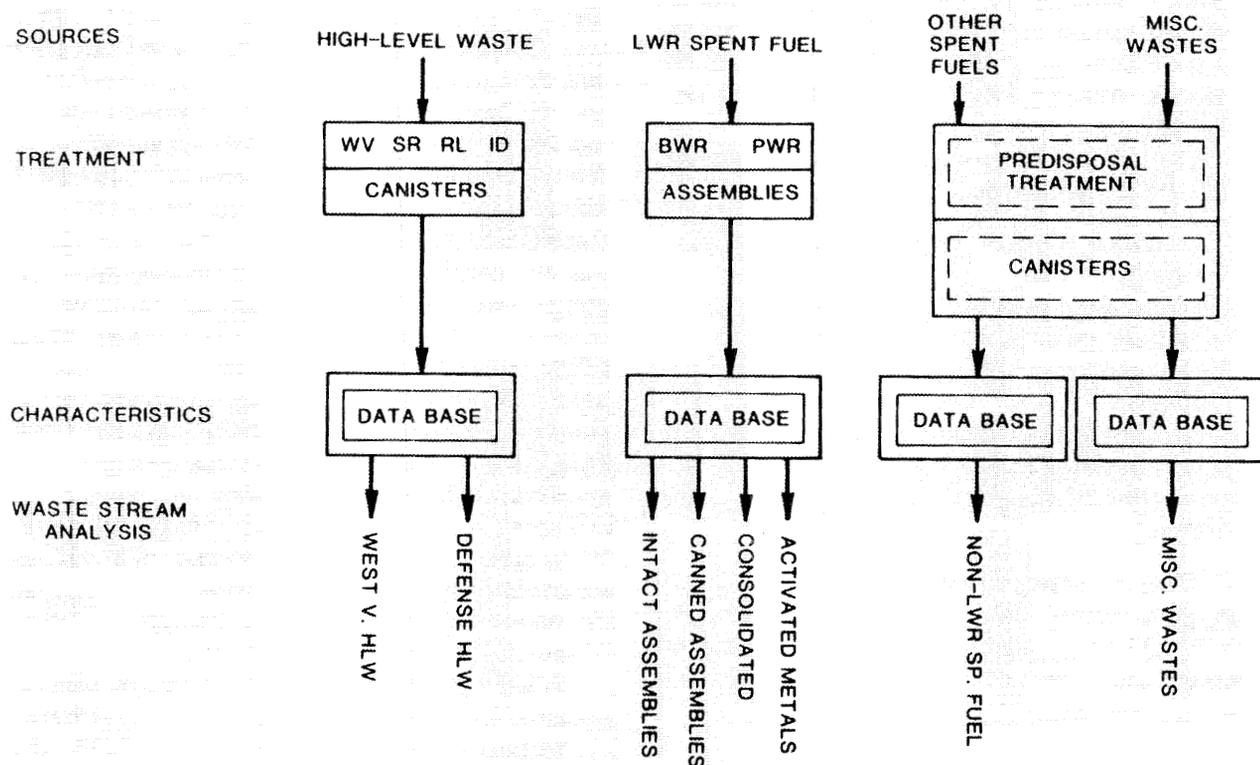


Fig. 2.2. Major input sources to the Waste Characteristics Data Base and derived output categories.

source, encompasses four of these. The four major categories, plus a summary, comprised the five major sections of the first characteristics report, which was issued as an eight-volume set.⁸ Six of these volumes consist of 16 appendixes that provide supporting information to the other two volumes.

Five of the appendixes are Users' Guides to as many menu-driven PC data bases. No programming skills are required to use these data bases designed for use on IBM-PCs or PC-compatibles and available through ORNL (ordering information is given on p. v of ref. 8). These menu-driven data bases cover the following topics:

1. LWR radiological data,
2. LWR assemblies and spent fuel disassembly (SFD) hardware descriptions,
3. high-level waste data,
4. LWR nonfuel assembly (NFA) hardware descriptions, and
5. LWR spent fuel quantities.

The previously mentioned data bases, as well as many of the internal working files, are written in dBASE III, which permits tabulation of special reports and interactive output. Mainframe files are used to generate some of the files as well as some of the hard-copy reports. Their use involves Statistical Analysis System (SAS) and FORTRAN programming.

The radiological properties are calculated using ORIGEN2⁹ and include:

1. quantities of each nuclide (grams or gram-atoms);
2. radioactivity, total and by nuclide (curies);
3. thermal power, total and by nuclide (watts);
4. photon energy spectra, by energy group (18 groups);
5. neutrons from spontaneous fission (per second);
6. neutrons from (α ,n) reactions (per second); and
7. quantity of each element (grams or gram-atoms).

These properties are decayed, over time, for 24 (or more) periods from 1 to 1 million years. For LWR SNF, the integral heats are also available between any 2 times out of a set of 38 times, also spanning periods of 1 to 1 million years.

Recent Work

Work during this reporting period was directed toward upgrading, updating, and expanding the existing data files and data bases in preparation to

reissue both the hard-copy report and the supporting PC data bases during FY 1990. Some of this work has been published, and some is in the process of being formally reported. The topics covered include:

1. an improved classification scheme for LWR fuel assemblies,¹⁰
2. a description of shipping and storage casks for SNF,¹¹
3. physical descriptions of General Electric Company (GE) boiling-water reactor (BWR) fuel assemblies,¹²
4. sensitivity of ORIGEN2 to various input parameters,¹³ and
5. characteristics and criticality aspects of non-LWR and special-case LWR spent fuels.¹⁴

Each of these topics is described briefly in the following paragraphs.

Because over 100 LWRs operate nationwide, representing designs by four primary vendors, and reload fuel manufactured by both these vendors and additional suppliers, a wide variety of fuel assembly types are in existence. The Waste CDB project required a national classification scheme for these fuels. To develop the classification scheme, extensive information on the fuel assemblies that have been and are being manufactured by the various nuclear fuel vendors was compiled, reviewed, and evaluated. It was determined that it is possible to characterize assemblies in a systematic manner, using a combination of physical factors. A two-stage scheme was developed consisting of 79 assembly types that are grouped into 22 assembly classes. The assembly classes are determined by the general design of the reactor cores in which the assemblies are, or were, used. The general BWR and pressurized-water reactor (PWR) classes are divided differently, but both are based on reactor core configuration. This system includes all known existing fuel types and is adaptable to take into account any future additions. If desirable or necessary, a third level of detail can be added to the classification scheme. This scheme can be applied to other areas and is expected to be of value to many OCRWM programs.

A wide variety of shipping and storage casks is available for SNF; their dimensions and capabilities are frequently of interest to users of the CDB. The physical descriptions of 6 transport casks and 11 storage casks were tabulated, along with drawings and photographs.

The physical characteristics of fuel assemblies manufactured by GE for BWRs were obtained from

various literature sources. These assembly descriptions were classified into assembly types based on the GE reactor product line, the CDB assembly class, and the GE fuel design. Thirty production assembly types were identified. Descriptions of special (nonstandard) fuels were also reported.

Extensive applications of the LWR radiological data base uncovered certain areas where the tabulated data exhibited discontinuities, primarily for the higher actinides, in the burnup region where the transition was made from standard to high-burnup ranges. A series of sensitivity tests showed the two major improvements needed to correct this were (1) a more precise use of uranium enrichment values and (2) utilization of newly recalculated effective cross sections. In addition, cycle data reflecting current practices and updated structural material descriptions will be used in revising the ORIGEN2 calculations for LWR spent fuel.

Two important categories of future repository wastes, non-LWR spent fuels and special LWR spent fuels, were evaluated regarding their quantities and packaging requirements, including criticality aspects. Assuming that canisters similar to those planned for vitrified HLW and conventional LWR fuel assemblies prove to be acceptable for these fuels, it is estimated that about 1400 such canisters will be needed for the non-LWR and special LWR spent fuels on hand and projected through the year 2020. Approximately half of these canisters are for high-temperature gas-cooled reactor (HTGR) spent fuel (Peach Bottom-1 and Fort St. Vrain), a quarter are for TMI-2 spent fuel and core debris, and the balance are fuels from a score of diverse sources. Two common canister sizes were adequate for all of these fuels: 24-in. OD by 12 ft long, and 28-in. OD by 15.6 ft long. Since many of these fuels are of higher enrichment or lower burnup than standard LWR spent fuel, estimation of the number of fuel assemblies that could be safely placed in a canister required preliminary calculations of neutron multiplication factors to ensure noncriticality. These estimates showed that the canister configurations used would provide more than adequate volume for the neutron poisons needed for this purpose. Thermal outputs per canister were estimated for Fort St. Vrain, TRIGA, and Pulsar fuels; the results showed that these fuels had thermal outputs per canister much lower than those of standard LWR spent fuels and vitrified HLW.

System Analysis Capability Development

The OCRWM Integration Branch has the responsibility of assessing the performance of various FWMS configurations and operating strategies from a system-wide perspective. These assessments require analysis of several performance characteristics, such as system cost and system operability. To effectively perform these assessments, a systems analysis capability has been developed that will consistently assess performance for all FWMS components. ORNL is currently engaged in supporting these OCRWM efforts by developing two logistics models: (1) the Waste Stream Analysis Model (WSA) and (2) SOLMOD. These models form integral parts of SIMS. The logistics models and their application as part of the SIMS will be briefly described.

Waste Stream Analysis Model

WSA simulates the movement of nuclear wastes on an annual basis through the major elements of the FWMS based upon a preselected set of operating rules. WSA tracks the history of each assembly from the time of its discharge from the reactor to the time it is emplaced in a repository. The model is designed to select the appropriate shipping cask or waste package based on the radiological and physical characteristics of the waste. WSA characterizes spent fuel inventories, movements, and containerization in terms of fuel type, burnup, age, heat, individual isotopes, and gamma and neutron radiation.

WSA provides the user with a broad spectrum of scenario options. Fuel selection can be based on age (oldest or youngest), heat, form (consolidated or intact), location (in pool or in dry storage), and so forth while maintaining individual reactor acceptance rights. The user can specify the transportation mode between facilities and whether unit trains are utilized. Fuel-handling processing at the repository and the MRS, if included in the system, are also simulated. Fuel may be packaged intact or consolidated at either the MRS or repository. The amount of waste that is placed in fuel packages may be constrained to comply with thermal and radiation limits at the various facilities.

The WSA reporting system is a large collection of standard reports that summarize the time- and location-dependent characteristics of spent fuel and other wastes. Reports are produced at various levels of detail in both tabular and graphical forms.

SOLMOD

SOLMOD permits the detailed analysis of operations and logistics functions by tracking in detail the movement and processing of individual waste packages. This technique permits the determination of average and instantaneous utilization factors of FWMS system elements and equipment and the identification of major pinch-points.

SOLMOD is a discrete-event simulation model that emulates the movement of waste packages through the FWMS from pickup at reactors to emplacement at the repository. The interaction of the waste packages with various process resources (e.g., cranes, unloading bays, or other equipment) is also simulated. SOLMOD can be used to show how different operating schedules and rules, system configurations, and inventories of resources affect the performance of the FWMS by identifying bottlenecks and the utilization of available capacity

or staff. In short, SOLMOD can help planners assess how well the FWMS will work and thus help improve its efficiency.

Model output includes the amount and characteristics of waste stored at selected points within the FWMS and the utilization of resources needed to transport and process the waste. The output routines in SOLMOD have been designed to allow the user to custom design the most appropriate tabular or graphical output for the application.

SIMS

SIMS is an integrated decision-support tool designed for system-wide studies and analysis. As shown in Fig. 2.3, SIMS has a number of components, including models and data bases, which operate either independently or collectively to provide consistent integrated analysis. The six basic parts of SIMS are (1) Operational Scenario and Assumptions Specifications, (2) Systems Descriptions Capability, (3) CDB, (4) WSA, (5) SOLMOD, and (6) System Engineering Cost Analysis Capability.

The ability of SIMS to simulate various FWMS configurations was demonstrated in FY 1989 by

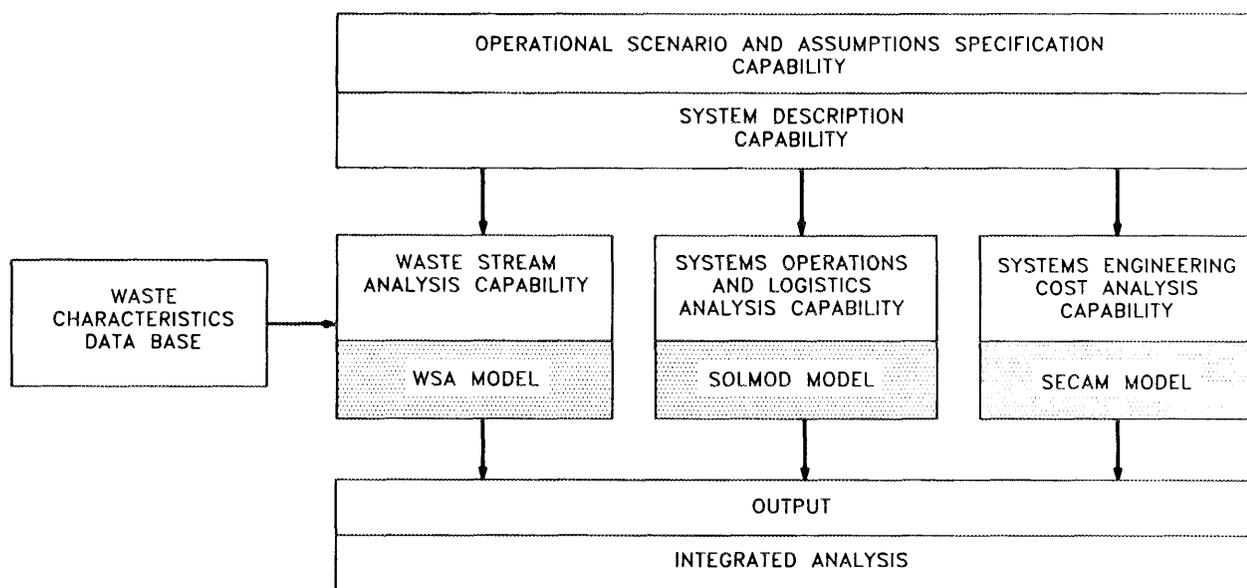


Fig. 2.3. Systems Integration Modeling System.

analyzing three scenarios. Case 1 included an integrated MRS for spent fuel consolidation and storage prior to shipment and emplacement in a repository. In Case 2, the MRS design was modified so that it would store only a portion of the spent fuel as intact assemblies before it is shipped to the repository for containerization and emplacement. The third case contained only a repository that contained and emplaced intact spent fuel assemblies.

A number of evaluation criteria or measures of merit were defined to assess the three cases. In defining the measures of merit it is necessary to determine the type of information and data needed to support the analysis and then choose those particular SIMS outputs that are most relevant. Thus, specific measures of merit are tailored to provide the planners with the pertinent data for making decisions. Two typical measures of merit designed to help assess repository operation and repository emplacement and performance are illustrated in Table 2.2 and Fig. 2.4, respectively.

One of the factors used to assess repository operation was the utilization of the cask unloading cells. Utilization rates, defined as the number of hours a facility is used relative to the number of hours available, provide insight as to the efficiency of the design. Average utilization of the repository cask unloading cells, shown in Table 2.2, varies significantly between cases but for any one case is relatively stable. Rates are high in Case 1, when only four cells are available, raising a question about the ability of the unloading cells to accommodate unscheduled events. The repository design for Case 2, on the other hand, had five unloading bays; the much lower utilization raises a possible question about efficient facility use.

One of the measures of merit selected to illustrate repository emplacement and performance was the integrated heat output over a 350-year period after emplacement. Different fuel selection criteria were used in each of the SIMS demonstration cases: (1) oldest fuel first (OFF); (2) youngest fuel greater than 5 years (YFF5); and

Table 2.2. Average annual cask unloading cell utilization (%)

	Number of cells	Year					
		2008	2009	2010	2011	2012	2013
Case 1	4	84	86	84	85	82	81
Case 2	5	47	43	51	48	57	55

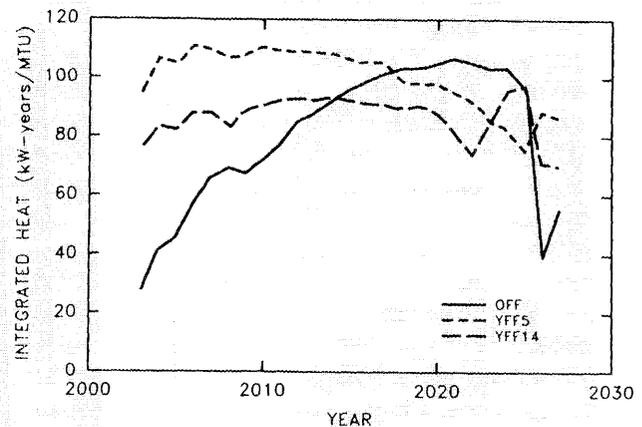


Fig. 2.4. Comparison of integrated heat for 350 years after spent fuel emplacement. OFF = oldest fuel first; YFF5 = youngest fuel greater than 5 years; YFF14 = youngest fuel greater than 14 years.

(3) youngest fuel greater than 14 years (YFF14). Figure 2.4 shows that the YFF14 selection scheme resulted in a more moderate and stable behavior of the thermal characteristics of interest to repository designers. Furthermore, these benefits were achieved with only a 2.5% increase in average heat output and only a 0.8% increase in emplacement area requirements relative to the OFF selection.

Model Assessment, Verification, and Validation

OCRWM, through its many contractors, uses computer models and software programs to assess facility design safety, perform environmental assessments, and support licensing activities. Because of the disparate nature of the programs it supports and the dispersed locations of the participants in the overall system, OCRWM has realized that a program is needed to (1) identify these generic software programs and methodologies; (2) assess their current status with respect to OCRWM and Nuclear Regulatory Commission (NRC) licensing requirements; (3) determine the necessary development and QA needs; and (4) perform the required computer code development, verification, and validation. Chem Tech has called upon several divisions at ORNL (Analytical Chemistry, Computing and Telecommunications,

Environmental and Health Protection, and Health and Safety Research) to assist in this activity.

Beginning in FY 1986, ORNL was directed by OCRWM to manage the "system modeling" program to evaluate the status of computer codes used with the FWMS. In coordination with the Technical Code Coordinating Group (TCCG) and the Spent Fuel Working Group (SFWG), ORNL began the assessment of seven initial code categories: radiation shielding, thermal analysis, criticality, radionuclide inventory (i.e., ORIGEN-type),¹⁵ dispersion/deposition, biosphere transport, and health effects (i.e., dose-to-man). During this reporting period, Chem Tech managed two projects for the OCRWM Office of Systems Integration and Regulations that supported the assessment and validation of computer codes used in the OCRWM program. The first project, entitled "Model Assessment," was funded by the Integration branch, and the second project, entitled "ORIGEN-Type Code Development and Validation," was funded by the Regulatory Compliance branch.

Model Assessment

From a survey of the project offices and contractors, ORNL has compiled and published a compendium of technical computer codes used in support of OCRWM programs.¹⁶ This report contains descriptions of 318 computer codes divided into 22 separate categories. Assessments of the seven initial categories selected by the TCCG have been completed, and reports documenting the code assessment have been prepared for criticality, shielding, thermal analysis, and radionuclide inventory.¹⁷⁻²⁰ A preliminary assessment of atmospheric dispersion, food-chain transport, and dose-to-man computer codes has also been completed.²¹ The Model Assessment project was completed at the end of FY 1989.

ORIGEN-Type Code Development and Validation

For OCRWM to design and construct properly a mined geologic repository for the disposal of SNF and HLW, they and their supportive contractors will need information on the radiological characteristics necessary for design and licensing assessments. The types of data required and their relevance include (1) radionuclide composition (criticality); (2) thermal power (package/cask/facility heat transfer);

(3) radioactivity (performance/risk/safety assessment); (4) photon source (shielding); and (5) neutron source (shielding and criticality). The data could be obtained by analyzing each waste component, but this would be too expensive and time-consuming. A number of computer codes, called radionuclide generation/depletion or source term codes, have been developed for simulating many of the nuclear fuel cycles and for estimating the nuclear compositions and characteristics of these materials. Before one of these codes can be used for licensing applications, it should be verified and validated.

ORNL is conducting each activity (i.e., assessment, verification, validation, and enhancement) under a graded QA approach as specified in NQA-1. During this reporting period, the DOE/HQ-approved QA plan was updated.²²

Verification

Because computer codes are capable of modeling many complex systems, techniques are necessary to document their computational features, capabilities, and accuracy. One excellent method to determine whether a code correctly performs its stated capabilities is a process called verification. If an analytical solution is available for a specific problem, then a direct check of a code's output can be determined. However, in most cases, the systems that are to be modeled are too complicated to lend themselves to analytical solutions. Under these circumstances several codes can be used to model the same problem, and then the resulting outputs can be compared.

A cooperative effort was initiated between Los Alamos National Laboratory and ORNL in order to compare the predictive capabilities of three radionuclide generation/depletion computer codes, CINDER-2, ORIGEN2, and ORIGEN-S.²³⁻²⁵ Of the five cases chosen, two modeled typical BWRs at burnups of 27.5 and 40 GWd/MTU, and two represented typical PWRs at burnups of 33 and 50 GWd/MTU. In the fifth case, identical input data were used for each of the codes to examine the results of decay only and to show differences in nuclear decay constants and decay heat rates. Comparisons were made for several different characteristics (mass, radioactivity, and decay heat rate) for 52 radionuclides and for 9 decay periods ranging from 30 d to 10,000 years. Only fission products and actinides were considered.

Of the more than 21,000 individual comparisons made for the 3 codes (taken 2 at a time), nearly half (45%) agreed to within 1%, and an additional 17% fell within the range of 1 to 5%. Approximately 8% of the comparison results disagreed by more than 30%. However, relatively good agreement was obtained for most of the radionuclides that are expected to contribute the greatest impact on waste disposal. While not without limitations, each of the codes in the comparison appears to produce respectable results.

Very good agreement was observed in the photon source term predicted by ORIGEN2 and ORIGEN-S. The absence of bremsstrahlung radiation in the CINDER-2 calculations resulted in large differences in both the production rates and spectra in comparison with the ORIGEN2 and ORIGEN-S results. A comparison of the CINDER-2 photon production rates with an ORIGEN-S calculation neglecting bremsstrahlung radiation showed good agreement. An additional discrepancy was observed in the photon spectra predicted from the CINDER-2 calculations and has been attributed to the absence of spectral data for ^{144}Pr in those calculations.

The neutron spectra predicted by ORIGEN-S and CINDER-2 were in good agreement for the decay-only case. The differences observed for the other cases reflect the disagreement seen in the actinide inventories (particularly ^{244}Cm).

Validation

The most reliable method to authenticate the computational output from a computer code is to compare it with experimental data. Such a process is known as validation. The first step in this process is to determine if current experimental data are sufficient to provide validation to the level of accuracy required. A literature survey was performed that brought together the experimentally determined LWR spent-fuel data comprising radionuclide composition, decay heat, and photon and neutron generation rates.²⁶

Although substantial fuel composition data were identified, it is difficult to compare these values with results calculated by source term codes because many of the necessary parameters are unavailable for fuel (burnup and power history) and the analytical analyses performed (accuracy and method). Also, the information was not produced under currently required QA procedures.

Current data on decay heat generation rates are reasonably well characterized. Comparisons of measured values with those predicted by ORIGEN agree to within $\pm 10\%$; however, the experimental data exhibit about the same amount of scatter. Also, only a limited number of cooling times and burnups are covered.

Literature citations were found in which one of the earlier versions of ORIGEN2⁹ was used to calculate the intensities of the gamma emissions, and the geometric code QAD-F²⁷ was used to determine the exposure rate.²⁶ Reasonable agreement (a factor of 2) was found between predicted and measured values for locations near the sides of fuel; however, there were large discrepancies (greater than an order of magnitude) when the codes were applied to locations above the fuel. The authors concluded that these differences may be due to deficiencies in the shielding model.

In another literature citation, ORIGEN2 was used to calculate the neutron source strength and the DOT computer code²⁸ was used to calculate the neutron dose rates on the outer surface of a storage cask containing spent fuel. The DOT predictions were found to be a factor of 2 to 4 higher than the measured values. The differences were ascribed to too high a source strength predicted by ORIGEN2.

Experimental

Because all of the information retrieved in the literature survey failed on at least one of the five qualifying standards considered critical to a validation effort, Chem Tech staff members have initiated a joint effort with the Materials Characterization Center (MCC) at Pacific Northwest Laboratory (PNL) to generate analytical data that will be used at ORNL to validate one of the source term codes. In the first phase of a two-phase approach, the analytical techniques were identified and tested, and the sensitivities of the method for each of the important radionuclides were estimated.

For the second phase, five samples, consisting of powdered uranium dioxide from commercial LWRs and representing both BWRs and PWRs, were received from the MCC. The composition values for certain radionuclides have been determined using thermal emission isotope dilution mass spectrometry. Comparison has not yet been made with the output from one of the source term codes (awaiting reactor operational data); however, the results agree favorably with those obtained by

the MCC on an adjacent sample (Figs. 2.5 and 2.6). The schedule for FY 1990 includes the application of spark source mass spectrometry and selected radiochemical analyses.

Enhancement

The assessment of radionuclide inventory codes indicated that the preferred code for use by OCRWM was ORIGEN2 or ORIGEN-S and recommended that enhancements be performed to provide the "best of both."²⁰ One shortcoming identified was the absence of an extended-burnup model for BWRs in the ORIGEN2 code. In response to this, ORNL has revised the LWR models available for the ORIGEN2 computer code. A new model for extended-burnup BWRs [to 40 GWd/metric tons of initial heavy metal (MTIHM)] has been prepared. Using the same computational techniques, the standard-burnup BWR model (to 27.5 GWd/MTIHM) and the standard- and extended-burnup PWR models (to 33 and 50 GWd/MTIHM, respectively) were updated.²⁹

RCRA Studies

Although source, special nuclear, and by-product material are exempt from RCRA, the non-radioactive constituents of by-product materials are

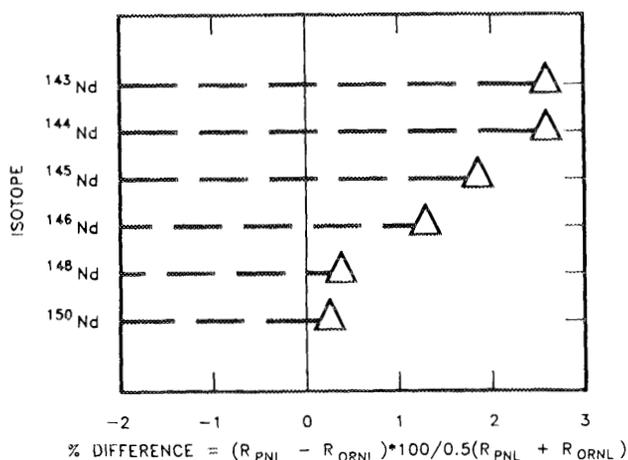


Fig. 2.5. Comparison of neodymium analyses by ORNL and Pacific Northwest Laboratory for nuclides in LWR fuel (ATM-104) in peak-burnup region (thermal-emission mass spectrometry). Note: this sample is adjacent to that shown in Fig. 2.6.

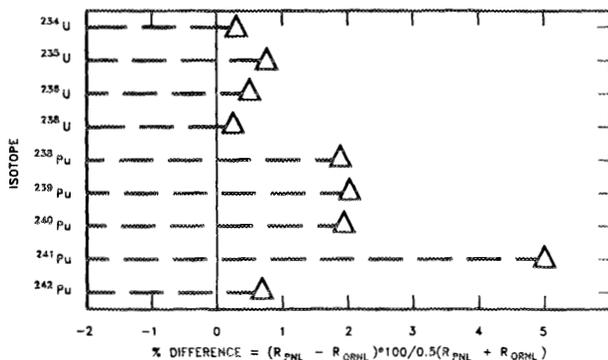


Fig. 2.6. Comparison of actinide analyses by ORNL and Pacific Northwest Laboratory for nuclides in LWR fuel (ATM-104) in peak-burnup region (thermal-emission mass spectrometry). Note: this sample is adjacent to that shown in Fig. 2.5.

not. Facilities that dispose of hazardous wastes are subject to regulation by the Environmental Protection Agency (EPA) under RCRA.² As part of an effort by OCRWM to assess the potential impacts of RCRA on planned OCRWM facilities, the status of non-LWR spent fuel and miscellaneous radioactive wastes under RCRA was evaluated. The wastes considered in this study were the miscellaneous materials that may be disposed of in a repository, including non-LWR power reactor fuels, test, research, and university reactor fuels, wastes from reactor operations, nonfuel assembly hardware, wastes from decommissioning, and wastes from other fuel cycle facilities. Although these materials represent a small fraction of the potential repository wastes, they are comprised of a much greater variety of materials, compositions, and physical forms than LWR fuel and HLW and thus present special problems.

Determining which wastes are hazardous is a key question, because only those wastes that are hazardous are subject to Subtitle C regulations of RCRA.² The Subtitle C program is designed to ensure that hazardous wastes are not mismanaged and creates a cradle-to-grave management system. Under Subtitle C regulation, a solid waste is defined as hazardous if it meets one of the following three conditions: (1) it exhibits one of four characteristics [ignitability, corrosivity, reactivity, or extraction-procedure (EP) toxicity], (2) it is listed, or (3) it is a mixture containing listed wastes. Wastes that meet

one of these three conditions may still be specifically excluded. The characteristic of EP toxicity is determined by the concentrations of certain toxic constituents in the extract of a leaching procedure designed to simulate the leaching actions that occur in landfills. Metals having EP-toxicity regulatory thresholds include As, Ba, Cd, Pb, Hg, Se, and Ag.

A review of the technical literature and readily available data bases was conducted to identify relevant "miscellaneous" radioactive wastes and to collect characterization data, in particular, data on composition, chemical and physical form, leach performance, reactivity, and physical properties. The applicability of RCRA was then assessed. In the non-LWR spent fuel category, Fort St. Vrain fuel is estimated to contain enough cadmium to come under RCRA based on 100% leachability in the EP-toxicity test. The estimated quantity is near the regulatory threshold, and a more detailed estimate may be warranted. However, such an estimate would require that trace impurity data be obtained and that an appropriate ORIGEN2 reactor model be developed. In the greater-than-class-C category, several wastes were identified as potentially RCRA-hazardous. Chromium and lead present in sludge taken from the TMI-2 auxiliary building sump tank are sufficient to exceed their EP-toxicity regulatory thresholds assuming 100% leachability. Alloys of Ag-In-Cd used in LWR control rod assemblies could also be EP-toxic. Analysis of sludge from the Idaho Chemical Processing Plant fuel pool floor indicates that the amount of chromium and lead present could exceed EP-toxicity limits if the sludge were moderately soluble in the leach solution.

Previous studies evaluated waste streams generated by OCRWM facilities and the major incoming waste streams—LWR spent fuel and HLW glass. Both spent fuel and HLW contain RCRA constituents. If these constituents leach sufficiently during an EP-toxicity test, the waste would have the characteristic of EP toxicity and thus be defined by RCRA as hazardous. A draft plan for conducting EPA leach tests on LWR spent fuel and HLW glass was prepared. The test sample population must be complete enough to satisfy the RCRA representativeness criteria. At a minimum, this will require tests with high- and moderate-burnup PWR and BWR spent fuel and tests on HLW glass samples with compositions representative of each major HLW source.

The Siting Record

This record of siting a geologic repository for high-level radioactive wastes describes the many investigations that culminated on December 22, 1987, in the designation of Yucca Mountain, Nevada, as the site to undergo detailed geologic characterization. In addition, this record recounts the important issues and events that have been instrumental in shaping the course of siting over the last three and a half decades. During this long task, which was initiated in 1954, more than 60 regions, areas, or sites involving 9 different rock types have been investigated. This effort became more focused in 1983 with the identification of nine potentially suitable sites for the first repository. From these nine sites, five were subsequently nominated by DOE as suitable for characterization, and then in 1986, as required by the NWPA of 1982,¹ three of these five were recommended to the President as candidates for site characterization. President Reagan approved the recommendation on May 28, 1986, and DOE developed site characterization plans for the three candidate sites, namely Deaf Smith County, Texas; Hanford Site, Washington; and Yucca Mountain, Nevada. As a consequence of the 1987 Amendment to the Nuclear Waste Policy Act, only the latter has been authorized to undergo detailed characterization.³⁰

In general, the siting of a geologic repository for HLW wastes can be divided into three phases. The first phase (1954-1971) involved the search for a site in rock salt in Kansas and in the northeastern states and concluded with the termination of activities at Lyons, Kansas. During the years of the second phase (1972-1981), many rock types and locations in the country were investigated for repository sites. A number of different sets of siting criteria were developed during this time, along with plans to have states and other organizations participate in the siting process. This phase ended with the signing into law of the NWPA.¹ The third phase of siting (1982-1987) covered the procedures and schedules that are prescribed by the Act and by its amendment.³⁰ Included here are the identification of nine potentially suitable sites for the first repository, the subsequent nomination of three sites recommended for characterization, and the decision to proceed with detailed site characterization at only one site, Yucca Mountain in Nevada.

2.2 Waste Management Technology

During this report period, Chem Tech continued to expand and broaden its research, development, and demonstration (RD&D) efforts in waste management both for DOE site-related wastes and other federal agencies' wastes.

These efforts ranged from applied R&D of physical, chemical, and biological treatment methods for specific and generic waste problems to design and engineering support for the development of technical information needed for remedial actions and compliance strategies. This includes test and evaluation demonstrations for new and innovative waste treatment technologies. Significant initiatives are also ongoing in areas such as transportation related to waste management, major program management support, and waste management issues related to the nuclear fuel cycle. In all of these activities, Chem Tech emphasized multidisciplinary and multidivisional approaches, drawing on or providing the appropriate skills, as needed.

2.2.1 DOE Site-Related Waste Technology

Chem Tech's major activities and involvement in DOE site-specific problems are discussed in this section. Some work includes bioremediation technology, wherein major initiatives have begun related primarily to the use of microorganisms for detoxifying and destroying a wide variety of organics. Principal efforts during this report period concentrate on destruction of polychlorinated biphenyls (PCBs) and mixtures of hazardous organic chemicals. Major effort has been expended toward design support for on-site air, water, and solid waste environmental projects and on R&D in support of these environmental projects. The largest of these environmental projects is the Waste Handling and Packaging Plant (WHPP). A multidisciplinary divisional RD&D program has been initiated related to the process development needs of the WHPP. Programs related to D&D of facilities and hazardous waste transportation have also been expanded and developed during this reporting period.

Bioremediation Projects

New technologies for bioremediation of hazardous waste sites are being developed in Chem Tech in collaboration with the Environmental Sciences Division, the University of Tennessee, the Oak Ridge Research Institute, and Oak Ridge Associated Universities. In this section, progress is summarized in the areas of bioremediation of PCB-contaminated soil and trichloroethylene- (TCE)-contaminated groundwater, biosorption of heavy metals, biodegradation of mixtures of hazardous chemicals, and bionitrification of contaminated groundwater.

Bioremediation of PCB-Contaminated Soil

PCB contamination is a significant environmental problem on the ORR, as it is at many government and industrial installations. Oak Ridge sites include the oil landfarm and other sites within the Y-12 burial grounds, portions of the old Z-oil system at Y-12, and at least one location along the floodplain of Bear Creek. There are undoubtedly other, as-yet-unidentified, contaminated sites on the reservation.

Until recently, it was widely believed that PCBs were wholly resistant to biodegradation. It is now known, however, that many of these chlorinated compounds can be broken down biologically, and, in some cases, the carbon can be converted to CO₂. To date, no technology for PCB biodegradation has been successfully demonstrated in actually cleaning up a contaminated site. However, research progress in biodegradation of PCBs and PCB-like compounds has been sufficiently positive to lead most investigators to believe that effective technologies can be developed.

We collected soil and sediment samples from PCB-contaminated sites on the ORR in late CY 1988. The sample identifying numbers and their sites of origin are given in the following:

1. ES-1 (Environmental Sample-1), soil, Y-12 oil landfarm;
2. ES-2, sediment/sludge, Y-12 landfarm oil pond;
3. ES-3, sediment/sludge, Y-12 landfarm oil/sludge pit;
4. ES-4, sediment/sludge, New Hope Pond, Y-12;
5. ES-5, sediment/sludge, New Hope Pond, Y-12;
6. ES-6, sediment, Bear Creek floodplain; and
7. ES-7, sediment, Bear Creek floodplain.

Enrichment cultures were prepared from the environmental samples by inoculating ~0.5 g of soil or sediment into 20 mL of enrichment medium containing only mineral salts and biphenyl. These cultures were incubated anaerobically at 32°C for at least 1 month and monitored visually for growth.

The bacteria from each of the enrichment cultures were screened for their ability to degrade a single PCB congener, 2,2',3,3',5,5'-hexachlorobiphenyl, at 10 ppm. A single congener was chosen instead of an Aroclor mixture because detection of a positive result at this early stage in the research would be simplified if the analysis initially involved only one compound and its possible degradation products. Controls were inoculated with sterile, distilled water. Following a 1-month incubation period, the PCBs in these cultures were extracted with acetone and hexane, and the extracts were analyzed by gas chromatography (GC) to determine if degradation products were present.

Bacterial growth was observed visually during the 1-month period in all of the enrichment cultures prepared. This confirmed that anaerobic bacteria existed in all soils and sediments collected and that they could be encouraged to grow in the medium chosen for use.

When these cultures (assumed to be mixed bacterial cultures) were incubated with 2,2',3,3',5,5'-hexachlorobiphenyl, with glucose as a carbon source, only the ES-3 culture showed peaks during GC analysis, which could be assumed to be degradation products of this congener. In addition to the peak corresponding to the original 2,2',3,3',5,5'-hexachlorobiphenyl, two other prominent peaks were observed. One of these peaks, when compared with standard data on relative retention times for GC analysis of chlorinated biphenyls, corresponded to a pentachlorobiphenyl, and the other to either a penta- or a tetrachlorobiphenyl.

A repeat of the previously described experiment with ES-3 bacteria was set up using both 2,2',3,3',5,5'- and 2,2',4,4',5,5'-hexachlorobiphenyls in separate incubation vessels. After incubation for 3 weeks, GC analyses revealed that the 2,2',3,3',5,5'-congener had again been degraded to two different products (the same as seen in the first experiment), and that the 2,2',4,4',5,5'-congener had also been degraded to at least one new compound (presumably a lower-chlorinated PCB). The exact identities of the new compounds formed are not known and await analysis by GC-mass spectrometry.

We are confident, however, that these new peaks seen after anaerobic incubation represent dechlorination of the added hexachlorobiphenyls.

Control experiments, using bacterial inocula that have been killed in an autoclave before inoculation, have shown that there is no degradation under such circumstances. It remains to be seen whether the particular bacterial species responsible for this degradation can be isolated from the mixed culture from ES-3.

The results obtained to date on anaerobic metabolism of PCBs are encouraging. Further experiments to expand our knowledge of this phenomenon are under way. We plan to test the ES-3 bacteria for dechlorination activity against a range of higher-chlorinated PCB congeners (6 to 10 chlorine atoms). Later studies will use bench-scale soil slurry bioreactors and field tests with lysimeters.

Biological Degradation of TCE in Groundwater

Previous laboratory-scale studies led to the demonstration of the technical feasibility of a methanotrophic bioreactor for the remediation of TCE and 1,2-trans-dichloroethylene (DCE) contamination in groundwater. These studies, conducted by personnel from the Chem Tech and Environmental Sciences Divisions at ORNL, have resulted in one published paper³¹ and an ORNL technical memorandum.³²

Laboratory work has been only minimally funded since the last reporting period. A larger reactor system (0.01 × 1 m) was constructed to test the utility of 1.7-cm-diam × 1.75-cm-long polyethylene Pall rings as an alternative support matrix for the biofilm. These Pall rings appear very satisfactory for supporting biofilm growth and should minimize plugging and channeling within the bioreactor. The system is being checked periodically for the ability of the microbial population to degrade TCE.

A small, pilot-scale system (1 gal/min) was designed at ORNL for construction and testing at the Savannah River Laboratory in Aiken, South Carolina. The unit is currently under construction.

Biotreatment of Mixtures of Hazardous Organics

Bioremediation is presently a viable technology for hazardous waste sites contaminated with a single

chemical or a mixture of similar chemicals such as gasoline. However, many complex mixtures of hazardous organic chemicals found in lagoons, landfill leachates, etc., are presently viewed as poor candidates for bioremediation because of their chemical diversity and complexity. The purpose of this ORNL Director's Fund project is to determine whether these complex mixtures can be microbially degraded using a sequential batch process, wherein a microbial culture adapts to and degrades one or more chemical species at a time over a certain period. This sequential adaptation process, aided by adjustment and control of the process conditions to favor biodegradation of each particular constituent(s) in turn, is believed to be the key to the successful treatment of such complex chemical mixtures. If general guidelines can be developed, they will be useful for evaluating the potential applicability of bioremediation for many hazardous waste sites without the need for exploratory treatment studies at each site.

Five 1-L batch bioreactors (Fig. 2.7) are being operated with cultures growing aerobically on binary mixtures of acetonitrile, benzene, naphthalene, bis(ethylhexyl)phthalate (BEHP), or 1,2,4-trichlorobenzene (TCB) as the sole carbon sources. Two bioreactors are being maintained on mixtures of all five of the compounds. These compounds were chosen as representative organics because there is evidence in the literature that they can be degraded. The medium is supplemented with a mineral salts solution and trace elements. The reactors were originally inoculated with a mixed culture that had been maintained on a variety of hazardous organic compounds. The culture was obtained from L. Grady, Clemson University, Clemson, South Carolina.

A considerable effort has been expended to develop methods to measure the organic compounds. Each has substantially different properties, which complicates both the addition of the compounds to the bioreactors (because of solubility limitations) and the subsequent analyses. A semi-automated method for analyzing the organic compounds is now being used. The compounds are extracted from the aqueous medium with heptane. A more suitable solvent or solvent mixture for extracting acetonitrile is being sought. These extracts are analyzed by gas chromatography using a flame ionization detector and a DB 1+ Megabore column (J&W Scientific, Rancho Cordova, California) for separation.

Although it has been evident, from visual observations of growth, that the microbial populations are using one or more of the organic compounds, we are just recently able to measure the degradation of most of the individual compounds. Table 2.3 shows some preliminary results of analyses of the individual reactors over a 1-week period. Although there is evidence that acetonitrile is being biodegraded, the data have been omitted because we have yet to develop a quantitative extraction procedure for this compound.

It is evident from Table 2.3 that benzene is used in the presence of naphthalene (Reactor 2) and BEHP (Reactor 5). Both of the latter compounds are cointilized in the presence of benzene. There may be some inhibition of benzene degradation by naphthalene. Benzene utilization is also somewhat reduced in Bioreactors 6 and 7, where all compounds are present. Acetonitrile appears to retard the utilization of naphthalene (Reactor 1 vs 2). It is uncertain whether TCB is being used to a significant extent. BEHP is utilized

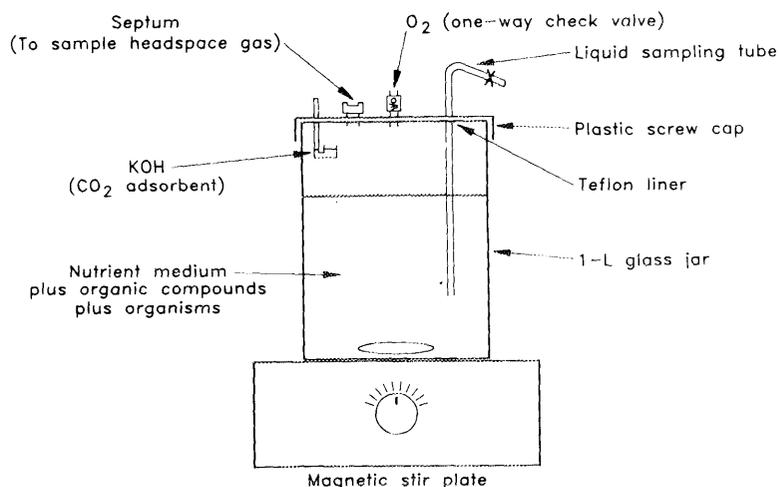


Fig. 2.7. Bioreactor for degradation of hazardous organics.

Table 2.3. Biodegradation of hazardous organic compounds soluble organic concentration* (mg/L).

Day	Reactor									
	1		2		3		4		5	
	Aceton.	Naph.	Benzene	Naph.	Aceton.	TCB	TCB	BEHP	Benzene	BEHP
0	N.A. ^b	13.0	27.1	8.2	N.A.	8.0	5.4	1.2	24.8	12.3
1	N.A.	11.6	23.8	7.2	N.A.	7.1	4.4	N.D. ^c	9.5	1.0
2	N.A.	10.8	13.8	6.0	N.A.	6.4	3.1	3.6 ^d	N.D.	0.5
3	N.A.	11.1	1.1	1.9	N.A.	7.1	3.3	0.6	N.D.	N.D.
4	N.A.	10.4	N.D.	N.D.	N.A.	7.1	3.5	0.5	22.4 ^e	15.6 ^e

Day	6					7				
	Aceton.	Benzene	TCB	Naph.	BEHP	Aceton.	Benzene	TCB	Naph.	BEHP
0	N.A. ^b	24.3	3.8	10.4	27.8	N.A.	26.8	3.2	8.5	14.0
1	N.A.	24.1	5.0	12.8	17.7	N.A.	24.0	2.2	7.6	10.3
2	N.A.	21.0	5.1	13.0	21.7	N.A.	19.9	1.9	7.4	10.0
3	N.A.	18.0	3.4	11.4	6.5	N.A.	18.2	1.9	7.3	7.5
4	N.A.	17.9	2.3	10.0	1.8	N.A.	15.0	1.4	6.4	7.3

*Aceton. = acetonitrile; TCB = 1,2,4-trichlorobenzene; Naph. = naphthalene; BEHP = bis(ethylhexyl)phthalate.
^bN.A. data not available.
^cN.D. = not detected.
^dAdditional organic compound added.

readily, although when all compounds are present (Reactors 6 and 7), there may be some inhibition by one or more of the other organics.

Future studies will continue to be directed at a detailed analysis of what is occurring in these reactors. Included will be a determination of the fate of remaining organics when one (Reactors 1 through 5) or more (Reactors 6 and 7) compounds are depleted, [i.e., does the culture adapt to and use the more recalcitrant carbon source(s)?]. We also plan to obtain pure cultures known to degrade the individual compounds and determine their influence when added to the bioreactors.

Enhanced Biosorption of Heavy Metals

One limitation to the use of microorganisms as biosorbents for removing/isolating heavy metals from waste streams is their limited capacity for certain metals when compared with other sorbents. The goal of this project, funded through the ORNL Exploratory Studies Program, was to isolate yeast cells, which have an increased capacity for uranium. The experiments were based on the previously noted preferential binding of certain yeast cells to garden pea phytohaemagglutinin via specific cell-surface carbohydrates such as mannose and glucose. In earlier studies, when a "normal" population of cells of the yeast *Saccharomyces cerevisiae* NRRL Y-2574 was exposed to garden pea phytohaemagglutinin, approximately the same percentage (30 to 50%) of yeast cells was bound to the agglutinin as those

within the population that accumulated uranium, as shown by electron microscopy.

A population of cells of *S. cerevisiae* was contacted, batchwise, with either agarose bead-immobilized garden pea phytohaemagglutinin or Concanavalin A, a phytohaemagglutinin with a more general binding pattern. Nonbinding cells were washed from the immobilized agglutinins with distilled water. Bound cells, which were microscopically observed on the surface of the agarose beads, were eluted from the immobilized phytohaemagglutinins with a solution of 0.2 M mannose (garden pea) or with 0.2 M glucose (Concanavalin A). The eluted cells were isolated by plating on YM agar. Several colonies were picked at random, subcultured, and then tested for uranium uptake.

The uranium uptake test involved shaking (100 rpm) a suspension of washed, resting cells in a solution of uranyl nitrate (100 mg/L, pH 4.0) at 40°C. The rate of uptake and equilibrium cell capacity (g U/g cells, dry wt) of each isolate was compared to the original parent population.

None of the 32 isolates tested showed an increased capacity for uranium compared with the parent population (i.e., g U/g cells).

Two variants, isolated using garden pea phytohaemagglutinin, initially exhibited a slightly increased rate of uranium uptake compared to the parent population. However, retesting of these variants 2 d later showed that the rates of uranium uptake were the same as those of the parent population.

One variant, isolated using Concanavalin A, had a significantly greater rate of uranium uptake than the parent population in the initial testing. This variant also reverted to the same rate of uptake as the parent population when retested at a later date.

The fact that such variants could be isolated from a general population indicates that potentially they were genetically "different" from other cells in the population. If these variants had unique qualitative or quantitative differences in cell surface components responsible for uranium complexation, the characteristic was unstable and not maintained during subsequent culture storage and/or subculture. Such a genetic reversion is not particularly unusual. It should also be noted that nutritional or other factors associated with cell growth that could affect cell-surface, uranium-complexing moieties could be involved as well. Additional studies were not possible because of the project time constraints.

Biodenitrification of Hanford Groundwater

The DOE Hanford site has groundwater contaminated with nitrate and organic chemicals. To treat this groundwater, PNL is considering a biological denitrification process. For this application, Chem Tech was asked to collaborate with PNL in the evaluation of the ORNL fluidized-bed bioreactor (FBR) process.

Chem Tech provided design and start-up support for a laboratory-scale FBR constructed at PNL. A mixed microbial culture isolated from the Hanford groundwater was used to inoculate the bioreactor. The system was operated at PNL for several months during the summer of 1989. The performance of the FBR was compared with the performance of a suspended-growth (stirred-tank) bioreactor built and operated earlier at PNL. Chem Tech also provided a generic comparison of suspended-growth and fluidized-bed bioreactors to aid in the comparative evaluation of the two types of bioreactors.

From these studies, PNL concluded that the fluidized-bed system would be suitable for treatment of the Hanford groundwater. Further work is scheduled by PNL to assess other operational factors for the two types of bioreactors, which will lead to the selection of a bioreactor for demonstration in the field.

In Situ Remediation of Radionuclides in Groundwater

Surface waters and shallow groundwaters become contaminated with radionuclides when these waters come in contact with contaminated soils and equipment, such as abandoned or leaking tanks and pipes. When these waters accumulate in dry wells, abandoned tanks, and natural drainage areas, they must be treated to meet National Pollution Discharge Elimination System standards before release to public waters. In some cases, an in situ treatment process might be cost effective if appropriate technologies can be identified and/or developed.

Under the auspices of the Remedial Action Program, Chem Tech evaluated various existing technologies for potential in situ removal of radionuclides from aqueous streams. The conventional technologies considered were ion exchange, sorption processes, reverse osmosis, and precipitation/coagulation. The principal criteria for evaluation were:

1. passive operation requiring minimal operator attention,
2. minimal hardware requirements, and
3. compatibility with ambient conditions of the waters to be treated.

None of these conventional technologies appear to be directly applicable to in situ removal of radionuclides. However, a process concept for in situ treatment was formulated based on sorption phenomena. A modest development program was suggested to determine the technical feasibility of the concept.

Waste Processing Design Support

Liquid Low-Level Waste (LLLW) Characterization

A sampling and analysis study is being conducted to determine the characteristics of the LLLW generated at ORNL. The result of this study will provide the environmental assessment data and design data for the WHPP being developed. It will also provide data in support of R&D activities in developing waste management alternatives.

The sampling plan for the first phase covers the collection of samples from the supernate and a "core" of sludge from six of the Melton Valley Storage Tanks (MVSTs) and two of the Bethel Valley (BV) storage tanks. Samples will be analyzed for major chemical constituents, radionuclides, total organic carbon, metals listed as hazardous under RCRA,² and some physical properties. A scoping survey for organic constituents will also be made. Because of limited access into the tanks and the relatively high radiation levels of the waste, only one location in each tank will be sampled. Information from these studies may be used in support of flowsheet development and systems design for the solidification and disposal of these wastes. A full characterization of the wastes under RCRA is planned prior to the time the wastes are processed for disposal.

The planning documents for the first phase have been prepared and the necessary approvals obtained. These documents include the sampling and analysis plan, sampling procedures, task QA plan, safety summary, waste management plan, contingency plan, and environmental ALARA review letter. Sampling equipment has been fabricated. Liquid samples have been collected from six of the MVSTs and sludge samples from four

MVSTs (as of November 1989). Analysis of these samples is in progress.

The second phase is to sample the supernatant liquid in tanks W-29 and W-30 in preparation for a campaign to remove supernate from the tanks and immobilize it in a cement matrix. This work is in support of the LLLW Solidification Project. These tanks were modified at the time of the Emergency Avoidance Solidification Campaign and cannot be accessed by the general procedures for the other tanks. Samples of the supernate will be drawn through the pump module suction leg using the pump module sampler. The analysis plan is similar to the plan for the other tanks except that both W-29 and W-30 will be analyzed for semivolatile and volatile organics. The necessary documents have been prepared and approvals obtained to sample tanks W-29 and W-30. It is expected that this work will begin in the near future. Because of the tank modifications, it is not possible to gain access to sample the sludge in these particular tanks.

Contingency Planning for LLLW Management

The primary sources of LLLW at ORNL include a number of facilities that perform R&D activities involving use and/or production of radioisotopes, and certain systems that support these R&D facilities (Fig. 2.8). The LLLW is collected in underground tanks (see Dilute LLLW Collection, Fig. 2.8) and, in turn, are transported to the evaporator system where the liquid is concentrated by evaporation. The LLLW concentrate is then stored in the concentrate storage tanks (four evaporator service tanks and eight MVSTs) that have a total capacity of 2160 m³ (570,000 gal). The evaporator condensate containing trace amounts of radionuclides is transported to the Process Waste Treatment Plant (PWTP) for further treatment.

No alternative method for LLLW disposal has been implemented since use of hydrofracture was discontinued in 1984; thus, the LLLW concentrate

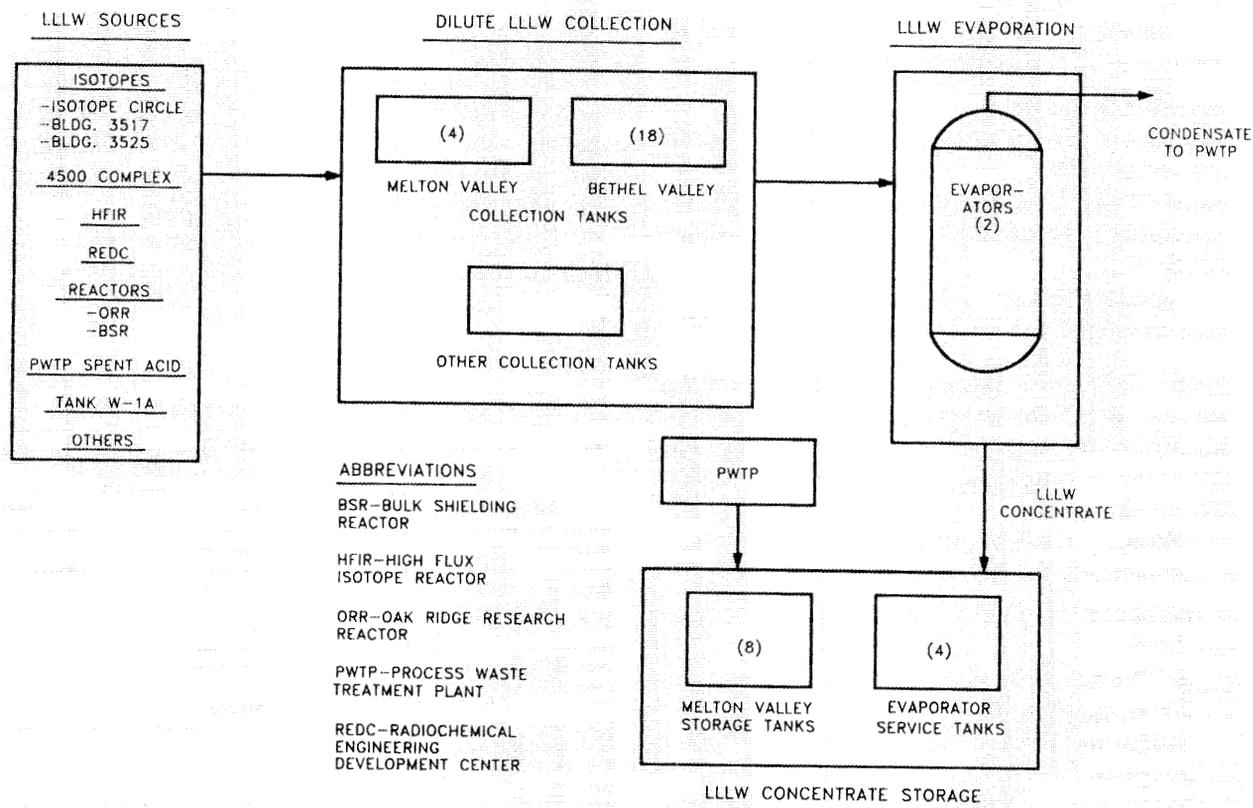


Fig. 2.8. Schematic flow diagram of ORNL LLLW system.

has been accumulating in 12 underground storage tanks. The volume of stored LLLW concentrate, as of March 1989, amounts to ~1676 m³ (442,800 gal), which leaves ~481 m³ (127,200 gal) of free space. The Operational Safety Requirements, however, require that a minimum of 189 m³ (50,000 gal) of storage space must be left unused, leaving only ~292 m³ (77,200 gal) of space available for storage. Further, an additional restriction in the available storage space is imposed by the Operational Flexibility Range [378 to 568 m³ (100,000 to 150,000 gal) of unused storage space] used as the "trigger" point at which some means for reducing the volume of LLLW concentrate must be implemented.

In view of the previously mentioned situation, two methods (solidification campaign and in-tank evaporation) are scheduled for implementation as interim solutions to the storage space problems. However, despite such efforts, there still is a possibility that the storage space may be depleted sooner than expected because of factors that are beyond our control. Thus, the goal of this task is to develop specific plans of action to be implemented should the available storage space for the LLLW concentrate be depleted.

A number of conditions that would decrease the available storage space were considered:

1. normal operation;
2. excessive, unexpected generation of LLLW concentrate;
3. loss of one LLLW concentrate storage tank;
4. excessive, unexpected generation of dilute LLLW;
5. evaporator failure; and
6. heavy rainfall.

The first three scenarios affect the storage situation after the evaporator (i.e., storage of concentrated waste), and the last three scenarios affect the storage of dilute LLLW before evaporation.

Following are the options that were considered for implementation:

1. use of inactive gunite tanks for storage of dilute LLLW,
2. use of tank vaults in MVST area for storage of LLLW concentrate,
3. installation of new storage tanks for dilute LLLW or LLLW concentrate,
4. shutdown or curtailing the operation of selected LLLW generators,
5. additional solidification campaigns, and

6. use of a mobile ion-exchange unit to remove radionuclides from the LLLW.

These options were evaluated as solutions for each of the scenarios. Among the major factors considered in the evaluation were the time and the costs required for implementation. The normal operation scenario represents not only the normally scheduled operations (including the restart of HFIR) but also nonroutine D&D activities (e.g., for inactive tanks), implementation of the in-tank evaporation technology, and one solidification campaign.

The results of the evaluation are summarized in Table 2.4. The option to use a mobile ion-exchange unit was excluded from further consideration primarily because of the long lead time and high costs required for implementation, including those for safety and regulatory documentation.

Gaseous Waste System Upgrade

Support is being provided to the Environmental Projects program through management of capital projects designed to upgrade the radioactive air-emission systems at ORNL. The goals of the upgrade program are to ensure that the air-emission systems are in compliance with applicable regulations, to improve performance, to ensure long-term reliable service, and to upgrade the monitoring systems. Projects that are under way or scheduled are described in the following paragraphs.

1. A project to install two additional sets of high-efficiency particulate air (HEPA) filters in the

Table 2.4. Evaluation of contingency options for given scenarios

Scenario	Contingency options selected	
	Options	Priority
Normal operation	Solidification campaign	1
Excessive generation of LLLW concentrate	Solidification campaign	1
	Use of tank vaults	2
	Construct new tanks	3
Loss of LLLW conc. storage tanks	Solidification campaign	1
	Use of tank vaults	2
	Construct new tanks	3
Excessive generation of dilute LLLW or heavy rainfall	None required	
Evaporator failure	Shutdown selected generators	1
	Use inactive Gunite tanks	2

cell ventilation system for Building 3517 is nearing completion. This addition will bring the system into compliance with DOE requirements for double HEPA filtration with greater than 99.95% removal efficiency.

2. Design is under way for an enclosure over the underground filter pits that serve HFIR. The enclosure will eliminate rainwater inleakage and provide an enclosed area for filter changes.

3. The ventilation equipment at the 3039 stack area is being upgraded to replace obsolete steam turbines that drive cell ventilation fans and to provide a new pressure-control system for the process off-gas scrubber system.

4. The monitoring systems for the 2026, 7512, 3020, and 6010 stacks are being upgraded to provide real-time monitoring and sampling of the radioactive effluents and to improve data transmission and collection.

5. Installation of four ORNL plant perimeter air monitoring stations is under way to replace obsolete stations. The new stations will detect rapid changes of radioactivity in the environment and will provide data needed to assess radiation exposures and demonstrate compliance with national Emission Standards for Hazardous Air Pollutants.

6. Future projects include the upgrade and repair of underground ventilation ducts to the 3039 stack to eliminate water inleakage.

LLLW and Process Waste System (PWS) Upgrade

Environmental projects under the LLLW and PWS upgrade program are directed toward establishing the improvements needed for continued protection of the environment and the health and safety of on-site workers and the public and for compliance with current and future environmental regulations. They have also involved improving or developing waste collection, treatment, storage, and disposal capabilities for all ORNL-generated waste and effluent.

Chem Tech continues to provide technical management and other support for a number of the ORNL water pollution control projects. These projects are directed at collection, treatment, and disposal of a wide variety of liquid wastes generated by the diverse activities at ORNL. These projects can be divided into three congressional line-item projects and general plant projects (GPPs).

The three line-item projects are:

1. The BV LLLW Collection-and-Transfer (CAT) System Upgrade,
2. The Isotopes Area (IA) low-level waste- (LLW-) CAT System Upgrade, and
3. The Melton Valley (MV) LLW-CAT System Upgrade.

The BV LLW-CAT, the IA LLW-CAT, and the MV LLW-CAT projects are currently planned to upgrade the aged ORNL LLW-CAT systems in both the BV and the MV areas. Each project is specifically concerned with upgrade of a particular portion of the overall ORNL LLW system.

These three projects have a number of common features, and management of the projects is being coordinated to take advantage of these commonalities. The three projects will provide an overall LLW system that has used the best available technology for satisfying regulatory requirements.

New underground pipelines, serving major source facilities on the ORNL LLW system, are to be provided. These pipelines will be completely doubly contained and will be cathodically protected against corrosion. The annulus of the doubly contained pipelines will be pressurized with nitrogen and monitored for leak detection in both the primary inner pipe and the outer secondary pipe (i.e., a leak in either will alarm).

The BV LLW-CAT line item is an FY 1988 project with a total estimated cost (TEC) of ~\$35 million. The validated IA LLW-CAT line item is an FY 1991 project with a TEC of ~\$40 million. The proposed MV LLW-CAT line item is an FY 1992 project with a TEC of ~\$25 million. (The TEC is a preconceptual design estimate and is low.)

New Monitoring and Control Stations (MCSs), which are associated with the source facilities to be served by the project systems, will be provided. The MCSs will consist of below-grade, vaulted collection tanks; monitoring instrumentation, including indication of pH, temperature, pressure, and tank level; a caustic addition system; and an above-grade control room that contains a sampling system. Generally, waste will flow from the source facility to the MCS tank by gravity. At the MCS, tank contents will be monitored and sampled, and the pH will be adjusted as necessary. Periodically, tank contents will be discharged to the downstream LLW system by means of steam-jet ejectors.

The BV LLW-CAT project will provide underground piped services to five facilities and a

new central facility for receiving and discharging liquid waste that is transported in small volumes in bottles and in larger volumes using a tank truck. The facility will have diked floors, hoods, and necessary ventilation and handling equipment to ensure operator safety and environmental compliance. The bottles and truck used for this effort will meet ORNL and DOE regulations for the transportation of regulated substances. The upgrade project includes ~2 km of doubly contained, stainless-steel pipeline and five MCSs.

The underground piped service to be provided by the IA LLW-CAT project interfaces with six facilities, which generate and process 9.46 m³ (2500 gal) of radioactive liquid waste per month, and will also have the capacity to service two facilities where wastes will be collected and bottled for transport to treatment facilities in trucks. The upgrade project includes ~2 km of doubly contained, stainless-steel pipeline, three 0.95 m³ (250-gal) stainless-steel intermediate collection tanks, and one MCS.

The MV LLW-CAT project will provide underground piped service to three facilities that generate and process ~10 m³ (2640 gal) of radioactive liquid waste per month. The upgrade project includes a pipeline that will connect the MV area with the treatment facilities in the BV; two MCSs will be provided.

The GPPs are directed toward connection of two ORNL facilities to the BV LLW-CAT system. The first project, the Bulk Shielding Reactor (BSR) Drain Segregation GPP, with a TEC of <\$100,000, will remove all process drainlines from the storm-sewer system. The second project, the BSR/Oak Ridge Research Reactor LLW-CAT System Upgrade, with an estimated cost of \$1.1 million, will provide a system for collection of the LLW from BSR operations and Oak Ridge Research Reactor decommissioning activities.

Transuranic (TRU) Waste Inventory Coordination

The TRU Waste and Integration Program for planning, integration, and technical development for TRU waste management is centered in the DOE's Albuquerque Operations Office. The program's goal is to implement permanent disposal of TRU wastes from all defense waste locations. The Waste Isolation Pilot Plant (WIPP), located in southeastern New Mexico, is under construction to serve as this central, permanent geologic depository.

Retrievable emplacement tests are now being conducted at the site to demonstrate that WIPP can be approved as a permanent disposal facility for TRU waste. In support of this activity, Environmental and Health Protection Division at ORNL has the primary responsibility of preparing certification documentation, design, and construction of a WHPP, and shipment of the packaged waste to WIPP. Chem Tech supported the effort during this reporting period by upgrading ORNL's TRU waste records and providing input to the WIPP Operation's Office for the Integrated Data Base Report.³³

Early records and files of ORNL's solid waste information management system (SWIMS, an ORNL computer data base that is updated daily), were searched to locate misplaced records and documentation of earlier burials. This search resulted in many packages being switched from the remote-handled (RH) TRU category (having >200 mR/h at the surface) to the contact-handled (CH) TRU waste category (with <200 mR/h at the surface). Additionally, documentation and research resulted in 22 RH-TRU casks being moved from the earlier records of the SWIMS into the official, active data base.

A search was run on the SWIMS to identify CH-TRU waste packages that were listed as containing excessively high radioactivities, resulting in isolation of 70 packages out of more than 2400. However, this small number of packages skews the CH-TRU waste records so much that it causes ORNL's CH-TRU waste to appear more radioactive than that at any other DOE site and even more radioactive than our casked RH-TRU waste. Corrections were made to these records. Until actual assays are performed on the CH-TRU waste packages, little can be done to correct the inconsistencies for CH-TRU waste in the SWIMS. A step in this direction was taken by providing the Stoller Corporation with information that will help them to evaluate the user friendliness of the SWIMS.

The 1989 Integrated Data Base (IDB) Report³³ update was prepared and submitted to WIPP on schedule. The volume of stored RH-TRU waste (including 6.8 m³ of special waste stored in SS-wells) is 1,311 m³ with a reported total radioactivity of 112,963 Ci (including the special-case waste's 66,000 Ci). The volume of stored CH-TRU waste is 625 m³, with a reported total radioactivity of 56,189 Ci. As of December 31, 1989, the stored TRU waste consisted of 353 RH-TRU casks, 2,441 CH-TRU

waste drums, and 39 boxes containing CH-TRU wastes. The projected generation rates are 6 RH-TRU casks per year, 75 drums per year, and 4 boxes per year, respectively.

A document,³⁴ based mainly on SWIMS' input but supplemented greatly with generator information and guidance, was published to lend support to the WIPP certification effort. This document will also facilitate the structuring and layout of the WHPP by providing more detail on the contents of the individual stored casks. Its PC-based data base was tabulated in FrameWork III and is compatible with dBASE.

ORNL Liquid and Gaseous Treatment

The Waste Management and Environmental Compliance programs were established at ORNL to provide the Laboratory with the capability for complying with existing and future environmental regulations and improved methods of treating hazardous and radioactive waste at the plant. These programs are directed toward identifying the improvements needed for continued protection of the environment and the health and safety of on-site workers and the general public. The ORNL Waste Management Program Office (WMPO) is responsible for the development, maintenance, and implementation of effective management strategies for handling ORNL's waste streams, while Environmental Projects is responsible for managing capital projects needed to install system upgrades.

ORNL Gaseous and Liquid Waste Management

Chem Tech provides programmatic support for the WMPO in two major areas—management of Air Pollution Control Projects and management of Water Pollution Control Projects. The purpose of the program is to provide the administrative and programmatic support for technical programs needed to define process modifications or capital projects for improving the air and water pollution control systems. The program also includes the development of waste treatment strategies and long-range plans for upgrading ORNL's waste treatment systems to meet regulations and optimize the types and amounts of waste generated for safe permanent disposal. The gaseous waste projects included

implementing a preventive maintenance program for fans and motors on the main stacks, which discharge radioactive emissions; remotely inspecting ventilation ducts to assess their structural integrity; and testing the uniformity of HEPA filter bank flows. The technical projects described in the following sections are in support of the long-term strategies for improved treatment and disposal of liquid wastes.

ORNL Liquid Waste Systems Analysis

A model of the overall ORNL liquid waste system is being developed to determine the effects that each waste generator and waste treatment step has on the composition and volume of the final waste form produced for permanent disposal. The model is being used to determine where improvements should be made in the liquid waste system. A comprehensive survey of the liquid waste generators was performed to determine the amount and type of waste produced at ORNL and to determine how these streams are being routed for treatment. This information is being coupled with a technical systems analysis to develop strategies for effectively improving the ORNL waste systems.

The generator survey and systems analysis identified three simple upgrades that could reduce the ORNL liquid waste system's production of solid LLW by 33% and the LLLW concentrate by 45%. In addition, the system analysis showed that only three current operations at ORNL—the PWTP, the Fission Products Development Laboratory (FPDL), and the Radiochemical Engineering Development Center (REDC)—significantly impact the radionuclide, nitrate, and phosphate contents and the amount of dissolved solids in the LLLW concentrate. Characterization and treatability studies are being performed to support implementation of capital projects to reduce waste generation rates in these areas by (1) treatment at the generation site, (2) modification of the processes generating the wastes, and/or (3) improved operations at the centralized facilities.

PWTP Upgrades

Process development studies are under way to support upgrades of the PWTP to increase the plant's capacity, improve the decontamination capabilities of the facility, and reduce the amount of secondary waste generated by the system. Recent

evaluations indicate that the installation of a pH adjustment system and an extra holding tank in the existing PWTP evaporator loop will reduce the LLLW concentrate by 4000 gal/year. Improved zeolite loading facilities will be added to reduce worker exposure to potentially harmful zeolite dust and to improve the overall safety of the operation. Other proposed improvements include the use of an excess clarifier at the Nonradiological Wastewater Treatment Plant (NRWTP) in the PWTP flowsheet to increase plant capacity, and disposable zeolite ion-exchange units to increase ^{137}Cs decontamination capabilities. The ultimate objective is to replace the existing plant with a zeolite ion-exchange system that will remove both ^{137}Cs and ^{90}Sr and produce only solid LLW secondary waste. This process will reduce the secondary waste generation rate by one-third and will eliminate all LLLW generated by the process waste system.

The strategy for installing the zeolite system was developed in conjunction with Environmental Projects. It will allow enhanced metals-removal capabilities, implementation of improved radioactive treatment capabilities 5 years ahead of schedule, reduction of system improvement costs by \$3 million, and reduction of operating costs for the two facilities. Development studies are under way to design the zeolite system. A mathematical model is being developed to assist in design of the zeolite columns and to set plant operating conditions to maximize usage of the zeolite material. Pretreatment requirements and hardware design for the columns are also being investigated.

NRWTP Support Studies

A study was undertaken to determine how ^{60}Co should be removed from HFIR process wastewater to meet the NRWTP acceptance criteria. Analysis of the HFIR system indicates that the addition of a holding tank and modification of the ion-exchange column regeneration procedures would allow recycling of the process wastewater. This will eliminate the problem process wastewater without increasing the amount of LLLW generated by the facility.

On-line radiation and metals monitors have been installed at various manholes throughout the ORNL complex and will be used to control the routing of waste streams to the appropriate treatment plants. A study was undertaken to determine if the monitors could be used to control the routing of wastewater, based on readings from these instru-

ments, and to develop alternative methods if the existing equipment was inadequate. The on-line radiation monitors were calibrated, and recommendations were made to set the alarm levels so that there would be few, if any, false-positive diversions. At these levels, the gamma coils will divert at a concentration below the Derived Concentration Guide (DCG) value for discharge of ^{137}Cs (111 Bq/L) in DOE Draft Order 5400.XX,³⁵ but neither the Phoswich nor the baggie beta monitors will detect the ^{90}Sr DCG value of 37 Bq/L. Routine sampling will be required to detect long-term low-level ^{90}Sr contamination in the streams, while the monitors will detect and divert any large spills.

An On-line Voltametric Wastewater Analyzer was developed for measuring trace quantities of heavy metals in wastewater. The monitors had excellent analytical accuracy but required frequent maintenance and well-trained operators. Therefore, diversion based on pH was considered as a simple alternative. A pH-based sampling system has shown that there is a high correlation between pH and metals contamination for the wastewater from the BV plant area. It was recommended that pH be used as the diversion criteria for wastewater. Any wastewater with a pH <6.0 or >9.0 should be sent for metals removal. Based on the results of 29 weeks of sampling, it is expected that ~9500 gal/week of wastewater from the BV complex will be collected for treatment. None of the wastewater collected from the MV complex required treatment for metals.

Continuous Countercurrent Ion Exchange

The calcium and magnesium in contaminated waste streams compete with strontium in most decontamination processes, making it difficult to efficiently remove strontium from wastewaters. Continuous countercurrent ion exchange (CCIX) has been used in some applications to selectively remove a preferred component from multi-component streams when fixed-bed operations resulted in extremely low separations. A 3-gal/min mobile pilot-scale CCIX system was operated to evaluate selective removal of strontium from ORNL process wastewater and contaminated groundwater. Results from more than 700 h of operation indicate that the CCIX can remove strontium from ORNL process wastewater to meet discharge requirements. The tests also indicate that the performance will be affected by changes in feed stream compositions and

flow rates. Additional long-term operating data and development of treatment methods for secondary waste are needed to determine if the CCIX should be considered for wastewater treatment.

Chem Tech Waste Minimization

A comprehensive waste-reduction program has been initiated in Chem Tech, with the immediate goal of providing an understanding of the waste streams produced in the division and identifying potential waste-reduction projects. The next phase of the project will be to initiate waste-reduction programs identified in the first phase of the program.

A report has been completed detailing the volumes and characteristics of the major waste streams produced in the division. Two areas, the REDC and the IA, are responsible for the majority of the waste produced by Chem Tech. Possible waste-reduction projects have been recommended and include the following:

1. creating a Chem Tech waste characteristic data base;
2. treating the LLLW at the sources, the REDC and FPDL;
3. identifying process waste generators in the IA; and
4. eliminating gaseous waste service from Chem Tech buildings, where practical.

WHPP Technology Development

The WHPP is a proposed \$245 million FY 1993 line-item project. The facility will prepare RH-TRU liquid and solid waste for shipment to WIPP in Carlsbad, New Mexico. A simplified flowsheet of the preliminary conceptual design for slurry processing is shown in Fig. 2.9.³⁶ Engineering development has been directed toward improving the slurry processing flowsheet. Slurry processing includes sludge mobilization and conventional and microwave evaporation. A goal of the WHPP

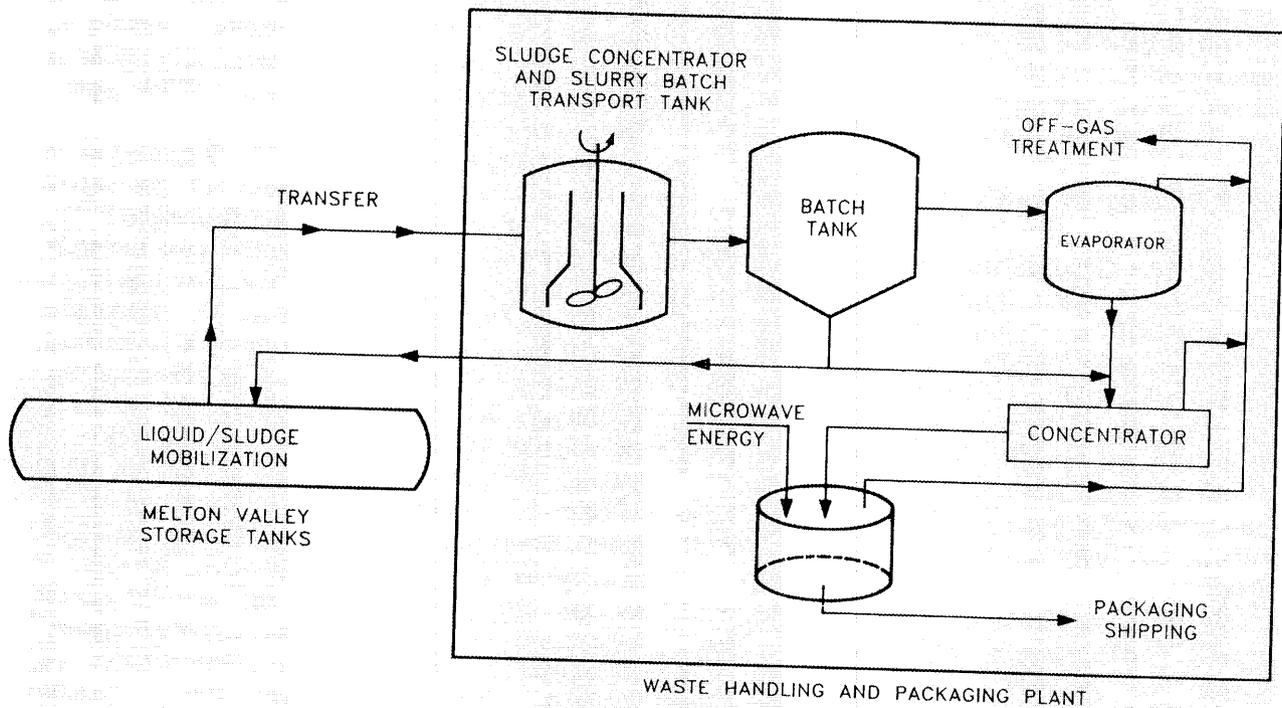


Fig. 2.9. WHPP slurry processing conceptual flow diagram as included in the conceptual design report.

technology development team is to develop a flexible slurry treatment process so that inclusion of long-term LLLW treatment is feasible.

Long-Term Development of LLLW Treatment Options

The ORNL LLLW treatment strategy stresses the need for centralized treatment facilities to process the various LLLW streams generated at the Laboratory. The goal of the long-term processing task is to identify processes that will render the bulk of the ORNL-contaminated liquid waste acceptable for final disposal or safe release to the environment.

Decontamination of the LLLW and the supernate in the MVSTs is an important step in the long-term process selection. Significant progress has been made in selecting a process for the removal of ^{137}Cs from these waste streams. Ferrocyanide ion exchangers have been tested using actual MVST supernate and have removed >99% of the ^{137}Cs present in these solutions. Further testing of ferrocyanide ion exchangers in conjunction with ultrafiltration is planned.

Progress is also being made in identifying resins to remove ^{90}Sr from the MVST supernate solutions. In particular, sodium titanate has generated decontamination factors >100 for ^{90}Sr removal. Further work is planned using organic ion-exchange resins for ^{90}Sr removal from the MVST supernate.

WHPP Development Facility (WDF)

The WDF is being constructed in Building 2528 at ORNL to provide engineering data needed for the design of the slurry processing systems that will be installed in the WHPP. The WDF will be used to select and evaluate the methods and equipment that are suitable for mobilization, transport, and solidification of the TRU sludge contained in the MVSTs. Tests will be conducted using a nonradioactive surrogate sludge that simulates the chemical and physical properties of the MVST sludge.

The WDF will include a 25,000-gal mockup of the MVST and supporting equipment such as tanks, pumps, and instrumentation for testing and evaluation of methods of sludge mobilization. The system initially will be set up for testing fixed-point jets (water or supernate) and jets attached to a scavenging robot for sluicing the sludge from the

tank. The WDF will also include base-line equipment (tanks, pumps, and condensers) for development and testing of processes for concentration and drying of the sludge and supernate. Processes that will be evaluated for sludge solidification include combinations of thin-film evaporation, microwave drying, and extrusion. The facility will also be used for development and evaluation of instrumentation critical to the certification of the final waste product.

Process flowsheets have been developed; preparation of specifications for instrumentation has been initiated; the 25,000-gal model tank has been installed; and installation of utilities such as water, steam, and electricity has begun. Equipment requiring long delivery time has been ordered.

Sludge Mobilization Development

A common problem of long-term storage of nonacidified liquid wastes is the formation of sludges³⁷ of high curie content. This is true of the ORNL MVSTs, which have all been found to contain sludges of various depths, consistencies, and compositions. The objectives of this development task are threefold: (1) to remove the waste sludges from the tanks, (2) to transport them to the proposed WHPP, and (3) to prepare a mixture of sludge and supernate there that will yield a solidified product that meets the WIPP waste-acceptance criteria.

The MVSTs are 18.3-m- (60-ft-) long horizontal cylinders, 3.7 m (12 ft) in diameter, with one access manhole 4.6 m (15 ft) from the tank center. The placement of this manhole, geometry of the tank, and obstructions within the tank pose significant challenges to sludge mobilization. Two techniques have been under study in a one-sixth linear-scale model of the MVSTs to mobilize and remove sludge in this geometry. The first technique involves the introduction of a high-pressure solid stream spray nozzle into the tank through the manway under conditions in which the tank is nearly empty. The nozzle is manipulated to direct the slurry of liquid and solids toward the manhole area where it is pumped from the tank. This was the method used successfully in the Guniting Tank Cleaning Program,³⁸ although the shape of the Guniting tanks was different. The second technique is more developmental and involves the use of an industrially available robot that travels along the bottom of the tank, pulling its own suction line and

carrying a very high-pressure water jet to break apart extremely hard sludge. Both techniques will be tested in the WDF. The sludge mobilization flowsheet of the WDF is presented in Fig. 2.10.

A highly instrumented experimental pipe loop has been built to study the flow of simulated slurries, which are formulated based on data from samples of MVST sludges. The simulated slurries are formulated based on the hindered settling velocities and rheological properties of the sludges. These properties, in turn, depend on the state of particle aggregation, size, and density. The simulated slurries are designed to be more difficult to mobilize and transport than the actual sludges. A Kingston fly ash and water slurry has been found to be slightly more rapidly settling and, therefore, a good surrogate for slurry transport studies of the soft MVST, W-26, sludge (the only MVST sample for which an analysis has been completed). Pipe flow data, head loss through fittings, and minimum transport velocity data have been obtained. Instrumentation is being evaluated for its sensitivity to process parameters such as density. A typical plot

of pressure drop vs flow rate with concentration as a parameter is shown in Fig. 2.11. Preliminary results are consistent with Thomas's observation³⁹ that slurries exhibiting low Bingham plastic-yield stresses ($<240 \text{ dyn/cm}^2$) can be correlated using the Moody plot of Fanning friction factors.

Conventional Evaporation Development

The Preliminary Conceptual Design Report³⁶ for WHPP calls for evaporation of the water from the sludge that is mobilized from the MVSTs. The solidified sludge will be transported to lined 55-gal drums before being transferred to WIPP. Two technologies are being considered for concentration of the sludge from the MVSTs—conventional evaporation and microwave energy. Development efforts will determine if the WHPP will contain both an evaporator and a microwave system or if the process will rely on only one technology. The evaporation product will be a stable waste form that meets the waste acceptance criteria for WIPP.

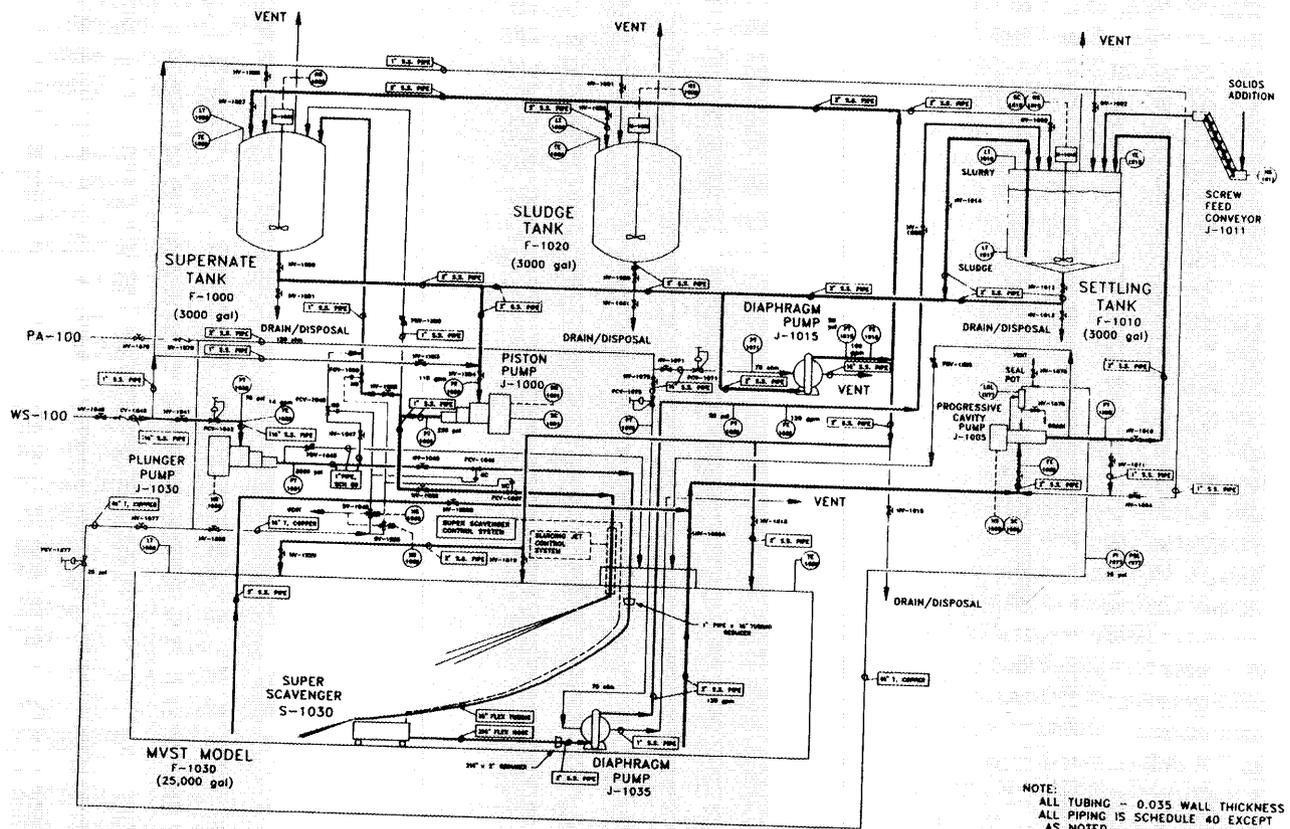


Fig. 2.10. Sludge mobilization flowsheet to be installed at the WDF.

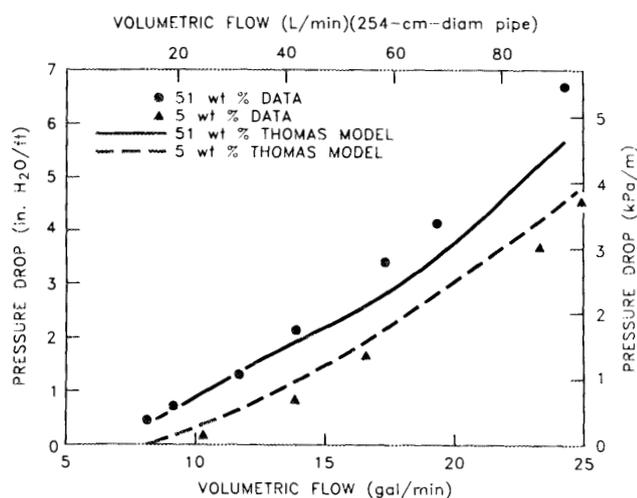


Fig. 2.11. Comparison of Thomas model predictions with slurry flow data.

The primary objectives of the conventional evaporation development task are to determine the feasibility of using conventional evaporation to concentrate the MVST sludges and to evaluate the different evaporator options. To accomplish this, a surrogate feed has been used for vendor demonstration tests. These tests have used facilities at various manufacturers to determine the applicability of their equipment for concentrating the surrogate waste. Those items that are most promising will be evaluated in pilot tests at the WDF. To date, vendor demonstration tests have been conducted using a vertical thin-film evaporator, a horizontal thin-film evaporator, and an extruder.

The agitated thin-film evaporator uses a rotor to spread a turbulent thin layer of solution over the heated surface. Vendor tests of both vertical and horizontal units indicate that the thin-film evaporators can be used to produce either a thick paste or a powder. Use of a thin-film evaporator may eliminate the need to evaporate the liquid and solid streams separately and may replace the kettle evaporator specified in the Preliminary Conceptual Design Report.³⁶ Thin-film evaporators will be further evaluated in the WDF.

A twin-screw extruder/evaporator was evaluated as a process candidate for drying and melting MVST waste to meet waste-form criteria. Tests were structured to evaluate the versatility of the extruder to (1) dehydrate the surrogate waste and produce a powdered/granular product that was melted in a second extruder, and (2) dehydrate and melt the slurry in a single step. The extruder proved capable

of operating in both modes. In each mode, the process yielded a product that met the waste-form criteria. The primary disadvantages of the extruder are the cost and complexity of operation. A decision has not been made whether to include or exclude the extruder from the WDF testing.

The conventional evaporation flowsheet of the WDF is presented in Fig. 2.12. All major components for this system have been ordered, and construction has been initiated.

Microwave Process Development

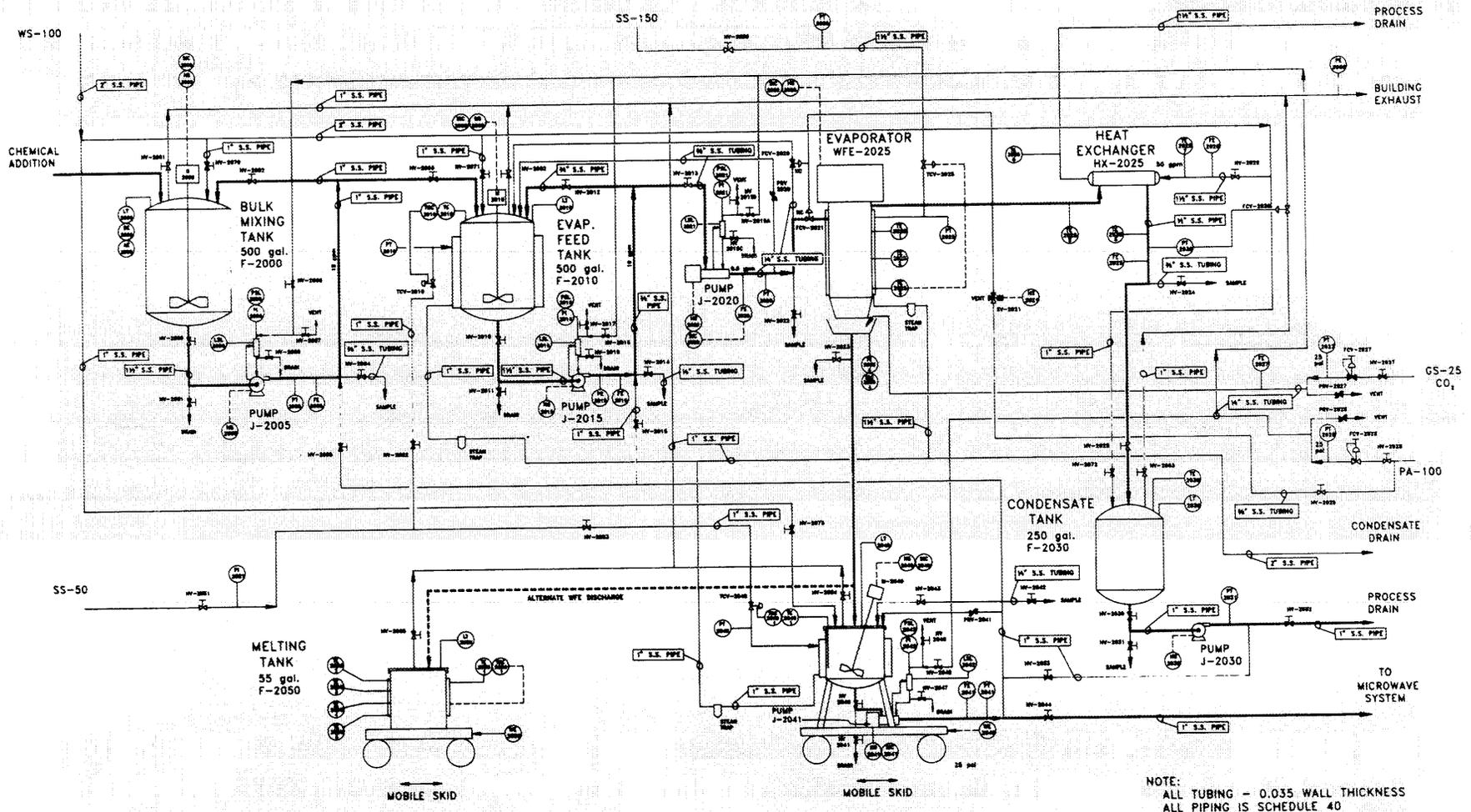
Microwave processing of surrogate RH-TRU sludges and liquids is being investigated as an alternative to conventional evaporation technology. Results of bench-scale tests using a 1-kW, 2450-MHz oven showed that the surrogate can be dried and then melted in a single process. An acoustic emission feedback control was developed to regulate the amount of boiling in the surrogate. A one-third-scale applicator (6 kW at 2450 MHz) for in-can evaporation and melting has been fabricated. The applicator will use a microwave field configuration that results in improved dependability. Both batch and continuous processing modes will be investigated, and a wide range of process controls in a PC-based data acquisition and control system will be used. The one-third-scale microwave system flowsheet is shown in Fig. 2.13.

A full-scale applicator capable of processing a 55-gal drum of surrogate has been designed and will operate at 60 kW and 915 MHz in the WDF. Data from the one-third-scale experiment will be used to optimize the design of the full-scale equipment.

Waste Characterization and Product Certification

The LLLW contained in the MVSTs is composed of an alkaline, sodium nitrate-based waste with a TRU sludge on the bottom of each tank. Plans are to evaporate a blend of the sludge and nitrate-based aqueous waste by either a low- or high-temperature process to form a salt monolith for shipment to WIPP. In preparation for this disposal, laboratory tests have been run on salt products generated from either the low- or high-temperature processes to observe product properties for WIPP certification.

Laboratory testing has been performed on salt products developed from a nonradiological surrogate



NOTE:
 ALL TUBING - 0.035 WALL THICKNESS
 ALL PIPING IS SCHEDULE 40

Fig. 2.12. Flowsheet of conventional evaporation system to be installed at the WDF.

waste, which represents the contents of the MVSTs. An extensive sampling campaign of tanks is in progress, and the surrogate used in testing has been formulated based on chemical, radiological, and physical waste characteristics of tank W-26. As more waste characterization data are available, a number of different surrogates will be used in product certification testing to observe the effects of various significant differences.

Laboratory and field processing scenarios include evaporating at 110 to 125°C to produce a salt monolith or evaporating and melting the salt at 310 to 340°C. Salt products developed by both methods have been studied to ensure that the product meets WIPP waste acceptance criteria and does not exhibit any undesirable properties. The primary tests used to study salt product properties are shown in Table 2.5.

Results to date show that both high- and low-temperature salt products are extremely hygroscopic due primarily to the presence of sodium hydroxide and calcium or magnesium chloride. The melted product presents a much lower salt surface area to humid air and, therefore, absorbs water at a lower rate when the residual water content is the same in both types of products. The melted salt product also presents a better opportunity for a higher volume reduction because, based on bulk densities, the melted product is between 15 and 30% more dense.

D&D Technology

The major objective of this task is to provide technical support and planning to the D&D Program at the Oak Ridge Gaseous Diffusion Plant (ORGP) and, ultimately, to the similar programs at the DOE enrichment plants at Portsmouth and Paducah. Another objective is to provide interface and coordination between the D&D Program and

divisions at ORNL that can make potential contributions in carrying out RD&D in support of the program.

R&D Support for the K-25 D&D Program

The D&D Program for ORGP enrichment facilities began ~2 years ago when separative work projections indicated a restart would not be forthcoming. Conversion of the site from standby to long-term protective storage in Phase I of the D&D Program began immediately to bring the facilities into legal compliance. Surveillance and maintenance (S&M) will also be carried out in Phase I, which is projected to last beyond the year 2000. Phase II will involve the ultimate disposition of large quantities of contaminated equipment and materials.

Phase III would involve the ultimate disposition of the buildings themselves and the remediation by-products associated with the site. Projections indicate an overall cost of several billion dollars, so that the potential for cost savings is enormous as a result of near-term planning and RD&D. This task is in direct support of this potential for cost savings, and associated activities that began about November of 1988 are summarized below.

Initial efforts were aimed at developing a prioritization of D&D RD&D needs, which began by dividing the projected D&D efforts into a detailed work breakdown structure (WBS). Several Energy Systems personnel knowledgeable in D&D were asked to rank each WBS element according to RD&D needed in that area, and, as a result, a composite needs ranking was established which would be used later in developing an RD&D plan. Prioritization was based on three considerations: (1) Is technology available to accomplish D&D objectives? (2) Is there a potential for cost savings for timely RD&D? and (3) Are there risks to be mitigated? Three major areas were considered for each WBS element: (1) treatment of the material, (2) management of secondary wastes, and (3) disposition of the material.

Shortly thereafter, a series of workshops (12 to date) was initiated with the objective of generating detailed alternative diagrams for the many WBS elements associated with the older, high-enrichment facilities (K-25 and K-27), which are expected to be processed first. The workshop utilized multidivisional skills and the expertise of retired ORGP employees who have been retained for their historical knowledge. These workshops were

Table 2.5. Laboratory tests of TRU salt products

Physical stability (expansion/contraction)
Hygroscopic properties at 64 and 84% RH
Chemical segregation
Residual water
Particle sizing
Thermal gravimetry
Bulk density
Potential for NO _x generation
Radiolysis and criticality potential

also used as a forum for discussion of a variety of issues identified in generating the alternative diagrams. To date, more than 30 diagrams have been identified, and more than 20 are complete or in draft form.

Two major documents were prepared by the D&D Program for DOE/HQ, DOE/ORO, and Energy Systems. A multidivisional team, which included Chem Tech staff, generated the *D&D Five-Year Plan*,⁴⁰ meeting a March 1989 milestone. Later in the year, a second document, the *D&D Strategy Paper*,⁴¹ was issued for management review, meeting another significant milestone.

A third major document has been prepared in draft form—a plan outlining the RD&D needed to support an ORGDP D&D Program with the endpoint scenario of reusing the buildings at ORGDP after equipment removal. Beginning with the prioritization of needs described earlier, the next step was soliciting proposals for projects from RD&D organizations within Energy Systems to match the prioritized needs. The final ranking of the 53 proposed projects submitted was determined by three factors: (1) technical merit, (2) project cost, and (3) need, as determined by the prioritization exercise. The highest-ranking projects were then organized into the RD&D Program to match funding levels in an early draft of the *DOE Five-Year Plan*. The RD&D plan is now in draft form and will be receiving management and program review.

Other activities in support of the D&D Program include definition of potential D&D RD&D projects for submission to the *DOE RDDT&E Plan* and for consideration for funding from the HAZWRAP Program. Technical support and planning have also been provided to the program for defining and arranging demonstrations of private sector technology that has potential D&D benefits. Internally, a demonstration was also arranged and completed through the Center for Engineering Systems Advanced Research (CESAR) Laboratory at ORNL to apply robotics technology to the surveying and mapping of contaminated floor surfaces at ORGDP. This mapping effort is expected to continue.

K-25 D&D Cost Model

The main objective of this task is to develop a model for cost estimation for the D&D of buildings K-25, K-27, K-29, K-31, and K-33 in the ORGDP

complex that can be used to compare alternative D&D technologies and to determine the changes in total project cost, schedule, and waste production in an interactive real-time mode.

The first phase of the task involves the gathering of information about location, existing equipment, decontamination methods, and cost and process models. Physical characteristics of the buildings and the corresponding equipment are now available. Partial-cost models for the decontamination of some equipment have been obtained. Existing decontamination processes have been analyzed, and process modeling for waste production has been started. Most of the information will be stored in a data base system.

The second phase of the task is related to the construction of a data base that will store most of the information obtained in the first phase. Two systems have been selected to build the data base: dBASE® IV and RdB. The dBASE IV will allow delivery of the package on an IBM-PC-compatible machine; RdB will allow delivery of the package on a MicroVax machine.

Most of the information found in the first phase has been included in the dBASE IV data base. This data base is integrated with an expert system using VPEXPERT, a commercial shell for IBM-PC-compatible machines. Text files that deliver additional information on the decontamination processes are integrated with the same system. The process modeling will be performed with conventional computer programs that will also be integrated with the expert system.

Efforts have been started to put the information found in the first phase into the MicroVax environment using the data base management system RdB. This system has a natural language interface, Intellect, that will allow the data base to be queried in common English language, thereby satisfying the user needs for different methods of questioning the data base.

D&D Technology Demonstrations

The main objective of this task is to demonstrate new D&D technologies on actual facilities, equipment, and waste streams that would be encountered in ORR D&D activity.

The first of the demonstrations is to evaluate several technologies for decontaminating highly radioactive and contaminated hot cells in the ORNL complex. All of the proposed technologies

use some type of recycle system or volume reduction for the decontamination agent to reduce the amount of secondary radwaste generated during the decontamination activity. Two of the technologies have ion-exchange systems that will selectively remove isotopes of interest or separately process PCBs, which eliminates producing a mixed waste. Bench-scale testing of artifacts from the hot cells is progressing with the evaluation of the surface finishes and contamination levels prior to the testing. The candidate technologies are (1) mobile ultrahigh-pressure water systems [$\geq 35,000$ psi (≥ 241 MPa)] with a liquid recycle system and waste processing module; (2) mobile liquid abrasive blasting with a recycle system for the abrasives and water; and (3) a pelletized carbon dioxide decontamination system. The mobile systems with a recycle capability will eliminate or significantly reduce the amount of secondary radwaste in liquid or solid form. Two hot cells have been selected to be decontaminated with the technologies that successfully pass the bench-scale testing by producing sufficient decontamination factors that will allow evaluation of the recycle systems for effectiveness, efficiency, and safety. The technology selected will be used to reduce the radiation and contamination to acceptable levels where workers can enter the hot cells and upgrade the waste drain system.

The second demonstration area is the destruction of organic waste streams generated in the D&D process. The 816 coolant from ORGDP has been selected as one of the test waste streams from the K-25 building. A sample of this material was provided to a contractor laboratory for bench-scale destruction of the coolant by a ozone/high-energy-light discharge system. In addition, waste streams have been identified and selected at ORNL for use of this technology. It could be used to destroy the organics in the water of cooling towers, thus negating the use of chromates and biocides for the control of corrosion and bacterial growth in the circulating water. Follow-on work in this area will demonstrate using point-source liquid waste streams in the cooling towers and destroying the waste, in addition to controlling the quality of the cooling water. Initial bench-scale tests have shown significant promise with regard to the destruction of organics. However, data from independent evaluation of the prototype equipment have shown problems in scaleup.

Molten Salt Reactor (MSR) Surveillance and Maintenance Project

In response to tightening environmental standards at ORNL facilities, the aging containment features of the MSR Facility were evaluated and upgrades were planned for sustained surveillance and maintenance with the solidified fuel salt remaining in the drain tanks. The improvements to be made were divided into the following six work elements:

1. replace the stack fans, which have exceeded their design life;
2. provide continual negative ventilation for the reactor drain tank (RDT) cell;
3. provide a means for leak-testing the RDT cell under vacuum conditions rather than at an elevated pressure, as is the current practice;
4. empty the liquid waste tank;
5. upgrade the surveillance instrumentation by grouping signals and adding alarms; and
6. install a monitor to determine the F_2 concentration of the gas exiting the RDT containing the fuel salt.

The control documentation (e.g., QA Plan, Safety Assessment) necessary for performing this work was completed, approved, and issued during the report period.

Two of the six work elements enumerated above were completed. Since the housings and support structures of the stack fans were found to be in good condition, only the wheel, shaft assemblies, and bearings were replaced (work element No. 1). Work element No. 5 was accomplished by installing a new electronic recorder that monitors temperatures, pressures, and radioactivity levels. When a value outside the normal range is detected, the chart drive is automatically activated and the alarm for that group of parameters is displayed at the Waste Operations Control Center. This arrangement enabled around-the-clock monitoring, eliminated the need for daily patrols, and increased the number of parameters monitored.

Another work element was carried to ~80% completion of the design stage. The full implementation of work element No. 3 would have entailed the installation and operation of equipment not in use when the safety analysis was performed for reactor operation. Consideration of this and other safety issues, especially those raised by DOE

Order 6430.1A,⁴² caused the program manager to suspend further work and defer the other three work elements until their impacts on schedules and budgets could be assessed.

Metal Recovery Facility, Building 3505, D&D

The Metal Recovery Facility (MRF) at ORNL was a pilot- and small-scale production reprocessing plant that was operated during the 1950s. The facility was primarily used to recover fuel and other nuclear materials from a variety of low-burnup feed materials. Since facility shutdown in 1960, the MRF has been maintained in a protective storage mode, and in recent years it has been part of the ORNL Surplus Facilities Management Program (SFMP).

In 1986, all process piping and equipment were removed from the facility cells and disposed of as radioactive waste. The initial plan for this reporting period was to decontaminate one of the facility cells (Cell G) by scarification of the surfaces with high-pressure water jets. This decontamination activity was scheduled to be conducted by Chem Tech's Isotopes Section personnel in early FY 1989. To support planning of the decontamination activities, samples of the concrete in Cell G were taken and leached at 95°C for 2 h with either water or a dilute HNO₃ solution. Aliquots of the leachates were analyzed to determine the soluble radioisotopes present; the results are given in Table 2.6. These results indicate that the radioactive species present are largely insoluble in water and could likely be effectively removed from the water used for scarification by simple filtration. Alpha scans of the acid leachate indicated that >90% of the alpha contaminants were ²³⁸Pu and ²³⁹Pu.

Unfortunately, in early FY 1989, the funding sponsor decided to use an outside contractor, rather than Chem Tech personnel, to conduct the decontamination activities. Activities to conduct

actual decontamination were diverted to preparation and review of statements of work and bid proposals. As of the end of this recording period, actual decontamination activities have not begun in the facility. The contractor is scheduled to begin work in late CY 1989 and is proposing a dry scrubbing method for decontamination. Division personnel will coordinate the contractor activities and package the waste generated by the contractor for on-site disposal.

Hazardous Materials Transportation

The Chemical Technology Division has been very active in supporting ORNL and Energy Systems in meeting the increasingly challenging requirements regulating the transportation of hazardous materials. Building on a core of expertise in this area, Chem Tech has provided assistance in formulating solutions to complex problems related to both packaging and transportation. During the report period, two specific problems were addressed, one specific to ORNL and the other Energy Systems-wide.

The Laboratory had come under increasing scrutiny from DOE/ORO in its management of radioactive materials packaging activities. This scrutiny resulted in a comprehensive evaluation of ORNL activities in this area. Chem Tech provided support for this effort by providing the chairman of the project team as well as expertise in the transportation needs and capabilities.

Energy Systems was simultaneously being criticized by DOE/ORO for packaging and transportation problems on a five-plant-wide basis. Chem Tech provided expertise to Energy Systems management to formulate a strategy for solving these problems. Energy Systems then turned to Chem Tech for assistance in implementing this strategy, and staff has been dedicated to support them in this area.

ORNL Transportation System Performance Improvement Project

The ORNL transportation system is a complex mix of organizations and interactions devoted to the safe and efficient movement of materials within and from the ORNL site. Materials which are transported include a wide variety of hazardous materials, such as radioactive and hazardous wastes,

Table 2.6. Analysis of leachates from the Metal Recovery Facility

Analysis	Water leachate (Bq/g concrete sample)	HNO ₃ leachate (Bq/g concrete sample)
Gross alpha	<10	2140
Gross beta	<10	75
Gamma scan		
¹³⁷ Cs	<20	<20
²⁴¹ Am	--	1340

as well as radioactive and hazardous products. The transportation system that exists has resulted from an evolution of organizational responsibilities and physical locations involved in shipping.

Several problems that were encountered raised the awareness that the transportation system was not working as well as it should. Problems surfaced relating to the continuation of package approvals required by regulation, the efficient and comprehensive flow of communication to the various organizations involved, and the lack of an ORNL focal point for these issues.

A Performance Improvement Process (PIP) project team was formed to evaluate the transportation system and to make recommendations for any needed improvements in the performance of ORNL transportation functions. The PIP team considered the full range of transportation functions performed at ORNL and the problems that had resulted in the creation of the project. There were several areas that could be excluded from the project since they were not associated with the identified problems. The project eventually focused on the off-site packaging and transportation of radioactive and hazardous materials (including wastes). Representatives from all of the major shipping organizations were assigned to the team, including Chem Tech, Quality Division, Environmental and Health Protection (E&HP) Division, Plant and Equipment (P&E) Division, Office of Operational Safety, Research Reactors Division (RRD), and Finance and Materials (F&M) Division. Chem Tech provided two representatives for the project: the chairman of the team and a person knowledgeable in the transportation activities of the division.

The team investigated the roles and responsibilities of the various organizations in making hazardous materials shipments. It became clear that the complexity and evolutionary nature of the ORNL transportation system presented several problems. It was not always clear on how specific functions were required to be performed. This was partially a result of incomplete assignment and documentation of responsibilities. To correct this, it was recommended that

1. comprehensive procedures be developed to control the performance of transportation functions,
2. roles and responsibilities be clearly delineated in transportation policies and procedures, and
3. policies and procedures be structured hierarchically for ease of comprehension.

Even if the problem of incomplete policies and procedures were corrected, the existing functional fragmentation of transportation activities would continue to cause difficulties. There are four major shipping divisions within ORNL, and some of these, such as Chem Tech, have multiple shippers. Without close coordination and communication, the diverse shipping activities of ORNL, as a whole, would not be easily improved and maintained in a quality manner. The following recommendations to improve the ORNL-wide management of these activities were made:

1. Strong centralized coordination of activities in both operations (actually making shipments) and safety oversight (compliance assurance) should be provided.
2. A broad-based advisory body should be formed to provide management with advice on operations matters (as the existing Transportation Safety Committee does for safety oversight matters).
3. The position of Transportation Safety Manager should be established in the Office of Operational Safety to formalize and provide coordination in the performance of safety oversight functions.
4. The position of Transportation Operations Manager should be established in F&M to formalize and provide coordination of operations functions.
5. Each of the four major shipping divisions (Chem Tech, RRD, E&HP, and F&M) should provide single points of contact for coordinating operations and safety oversight activities.
6. The F&M single point of contact should establish and advertise a program to assist "occasional shippers" in the preparation of shipments.
7. Every ORNL division should designate a person to be trained and to serve as a resource in hazardous materials transportation and to identify situations where hazardous materials under the division's control may be shipped.

Several of the problems that contributed to the formation of the project related to radioactive materials package designs which must be certified (approved) by regulatory agencies such as the NRC. These package designs must be rigorously documented, and the applications for approval must meet strict requirements on content and timeliness. The fragmentation of responsibilities for these packages had resulted in inconsistent quality and

schedule performance. The team reviewed these problems and suggested these actions:

1. The ORNL divisions should retain responsibility and be held strictly accountable for performing certification functions.
2. The responsibility for coordinating certification activities on an ORNL-wide basis should be assigned to the Transportation Operations Manager.
3. A written procedure should be developed for processing and approving package safety documentation within ORNL.

Compliance with the requirements for shipping hazardous materials is dependent on the level of training that personnel performing the functions have. The critical role of training was recognized, and to ensure that adequate training was provided, the following recommendations were made:

1. All employees should receive an introduction to hazardous materials transportation requirements (what is considered hazardous, what off-site transportation is, where to go for assistance) as part of the General Employee Training.
2. Appropriate training courses should be developed and given to all personnel involved in preparing hazardous materials shipments (graduated to the degree of their responsibilities in these functions).

The final area examined was that of QA. Quality assurance was found to be generally acceptable but needing improvement in relation to the procurement, inspection, and documentation of U.S. Department of Transportation (DOT) specification packagings. Many of the packagings are procured on a price-competitive basis, and insufficient attention to quality could leave ORNL in a noncompliance situation. It was recommended that a comprehensive vendor qualification and inspection program be established to ensure that procured packages fully comply with regulations.

The recommendations of the project team were presented to representatives of the major organizations that would be affected by them. When agreement on the recommendations was reached, a letter of agreement was prepared and signed, which provided the basis for proceeding to implement the recommendations. The project team then examined each recommendation and provided more detailed implementation recommendations. All of these were collected into the project's draft report.

The recommendations were presented to the ORNL Executive Committee for its information and concurrence. Some clarifications were made in the project's final report,⁴³ and it is now serving as the basis for implementing the changes needed. The report has been well received and is being smoothly put into place. Several of the organizational changes have been instituted, including the establishment of the Chem Tech transportation focal point. The work of the project team has also been recognized through presentation of the President's Award for Performance Improvement to the team members.

Support to Energy Systems Transportation Safety Management

Beginning with the third quarter of FY 89, Chem Tech has provided technical support to the central staff through the Transportation Technologies Group in the Engineering Coordination and Analysis (EC&A) Section. Involvement stems largely from earlier ORNL transportation and packaging PIP activities under the leadership of the Transportation Technologies Group. A number of PIP recommendations originally intended for ORNL were adopted by the central staff and are being implemented by the other four Energy Systems plant sites. Principal among these is the creation of the position of Transportation Safety Manager on the central staff and the creation of the positions of Installation Transportation Operations Manager (ITOM) and Installation Transportation Safety Manager (ITSM) at each of the five plant sites: ORNL, the Oak Ridge Y-12 Plant, and the Gaseous Diffusion Plants in Oak Ridge (K-25), Portsmouth, Ohio, and Paducah, Kentucky. The purpose of these new positions is to establish central management oversight of transportation and packaging safety matters and to create single points of contact for transportation and packaging operations and safety at each of the five plant sites. The TSM reports to the Facility Safety Manager (FSM) in the Environmental and Safety Activities (ESA) organization. The position was staffed in mid-August 1989.

Support from Chem Tech to the Energy Systems central staff includes one full-time employee from the Transportation Technologies Group. Two major support functions are provided: (1) independent review of Safety Analysis Reports for Packaging (SARPs) and (2) technical support to the TSM.

Fissile and other nuclear material whose radioactivity exceeds certain limits as defined in DOT regulations must be transported in packaging that meets NRC Type B requirements. Type B packaging must be able to provide containment, afford shielding, and, if applicable, prevent criticality of the contents, in both the course of normal transport conditions and in the event of a serious accident. Depending on use, Type B packaging is certified by either the NRC or by DOE (DOE Headquarters, DP 4.1, for nonweapons packaging and DOE Albuquerque Operations Office for weapons packaging) and receives a 5-year Certificate of Compliance when demonstrated, through a SARP, that all regulatory requirements are met.

DOE Order 1540.2/ORO⁴⁴ requires that all DOE/ORO-sponsored SARPs receive an independent review prior to submission for certification. The criteria for independent SARP review are twofold: (1) that the reviewers have had no role in the preparation of the SARP and (2) that the reviewers have no interest in the use of the package. Energy Systems SARP review emphasizes the independent confirmation of analytical methods and results.

Because management of independent SARP reviews through the central staff is a new initiative, Chem Tech support activity in the first quarter of FY 89 focused on a search for, and selection of, a qualified SARP review base. In addition to the two criteria mentioned above, a third selection criterion requiring Q-cleared personnel and facilities for classified document control evolved as a result of the significant numbers of classified Y-12 SARPs. After analyzing resources within Energy Systems, in the private sector in Oak Ridge and elsewhere, and at other DOE facilities, particularly Albuquerque and Richland, it was determined that the most qualified resources for conducting independent SARP reviews for Energy Systems reside in Oak Ridge and in-house.

In late July, ESA's FSM approved the use of ORNL's Applied Technology Division for SARP review. FY 89 and FY 90 funding was approved and established. The first SARP review, under the direction of Chem Tech support to the TSM, began in August 1989.

Chemical Technology Division technical support to the TSM has centered on administrative and organizational start-up activities and program definition. Principal among these is the reformation of the Energy Systems Transportation Safety

Committee (ESTSC) under the leadership of the TSM. The ESTSC, and its associated ad hoc subcommittees, is a primary means of communication between the five Energy System plant sites and the TSM. An important key feature of the central role of the TSM is a renewed ability to coordinate and to standardize, where appropriate, Energy Systems-wide site activities. During this reporting period, Chem Tech technical support to these ongoing activities has involved the development of procedures for the use of 6M DOT specification packaging, the use and disposition of obsolete packaging, the development of on-site transfer manuals, and the initiation of drug testing for vehicle drivers, all of these at each of the five plant sites. In addition, a second means of communication is the creation of a *Transportation and Packaging Bulletin*,⁴⁵ to be published on a regular basis. On a more formal level, with the assistance of Chem Tech technical support, an Energy Systems transportation and packaging policy is being developed and defined through the line organization. Policy documents, as they are developed, will be promulgated to plant managers under the signature of the ESA director.

2.2.2 DOE Generic Waste Technology

Three-Mile Island Support Studies

Chemical Technology Division staff members, as well as other ORNL staff, were integrally involved in many phases of the recovery operations at the Three Mile Island Unit 2 (TMI-2) Nuclear Power Station following the accident in 1979. In addition, Chem Tech managed all TMI-2 assistance provided by personnel at DOE facilities in Oak Ridge.

Defueling of the reactor vessel and the reactor containment building, packaging of the debris, and shipment to Idaho National Engineering Laboratory (INEL) were essentially completed during this report period. ORNL personnel from the Reactor and Fuel Cycle group of the Nuclear Engineering Applications Section, Computing and Telecommunications Division (C&TD), provided analyses of the potential for criticality during various phases of the defueling, packaging, and shipping operations, as well as for the Post

Defueling Monitored Storage (PDMS) phase, which is now beginning at TMI-2.

Decontamination operations in the reactor and auxiliary buildings at TMI-2 were also essentially completed during this report period. In support of part of this work, Chem Tech personnel completed and reported on the "Analysis of Data from Leaching Concrete Samples Taken from the Three Mile Island Unit 2 Reactor Building Basement."⁴⁶ The DOE financial support for the TMI-2 recovery project ended on September 30, 1989.

Gas Separations Using Inorganic Membranes

The overall objective of this program is to develop and evaluate inorganic membranes for separating gases at high temperatures and pressures and/or in hostile process environments. The program will apply porous membrane technology to the separation of various gases present in coal gasification processes. Major parts of the program include (1) assessing the status of R&D activity related to gas separations using inorganic membranes; (2) identifying and selecting candidate membrane materials; (3) fabricating and distinguishing physical characteristics of candidate membranes using specialized techniques available at ORGDP; and (4) evaluating the separations capability of the fabricated membranes in terms of permeabilities and fluxes of gases.

Synthesis gas produced in coal gasification processes may contain hydrogen, carbon monoxide, nitrogen, water, carbon dioxide, hydrogen sulfide, and other gases, depending on the particular gasification process. Inorganic membranes could potentially be developed to separate these gases under gasifier exit gas conditions, significantly improving process efficiency and economics. Several porous inorganic materials that could be used as membranes are commercially available, but the minimum pore diameter is in the range of 3 to 4 nm. While some gas separations can be achieved with these materials, it is generally accepted that smaller pore diameters or other membrane modifications will be needed for efficient gas separations.

The available literature and other information on inorganic membrane R&D in gas separations were compiled in a topical report, *Using Inorganic Membranes to Separate Gases: R&D Status Review*,⁴⁷

during this reporting period. Inorganic membranes are being investigated worldwide for separating gases on a laboratory scale. Membrane materials include metals, ceramics, and glass. Most of the membranes have pore diameters >2 nm, and the primary transport mechanism is Knudsen diffusion. Membrane preparation methods are based on sol-gel, slipcasting, anodic oxidation (metallic oxides), and phase-separation/leaching (porous glass) techniques. Metallic membranes have been used primarily for separating hydrogen isotopes and for separating hydrogen from other gases. Some of the gases that have been separated using porous glass membranes include hydrogen, helium, argon, nitrogen, oxygen, carbon dioxide, carbon monoxide, and light hydrocarbons. The most frequently used materials for metallic oxide membranes are alumina and silica. Some membranes contain mixtures of these along with other oxides such as zirconia or titania. Alumina is also frequently used as the support for other membrane materials. The permeation behavior of several gases has been studied using alumina or alumina-containing membranes. These gases include hydrogen, nitrogen, carbon dioxide, helium, argon, oxygen, hydrogen sulfide, sulfur dioxide, water, alcohols, and light hydrocarbons.

The design and construction of a test apparatus for measuring the gas permeabilities of membranes at high temperatures (up to 500°C) and pressures (up to 4 MPa) were completed. A flow diagram of the apparatus is shown in Fig. 2.14. Gases are supplied from cylinders through high-pressure regulators and an associated manifold. Pressures are controlled by pressure control valves, and gas flows are measured with differential pressure transmitters. The gases are preheated in a tubular furnace which will also house the membrane enclosed in an outer tube. Temperatures are measured with thermocouples. Exit gases are cooled with heat exchangers and analyzed by gas chromatography. Shutoff valves are operated by air-controlled actuators, supplied through solenoid-operated valves. The recorder/controllers, thermocouple readouts, and valve switches are installed in a separate instrument cabinet. Efforts are now directed toward developing the necessary metal-to-ceramic seals for attaching the membrane specimens to the test apparatus.

During the past year, significant improvements were made in fabricating alumina membranes at ORGDP. The average pore radius of the

for these activities is completed and coupled to the expert systems, thousands of combinations of use/decontamination/disposal scenarios can be evaluated rapidly using "English" queries by personnel not trained in data base or modeling procedures.

Neural network applications

A number of small projects this past year have demonstrated the usefulness of neural nets in solving engineering problems. Among these are:

Forecasting—Two PC-compatible neural network shells, NeuroShell and NeuralWork, were used to forecast LLW production for the X-10 site. More accurate predictions were obtained by both neural nets than by the standard approach, a regressive statistical model. The NeuralWork program provided the best results of the three methods.

Troubleshooting a sanitary waste treatment system—A "hybrid system," a combination of an expert system and a neural network, was used to develop a problem diagnostic and solving system for an activated-sludge sanitary waste treatment system. By focusing on settling rates and appearance factors associated with the clarifier, the neural network can determine whether a problem exists and identify its causes. Once a problem appears, the expert system/neural net identifies the causes of the problem and indicates the proper procedure for the operator to take to correct the problem.

International Technology Programs (ITP) Support

The ITP of Energy Systems provide technical support to DOE and other federal agencies concerning foreign nuclear technologies and their implications for international nonproliferation agreements and multinational organizations involved in furthering the peaceful uses of nuclear energy. Chem Tech is providing technical support to ITP in areas related to the nuclear fuel cycle, including waste management. Although Chem Tech has a longstanding relationship with ITP, recent events have enhanced needs such that multiple full-time employees are working on ITP matters.

Chem Tech staff contributed to an evaluation of safeguards and security considerations for the DOE/OCRWM waste management system.⁴⁸ This

work evaluated the reference OCRWM system against applicable safeguards and security requirements. Seventeen activities were recommended to resolve the various issues identified.

More recently, Chem Tech staff was centrally involved in formulating conceptual criteria concerning the termination of safeguards on radioactive wastes as a basis for the U.S. position in meetings at the International Atomic Energy Agency (IAEA). The specific issue addressed was identifying the basis for terminating safeguards on an arbitrary waste stream and then quantifying this as measurable criteria. During the course of the work, the basis and quantification were accomplished, documented,⁴⁹ approved by the United States government, and presented as a working paper at the IAEA.

Chem Tech has a continuing involvement in providing technical input to an ITP project to evaluate Atomic Vapor Laser Isotope Separation (AVLIS) technology as it relates to the need to classify and protect technology details because of proliferation and safeguards concerns. This evaluation is being conducted in phases. The most recently completed phase involves developing and documenting a baseline description of the AVLIS process suitable for further evaluation.⁵⁰

DOE Waste Management Applied Research, Development, Demonstration, Testing, and Evaluation (RDDT&E) Planning Support

During 1989 the newly named Secretary of Energy very quickly changed DOE's approach to addressing the waste problems on its many sites. Specifically, he took measures to (1) centralize management of waste-related matters at DOE/HQ, (2) increase the budget for waste management and environmental restoration on DOE sites, and (3) greatly enhance the importance and budget of waste-related RDDT&E. With specific regard to the latter, DOE initiated the preparation of a comprehensive RDDT&E plan. This was accomplished by establishing 17 teams comprised of technical staff members representing all DOE sites. These teams were typically defined according to a specific type of waste-related technology. Each team was responsible for identifying DOE needs related

to its technology area, assessing the technology status in that area and making recommendations for RDDT&E that would reach fruition in 2 to 5 years.

Chem Tech was asked to manage five of these teams related to the treatment of DOE wastes. The technologies addressed by the teams were chemical processing, thermal/photo processing, soils treatment, D&D, and biological processing. Chem Tech also provided the team leader for the chemical processing team, as well as team members for two of the other teams.

The input to the plan was developed on an extremely tight schedule (the teams had about a month between the organizational meeting and the completion date). The schedule was met, and a draft RDDT&E plan was assembled on an equally tight schedule. This plan is now being circulated for public comment.⁵¹ A companion report containing the details of the efforts of the 17 teams will be issued shortly.

Radioactive Waste Partitioning-Transmutation (P/T)

An advanced fuel cycle for LWRs and advanced liquid-metal reactors (ALMRs) that incorporates waste P/T is being investigated. As applies herein, partitioning is the recovery of long-lived radionuclides (Np, Pu, Am, Cm) from wastes. Transmutation is the conversion of these radionuclides by fissioning in a reactor to short-lived radionuclides. The P/T process assists waste management by eliminating the long-lived radionuclides in HLWs that would ordinarily go to a repository.

The proposed fuel cycle is shown in Fig. 2.15. LWRs are fueled with uranium oxide fuel (current practice). The LWR spent fuel is processed to recover uranium, plutonium, and the other actinides. These actinides are converted and are fabricated into fuel assemblies which provide the initial fuel for an ALMR. The plutonium and higher actinides produced in existing LWRs in the United States in 1 year are sufficient to start up one ALMR. ALMR spent fuel may be processed using a new pyroprocess developed at ANL with plutonium and actinides recycled into new ALMR fuel. The characteristics of the ALWR allow both the

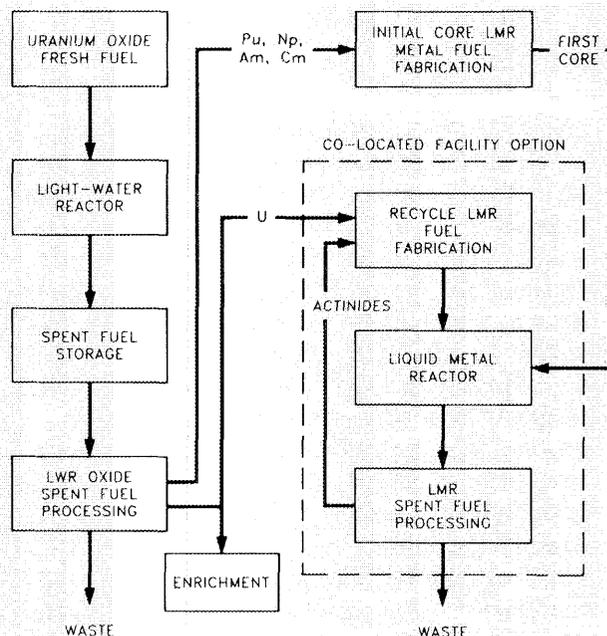


Fig. 2.15. Conceptual LWR/ALMR fuel cycle.

plutonium and higher actinides to be used as fuels while destroying these long-lived radionuclides.

Chem Tech, in cooperation with other divisions, is undertaking a series of studies on this advanced fuel cycle, including the following:

1. A review of historic work on waste partitioning was completed.⁵² This provided an update on P/T research and issues worldwide since the large-scale studies on P/T at ORNL in the late 1970s.⁵³
2. A preliminary study was completed on the preferred processing methods for LWR fuel to provide metal plutonium and higher actinides for LMR fuel.⁵⁴ The requirements for this step differ significantly from those in typical reprocessing plants in several ways:
 - a metal rather than oxide product is required,
 - plutonium and all the higher actinides are to be recycled, and
 - the LMR fresh fuel can have very high loadings of various fission products.

Additional studies on this new fuel cycle are planned.

Transportation Support to DOE/ES&H

The DOE Office of Environment, Safety, and Health (ES&H) has the oversight responsibility to ensure that all aspects of environmental, safety, and health-related DOE activities are adequately performed. DOE Order 5480.3⁵⁵ establishes the requirements for the packaging and transportation of hazardous materials, substances, and waste applicable to all DOE operations. Among the ES&H responsibilities delineated in the order (and a subsequent draft of a replacement order) are:

1. provide an independent safety and compliance assessment of DOE's transportation operations, including the packaging and transportation of hazardous materials;
2. serve as the DOE point of contact and coordination on safety matters between departmental elements and other agencies;
3. ensure that all packages meet applicable safety standards and regulations;
4. provide technical assistance and support to all DOE/HQ and field organizations in matters pertaining to packaging and transportation; and
5. support safety appraisals of DOE nuclear facilities.

To provide ES&H a mechanism to implement its obligations as defined in a proposed revision to DOE Order 5480.3, Energy Systems was selected to provide technical and management support. The Chem Tech Transportation Technologies Group (TTG) was assigned these tasks and serves as the integrating contractor.

Program Management

The Packaging and Transportation Safety (PATS) Program was initiated by Chem Tech's TTG in October 1988. A project logic sequence, Fig. 2.16, was defined for major PATS activities. The critical path for key activities is shown in the center sequence on the logic diagram. The other two sequences display parallel activities supporting the prime sequence. The top sequence depicts the prioritization and scheduling necessary to direct PATS activities in a manner responsive to the program manager's needs. The bottom sequence depicts activities required to develop the resource base necessary to support the program. From the logic diagram, the PATS Program was divided into a Work Breakdown Structure (WBS), milestones were defined, and a schedule and budget were developed. A PATS Program Management Plan was

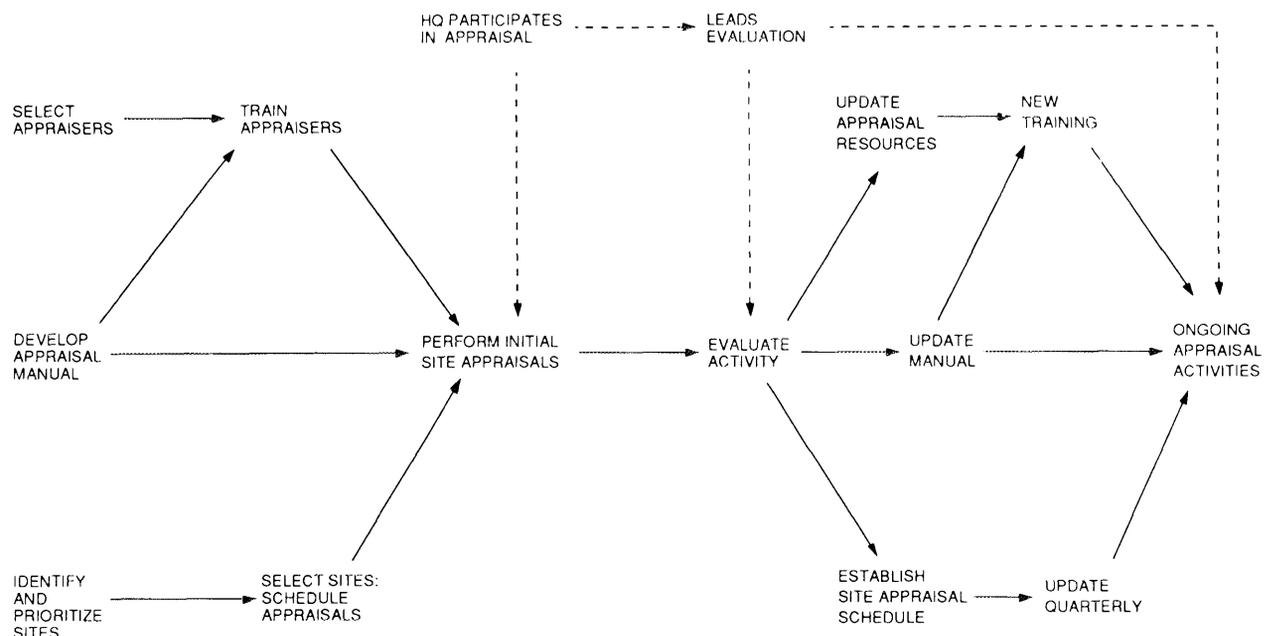


Fig. 2.16. A logic sequence for the PATS Program.

developed that describes the WBS work plans and associated milestones, schedule, and budget in accordance with DOE Order 4700.1, Project Management System Requirements.⁶⁶ The Program Management Plan was completed in late June 1989 and has been favorably received by ES&H.

A PATS Quality Assurance Plan (QAP) was prepared to establish QA requirements for the PATS Program and the procedures to implement those requirements. The QAP functions under the Waste Research and Development Quality Assurance Program and has been approved by the QA specialist and the ORNL Quality Program Manager. The PATS Program was determined to be a Quality Level 2 program. The QAP was completed and approved in August 1989.

Appraisal Manual

It was apparent that consistent evaluations of transportation operations would require an appraisal manual to train appraisers and to serve as a guide in the procedures, criteria, objectives, and regulations for an appraisal of a DOE facility's packaging and transportation program. The work on the appraisal manual was transferred to ORNL by ES&H in July 1989. Previous preliminary drafts and outlines had been prepared by a Washington-area contractor. A new outline of the manual was developed by PATS staff, and some draft chapters were prepared and presented to ES&H in September. A system for judging the severity of any concerns developed in the course of an appraisal was also developed as well as a "model" appraisal report. The "model" report describes the elements necessary for the draft and final appraisal reports and will be used to expedite report writing in the field. These systems will be field tested by ES&H in the first functional appraisal of a facility packaging and transportation program. Other chapters containing performance objectives and criteria, as well as checklists, are also under development. Drafts of these chapters are expected in the first quarter of FY 1990.

Completion of the appraisal manual is a major milestone and will enable the PATS Program to move into the appraiser training phase, as indicated on the logic diagram (Fig. 2.16). In anticipation of this training program, computer- and/or video-assisted training programs are being investigated. These will be developed specifically for the PATS Program for use with the appraisal manual.

Packaging Reviews

The PATS staff completed several reviews of technical proposals for transportation operations that would be excepted from certain regulatory requirements. The proposals were evaluated for reasonableness and the level of transportation safety that would result if the exceptions were granted. A proposal to continue the use of ATMX railcars for the transportation of CH TRU waste was evaluated. This review coordinated the results of a Peer Review Panel commissioned by ES&H in mid-1988. The panel met with the ATMX Railcar Safety Assessment Committee, responsible for preparation of the system safety assessment document, and submitted comments during the development of the draft safety assessment.

An evaluation of the "Safety Analysis Report for Packaging—UNH Trailer" was performed and comments supplied to ES&H. The approach used in the report was basically flawed in its interpretation of what packaging would normally be required for this material under DOT regulations. Consequently, a comparison to "equivalent" standards was not performed and a justification for allowing the use of the trailer and its equivalence to currently required packaging was not made. These conclusions helped guide DOE in the selection of alternatives that would not require the transport of this material.

An application for the transport of the general-purpose heat source—radioactive thermoelectric generators (GPHS-RTGs) from Mound Laboratories in Miamisburg, Ohio, to the Kennedy Space Center at Cape Canaveral, Florida, was reviewed for transportation safety provisions. The GPHS-RTGs are plutonium-fueled thermoelectric generators that are utilized in the Galileo and Ulysses space missions as power supplies for the planetary probes. The RTGs are significant heat sources (~4 kW) and must also be protected from theft and sabotage. DOT- or NRC-approved shipping containers for highway transport were not available, nor were they feasible, within the time constraints of the space launch.

The application, accompanied by an SAR, proposed a transport system utilizing the DOE safe secure trailer (SST) and operational controls to ensure safety equivalent to that required by the regulations. A critical examination was made of all aspects of the proposed system, including operations of the SST, loading and transport preparation operations at Mound, and emergency response

procedures for transport. Visits were made to the Southeast Couriers Section at the Y-12 Plant and the Transportation Safety Division at the Albuquerque Operations Office to evaluate response and recovery plans. PATS staff made recommendations for additional transport controls and emergency and operating procedures to mitigate the potential safety problems resulting from the unique nature of the package and transport vehicle. ES&H was provided with a detailed evaluation of the System Safety Assessment document prepared for the shipments, and a visit was made to the Transportation Safety Division at the Albuquerque Operations Office was performed to verify that the recommendations for transport controls and emergency and operating procedures were executed. ES&H authorized the first of these shipments, and they were completed in August without incident.

Regulatory Analyses

Activities related to regulatory responsibilities of the PATS program have included (1) analysis of DOT regulation 49 CFR Pt. 173.474³⁷ and its interpretation by DOE for first usage of foreign-constructed UF₆ packagings; (2) transportation regulatory impacts for the DOE Nuclear Weapons Complex Modernization Study; and (3) comments on changes to the DOT Emergency Response Guidebook.

Additionally, the PATS staff has participated in IAEA, International Standards Organization, and American National Standards Institute committee activities.

Information Management

At ORNL, C&TD is working with Chem Tech PATS staff to develop a data base for the PATS information management. Data elements have been defined, and investigation into a computer network system for the finished data base has been initiated. The data base will contain information related to appraisal activities and resources, including:

1. information on DOE facilities, their transportation programs, and unique characteristics of their hazardous materials transportation activities;
2. information on completed appraisals, with open items, unresolved concerns, and schedule for appraisal visits; and.

3. data on PATS appraisers, their training, participation in appraisals, and other pertinent information.

The Transportation Capability Assessment System was transferred from Oak Ridge Associated Universities (ORAU) to Chem Tech. The data base contains the resumes of experts in many transportation-related fields and is being used to help in selecting technical specialists for the PATS Program.

Waste R&D Program Management

During the last several years, Chem Tech has had a leadership role in the waste-related R&D for all of ORNL. This has occurred because of Chem Tech's historical involvement in waste management dating back to the first days of fuel reprocessing and the need to manage the resulting wastes.

During this report period, Chem Tech staff were responsible for directing this entire program area, constituting ~\$25M/year and 80 identifiable tasks. Changes in the Chem Tech organizational structure and the greatly accelerated pace of waste-related R&D activities resulted in the transfer of this responsibility out of Chem Tech and the creation of a separate waste management R&D program office in the middle of CY 1989. However, Chem Tech continues to be heavily involved in the management of ORNL waste-related R&D activities because most of the program managers comprising the new waste management R&D program are Chem Tech staff.

2.2.3 Work for Others Waste Technology

Oak Ridge National Laboratory has fostered significant process design, development analysis, and demonstration capabilities to handle environmental restoration and waste management problems. This capability has been offered to other federal agencies by working through the Hazardous Waste Technology Program (HWTP) (see Sect. 3.1.3 of this report), and several large-scale demonstrations were conducted for Air Force and Navy sponsors during the review period.

These types of projects make excellent use of Chem Tech capabilities and provide a synergistic effect for both the DOD sponsors and for DOE.

Technology and lessons learned from these demonstrations are being transferred to new demonstration projects on the ORR where the experience gained can be put to use remediating leaking burial grounds. The WFO projects provide an opportunity to develop diverse training, conduct and publish exciting work, and extend the capability of Chem Tech.

Selected projects are described in this section. The air-stripping project was successful in evaluating several unit operations in a field system. This work is being expanded to include an Air Force base (AFB) in Michigan. The soil-venting project demonstrated the versatility of this technology at a base in Utah. Reverse osmosis, a novel approach to cleaning metal plating waste, was demonstrated for the Navy. Finally, the analysis capability of ORNL was used to support environmental surveys at several AFBs over the review period.

Air Stripping with Emissions Control

The primary objective of this project, which was conducted for the Department of the Air Force, was to field test innovative air-stripping techniques, such as rotary air strippers (RAS), and materials in concert with emissions control technologies (catalytic incineration and granular activated carbon) for the treatment of groundwaters contaminated with organic compounds.

The ultimate goal of the program was to provide managers and engineers who are responsible for groundwater cleanup with design information for full-scale implementation and to provide a direct comparison with technologies and materials tested in previous Air Force studies. A secondary objective was to develop a manual to aid the major commands and base-level decision makers in the selection of an appropriate remedial action. This guidance manual will focus on comparing pump-and-treat technologies but will briefly describe other viable alternatives.

A packed-tower air-stripping system, which included an activated carbon adsorption unit, was designed, fabricated, and installed on a portable skid at ORNL. Also, a trailer was outfitted with analytical equipment and a data acquisition and analysis system. The skid and trailer, along with a purchased RAS and a leased catalytic incineration unit, were shipped to Eglin AFB in Florida in August 1988. The test site at the base was the fuels storage area where the groundwater had been contaminated by a spill of JP-4 jet fuel.

After startup operations were completed in October 1988, an experimental program was conducted through May 1989. The objectives of the air-stripper tests included evaluation of new packings, evaluation/development of performance correlations, and demonstration of long-term operability. In the carbon adsorption and catalytic incineration tests, the objectives involved evaluation of different types of catalysts, performance correlation evaluation, and determination of the behavior of a multicomponent feed to these emissions control units.

In the packed-tower air-stripper tests, four different packings were evaluated. Pall ring packing, a standard random packing for which performance correlations are available, was used as the base case. The experimental results were in good agreement with the predictions of the Pall ring correlations. The performance characteristics of three structured packings, Delta SH, KOCH/SULZER, and KOCH Flexaramic packings, were determined. For the eight volatile organic compounds (VOCs) of jet fuel that were specifically monitored, the KOCH/SULZER packing produced the lowest values for the height of a transfer unit and, consequently, the highest removal efficiencies. All packings showed evidence of iron deposition [the groundwater contained ~10 ppm_w (10 mg/kg) of iron]. In particular, after 1 month of continuous operation, a significant fraction of the cross-sectional area of the Delta SH packing was plugged by iron deposits. For air stripping of groundwaters containing high concentrations of iron, pretreatment of the groundwater or frequent cleaning of the stripper packing will be necessary.

In the RAS, groundwater is driven through the packing by centrifugal force. Two types of packing, a wire mesh and a metal gauze packing, were evaluated in the RAS. The RAS produced very high removal efficiencies. For example, a certain fractional removal of a VOC could be effected over a radial distance (in the contacting element of the RAS, fluid flow is in the radial direction rather than the axial direction as in the packed tower) of ~3 cm in the RAS, as compared with a height of ~60 cm in the packed-tower stripper with Pall ring packing. Because of the small hydraulic diameter of the packing, the RAS was particularly susceptible to iron deposition. As in the case of the packed-tower stripper, pretreatment or cleaning will be required when utilizing the RAS to treat groundwater with a high iron content.

The performance of the catalytic incinerator, used to destroy the VOCs in the effluent air from

the air stripper, was adversely affected by the presence of volatile sulfur compounds in the groundwater [sulfur concentrations as high as 800 ppb_w (0.8 mg/kg) were measured]. Conversions were only on the order of 20 to 30%. The catalyst evaluated was a noble metal-based catalyst. A sulfur-tolerant catalyst will be required if catalytic incineration is utilized for emissions control when a significant sulfur concentration exists in the groundwater.

In the tests with activated carbon, used to adsorb the VOCs in the effluent air from the air stripper, a significant increase in bed capacity was obtained by operating at a humidity of ~40% (obtained by preheating the feed to the adsorber bed). A molecular sieve adsorbent, which was also evaluated, had a lower capacity than activated carbon. A potential advantage of this adsorbent is that it can be regenerated in situ with ozone.

A draft of the final report was completed and delivered to the Air Force project officer for review. As part of the guidance manual, a spreadsheet was developed that will allow AFB personnel to perform preliminary design and cost estimates for an air-stripping system with emissions control. The user will have the options of the type of air stripper, which may be a conventional packed tower or an RAS, and the type of emissions control, which may be carbon adsorption or catalytic incineration. In order to study the effect of uncertainties in the input parameters, the spreadsheet may also be used to do sensitivity analyses. A draft of the guidance manual will be delivered to the Air Force project officer by the end of 1989. This project will be completed when the comments have been incorporated into the final report and the guidance manual.

In Situ Soil Venting

A promising technology for the remediation of unsaturated zone soils contaminated with VOCs is in situ soil venting. In this technique, a large volume of air is passed through the soil. The air flow sweeps out the gas that is present in the soil, disrupting the equilibrium that exists between hydrocarbons that (1) are sorbed onto soil particles, (2) are dissolved in the water in the soil pore space, (3) exist as a liquid hydrocarbon phase, and (4) are present as vapor. This results in volatilization of the contaminants and subsequent removal in the airstream. In situ soil venting, which is most

applicable to soils of high permeability and contaminants of high volatility, may be a cost-effective decontamination technology.

A project is being conducted for the Air Force Engineering and Services Center, Tyndall AFB, Florida, to demonstrate in situ soil venting at a fuel spill site. Chem Tech is performing the study in conjunction with the Environmental Sciences Division (ESD) and the Health and Safety Research Division (HSRD). The objectives are to determine cleanup efficiencies attainable by soil venting and to define operating, cost, and system design parameters for application of soil venting at other sites.

The site selected for the in situ soil-venting study was a fuel storage area at Hill AFB, Utah, where 102,000 L of JP-4 was spilled from an underground storage tank in January 1985. Based on information from a site characterization and a one-vent pilot test that was conducted in January 1988, a full-scale in situ soil-venting system was designed for collection of data and cleanup of the JP-4 contaminated soil. The venting system consisted of three subsystems (see Fig. 2.17): (1) a vertical vent array in the area of the spill, (2) a lateral vent system under a new concrete pad and dike that were constructed for the tanks after the spill, and (3) a lateral vent system in the pile of soil that was retained after excavation of the tanks. This design included features that permit evaluation of several factors affecting contaminant transport and subsurface airflow.

A blower/emission control system was installed for inducing airflow to the three subsystems and for treating emissions as necessary to meet regulatory requirements of the state of Utah. Two rotary-lobe blowers provided the capability for extraction of up to 57 std m³/min (2000 ft³/min) of gas from the three subsystems. In order to protect against potential hazards presented by combustible gas mixtures, flame arrestors were installed at the inlet to each blower. The blowers were controlled by an automatic shutdown system based on a combustible gas detector. Two catalytic oxidation units were used for conversion of the jet fuel hydrocarbons to carbon dioxide and water before discharge into the atmosphere. The propane-fired units differed in the configuration of their catalyst beds, one having a fluidized-bed design and the other a fixed-bed design. The units were evaluated in terms of economics and reliability, as well as hydrocarbon destruction efficiency. A knockout drum, flowmeters, and gas monitors were also included in the system.

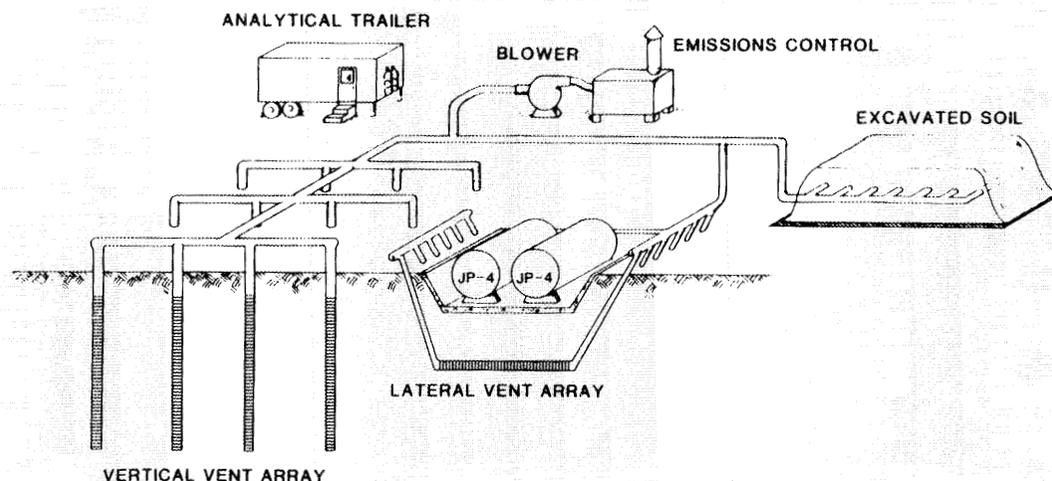


Fig. 2.17. Conceptual drawing of the in situ soil-venting demonstration system at Hill AFB.

The ORNL operation of the Hill AFB full-scale in situ soil-venting system began in December 1988. During the period of ORNL operation from December 16, 1988, to October 8, 1989, a total of ~47.7 Mg (105,000 lb) of hydrocarbons was extracted in ~4.8 million m³ (170 million ft³) of soil gas. During this period, the extracted soil gas concentrations decreased from an initial level of 38,000 ppm (38 g/kg) hexane equivalent to a final concentration range of 15 to 505 ppm (15 to 505 mg/kg) hexane equivalent for the vertical vents. The final average concentration of the gas extracted from the vertical vents was 157 ppm (157 mg/kg).

Figure 2.18 presents the hydrocarbon concentration measured in the extracted gas as a function of the extracted gas volume. The data are characteristic of soil-venting operations, with a rapid decrease in concentration from initially high hydrocarbon levels, followed by an extended period with a much more slowly decreasing removal rate. The curve shown in Fig. 2.18 results from a simple equilibrium model based upon Raoult's law. The initial spill size and measured hydrocarbon weight fractions are inputs to the model. Given the simplicity of the model, agreement with the experimental data is excellent. Thus, a simple model may be useful in prediction of a soil-venting system behavior for sites without significant mass transfer resistances. As operation progresses, deviation from the model is expected due to imperfect contact of contaminants and air flow, greater relative amounts of contaminants in less permeable soil zones, soil

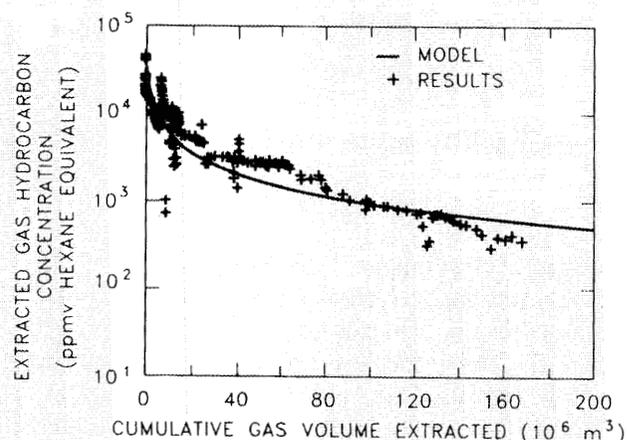


Fig. 2.18. Hydrocarbon concentration as a function of the extracted gas volume from soil venting, comparing results and the model.

moisture, bioactivity, contaminant sorption on soil, and other factors.

Further support for the equilibrium model may be seen from the relative weight fractions of hydrocarbons in extracted soil gas samples, as shown in Fig. 2.19. The results indicate a shift toward heavier compounds in the extracted gas as venting progressed. This shift agrees with the assumption of equilibrium volatilization of a contaminant mixture.

A potentially significant means of hydrocarbon removal due to in situ soil venting is enhanced biodegradation. The increased oxygen levels in the soil gas due to infiltration of atmospheric air may

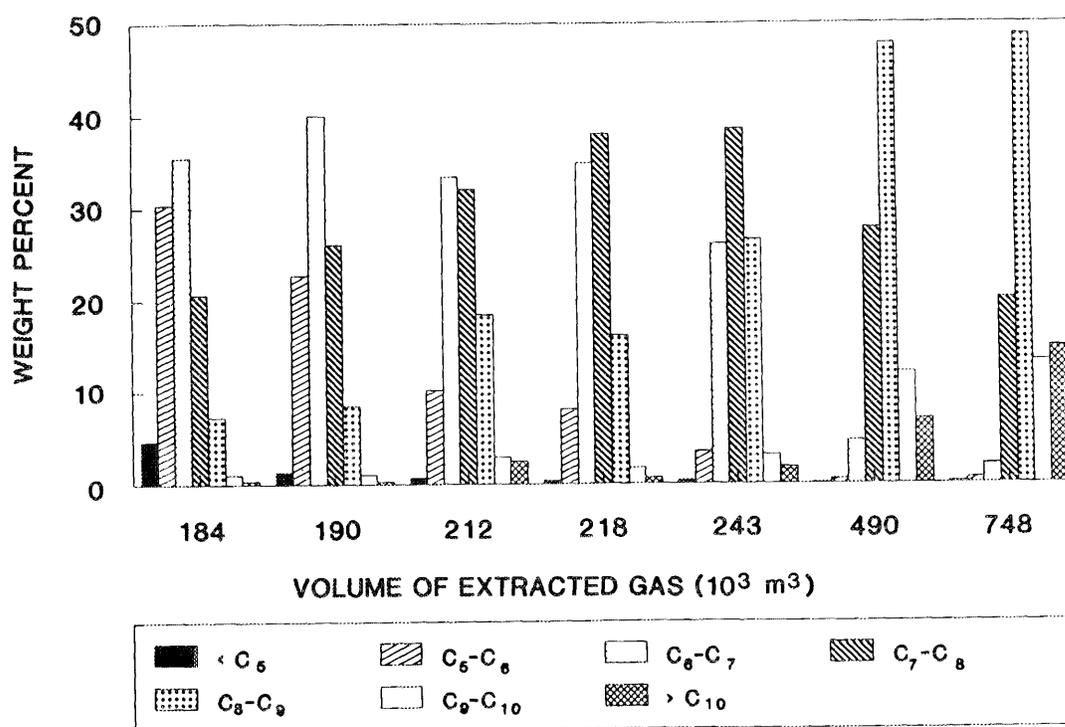


Fig. 2.19. Relative weight fractions of hydrocarbons in extracted soil gas samples from soil venting.

considerably stimulate biological activity. To evaluate this factor, carbon dioxide and oxygen were measured in the extracted gas. As seen in Fig. 2.20, high CO₂ (11%) and low oxygen (1%) levels were initially measured in the soil gas. As venting continued, the CO₂ levels decreased and oxygen levels increased. Carbon dioxide levels continued to be an order of magnitude higher than background. Integration of CO₂ concentrations up to June 1989 indicated that bioactivity contributed up to 28% of total hydrocarbon removal. These results are important because they show that biological activity may continue to be significant despite no addition of nutrients. Biodegradation may prove to be the means for decontamination of heavier compounds that are not readily volatilized.

The results obtained in the Hill AFB in situ soil-venting demonstration have shown that this technique is very effective for removal of large amounts of jet fuel from soil in a relatively short period of time. Soil samples that are to be taken after the termination of ORNL operation of the system will allow evaluation of the effectiveness of hydrocarbon removal to that point and will provide the basis for determination of site remediation economics. After the site characterization activities

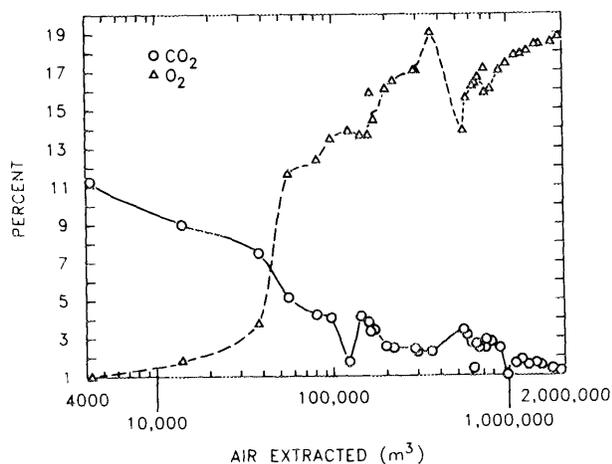


Fig. 2.20. Variation of oxygen and carbon dioxide concentrations in extracted soil gas from soil venting.

have been completed, the system will be operated by Hill AFB personnel for remediation of the site.

Publication of a literature review of soil-venting technology, a technical report of the demonstration, and a guidance manual for applicability of the technology at other sites is scheduled for early 1990.

Eglin AFB Air Stripping

Pump-and-treat remediation technologies are being used at two groundwater contamination sites at Eglin AFB, Florida. At the first site (denoted as "A-20" by the Air Force), a diesel fuel spill occurred, and the remediation treatment being used involves packed-tower air stripping followed by aqueous-phase granular activated carbon adsorption. The second site (Seventh Street BX) is a gasoline spill site, and the treatment is by packed-tower air stripping. At the A-20 site, the aqueous effluent is returned to the groundwater via exfiltration galleries. At the Seventh Street BX site, the air-stripper aqueous effluent is discharged to the local sewage treatment plant. The Air Force requested assistance from ORNL via HAZWRAP to analyze operation of both systems for 1 year, during which time data would be collected regarding system efficiency and operability.

Additional instrumentation and sample points were installed on the two air-stripping systems by ORNL in August 1988. After maintenance and operational testing procedures were completed by the CH2M-Hill Company, ORNL collected samples of the aqueous influent and effluent from the Seventh Street BX air stripper through October 1989. These samples were analyzed for VOCs. The compounds identified in the groundwater were primarily benzene, toluene, and xylenes. The air stripper demonstrated a high removal efficiency during the operational period, with no evidence of any adverse effect from iron deposition. The CH2M-Hill Company had identified several maintenance tasks which should be performed before starting up the A-20 unit. However, these tasks were not completed, and the A-20 air stripper was not operated during this period. A final report for this project will be completed by the end of 1989.

Environmental Evaluation of Air Force Fire-Training Pits

The Energy Systems Engineering Organization at ORNL had undertaken the design, construction, and testing of a new Environmentally Acceptable Fire-Training Facility (EAFTF) for the Department of the Air Force for the purpose of providing live fire-training capabilities without harming the environment. The specific goals of the EAFTF were to (1) eliminate contamination of area soil,

groundwater, and surface water by the loss of JP-4 jet fuel and fire suppressants; (2) provide enhanced training and improved safety for fire-fighting personnel; and (3) minimize water pollution impact by treatment, recycling, and reuse of resultant effluent/wastewater and recovered fuel. At present, EAFTFs are planned for the Davis-Monthan, Grand Forks, Mountain Home, and Tyndall AFBs. The purpose of Chem Tech involvement was to evaluate the wastewater treatment systems of the training facilities. The wastewater treatment system operates as a closed loop and consists of (1) the burn pit, (2) the fuel/water separator, and (3) the holding pond.

The burn pit is a specially prepared area used to contain the water, jet fuel, and fire suppressants used during a fire-training exercise. After a jet fuel fire has been extinguished, the remaining mixture of water, fire suppressants, and unburned jet fuel is drained and flushed to the fuel/water separator. The separator is a three-stage concrete unit designed to reduce the concentration of oils and greases to 25 mg/L in the effluent. The unburned fuel is skimmed off and held for later addition to the burn pit. The effluent from the separator is pumped to the holding pond. The holding pond is used to supply the water that is used to flush the burn pit following a training exercise.

The EAFTF studies, originally scheduled for Tyndall AFB, Florida, were performed at Davis-Monthan AFB, Arizona. The tests involved (1) taking a set of background samples at Davis-Monthan AFB, (2) allowing the Air Force to conduct a series of fire-training exercises, and (3) taking a set of samples from the fuel/water separator and the holding pond immediately following each training exercise to determine the effect the exercises had on the wastewater. In addition, the burn pit was sampled to determine if the waste remaining after the training exercises could be considered hazardous. The separator and holding pond were also allowed to sit undisturbed for a period of ~60 d. During this period, the separator and holding pond were sampled periodically to determine if biodegradation and/or natural stripping were occurring in the water.

The studies were completed, and a final report was submitted to the Air Force. The major conclusions from this study were (1) volatilization and biodegradation of dissolved organic components were ineffective in reducing organic concentrations, and (2) the burn pit would not be considered a RCRA hazardous waste site if the pit was properly

flushed after the completion of a series of fire-training exercises. Recommendations made to the Air Force included (1) modifying the flushing procedure in order to obtain a cleaner pit following the training exercises, (2) allowing a proper amount of time between training exercises to promote the separation of organics from the water, and (3) conducting bench-scale studies to determine methods to stimulate biological activity.

Evaluation of Catalytic Incinerator at Wurtsmith AFB

The objective of this project, which is a subtask to the "Air Stripping with Emissions Control" project, is to analyze, evaluate, and report on operating data from a full-scale catalytic destruction unit at Wurtsmith AFB, Michigan. The catalytic incinerator is used to control the gaseous emissions from an air-stripping unit that is removing TCE and DCE from groundwater. The evaluation will determine the destruction efficiencies of the preheater and the catalytic reactor. The catalytic destruction unit will also be studied to determine if there are any products of incomplete combustion in the effluent of the preheater or in the effluent of the catalytic reactor. The evaluation will include the collection of accurate costs on the operations and maintenance of the catalytic system. The project will require preparing a test plan, performing the tests described by the test plan, submitting monthly progress and cost reports, and producing interim and final reports.

The test plan prepared for this project was designed to obtain samples that will determine the destruction efficiency of the incinerator as a function of (1) the operating temperature and (2) the service time of the catalyst. The study will involve evaluation of a catalyst that has been in service for ~18 months and evaluation of a new batch of catalyst for comparison purposes. Sampling of the incinerator by EPA Method 30, known as the Volatile Organic Sampling Train (VOST), will be performed on an approximately quarterly basis over a period of ~20 months. The VOST collects VOCs on tenax and tenax/carbon traps. The organic compounds are removed from the traps by thermal desorption or by methanol extraction, depending on the anticipated amount of components to be adsorbed on the traps, and analyzed by gas chromatography. A total hydrocarbon analyzer will be used on-site to continuously monitor the

concentration of organics in the airstreams. Since the primary combustion products of chlorinated hydrocarbons are hydrochloric acid and chlorine, the Research Triangle Institute was subcontracted to sample for these components using a new method of analysis it had developed.

The first series of sampling has been completed, and the samples are being analyzed. It is anticipated that six more sampling series will be conducted to obtain the necessary data for the evaluation of the catalytic destruction unit.

Hazardous Waste Minimization at the Naval Ordnance Station (NOS) at Louisville, Kentucky

The NOS is an industrial facility that produces new ordnance hardware and reconditions hardware already placed in service. Operations in the NOS include (1) machine and tool shops, (2) a complete plating facility, (3) a variety of painting and stripping areas, (4) assembly areas, and (5) test and evaluation areas. Approximately 2300 m³ (600,000 gal) of wastewater per day is produced by these processes within the NOS. This wastewater is discharged to the local sanitary sewer, which is regulated by a local Metropolitan Sewer District (MSD). The MSD had established discharge limits for the NOS, and reports have indicated that the major source of wastewater (and primary area of concern) is the discharge from the industrial plating shop within the NOS. This plating shop generates ~1100 m³ (300,000 gal) of wastewater per day.

Because construction of a new plating shop was planned, complete renovation of the existing plating shop was not economically feasible. As a result, the Naval Energy and Environmental Support Activity (NEESA) was given the task of demonstrating the use of innovative wastewater treatment technology that would minimize the volume of wastewater generated from the plating processes and maintain compliance with the MSD discharge limits until the new plating shop is put into operation.

Chem Tech was contracted by NEESA to (1) analyze the problems at the NOS plating shop, (2) recommend solutions to these problems, and (3) implement these solutions.

Studies at NOS identified chromium as the major pollutant from the plating shop and identified the major sources of chromium contamination within the plating shop. Several improvements in the operation of the plating shop were implemented

which reduced the quantity of chromium lost from the plating shop by >85%. These improvements included (1) installation of a closed-loop rinsing system with zero aqueous discharge on the chrome plating line, (2) utilization of a "new bright dip formula" that contains ~75% less chromium than the previous formula, and (3) replacement of the dichromate deoxidizer on the chromate conversion line with a chromium-free deoxidizer.

With these improvements, the remaining major sources of chromium contamination are the rinse waters from the gun-barrel line and the bright dip line. A system consisting of reverse osmosis (RO) coupled with concentrate evaporation was recommended to treat these streams. The RO unit will meet NEESA's objective of demonstrating innovative wastewater treatment technology that will produce information useful for treating similar wastewaters at other naval facilities. Evaporation of the RO concentrate will maximize waste-volume-reduction-process flexibility.

Pilot-scale tests were conducted at NOS to evaluate the feasibility of RO treatment of these waste streams. The tests indicated that the rejection of chromium for both waste streams exceeded 95% at recoveries up to 95%. These tests indicated that the gun-line rise could be successfully treated, with minimal fouling, utilizing a 5- μ m prefilter. Some fouling of the membrane occurred when the RO unit operated on the bright dip rinse stream; however, cleaning the membranes with EDTA proved adequate for restoring the effectiveness of the membrane.

The full-scale system was designed to take into account the decrease in permeate flux that occurred when the RO unit operated on bright dip rinse waters. The full-scale unit was constructed at ORNL and transported to NOS. Chem Tech personnel started the full-scale unit, ensured that the unit was operating properly, and trained NOS personnel to operate the unit.

Environmental Surveys

A multidivisional team of engineers and scientists from the Chem Tech, Energy, and Environmental Sciences divisions is participating in the Environmental Compliance Assessment Program (ECAMP). ECAMP is a tool for the assessment of the compliance status of Air Force installations with federal, state, and local regulations, as well as with relevant DOD and U.S. Air Force directives and

regulations. The program consists of assessment protocols in every major environmental area: air, wastewater, solid waste, hazardous waste, pesticides, PCBs, drinking water, petroleum-based fuels and lubricants, hazardous materials, natural resources, and historic sites. Environmental assessment at each AFB consists of a week-long site evaluation by the ORNL team and a published comprehensive report concerning the AFB compliance status. The ECAMP is a proactive method for permitting the Air Force to assess compliance status and to correct any deficiencies. During this report period, environmental assessments were conducted by the ORNL team at Sonderstrom AFB, Greenland; Thule AFB, Greenland; Lowry AFB, Denver, Colorado; and Nellis AFB, Las Vegas, Nevada.

2.3 Advanced Reactor Systems

2.3.1 ANS

The ANS reactor is being designed to be the most advanced research reactor in the world. Chem Tech's heat removal studies provide thermal analyses of key components of the reactor. Both forced-convection and natural-circulation cooling modes are addressed.

Preconceptual core design activities (through early 1989) included the initial development of a suitable critical heat flux correlation, a reinterpretation of the rate at which oxide forms on the aluminum fuel-plate cladding, and calculation of the incipient-boiling power levels of a number of candidate core designs.⁵⁸⁻⁶⁰ Values were recommended for the initial core inlet pressure and for the minimum nominal coolant gap. A potential method of enhancing the rigidity of parallel-plate fuel assemblies was conceived and described.

Since selection of the preconceptual reference core design, experimental data for the critical heat flux in low-pressure, natural-circulation water systems have been recorrelated and applied to evaluations of decay heat removal. Additional correlations for single-phase, laminar-flow heat transfer rates in the ANS geometry and for single-phase friction coefficients for all flow regimes have been evaluated and recommendations made. The probable fuel-plate hydrodynamic roughness at beginning-of-cycle was evaluated, and a study was

begun of buoyancy effects on vertical-heated, single-phase, core-flow, heat transfer rates.

2.3.2 Developmental LWR Program

The Developmental Light-Water Reactor (DLWR) program is an ongoing activity in Chem Tech. Its objectives are to determine the feasibility of creating an LWR with major improvements in economics and radical improvements in safety. The program was initiated within Chem Tech because of a series of inventions by its staff.

A developmental reactor, by definition, refers to a reactor whose characteristics are advanced enough that a test or demonstration reactor may be necessary to prove feasibility. This implies initial application in the 2000 to 2020 time frame.

The DLWR is not a single reactor concept; it is a set of goals, including PRIME safety goals. PRIME is defined as:

1. Passive safety. The DLWR should be designed to have only passive safety systems (no electrical or moving mechanical components). This eliminates the potential for most mechanical failures and operator errors.
2. Resilient Safety. The DLWR safety and operating systems must be resilient (i.e., activate when required, but not during normal plant transients). Safety systems should not interfere with normal operations or create other incentives to be bypassed or disabled by operating and maintenance staffs.
3. Inherent safety. The DLWR safety systems should have inherent safety whenever possible; that is, materials and structural configurations should be selected to eliminate classes of accidents (such as those caused by chemical reactions) and their associated safety systems.
4. Malevolence resistance. The DLWR should be capable of passively withstanding malevolent acts of man (conventional off-the-shelf munitions and short-term power plant takeover by terrorists) without significant release of radionuclides to the environment. For public health and safety, plant security should depend primarily on passive rather than active (guards, security checks, etc.) techniques. In practice, this design objective also covers all operator errors and inaction.

5. Extended safety. DLWR safety should be ensured under the above conditions for extended time periods (>1 week) after an accident without human intervention.

Program Strategy

Any DLWR will be composed of structures, systems, and components (SSCs) that meet PRIME safety goals and are the building blocks for a power reactor. Various SSCs are designed to meet the functional requirements for plant operation or to prevent, stop, or mitigate a reactor accident. The program has three components to achieve its goals:

1. identify, invent, and develop power reactor SSCs that meet PRIME safety goals;
2. integrate SSCs into several mainline DLWR concepts; and
3. evaluate SSCs and reactor concepts to define preferred options.

Example DLWR

An example of a single DLWR concept (Fig. 2.21) can provide (1) an understanding of SSCs, (2) an example to clarify directions of research, and (3) a definition of how this activity relates to other programs.

The Process Inherent Ultimate Safety Boiling-Water Reactor (PIUS/BWR), which was invented by Chem Tech staff members, has a conventional nuclear reactor core and steam cycle [750 MW(e)] but a radically different set of safety systems.⁶¹⁻⁶³ These three systems are (1) the Prestressed Concrete Reactor Vessel (PCR/V), (2) the Fluidic In-Vessel Emergency Core Cooling System (FIVES), and (3) the Core Melt Source Reduction System (COMSORS).

In the PIUS/BWR, all reactor safety systems are located within a large PCR/V that protects the reactor from violent internal and external threats. This particular PCR/V design is based on work in Sweden for another type of DLWR.⁶⁴ The interior of the reactor vessel has a shape similar to that of a soft drink bottle with a diameter of 13.5 m and a height of 35 m. Vessel walls are 7 m thick. The PCR/V is held together with both prestressed steel cable and steel rebar, but it is designed so the loss of either cables or rebar will not result in vessel failure. Multiple design features ensure leak-tight integrity from the vessel. All pressure vessel

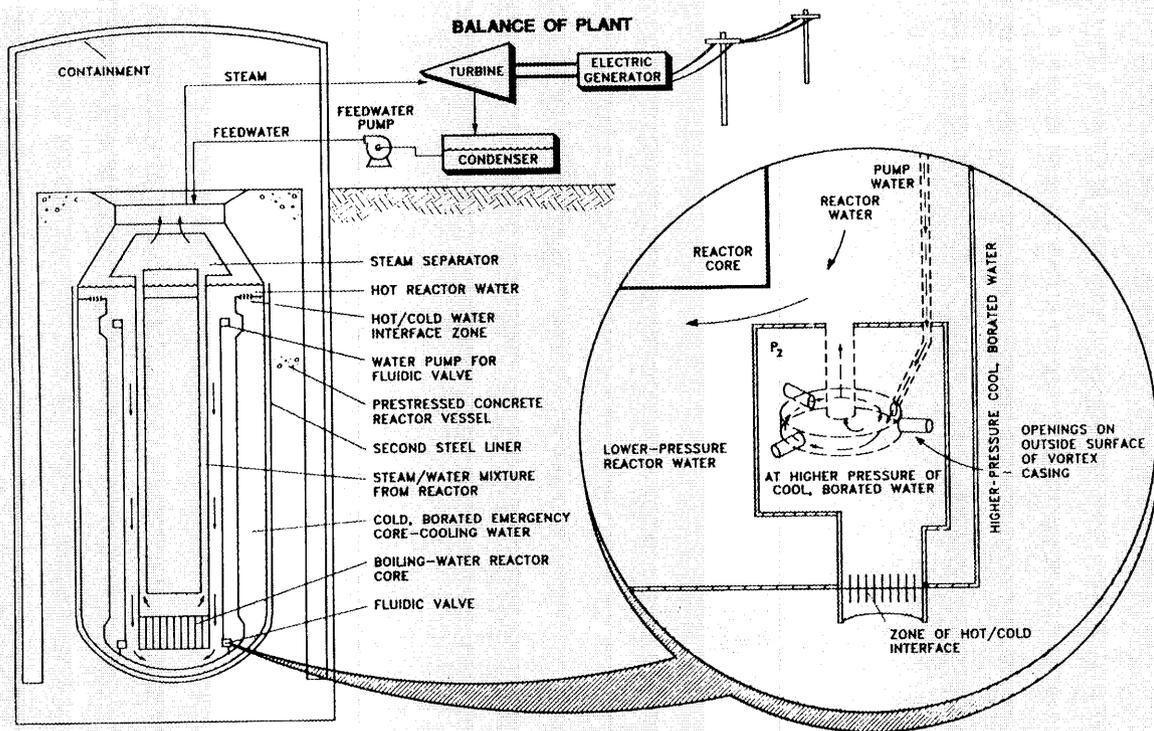


Fig. 2.21. Example of a Developmental Light-Water Reactor (DLWR) concept: The Process Inherent Ultimate Safety Boiling Water Reactor (PIUS/BWR).

penetrations are at the top of the vessel, and the vessel has multiple interior liners. From inside to outside the PCRV, the vessel wall includes (1) a steel liner, (2) 1 m of concrete, (3) a second steel liner, and (4) 6 m of concrete. Under severe accident conditions, the vessel is designed to depressurize (but not to leak water) by seal failure at the top of the PCRV.

The primary safety concern in an LWR is to prevent a reactor core meltdown. This can be done by keeping the reactor core underwater at all times. The PIUS/BWR uses an emergency core cooling system, called FIVES, which is, in part, based on earlier work on nuclear fuel reprocessing plants.

The inside of the PCRV is divided into two zones. The first contains the BWR primary system (clean primary coolant water, reactor core, downcomer, and steam separator). The second contains FIVES, which includes a fluidic valve, a hot/cold water interface zone, a small water pump, and sufficient borated water to shut down the reactor and cool it for 1 week by water boiloff.

If the reactor core is short of water, the borated water enters the reactor core, shuts down the

reactor, and removes heat by boiling off water. The borated water during normal operations is in contact with the reactor primary coolant at two locations. Near the top of the PCRV, the two water zones are in direct contact with each other through a hot/cold water interface; the cold, high-density, borated water is below the low-density, clean, hot water. Appropriate design reduces the diffusion of boron across this water interface zone. Near the bottom of the PCRV, a fluidic valve (a valve with no moving parts) separates cold borated water from hot reactor water. The valve remains closed only if it receives high-pressure water from a water pump high in the downcomer. If the reactor water is low or a power failure occurs, this pump fails; the result, then, is that no water is sent to the fluidic valve, the valve opens, and the reactor core is shut down and flooded with FIVES cool, borated water.

The central component of FIVES is the vortex fluidic valve assembly, which is a modified vortex fluidic amplifier operated as a valve. This arrangement is similar to a conventional centrifugal pump with a blocked exit line. The incoming FIVES water is injected tangentially at high

velocities into the vortex casing, causing the water to move in a circle. The centrifugal forces create higher pressures near the outside surface of the vortex valve casing and lower pressures near the inside. The outside surface has holes (short lengths of tubing) that connect it to a zone of clean, higher-pressure reactor water, which, in turn, is in contact with the borated water zone at the hot/cold water interface zone. The center of the vortex casing is connected to the downcomer and exhausts FIVES water to it. By adjusting FIVES water-pump output, pressures across the vortex valve can be made to match those of the two water zones. In effect, a valve exists that uses the dynamic forces of water rather than pieces of metal to prevent flow through the valve.

The hot/cold water interface that separates borated water from reactor coolant is located below the fluidic valve. Water temperature and boron concentration sensors determine the location of the interface zone. The sensors are used to control the speed of the FIVES water pump system during normal operation and, hence, the interface position.

A third proposed feature (SSC) of the PIUS/BWR is the COMSORS, which is designed to limit the maximum potential release of radioactivity to the environment after an accident by control of core meltdown chemistry. It provides a form of inherent safety by limiting maximum accident size. The system consists of carefully selected concrete aggregate under the reactor core. It is based on two observations:

1. Experiments and theoretical calculations in Chem Tech and at Sandia National Laboratories (NRC program) indicate that the quantity of radioactive vapor and aerosols released after a reactor accident depends on the chemical reactions between molten core and concrete. For example, if the concrete contains limestone aggregate, the molten core and concrete interactions generate carbon dioxide, which bubbles through the molten mixture, creating aerosols and stripping volatile fission products from the melt to the atmosphere.
2. Research on solidification of radioactive wastes from nuclear fuel reprocessing plants indicates that control of waste chemistry can encourage fission products to remain in the melt and minimize aerosols in the off-gas systems.

These diverse lines of research suggest that proper selection of aggregate (chemical composition) for concrete under the reactor core can

significantly reduce the release of radionuclides to the containment if a core melt accident occurs. This reduces the challenge to containment, thus decreasing the probability of containment failure. If containment failure occurs, the proper aggregate reduces radionuclide releases to the environment. It may also reduce containment costs if maximum accident pressures and temperatures in containment after an accident are reduced.

Activities During FY 1989

The specific accomplishments during this report period have included:

1. Completed *Proposed and Existing Passive and Inherent Safety-Related Structures, Systems and Components (Building Blocks) for Advanced Light-Water Reactors*.⁶⁵ This ORNL report was the major program activity in FY 1989. It is a compendium of LWR safety-related passive and inherent SSCs, the "building blocks" that combine to create a complete reactor design. The concepts described range from current commercial technologies to speculative ideas and include both chemical and mechanical SSCs. This report provides a basis to define future directions of research and documents the large base of technology available to create DLWRs with PRIME safety characteristics.
2. Invented or identified possible technologies to eliminate size restrictions for all DLWRs. Nuclear core instabilities at larger core sizes limit some designs of DLWRs to ~700 MW(e), not a concern because of different core designs and because design criteria allow the use of control rods to limit such instabilities. Because of the PRIME design criteria, control rods in DLWRs cannot be used for safety, although they are allowed for operations. Control rods as active components have potential failure modes and create possibilities for misoperation or sabotage. For this reason, it is desirable not to include them as a safety system in very advanced reactors.

As a part of the DLWR program, patent reports of possible subject invention have been filed on passive devices to limit nuclear reactor core instability problems. If these devices are practical, there are no theoretical reactor core size limits for DLWRs. This could potentially have a major impact on economics of DLWRs and creates the option of building such reactors

in any desired size to match utility interests. The devices may also have applications to other reactor types.

3. Invented or identified passive decay heat cooling system applicable to current and future LWRs. Patent reports of possible subject invention have been filed on a passive decay heat removal system applicable to all water-cooled reactors. This passive safety system would prevent core damage after a loss-of-heat-sink accident or after most small- and medium-size loss-of-coolant accidents. Such a system would prevent core damage in an accident such as the one at Three Mile Island. The system appears applicable to both near-term and more advanced reactor designs, and it can be applied to PWRs and BWRs.

Observations

The DLWR program is an exploratory research effort that has aggressive long-term goals. Program success will depend on cooperation with many divisions within ORNL, various national laboratories, universities, and private industry because the radical technical approaches being investigated for safety systems do not fit within traditional disciplines or divisions.

2.4 Integrated Data Base Program

The IDB is the official DOE data base for SNF and radioactive waste inventories and projections through the year 2020. The IDB Program is jointly sponsored by DOE's OCRWM, the Office of the Assistant Secretary for Defense Programs, and the Office of the Assistant Secretary for Nuclear Energy. The purposes of the IDB program are to create and to maintain a reliable baseline of quality data and information to be used for national program planning, decision making, and other management activities. As the official data base for DOE, the IDB is readily available to the DOE community and DOE-sponsored contractors.

The IDB program provides access to information on SNF and radioactive waste inventories and characteristics, including volume and mass, age, radioactivity, heat generation, chemical and physical properties, location, packaging, and

nuclide composition. The IDB, which uses a modular system of computer codes, also provides projections based on anticipated growth rates, schedules for new facilities, waste generation source terms, and treatment assumptions. Projections for commercial waste are based on official DOE/Energy Information Administration electrical growth projections.

2.4.1 Annual Inventory Report for 1989

The annual report of the IDB Program presents summary-level tables and figures that compare volumes and radioactivity levels of all forms of SNF and radioactive waste (HLW, TRU waste, LLW, and mixed waste). These materials are generated by commercial fuel cycles (including D&D activities), defense programs, remedial action programs, and institutional and industrial operations. *Integrated Data Base for 1988; Spent Fuel and Radioactive Waste Inventories, Projections and Characteristics*⁶⁶ was printed in September 1988, and over 1500 copies were distributed during 1989. Alternative forms of the same information were also generated and distributed, including a one-page foldout data summary and a PC version. The former provides an easy-to-find summary of the available data, and the latter gives a menu-driven data base that allows detailed tables and graphics to be generated.

The 1989 version of the annual inventory report was compiled and sent to printing in November 1989;³³ distribution is planned for December 1989. Tables 2.7 and 2.8, taken from this publication, show the kinds of data the report provides. Alternative data formats for this report are also being produced.

2.4.2 Mixed Waste Data Base

Recent initiatives by DOE/HQ have resulted in the decision to proceed with the incorporation of mixed waste data in the IDB program. Recent efforts by members of the IDB staff in support of the former five-site Martin Marietta Energy Systems Hazardous Waste Development, Demonstration, and Disposal Program led to a prototype Mixed Waste Data Base. An outline of a new chapter for the

Table 2.7. Spent fuel and radioactive waste inventories as of December 31, 1988

UMTRA = Uranium Mill Tailings Remedial Action; FUSRAP = Formerly Utilized Sites Remedial Action Project; SFMP = Surplus Facilities Management Program; GJRAP = Grand Junction Remedial Action Project

Waste category	TRU isotopes (kg)	Mass (MTHM)	Volume (m ³)	Activity ^a (10 ⁶ Ci)	Thermal power (10 ³ W)
Spent fuel (commercial)					
BWRs		6,756	3,215 ^b	5,179	18,900
PWRs		10,850	4,751 ^b	13,476	51,600
High-level waste					
Savannah River (DOE)			128,400	661	1,830
Idaho (DOE)			11,000	67	196
Hanford (DOE) ^c			243,500	446	1,307
West Valley (commercial)			2,130	29	87
Transuranic waste (DOE)					
Buried TRU waste	771		190,837	0.06	1.8
Stored TRU waste	2,055		59,681	3.88	66.6
Stored LLW ^d	14		36,123	c	c
Low-level waste (total area of all sites)					
DOE sites (262 ha) ^f			2,471,600	13.42	18.61
Commercial sites (74 ha)			1,298,500	4.79	18.09
Uranium mill tailings (commercial)					
Licensed mill sites ^g			116,900,000	c	c
Remedial action activities (DOE) ^h					
UMTRA Project (25 sites)					
Mill tailings and other waste (permanent storage)			4,915,500	c	c
FUSRAP (30 sites)					
LLW (permanent and interim storage)			187,760	c	c
Defense D&D Program (230 sites)					
TRU waste			i	c	e
LLW			i	c	e
Mill tailings			i	c	e
SFMP (civilian projects; 30 sites)					
TRU waste			i	e	e
LLW			i	e	e
Mill tailings			i	e	e
GJRAP (593 sites)					
Mill tailings			52,070 ^j	c	c
Reactor decommissioning			k	k	k
Mixed LLW					
DOE		70.2 ^l	52,140	c	c
Commercial		c	c	c	c
Miscellaneous, highly radioactive materials		252.5	c	c	e

^aActivity data are calculated values as of December 31, 1988.

^bIncludes volume of spacing between the fuel rods of each assembly.

^cAll tank waste has been declared HLW. However, this categorization could change after the work characterizing this waste has been completed.

^dTRU-contaminated waste in interim storage that will be managed as LLW after retrieval and assay for certification.

^eInformation not available.

^fha = 1 hectare = 10,000 m² or 2.47 acres.

^gIncludes contributions from 26 licensed mills. At the end of 1988, only three mills were active.

^hUnless otherwise indicated, inventories reported in this table for remedial action activities include only contributions from projects completed at the end of 1988.

ⁱShould include only inventories of wastes stored at remedial action sites. This information is currently not available.

^jMill tailings stabilized by the Grand Junction Remedial Action Project through 1982.

^kMost of this activity has involved small test reactors. The LLW collected to date from such small reactors are included in the LLW inventories listed above.

^lMass of mixed LLW is expressed in metric tons (t) and includes other elements in addition to heavy metals.

Table 2.8. Current and projected cumulative quantities of radioactive waste and spent fuel

UMTRA = Uranium Mill Tailings Remedial Action; FUSRAP = Formerly Utilized Sites Remedial Action Project; SFMP = Surplus Facilities Management Program; GJRAP = Grand Junction Remedial Action Project; WVDP = West Valley Demonstration Project

Source of material and type	End of calendar year			
	1988	2000	2010	2020
DOE/defense, 10 ³ m ³				
HLW				
Interim storage				
Glass ^a	383	323	310	306
TRU ^b	0	2.0	3.3	3.5
Buried				
Stored	191	191	191	191
LLW	60	90	115	c
Remedial action activities ^d	2,472	4,250	5,591	6,869
UMTRA Project and GJRAP				
Mill tailings and other waste ^{e,f}	4,968	29,613	29,613	29,613
FUSRAP				
LLW ^g	188	1,258	1,286	1,286
Defense D&D Program				
TRU waste	h	0.2	0.4	0.4
LLW ^g	h	54.5	64.4	64.9
Mill tailings	h	65.4	65.4	65.4
SFMP (civilian projects)				
TRU waste	h	0.9	1.1	1.1
LLW ^g	h	588.4	869.7	869.7
Mill tailings	h	1,529	1,529	1,529
Mixed LLW	52.1	c	c	c
Miscellaneous, highly radioactive materials, MTHM	252.5	c	c	c
Commercial, 10 ³ m ³				
LWR spent fuel, MTHM ⁱ (no reprocessing)				
No New Orders Case	17,607	40,100	58,100	75,100
Lower Reference Case	17,607	40,200	58,400	82,400
Commercial HLW glass (WVDP)	0.0	0.210	0.210	0.210
LLW (no reprocessing)	1,299	1,762	2,160	2,471
D&D (LLW) ^j				
Classes A, B, and C LLW		0.00	7.67	645.95
Greater-than-Class-C LLW		0.00	0.06	3.55
Mill tailings (no reprocessing)				
Mixed LLW	116,900	120,100	c	c
	c	c	c	c

^aIncludes projections for glass only at Savannah River Site.

^bProjections are updated mainly as a result of improvements in detection methods. Approximately 38% of the currently stored volume will be managed as LLW.

^cInformation not available.

^dThese activities involve remedial actions on existing wastes. Projections are based on the scheduled completion of most remedial action activities by the year 2000.

^eMill tailings stabilized from both GJRAP and UMTRA Project activities.

^fIncludes windblown contaminated soil and stabilization material from sites that may require remedial action.

^gProjected LLW volumes from remedial action activities are not included in the DOE/defense LLW volumes reported above.

^hShould include only inventories of wastes stored at remedial action sites. This information is currently not available. Projected data that follow are cumulatives from future remedial action activities.

ⁱHistorically, spent fuel has been measured in units of mass (MTHM) rather than units of volume.

^jProjected D&D wastes from light-water reactors shut down after 1988. Wastes collected from historical D&D of reactors are included in the LLW inventories listed above.

annual IDB inventory report was presented to the IDB Steering Committee at its January 1988 meeting as a prototype for inclusion in the 1989 edition of the annual IDB report. This prototype was also factored into a national HAZWRAP proposal for acquiring mixed waste data for the annual report. The IDB program staff was, and continues to be, highly involved in various quality assurance aspects of acquiring and manipulating the mixed waste data. Two mixed waste tables from this year's annual report are shown in Tables 2.9 and 2.10. It is anticipated that this effort will continue in future years and, in fact, be expanded to include purely hazardous wastes from the DOE sites as well as the currently reported mixed wastes.

Mixed HLW and TRU wastes are not included in the mixed waste data base but are included in the HLW and TRU waste inventories and

projections. This data base does not consider the unique features of mixed HLW and TRU wastes because their dominating radioactive characteristics alone dictate the methods by which these materials need to be treated, handled, stored, and disposed of. In contrast, the hazardous characteristics of mixed LLW are subject to RCRA.² Thus, the treatment, handling, and disposal of these wastes are subject to the regulations of the EPA as well as to those of DOE and NRC.

Typically, mixed waste at DOE sites includes a broad spectrum of contaminated materials, such as air purifiers, cleaning solutions and cleanup materials, engine oils and grease, epoxies and resins, gravel, laser dyes, paint residues, photographic materials, soils, asphalt, roofing compounds and wall materials, water treatment chemicals, and decommissioned weapons manufacturing equipment.

Table 2.9. Cumulative volume (m³) inventories of DOE site mixed LLW, by physical category, through 1988^a

Site	Solid	Liquid	Gas ^b	Sludge	Other ^c	Total
PAD ^d	1,032.5	36.1	0	0.6	0	1,069.2
ORGDP ^d	942.4	445	0	9,040.3	0	10,427.7
PORTS ^d	130.3	164.3	0	1,885	0	2,179.6
ORNL ^d	3.3	84.1	<0.1	0	0	87.4
Y-12 ^d	7,556.4	129.7	0	3,185	0	10,871.1
HANF ^{d,e}	715	593	0	0	69.6	1,377.6
SRS	492.9	8,525.4	0	5,680	0	14,698.3
LLNL	42.7	86.7	0	0	0	129.4
SNLL	0.2	1.23	0	<0.01	0	1.43
LANL	44.5	136.6	0.5	32.8	<0.1	214.4
RFP	9,349.2	6.3	0	0	0	9,355.5
MOUND	5.6	42.5	0	0	0	48.1
PANT	83	3	0	0	0	86
SNL	68.5	1.2	0	0.3	0	70
Allied-Signal	2.65	0	0	0	0	2.65
ITRI	0	28.8	0	0	0	28.8
Pinellas	0	0	0	0	6.47	6.47
INEL	33.6	9.7	0	3	0	46.3
NTS	0	0	0	0	22.1	22.1
FMPC ^f	217	54	0	0	0	271
Ames ^f	0	0	0	0	0	0
ANL-E ^f	37.7	34.9	0	0	0	72.6
ANL-W ^f	13.23	0	0	0	0	13.23
BNL ^f	0	1,061	0	0	0	1,061
Total	20,770.68	11,443.53	0.5	19,827.0	98.17	52,139.88

^aMaterials may be in interim storage awaiting treatment. PAD = Paducah Gaseous Diffusion Plant; ORGDP = Oak Ridge Gaseous Diffusion Plant; PORTS = Portsmouth Gaseous Diffusion Plant; Y-12 = Y-12 Plant; HANF = Hanford Reservation; SRS = Savannah River Site; LLNL = Lawrence Livermore National Laboratory; SNLL = Sandia National Laboratories, Livermore; LANL = Los Alamos National Laboratory; RFP = Rocky Flats Plant; MOUND = Mound Plant; PANT = Pantex Plant; SNL = Sandia National Laboratories; ITRI = Inhalation Toxicology Research Institute; INEL = Idaho National Engineering Laboratory; NTS = Nevada Test Site; FMPC = Feed Materials Production Center; ANL-E = Argonne National Laboratory-East; ANL-W = Argonne National Laboratory-West; BNL = Brookhaven National Laboratory.

^bStored in cylinders.

^cA mixture of two or more of the solid, liquid, gas, or sludge categories.

^dDensities of 1000 kg/m³ for liquids and 1500 kg/m³ for solids, sludges, and others were assumed to calculate volumes.

^eHANF inventories are 1987 and 1988 data only.

^fData for these sites were provided by a DOE/IIQ/DP effort in support of a study on land disposal restrictions (LDRs) recently imposed on the DOE. LDRs require phased-in treatment of hazardous mixed wastes before disposal.

Table 2.10. Volume generation rates (m³/year) of DOE site mixed LLW, by physical category, for 1988^a

Site	Solid	Liquid	Gas ^b	Sludge	Other ^c	Total
PAD ^d	187.3	11.0	0	0	0	198.3
ORGDP ^d	72.3	46.6	0	2.0	0	120.9
PORTS ^d	91.3	29.5	0	35	0	155.8
ORNL ^d	1	21.2	<0.1	0	0	22.2
Y-12 ^d	445	1,663.5	0	15.4	0	2,123.9
HANF ^d	373.4	4.8	0	0	0	378.2
SRS	192.3	2,702.3	0	1,800	0	4,694.6
LLNL	15.8	155.5	0	0	0	171.3
SNLL	<0.1	0.43	0	<0.01	0	0.53
LANL	16.1	49.6	0.1	19.3	0	85.1
RFP	3,104.1	0.4	0	0	0	3,104.5
MOUND	1	3	0	0	0	4
PANT	95	3	0	0	0	98
SNL	73.8	1.6	0	0.6	0	76
Allied-Signal	0.42	0	0	0	0	0.42
ITRI	0	1	0	0	0	1
Pinellas	0	0	0	0	0	0
INEL	10.8	6.9	0	3	0	20.7
NTS	0	0	0	0	0	0
FMPC ^e	15	30	0	0	0	45
Ames ^e	0.5	0	0	0	0	0.5
ANL-E ^e	1	786	0	0	0	787
ANL-W ^e	1.3	0	0	0	0	1.3
BNL ^e	0	267	0	0	0	267
Total	4,697.52	5,783.33	0.1	1,875.3	0	12,356.25

^aValues do not reflect any treatment that may, or will, occur prior to interim storage. PAD = Paducah Gaseous Diffusion Plant; ORGDP = Oak Ridge Gaseous Diffusion Plant; PORTS = Portsmouth Gaseous Diffusion Plant; Y-12 = Y-12 Plant; HANF = Hanford Reservation; SRS = Savannah River Site; LLNL = Lawrence Livermore National Laboratory; SNLL = Sandia National Laboratories, Livermore; LANL = Los Alamos National Laboratory; RFP = Rocky Flats Plant; MOUND = Mound Plant; PANT = Pantex Plant; SNL = Sandia National Laboratories; ITRI = Inhalation Toxicology Research Institute; INEL = Idaho National Engineering Laboratory; NTS = Nevada Test Site; FMPC = Feed Materials Production Center; ANL-E = Argonne National Laboratory-East; ANL-W = Argonne National Laboratory-West; BNL = Brookhaven National Laboratory.

^bStored in cylinders.

^cA mixture of two or more of the solid, liquid, gas, or sludge categories.

^dDensities of 1000 kg/m³ for liquids and 1500 kg/m³ for solids, sludges, and others were assumed to calculate volumes.

^eData for these sites were provided by a DOE/IHQ/DP effort in support of a study on land disposal restrictions (LDRs) recently imposed on the DOE. LDRs require phased-in treatment of hazardous mixed wastes before disposal.

References

1. U.S. Congress, The Nuclear Waste Policy Act of 1982, Pub. L. 97-425, January 7, 1983.
2. U.S. Congress, Resource Conservation and Recovery Act of 1976, Pub. L. 94-580, Oct. 21, 1976, as amended by the Hazardous and Solid Waste Amendments Act of 1984, Pub. L. 98-616, November 9, 1984.
3. L. B. Shappert et al., *Transportation Operations Functions of the Federal Waste Management System*, ORNL/TM-10811 (ORO/TOP-5403.0), Oak Ridge National Laboratory, June 1988.
4. Oak Ridge National Laboratory, Quality Assurance Plan for Drop Testing, QAP-X-89-WMRD, 001, May 12, 1989.
5. Nuclear Assurance Corporation, *Facility Interface Capability Assessment Project Interim Topical Report*, ORNL/Sub/86-97393/5, to be published at Oak Ridge National Laboratory.
6. E. R. Johnson Assoc., Inc., *The Incentives and Feasibility for Direct Measurement of Spent Nuclear Fuel Characteristics in the Federal Waste Management System*, ORNL/Sub/86-SA094/3, Oak Ridge National Laboratory, August 1989.

7. *Report of System Integration Modeling System (SIMS) Exercise*, DOE/RW, draft, March 1989.
8. *Characteristics of Spent Fuel, High-Level Waste, and Other Radioactive Wastes Which May Require Long-Term Isolation*, DOE/RW-0184, Vols. 1–6, December 1987, and Vols. 7–8, June 1988.
9. A. G. Croff, *ORIGEN2—A Revised and Updated Version of the Oak Ridge Isotope Generation and Depletion Code*, ORNL-5621, Oak Ridge National Laboratory, July 1980.
10. R. S. Moore, D. A. Williamson, and K. J. Notz, *A Classification Scheme for LWR Fuel Assemblies*, ORNL/TM-10901, Oak Ridge National Laboratory, November 1988.
11. E. R. Johnson and K. J. Notz, *Shipping and Storage Cask Data for Spent Nuclear Fuel*, ORNL/TM-11008, Oak Ridge National Laboratory, November 1988.
12. R. S. Moore and K. J. Notz, *Physical Characteristics of GE BWR Fuel Assemblies*, ORNL/TM-10902, Oak Ridge National Laboratory, June 1989.
13. K. J. Notz, T. D. Welch, and R. J. Andermann, *ORIGEN2 Sensitivity to Enrichment and Other Factors*, ORNL/TM-11333, Oak Ridge National Laboratory, in press.
14. R. Salmon and K. J. Notz, *Non-LWR and Special LWR Spent Fuels: Characteristics and Criticality Aspects of Packaging and Disposal*, ORNL/TM-11016, Oak Ridge National Laboratory, December 1989.
15. M. J. Bell, *ORIGEN—The ORNL Isotope Generation and Depletion Code*, ORNL-4628, Oak Ridge National Laboratory, May 1973.
16. A. F. McBride et al., *Compendium of Technical Computer Codes Used in Support of the DOE Office of Civilian Radioactive Waste Management*, ORNL/TM-10919, Oak Ridge National Laboratory, April 1989.
17. R. M. Westfall, H. R. Dyer, and L. M. Petrie, *Assessment of Criticality Computational Software for the U.S. Department of Energy Office of Civilian Radioactive Waste Management Applications*, ORNL/CSD/TM-247, Oak Ridge National Laboratory, October 1988.
18. C. V. Parks et al., *Assessment of Shielding Analysis Methods, Codes, and Data for Spent Fuel Transport/Storage Applications*, ORNL/CSD/TM-246, Oak Ridge National Laboratory, July 1988.
19. P. T. Williams, et al., *Assessment of Thermal Analysis Software for the DOE Office of Civilian Radioactive Waste Management*, ORNL/TM-10940, Oak Ridge National Laboratory, July 1989.
20. A. D. Kelmers et al., *Identification and Evaluation of Radionuclide Generation/Depletion Codes for Potential Use by the Department of Energy's Office of Civilian Radioactive Waste Management*, ORNL/TM-10404, Oak Ridge National Laboratory, February 1989.
21. K. J. Riggle and J. W. Roddy, *A Preliminary Assessment of Selected Atmospheric Dispersion, Food-Chain Transport, and Dose-To-Man Computer Codes for Use by the DOE Office of Civilian Radioactive Waste Management*, ORNL/TM-10915, Oak Ridge National Laboratory, February 1989.
22. Oak Ridge National Laboratory, *ORIGEN-Type Code Work Quality Assurance Plan*, QAP-RD-011-R1, January 1989.
23. O. W. Hermann et al., *Multicode Comparison of Selected Source-Term Computer Codes*, ORNL/CSD/TM-251, Oak Ridge National Laboratory, April 1989.
24. M. C. Brady, O. W. Hermann, and W. B. Wilson, *Comparison of Radiation Spectra from Selected Source-Term Computer Codes*, ORNL/CSD/TM-259, Oak Ridge National Laboratory, April 1989.
25. M. C. Brady, O. W. Hermann, and W. B. Wilson, "Comparison of Radiation Sources from Selected Point Depletion Codes," presented at the ANS/ENS International Conference, Washington, D.C., October 30–November 3, 1988.
26. J. W. Roddy and J. C. Mailen, *Radiological Characteristics of LWR Spent Fuel: A Literature Survey of Experimental Data*, ORNL/TM-10105, Oak Ridge National Laboratory, December 1987.

27. R. E. Malenfant, *QAD: A Series of Point-Kernel General-Purpose Shielding Programs*, LA-3573, Los Alamos Scientific Laboratory, Los Alamos, NM, April 1973.
28. W. A. Rhoades and R. L. Childs, *An Updated Version of the DOT 4 One- and Two-Dimensional Neutron/Photon Transport Code*, ORNL-5851, Oak Ridge National Laboratory, April 1982.
29. S. B. Ludwig and J. P. Renier, *Standard- and Extended-Burnup PWR and BWR Reactor Models for the ORIGEN2 Computer Code*, ORNL/TM-11018, Oak Ridge National Laboratory, to be published.
30. U.S. Congress, The Nuclear Waste Policy Amendments Act of 1987, Pub. L. 100-203, Title V, Subtitle A, December 22, 1987.
31. G. W. Strandberg, T. L. Donaldson, and L. L. Farr, "Degradation of Trichloroethylene and trans-1,2-Dichloroethylene by a Methanotrophic Consortium in a Fixed-Film, Packed-Bed Bioreactor," *Environ. Sci. Technol.* **23**, 1422-1425 (1989).
32. S. B. Garland et al., *The Use of Methanotropic Bacteria for the Treatment of Groundwater Contaminated with Trichloroethylene at the Department of Energy Kansas City Plant*, ORNL/TM-11084, Oak Ridge National Laboratory, November 1989.
33. U.S. Department of Energy, *Integrated Data Base for 1989: Spent Fuel and Radioactive Waste Inventories, Projections, and Characteristics*, DOE/RW-0006, Rev. 5, Oak Ridge National Laboratory, November 1989.
34. R. C. Stewart et al., *Remote-Handled Transuranic Solid Waste Characterization Study: Oak Ridge National Laboratory*, ORNL/TM-11050, Oak Ridge National Laboratory, June 1989.
35. U.S. Department of Energy, *Radiation Protection of the Public and the Environment*, DOE Draft Order 5400.XX, Washington, DC, March 18, 1988.
36. *Preliminary Conceptual Design Report for the Waste Handling and Packaging Plant*, X-OE-453, Oak Ridge National Laboratory, May 1989.
37. M. Benedict, T. Pigford, and H. Levi, *Nuclear Chemical Engineering*, McGraw Hill, New York, 1981, p. 576.
38. H. O. Weeren, "Sluicing Operations at Gunite Waste Storage Tanks," ORNL/NFW-84/42, Oak Ridge National Laboratory, September 1984.
39. D. G. Thomas, "Significant Aspects of Non-Newtonian Technology," *Progress in International Research on Thermodynamic and Transport Properties*, ASME, New York, 1962, p. 669.
40. R. L. Faulkner et al., *D&D Five-Year Plan*, K/SS-523, Oak Ridge Gaseous Diffusion Plant, March 1989.
41. R. L. Faulkner et al., *D&D Strategy Paper*, Oak Ridge Gaseous Diffusion Plant, September 1989.
42. U.S. Department of Energy, *General Design Criteria Manual*, DOE Order 6430.1A, Washington, DC, April 6, 1989.
43. R. R. Rawl et al., *Oak Ridge National Laboratory Transportation System Performance Improvement Project: Final Report*, Oak Ridge National Laboratory, June 1989.
44. U.S. Department of Energy, *Hazardous Material Packaging for Transport*, DOE Order 1540.2/ORO, Washington, DC, September 30, 1986.
45. B. W. Welles, *Transportation and Packaging Bulletin*, Martin Marietta Energy Systems, November 1989.
46. E. D. Collins et al., "Analysis of Data from Leaching Concrete Samples Taken from the Three Mile Island Unit 2 Reactor Building Basement," *Nucl. Technol.* (December 1989).
47. B. Z. Egan, *Using Inorganic Membranes to Separate Gases: R&D Status Review*, ORNL/TM-11345, Oak Ridge National Laboratory, November 1989.

48. B. W. Moran et al., *Safeguards and Security Recommendations for the OCRWM Federal Waste Management System*, K/ITP-102, August 1988.
49. B. W. Moran et al., *A United States Sponsored Working Paper on Criteria for the Termination of International Atomic Energy Agency Safeguards on Nuclear Materials in Waste*, K/ITP-273, 1989.
50. C. W. Forsberg et al., *AVLIS Technology Significance Study Part 2: Description of Systems*, K/ITP-169, November 1989.
51. U.S. Department of Energy, *Applied Research, Development, Demonstration, Testing, and Evaluation Plan (Review Draft)*, November 1989.
52. C. W. Forsberg et al., *Historical Perspective, Economic Analysis, and Regulatory Analysis of the Impacts of Waste Partitioning—Transmutation on the Disposal of Radioactive Waste* (September 30, 1988).
53. A. G. Croff, J. O. Blomeke, and B. C. Finney, *Actinide Partitioning—Transmutation Program Final Report. I. Overall Assessment*, ORNL-5566, Oak Ridge National Laboratory, June 1980.
54. J. T. Bell et al., *Draft: Evaluation of Conceptual Flowsheets for Incorporating LWR Fuel Materials in an Advanced Nuclear Fuel Cycle* (1989).
55. U.S. Department of Energy, *Safety Requirements for Packaging and Transportation of Hazardous Materials*, DOE Order 5480.3, Washington, DC, July 1985.
56. U.S. Department of Energy, *Project Managements System Requirements*, DOE Order 4700.1, Washington, DC, 1988.
57. U.S. Department of Transportation, "Quality Control for Construction of Packaging," *Code of Federal Regulations*, 49 CFR Pt. 173.474, Oct. 1, 1987.
58. W. R. Gambill and T. Mochizuki, "ANS Design: Burnout Heat Flux Correlation Development," *Amer. Nucl. Soc. Trans.* **57**, 98–300 (October 1988).
59. W. R. Gambill and J. H. Lienhard, "An Upper Bound for the Critical Boiling Heat Flux," *J. Heat Trans.* **111**(3), 815–818 (August 1989).
60. W. R. Gambill, "A New Interpretation of Oxide Formation Rates and Thickness Limits," *Report of the ANS Aluminum Cladding Corrosion Workshop, Nov. 26–27, 1988, Idaho Falls, Idaho*, ORNL/CONF-8811203, Oak Ridge National Laboratory, February 1989.
61. C. W. Forsberg, "A Process Inherent Ultimate Safety Boiling Water Reactor," *Nucl. Technol.* **72**, 121 (February 1986).
62. C. W. Forsberg, "Passive Emergency Cooling Systems for Boiling Water Reactors," *Nucl. Technol.* **76**, 185 (January 1987).
63. C. W. Forsberg, "Boiling Water Neutronic Reactor Incorporating Process Inherent Safety Design," U.S. patent 4,666,654, May 19, 1987.
64. K. Hannerz, "Adapting the LWR to Future Needs: Secure P (PIUS)," *Power Eng.* **89**, 54 (September 1985).
65. C. W. Forsberg et al., *Proposed and Existing Passive and Inherent Safety-Related Structures, Systems, and Components (Building Blocks) for Advanced Light-Water Reactors*, ORNL-6554, Oak Ridge National Laboratory, December 1989.
66. *Integrated Data Base for 1988: Spent Fuel and Radioactive Waste Inventories, Projections, and Characteristics*, DOE/RW-0006, Rev. 4, Oak Ridge National Laboratory, September 1988.

3. Waste Management Technology Center

During this report period, the Waste Management Technology Center (WMTc) provided technical support to several Martin Marietta Energy Systems, Inc. (Energy Systems) sites and major program areas; conducted research and development (R&D) in several waste management areas; managed and conducted demonstrations; and organized information-exchange workshops.

3.1 Technical Support to Programs

The WMTc provided management assistance to two major programs: the Low-Level Waste Disposal Development and Demonstration (LLWDDD) Program and the Hazardous Waste Development, Demonstration, and Disposal (HAZWDDD) Program. Engineering and technical support was provided to the Toxic Substances Control Act (TSCA) Incinerator Operations. Technical assistance was given to the Oak Ridge Gaseous Diffusion Plant (ORGDP) and the Y-12 Plant for development of programs to involve the private sector in waste management efforts. Technical support was also provided to the Paducah and Portsmouth Gaseous Diffusion Plants to facilitate site preparation of plans ensuring compliance with the DOE Order 5820.2A¹ directive. The WMTc continued its management support role for Chem Tech in the ORNL Historically Black Colleges and Universities/ Minority Educational Institutions (HBCU/MEI) Program.

3.1.1 LLWDDD

The WMTc/Waste Management Technology Support Group (WMTSG) provided the program management and the management of major work elements of the LLWDDD Program. The purpose of the LLWDDD Program is to develop new and improved waste disposal facilities for the management of low-level solid wastes generated on the Oak Ridge Reservation (ORR). The task is needed both because current disposal capacities are being exhausted and because improved waste management practices are required to satisfy regulatory groups.

The scope of the LLWDDD Program encompassed the following:

1. developing and securing approvals for a low-level waste (LLW) management strategy for the ORR;
2. assisting sites in the development of site-specific LLWDDD strategy implementation plans and assisting the Central Waste Management Office (CWMO) by reviewing these plans and integrating them into an overall LLWDDD plan;
3. defining the functional requirements for new disposal facilities (to include characteristics of waste streams or waste packages to be routed to the disposal facilities, a schedule for needed new disposal capacity for various waste streams or waste packages, and performance requirements for disposal facilities);
4. siting new disposal facilities and satisfying National Environmental Policy Act² requirements for disposal facilities;
5. conducting demonstrations of treatment and packaging, storage, and disposal technologies to reduce technical and economic uncertainties associated with deployment of new disposal facilities;
6. supporting improved LLW management operations conducted by waste management organizations at plants on the ORR (to include assistance in the design of disposal facilities, development of monitoring plans, and evaluation of performance); and
7. preparing new sites and constructing new disposal facilities for operation by waste management organizations at plants on the ORR.

The LLWDDD Program was involved with an Energy Systems task team in evaluating the feasibility of combining the Y-12 and ORNL waste disposal line-item projects and having one organization responsible for the design, construction, and operation of the new LLW disposal facilities for the ORR. The task team consisted of waste management and engineering

representatives from the three ORR plants, a representative from the CWMO, and one from the LLWDDD core program. The task team presented its results to DOE/Oak Ridge Operations (DOE/ORO), and the proposed organizational changes took place in May 1989. As a result of the new organization, the LLWDDD core program became part of the Reservation Waste Management Division (RWMD), and the tasks were reoriented to accomplish the mission of the new division. The RWMD reports to the manager of the ORGDP.

3.1.2 HAZWDDD Waste Development

Chem Tech provided technical and managerial support to the Energy Systems Environmental and Safety Activities Department in the year-long preparation of the HAZWDDD Program Plan, which was written to ensure that Energy Systems' needs for treatment and disposal of all of its hazardous and mixed wastes have been identified and corrective actions planned. A multifaceted approach to developing and implementing this plan is given in this report, including complete plans for each of the five installations and an overall integrated plan. This plan is in response to DOE/ORO directives and provides information useful in addressing DOE Order 5820.2A¹ concerning waste management.

The HAZWDDD Plan accomplishes the following: (1) provides both background and organizational information; (2) summarizes the 402 hazardous and mixed-waste streams from the 5 installations by grouping them into 13 general waste categories; (3) presents current treatment, storage, and disposal capabilities within Energy Systems; (4) develops a management strategy by outlining critical issues, presents flow sheets describing management schemes for problem waste streams, and builds on the needs identified; (5) outlines specific activities needed to implement the strategy developed; and (6) presents schedule and budget requirements for the next decade.

The HAZWDDD Program addresses current and future technical problems and regulatory issues and uncertainties. Because of the nature and magnitude of the problems in hazardous and mixed-waste management, substantial funding will be required. Delays in funding the identified activities

will end in increased costs resulting from the need for additional storage capacity and from potential punitive actions by regulating agencies. Implementation of the HAZWDDD strategy, as developed in this document, ensures that decisions concerning hazardous and mixed-waste management are made in a coordinated and integrated manner among the installations and that these decisions have sound technical bases.

Since its issue in December 1988, the HAZWDDD Plan has been used as a building block for the privatization efforts that Energy Systems has undertaken to place treatment projects out for contract. The HAZWDDD Plan has been the basis for several short courses at Roane State Community College, and we have participated in providing lectures for these courses. The HAZWDDD Plan was highlighted in presentations at the 1988 DOE Model Conference in Oak Ridge and at the WATtec meeting held in Knoxville in February 1989.

3.1.3 Hazardous Waste Technology Program (HWTP) Overview

The HWTP has grown from a single \$250,000 project in FY 1986 to a \$10 million program in FY 1990. This program, which matches the needs of various federal sponsors with the waste and environmental researchers and engineers at Oak Ridge, will continue to aggressively expand as DOE and Congress place greater emphasis on the environmental restoration, waste management, and technology development issues. The HWTP has become the "clearinghouse" for hazardous waste technology research and development in the Energy Systems research divisions.

The HWTP serves as the point of contact from Oak Ridge to the Hazardous Waste Remedial Action Program (HAZWRAP) Support Contractor Office of DOE and interfaces closely with the newly evolving Office of Technology Development on waste issues. Research projects sponsored by HAZWRAP during the review period include paramagnetic separation of wastes, bioremediation of methyl mercury, bioremediation of polychlorinated biphenyl- (PCB-)contaminated soil, bio-

remediation of trichloroethylene- (TCE-)contaminated water, and development of simple analytical chemistry sampling tubes. Demonstrations conducted include testing of several processes to treat mixed-waste-contaminated groundwater and a process to substitute a nontoxic water-based metal cleaner for the historic TCE process. Several expanded R&D and demonstration projects are also scheduled to begin in FY 1990.

For research sponsors such as the U.S. Air Force, the Navy, or Environmental Protection Agency (EPA), the HWTP can coordinate a team effort that will harness the necessary technical strengths of the various divisions to solve a particular waste problem. Working across organizational boundaries, the HWTP is responsible for ensuring that waste research results are of high quality and responsive to the sponsors' needs. This integrated approach has proved especially useful in solving hazardous waste problems, which tend to be complex and frequently require the expertise of researchers from several disciplines. The HWTP also handles the business aspects and much of the "red tape" for waste management projects, helping to prepare the extensive documentation required to allow the sponsored work to be done.

3.1.4 ORGDP Support

The WMTC provided engineering and technical support to the ORGDP in the following areas: (1) process engineering support in the startup and operation of the K-1435 TSCA Incinerator, and (2) the development of the ORGDP plan for privatizing the potential treatment and disposal of the plant's wastes.

TSCA Incinerator Support

Since May 1989, at ORGDP's request, the WMTC provided experienced staff familiar with waste management technology operations to assist the TSCA incinerator staff with the operation of the K-1435 TSCA mixed-waste incinerator. Technical assistance has been and is continuing to be provided in the following areas:

1. Process engineering support for the startup, testing, and operation of the incinerator.
2. Development, design, and implementation of a computer-based management information system and the burn plans for the operation of the

incinerator. The computer system, called the TSCA Incinerator Management Information System, when fully operational, will perform, for example, the following functions for the incinerator:

- scheduling and tracking the wastes to be incinerated from the moment they are destined for the TSCA incinerator until their eventual destruction;
- defining incinerator operating parameters for different incineration campaigns, recognizing the many permit limits placed on the operation, the variety of wastes to be incinerated, and the capabilities of the physical plant; and
- preparing, on demand, a multitude of reports on the incinerator operations, such as environmental reports required by the regulatory agencies, management reports, waste destruction certificates, process data on the secondary wastes generated during the incineration operation, and preventive maintenance schedules for the incinerator to ensure optimum operations.

3.1.5 Y-12 Support

The WMTC has provided and continues to provide technical support to the Y-12 Plant in developing and demonstrating treatment technologies from the private sector for treating the large volume of mixed-waste contaminated soils and sludges generated at the Y-12 Plant. These soils and sludges are predominantly contaminated with depleted uranium, mercury, and/or organics that preclude their disposal in a Class I industrial landfill. The technical support consists of preparing requests for proposals (RFPs) that invite the private sector to recommend and demonstrate treatment technologies that will separate the radioactive and/or hazardous constituents from the soils and sludges, thereby permitting the treated soils/sludges to be disposed of in an industrial landfill or in radioactive disposal sites that are in compliance with all applicable environmental regulations. The reduced volume of the removed radioactive and hazardous constituents can be disposed of separately in compliance with all applicable environmental regulations.

Working in close cooperation with the Y-12 Plant Waste Management Division, the following

actions have been completed on this project during the reporting period.

1. The problem was scoped to define its magnitude, the nature and size of the treatment demonstrations, and the desired contaminant removal levels for the soils and the sludges to permit their disposal in an industrial landfill.
2. RFPs were issued inviting the private sector to submit competitive bids to demonstrate technologies that could treat the contaminated soils to the desired treatment level.
3. After the responses had been evaluated, contracts were let to three private sector companies to demonstrate their proposed treatment processes.
4. Activities are currently under way by these companies to demonstrate their treatment processes.

The results from these demonstrations will be used to define the waste treatment process for the contaminated soil.

A similar methodology is being followed to develop the treatment option for the toxic metals-contaminated sludge.

3.1.6 MEIs

Chem Tech is proactive in developing joint R&D projects with MEIs, and the WMTC has coordinated the division efforts. During this reporting period, progress has been made in collaboration with three universities.

North Carolina A&T State University

Two collaborative studies in waste management were continued between North Carolina A&T State University and ORNL during FY 1989. One study focused on the application of systems optimization as a waste management tool, and a paper on systems optimization for nuclear waste disposal was presented at the Seventh International Conference on Mathematical and Computer Modeling,³ which was held in Chicago. The second study focused on the theory and evidence for economies of scale in the development of waste management systems. A paper from this study was presented at the DOE Model Conference in Oak Ridge.⁴

Southern University

The WMTC is supporting a project being conducted at the Southern University and A&M College in Baton Rouge, Louisiana, studying the in situ treatment of contaminated soils using air stripping. The project is being performed by a graduate student in partial fulfillment of a master's degree in environmental science. The project scope consists of: (1) conducting a literature review of past air-stripping activities to develop an experimental investigation plan; (2) building an experimental system to study air stripping of different types of soil; (3) and conducting experiments to develop correlations for extracting TCE and toluene from soils, using air as the stripping medium.

Florida International University (FIU)

A contract has been given to FIU to assist in the evaluation of the In-Tank Evaporation (ITE) Process. The objective of the ITE project is to concentrate the liquid low-level waste (LLLW) currently stored in the Melton Valley Storage Tanks (MVSTs) by sparging with dry air. The ITE process has the potential to remove ~568 m³ (150,000 gal) of water from the stored LLLW before the startup of the Waste Handling Pilot Plant (WHPP) process, thus allowing adequate storage space for future LLLW generated at ORNL.

FIU has been charged with the development of a performance model of the ITE process. Accomplishments to date have included a detailed bubble model, complete heat and mass transfer models, and a detailed analysis of heat losses from the MVST vault. These programs are packaged in a user-friendly, menu-driven personal computer program. Publications resulting from the work are being reviewed at ORNL.

3.2 Technology Development

The Waste Technology and Development Research Group of the WMTSG conducts developmental studies to solve waste problems. This is primarily a laboratory group that supports various programs in the areas of waste-form development and separations sciences. The group is located in

Building K-1006 at ORGDP and has a close and mutually beneficial working relationship with the Quality and Technical Services Division there. This group has evolved over the past 20 years, from the solidification of high-level waste (HLW) to a multiprogram, broadly based technology development role, with projects in HLW, LLW, hazardous and organic waste, and municipal waste disposal. Recently, this broad effort has been recognized by various sponsors, and we now work in close coordination with the EPA Risk-Reduction Engineering Laboratory, the DOE/Richland Operations Office, the Energy Systems LLWDDD Program, Air Force sponsors, and private sector sponsors looking for waste-form development. This group has submitted several proposals to the new DOE Environmental Restoration and Waste Management Office of Technology Development Research, Development, Demonstration, Testing, and Evaluation. The future for this group is bright, and significant expansion is envisioned within the next 2 years.

Following is a review of several programs that the group supported during the report period. Hanford Support is a long-term activity to help solve the most pressing problem at the DOE Hanford Reservation. The work on immobilization of volatile organic compounds (VOCs) is a recent project designed to develop and report work on the "cutting edge" of waste immobilization and has received significant attention in the waste-form development technical community. The Weldon Springs study showed the capabilities of this group in waste form development. The Waste Immobilization Technology (WIT) Program is a new way of doing business for DOE. The program studies and reports the performance of several waste forms developed by private vendors. The work is vital in ensuring that the best technology is finally selected for treatment of DOE wastes. Then, several innovative, smaller studies, such as a novel encapsulation method, are discussed.

3.2.1 Hanford Grout Support Studies

ORNL has been involved in the Hanford Grout Disposal Program (HGDP) since 1981. The HGDP will provide the technology and equipment for disposal of the low-level fraction of radioactive liquid double-wall tank waste in a cement-based

grout. The equipment design and construction were completed in FY 1988. ORNL was previously involved in the Transportable Grout Facility (TGF) design and construction effort, which is now complete, engineering investigations at Pacific Northwest Laboratory, and waste formulation development work performed at ORNL. ORNL efforts during this reporting period focused on identifying methods to reduce the heat generation of a current grout formula and developing a grout testing meter. These projects are discussed in more detail in the following paragraphs.

Grout Calorimetry

ORNL provided assistance to the Westinghouse Hanford Company (WHC) in identifying methods to reduce the heat generation of double-shell slurry feed (DSSF) grouts. The current DSSF grout formulation generates $\sim 372 \text{ MJ/m}^3$ ($\sim 10,000 \text{ Btu/ft}^3$) of grout. Practical transportable grout equipment and vault-filling operations require a heat generation of 223 MJ/m^3 ($< 6,000 \text{ Btu/ft}^3$) of grout.

Isothermal calorimetric data obtained at 35, 50, 60, and 80°C showed that individual blend components (i.e., fly ash and granulated blast-furnace slag) exhibited different total heat releases based on material source. A material characterization via X-ray diffraction (XRD), scanning electron microscopy (SEM), energy-dispersive X-ray spectroscopy (EDS), electron spectroscopy for chemical analysis (ESCA), and 24-h leach indicated that the observed differences did not result from surface reactions. XRD characterization of the slags found a higher gypsum content in the slag exhibiting the lowest total heat release, indicating that partial hydration may have occurred during sample shipping and handling. One of the most interesting observations from the study occurred when the total heat releases at 60 and 80°C from grouts prepared with waste and tap water were compared. The comparison revealed that there was a synergistic effect between the waste constituents and temperature, which affected the rate of heat release but not the total heat release.

Although additional data are needed to quantify the principal factors contributing to the excessive heat and, hence, identify an optimum solution, the data do provide the basis for three recommendations to control the heat release. The three recommendations are (1) evaluating the material source to determine which raw materials can

contribute to lower heat release; (2) partially pre-hydrating the raw materials to reduce the heat generation in the grout product; and (3) enhancing the formula by substituting fly ash for the more heat-generating component, granulated blast-furnace slag. Each of these recommendations needs additional study for optimization, but at present, all appear to be viable options to facilitate heat reduction.

Grout Meter Development

Limited work at ORNL during this reporting period identified the potential for developing a meter that could be used to continuously monitor grout density during TGF operations. The development effort involved the use of a pumping loop in conjunction with the grout meter to study the effects of mix ratio, waste composition variations, and run time. This grout meter, which has been patented by ORNL, consists primarily of (1) sensors, presently based on the electrical resistance/capacitance of the grout; (2) a recording device, and (3) calibration standards. The grout meter is made of polyvinyl chloride (PVC) in the shape of a right circular cylinder with alternating rings of PVC and metal. The metal rings serve as the electrodes, and the PVC rings serve as spacers between the electrodes. Metal screws contacting the metal rings through the side of the meter provide the means to induce a current and obtain readings. Thus, this design uses the metal rings, which are flush with the pipe walls, as the "probe" and thereby eliminates the need for a probe protruding into the grout. Collectively, data obtained to date show that electrical response is linear with respect to mix ratio over a wide range of operating conditions. In addition, no visible signs of caking were observed during the 24-h operation period. Thus, this simple instrument, which can be readily installed in most existing grout facilities, can be used for improved process control and can be cleaned at the end of two 8-h shifts by simply flushing with water.

The Energy Systems Technology Transfer Group has recognized this invention as being useful to industry and has made it available for licensing.

Hanford Technical Support for Waste Receiving and Processing Facility

WMTSG staff provided consulting services to WHC concerning low-level, remote-handled,

transuranic, and hazardous waste processing systems that pertain to the Waste Remedial Action Program (WRAP) facility; participated in design reviews as requested; and monitored national and international technology development in solid waste treatment and disposal.

During FY 1989, sites were visited in West Germany, Sweden, and Finland in support of this effort.

WRAP is part of the triparty agreement among the EPA, DOE, and the state of Washington. The facility is scheduled to become operational in the late 1990s.

Efforts are under way to exchange solid waste technology information with other DOE sites.

3.2.2 In Situ Immobilization of Volatile Organic Compounds

In 1988, a study to evaluate the technical feasibility of grouting technology as a potential remediation option for the Zone 1 National Priorities List site at Robins Air Force Base (RAFB) in Warner Robins, Georgia, was initiated at ORNL through HAZWRAP. The objective of the 2-year study was to establish the technical and regulatory credibility of grouting technology as it would be applied to the sludge lagoon at the RAFB Zone 1 site.

The sludge lagoon and Landfill No. 4 are contiguous sites located adjacent to a swampy area on the east side of RAFB. The landfill, which occupies ~18.2 ha (45 acres), was operated from 1965 through 1978 and was used for disposal of general refuse and occasional disposal of industrial wastes. The sludge lagoon, located adjacent to the north side of the landfill, was used for disposal of industrial wastewater treatment plant sludges and occasional disposal of other liquid industrial wastes from 1968 until 1978. The lagoon was unlined and bordered by an elevated earthen dike. The lagoon surface area covered ~.61 ha (1.5 acres), and it was ~1.8 m (6 ft) deep.

Site investigations to date indicate that inorganic contaminants are present in the lagoon leachate. Organics, including VOCs, have been detected in the lagoon leachate, as well as in the groundwater underlying Zone 1. The in situ stabilization option is economically attractive because the cost of implementation is estimated to be ~\$5 million. In comparison, incineration was

estimated to require a \$10 million to 20 million capital investment and generated a hazardous side stream (i.e., the ash).

In our study, the technical performance of the waste forms was evaluated mainly from the standpoint of the VOC immobilization potential. Immobilization in this sense means retention of VOC or retardation of release to a leachant. Grout formulas and materials were obtained from four vendors believed to be representative of available commercial technology. Grout products were then prepared and evaluated for their suitability in retaining VOCs. The results of this evaluation involved several subtasks: (1) conducting a screening study to provide guidance to the final experimental design, (2) developing laboratory quality assurance and quality control procedures to quantify the VOC retention during sample handling and preparation, (3) evaluating grout product performance with regard to leachability and selected physical integrity criteria, and (4) evaluating the compatibility of commercially available processing equipment with regard to site regulatory criteria and cost.

One of the major criticisms of immobilization of VOC in cementitious waste forms was that most or all of the VOC escapes during mixing of the grout. A major effort in this study was estimating the amount of VOC each sample retained after mixing and curing. Emphasis in this area makes this study unique in the technical validity and credibility of the data obtained and its subsequent interpretation.

The results of this study indicate that grouting technology is a viable option for remediating the RAFB sludge lagoon and should be kept as an action option in the ongoing regulatory procedures for closing the site. Data show that commercially available waste forms can (1) obtain a leachability index⁵ of at least 7 for acetone, 1,2-dichloroethylene, chloroform, methyl ethyl ketone, TCE, benzene, perchloroethylene, and chlorobenzene; and (2) pass the EPA Toxicity Characteristics Leaching Procedure test for sludge concentrations representative of the RAFB sludge lagoon.

3.2.3 Weldon Springs Studies

In 1956, the Atomic Energy Commission acquired ~89 ha (220 acres) of the original Weldon Spring Ordnance Works property located in St.

Charles County, Missouri, from the Department of Army for use as the Weldon Spring Uranium Feed Materials Plant (WSUFMP). The WSUFMP operated between 1957 and 1966, processing uranium ore concentrates and recycled scrap to produce pure uranium trioxide, uranium tetrafluoride, and uranium metal. An average of 14.5 Gg (16,000 tons) of uranium materials were processed at this plant every year. In addition, thorium ore concentrates were also processed. These operations generated several chemical and radioactive waste streams, including raffinate streams from the refinery operation and magnesium fluoride slurry streams (washed slag) from the uranium recovery process. These streams were slurried to four raffinate pits, designated as raffinate pits 1, 2, 3, and 4, where the solids settled out and the supernatant liquids drained to the plant process sewer that drained off-site to a natural drainageway and ultimately to the Missouri River. The solids remaining in the pits consist of silica and other insolubles associated with the yellow cake ore feed materials, along with hydroxides and other precipitates formed from the pH neutralization of the raffinates with lime. Washed slag residues from the uranium metal production operation were also discharged to the pits. These pits contain an estimated 170,000 m³ (6,000,000 ft³) of waste sludge, including ~136 Mg (150 tons) of uranium and 68 Mg (75 tons) of thorium. DOE has the responsibility for maintenance of the entire site, including the 210-ha (518.9-acre) portion that contains the four raffinate pits.

Solidification/stabilization technology has the potential to provide a regulatory-acceptable and cost-effective remedy for hazardous and/or LLW sites. ORNL conducted a study to assess the applicability of cement-based solidification/stabilization technology as a remedial action option for the Weldon Spring Raffinate Pits 1, 2, 3, and 4.

Three dry-solids blends were evaluated: (1) Blend A, consisting of 20 wt % Type II Portland cement and 80 wt % American Society for Testing Materials (ASTM) Class F fly ash; (2) Blend B, consisting of 40 wt % Type II Portland cement and 60 wt % ASTM Class F fly ash; and (3) Blend C, consisting of 60 wt % Type II Portland cement and 40 wt % ASTM Class F fly ash. The blends were combined with the raffinate pit sludge at mix ratios (grams of dry-solids blend per gram of waste) of 0.4, 0.6, and 0.8 g/g, respectively.

Waste forms were prepared with composite waste samples from pits 1, 2, 3, and 4 as well as

with composite samples diluted to 20 wt % solids content. All of the waste forms prepared with Blends B and C met the performance criteria of (1) no drainable water within 28 d, (2) unconfined compressive strength of 60 psi, and (3) resistance to thermal cycling. Volume increases (vs the original waste) were consistent for all three blends at any one mix ratio and were 22, 32, and 40 vol % at mix ratios of 0.4, 0.6, and 0.8 g/g, respectively.

Collectively, the data indicate that the rate of set as determined by penetration resistance, drainable water, and unconfined compressive strength can be controlled by minor changes in the dry-solids blend composition or mix ratio. In addition, the effects of waste solids content on these properties can be controlled in the same manner. Thus, these grouts (Blends B and C), using no proprietary additives, can accommodate expected variations in the waste composition or future, more stringent performance criteria by the use of minor process operating changes (i.e., blend composition or mix ratio), which are well within the capability of standard commercially available technology.

Based on the available data, it is recommended that grouts prepared with Blend B at a mix ratio of 0.6 g/g be used as a reference formula for preliminary design and evaluation purposes. Waste forms prepared with this formula would be characterized by

1. weight increase (vs the original waste) of 60%;
2. volume increase (vs the original waste) of 32%;
3. no free liquid within 21 d;
4. penetration resistance of 4000 psi within 14 d; and
5. unconfined compressive strength >200 psi.

3.2.4 WIT Program

The WIT Evaluation Program was a subtask of the LLWDDD Program. The major objective of the WIT effort was to evaluate waste forms generated by stabilization/solidification of selected waste sludges generated by wastewater treatment operations within Energy Systems. The product waste forms should be suitable for interim storage for periods up to 10 to 15 years and disposal in a land-based facility. The WIT program sought to identify the physical requirements for solidified waste forms to be acceptable for land disposal and the appropriate testing protocol to measure waste-form characteristics. These performance criteria

would establish standards for solidified waste forms destined for land disposal on the ORR.

The WIT program consisted of three phases, with the first two phases consisting of screening studies to be performed before the development of a full-scale demonstration. Phase 1 involved the testing of laboratory-scale specimens using surrogate waste formulas representative of a generic family of specific waste sludges. Phase 2 was the bench-scale evaluation of specimens containing actual waste, using processes that had been found to be acceptable under Phase 1 testing. Phase 3 was the large-scale demonstration treating a statistically acceptable volume of actual waste using a process demonstrated to be acceptable in Phase 2.

Phase 1 of the WIT effort was completed before the cancellation of the program, and the results of the evaluation program will be published. Commercially available technologies using cement and bitumen as binding agents immobilized waste sludges in solidified waste forms. The capability of test protocols to measure performance and the ability of these waste forms to meet a selected performance criteria were evaluated. The testing protocol was based on regulatory requirements and suggested guidelines to assess the performance characteristics of the waste forms. The waste form characteristics measured included mechanical integrity, chemical stability, radiation resistance, biodegradation resistance, homogeneity, corrosivity, and leachability. In evaluating the product waste forms, it was found that the specified performance criteria were not sufficient to distinguish between the various products. More stringent performance criteria are required to rank the waste form products. A revised testing protocol is recommended to evaluate waste-form performance that will (1) generate defensible estimates of characteristics needed for the assessment of waste-form performance, and (2) reduce the costs associated with the evaluation program.

3.2.5 Development of a New Polymer-Impregnated Grout

A U.S. patent on a novel process for unifying a modern-day polymer, such as polystyrene, with a cement-based grout without the need to first remove or evacuate water was granted DOE in May 1989,

and patent rights have been waived to Energy Systems for technology transfer purposes. Work is in progress with two prospective licensees interested in using the new process to more efficiently immobilize ion-exchange media in cement-based grout. Test programs are in progress by these prospective licensees, and Energy Systems staff members aid in outlining the direction of tests at the company laboratories.

A simple way to effect polymer entry into materials, such as dense cement-based grout containing liquid waste following curing of the grout, has been developed and has potential applications using other materials and various polymers. The method, which has rendered alkaline, cement-based grout impervious to attack by even concentrated acid, is still being evaluated. Entry is based on the use of pore-entry particles that open a network of channels inside very dense materials despite the presence of water, facilitating rapid and complete entry by a polymer. The method requires no special equipment and appears to be easily adaptable to practical industrial scale-up. A schematic diagram is shown in Fig. 3.1 in which the possible use of the pore-entry technique is used with cement-based grout containing an aqueous-based waste.

3.2.6 Technology Transfer

Grouting technology has long been the dominant disposal option for LLW. It is rapidly

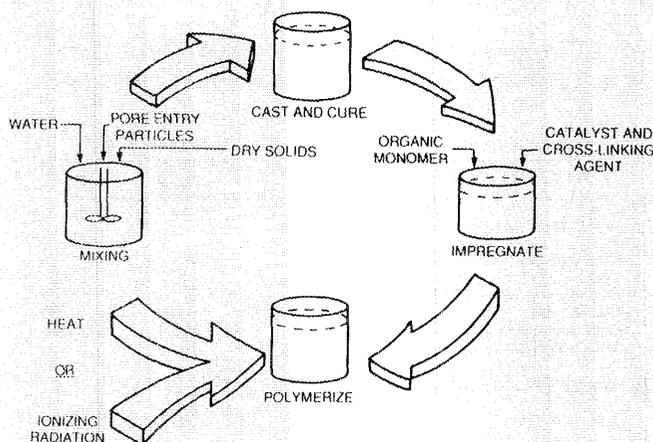


Fig. 3.1. A schematic process diagram showing the use of the polymer entry technique in the waste immobilization industry.

emerging as a principal disposal/remediation option for hazardous wastes as well. Division personnel have become the focal point for technology transfer between researchers involved in this technology from the hazardous waste and nuclear waste communities.

Division personnel working in concert with EPA and ASTM have established a symposium series specifically designed to provide a forum for technology transfer between the two scientific communities. The first symposium, held in Atlanta, Georgia, had 64 presentations, representatives from 11 countries, and an attendance of 250. The "Second International Symposium on Stabilization/Solidification of Hazardous, Radioactive, and Mixed Wastes," is to be held in Williamsburg, Virginia, May 29 through June 1, 1990.

This technology transfer effort has led to a greater utilization by EPA of expertise within Chem Tech. Most recently, division personnel were invited to contribute to a document that is to be issued by the EPA Risk Reduction Engineering Laboratory in Cincinnati, Ohio. The document is intended to provide guidance on assessing the suitability of the technology as a disposal/remediation option.

3.3 Demonstrations

In pursuit of one of its principal founding objectives, the WMTC continues to actively initiate, develop, manage, and conduct demonstrations of innovative processes and operations for the treatment of hazardous and/or radioactively contaminated wastes. To the extent possible, these demonstrations are designed to seek and evaluate the efficacy of innovative solutions from the private sector for treating and managing these types of wastes that exist at the DOE sites. During this reporting period, the WMTC was actively involved in the following projects:

- Y-12 Sludge Detoxification Demonstration
- International Technology (IT) Corporation's Thermal Treatment of Soils Demonstration
- Groundwater Treatment Demonstration
- Solvent Substitution Demonstration

Further details on the above demonstrations follow.

3.3.1 Y-12 Sludge Detoxification Demonstration

The Sludge Detoxification Demonstration was conducted for HAZWRAP by the WMTC at ORNL. This is the first technology demonstration project selected by HAZWRAP and involves demonstration by a private-sector contractor, Chem-Nuclear Systems, Inc. (CNSI), of technology for the treatment of mixed-waste sludges. These sludges result from the treatment of various wastewater streams at the Y-12 Plant and ORGDP.

The technology demonstrated involved an innovative combination of thermal treatment of the sludges followed by fixation of the dried solids in a cement-based grout matrix. A thermal separation process is employed in which the sludges are indirectly heated in an inert atmosphere, driving off the water and organic contaminants and leaving a dry-solids residue behind. The volatilized components are transferred with a nitrogen carrier gas to an off-gas system where they are condensed and collected for further processing. A liquid separation is then performed on the condensed liquids from the thermal unit to separate the organic phase from the aqueous phase. The plan is to return the cemented waste form and the organic phase to Energy Systems for disposal. The waste form is expected to eventually be "delisted" under Resource Conservation and Recovery Act⁶ (RCRA) provisions. Fig. 3.2 is a block-flow diagram of the treatment process.

The project was initiated in June 1987, and a summary of the FY 1988 activities was published.⁷

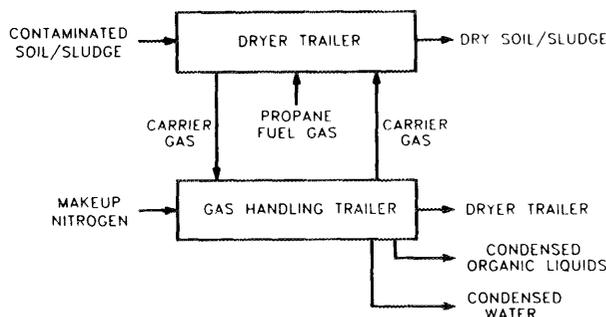


Fig. 3.2. Diagram of treatment process.

The X*TRAX® process equipment was brought to the demonstration site at ORGDP on August 4, 1988. Upon receipt of approval from DOE/ORO to proceed, the demonstration was conducted from September 27, 1988, through October 23, 1988. During this period, the 24 drums of the K-1232 treated sludge were successfully processed. However, while processing the Central Pollution Control Facility (CPCF) sludge, the off-gas condenser in the gas-handling trailer plugged after operating for 2.5 h. Efforts were made to correct the problem; however, they were not successful. A decision was then made to not process any additional CPCF sludge but to continue to process the K-1232 treated sludge. Summary data are given in Table 3.1 on the results of the demonstration.

The closeout of the demonstration was initiated by arranging for the treatment and the disposal of the demonstration-related wastes. In addition, CNSI and Energy Systems staff prepared immobilized samples of the dry residue and submitted those to the ORGDP Analytical Chemistry Department to perform the leaching tests in accordance with RCRA⁶ protocols. The RCRA leaching tests are currently being conducted. The final project report will be prepared following analysis of the leaching test results.

3.3.2 IT Corporation Thermal Treatment of Soils Demonstration

IT Corporation and Energy Systems participated in a joint-venture testing program to demonstrate

Table 3.1. Summary data on the Y-12 sludge detoxification demonstration

	K-1232 ^a	CPCF ^b
<u>Feed sludge characteristics</u>		
Total solids (wt %)	29.0	40.3
Density (g/ml.)	1.16	1.21
Oil and grease content (wt %)	0.7	10.2
Phenols (ppm)	54,000	6,300
Other semivolatiles (ppm)	3,525	5,752
Tetrachloroethene (ppm)	2.4	5.9
<u>Treated dry-residue characteristics</u>		
Total solids (wt %)	99.95	98.8
<u>Removal efficiencies</u>		
Oil and grease (%)	86.3	74.4
Phenols (%)	99.6	92.9

^aArea K-1232 at ORGDP.

^bCentral Pollution Control Facility.

the removal of organics (PCBs) from soils also contaminated with uranium and technetium. The testing program involved treating soils from DOE's Portsmouth and Paducah Gaseous Diffusion Plants. The IT-developed process used a rotary-kiln dryer to heat the contaminated soils and to drive off the organics, leaving a residue that contains inerts, radionuclides, and nonvolatile metals. The equipment layout of the process is shown in Fig. 3.3. The testing program was conducted at the IT facility on Bear Creek Road in Oak Ridge, Tennessee, in August 1988, with the final report being completed in November 1988.

Qualitatively, the demonstration showed that the process effectively removed PCBs and mercury from the contaminated soil. However, it was concluded that additional testing would be required for a quantitative evaluation of the process.

3.3.3 Groundwater Treatment Demonstration

The objective of this project is to demonstrate treatment processes to remove the radioactive and hazardous constituents from groundwater found on DOE/ORO sites operated by Energy Systems. Multiple fixed-price contracts will be awarded by Energy Systems to firms that meet the qualification criteria to conduct off-site pilot-scale demonstrations of several different innovative treatment processes (such as chemical, mechanical, and/or biological treatment). The project objective is to identify and evaluate groundwater treatment technologies that will be acceptable to the regulatory agencies, the public, and DOE.

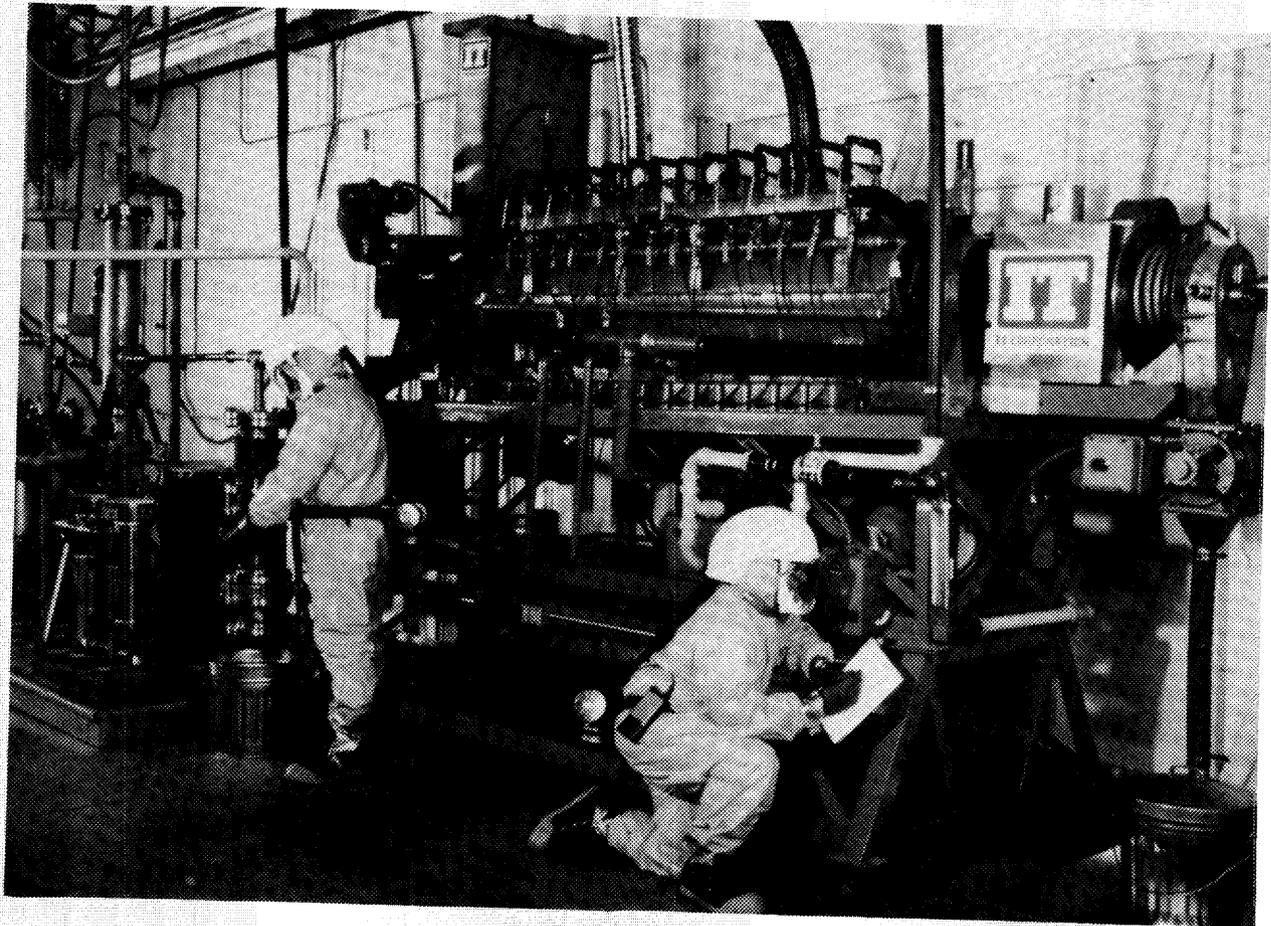


Fig. 3.3. IT Thermal Separation Process.

The groundwater treatment demonstrations are expected to provide the following information useful to groundwater cleanups at DOE sites:

1. evaluation of the performance and capabilities of the selected groundwater treatment methodologies provided by private-sector subcontractors,
2. comparison of costs for the removal of the hazardous and radioactive components from groundwater,
3. identification of problems associated with groundwater treatment, and
4. evaluation of volume of waste produced by the treatment technology.

The technologies demonstrated as part of this project are expected to be directly applicable to solving similar groundwater contamination problems known to exist at numerous DOE and Department of Defense facilities and Comprehensive Environmental Response, Compensation, and Liability Act sites and should be applicable to groundwater remediation programs nationwide.

Multiple fixed-price contracts will be awarded by Energy Systems on a low-bid basis to firms that meet the qualification criteria. The groundwater treatment demonstrations will be conducted at the vendor's site. The groundwater to be treated by the proposed demonstrations will be pumped from groundwater well GW-253 located in the vicinity of the S-2 site at the west end of the Y-12 Plant. Groundwater monitoring data collected from GW-253 show heavy metal, organic, nitrate, and radioactive contamination.

During this reporting period, an "Expression of Interest," which specified the qualification criteria required to conduct the proposed groundwater treatment demonstrations, was issued to the private sector in June 1989 to solicit their response to the project. Several replies were received, and a list of potential vendors for the groundwater demonstrations was developed for issuance of the statement of work and RFP.

Shortly after continued FY 1990 funding for the project is received from DOE, the RFP will be issued to those firms that responded positively.

3.3.4 Solvent Substitution Demonstration

This demonstration is being carried out for HAZWRAP as part of its compliance technology

evaluations. There are three main purposes for the work:

1. design, procurement, installation, testing, and demonstration of a process from the private sector to replace a chlorinated solvent (HydroSeal) and Freon® with a water and detergent solution in an ultrasonic system in the maintenance operations of the Y-12 Plant Research and Support Division;
2. documentation for the mutual benefit of the DOE defense program solvent substitution programs carried out at Y-12 and funded by Albuquerque Operations over the past several years; and
3. interfacing with nationwide DOE defense program nuclear weapons complex chlorinated solvent substitution programs over the life of this task through established working committees.

The ultrasonic cleaning system will be installed in the vacuum pump shop (9720-13) at the Y-12 Plant. The shop is operated by personnel of the Plant Research and Support Division at Y-12, the organization responsible for most of the activities associated with this demonstration.

The work is especially timely in that the methylene chloride ingredient of the HydroSeal solvent is a recognized carcinogen and will soon be covered under DOE Order 5480.10,⁸ which specifies the DOE carcinogen control program. In recent years, the nuclear weapons complex has been actively involved in a program to eliminate the use of chlorinated solvents for the weapons manufacturing process. During this time, much progress has been made, most of which is generally unavailable outside the nuclear weapons sector of the Defense Program within DOE. Thus, the documentation of the solvent substitution work throughout the nuclear weapons sector of the Defense Program is an important aspect of this project.

A number of nationwide and Y-12 committees have been active in the solvent substitution effort, and personnel from Y-12 have been actively interfacing with the following groups:

1. Chlorinated Hydrocarbon Solvents Coordinating Committee of the nuclear weapons complex, which was chartered to exchange technical information;
2. Chlorinated Solvents Action Committee at Y-12, which is addressing substitutions of chlorinated solvents through a series of

- activities governed by long-range planning; and
3. Interagency Manufacturers Operating Group, which is addressing cleaning and contamination control on a weapons-complex-wide basis.

Initial activities, led by the WMTC, centered on project definition and planning, based on experience gained in carrying out other demonstrations. A work breakdown structure was defined, and an estimation of costs for each element was formulated. Based on the definition and costing, the project was proposed to HAZWRAP and selected based on its merits and cost benefits. Subsequently, a field work proposal was prepared and submitted.

The ultrasonic system has been designed and specified through Y-12 Engineering, and the specifications are at Energy Systems being incorporated into a procurement package. The 9720-13 pump shop has been readied for installation of the equipment. Preparation of the report covering previous Y-12 solvent substitution efforts is in progress and is expected to be published in early 1990. The equipment is to be installed and tested in February and March 1990, and the actual demonstration will be carried out in April 1990. Video documentation of this demonstration and other solvent substitution efforts is planned, and a final report is scheduled for issuance in August 1990.

3.4 Information Exchange

During this report period, WMTC/WMTSG continued management assistance to the national DOE Uranium Task Force (UTF); also, several nationally significant workshops, for example, the off-site release criteria, contaminated soils, and leaching tests workshops, were organized and conducted.

3.4.1 UTF

The recently issued DOE Order 5820.2A¹ defines uranium wastes (excluding mining and mill tailings) as LLWs and requires that their disposal meet all applicable federal, state, and local regulations. All disposal sites will have to verify, through a formal performance assessment, that the uranium wastes meet these requirements before disposal.

The proposed 40 CFR 193⁹ is expected to identify uranium wastes as LLW and to require that

disposal of such waste "be conducted in such a way that assures that no individual receives a total dose from release to the general environment in excess of 25 mrem/year from all the LLW disposal in the U.S." Furthermore, wastes with concentrations of radioactivity low enough to result in annual doses of 4 mrem/year or less to individuals could be disposed of as nonradioactive wastes.

Both the requirement for a performance assessment by the DOE and the EPA's establishment of performance objectives for uranium wastes represent changes from the past.

Because uranium and its daughters have very long half-lives and because the chemical properties of uranium make it potentially very mobile in the geosphere, a framework is needed to develop, refine, and maintain programs to satisfy DOE performance objectives for long-term safety. Uranium wastes, on a volume and weight basis, make up a significant portion of all LLW.

To help meet these performance objectives, the Defense Low-Level Waste Management Program organized the UTF. The primary objectives of the UTF are: (1) developing an understanding of the problems associated with the disposal of uranium wastes, (2) identifying options available for resolution of these problems, and (3) developing recommendations for the consistent and safe treatment and disposal of uranium wastes.

Although three of the DOE facilities account for the bulk of the uranium wastes, many of the other sites generate some uranium wastes and must, therefore, comply with current and pending regulations for these wastes. By reviewing and assessing the status of uranium waste generation, storage, and disposal at all sites, the necessary data will be assembled to facilitate an integrated and coordinated approach to the new and proposed, regulator-approved and performance-based systems for uranium waste disposal.

Large volumes of uranium wastes have been generated since the early 1940s, beginning with the commission of the Manhattan Project. In addition to the great volumes of these materials, it is significant that the wastes are generated at various types of facilities, such as production and processing installations, research laboratories, weapons plants, fabrication plants, and testing facilities. The waste may be solid, liquid, or gas and of various compositions. It may be entirely a low-level radiological waste or various categories of a mixed radiological-hazardous waste. The points to be noted are that there are various facilities of waste

generation, sources within a facility, operations within a source, forms generated from a source of operation, and types of waste of different physical state, chemical, and radiological composition. Further, the waste type and generation rate will vary from one facility to another. Thus, the complexity of the problem of uranium waste management can easily be discerned.

There are 17 facilities that are managed by the DOE and generate, treat, store, and/or dispose of uranium and other LLWs. Many of these facilities were constructed in the 1940s and 1950s and were not designed to accommodate the waste minimization standards of the 1990s. Thus, many of these facilities are using outdated equipment and/or obsolete technologies, while some have upgraded their processes to meet the rapidly changing standards of waste generation, treatment, storage, and disposal.

It is necessary to look at the approach to uranium waste management at each facility in the context of an integrated picture of the entire DOE system. This is imperative now because of the implementation of DOE Order 5820.2A¹ and the proposed 40 CFR 193⁹ by EPA.

At present, site-specific data on the management of uranium at some 17 facilities have been assembled and are being analyzed to develop a comprehensive report on uranium processes, treatment, storage, and disposal on a DOE-wide basis.

3.4.2 Contaminated Soils Workshop

The management of large quantities of soils contaminated with generally low concentrations of fission products and/or other radioactive materials is a major problem at many DOE-owned facilities because interim policies for their disposition have been slow to develop. To initiate work throughout DOE toward common solutions to this perplexing and difficult problem, a workshop on "The Management of Radioactively Contaminated Soils" was held at the Knoxville Hilton Hotel in November 1988. The meeting was sponsored by Energy Systems for DOE and was hosted by the WMTC. Fifty-eight scientists and engineers attended the 1-d event, which included a morning plenary session and panel discussion with four concurrent topical sessions in the afternoon, followed by a

wrap-up session in the evening. Representatives from 16 sites with contaminated soils problems attended, along with scientists and engineers affiliated with the DOE, Tennessee Department of Health and Environment, Texas Bureau of Radiation Control, and seven private companies. The size and composition of the group were considered to be optimum for the topical sessions as there was full participation with lively discussions and interchanges of ideas. With selected representatives from all of the DOE sites that possess and/or generate contaminated soils and with experts in regulatory affairs and soil characterization, many of the important issues that bear on the solutions to problems of contaminated soils were addressed. Topical sessions focused on were: (1) characterization, (2) present practices, (3) regulations, and (4) interim and performance-based policies.

In this first national meeting on the management of contaminated soils, there was unanimous agreement that dose-based performance objectives can be used in deriving soil concentration limits from pathway analysis models. This is an important result, as this approach would bring about the desired consistent and rational management of contaminated soils at all DOE sites. Although there was agreement on the primary approach to the management of these wastes, many secondary issues were discussed without clear resolution. These issues included:

1. setting acceptable confidence intervals for certification of soils;
2. deciding whether to excavate soils or leave them in place for decay, as appropriate;
3. deciding if all or parts of reservations or plant sites should be surveyed in search of contamination; and
4. deciding the extent to which personnel exposure should be taken into account in remediation decisions.

The characterization of contaminated soils is a complex, time-consuming, and often costly first step in the management of these materials. As the data generated in this process provide the basis for the succeeding steps of remediation and certification, they must be thoroughly accurate and cost effective. The difficulty in the characterization process stems, in large part, from the inhomogeneity of contamination that is spread over large areas and the necessity to make measurements at or near the ambient background levels of natural or fallout

radioactivity. In the planning phase of characterization, it is essential that a team-effort approach be used among health physicists, radiochemists, statisticians, etc., as existing records, maps, surface surveys, and other pertinent data are assembled to lay out a representative sampling program. Once the sampling phase is completed, it is imperative that the analytical work be carried out under rigid quality assurance and quality control principles.

Present practices for the handling and disposition of contaminated soils include box-and-bury on-site and/or box-and-ship off-site methods. Treatment processes that may be used in the cleanup of contaminated soils include incineration, solidification, stripping, and bioremediation. Although these treatments have been used with varying degrees of success in hazardous and toxic waste streams, there are limited and largely unknown effects in their application to the treatment of radioactively contaminated soils. All of these tests and procedures are likely to be used, though no single technique is capable of adequately treating all types of contaminated soils.

There are rules and regulations, either in effect or proposed, that address the management and disposal of contaminated soil, once it is declared a "waste." DOE Order 5820.2A,¹ Chapter III, addresses the management of radioactively contaminated materials, as does the EPA's draft proposed regulations for LLW disposal. The latter would establish national environmental standards for the management, storage, and land disposal of contaminated soil classified as LLW or naturally occurring accelerator-produced radioactive waste (40 CFR 193⁹). Soils contaminated with both radionuclides and hazardous materials are also subject to the rules and regulations promulgated by EPA under RCRA.⁶

It was clear from all discussions at the workshop that interim and long-term policies should be set up on the dose-based performance objectives of DOE Order 5820.2A¹ and EPA's draft proposed 40 CFR 193.⁹ As an example using this approach to dispose of these wastes, a strategy was presented to develop a dose-based, soil-waste classification system for the Portsmouth Gaseous Diffusion Plant.

3.4.3 Off-Site Release Criteria Workshop

At the request of staff from Energy Systems' Environmental and Safety Activities Division, the

WMTC arranged and convened a 1.5-d workshop in May 1989 at the Pollard Auditorium in Oak Ridge, Tennessee, to discuss the subject of off-site release criteria for contaminated materials. Seventy-two people attended the workshop. The attendees were mainly from DOE facilities faced with managing contaminated materials but also included experts from industry and the regulatory agencies. The goal of the workshop was to develop a strategy for the development of reasonable release criteria for slightly radioactively contaminated materials for disposal off-site. Some of these materials are currently being generated and stored at DOE sites as a result of decontamination and decommissioning operations.

The workshop consisted of six opening presentations and a panel discussion. The presentations were: (1) Nature and Magnitude of Problems, (2) Regulatory Issues, (3) DOE Order 5400.XX¹⁰ Impact, (4) Current Industry Practices, (5) Chemical Wastes Acceptance Criteria, and (6) Modeling Studies to Develop Release Criteria. Four topical sessions were held during the afternoon of the first day to allow the participants to examine the issues in depth. The four topical sessions were entitled (1) Problem Scope, (2) Regulatory Issues, (3) Decontamination Technologies, and (4) Strategic Options. A summary session was held the next morning to allow the participants to assimilate the various components of the workshop. At the summary session, the chairperson of each of the four topical sessions summarized the discussion at his session, and then the floor was opened to the participants to further debate the issues.

The workshop adjourned with the unanimous agreement that there is an urgent need to develop reasonable release criteria, and a plan was developed for DOE to undertake the development of these criteria working with other regulatory and standards-setting bodies such as the Nuclear Regulatory Commission, EPA, the American National Standards Institute, the National Council of Radiation Protection, and the ASTM.

3.4.4 Alternative Treatment Technologies Workshop for Superfund Sites

The Alternative Treatment Technologies Workshop for Superfund Sites was held in November 1988 at Oak Ridge Associated

Universities in Oak Ridge, Tennessee. It was presented by EPA in cooperation with the DOE/ORO and Energy Systems. Local arrangements were made by the WMTC and HAZWRAP. This 2-d workshop was an in-depth "hands on" experience in evaluation of treatment technologies and decisionmaking regarding application of appropriate technologies to selected cases. An overview of each technology group, namely physical, chemical, biological, and thermal processes, was followed by working sessions in which attendees investigated the application of technologies to specific wastes defined by hypothetical site conditions. Technologies being evaluated by the Superfund Innovative Technology Evaluation Program were also discussed.

3.4.5 Leaching Test Workshops

Because of the importance of leaching tests in waste management, the WMTC is sponsoring a series of four workshops on leaching tests and their relation to disposal conditions and waste-form durability. Most of the currently viable tests were considered in the first workshop (July 1987) on scientific basis and applicability of leaching tests. The second workshop (October 1988) continued to develop the scientific basis and applicability of leaching tests with primary emphasis on short-term tests to reliably predict long-term durability of waste forms.

The third (tentatively scheduled for the fall of 1990) and fourth workshops will address leaching

under disposal conditions and durability of waste forms, respectively. The ultimate goal of the workshop series is to provide information and data on the durability of waste forms and how to prove or predict the durability of stabilized/solidified waste forms.

3.4.6 Symposia Cosponsored by National Professional Societies

Under the auspices of the American Chemical Society (ACS), symposia have been organized by the WMTC and held in conjunction with National ACS meetings.

The symposium on Innovative Waste Management Technologies was held in April 1989 in Dallas, Texas. Developing waste treatment and disposal methods were compared to historical and conventional industrial practices. Presentations were made on recently developed innovative methods such as microwaving, in situ immobilization, and the use of engineered waste forms to meet requirements of specific geologic sites.

The symposium on the Environmental Chemistry of Groundwater was held in October 1989 in Winston-Salem, North Carolina. The symposium included a tutorial on groundwater chemistry and hydrogeology, and it also dealt with pollution problems and movement of contaminants.

References

1. U.S. Department of Energy, *Radioactive Waste Management*, DOE Order 5820.2A, September 26, 1988.
2. U.S. Congress, National Environmental Policy Act, 42 U.S.C. 4321 *et. seq.*, January 1, 1970.
3. S.-Y. Chang and A. L. Rivera, "A System Optimization Model for Low-Level Nuclear Waste Disposal," Proc. 7th Int. Conf. on Mathematical and Computer Modeling, Chicago, IL, Aug. 2-5, 1989.
4. S.-Y. Chang and A. L. Rivera, "Theory and Evidence of Economies of Scale in the Development of Waste Management Systems," presentation at DOE Model Conf., Oak Ridge, TN, Oct. 2-6, 1989.
5. ANSI/ANS-16.1-1986, Measurement of the Leachability of Solidified Low-Level Radioactive Wastes by a Short-Term Procedure, American Nuclear Society, La Grange Park, IL, 1986.
6. U.S. Congress, Resource Conservation and Recovery Act of 1976, Pub. L. 94-580, Oct. 21, 1976, as amended by the Hazardous and Solid Waste Amendments Act of 1984, Pub. L. 98-616, Nov. 9, 1984.

7. L. D. Eyman, *Hazardous Waste Remedial Action Program Annual Progress Report for the Period Ending September 30, 1988*, DOE/HWP-74, Oak Ridge, TN, July 1989.
8. U.S. Department of Energy, *Contractor Industrial Hygiene Program*, DOE Order 5480.10, February 25, 1986.
9. U.S. Environmental Protection Agency, "Environmental Standards for the Management, Storage, and Land Disposal of Low-Level Radioactive Waste and Naturally Occurring and Accelerated Producer Radioactive Waste," *Code of Federal Regulations*, 40 CFR 193, draft proposed rule, 1988.
10. U.S. Department of Energy, *Radiation Protection of the Public and the Environment*, DOE Draft Order 5400.XX, Washington, DC, March 18, 1988.

4. Radiochemical and Isotope Programs

Chem Tech performs chemical process R&D related to most segments of the nuclear fuel cycle, such as fuel and target processing for recycle and recovery, radioactive waste minimization and treatment, and fission product transport under reactor accident conditions. The division also processes, packages, and distributes a wide variety of electromagnetically separated stable isotopes and many radioactive isotopes from the lightest (tritium) through transuranium elements in many special forms for use in research and in applications for medicine and industry. During the period of this report, the division's special capabilities in remote radioactive materials handling and processing were sought for the emergency recovery of a leaking radioactive source from a commercial irradiation facility.

4.1 Radiochemical Engineering Development Center (REDC) Programs

The Radiochemical Engineering Development Center (REDC) includes the remotely operated and maintained hot cells and glove box laboratories in Bldgs. 7920 and 7930, located in the Melton Valley area of ORNL. The REDC has unique shielding, containment, and equipment replacement capabilities, which enable radiochemical engineering R&D activities to be carried out on all types of high-level radioactive materials, including alpha, beta-gamma, and neutron emitters. The major programs at the REDC are (1) the Transuranium Element Processing Program, (2) the ^{252}Cf Industrial Sales/Loan Program, and (3) the Mark 42 Processing Program. All of these involve radiochemical separations of transuranium elements.

The purpose of both the Transuranium Element Processing Program and the Mark 42 Processing Program is to develop and use radiochemical processes for the recovery of transuranium elements and specific radionuclides and to supply the recovered materials to U.S. DOE-sponsored programs. The purpose of the ^{252}Cf Industrial Sales/Loan Program is to sell bulk Cf_2O_3 and palladium-californium cermet or alloy to foreign and domestic commercial users and to fabricate and loan ^{252}Cf neutron sources to universities and government agencies.

4.1.1 Transuranium Element Processing Program

The REDC serves as the processing, storage, and distribution center for transuranium elements used in the heavy-element research program in the United States. The isotopes of these elements continue to be used nationally and internationally in basic studies of physics and chemistry, including experiments designed to produce new, neutron-rich isotopes of the heavier elements. Activities at the REDC are carried out in collaboration with those in other divisions at ORNL and at other national laboratories where research on actinide and transactinide elements is performed. All of the ^{252}Cf feed material for the DOE ^{252}Cf Industrial Sales/Loan Program is recovered in the REDC.

Typical Processing Steps

Target rods containing americium and curium are remotely fabricated at the REDC; irradiated in the High Flux Isotope Reactor (HFIR); and then processed at the REDC for the separation, recovery, and purification of the heavy actinide elements, which are then distributed to laboratories throughout the country for use in research. The isotopes recovered directly from the mainline chemical processing of targets irradiated in the

HFIR are ^{249}Bk , ^{252}Cf , ^{253}Es , and ^{257}Fm . Other products are separated decay daughters. Each of these is recovered by purifying the radioactive parent, allowing the parent to decay, and then processing to recover the daughter. The separated daughters include ^{248}Cm , ^{249}Cf , and high-isotopic-purity ^{253}Es . Products are usually highly purified prior to shipment and are frequently provided in special chemical forms or in special devices required by the experimenter.

Processing of the targets irradiated in the HFIR is accomplished by a long sequence of individual steps that begins in the heavily shielded, remotely operated hot cells in Bldg. 7920. The processing steps generally tend to separate the transuranium elements as a group from aluminum, metallic impurities, activation products, and fission products before partitioning and purifying the individual elements.

The sequence of processing steps is called a "campaign." All of the target rods for a single campaign (usually a group of 10 to 15) are put into a dissolver vessel and solubilized in two steps. The aluminum is preferentially dissolved in caustic nitrate solution, which is decanted through a filter. Then the residual actinide and fission product oxides are dissolved in 5 to 6 M HNO_3 . Various metallic impurities are removed by means of the Cleanex batch extraction process, which uses di-(2-ethylhexyl)-phosphoric acid as the extractant. The Cleanex product solution is subsequently treated in two steps to remove rare-earth fission products and separate the curium and americium from the transcurium elements: (1) a rough separation is made, using the Tramex batch extraction process, in which the extractant is a tertiary amine; and (2) more complete decontamination from fission products and separation of americium and curium from the transcurium elements are accomplished by means of the LiCl-based anion-exchange process. The transcurium elements are then separated from each other by means of a pressurized ion-exchange process using chromatographic elution with α -hydroxyisobutyrate.

The curium is converted to curium oxide microspheres for fabrication into targets for irradiation in the HFIR. The transcurium products are transferred from the main cell bank to other cells and glove boxes for final purification. Product purification operations for a particular actinide element may include cation-exchange, anion-exchange, solvent extraction, and precipitation

techniques in various combinations to provide the purity and chemical form required by the experimenter.

Current Operations

During this report period, the HFIR operated for only a very short time and at low power. This was not sufficient to produce needed amounts of transuranium elements in the existing 15 targets. Approximately 100 d of irradiation at the new full-power level of 85 MW will be required to restore all short-lived transuranium elements to the desired amounts.

An extensive program was continued to upgrade the equipment in the Bldg. 7920 hot cells for remote fabrication of targets for irradiation in the HFIR. Primary emphasis was on the target pellet fabrication equipment in Cell 3, which had become very unreliable. The entire sequence of operational steps required for pellet fabrication was reviewed. For some of the steps, it was determined that the existing equipment design was adequate, but the equipment was worn out. For those items, it was only necessary to fabricate new equipment with few, or no, design changes. Equipment for other fabrication steps had to be redesigned in order to meet functional requirements. A major change involved the use of precision positioning equipment that has become available commercially since the original pellet fabrication equipment was designed. The new equipment has been fully automated and is computer driven. A mockup of the in-cell equipment support stand was erected and used for out-of-cell testing of the assembly of the new equipment. This testing was successfully completed, and the new equipment is being installed in Cell 3.

A batch of americium-curium product, previously recovered in Campaign 66, was purified and converted to oxide microspheres for use in fabrication of new HFIR targets. A total of 122 g of oxide containing 7 g of americium and 86 g of curium was prepared.

The ^{252}Cf produced each year (~ 450 mg when the HFIR is in full operation) greatly exceeds annual needs. The majority of this product nuclide is highly purified of all curium isotopes and set aside for several years to allow ingrowth of the ^{248}Cm daughter. Isotopically pure ^{248}Cm is quite valuable in both chemical and nuclear research. The ^{252}Cf loan program was established to allow productive use of the ^{252}Cf neutrons while the ^{248}Cm

is forming. Approximately 119 mg of ^{248}Cm was separated from stored ^{252}Cf during this report period. Also, ~55 mg of ^{248}Cm that had been previously separated from ^{252}Cf in 1984 was purified and made ready for shipment. The processing steps included (1) elution of the actinides from the storage packages, (2) concentration and purification of the actinides by means of cation-exchange techniques, (3) removal of residual ^{252}Cf from ^{248}Cm by selective chromatographic elution from cation-exchange resin using α -hydroxyisobutyric acid, and (4) final removal of cationic and anionic impurities. The final product contained 54 mg of ^{248}Cm , with <50 ng ^{252}Cf per g of ^{248}Cm .

During the report period, 50 shipments were made. These include 162.5 mg of ^{252}Cf in 19 sales and 18 loans under the auspices of the ^{252}Cf Sales/Loan Program. The total quantities of each type of product shipped are listed in Table 4.1.

4.1.2 Californium Neutron Sources

Californium-252 is an ideal material for making portable neutron-source capsules. The half-life of ^{252}Cf is 2.65 years, and, although the primary decay mode is by alpha emission to ^{248}Cm , ~3% of the decay occurs by spontaneous fission. This very intense neutron source generates 2.3×10^{12} neutrons $\cdot \text{s}^{-1} \cdot \text{g}^{-1}$. Compared with conventional (α, n) neutron sources, ^{252}Cf neutron sources have

smaller dimensions. As with all isotopic neutron sources, ^{252}Cf sources do not require elaborate power supplies, control systems, or maintenance programs. Californium-252 neutron sources have many applications, including cancer therapy, reactor startup sources, and nuclear fuel rod scanners. There are also applications in the fields of neutron radiography and neutron activation analysis, as well as some special applications, such as in the new thermal neutron analysis-based airport luggage monitors.

During this reporting period, a loan of 100 mg of ^{252}Cf , in six encapsulated neutron sources, was made to the McClellan AFB. These sources are the heart of a new, robotic neutron radiography facility. The new facility is expected to speed up aircraft maintenance by quickly detecting areas of corrosion or fuel contamination that will require further inspection or repair. The Air Force expects this new facility to return aircraft to the flight line more quickly, thus "buying" the equivalent of one new F-111 fighter plane. If experience confirms this expectation, other Air Force, Navy, and possibly even civilian maintenance depots will become interested in neutron radiography.

4.1.3 ^{252}Cf Industrial Sales/Loans Program

Beginning in 1986, ORNL assumed responsibilities for the DOE ^{252}Cf Industrial Sales/Loans Program, which involves loans of ^{252}Cf neutron sources to agencies of the U.S. government and sales of ^{252}Cf in the form of bulk oxide and palladium-californium alloy pellets or wires. The program had been operated from 1968 through 1986 in temporary facilities at the Savannah River Laboratory (SRL). These activities were then transferred to the REDC Californium Facility in Bldg. 7930. The Californium Facility has been operated since 1970 by the staff at REDC as part of the DOE Heavy-Element Research Program. Additional equipment and facilities were provided in the Californium Facility to accommodate the Sales/Loan Program activities while continuing the Transuranium Element Processing Program work.

The improved Californium Facility can purify and package bulk californium, as well as fabricate neutron sources, in all of the physical forms required by both the research and industrial sales/loans programs.

Table 4.1. Total quantity of each type of product shipped from the REDC between July 1, 1988, and September 30, 1989

Product nuclide	Shipment	Total quantity (mg)
^{243}Am	3	133
^{244}Cm	8	2.75
^{248}Cm	2	10.7
^{252}Cf (Sales)	19	49.6
^{252}Cf (Loans)	18	112.9
	<u>50</u>	

As of April 1, 1988, ORNL assumed responsibility for administering the contracts for all of the loans of ^{252}Cf sources that had previously been administered by SRL. At that time, 76 contracts covering 179 different items were in place with 73 different institutions. Concurrently, ORNL had 43 loan contracts with 28 institutions covering 83 items. At the end of this report period, a total of 117 contracts covering 272 items were in place with 91 institutions. The 272 items contain a total of 240 mg of ^{252}Cf .

4.1.4 Mark 42 Processing Program

A new transuranium element project called the Mark 42 Processing Program was begun in November 1988 to recover ^{242}Pu , ^{243}Am , and ^{244}Cm for use in weapons diagnostic studies. Activities to process one Mark 42 target per year, beginning in FY 1990, will be performed along with the Transuranium Element Processing Program activities at the REDC.

The Mark 42 target assemblies were originally fabricated and irradiated at SRL and were discharged from the reactor more than 5 years ago. Each target, now containing ~250 g of ^{242}Pu , 75 g of ^{243}Am , and 85 g of ^{244}Cm , consists of a set of three concentric tubes formed from a $\text{PuO}_2\text{-Al}$ cermet extrusion with aluminum cladding. When the Mark 42 target assemblies were removed from the reactor for storage, they were cropped to a length of ~4.3 m (~14 ft) [the active section is 3.86 m (152 in.)]. Before the Mark 42 target assemblies can be processed in REDC, they must be cut into eight segments that are nominally ~51 cm (~20 in.) long.

During this report period, initial activities were carried out to prepare for shipment and disassembly of the Mark 42 targets. Arrangements were made to ship the targets in a commercial spent fuel cask, the NLI-1/2, owned by Nuclear Assurance Corporation. Since neither SRL nor ORNL currently has suitable facilities for disassembly, arrangements were made for segmenting at Pacific Northwest Laboratory (PNL). After disassembly, each segment will be sealed into a welded stainless steel can for shipment from PNL to REDC, again using the NLI-1/2 shipping cask.

Several new equipment items or modifications are needed to unload the NLI-1/2 at REDC Bldg. 7920. These tasks are in progress and include (1)

raising the elevation of the outside 45-Mg (50-ton) crane to provide clearance to lift the NLI-1/2 off the transport trailer, (2) fabrication of a new dolly and tracks to move the NLI-1/2 into the building, (3) modifications of the carrier stand to hold the NLI-1/2, (4) fabrication of a new adapter ring to position the existing LRL 25T ("Q-ball") transfer cask over the NLI-1/2, (5) fabrication of a temporary shield plug for the NLI-1/2 unloading operations, and (6) fabrication of a new top plug for the Q-ball. An integrated test of the unloading equipment and procedures will be made using an empty NLI-1/2 cask before the segmented Mark 42 pieces are shipped to ORNL.

Initially, processes and equipment currently being used for processing transuranium targets at REDC will be used for the Mark 42 targets. Improved processes and equipment are being developed to increase the efficiency of Mark 42 processing and will be implemented as soon as possible.

A significant effort was made during this report period to develop an improved method for separation and purification of americium. The Mark 42 Program will require this separation, whereas it is not done routinely in the Transuranium Element Processing Program. A carbonate precipitation process has been used previously and is the reference flowsheet pending new developments. Several years ago, tests were made using a more efficient process in which americium in the feed solution was oxidized to Am(V) and the solution passed through a cation-exchange process to sorb essentially all cations except Am(V) . Those tests were performed on americium that contained only ~4 mg/g of curium, and only 50 to 60% yields of purified americium were obtained. Current tests have been made using controlled oxidation to Am(VI) with ammonium persulfate and controlled reduction of the Am(VI) to Am(V) using hydrogen peroxide. Beginning with equal concentrations of americium and curium, the improved process has enabled 60 to 70% yields of purified americium while using an organic cation-exchange resin in the hot cell. Moreover, yields of >90% have been obtained in glove box tests at tracer level concentrations when using an inorganic cation exchanger, such as zirconium phosphate or polyantimonic acid. Sol-gel techniques are being tested with the goal of preparing these types of inorganic sorbents with sufficient loading capacity to permit higher concentrations of americium and curium to be processed.

4.2 HTGR Fission Product Transport

Chem Tech's activities under the HTGR Program include investigating the behavior of volatile fission products in the helium circuit of a gas-cooled reactor and, in particular, interactions with the metallic parts of the circuit. This work is not reported here this year in compliance with regulations stipulating that such information must be distributed solely as applied technology.

4.3 ^{233}U Program

This program has existed at ORNL's Radiochemical Development Facility (RDF) for almost three decades and has provided DOE with (1) a national repository for ^{233}U , (2) uranium purification (for removal of ^{232}U decay daughters) and oxide preparation, and (3) distribution services to various users of ^{233}U . In recent years, however, receipts and shipments have diminished. Because of this decreased demand and because the associated processing equipment is obsolete, Chem Tech recommended to DOE's Office of Nuclear Materials Production that (1) the existing ^{233}U processing equipment be placed in long-term standby condition by FY 1989 and (2) modern equipment be installed in better hot cell facilities at Bldg. 7930 if needs for ^{233}U are renewed.

4.3.1 Current Operations

Work during the period between July 1988 and October 1989 was performed to (1) improve RDF documentation, (2) reduce personnel exposures, and (3) improve personnel access control during ^{233}U operational standby surveillance activities. Also, discussions were initiated and planning has begun for preparing ^{233}U stored at Mound Laboratories to be transported to RDF for long-term storage. Although Mound has shut down its ^{233}U handling facilities, this material must be repackaged to meet current shipping regulations and RDF storage requirements. An acceptable overpack is being sought that will allow transport with only minimal repacking and still permit the necessary repackaging for storage at RDF.

In response to the changing technical safety requirements, activities are currently focusing on

improving safety and quality documentation, safety-related signs, contamination control, and general operational safety. To reduce the radiation background and associated personnel exposures during surveillance of standby equipment, glove boxes were cleaned of inside-surface contamination resulting from many years of ^{233}U processing and secured to eliminate some of the components (e.g., gloves) vulnerable to aging by radiation or air drying. Additionally, selected glove boxes were disposed of, since they were (1) either deteriorating or unneeded for operational standby status and (2) were imposing an additional resource drain and exposure burden as part of surveillance operations.

A new safeguards plan was developed, approved by DOE, and implemented to redefine the personnel access area for stored ^{233}U . Since essentially all ^{233}U at RDF is securely stored in one area, recently revised DOE safeguards regulations enabled redefinition of area controls and, hence, more efficient surveillance activities.

4.3.2 Facility Upgrades

Upgrades are continuing to improve control of contaminated liquid wastes. A survey of RDF identified six pathways for undetected leakage of water into contaminated areas, with the potential for subsequent leakage of contaminated liquid to the environment. Work has begun to isolate and instrument these areas so that continual monitoring by operating personnel will provide timely detection and response to such a buildup.

Also, preparations have begun for tie-in of RDF's liquid low-level waste line from its secondary containment tray to a new doubly contained line. Project control documentation has been completed, and design is about 90% complete for anticipated construction during the next report period. This tie-in is necessary to dispose of ^{233}U -contaminated liquids because the integrity of the secondary containment tray is no longer ensured.

4.3.3 Facility Cleanout

A concerted effort was expended to clean up and dispose of $>100\text{ m}^3$ of contaminated process-related materials that had accumulated over the years at RDF in out-of-the-way places. Extensive monitoring, documenting, and packaging were required for disposal of the transuranic, low-level,

hazardous, and mixed wastes. Included in this was a special task to solidify (by grouting) and dispose of ~100 L of ion-exchange resins and organic-coated extraction chromatography packings used during the last report period to purify residual ^{233}U solutions.

4.4 Purex Flowsheet Development

4.4.1 Purex Solvent Treatment

Purex solvent is degraded by both radiation and nitric acid. Standard solvent treatment, typically with a basic solution, removes the major portion of the degradation products. Some degradation products formed by degradation of the diluent are not soluble in aqueous solutions and slowly build up in the solvent until they adversely affect the operation of the solvent extraction system. Earlier work has shown that treatment of the solvent with activated alumina, particularly if the solvent is pretreated to remove water, is very effective in returning the degraded solvent to near-pristine condition. Recent efforts were aimed at finding the best type of commercially available activated alumina for removal of the diluent degradation products and retained ruthenium from first-cycle SRL solvents that had been previously washed with sodium carbonate solution. The efforts were particularly aimed at finding activated aluminas that would be effective without predrying the solvent.

A macroporous activated alumina with a high sodium loading (A-204-4 from LaRoche Chemicals) was the best of the commercially available activated aluminas tested. This activated alumina was effective in treating degraded solvent when saturated with water, eliminating the need to dry the solvent; was more effective in the removal of the long-chain degradation products and had a higher capacity for them; and was capable of removing all the chemical forms of retained ruthenium. The effects of temperature on the equilibrium and kinetics of solvent treatment were determined.

4.5 AVLIS Processing

Chem Tech's activities under the AVLIS Program include the production of UCl_4 from

uranium oxide and electrochemical reduction of uranium from molten salts. This work is not reported here in compliance with regulations stipulating that such information must be distributed solely as applied technology.

4.6 RSI Cesium Capsule Recovery

In mid-June 1988, a release of ^{137}Cs from one or more sealed sources was experienced by Radiation Sterilizers, Inc. (RSI) at their facility outside Atlanta, in Decatur, Georgia. The sources, used to sterilize medical supplies, were leased to the firm by DOE. This site, one of five supplied cesium under the loan program, contained 252 doubly encapsulated CsCl sources from the Waste Encapsulation Storage Facility (WESF) in Hanford.

The state of Georgia requested emergency aid from DOE, and, in turn, ORNL was asked to respond. The initial response dealt primarily in the health physics/impact analysis/emergency action areas. Following the initial response, the role of ORNL continued to evolve and included providing an on-site senior technical advisor, loading and shipping suspect capsules to ORNL, and identifying and removing the failed capsules from the facility. ORNL's current responsibilities in connection with the RSI Recovery Project are (1) to maintain a subcontract with a firm to decontaminate the facility, (2) to provide on-site technical and operational management, and (3) to perform capsule examination to determine the failure mode of the leaking capsules.

4.6.1 On-Site Recovery Activities

The 252 WESF capsules, each containing ~50 kCi of ^{137}Cs as CsCl encapsulated in stainless steel tubes, were stored in two racks at the facility. A wet-dry operational mode was used in which the racks were submerged in a temperature-controlled demineralized water pool when not in use, and irradiation was accomplished by raising the racks out of the pool and circulating pallets holding the product through the radiation field in the irradiation cell and then returning the racks to pool storage following a run.

The event involved the release of 6 to 10 Ci of activity, with ~4 Ci reporting to the pool and the remainder distributing on surfaces in the irradiation cell and pallet loading and unloading area. Chem-Nuclear Systems, Inc., (CNSI) was retained by RSI, and subsequently by ORNL, to provide facility and pool decontamination.

A team from Westinghouse Hanford Company (WHC) conducted a visual examination, using underwater video equipment, of each source and identified 27 capsules that were judged to show potential corrosion of the weld areas. Further, underwater ultrasonic tests were made, and two capsules yielded signals that could be interpreted as an indication of water in the annular region between the inner and outer shells.

The DOE/ORO on-site recovery team requested that ORNL provide support for transporting sources from Decatur to ORNL's hot cells for examination. A General Electric Model 600 cask was selected for the shipment, and the Nuclear Regulatory Commission required that the capsules be contained in a metal capsule overpack (CO) that should be purged of water and dried, and that the CO be tested for leaks while underwater. The overpacks were designed and fabricated by RSI. The first shipment containing the source judged to show the highest degree of corrosive attack was made, and the CO was found to contain water when it was opened at ORNL. Examination of the source in the hot cell revealed that there was no obvious evidence of corrosive attack but that discoloration in the weld areas could be interpreted, in an underwater environment, as corrosion.

ORNL undertook the task of modifying the overpack equipment and loading procedure to provide a vacuum drying system and incorporating features to enhance the safety of the operation into the design. In the second shipment two capsules were in individual overpacks, which showed ultrasonic test indications of water in the annular region. The CO was dry when this shipment arrived. Examination of these sources in the hot cell, including helium leak checking and sampling of the annular gas, revealed no evidence of leaking nor any artifacts of water in the annuli. Further, ultrasonic tests using the same equipment employed in the RSI pool on the source end-caps produced the same indication of water even though the caps were dry.

The activity of the water in the pool continued to decrease as a result of the operation of the ion exchangers installed by CNSI. The pool activity was

monitored by the state of Georgia, and their data indicated that, with time, an equilibrium state was achieved more than an order of magnitude above that of the demineralizer effluent. The interpretation was that one or more sources was releasing activity into the storage pool at the rate of ~25 Ci/h.

As a result of this conclusion, ORNL designed an in situ leak detector that could isolate three sources at a time from the pool water and sample the water in contact with these sources for increased activity. Using this equipment, an ORNL team tested the remainder of the suspect sources, as well as several others, with negative results. During this period, it was observed that the apparent leak rate from a source, or sources, increased from 25 to ~150 $\mu\text{Ci/h}$.

A new, more aggressive leak detection device was designed and fabricated at ORNL to incorporate the ability to isolate six sources from the pool water at a time and to subject them to cyclic elevated hydrostatic pressures and elevated temperatures. The water in contact with the capsules was continuously monitored for any increase in activity. During the loading of the leak detector, it was noted that one of the six sources would not fit within the detector sheath. Visual observation of this source with underwater video equipment showed that the source was swollen ~10 cm (4 in.) from one end. This source, Capsule 1502, was isolated in a capsule overpack, and the pool water activity immediately began to decrease. Testing of the remainder of the capsules continued until all 249 sources were tested. One additional source, 1504, was also found to be swollen, but isolation of this capsule had no effect on the pool water activity.

Prior to the shipment of these two sources to ORNL, the water in each CO was sampled for activity. The results showed a significant increase of activity associated with Capsule 1502 but not for 1504. These sources were transported to ORNL in December 1988 for failure mode analysis.

Since the indicator of a failure is a swollen capsule and since in the as-manufactured condition the inner capsule is free to move within the outer capsule, two additional tests were devised. The first was a simple set of go, no-go ring gauges to determine swelling, and the second was a "clunk" test using underwater microphones to listen for the free movement of the inner capsule. A team from WHC has performed these tests on all remaining

capsules at the Decatur facility, at a sister RSI facility in Westerville, Ohio, and in the WESF storage pool. Nine nonclunking sources were found in Decatur, one in Westerville, and none at WESF. These ten additional capsules have been transported to Hanford.

The Laboratory continues to provide on-site technical oversight for DOE/ORO, oversight of the cleanup activities at the site by CNSI, and coordination of shipping the remaining sources to Hanford by WHC.

4.6.2 Capsule Examination

Examination of the two failed capsules (Fig. 4.1) and failure mode analysis were initiated in the

hot cells of Bldg. 3525. Broadly, the examination consisted of two phases. The first was nondestructive and included all tests up to the actual removal of the outer containment shell. The nondestructive phase has been completed for both capsules. The second phase, the destructive examination directed toward obtaining salt samples, has been delayed pending approval to restart activities in the shutdown hot-cell facilities of Bldg. 3517.

The nondestructive examinations can be grouped into five areas. In developing the examination plan, care was taken to ensure that the tests performed in one group would not adversely affect the results desired from tests in other groups. On receipt at the hot cells, a complete set of photographs of each capsule was taken at low magnification.

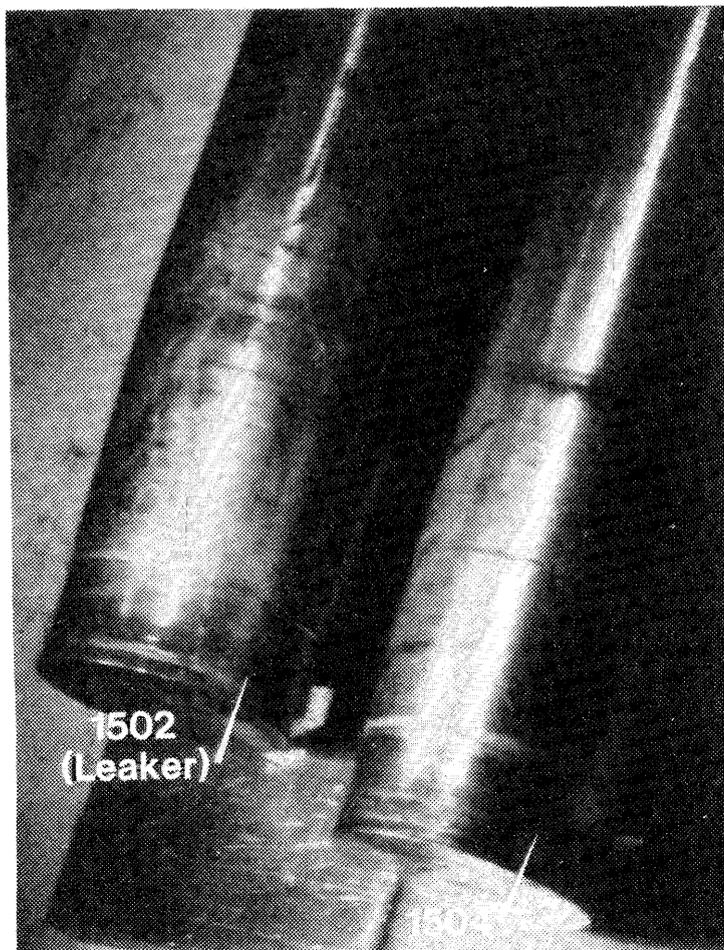


Fig. 4.1. Two failed capsules found at the Radiation Sterilizers, Inc., facility in Decatur, Georgia. Both sources show swelling at one end. Capsule 1502 is designated as a leaking source based on tests performed at the facility. Capsule 1504 did not appear to leak activity into the environment.

Dimensional profiles of the capsules were obtained initially and after they had undergone several thermal cycles as a result of other tests. Distortion was observed at only one end of the capsules beginning just above the end-cap weld, reached a maximum ~10 cm (4 in.) from that end, and disappeared at ~25 cm (10 in.) from the end. The remainder of the 53-cm (21-in.) capsule did not appear to be swollen. The nominal diameter of an unswollen capsule is 6.67 cm (2.6 in.), and at the maximum of the swollen region, the diameter was ~6.99 cm (2.75 in.). Analysis of the initial measurements showed a slight circumferential asymmetry, and, after thermal cycling, the asymmetry decreased, but no evidence of circumferential expansion or continued swelling was found. Temperature profiles, in free air, were also made. The temperatures rose from a minimum of about 130°C at the unswollen end to a maximum of 250°C in the swollen region.

Autoradiographs of the capsules were made on both an axial and a radial grid. From analysis of these data, it was concluded that there is a 15-cm (6-in.) void above the salt column, the salt fills the swollen volume, and, in one capsule, a 2.5-cm (1-in.) length at the top of the salt is free to move. These conclusions indicate that the failure mechanism must account for both a densification and an expansion of the salt. Further, there is evidence of the presence of CsCl in the annular region, that is, outside the inner capsule, at the swollen end of both sources.

Penetration into the annular regions was accomplished by drilling into the caps at each end of the capsules. Gas samples were withdrawn at the time of entry and analyzed for radiolytic decomposition products of water. The results for Capsule 1502, the leaker, show 85% hydrogen, whereas the hydrogen content of the gas from the annulus of Capsule 1504 was only 5%. This latter measurement is consistent with the gas withdrawn from the three previously demonstrated nonleakers. Low oxygen concentration was observed for both capsules and is consistent with the gettering action of the barium decay product of cesium observed to be in the annulus from the gamma scans. The annular volume of both capsules was ~60% of the normal 180 cm³, and it was established that there was a gas pathway, albeit restricted, between the ends of the capsules.

Helium leak detection was performed on both capsules to ascertain if there was a breach in the

outer containment cylinder and, in the case of Capsule 1502, to determine the leak site. All tests were conducted at 0.34 MPa (50 psig) helium and included (1) external pressurization followed by external vacuum, (2) external pressurization with vacuum applied to the annular region, and (3) pressurized annuli with external vacuum. No evidence of a leak was found in either source with a test sensitivity of 10 to 1 mPa (1×10^{-7} to 1×10^{-8} atm) · cm³ · s⁻¹.

Additional tests were made on Capsule 1502 in an attempt to locate the leak site. These included application of a "wet blanket" to the surface of the capsule in an attempt to dissolve any salt plugging the site. Radiographs of the blanket showed spots of surface activity but were not definitive in locating the leak site. The capsule was also submerged in a temperature-controlled water bath with the annulus pressurized to 0.34 MPa (50 psig) helium. A sniffer probe was located above the water to indicate the presence of escaping gas. These results were also negative.

A high-temperature dye penetrant was applied to the entire surface of both capsules. No evidence of cracks or holes was found for either source.

Analysis of the manufacturing data for the two failed capsules examined at ORNL and those capsules that failed the clunk tests revealed no underlying pattern. The manufacturing dates spanned the period from August 1980 to June 1983, and each capsule contained a different batch of salt. There also appears to be no correlation between the weight of the CsCl filling and failure, since the failed list contains both the lightest and heaviest capsules manufactured. Further, there appears to be no evidence for stratification of contaminants during the pouring since the pour spout used to fill these capsules was randomly distributed among the seven used in the batch pouring operation.

4.7 Metallurgical Examination Hot Cells

The Metallurgical Examination Hot Cells (MEHC) Department provides technical services for nuclear materials research programs at ORNL. Capabilities for nondestructive and destructive examination of irradiated materials, including nuclear fuels, are maintained in support of client program objectives. The MEHC resource base for

experimental operations includes four hot-cell facilities: the Physical Examination Hot Cells, Bldg. 3025E; the Segmenting Hot Cells, Bldg. 3026D; the Oak Ridge Research Reactor Hot Cells, Bldg. 3042; and the High-Radiation-Level Examination Laboratory, Bldg. 3525. In March 1989, responsibility for operation of the Bldg. 3025E hot cell facility, which was utilized primarily for remote mechanical properties testing of irradiated metals and alloys, was transferred to the Metals and Ceramics Division.

4.7.1 High Temperature Gas-Cooled Reactor (HTGR) Fuels Qualification Program

ORNL is the DOE lead laboratory in the qualification of nuclear fuel forms being evaluated for the modular HTGR technology development program. The MEHC conducts postirradiation examination of prototypic coated-particle fuels in collaboration with the Fuels Qualification Group of the Metals and Ceramics Division. Irradiation of experimental HTGR fuels at ORNL has been disrupted by the shutdown status of the HFIR since 1987. However, the United States is participating in a joint venture with the Federal Republic of Germany (FRG), and irradiated HTGR fuel from the High Flux Reactor, located at Petten in the Netherlands, and from the Arbeitsgemeinschaft Versuchsreaktor, located at Jülich in the FRG, is currently being examined in the Irradiated-Microsphere Gamma-Analyzer Cell in Bldg. 3525. Selected subsets of microspheres from the FRG Set 4 fuel inventory are processed through the particle singularizing and gamma spectrometry systems for fission product measurement and segregation of "failed" fuel particles.

4.7.2 Fusion Materials Program

One of the major tasks in the Magnetic Fusion Energy (MFE) Program is the development of engineering materials for use in the prospective construction of magnetic-confinement fusion

reactors. In collaboration with the Structural Materials Group of the Metals and Ceramics Division, MEHC disassembles reactor experiment capsules for, and conducts physical and mechanical properties testing of, the irradiated metallurgical specimens. The MFE-6J and MFE-7J experiment capsules irradiated in the Oak Ridge Reactor and the CTR-49, CTR-50, CTR-52, CTR-53, CTR-54, CTR-55, and CTR-56 experiment capsules irradiated in the HFIR were disassembled in the South Cell at Bldg. 3042. Metallurgical test specimens recovered from these experiments, along with others from previous irradiations in the Oak Ridge Reactor, HFIR, and the Fast Flux Test Facility at Hanford, were tested in Bldg. 3025E. The test program included metrology, densitometry, tensile testing, and fatigue testing. Selected irradiated specimens from the ORR-MFE-6J and ORR-MFE-7J experiments were assembled into two new experiment capsules, the HFIR-MFE-60J and HFIR-MFE-330J, for reirradiation when the HFIR resumes operation.

4.7.3 Examination of Cesium Capsules from Radiation Sterilizers, Inc.

The examination of the ^{137}Cs sources recovered from the RSI facility in Decatur, Georgia, was an important project conducted in the High Radiation Level Examination Laboratory, Building 3525. This project is described in Sect. 4.6.

4.7.4 Reduced Enrichment Research Test Reactor (RERTR) Program

The objective of the RERTR Program, directed by Argonne National Laboratory (ANL), is to qualify the performance of low-enriched uranium fuel for application to research reactors in support of United States nuclear nonproliferation initiatives. Use of uranium silicides, rather than oxides, in the fabrication of the prototypic fuel provides for high ^{235}U density with a ^{235}U enrichment of only 20 to 40%. The program, active since the 1970s, will be completed with the evaluation of fuel irradiated in

the Oak Ridge Reactor full-core demonstration project.

Selected fuel elements were disassembled at the Bldg. 3042 hot cells, and individual plates recovered from them were transferred to Bldg. 3525 for thickness measurements and fission product profiling by gamma scanning. Samples were sheared from these fuel plates for shipment to ANL, where metallographic examination and burnup analyses will be completed. Other fuel plates from prior Oak Ridge Reactor irradiations were recovered from the Solid Waste Storage Area retrievable-storage facility for sectioning of additional samples to be included in the ANL shipment.

4.7.5 Facility Upgrade/Consolidation Activities

In addition to R&D activities conducted by MEHC, significant resources have been committed to a multiyear facility upgrade program. The major objective of the program is to modify Bldg. 3525, which has operated since 1964 primarily as a fuels examination laboratory, to provide a multipurpose facility that, along with the Bldg. 3025E hot cells, will serve Chem Tech's and ORNL's long-range needs for postirradiation examination and testing of both fuel and nonfuel materials. Irradiated nonfuel material examinations previously performed in Bldgs. 3042 and 3026D, which are aged and substandard by current nuclear facility design criteria, are being consolidated into Bldg. 3525 as the transition progresses.

Facility upgrade initiatives in Bldg. 3525 were somewhat disrupted by the unanticipated task of examining WESF cesium capsules, suspected of leaking, recovered from RSI. Methodical operations directed toward packaging radioactive solid waste for disposal, cell decontamination, and equipment refurbishment were supplanted by a crash effort to prepare part of the facility for conducting the RSI capsule examinations. Rigid temporary exhaust ducts were installed in the basement, replacing flexible temporary ducts, to ensure containment integrity while the high-activity sources were handled in the hot cells. The Cell Ventilation Upgrade Project in Bldg. 3525 was completed in November 1988 with the installation of permanent replacement ducting during a brief lapse in the RSI work. The overall

impact of the RSI task on Bldg. 3525 upgrades is an estimated 6-month setback in progress relative to established objectives.

An intensive campaign to prepare Bldg. 3026D for placement in safe standby condition pending D&D was begun in May 1989. The objectives of the campaign are to dispose of contaminated equipment items from cell access areas, decontaminate those areas accessible to personnel to reduce risk of contamination exposure, remove radioactive metallurgical test specimens from the hot cells to alternative storage sites, dispose of debris and contaminated equipment items from the cells, and perform a preliminary washdown of the cell interior surfaces to reduce levels of transferable contamination. It is estimated that 6 to 12 months will be required to accomplish the task, dependent on availability of funding and manpower resources.

4.8 Isotopes Research Materials Laboratory

The Isotopes Research Materials Laboratory (IRML) is one of the most diversified isotope materials processing facilities in the world. Since it was established in 1961, it has been a preparation center for custom-ordered materials enriched in various isotopes to meet special research needs. The IRML staff has extensive experience and unique facilities for performing vacuum, metallurgical, and ceramic processing of enriched isotopic materials. Isotopes of virtually every element in the periodic table, both stable and radioactive, have been processed for distribution to the worldwide research community on a cost-recovery basis. In addition, the IRML materials R&D effort has developed or improved the materials processing methods used to prepare the research material samples. IRML's R&D capabilities are also used to assist other Energy Systems programmatic efforts.

In addition to carrying out hundreds of "routine" research materials preparations (many of which would be impossible to do anywhere else in the world), IRML has produced a number of unique products during the period covered by this report. For example, the development of full-scale particle beam neutralizer foils for use in the Strategic Defense Initiative (SDI) Program was completed. A small-scale version of these foils is shown in Fig. 4.2. They are made by flash-evaporating a thin layer of metal (usually aluminum) onto a glass plate that

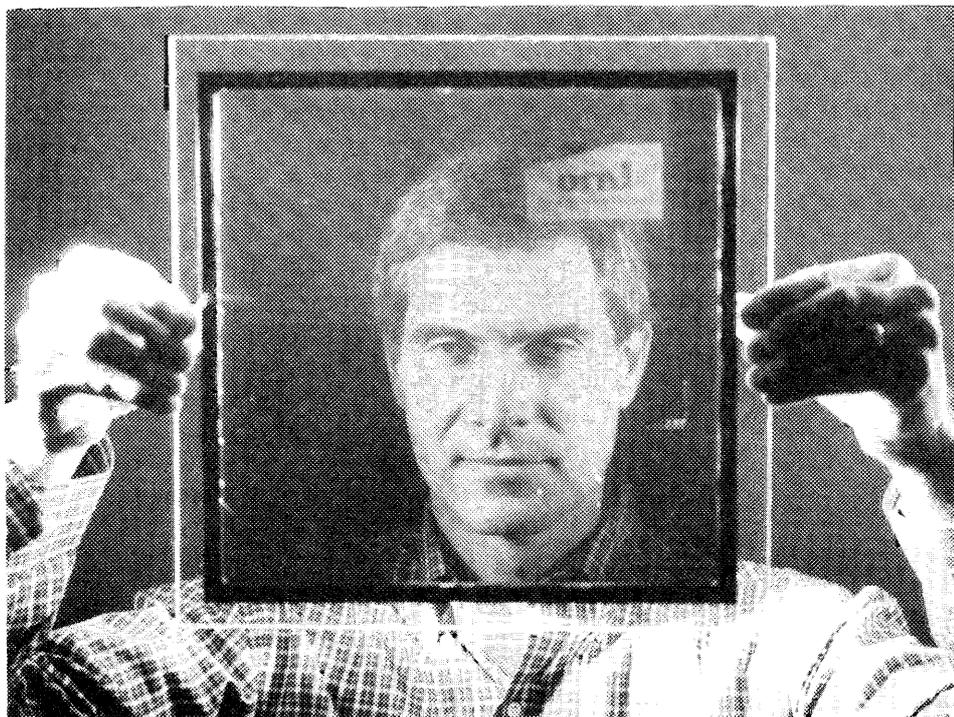


Fig. 4.2. This aluminum foil, developed for the Strategic Defense Initiative Program, is thin enough to see through.

is coated with a parting agent. The thin foil [on the order of 300 Å (30 nm) thick] is floated off in a large tank, then draped over a nickel-mesh frame assembly that is more than 90% transmissive. These foils have since been successfully tested by other participants in the SDI Program.

In support of a cancer therapy development program at Brookhaven National Laboratory (BNL), a small (500-mg) batch of normal Sm_2O_3 was converted to metal by the reduction/distillation process. This material is being evaluated at BNL to determine its suitability for loading in small titanium seed capsules. If BNL decides the metal is satisfactory, the entire U.S. inventory of $^{144}\text{Sm}_2\text{O}_3$ (500 mg), which was purchased from ORNL several years ago, will be returned and converted to metal.

Two high-output (10^8 neutrons/s) neutron sources were prepared by mixing 4.1 g of $^{238}\text{PuO}_2$ with 15 g of beryllium metal powder and ball-milling in a hot cell. The development of an improved ^{244}Cm - ^{13}C monoenergetic gamma source was also completed. In previous sources, the Cm-C mixture was a loose powder that was free to move around inside the source capsule. A need developed for a source in which the mixture was

fixed in one location. This was accomplished by loading the ^{244}Cm - ^{13}C mixture into an aluminum capsule, covering it with a layer of aluminum powder, and hydraulically pressing the capsule in a die. This procedure yields a monolithic source that is permeable enough to allow the escape of decay-produced helium but that can be securely held in place in the finished capsule.

A major job involved the preparation of ^{44}Ca plates for Los Alamos National Laboratory (LANL). A total of 76.79 g of ^{44}Ca that had been on loan to LANL from the Isotopes Distribution Office was returned to ORNL in the form of $^{44}\text{CaCO}_3$. Since ^{44}Ca has a value of \$7.30/mg, this represented \$560,567 worth of material. The carbonate was converted to the oxide and subsequently reduced to the metal with an excellent yield of 94.9%. The metal was split into smaller portions, arc-melted, and cold-pressed into plates for subsequent rolling in an inert gas box. A total of seven 5- by 10-cm plates were rolled; three were 2 mm thick, and four were 0.6 mm thick.

In a major actinide project, a 1-g pellet of ^{244}Cm metal was prepared for use as a diagnostic sample for LANL. The metal was prepared by

IRML's vacuum-pyrochemical process of reduction/distillation after which it was melted and cast into a cylinder in a boron nitride mold, then doubly encapsulated in thin-wall vanadium cans.

In addition to production and R&D activities, a major consolidation of IRML facilities was begun during this period.

4.9 Isotope Enrichment

4.9.1 Stable Isotope Enrichment

Stable and actinide isotope enrichment activities are carried out in the Isotope Enrichment Facility (IEF), located in Bldg. 9204-3 at the Y-12 site, using electromagnetic isotope separators (calutrons). Sixty-two of these high-current mass spectrometers are available for isotope enrichment. Eight are housed in a containment area and can be used for low-specific-activity beta-gamma and alpha-active separations. The latter units are called the "Actinide Facility" and are currently in standby condition.

The calutrons are physically arranged in two rectangular "tracks" (Fig. 4.3). Each track is ~30 m long and contains two parallel lines of mass spectrometer units. The two parallel lines are joined by magnetic shunts that subdivide the track into segments, and the ends of each track are joined by an iron yoke. Each segment forms a closed magnetic loop linking its group of separators in a common magnetic field produced by excitation of its electromagnetic coils. Different field values can be

obtained by exciting the coils in each segment independently to enable separation of the isotopes of different elements simultaneously. Most of the mass spectrometer units are the type having a 180°, 61-cm-radius ion path. However, as indicated in Fig. 4.3, several of the spectrometers have a different geometry designed to enable greater physical separation of the isotopes, but at the expense of a slower rate of separation.

The purposes of the Isotope Enrichment Program are (1) to develop separations processes and related technology for preparation of needed enriched isotopes; (2) to operate the processes to supply, on a worldwide basis, enriched isotopes for research, medical, and industrial applications; (3) to improve the processes where possible to reduce costs; (4) to maintain the equipment and facilities; and (5) to incorporate new equipment (such as PSP, AVLIS, Centrifuge, etc.) to enable improvement of specific isotope separations and capacities.

During this report period, the calutrons were operated continuously (on a 5-d-per-week basis), using one or two segments, and stable isotopes of the elements Ca, Sb, Tl, Sn, Te, and Fe were separated. A summary of the enrichment campaigns is given in Table 4.2.

Two types of activities occur regularly in the chemical separations areas. First, appropriate chemical forms of the various elements are prepared for use as feed material to the electromagnetic separators. These feed forms must be such that the metal or compound can be uniformly vaporized and ionized at a suitable rate. Also, the feed form must be highly purified to minimize the deterioration of the separation by ionic and neutral impurities. Second, collector pockets from the mass spectrometer units containing each separated isotope product are leached or ignited. They are then processed through various chemical and physical steps to recover and purify the enriched isotope product and convert it into the chemical form desired by the user.

Development activities are inherently required to some extent for each element separated and for each enriched isotope product that is recovered. This may include feed material preparation, vaporization and ionization techniques, baffling and other ion-path control methods, collector pocket or collection method design, and development of chemical separation and conversion processes.

A modification of the electrodes used for ionizing thallium was made to lower the cost of

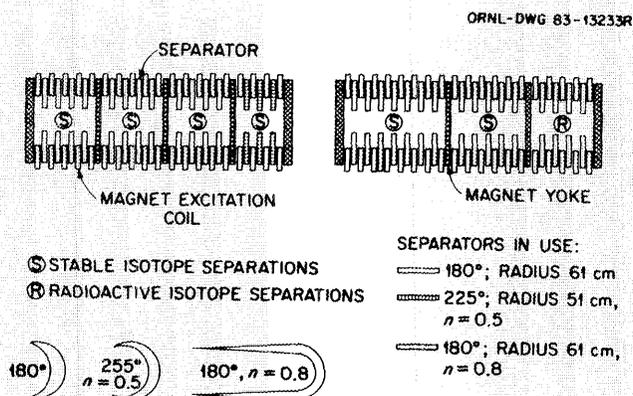


Fig. 4.3. Diagram of the physical arrangement of the calutrons in the Isotope Enrichment Facility.

Table 4.2. Summary of stable isotope enrichment campaigns

Element separated	Date completed	Campaign length (months)	Cumulative calutron tank-hours	Efficiency of separation (%) ^a	Amount processed (g)
Calcium	March 89	6	20,544	75	1,752
Antimony	May 89	2	840	72	66
Thallium	June 89	11	48,360	107	8,997
Tin	July 89	4	14,328	81	323
Tellurium	September 89	3	9,936	105	514
Iron		2	8,448	75	491 ^a

^aThe efficiency rating is a comparison of the product of the average total throughput and the fraction of time in actual operation with similar data from a previous reference separation.

electrode fabrication and to enable easier changout of the negative electrode isolation insulator. The modification included a completely new potential stress relief system for the negative extraction electrode and required redesign of the grounded secondary focusing electrode mount.

During the campaign to enrich the isotopes of tin, three feed forms were evaluated: SnCl₄, SnS, and SnS with added H₂S. Data collected during the campaign (Table 4.3) indicated that the SnS feed with added H₂S enabled the highest product recovery, while SnCl₄ enabled the highest isotope enrichment.

In the chemical separations area, an electrochemical process was developed to remove chemical impurities from enriched strontium isotopes. In this process, Sr(NO₃)₂ solution was electrolyzed using a platinum anode and a platinum cathode. Product recovery was >99.5%, and the key impurities (copper, lead) were reduced to <10 mg/kg. Also, an electrolytic process was developed

for converting enriched isotopes of mercury from HgO to metal. This process utilizes a uniquely designed platinum cathode. Compared with previously used methods, the new process enables higher product yields (>93%), a reduced number of processing steps, and a reduced processing time (less than half of that previously required).

A catalytic process for conversion of SF₆ to H₂S or metal sulfides has been developed. This process will be used to enable sulfur isotopes to be pre-enriched in gas centrifuges, in the form of SF₆, and to be further enriched in the calutrons using other sulfur compounds. Overall, this will enable relatively large amounts of highly enriched sulfur isotopes to be produced.

4.9.2 Special Separations

New approaches to chemical separations of isotopes are being investigated. Initial experimental

Table 4.3. Comparison of feed materials for enrichment of tin isotopes

Feed material	Length of enrichment (tank-hours)	Recovery (%)	Final isotopic concentration of ¹¹² Sn (at. %)
SnCl ₄	3810	60.0	84.39
SnS	5247	62.2	77.66
SnS + H ₂ S	3847	74.8	81.24

efforts have explored promising extractants, and one extractant appears to be particularly interesting but was not available in sufficient quantities for reliable and accurate research. Useful quantities of this extractant are being prepared. Several other alternative extraction media have also been identified that would use more conventional chemicals, and initial investigations of these extractants have begun.

Methods for introducing extractants into high-efficiency contacting systems have been explored theoretically, and preliminary experiments have been initiated. Major innovations in the contacting systems that would give even more rapid and effective extraction are also being explored, and experimental development of such approaches will proceed as time and resources permit.

4.9.3 Plasma Separation Process (PSP)

The PSP is a process for separating isotopes of metallic elements in large quantities. It is based upon the selective heating of isotopes by ion-

cyclotron resonance. In order to use the ion-cyclotron principle, a PSP separation module must vaporize the feed material and then produce an electrically neutral plasma of ions and electrons. Once the plasma is formed and the isotopic ions are selectively heated, the separator uses the difference in average energy between the different ionic species to effect a physical separation.

PSP was developed by TRW, Inc., for DOE during the period 1976–1987. To date, DOE has invested more than \$160 million in the PSP program. During FY 1988 and 1989, the large-scale PSP equipment has been disassembled and shipped to ORNL, where it has been placed in storage pending identification of new isotope production missions.

Chem Tech has been responsible for receiving the PSP hardware and placing it into storage; transferring from TRW reports, manuals, and procedures that document the technology base; and evaluating market needs for PSP-produced isotopes.

The major equipment item in a PSP module system is the superconducting magnet coil in which the separations take place. The large-scale PSP magnet system, shown in Fig. 4.4, is ~8 m long, has an inner bore diameter of 1 m, weighs 39.5 Mg

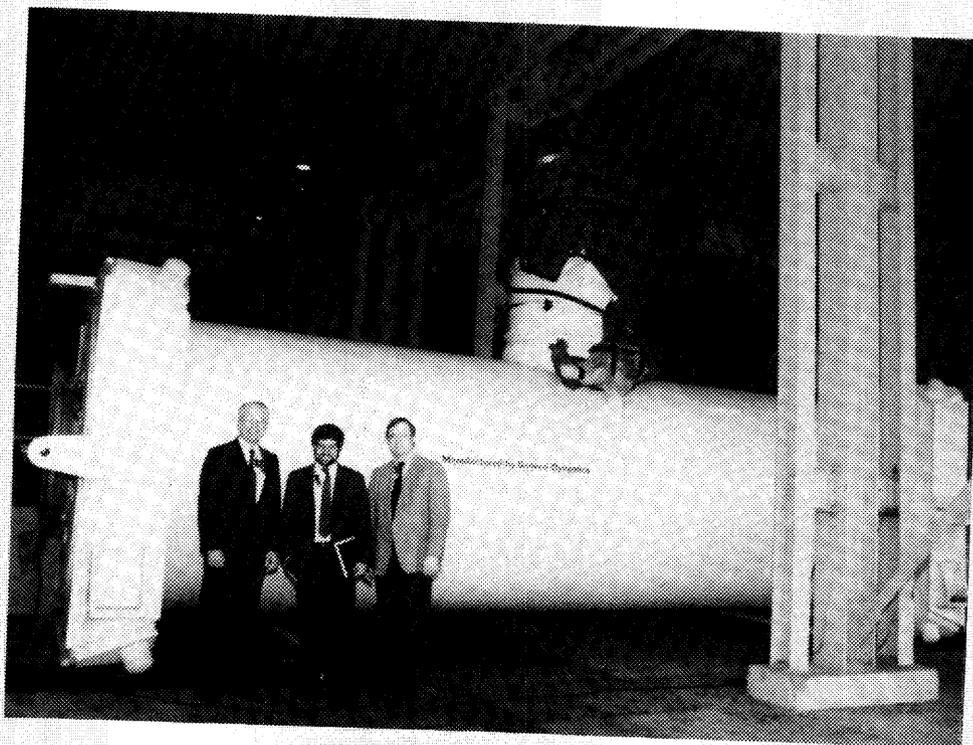


Fig. 4.4. The large-scale PSP magnet system is the largest homogeneous field magnet in existence.

(87,000 lb), and is capable of producing a field of 2 T with a homogeneity of $\pm 0.1\%$. Estimated to have a replacement cost of \$9 million to 10 million, this system is the largest homogeneous field magnet in existence. Its high value and relative fragility required very special operations in order to transfer it to Oak Ridge and to unload it from the truck. During January 1989, the magnet system was received and placed into storage in Bldg. 1423 at the Oak Ridge K-25 site.

In addition to the magnet system, 15 tractor-trailer loads of PSP equipment with an estimated capital value in excess of \$20 million were transferred from TRW to ORNL. Included in the transferred equipment were several high-power microwave systems, such as two 1-MW, 28-GHz systems; one 1-MW, 56-GHz system; and several lower frequency systems. The 56-GHz microwave system has been placed on a 2-year loan to Princeton Plasma Physics Laboratory to aid in fusion energy research. A customized high-amperage power supply system has been loaned to the ORNL Superconducting Motor Program. The remainder of the equipment has been placed into storage in Bldg. K-31 at K-25.

PSP technology transfer activities were begun in September 1989 as members of ORNL's Fusion Energy and Chemical Technology divisions visited the TRW site to observe and record the disassembly of the large isotope separation module. During the spring of 1989, over 500 PSP program documents were received from TRW, including test procedures, test reports, system requirement reports, theory reports, and operating procedures. A library system was established to enable the future user to sort and retrieve information.

A first-cut, highly preliminary cost estimate was generated for the reassembly and startup of the PSP facility at Oak Ridge. It was estimated that 3 years and ~\$10.5 million would be required to reassemble the separator module, replace old and missing equipment (diagnostics and controls), and bring the system to the point where production operations could commence. This reassembly cost remains as the principal obstacle to deployment of the PSP technology on new isotope separation missions.

PSP Isotope Separation Missions

In the past, the PSP separator has been used for large-scale isotope separation tasks involving isotopes of nickel and palladium. Separation

performance has also been demonstrated on small quantities of isotopes of U, Pb, In, K, and selected lanthanide elements. This performance experience, as well as reliability data from previous production operations, was used as a basis for evaluating potential new PSP isotope missions. A market assessment of stable isotope needs was undertaken to identify these new missions and to forecast emerging isotope needs.

Potentially significant PSP missions are summarized below. In some instances, these missions had not been previously identified or have become more exigent as circumstances have changed.

1. **Radiological seeds for cancer therapy.** A novel cancer therapy employs PSP-enriched ^{102}Pd as a precursor for making ^{103}Pd and is apparently gaining wide acceptance for treatment of prostate cancer. Current inventories of enriched ^{102}Pd will be depleted in several years; a restarted PSP system appears to be the only viable production system to support this application.
2. **Advanced neutron optics.** A multilayer coating system composed of ^{58}Ni and ^{62}Ni isotopes is projected to be capable of increasing the transmission of cold neutrons to neutron-scattering instruments in research reactors by a factor of 3 to 9. The availability of these PSP isotopes would revolutionize the economics of research reactors and neutron-scattering instrumentation.
3. **Long-life "betacells."** An electric power supply system that employs the beta-emitting radioisotope ^{63}Ni as its "fuel" has the potential for providing high-specific power with long life (>40 years) and with minimal radiological hazards. The operation of PSP to enrich the precursor ^{62}Ni isotope is critical to the production economics of these devices. These betacells are seen to have important space and military applications.
4. **Improved nuclear reactor materials.** The PSP technology has unique capabilities to enrich large quantities of rare earth isotopes. In particular, the use of ^{157}Gd as a burnable poison in commercial light-water reactors would have the benefits of improving their operational safety and economics.

In principle, there is a strong rationale for conducting a PSP isotope separation program at

Oak Ridge. It appears that the costs of the operation of a PSP program could be fully recovered by the revenues created from sales of enriched isotopes. In practice, the potential benefits of the PSP separations technology are dispersed among several user communities, and no single organization or community appears to be positioned to serve as sole sponsor for the restart of PSP. Future Chem Tech activities will be directed toward obtaining commitments of partial sponsorship from several user organizations and toward further investigation of new PSP isotope missions.

4.10 Radioisotope Processing

Radioactive gas processing activities are part of the radioisotopes processing activities at ORNL and encompass providing tritium and ^{85}Kr , both normal and enriched, to the medical, R&D, and industrial communities on a full-cost-recovery basis. Activities include purification of the gases to remove decay products and other unwanted contaminants, enrichment of ^{85}Kr to levels of concentration required by certain operations, and packaging of the materials in cylinders and uranium trap containers in quantities useable or capable of being handled by customers. A summary of activities in each of the areas of tritium operations, enriched ^{85}Kr , and normal ^{85}Kr and follows.

4.10.1 Tritium

A total of 783,762 Ci of tritium was processed and shipped to customers during the reporting period. During the last two months of the reporting period, the tritium processing system was shut down to investigate discrepancies between quantities shipped and those received by customers.

4.10.2 ^{85}Kr

Two enrichment campaigns, producing a total of 815 Ci of enriched ^{85}Kr products, were completed. A third campaign, which is expected to produce material >25% enriched, was still in progress at the end of the reporting period.

Two batches of normal ^{85}Kr feed materials totaling ~10,000 Ci of material were processed.

Sales of this material continued during the reporting period; about 5000 Ci of 2.5%-enriched material were sold.

4.10.3 ^{90}Y

Yttrium-90 is an important radioisotope that is undergoing extensive testing for the treatment of several forms of cancer. The ^{90}Y is extracted and purified from the decay of ^{90}Sr . Current work includes weekly production commitments to customers in the medical community for patient treatment as well as R&D of new treatment methods. R&D on process improvements to reduce the levels of ^{90}Sr and trace elements is also carried out.

Demand for ^{90}Y grew steadily during this report period, and average monthly shipments are approaching 2000 mCi per month. This demand is expected to continue increasing over the next year as patient trials begin at several medical research institutions. A variety of process improvements have been implemented in recent months to reduce worker radiation exposure and maintain the quality of the product while meeting the increased production needs.

4.10.4 ^{153}Gd

Gadolinium-153 has important applications in both medicine and industry. As a dual-photon source (X-ray and gamma), this gadolinium isotope has been used in a dual-photon absorption machine to determine the loss of bone minerals in patients suffering from osteoporosis. Industrial applications include thickness monitoring and X-ray fluorescent applications. Present production of this radioisotope is by neutron irradiation of natural europium. The objective of this work is to supply the customer demands for this isotope, continue process improvement development, and develop other uses for this isotope.

The largest demand for ^{153}Gd was for use in dual-photon sources. Unfortunately, this application largely disappeared when the insurance industry refused to reimburse the cost of diagnosis using this procedure. In addition, dual-photon X-ray tubes were developed that can be used for the same diagnostic procedures. Demand is now limited to industrial applications that require a relatively small amount of material.

4.10.5 ^{63}Ni

Nickel-63 is an isotope with a very weak (69-keV) beta activity. It is used in thickness gauging and may have application in nuclear battery development. Two experimental ^{62}Ni targets that had been irradiated in the Advanced Test Reactor to produce ^{63}Ni were successfully transported to ORNL, opened, and processed. Several radioactive contaminants were removed by ion exchange. A total of 224 Ci of ^{63}Ni valued at \$324,800 was recovered from the two targets.

4.10.6 ^{14}C

The radioisotope ^{14}C is not currently being produced in the United States. The world market demand of approximately \$1,000,000 per year is supplied by Canada. The objective of this project is to evaluate the feasibility of producing and processing ^{14}C by using HFIR-irradiated aluminum nitride (AlN). Several targets were irradiated, but the first pellets to be analyzed showed a ^{14}C assay of only 60 to 70 at. % vs the expected yield of greater than 90% (the balance being ^{12}C). An extensive review of the AlN pellet fabrication and handling procedures, the ^{12}C analysis of the starting material, and the recovery procedures showed negligible amounts of contamination by normal carbon. However, the pellets that were processed and analyzed had been exposed to air for several months, and it is theorized that they reacted with the water and carbon dioxide in the air to form $(\text{NH}_4)_2^{12}\text{CO}_3$ and thus contaminated the ^{14}C produced in the reactor. New procedures are now being developed to prevent this reaction. If successful, this project may provide an economical domestic source for ^{14}C .

4.10.7 ^{234}U

Uranium-234 is a decay product of ^{238}Pu and is principally used as a dosimeter (flux monitor) material for reactor operations. A multistage ion-exchange process is used to separate and purify the product. The residual ^{238}Pu is repackaged and

stored. ORNL is the sole domestic source of this important tool for reactor dosimetry, and we are currently producing ~10 g/year.

4.11 Isotopes Packaging and Shipping

The Isotope Packaging and Shipping operations encompass activities necessary to provide support required for the receipt and shipping of radioactive and stable isotopes. Activities include the actual packaging of isotopes products produced by ORNL, completion of packaging and labeling of radioactive materials shipments required for research activities conducted by other ORNL divisions and reactor operations, receipt of all radioactive materials shipments coming into ORNL, and maintenance and quality assurance (QA) of shipping packages. Maintenance and QA activities are performed in conjunction with the packaging and shipping activities. This operation requires a highly trained staff that is knowledgeable in Department of Transportation regulations regarding the packaging and shipment of radioactive and other hazardous materials as well as skilled in the handling of radioactive materials. A summary of the type of activities in this area is presented in Table 4.4. In addition to the 3324 radioactive shipments indicated in Table 4.4, ~500 shipments of stable isotopes are typical.

Table 4.4. Radioactive materials shipments
July 1, 1988, through September 30, 1989

	Number of shipments
Type of container	
Nonreturnable	2549
Returnable	324
Customer-owned	451
Type of material	
Solid	1086
Liquid	681
Gas	753
Empty	88
Total radioactive materials shipments in a 15-month period:	3324

4.12 Isotope Distribution Office

4.12.1 Business and Distribution Activities

The Isotope Distribution Office is responsible for the business and distribution activities of the Isotope Production and Distribution Program. The objectives are (1) to administer DOE's Isotope Production and Distribution Program for the sales and loans of isotopes within the regulatory requirements of state, federal, and international agencies and of Martin Marietta Energy Systems, Inc.; (2) to market the isotopes and services available through the program and forecast the needs of the user community; and (3) to direct the production of isotopes and various accelerators, reactors, and other production facilities available within DOE.

Sales of Products and Services

ORNL's Isotope Distribution Office offers the products and services available from DOE's Isotopes Program. DOE operates a network of research facilities with a wide variety of unique, versatile equipment assistance to a broad base of R&D activities in medicine, science, education, agriculture, and industry. Most of the products and services are offered routinely, but specialized technical services are also available through the program.

Sales of products and services for July 1988 through September 1989 are summarized in Table 4.5. Sales for individual items are shown in Table 4.6.

Top-Selling Isotopes and Top Customers

The top-selling isotopes for the period are shown in Table 4.7, and the top customers are shown in Table 4.8.

Table 4.5. Sales of isotope products and services

Classification	Revenue July 1988 through September 1989 (\$)
Radioisotopes	8,323,823
EM Isotopes ^a	6,411,702
Non-EM stable isotopes	136,347
Heavy elements and other special materials	<u>2,731,893</u>
Total	17,603,765

^aEM = electromagnetically separated.

Table 4.6. Itemized sales of isotopes products

Products and services	Revenue July 1988 through September 1989 (\$)
<i>Radioisotopes</i>	
Krypton	1,091,630
Tritium	2,830,307
Other inventory items	(78,916)
⁶⁰ Co	688,032
¹⁵³ Gd	281,190
¹⁹² Ir	127,455
Other major products	422,181
Miscellaneous processed materials	7,684
Argonne National Laboratory products	3,612
Brookhaven National Laboratory accelerator products	169,155
²⁸ Mg	8,843
¹²⁷ Xe	169,359
General Electric products	34,495
Los Alamos National Laboratory/ Los Alamos Meson Physics Facility	963,726
Handling charges	39,942
Other services	<u>1,565,128</u>
Total	8,323,823
<i>Non-EM stable isotopes^a</i>	
Lithium-6 and -7	103,773.00
Services	<u>32,574.00</u>
Total	136,347.00

Table 4.6. (continued)

Products and services	Revenue July 1988 through September 1989 (\$)
<i>Heavy elements</i>	
Radioactive isotope handling charge	39,942
²⁵² Cf	1,290,593
²⁴⁴ Cm	250,300
²³⁹ Pu	66,500
²⁴¹ Pu	16,735
²⁴² Pu	86,550
²³³ U	10,414
²³⁵ U	12,872
Other products	(162,996)
Services	399,889
Total	2,010,799
<i>EM Stable isotopes</i>	
⁴⁸ Ca	77,059
⁵⁸ Fe	40,154
⁶² Ni	532,088
¹²⁴ Te	63,107
²⁰³ Tl	3,140,070
¹⁶⁸ Yb	222,686
⁶⁸ Zn	655,808
Other products	1,213,570
Services	467,160
Total	6,411,702
<i>Other special materials</i>	
²⁴¹ Am	32,921
²⁰⁷ Bi	634,387
²³⁴ U	38,451
²³⁷ Np	2,425
²³⁸ Pu	12,910
Total	721,094
<i>Sales summaries</i>	
Total radioisotopes and non-EM stable isotopes	8,460,170
Heavy elements and EM stable isotopes	422,501
Other special materials	721,094
Total sales	17,603,765

^aEM = electromagnetically separated.

4.12.2 Marketing and Off-Site Production

Marketing Activities

The marketing function within the Isotope Production and Distribution Program generally focuses on promotion and public awareness activities. These activities consist of five primary components:

1. exhibition at national and regional meetings of technical and professional organizations;
2. production and distribution of publications, including a catalog and newsletters;
3. advertising in technical and professional publications;
4. public relations activities; and
5. market research into the uses of and needs for isotopes.

The Isotope Production and Distribution exhibit was displayed at the American Nuclear Society Topical Meeting on Industrial Radiation and Radioisotope Measurement Applications at Pinehurst, North Carolina; at the Tenth Conference on the Application of Accelerators in Research and Industry at North Texas State University, Denton, Texas; and at the 36th Annual Meeting of the Society of Nuclear Medicine at St. Louis, Missouri.

Information published and distributed during the period included five issues of the newsletter, *Isotope News*; a major revision of the products and services catalog; and an ordering brochure.^{1,2} Advertising was done through display ads in publications associated with exhibition activities and listings in national and international supplier directories.

The following customers were visited: E. I. du Pont in North Bellirica, Massachusetts; Mallinckrodt in St. Louis, Missouri; and the University of Missouri in Columbia, Missouri. In addition, the Isotope Production and Distribution program was represented by Chem Tech personnel at the International Target Development Society meeting at Darmstadt, Germany, and the 7th International

Table 4.7. Top-selling isotopes and services for July 1988 through September 1989

Material name	Sales (\$)	Percentage of total sales
²⁰³ Tl	3,140,070	17.8
Tritium	2,830,306	16.1
²⁵² Cf	1,290,593	7.3
Los Alamos National Laboratory irradiations	963,726	5.5
⁶⁸ Zn	655,808	3.7
Special services	634,387	3.6
Encapsulation services	541,845	3.1
⁶³ Ni	532,089	3.0
Transuranic services	486,328	2.8
⁸⁵ Kr	466,137	2.6
Total sales	17,603,765	

Table 4.8. Customers for July 1988 through September 1989

Customer name	Sales (dollars)	Percentage of total sales
E. I. du Pont de Nemours & Co.	3,389,669	19.3
Amersham International Limited	1,832,497	10.4
Saunders-Roe Developments, Ltd.	852,079	4.8
General Electric Company	706,385	4.0
Medi-Physics, Incorporated	677,800	3.8
Oak Ridge National Laboratory	633,915	3.6
Safety Light Corporation	538,984	3.1
Teledyne Energy Systems	451,794	2.6
E. R. Squibb & Sons	434,380	2.5
Self-Powered Lighting, Ltd.	352,950	2.0
Total sales	17,603,765	

Meeting on Radiation Processing at Noordwijkerhout, The Netherlands.

Customer surveys to determine anticipated demand levels, impact of supply shortages, and consequences of various changes in the program were conducted for tritium, enriched ⁸⁵Kr, ⁹⁰Y, ¹⁵³Gd, high-output neutron sources, and the Research Materials Collection of stable isotopes.

Off-Site Production

LANL routinely produces several short-lived radioisotopes at the Los Alamos Meson Physics Facility, and BNL produces short-lived radioisotopes

at the Brookhaven Linac Isotope Producer. The operating cycles of these two high-energy accelerators are staggered to provide maximum product availability to the user community. ANL produces two isotopes of plutonium for the program. Reactor space for production of radioisotopes is provided by the Idaho National Engineering Laboratory's Advanced Test Reactor located in Idaho. Special tritiated target fabrications are supplied by the General Electric Neutron Devices Group at Largo, Florida. Production and distribution for these off-site materials and services are administered by Chem Tech's Isotope Distribution Office, and Table 4.9 lists the radioisotopes available from each facility.

Table 4.9. Radioisotopes from off-site facilities

Los Alamos Meson Physics Facility	Brookhave Linac Isotope Producer	Argonne National Laboratory	Advanced Test Reactor ^a
²⁶ Al	⁷ Be	²³⁶ Pu	⁶⁰ Co
⁷³ As	⁶⁷ Cu	²³⁷ Pu	¹⁵³ Gd
⁷ Be	⁵² Fe		¹⁹² Ir
²⁰⁷ Bi	⁶⁸ Ge		⁶³ Ni
⁷⁷ Br	²⁸ Mg		
¹⁰⁹ Cd	²² Na		
⁶⁷ Cu	⁹⁷ Ru		
⁶⁸ Ge	⁸² Sr		
⁸³ Rb	¹²⁷ Xe		
³² Si			
⁸² Sr			
¹²⁷ Xe			
⁸⁸ Y			

^aBelongs to Idaho National Engineering Laboratory.

5. Basic Science and Technology

The Chemical Technology Division's research and development efforts impact a number of diverse sciences, including biotechnology, engineering, chemistry, and physics. Fundamental and applied studies in the area of biotechnology focus on such areas as microbial solubilization of coal, the biological conversion of light into energy, immobilization of enzymes, and investigations into the kinetics of enzyme processes. Historically, this division has been at the forefront of separations research in the nuclear field; chemistry research efforts build upon this expertise in investigations of the actinide elements. The principal focus of the division has long been in the area of engineering because it usually serves as a bridge between fundamental studies and their application to meaningful processes. Improved mass transfer in separations processes is the goal of engineering studies on the effects of external electric fields in multiphase systems. Developing an understanding of the fundamental interactions between solvents and solutes close to the supercritical point and exploring applications of continuous annular chromatography are other focuses, also aimed at improved separations processes. Materials research efforts in the division are concerned with basic thermodynamic studies of energy-related materials, including an oxide superconducting ceramic, particle growth and nucleation, and measurement of transport properties of fluids in the critical region. Developments in one of the above engineering programs were applied to an innovative new method of synthesizing precursor particles for ceramics. Finally, research into an exciting new area known as "cold fusion" was initiated just after midyear with a multidisciplinary group of engineers, chemists, and physicists.

5.1 Biotechnology Research

Research and development (R&D) in biotechnology is carried out in four major areas: (1) biological interactions with coal in which microbiological and enzymatic methods to solubilize coal are studied; (2) biophotochemical research involving studies on electron transport mechanisms in living and nonliving systems for the conversion of solar energy into chemical energy and biomolecular electronics; (3) enzyme kinetics, in which the interaction of enzymes with solid surfaces is examined, illustrated by the cellulase/cellulose systems; and (4) bioengineering research that provides the bridge between the more fundamental work and processing systems.

5.1.1 Biological Interactions with Coal

Pioneering investigations at ORNL into the biological treatment of coal have been continued. Work in this area was initiated following the observation that certain microorganisms are able to interact with lower-ranked coals. This activity can

be exploited for the conversion of lower-ranked coals to high-quality fuels, as described below. A parallel research initiative involves the use of isolated biological catalysts (i.e., enzymes) toward this end. This work will ultimately contribute to the design of new, efficient technologies for coal utilization.

Microbial Coal Conversion

Some microorganisms modify lignites and bituminous coals, transforming them to a water-miscible liquid product. The principal product of this microbial activity is generally characterized as a heterogeneous, high-molecular-weight, acid-precipitable, polar material which is partially oxidized and which has a high oxygen content. Little information has been available on other product components or on the fate of biosolubilized coal in cultures; nor have the potential uses of the bisolubilized product been explored.

Preliminary work during this period focused on the characterization of a microbial solubilization product derived from a chemically pretreated subbituminous coal.¹ The test substrate was Wyodak subbituminous coal that had been oxidized with nitric acid. Two fungi previously identified as highly active against coal, namely *Candida* sp. ML13 and *Paecilomyces* sp. Li, were used in these tests.

Cultures of either organism grown in the presence of coal produced a high-molecular-weight, oxidized, oxygenated, presumably polyaromatic, fraction which was evident in aqueous samples prepared by acid precipitation of spent culture medium. The high-molecular-weight fraction may be at least partially derived from lower-molecular-weight material detected in dioxane extracts of the nonacidified cultures. The basis for its apparent acid-catalyzed polymerization is not known. Acidification of *Candida* cultures yielded increased amounts of low-molecular-weight material (in addition to the high-molecular-weight fraction). This behavior was not seen in *Paecilomyces* cultures, suggesting differences in product composition between the two organisms.

Elemental and ^{13}C -NMR spectroscopic analyses indicated that the high-molecular-weight product is enriched in oxygen bound to carbon. The oxidation level of the product increased over time, suggesting further metabolism of the biosolubilized coal in vivo, within these fungal cultures. Preliminary findings² suggest that this oxidized product may be converted to readily utilizable fuels via anaerobic microbial metabolism.

A mixed bacterial culture was developed that was apparently able to produce methane and carbon dioxide from biosolubilized coal (Fig. 5.1). The depolymerization and further degradation of the polymeric material contained within the solubilized coal substrate was rate-limiting for the overall conversion.

The work described above provides a basis for the development and optimization of a two-step process for coal conversion to methane. Future research will concentrate on development of a process for the direct anaerobic microbial conversion of coal to useful products.

Enzymatic Coal Conversion

Most of the research on biological solubilization of coal is still associated with direct microbial action on low-ranked coal particulates in an aerobic environment in which the solubilization product is a complex aqueous solution of predominantly aromatic compounds that cover a wide range of molecular weights. However, the emphasis of the recent research at ORNL has been on anaerobic solubilization of subbituminous and bituminous

coals utilizing both microorganisms and in vitro enzymes.

Earlier research indicated that, under some circumstances, enzymes used in vitro can also affect the solubilization or liquefaction of coal. Such a bioprocessing approach is of great interest, since potentially both oxidative and reductive enzymes could be used in either aerobic or anaerobic processes, thus allowing a variety of bioprocessing options. The use of an organic solvent instead of an aqueous medium in conjunction with the enzyme solubilization process may also be possible. Such a system may provide an environment that interacts more effectively with the coal particulates and is more conducive to a nonpolar solubilization product which would have properties that are similar to conventional thermal/chemical coal liquefaction products.

The use of isolated enzymes for coal solubilization has been further investigated, with the emphasis on enhancing enzyme activity, especially in organic solvents.^{3,4} Several oxidative enzymes have been studied for the solubilization of low-ranked coals (lignite and subbituminous) in both aqueous and hydrous organic media. Peroxidase has been shown to increase coal solubilization, particularly within an organic medium. However, the anaerobic solubilization of coal may be even more important than the oxidative interaction, especially if molecular hydrogen can be used at moderate temperatures.

Anaerobic enzymatic coal solubilization has now been demonstrated. The catalyst used in this work was hydrogenase, a reductive enzyme, which was complexed to polyethylene glycol. The stabilized enzyme was shown to interact with bituminous coal under a hydrogen atmosphere within a benzene matrix (Fig. 5.2). The product generated during this treatment was quite different from that produced under oxidative conditions.

Both oxidative and reductive enzymes can thus accelerate the solubilization of coal in organic media. The reacting systems are very complex, with numerous important operating parameters. In many cases, the effects are measurable but minimal and cannot yet be the basis for process development until additional research is carried out. The use of a stable hydrogenase in an organic medium under anaerobic conditions for the solubilization of higher-ranked coals shows the most promise, with the resulting product being much less polar than the oxidative product.

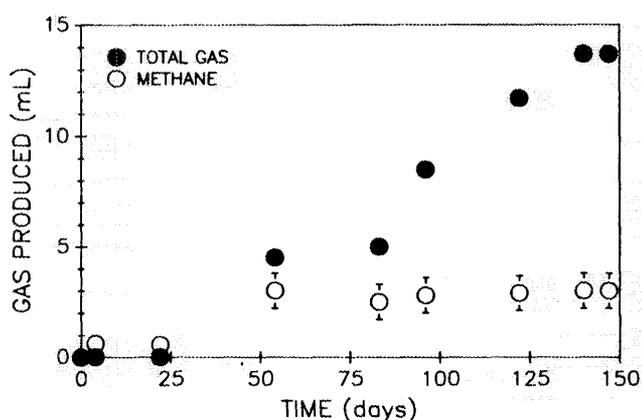


Fig. 5.1. Biogas production from a culture grown on solubilized coal.

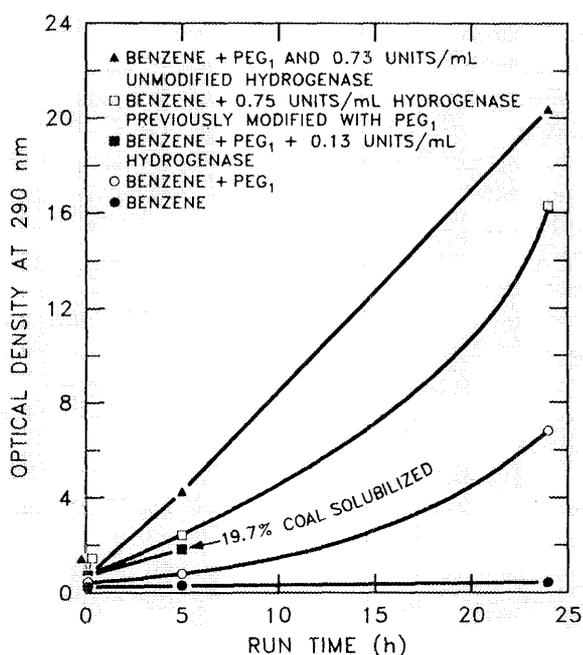


Fig. 5.2. Solubilization of Illinois No. 6 bituminous coal at 30°C in a hydrogen atmosphere at ambient pressure.

5.1.2 Biophotochemistry

Photosynthesis is vectorial photochemistry. Light quanta that are trapped in photosynthetic reaction centers initiate a primary electron transfer reaction that results in the spatial separation of electrostatic

charges across the photosynthetic membrane. The vectorial nature of photosynthesis lies in the intrinsic orientation of the reaction centers embedded in the membranes (See Fig 5.3). Electron flow is from the inner membrane surface of the flattened sac-like vesicles to the outer surface. It has been shown that colloidal platinum can be precipitated onto the surface of photosynthetic membranes such that water can be photobiocatalytically split into molecular hydrogen and oxygen upon illumination with light of any wavelength in the chlorophyll absorption spectrum. Since no electron mediator such as ferredoxin or methyl viologen is present, the colloidal platinum must be precipitated sufficiently close to the Photosystem I reduction site to allow interfacial electron transfer from the membrane to the platinum. The presence of a reticulated network of platinum particles embedded in the chloroplast matrix suggests that metal-like properties can, at least partially, be imparted to the chloroplasts. The property investigated in this work was the photoconductivity of the material, which was measured by the flow of photocurrent through the platinized chloroplasts from a point of laser irradiation to a linear platinum wire electrode.

A novel photobiophysical phenomenon was observed in isolated spinach chloroplasts that were metallized by precipitating colloidal platinum onto the surface of the thylakoid membranes. (See Fig. 5.3 for a schematic illustration of the structure of the surface-modified photosynthetic membranes.) A two-point irradiation and detection system was constructed in which a helium-neon laser was used

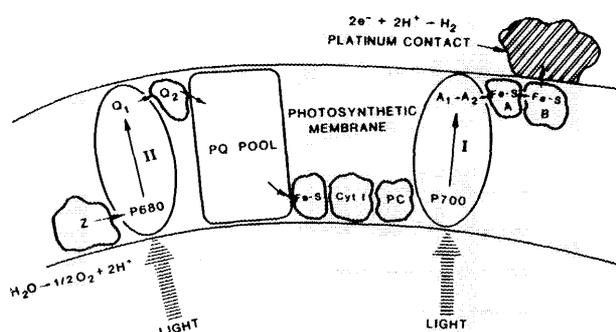


Fig. 5.3. Schematic illustration of metallized photosynthetic membrane. Colloidal platinum was precipitated onto the surface of photosynthetic membranes in such a way that electrical contact with the reducing end of Photosystem I was achieved.

to irradiate the platinized chloroplasts at varying perpendicular distances from a single linear platinum electrode in pressure contact with the platinized chloroplasts. A sustained steady-state vectorial flow of current from the point of laser irradiation to the linear wire electrode was measured. The absolute value of the measured photocurrent and the maximum perpendicular distance between the point of irradiation and the linear wire electrode depended critically on the experimental procedure used to prepare the platinized chloroplasts.

A schematic illustration of the experimental arrangement is shown in Fig. 5.4. This is a cross-section of the entrapped platinized chloroplasts and linear electrode viewed parallel to the axis of the electrode. The helium-neon laser and front-surface mirror are mounted on a linear translation stage whose position is controlled by a stepper motor. Type-C chloroplasts were prepared according to the procedure of Reeves and Hall.⁵ In this procedure, the chloroplast envelope is osmotically ruptured, exposing the photosynthetic membranes to the external aqueous medium. Solutions of chloroplatinic acid neutralized to pH 7 with NaOH were prepared. The amount of precipitated platinum could be controlled by adjusting the concentration of hexachloroplatinate.

A 5-mL suspension of spinach chloroplasts (containing 3 mg of chlorophyll) was used for these experiments. Platinization of chloroplasts is feasible because the chemistry of hexachloroplatinate allows it to be converted to metallic platinum at pH 7 and

room temperature. These experimental conditions preserve the functionality of the isolated chloroplasts. The platinized chloroplasts were entrapped on fiberglass filter paper (Millipore, AP40) and were moistened with Walker's assay medium which contained the chloroplast suspension. A silver/silver chloride reference electrode was placed in pressure contact with the filter paper; a single 0.25-mm platinum wire was placed in pressure contact with the entrapped platinized chloroplasts. The electrodes, platinized chloroplasts, and filter paper were held together with Lucite® plates and compression screws. In order to prevent the electrochemistry of atmospheric oxygen from interfering with cathode reactions by providing alternate pathways, the entire assembly was placed in a small glass chamber sealed with O-rings. The O-rings were pierced to allow the passage of small wires for establishing electrical contact with the electrodes. Helium gas flowed through the chamber to flush out atmospheric oxygen. After ~45 min, the oxygen concentration of the chamber was <~10 mg/L.

The platinum precipitation step was performed in a water-jacketed reactor cell maintained at 20°C. Molecular hydrogen was passed over the headspace of the reactor while the chloroplast suspension was gently stirred in a neutral hexachloroplatinate solution with a Teflon®-coated magnetic stirrer. Purge times of 30 to 60 min were used. After incubation and precipitation, the reactor chamber was opened to air, the contents were filtered onto the filter paper, and the cell was assembled as described above. This coprecipitation step was essential.

The novel observation of this research is the effect of the precipitated platinum on the photoconductivity of the chloroplast matrix. Figure 5.5 illustrates the current-vs-distance profile for a sample that contains 1.3 mg Pt/cm². Each data point represents a true steady state flow of current. Although steady state was achieved within a few minutes of each change in the laser beam position, a sustained current could be observed for hours. Each data point represents a dwell time of 15 to 20 min.

It can be seen in Fig. 5.5 that the photocurrent has a maximum value when the laser beam is directly over the linear platinum wire. The degree to which the laser beam can be moved from the wire and still allow photocurrent to be observed depends on the amount of platinum precipitated.

The origin of the photocurrent can be explained as follows. It has been previously shown that

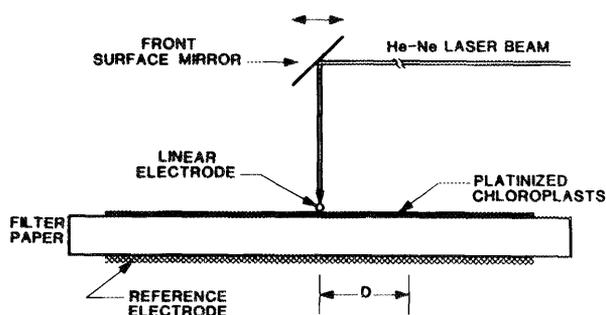


Fig. 5.4. Schematic illustration of the experimental setup. Shown is a cross-sectional view of the photobioelectrochemical cell viewed parallel to the linear platinum wire electrode. D is the perpendicular distance from the point of irradiation of the helium-neon laser beam to the linear wire electrode.

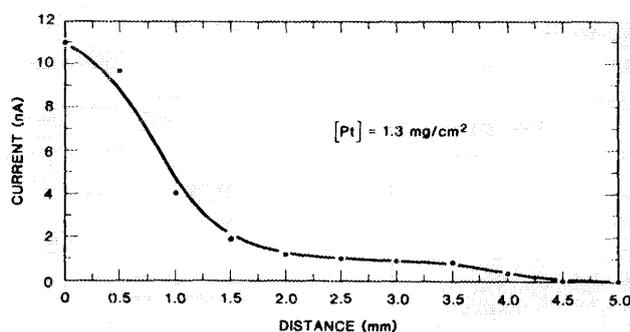


Fig. 5.5. Photocurrent-vs-distance profile in platinized chloroplasts; platinum concentration = 1.3 mg/cm^2 . Note that photocurrent can be observed at a distance of 4 mm. This represents photocurrent through the reticulated matrix of colloidal platinum embedded in the composite biomaterial structure.

colloidal platinum can make direct electrical contact with the reducing end of Photosystem I of photosynthesis. This contact was demonstrated in two ways: by the photocatalytic evolution of molecular hydrogen and through the observation of photocurrent in a sandwich-like photobioelectrochemical cell. The direction of flow of photocurrent in the sandwich-like cell was consistent with the molecular architecture of the photosynthetic membranes. Upon illumination, the platinum electrode in pressure contact with the platinized chloroplasts swung negative with respect to the silver/silver chloride electrode that was in pressure contact with the electrolyte-impregnated filter paper.

A reasonable model, based on the generally accepted structure of photosynthetic membranes, is that the colloidal platinum which is precipitated onto the external surface of the thylakoid membranes forms an isopotential surface whose distance is determined by the connectivity of the reticulated colloidal particles that comprise the metallic component of the chloroplast-metal composite matrix. This distance is a statistically determined parameter whose average value is decided by the nature of the platinum precipitation process. When platinum is precipitated it does so in a nonspecific manner on the external surface of the thylakoid membranes. There is, however, an electrostatic interaction between the negative charge of the hexachloroplatinate ion and the local positive charge of the lysine residues making up the polypeptide composition of the Photosystem I proteins.

The key observation of this research is that electrical contact with the reducing end of Photosystem I can be achieved by precipitating colloidal platinum in the presence of isolated chloroplasts. The presence of the platinum has a significant effect on the photoconductivity of this new metal-biological composite material. The technological significance of this work is that the photosynthetic reaction centers are nanometer-sized structures with picosecond switching times. This work demonstrates that the electron transport chain of photosynthesis can be electrically contacted and that the larger structural matrix of the platinized chloroplasts can possess enhanced photoconductivity.

5.1.3 Enzyme Kinetics

Fungal cellulases that catalyze the hydrolysis of insoluble cellulose to glucose are composed of cellobiohydrolases, endoglucanases, and β -glucosidases. They can be used for the hydrolysis of lignocellulosic materials to glucose; however, under industrial conditions, there is incomplete hydrolysis of the substrate, and much of the enzyme remains adsorbed to the lignocellulosic residue. Recovery and reuse of adsorbed cellulase components are recognized as important issues for energy conservation and reduction in costs associated with the enzymatic hydrolysis of cellulose.

Many compounds have been tested for their ability to elute cellulase protein components that are bound to microcrystalline cellulose. Sodium hydroxide, guanidine hydrochloride (Gdn HCl), and urea are the best eluants (>90% of bound protein eluted), but the disadvantage to their use is that cellulase activity was inactivated by these eluants. Since high concentrations of Gdn HCl (4 M) and urea (6 M) are required to effect elution of cellulase components from cellulose, the eluted components are denatured, or unfolded, and hence inactivated. Although many Gdn HCl-denatured proteins are capable of spontaneous refolding, the conditions under which Gdn HCl-denatured cellulase enzyme components are capable of renaturation and reactivation have not been studied.

The cellobiohydrolase I (CBH I) component comprises as much as 60% of the total protein in a crude cellulase preparation. Thus, the majority of cellulase protein bound to a cellulosic residue, at the completion of hydrolysis, will be CBH I. The recovery and subsequent reuse of CBH I are, therefore, particularly important, but the use of denaturants as eluants requires that the eluted

enzyme can be subsequently renatured with, hopefully, full recovery of activity.

Denaturation and Inactivation of CBH I by Gdn HCl

The key result of this work is that although the CBH I component of cellulase is partially denatured (unfolded) and inactivated by high concentrations of Gdn HCl (4 M), CBH I can be fully renatured and reactivated after removal of the eluant, as judged by fluorescence spectroscopy and activity measurements.

The fluorescence spectrum of a protein is an indication of its physical state and can be used to monitor denaturation, or unfolding, of its native structure. The effect of 4.5 M Gdn HCl on the fluorescence of CBH I is shown in Fig. 5.6. These data show that only a partial, or local, unfolding of CBH I occurs, even at high concentrations of denaturant (Fig. 5.6), and that the extent of unfolding is largely complete after 30 min. This would seem likely since this enzyme has been reported to contain 12 disulfide bridges. An extensive decrease in fluorescence or the unfolding of CBH I requires, presumably, the addition of a reducing agent, such as dithiothreitol or β -mercaptoethanol.

Almost complete inactivation of CBH I occurred at 0.25 M Gdn HCl. It appears, therefore,

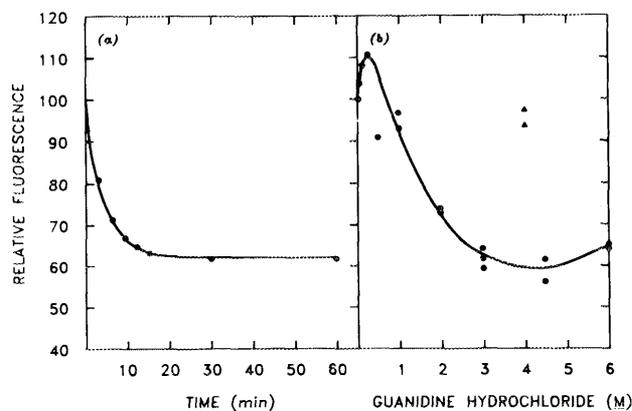


Fig. 5.6. The effect of Gdn HCl on the fluorescence of CBH I. Time course of the decrease in tryptophan fluorescence at 350 nm of CBH I by 4.5 M Gdn HCl: (a) effect of Gdn HCl concentration; (b) fluorescence measured 1 h after incubation with CBH I. The closed triangles represent the fluorescence of 4 M Gdn HCl-treated CBH I after its renaturation by gel filtration. Excitation of CBH I by 280 nm.

that low concentrations of Gdn HCl inhibit CBH I, resulting in an apparent loss in activity rather than partial unfolding, or denaturation, which requires much higher concentrations of this denaturant.

The adsorption kinetics for the binding of CBH I to Avicel (microcrystalline cellulose), given in Fig. 5.7, were completely abolished in the presence of 4 M Gdn HCl. Consequently, there was no hydrolysis of Avicel by CBH I in the presence of this concentration of Gdn HCl. The extent of binding of CBH I to Avicel decreased as the concentration of Gdn HCl present in the reaction mixture increased; binding was completely prevented by 2 M Gdn HCl. The finding that 4 M Gdn HCl prevents the binding of CBH I to Avicel suggests that partial unfolding, or denaturation, of CBH I, resulting in decreased tryptophan fluorescence, is the mechanism whereby binding is prevented.

Renaturation and Reactivation of CBH I

After treatment of CBH I with 4 M Gdn HCl, the solution was dialyzed overnight at 4°C to remove the denaturant. The dialyzed, denatured enzyme was assayed for its ability to hydrolyze *p*-nitrophenylcellobioside (PNPC) and Avicel to determine whether renaturation had occurred. The

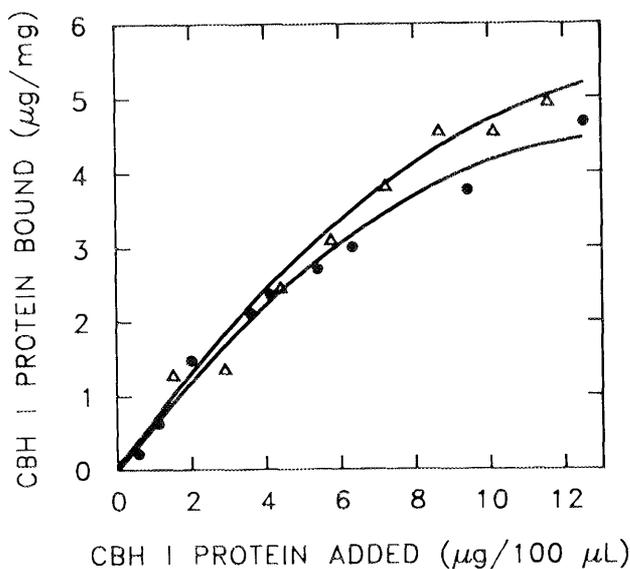


Fig. 5.7. The adsorption of native and renatured CBH I to (1% w/v) microcrystalline cellulose (Avicel). Native (●) or renatured (△) CBH I.

observed kinetics of the reaction of Gdn HCl-denatured CBH I (Fig. 5.8) indicate that maximum recovery of activity is obtained after 3 h. Also, there was no obvious correlation between the yield of reaction and the concentration of CBH I between 0.15 and 1.0 mg/mL.

The mass action constant (K_m) and the Avicel-binding capability of the renatured enzyme were also measured. The data in Table 5.1 indicate that there is little difference between the specific activity and K_m of the native and renatured CBH I with respect to PNPC hydrolysis. Renatured CBH I was similar to the native enzyme in its Avicel-binding and hydrolysis properties. It appears, therefore, that in terms of their catalytic activity, native and renatured CBH I are very similar to each other. The relative fluorescence of renatured CBH I (after denaturation with 4 M Gdn HCl) was ~95% of the native enzyme (Fig. 5.6), suggesting that the original conformation had been restored.

In conclusion, therefore, it has been shown that the purified CBH I component of *Trichoderma reesei* cellulase is partially unfolded at high concentrations

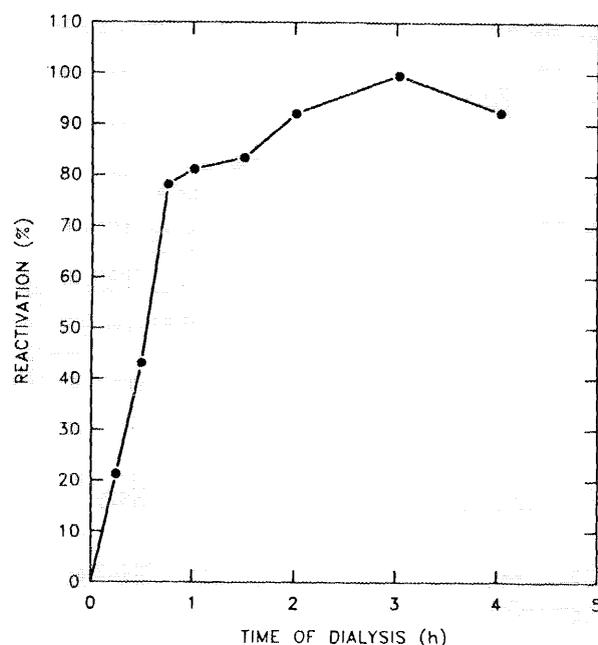


Fig. 5.8. Kinetics of the reactivation of Gdn HCl-denatured CBH I. Gdn HCl-denatured CBH I (350 $\mu\text{g/mL}$) was dialyzed against 50 mM sodium acetate buffer (pH 5.0). At the indicated times, 100- μL samples were withdrawn from the dialyate and assayed for their PNPC-hydrolyzing activity.

Table 5.1. Comparison of the specific activity and K_m of native and renatured CBH I with respect to PNPC hydrolysis

	Native	Renatured	Native ^a
Specific activity (nmol PNP/min per mg protein)	52	51	66
K_m , mM	2.2	2.9	n.d. ^b

^aSource of CBH I: Genencor.

^bn.d., not determined.

(≥ 2 M) of Gdn HCl. However, lower concentrations (0.25 M) are only required to abolish activity. Since this loss in CBH I activity by treatment with Gdn HCl cannot be correlated with a decrease in its intrinsic tryptophan fluorescence, inhibition of CBH I does not appear to be related to a gross conformational change in this enzyme. However, the loss of Avicel-binding capability of CBH I clearly can be correlated with at least a partial unfolding of its tertiary structure. The fact that CBH I can be renatured with full recovery of activity and conformation suggests that Gdn HCl can be used to elute cellulase from undigested cellulosic residues. The eluted enzyme can be subsequently reactivated by removal of the denaturant and hence reused.

5.1.4 Bioengineering Research

These projects have continued efforts in the study of immobilized and novel biocatalysts and of columnar bioreactors.

Biocatalysts

Production and characterization of gel-entrapped biocatalysts

Natural hydrocolloidal gels have been used to entrap biocatalysts such as microbes or enzymes within beads. In order to improve mass transfer, small uniform beads of <1-mm diam can be produced by the forced flow of the unsolidified gel through a small nozzle under an imposed vibration. These biocatalysts have been successfully used in

columnar reactors, as described below. In order to predict and scale up the overall kinetics of these systems, the mass transfer of substrates within the gel must be measured and correlated.

A transient equilibration technique was used to measure solute diffusion within gel beads of different compositions.⁶ Previously, it was determined that the diffusion coefficient of glucose in the gel approaches that in water for *k*-carrageenan gels of 1 to 4%. Other solutes have now been determined to give similar results (Table 5.2). Most importantly, the presence of additives such as microorganisms or inorganic oxides in the bead does decrease the diffusion coefficient. This was correlated for microorganisms from experiments using inactive *Pseudomonas aeruginosa* (Fig. 5.9).

Enzyme Solubilization in Organic Media

Increasing interest in biocatalysis in organic media has prompted research into methods for

Table 5.2. Comparison of diffusion coefficients of several solutes in 4% *k*-carrageenan gel beads and in water at 30°C

Solute	Molecular weight	Diffusion coefficient (10 ⁶ cm ² /s)	
		In gel ^a	In water
Ethanol	46	10.1	11.0
Glucose	180	6.7	6.8
L-tryptophan	204	5.8	6.6
α -lactalbumin	14,200	1.2	1.0

^aStandard deviation = $\pm 0.5 \times 10^{-6}$ cm²/s.

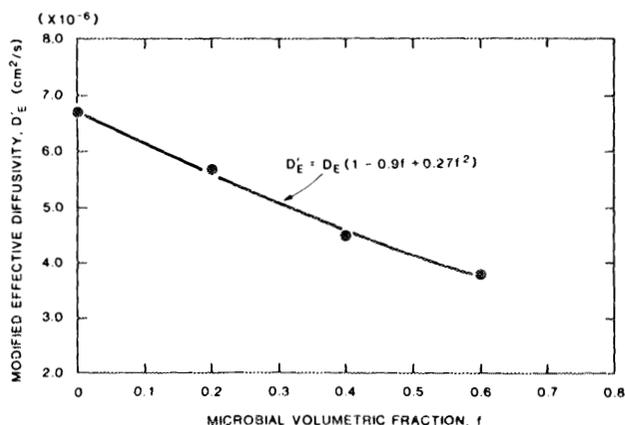


Fig. 5.9. Effect of microbial loading on glucose diffusion at 30°C in 4% *k*-carrageenan gel beads.

solubilizing enzymes in organic solvents so that they can retain their activity. An entirely new approach has been developed for the enhancement of enzyme solubilization in organic solvents in the absence of water. The approach is to chemically complex enzymes to support molecules that are less polar, thereby rendering them more hydrophobic and more soluble in organics. This can be done by binding the enzymes with a bridging chemical that should not affect activity. The bridging chemicals of interest here are triazine dyes, which are insoluble in organic solvent but can be bound to polyethylene glycol (PEG), a water-soluble polymer possessing some solubility in organic solvents.⁷ The PEG-dye-enzyme complex is being investigated as a vehicle to enhance enzyme solubility and activity in organic media.

The solubility of cytochrome C and of peroxidase in benzene could be increased using the polymer-dye complex. For example, the PEG-dye-cytochrome C complex was formed in aqueous buffer utilizing ~20 mg/mL of the PEG-dye complex and 5 mg/mL of the enzyme. The material was then dried and dissolved into pure benzene. Material that did not dissolve was removed by centrifugation and the enzyme concentration determined spectrophotometrically. Table 5.3 shows the significant increases in solubility with these enzymes. These enzymes maintained activity when back extracted into aqueous buffer.

Preliminary tests were performed with reduced cytochrome C in benzene to anaerobically interact with polyaromatic compounds including some of the components of coal. Approximately 150 mg/mL of powdered (~60 mesh + 100 mesh) bituminous coal was then added to the benzene solution in a shake flask at 30°C under a hydrogen atmosphere. After 4 h, it was noted that solubilization of coal components was enhanced from 8 to 18%.

Advanced Bioreactor Systems

Ethanol Production in a Fluidized-Bed Bioreactor (FBR)

The production of ethanol from dextrose by immobilized cells of *Zymomonas mobilis* has been shown previously⁸ to allow dramatic increases in productivity (50 to 100 g EtOH L⁻¹ h⁻¹) and yield (98% of theoretical) using this system. Current efforts focus on modeling and scaleup. This

Table 5.3. Solubility of enzymes in benzene

Enzyme	Concentration of enzyme (mg/mL)	Concentration of PEG-dye (mg/mL)	Activity of enzyme in benzene (U/mL)
Peroxidase	1	0	0.00
	1	1	0.76
	1	5	2.26
	1	10	3.06
Cytochrome C	1	10	<u>Solubility (mg/mL)</u> 0.8
	2.5	25	0.9
	5	50	1.2
	5	0	0.05

fermentation was carried out using immobilized *Z. mobilis* in the 7.6-cm-ID FBR. The bacteria were immobilized within small uniform beads (1.25-mm diam) of *k*-carrageenan at a high cell loading (<15 g dry wt/L). The actively fermenting bed could be operated without significant slugging at much lower liquid velocities than the 2.5- or 3.8-cm-ID columns used previously. The gas disengagement was better, and the gas holdup was <3%, as expected from the hydrodynamic tests described below. Productivities in the active portion of the reactor were ~ 70 g EtOH L⁻¹ h⁻¹, comparable to those in the smaller reactor.

Estimates of back mixing from our measurements have been incorporated into a total columnar reactor model and have improved the accuracy of the model with respect to experimental data. The model incorporating reaction and axial dispersion in a columnar bioreactor was used in a computer integration routine. This routine used complex kinetics, such as Monod kinetics, with product and substrate inhibition—appropriate for ethanol fermentation with *Z. mobilis*. The effect of the tapered entry section has been included, and the conversion profile matches that of the experimental data.

Solvent Fermentation with Immobilized Cells

A bioconversion of considerable interest is the anaerobic fermentation of sugars into organic solvents, such as acetone and butanol by *Clostridia* sp. Previous experimental results demonstrated the

successful use of oxygen-reducing biocatalytic cell fragments for rapid startup of anaerobic solvent fermentation systems, but these results suffered from the conversion of less than half of the 40 g/L glucose provided. A different microbial strain of *C. acetobutylicum* (a known active solvent producer provided by H. I. Adler of Oak Ridge Associated Universities) is being used to extend these results.

Two batch fermentations of 40 g/L of glucose were performed with the active solvent-producing strain. In one fermentation, the pH was uncontrolled and a final glucose concentration of 13 g/L was seen. In the run with the pH controlled at 5, the highest glucose conversion was observed, with a residual concentration of ~ 5 g/L out of a 40-g/L feed.

A continuous columnar reactor was set up with a biocatalyst containing both the immobilized *Clostridia* and the oxygen-reducing membrane fragments. The reactor was operated successfully with an oxygenated feed for >2 weeks. High gas formation rates were observed. Effluent glucose concentrations as low as 1 g/L were observed at the lower flow rates (1.3 mL/min). Acetone, butanol, and ethanol concentrations were >7 g/L, 8 g/L, and 3 g/L, respectively.

FBR Hydrodynamics

The incorporation of numerical estimates of the gas holdup and back mixing into the total columnar reactor model has considerably improved the accuracy in packed-bed models. It is expected that experimental measurements will allow similar

improvements in an FBR model. A model, nonfermenting, experimental three-phase system is being used to characterize and correlate these systems because of its capability to set the gas and liquid flow rates independently.

Electroconductivity is used to measure the gas holdup with this three-phase FBR system, using a nonintrusive probe constructed at ORNL. In essence, both the broth and the beads are equally conductive, due to the salts present, while the gas is not; thus, the conductivity decreases with the amount of gas present. In addition, the multiple axial probes can estimate the liquid axial dispersion coefficient from the response curves to a salt pulse. The results have been obtained with ~1-mm beads in both a 2.5-cm-ID and a 7.8-cm-ID column to examine the effects, particularly the wall effects, of scale on the system hydrodynamics. There are major qualitative differences in behavior between the 1- and 3-in.-ID columns when operated at the same superficial gas and liquid velocities. In the 2.5-cm-ID bed, the gas bubbles rapidly coalesce to fill the column diameter. This can cause slugging behavior, with increased back mixing and lower conversions. In the 3-in.-ID column, the bubbles reach an apparently stable size of 5-mm diam. This occurrence indicates operation in a turbulent regime where bubble coalescence and bubble breakup due

to shear are equal. The gas holdup in the larger system was always <3%. The correlation and the estimation of gas holdup in three-phase reactors have been explored. As expected, for liquid-solid fluidized beds the Richardson & Zaki equation⁹ works very well (Fig. 5.10). However, in fluidized-beds with low-density solids, the experimental observation that the liquid holdup remains constant for a range of gas velocities has important ramifications on the correlations used. These include a linear dependence of the particle-settling velocity on the liquid velocity. Some speculation on the underlying cause of this phenomenon has been presented but requires further research. The drift-flux model has been shown to correlate the experimental data very well (Fig. 5.11). Correlations of these types, when combined with the wake model, as described elsewhere,⁹ allow the accurate prediction of the gas-phase holdup in three-phase fluidized beds with low-density solids, such as biocatalysts.

Axial dispersion coefficients were calculated from the response to a salt tracer by the method-of-moments. Particle Peclet numbers (a ratio of convective to dispersive forces) are defined as

$$Pe = u_L \frac{d_p}{D}$$

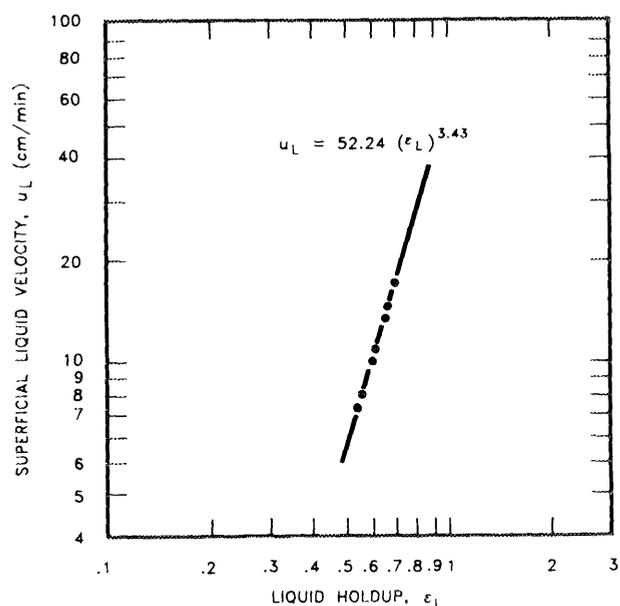


Fig. 5.10. Superficial liquid velocity vs liquid holdup in a two-phase solid-liquid FBR. Solid line represents data fit by correlation of Richardson and Zaki.⁹

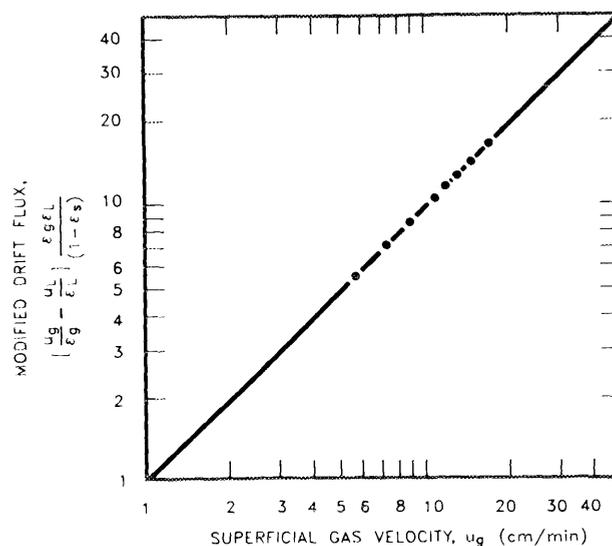


Fig. 5.11. Plot of modified drift-flux velocity, $v_{g,L}(1-\epsilon_s)$, vs the superficial gas velocity, u_g . This plot uses all three-phase FBR data in Table 5.2.

where d_p is the particle diameter, D is the liquid axial dispersion coefficient, and u_L is the superficial liquid velocity. The Peclet numbers are estimated to be between 10^{-1} and 10^{-2} and are comparable to those measured in the narrow column under similar conditions. The values obtained have been compared with several literature correlations, which range over an order of magnitude and bound the data. It should be noted that these literature correlations were developed with glass or metal beads, not with the near-neutral-density beads used here. Fluidized biological systems will require low-density biocatalysts and possibly new data and correlations for phase holdup and dispersion, such as presented in part here.

These results and the ones presented previously indicate that larger-diameter systems for high-productivity bioconversions will have improved operability over the smaller fermentation FBRs that we have successfully tested. Slugging behavior will not be a problem in the larger systems, as the bubbles remain small relative to the 7.6-cm-column diameter. Likewise, the wall effects, which were seen to be very important in the hydrodynamics and bubble coalescence in the narrower column, are less important in the larger column. If correct, the hydrodynamics of the 7.6-cm column should scale to even larger systems. In addition, the gas holdup, which was measured here to be <3%, can be neglected in a larger fermenting FBR. This will simplify the modeling and scaleup of the fluidized-bed fermentor system. Nevertheless, the gas flow will have a significant effect on the overall system behavior, in part due to bed expansion lowering the solids fraction.

Biparticle FBR

Fluidized-bed bioreactors with immobilized cells can increase the productivity of fermentations for useful products. An additional advantage of the multiphase FBR is the potential to add new phases to perform additional functions beyond the three present previously. These three phases are a solid biocatalyst, a liquid containing the substrate and products, and a gas which can be a reactant and/or product. A new approach is to add a fourth phase, which exhibits extractive capabilities for the desired product, to this system to both enhance productivity and accomplish a simultaneous separation. This extractive phase could be an immiscible liquid or a solid. Some work has been performed with liquid

extraction but generally has been limited by the solvent selectivity and toxicity. This exploratory approach included using a solid adsorbent for product removal in an FBR.

An important consideration of the use of another solid phase is the selective removal of this phase from the four-phase FBR. This can be accomplished by exploiting a well-known property of FBRs: their ability to stratify on the basis of size or density. However, the adsorbent itself may not have the desired physical or hydrodynamic properties. This potential difficulty may be overcome by immobilizing the solid adsorbent as a dispersed phase within a gel bead; the size and/or density of this bead can then be controlled to give it hydrodynamic properties that are significantly different from those of the solid biocatalyst. For example, the adsorbent bead can be made denser so that it could be introduced at the top of the column and removed from the base of the FBR. This would result in a countercurrent flow of the adsorbent with respect to the upward-moving liquid phase.

A biparticle system has been demonstrated to separate while continuously adding the heavier particles as a slurry to the top of the column with a pump and removing them from the base of the column below the liquid-feed inlets. This column has a 1.3-cm-ID inlet section which tapers to a 2.5-cm-ID section. At the top, the column expands to a 7.6-cm ID to decrease the superficial liquid velocity and thus retain the solids.

The biparticle system consisted of gel beads of 4% *k*-carrageenan, which were color coded to allow for visual differentiation. The lighter red beads contain 3% Fe_2O_3 and are 1.2 mm in diam, while the heavier tan beads contain 5% CeO_2 and are 1.6 mm in diam. In one set of experiments, the red beads were fully fluidized in the 2.54-cm-ID section of the column by the liquid feed at 6.9 L/h. The heavier solids slurry was added to the top of the column at a total rate of 1 L/h (or 260 beads/min) and removed at a rate of 1 L/h from the bottom. The effect of gas addition is shown in Table 5.4.

These preliminary results indicate that essentially only the desired beads can be removed from this system while leaving the other solids in the fluidized bed. Gas bubbles increase the mixing of the bed and decrease the rate at which the heavier solids collect at the bottom of the column.

Preliminary experiments were also performed in cocurrent operation. In these experiments, the heavier tan beads were fluidized at a liquid flow of 11 L/h. The slurry of lighter red beads was added

Table 5.4. Effect of gas on separation of a biparticle fluidized-bed^a

Gas flow rate (L/h)	Slurry flow		Bead rate		Purity of beads removed (% tan beads)
	Added to top (L/h)	Removed at bottom (L/h)	Added to top (beads/min)	Removed at bottom (beads/min)	
0.0	1.0	1.0	260	170	100
0.8	1.0	1.0	260	98	99
1.2	1.0	1.0	260	58	100
0.0	0.75	1.43	344	290	97
0.1	0.75	1.43	344	130	99
0.0	1.0	2.0	430	450	99

^aNet liquid flow rate through FBR is 6.9 L/h; beads described in text.

at the base of the column. The red beads quickly rose to the top of the bed. The expansion of the FBR to 7.6 cm was still sufficient to retain both types of beads. When this section was eliminated, the red beads could be collected and removed from the top of the bed. Cocurrent operation is feasible but will apparently require a greater density difference than that which existed between this pair of beads.

Industry has developed interest in using advanced bioreactors for production of liquid oxychemicals, such as organic acids, solvents and ethanol. The production of organic acids should provide a good test of this new system. Lactic acid is the primary candidate because of the relative ease of cultivation of the microbes, the availability of some promising data on adsorbents for lactic acid, and significant industrial interest and the possibility of cooperative research. Activated carbon has been chosen as an initial adsorbent. Initial capacity tests have been performed on two activated carbons and one anionic adsorbent. At a concentration of 1% lactic acid, the Filtrasorb carbon is expected to have a capacity of >0.1 g/g.

5.2 Chemistry Research

This research includes investigations into the fundamental chemistry of the actinide elements, their analogs, and fission products and studies on the effect of macrocycles on solvent extraction separations of the lanthanides and actinides. Small-angle X-ray and neutron-scattering studies of actinide and analog polymers, together with low-temperature infrared studies of matrix-isolated actinides, serve as the current principal focus of this

work, as the solvent extraction work has been de-emphasized.

5.2.1 Chemistry of the Actinides and Fission Products

This continuing research effort in actinide/fission product chemistry is concerned with the behavior of these elements in various separations schemes. Because the scope of the program is broad, numerous activities encompass many techniques and considerations. Currently, these efforts have been focused on (1) spectroscopic studies of matrix isolated actinide species, (2) small-angle scattering of neutrons and X-rays from hydrous polymers, (3) synergistic effects of macrocycles in the solvent extraction of lanthanides and actinides, and (4) thermodynamic studies of lanthanide and actinide coordination chemistry in room-temperature molten salts. The emphasis in this overall effort has been shifting toward the first two areas, with an associated de-emphasis in the latter two. Details of each of these efforts are given below.

Low-Temperature Matrix-Isolation Studies

In our spectroscopic studies of actinide and related compounds, we seek an understanding at the molecular level of structure, bonding, and chemical and photochemical reaction mechanisms. In recent

years, we have turned to low-temperature methods, particularly matrix isolation spectroscopy, in order to gain better control of reaction processes and to stabilize short-lived intermediate species. A new high-resolution Fourier transform infrared (FTIR) spectrometer has been acquired and installed for these studies, and the ability to generate and trap high-temperature species is being developed. Topics recently or currently under investigation include the reaction between uranium hexafluoride and water, photochemical reduction of uranyl ion, photo-induced electron transfer from solid xenon to metal hexafluorides, hydrogen bond interactions between metal hexafluorides, and hydrogen halides.

The hydrolysis of UF_6 was considered a possible means of producing matrix-isolated uranyl species for photochemical examinations. The reaction has been studied at ambient temperatures in the gas phase and at low temperatures in solid rare gas matrices and in thin films of pure reactants. The hydrolysis proceeds through several stages and intermediates and is relatively slow, even in the gas phase.

In related work, we have found that uv irradiation of UF_6 in water-doped xenon matrices produced a charge transfer species, UF_6^- . The molecular anion was positively identified by its infrared absorption spectrum, as shown in Fig. 5.12. The absence of any absorption attributable to a cation indicated that the corresponding positive charge was shared among several xenon atoms as a self-trapped hole. The "trapping" of the positive charge results in a charge separation that enables the system to store optical energy; the energy is recovered as fluorescence when the ions recombine. In undoped xenon, rapid recombination of charges prevents any observable buildup of the product species; thus, little energy storage occurs. We are investigating the impurity role in trapping the positive charge and are examining the charge transfer process in metal hexafluorides of varying electron affinities.

The complexes of hydrogen fluoride with sulfur hexafluoride and molybdenum hexafluoride have been studied using matrix-isolation infrared spectroscopy. The primary objective in characterizing the different complexes between HF and MF_6 is to examine the role that the hexafluorides play in determining whether HF serves as a Bronsted base ($\text{MF}_6 - \text{HF}$) or as a Lewis base ($\text{MF}_6 - \text{FH}$). Both types of HF complexes have been considered in numerous theoretical and

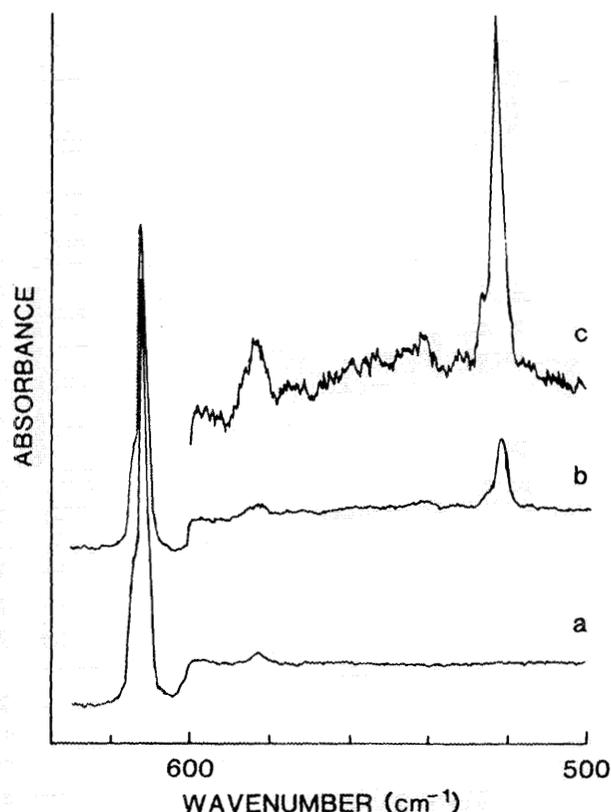


Fig. 5.12. Infrared spectra of UF_6 in xenon matrix before (a) and after (b,c) photolysis. The peaks around 610 cm^{-1} are due to UF_6 ; the peak that appears at 520 cm^{-1} after photolysis is due to UF_6^- .

experimental studies because of the role which hydrogen bonding and Lewis acid-base interactions exert in determining the physical properties of a wide array of complexes. A second objective of this study is to estimate the proton affinity of the hexafluorides. Earlier studies have shown a direct correlation between the frequency shift of the HF stretching mode in the HF complex and the base's proton affinity if the basic sites are similar. The proton affinity of SF_6 has not been accurately determined and yet is a very important parameter for kineticists since SF_6 is used extensively as a thermal electron scavenger in ionized systems. A third objective in this investigation is to identify and better understand these HF complexes in two important chemical systems: the hydrolysis of transition metal hexafluorides and MF_6 -doped HF chemical lasers.

A preliminary analysis of the infrared spectra of the complexes of HF with SF₆ has been completed. Based on the concentrations of the HF and SF₆, the annealing behavior, and the band positions of the HF modes, the 1:1 (SF₆:HF), 1:2, and 1:3 complexes have been identified. The normal 3919 cm⁻¹ position of the HF stretching mode, as seen in Table 5.5, is shifted in the 1:1 complex by only 14 to 18 cm⁻¹, which indicates a very weak interaction. While this weak interaction makes SF₆ an ideal fluorine atom source for HF chemical lasers, the weak interaction does not shift the frequencies of complexed SF₆ modes enough from the isolated SF₆ modes to be detected. In addition, the weak interaction accompanied by the limited diffusion of SF₆ in solid argon prevents determining the exact number of SF₆ molecules present in each complex (which here is assumed to be one). Based on earlier studies and the band positions of the 1:1 and the 1:2 complexes, SF₆ and HF form hydrogen-bonded complexes, SF₆-HF and SF₆-HF-HF. The 1:3 complex involves a HF cyclic polymer that is slightly perturbed by the hexafluoride. The position of the SF₆ with respect to the polymer cannot be determined from the IR spectra.

The initial interpretation of the HF complexes with MoF₆ based on infrared spectra has indicated that the MoF₆ and HF system is much more complicated than the SF₆ and HF system. Using the chlorine and HF complexes from an earlier matrix IR study as models, two 1:1 complexes, MoF₆-HF and MoF₆-FH, have been observed. Based on the IR band intensities, the anti-hydrogen-bonded complex is clearly the more stable arrangement

under these experimental conditions. In addition to the 1:1 complexes, a single 1:2 complex was observed and assigned to MoF₆-FH-FH, and the 1:3 and 1:4 cyclic complexes were characterized. Again, no MoF₆ modes due to the HF complexes were observed. The interaction in the MoF₆-FH complex is considerably stronger than that in the SF₆-HF complex. This stronger MoF₆ interaction greatly hinders its usefulness as an inert fluorine source for HF chemical lasers.

Small-Angle Scattering

Small-angle X-ray scattering (SAXS) experiments have been performed on Zr(IV) hydrous polymer suspensions of 0.035 to 1.0 M. A monodisperse species of a 0.487-nm radius of gyration has been observed for all of these solutions and is consistent with the existence of tetrameric Zr(IV) units. As the Zr(IV) concentration was increased from the lower limit, interference from interparticle scattering was observed in the Guinier plots of the data, as shown by the nonlinear portion of Fig. 5.13. The position at which the interference occurs, however, is consistent with the interparticle separation for each solution.

These data for monodisperse polymers have provided a basis for testing particle geometries by intensity calculations which are sensitive to changes in the geometry. Intensity distributions for spherical, ellipsoidal, and cylindrical particles have been fitted to the observed data. These distributions are based on the intensity of a scattered X-ray of given

Table 5.5. HF stretching mode band positions (in cm⁻¹) in hexafluoride/HF complexes produced on codeposition of hexafluorides and HF or DF with excess argon at 12 K

System				Assignment
SF ₆ + HF	SF ₆ + DF	MoF ₆ + HF	MoF ₆ + DF	
3905	2866	3911	2871	MF ₆ -HF
3903				"
3901				"
		3883	2848	MF ₆ -FH
		3879	2844	"
3818	2797			MF ₆ -HF-HF
		3777	2757	MF ₆ -FH-FH
3692	2712	3693	2712	MF ₆ -(HF) ₃
3690	2710			"
		3411	2525	MF ₆ -(HF) ₄

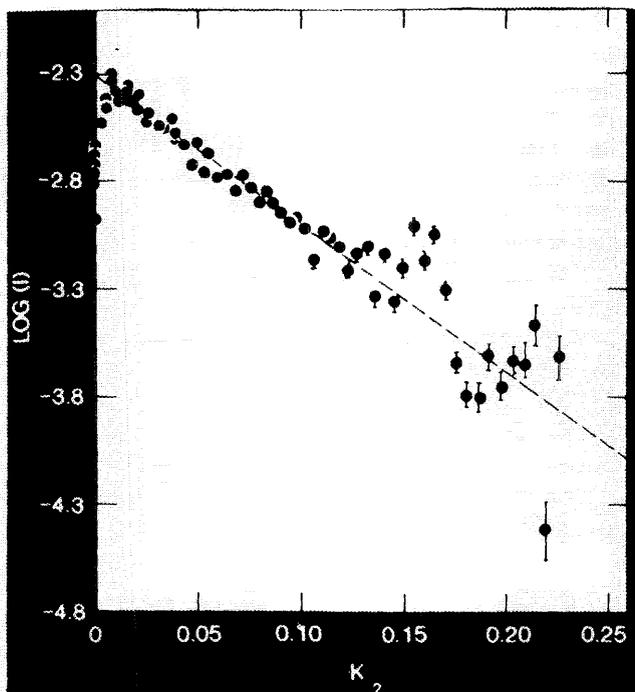


Fig. 5.13. Guinier plot of 0.035 M Zr^{4+} in aqueous solution showing monodisperse tetramers with radius of gyration = 4.87Å.

wavelength at a given angle for a particle of radius, R (for spheres), or diameter and aspect ratio (for ellipsoids), or diameter and length (for cylinders). The optimal fit was obtained for an ellipsoid of 0.487 nm and aspect ratio (major/minor axis ratio) = 2.8. These values are consistent with the shape and dimensions of a tetrameric Zr(IV) hydrous polymer unit, as shown in Fig. 5.14.

In collaboration with members of the Chemistry Division at the Argonne National Laboratory (ANL), small-angle neutron-scattering (SANS) experiments have been carried out on Pu(IV) hydrous polymers using the Intense Pulsed Neutron Source (IPNS) at ANL. The structures of Pu(IV) hydrous polymers were measured using SANS in aqueous media and in organic media (after solvent extraction). The scattering data from polymers occurring as aqueous colloidal suspensions indicate long, thin, rod-like particles. Pu(IV) hydrous polymers formed in the presence of uranium were shorter than those produced in the absence of uranium; this finding is consistent with earlier observations that uranium reduces the rate of Pu(IV) polymer formation. The Pu(IV) polymers were extracted into C_6D_6 solutions by alkyl esters of

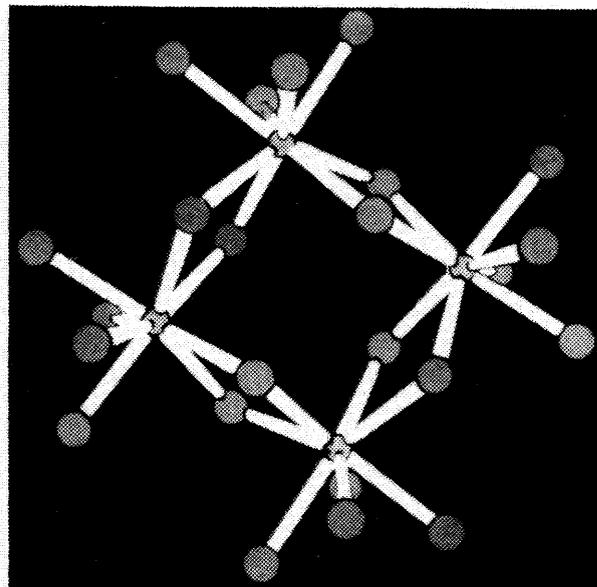


Fig. 5.14. Structure of Zr(IV) hydrous tetrameric units found in crystalline $Zr_4(OH)_8(H_2O)_{16}^{8+}$.

phosphoric acid. The extractant-Pu-polymer geometries were shorter than those in the aqueous media; their radii were increased by the lengths of the extended alkyl substituents of the extractants.

Synergistic Effects in Solvent Extraction of Lanthanides and Actinides

The goal of this research is the development and characterization of synergistic solvent extraction systems for the separation of trivalent lanthanides and actinides. The work is being carried out in collaboration with D. D. Ensor of Tennessee Technological University. A paper describing initial studies of the mechanism of extraction of U(VI) using thenoyltrifluoroacetone (TTA) in combination with 4,13-didecyl-1,7,10,16-tetraoxa-4,13-diazacyclooctadecane (22DD) was published along with a second more general paper describing the behavior of trivalent actinides and lanthanides. The most interesting result is that the nitrogen atoms in the macrocycle seem to be largely responsible for the synergistic effect; analogous crown complexes are ineffective. The results showed that the synergistic effect is largest for the ions with the greatest ionic radius and decreases as the radius decreases. An

excellent group separation could be achieved, but there were no significant separations of the individual elements.

These results were further confirmed by studies using nonmacrocylic nitrogen donors as synergists with TTA. Simple tertiary amines, such as tri-*n*-octyl amine, showed the same synergistic effect as the macrocylic donors. We have observed that TTA performs poorly as an extractant in hydrocarbon diluents unless a critical concentration of a synergist or other phase modifier is present. Specifically, the metal species tend to collect at the interface of the system rather than in the aqueous or organic phases. Under some conditions, a fine white precipitate can be seen at the interface, even in the absence of metal ions. The precipitate was isolated and identified as the ketohydrate of TTA by its infrared spectrum. It is likely that the addition of a synergist or other phase modifier dehydrates this TTA species, increasing its solubility in the organic phase and improving the performance of the system.

Studies of the synergistic solvent extraction of Am(III) and Eu(III) from a chloroacetate buffer with TTA and triisobutylphosphine sulfide (TIBPS) in cyclohexane were completed. The metals are extracted as two metal complexes, $[M(TTA)_3]$ and $[M(TTA)_2(TIBPS)(CLO_4)]$. For europium, the logarithms of the extraction constants are -7.7 ± 0.1 and -2.34 ± 0.01 , respectively. For americium, the values are -8.4 ± 0.2 and -2.57 ± 0.01 . The extraction of both Eu and Am is synergized by the addition of TIBPS, but the effect is smaller than that of an oxygen donor.

A study of the synergistic properties of bidentate ligands such as octyl(phenyl)-*N,N*-diisobutylcarbamoylmethylphosphine oxide (CMPO) was conducted. It was found that these ligands show a synergistic effect on the extraction of actinides and lanthanides at low synergist concentrations, but show an anti-synergistic effect at slightly higher ones.

Thermodynamic Studies of Lanthanides and Actinides

Highly purified $AlCl_3$ and 1-ethyl-3-methylimidazolium chloride (EMIC), which together form a molten salt at room temperature in acidic (>50 mol % $AlCl_3$), basic (<50 mol % $AlCl_3$), and neutral mixtures, were synthesized. The uranyl ion is stable in basic melts, but it is slowly (over a period of days) reduced to U(V) in acidic solutions.

This reaction was followed spectrophotometrically in an effort to determine the reaction rate constants. The data are consistent with a two-step mechanism. The relatively fast first step is a first-order process with a rate constant of $\sim 0.09 \text{ min}^{-1}$. The slower second step is also a first-order process with a rate constant of $\sim 0.0004 \text{ min}^{-1}$. These results were obtained by watching the growth of the U(V) band at 395 nm. Further examination of the uv-vis spectra obtained for the reaction mixture has revealed a band at 1515 nm that appears during early stages of the experiment and then disappears as U(V) is formed. This band is assumed to be due to the intermediate that forms from the fast reaction. These studies have been conducted in collaboration with G. R. Choppin of Florida State University.

5.3 Engineering Research

Fundamental studies are pursued in two distinct areas of engineering research: the exploration of novel approaches to the improvement of chemical separations and the development of fundamental knowledge which will lead to better understanding of separation phenomena and ultimately yield enhanced predictive methods. Enhanced mass transfer performance is sought in a variety of multiphase separation processes, such as solvent extraction, vapor-liquid systems (distillation), solid-vapor (vacuum sorption), and solid-liquid (chromatography and high-gradient magnetic field separations). In the fluid-fluid work, an important thrust is the enhancement of transport through the interaction of external fields with the interface. This has led to in-depth studies of dispersion and coalescence in liquid-liquid systems under the influence of electric fields as well as some fundamental mass transfer studies under oscillating and disruptive conditions. The overall program has led to developing and patenting of a new solvent extraction concept, the emulsion-phase contactor (EPC). The investigation of high-intensity, high-gradient magnetic fields has shown that there is an excellent potential for the separation of macromolecules and particulate entities. A wide-ranging study of continuous annular chromatography (CAC) as applied to bioseparations and to metals separations has been completed with technology transfer in full progress.

Finally, a new approach to the understanding of solute-solvent interactions in the supercritical

region has been developed as part of a study of supercritical extraction. The new theory, based on Kirkwood-Buff formalism, has successfully predicted behavior of extraction in the supercritical region. This has led to new initiatives in neutron scattering and to the development of a surface acoustic wave (SAW) device.

5.3.1 Effects of External Fields on Multiphase Systems

This program is comprised of several fundamental studies that address the use of electromagnetic fields to enhance the efficiency of multiphase separation processes. The primary aim is to improve mass transfer performance in solvent extraction by utilizing theoretical insight gained from in-depth analysis of electric field interactions with dispersed liquid-liquid systems. The goal of this portion of the program is to examine effects on transport processes through a basic understanding of the inherent electrohydrodynamical processes. A secondary thrust involves investigation of the use of high-intensity, high-gradient magnetic field interactions for the separation of macromolecules in liquids. This project involves understanding the role of magnetic and electrostatic forces in controlling macromolecular motion in liquids.

Supported Drops: Shapes and Stability of Pendant/Sessile Drops and Drop Formation in Electric Fields

Research in our laboratories has shown that electric fields can be used to reduce—by as much as a factor of 10 or more—and control the volume of drops forming at an orifice. A proper understanding of this phenomenon and its exploitation for beneficial use in chemical engineering operations requires that one be able to calculate drop shapes and volumes and electric field distributions around supported drops from first principles.

When the volumetric rate of flow of liquid issuing from an orifice is low, the electric field

required to form drops of a given size can be determined by means of a static analysis. This requires the simultaneous solution of the augmented Young-Laplace equation of capillarity for drop shape and the Laplace equation for electric field distribution. Here this coupled set of nonlinear equations has been solved by finite element analysis. Figure 5.15 shows results of sample calculations: sequences of drop shapes and electric potential and electric field distributions outside conducting drops are plotted as functions of increasing field strength. In Fig. 5.16, limits of stability of drops as a function of drop size are presented. The dimensionless drop-size parameter, D , ranges between -1 and 1 ; $D=0$ corresponds to the case in which the drop shape is hemispherical in the absence of an electric field. For a given drop size, the critical value of the electric field is the field strength at which a drop of given volume would become unstable and detach from the orifice.

Free Drops: Drop Oscillations in Liquid-Liquid Systems

A complete understanding has been developed of the problem of small amplitude oscillations of a liquid drop that is immersed in another liquid.¹⁰ When the relative importance of gravitational-to-surface forces is small, such oscillations are governed by the nonlinear dispersion relation derived by Miller and Scriven.¹¹ The dispersion relation has been solved numerically to determine the character of oscillations for arbitrary values of drop size, physical properties of the two fluids, and interfacial tension. The new theoretical results have determined the range of validity of various approximations that have been proposed hitherto and have also been shown to be essential for proper interpretation of many previously reported experimental results. New experimental measurements of natural frequencies of oscillation of water drops falling in 2-ethyl-1-hexanol, a system having properties characteristic of many others in solvent extraction, agree well with theoretical predictions when the drop radius is smaller than a critical size. Moreover, the research has shown that frequencies of oscillation of larger drops are better described by the dispersion relation due to Subramanyam,¹² which accounts for the relative motion of the two phases, as shown in Fig. 5.17.

FIXED CONTACT LINE, $D = 0$ ($V = 2\pi/3$), $H = 5$

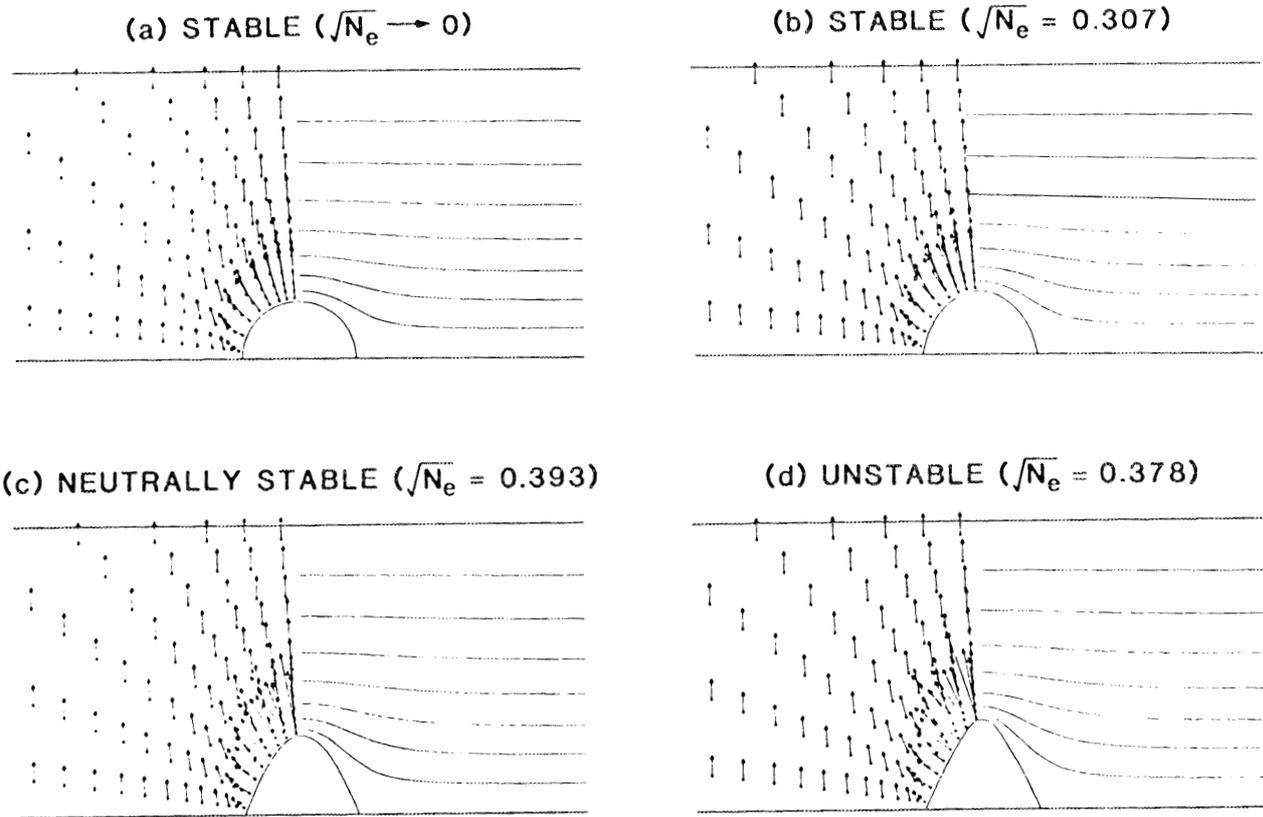


Fig. 5.15. Evolution of supported (pendant or sessile) drop shapes with electric field strength.

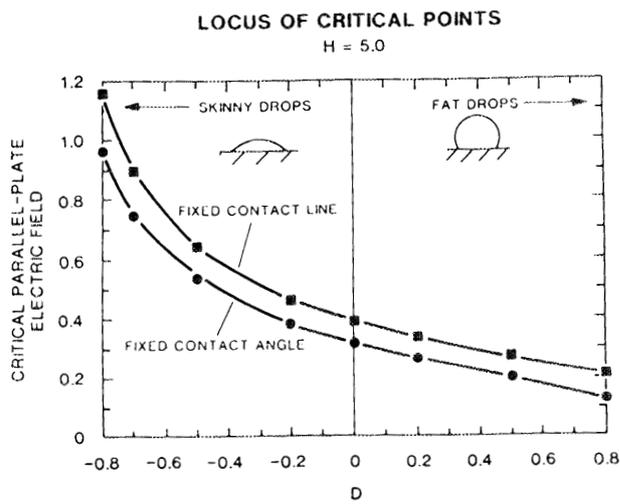


Fig. 5.16. Neutral stability curve in parameter space for supported drops: critical field strength for instability vs drop size for drops making either a fixed contact line or a contact angle with the supporting plate.

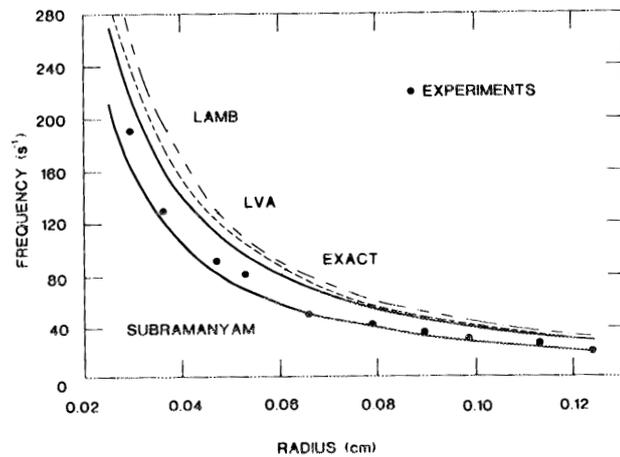


Fig. 5.17. Frequencies of oscillation as a function of radius for water drops falling in 2-ethyl-1-hexanol; a comparison of experimental and theoretical results.

High-Gradient Magnetic Field Separations (HGMS)

The imposition of a high-intensity, high-gradient magnetic field on a column packed with ferromagnetic spheres has allowed capture of the protein ferritin from an aqueous-based buffer solution. Ferritin is the major iron storage protein found in mammals. It is comprised of ~4500 atoms of ferric iron surrounded by a proteinaceous shell. The molecular weight of ferritin is 900,000, with the weight being roughly equally distributed between the iron core and the shell. The shape in solution is essentially spherical with a core diameter of 75 Å and an outer diameter of 125 Å. The isoelectric pH of ferritin is 4.4; therefore, at pH values above this point it has a net negative charge while below this point it has a positive charge. Due to the iron in the core, ferritin displays a strong interaction with magnetic fields (superparamagnetism). This makes it a logical candidate for investigating the effects of magnetic fields on macromolecular separations.

The design of the column in which capture was demonstrated is shown in Fig. 5.18. A 6-T magnetic field was imposed on a bed of ferromagnetic steel spheres (100- μm diam). Solutions containing ferritin at various pHs were passed through the column, and the effluent was monitored for ferritin breakthrough. Figure 5.19 is a set of three ferritin breakthrough curves which were taken at pH values

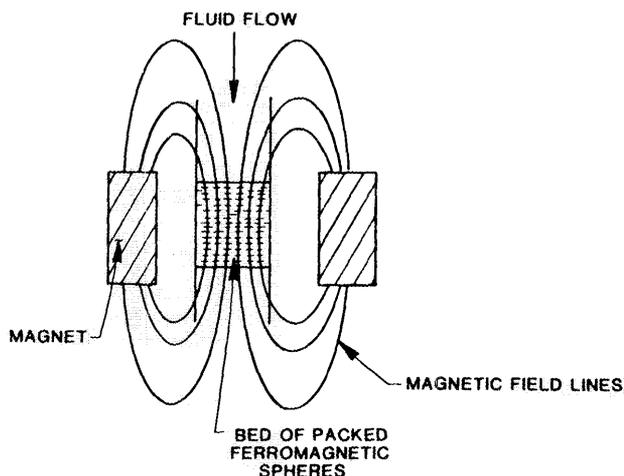


Fig. 5.18. Diagram of stainless steel column, packed with 100- μm ferromagnetic spheres, in which capture experiments were conducted.

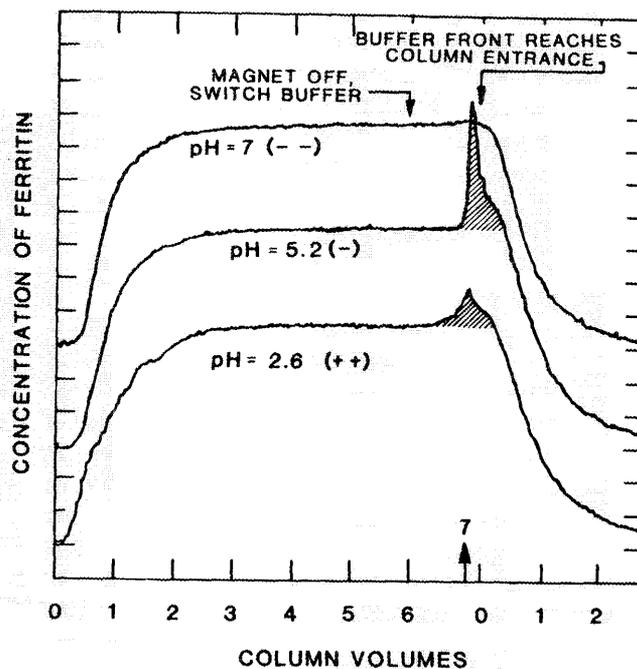


Fig. 5.19. Comparison of ferritin breakthrough curves for magnetic cases at pH values of 2.6, 5.2, and 7.

of 2.6, 5.2, and 7. These represent cases in which the protein is slightly positively charged, slightly negatively charged, and (relatively) highly negatively charged, respectively. As indicated in the figure, a small amount of protein capture was possible only near the isoelectric pH where the charge of the protein was diminished. It is also interesting to note that at the isoelectric pH (4.4), very rapid capture was observed and the column began to plug. This infers that the presence of electrical charge on macromolecular entities can control their behavior in magnetic field gradients. Hence, manipulation of the charge may prove to be a technique that is useful in the separation of such molecules from solution.

The Emulsion-Phase Contactor

High-intensity transient electric fields can be used to disperse aqueous-based liquids in relatively nonconducting immiscible organic liquids. The characteristic droplet size of this process is on the order of microns; hence, a vast amount of surface area is produced for interfacial mass transport and reaction. In addition, the electric field can be used

to control the motion of and induce coalescence/phase separation of the liquid-liquid dispersion. The EPC is a continuous, countercurrent flow, multistage extraction device that uses electric field interactions to control droplet formation, transition, and coalescence. Due to the vast amount of surface area, the mass transfer performance of the device has proved to be an order of magnitude better than other laboratory-scale devices reported in the literature.

While exhibiting outstanding mass transfer performance, the throughput of the preliminary system was extremely low. Improvements have been made in the electrode design and the type of electric field used in order to increase the throughput of liquids in this device. Figure 5.20 is a schematic diagram showing the new electrode design. Essentially, the EPC has two electrode sections—one which is best for dispersion of the aqueous phase out of nozzles and a second which controls the holdup and coalescence of the dispersion in the device. The dispersion section is comprised of charged electrodes that surround grounded nozzles through which the aqueous feed is introduced. This field effectively disperses liquid

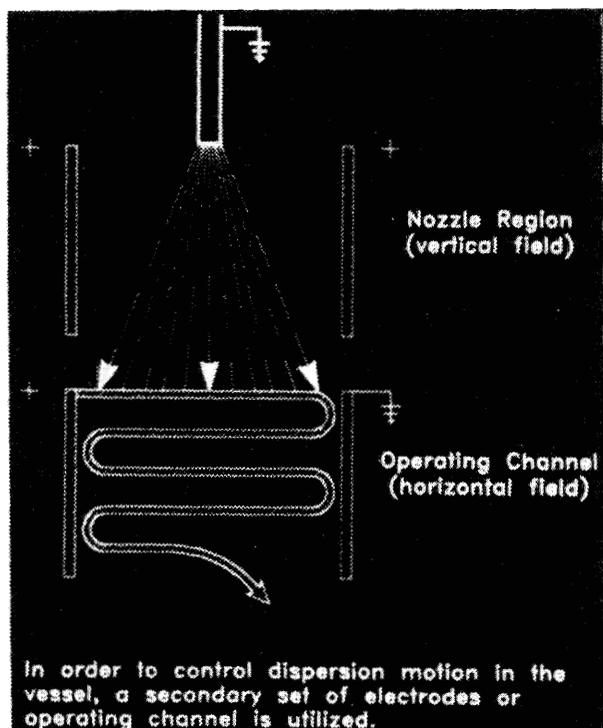
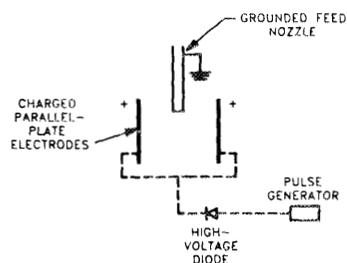


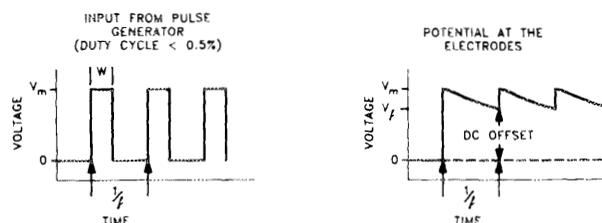
Fig. 5.20. Schematic drawing of the emulsion-phase contactor, showing the new electrode design.

from the nozzle and accelerates it downward. The second electrode set, or operating channel, is comprised of alternating charged and grounded flat plate electrodes. This section of the device gives the dispersion a significant side-to-side motion, thus increasing holdup in the system. In the operating channel, numerous redispersion and coalescence events take place.

Figure 5.21 illustrates the type of electric field used in the system. By utilizing a high-voltage diode in the circuit, electrical pulses sent to the electrodes can be substantially trapped on the electrodes. Loss of charge occurs from the grounded surfaces in the device; however, if one increases the frequency of the charging pulses, a dc-field offset occurs between the charged and grounded surfaces. The result is a composite electric field that has voltage spikes imposed on top of a dc-field offset. Utilizing this electrode design along with the composite field, the throughput of the system was increased by a factor of 100. This puts the throughput near the values of typical industrial devices.



A HIGH-VOLTAGE DIODE IS USED TO PREVENT BACKFLOW OF CHARGE FROM ELECTRODES TO THE POWER SUPPLY.



AS THE PULSE FREQUENCY IS INCREASED, A DC OFFSET DEVELOPS BETWEEN THE NOZZLE AND ELECTRODES. TYPICAL VALUES: $V_m = 6$ kV; $f = 2000$ Hz; $W = 2$ μ s; $V_f = 4$ kV.

Fig. 5.21. Schematic drawing of the emulsion-phase contactor, illustrating the type of electrical field used in its design.

5.3.2 Interactions of Solvents, Solutes, and Surfaces

Fundamental theoretical and experimental research focuses on relating macroscopic properties of solutions to intermolecular interactions and molecular distribution functions for the highly asymmetric systems characteristic of extraction by supercritical solvents and adsorption from supercritical solutions. These studies are aimed at an improved understanding of the molecular interactions and their influence on the basic phenomena that are important in practical separation processes.

Theory

Our theoretical approach has been within the framework of molecular distribution function theory of equilibrium statistical mechanics. Bulk properties are related to integrals of the molecular pair distribution function through the formalism of Kirkwood-Buff solution theory. Integral equations permit a priori calculation of the molecular distribution functions and resultant properties, including chemical potential, for which a new calculation method has been developed. In addition to bulk properties, these theoretical calculations provide detailed information on fluid microstructure, which can lead to important new insights.

Kirkwood-Buff Theory

Kirkwood and Buff¹³ developed a statistical mechanical theory of solutions which relates bulk properties to molecular distribution functions through the fluctuation integral, $G_{\alpha\beta}$, which is defined in terms of the pair distribution function, $g_{\alpha\beta}$, for species α and β :

$$G_{\alpha\beta} = G_{\beta\alpha} \equiv \int_0^\infty dr 4\pi r^2 [g_{\alpha\beta}(r) - 1] . \quad (5.1)$$

In addition to the familiar expressions for the partial molecular volume and the isothermal compressibility, we have derived a new expression for the solubility of a solute k in a solution of c components in solvent j :

$$\frac{kT d\rho_k}{\rho_k} = \left\{ (G_{kj} + \bar{v}_k) + \sum_{i \neq j}^c [\rho_i \bar{v}_i (G_{ki} - G_{kj})] \right\} dP, \quad (5.2)$$

where ρ_α is the number density of species α , P is the pressure, T is the absolute temperature, k is the Boltzmann constant, and \bar{v}_α is the partial molar volume of species α . In the common case of a solid, incompressible solute, \bar{v}_k is simply the reciprocal of the solid density.

The first term on the right-hand side provides the basis for development of accurate models of solubility in dilute solutions (below), and the second term makes explicit how solubility is affected by increasing concentration, by the presence of a second solute, or by the presence of an added cosolvent. This is the only theoretical model of supercritical solutions with such scope; in addition, it is the only theory of supercritical solutions with such rigorous foundations.

Useful practical models for dilute solutions result when the solute-solvent fluctuation integral, G_{jk}^0 , can be expressed in terms of the solvent-solvent fluctuation integral, G_{jj}^0 . The latter can then be calculated from an accurate equation of state for the pure solvent; for the common supercritical solvents (e.g., CO_2 , C_2H_4), very accurate equations of state are available. We have used this model for a priori predictions¹⁴ as well as for accurate fitting of solubility data.¹⁵⁻¹⁷ In addition, we have developed models for the variation of the fugacity coefficient with concentration^{18,19} and for the number of excess solvent molecules, beyond the bulk average, surrounding a dissolved solute.²⁰⁻²⁴

Solution Microstructure from Integral Equations

Molecular pair distribution functions can be calculated from the Ornstein-Zernike equation using approximate integral equation theories. We have made such calculations for representative supercritical solutions using the Percus-Yevick (PY) theory, hybrid mean spherical approximation (HMSA), and the reference hypernetted chain (RHNC) theory. In addition to providing the inputs for the theoretical expressions described above, these calculations yield detailed information about

the fluid microstructure. Figure 5.22 shows the number of excess CO_2 molecules, beyond the bulk average, that surround a dissolved naphthalene molecule as a function of density for an isotherm just above the critical temperature. These a priori theoretical calculations are consistent with a growing body of experimental data that indicate that solvent-solute clustering is the fundamental cause of the striking behavior of supercritical solutions. The integral equation calculations also indicate that there is a strong tendency for solute molecules to aggregate (see Fig. 5.23). Such behavior would be expected to have observable effects; indeed, independent spectroscopic studies²⁵ have shown a marked solute-solute aggregation in supercritical solutions. Other effects of potential practical importance (i.e., acceleration of rates of bimolecular reactions) can be predicted.

Theoretical predictions^{23,24,26-28} indicate that another type of supercritical solution must exist that shows some characteristics opposite those of the typical dilute supercritical solution. In typical supercritical solutions, the size and energy of solute-solute and solute-solvent interactions are large compared with the solvent-solvent interaction parameters: $\sigma_{kk}, \sigma_{jk} > \sigma_{jj}$ and $\epsilon_{kk}, \epsilon_{jk} > \epsilon_{jj}$. As a consequence of the clustering of solvent and solute molecules, as described above, these typical solutions are called dilute, *attractive* systems, and their behavior is characterized by large, *negative* solute partial molar volume and dramatic *decrease* in solute fugacity near the solvent critical point.

In contrast, there must exist dilute solutions where the size and energy of solute-solute and solute-solvent interactions are small compared with

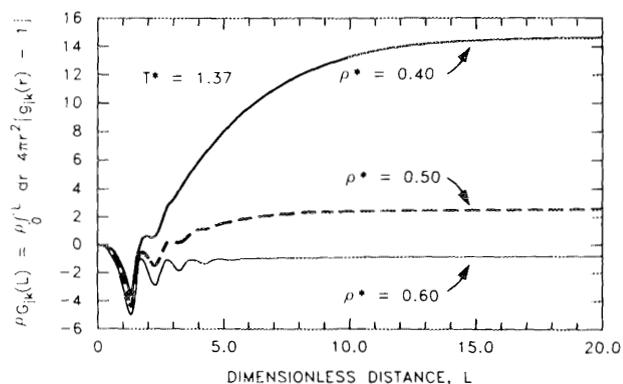


Fig. 5.22. Excess solvent molecules surrounding attractive solute molecule.

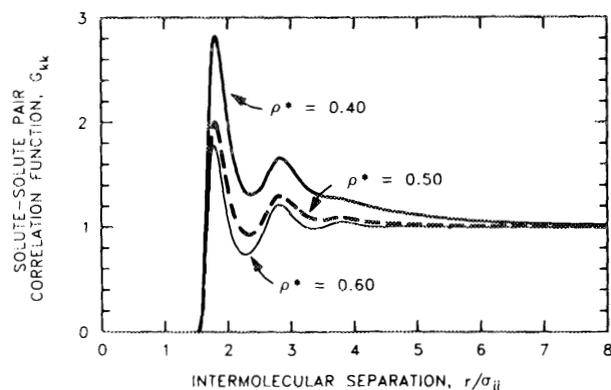


Fig. 5.23. Solute-solute pair correlation functions.

the solvent-solvent interaction parameters: $\sigma_{kk}, \sigma_{jk} < \sigma_{jj}$ and $\epsilon_{kk}, \epsilon_{jk} < \epsilon_{jj}$. One might guess that such systems would exhibit a deficit of solvent molecules surrounding a dissolved solute molecule rather than a cluster. Indeed, Fig. 5.24, calculated from integral equation theory, shows just such behavior. Such solutions have been called dilute, *repulsive* supercritical solutions. Such systems are predicted to be characterized by large and *positive* solute partial molar volume and dramatic *increase* in the solute fugacity near the solvent critical point. Unexpectedly, perhaps, the dilute, repulsive supercritical solutions exhibit solute-solute aggregation just like the attractive systems. Independent calculations by decorated-lattice gas theory,²⁶ thermodynamics,²⁷ molecular simulation,²⁸ and integral equations^{23,24} show qualitative and quantitative agreement in their predictions about

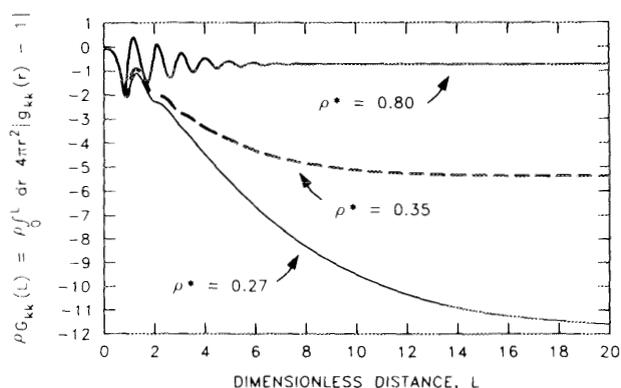


Fig. 5.24. Deficit of solvent molecules surrounding repulsive solute molecule.

dilute, repulsive supercritical solutions. Furthermore, recent measurements of the molar volume of aqueous solutions of Ar, C₂H₄, and Xe near the critical point of water²⁹ are strongly indicative that the solute partial molar volume would exhibit the predicted large, positive divergence, if measured. The predicted rapid increase in the solute fugacity for dilute, repulsive supercritical solutions may have important implications for practical separation schemes. Such possibilities are under investigation.

Chemical Potential

For modeling separations, chemical engineers are most interested in the fugacity (or, equivalently, the chemical potential) of the species in solution. Unfortunately, entropy (and, therefore, free energy and chemical potential) is not a mechanical variable (i.e., not an ensemble average of a pairwise quantity). Consequently, calculation of chemical potential from statistical mechanics requires orders of magnitude more computational effort than calculation of mechanical variables. Until recently, the magnitude of the computation was prohibitively large except for a few limited cases.

Recently, we have explored two theoretical approaches for more efficient statistical mechanical calculation of chemical potential from the distribution functions that can be calculated from integral equation theory. One approach³⁰ employs the zero-separation theorems of distribution function theory to relate the chemical potential, μ , to the coincidence value of the cavity correlation function, $y(0)$, and the potential distribution of a particle created by the overlap of two molecules, $\ln\langle\exp[-2\int(0)/kT]\rangle_{N-1}$:

$$2\mu/kT = \ln y(0) - \ln\langle\exp[-2\int(0)/kT]\rangle_{N-1} \quad (5.3)$$

By using the hypothesis of universality of the bridge function, then, one might make practical calculations using the known bridge function of a reference fluid (hard-sphere fluid). Unfortunately, the sensitivity of this calculation to deviations from the hypothesized universal bridge function has proved to be so large that accurate results will require a perturbative correction. Work on this approach is continuing.

A second approach^{31,32} employs the concepts of Widom's test-particle insertion method, scaled-particle theory, and Kirkwood charging integration to calculate chemical potential using

distribution functions for the solution and for the solution about the test particle calculated from integral equation theory. The method consists of first calculating the molecular distribution functions for the solution using an accurate integral equation theory. Then, using the Kirkwood charging integration, the change in the free energy is calculated for the hypothetical change of one particle in the solution into a particle which interacts with its neighbors only with the Weeks-Chandler-Anderson (WCA) repulsive reference potential. Then, the diameter of an equivalent hard sphere is calculated so that the free energy change is zero for transformation of the WCA particle into a hard sphere. The free energy change is then calculated as the diameter of the hard-sphere particle decreases from the equivalent diameter to zero diameter. Finally, the free energy change is calculated for the removal of the hard point particle from the solution. This charging path has been carefully specified so that the charging integration for each step requires a minimal number of calculations of the distribution functions of the solution about the test particle, and all calculations are performed at the same thermodynamic state as the first calculations of the distribution functions of the solution itself.

Extensive testing of this method against the results of computer simulations and against exact analytical expressions for simple cases has shown that the method is as accurate as possible based on the integral equation theory that is used, and some modern integral equation theories (RHNC and HMSA) have been found to be within the statistical uncertainty of simulation results for most thermodynamic states. With the HMSA integral equation theory, the new method is highly accurate for all systems tested at temperatures above the critical point, and for subcritical temperatures, the method with the HMSA theory is highly accurate at high density and at low density. At moderate density and subcritical temperature, the method with the HMSA theory is highly accurate for all pure fluids tested and for all mixtures tested except those where the size difference of the species is large. Figures 5.25 and 5.26 compare the results of our calculation with exact simulation results for two mixtures of WCA fluids with different particle size and for mixtures of Lennard-Jones fluids with different interaction energy, respectively. Figure 5.27 shows that at low temperature and moderate density, the calculated chemical potential for Lennard-Jones mixtures exhibits incorrect variation with particle

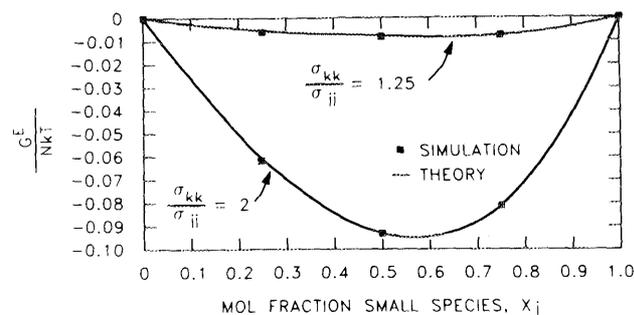


Fig. 5.25. Predicted excess Gibbs free energies of mixing in softly repulsive (WCA) fluids.

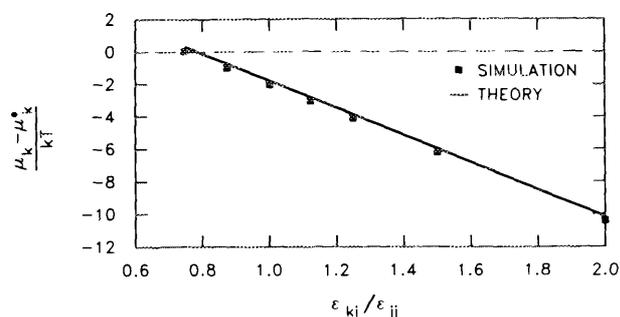


Fig. 5.26. Predicted residual chemical potential of a dilute solute-vs-energy ratio.

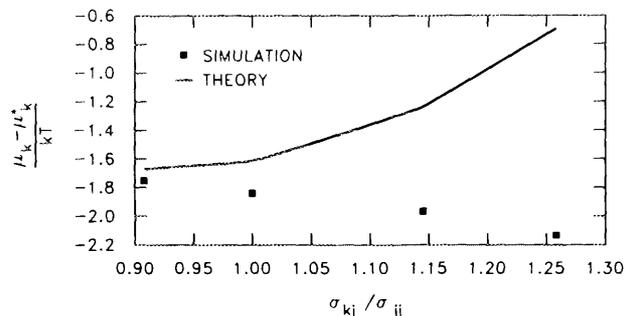


Fig. 5.27. Predicted residual chemical potential of a dilute solid-vs-size ratio.

size; this variance resulted from inaccurate calculations of the distribution functions of the solution molecules around the test particle by the HMSA theory.

Experiment

Our experimental studies of the effects of interactions among solvents, solutes, and surfaces have explored just those regions that are most challenging to theory. In measuring solubility in supercritical solvents, we have sought systems where the solvent-solute size ratio is different from that of previous systems studied. Our adsorption measurements anticipate future theoretical investigations of inhomogeneous fluids at moderate density. Our planned neutron-scattering experiments are aimed to test the predictions of integral equation theory for intermolecular separations between the first-neighbor distance (which has been tested with molecular simulation results) and very long range (which have been tested with exact asymptotic calculations).

Solubility of CCl_4 in Supercritical CF_4

We have completed measurements³³ of the solubility of solid carbon tetrachloride, CCl_4 , in supercritical carbon tetrafluoride, CF_4 . The CF_4 was compressed, brought to the desired equilibrium temperature, and then passed through a bed of crushed (~ 1 mm), solid CCl_4 in an equilibrium cell. The solution was sampled into an on-line mass spectrometer for analysis. After appropriate calibrations and verification of equilibration and reproducibility, solubility data were collected for two isotherms—at 244.0 and 249.0 K—as shown in Fig. 5.28. Data collected at two other isotherms—234.0 and 239.0 K—were not reproducible, giving strong indications that presence of a third, liquid phase interfered with obtaining representative samples of the supercritical fluid phase.

Both CF_4 and CCl_4 are almost spherical molecules, so that comparisons of experimental results with predictions from theory are not complicated by calculational details resulting from anisotropy. It should be noted that the fluctuation integral, used in our theoretical models, involves the spatial pair distribution function; thus, molecular anisotropy is handled by straightforward averaging over all orientation angles. Nonspherical, even polar, molecules can be modeled (e.g.,

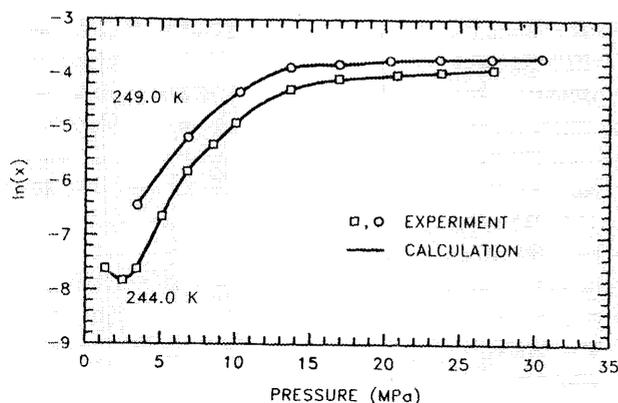


Fig. 5.28. Solubility of solid CCl_4 in supercritical CF_4 .

published calculations for benzoic acid and 1,4 naphthoquinone¹⁵). Nevertheless, the comparison with experimental results can be more nearly unequivocal without such complications.

The vapor pressure of solid CCl_4 is appropriate at the temperatures of the experiments, yielding equilibrium mole fractions ranging from 4.0×10^{-4} to 2.6×10^{-2} .

Compared with previously studied supercritical solutions, however, this system is unusual in that the ratio of solute to solvent molecular size is relatively small — $\sigma_{kk}/\sigma_{jj} = 1.25$ — while the ratio of solute to solvent molecular energy parameter is relatively large — $\epsilon_{kk}/\epsilon_{jj} = 2.15$.

The measured solubility data are in very good agreement with theoretically calculated solubility (see Fig. 5.28). Most of the dramatic changes in the fluid microstructure, hence in bulk properties, result from nearness to the solvent critical point; so, use of an accurate equation of state for the pure solvent³⁴ can yield accurate calculations, even with simple models, as long as the model has a sound theoretical basis.

Adsorption Measurements at High Pressure

We are seeking to demonstrate an experimental technique with which we can measure small changes in the mass on a surface with time, even at elevated pressure. First we must demonstrate the performance of the technique under conditions where other techniques are available for comparison; then we must quantify the effects of

pressure and temperature before we can make quantitative measurements at high pressure. Our aim is to generate data on adsorption equilibrium and kinetics over a range of pressures from vacuum conditions, to which currently available techniques are limited, to high pressures characteristic of supercritical solutions.

The approach we are following³⁵ is to measure, for a surface harmonic on a SAW transducer, the change in frequency that results from changes in the mass loading on the surface. A SAW transducer has been mounted on a vacuum flange and installed in a vacuum system for calibration against a Cahn microbalance. A similar SAW transducer, exposed to the same conditions (except for exposure to the adsorbate), is used as reference. We project that changes on the order of 0.001 of a monolayer per second can be measured.

Neutron-Scattering Measurements for Supercritical Solutions

Scattering experiments permit direct measurement of the structure factor of a material system, and careful neutron-scattering measurements employing isotope substitution can yield partial structure factors for mixtures. Pair correlation functions can be determined by Fourier transformation of partial structure factors measured over a wide \mathbf{k} range (e.g., this corresponds to a wide angular range for scattering of monochromatic neutrons), and fluctuation integrals can be determined by extrapolation of small-angle scattering results to $\mathbf{k}=0$. Thus, neutron-scattering experiments can provide data to test our distribution function-based theories in ways that are not otherwise possible. For data to be most useful in testing theory, it is important that discrepancies, if any, be clearly attributable to the theory and not ambiguously attributable to the theory plus the intermolecular potential energy function used. This suggests that the experiment should be performed with a system for which extremely accurate potential energy functions are available, such as the noble gases. Fortunately, one of the noble gas isotopes, ³⁶Ar, has an exceptionally large elastic scattering length for thermal neutrons, and scattering lengths have been measured for several isotopes of both argon and neon. Consequently, a solution of solid argon in supercritical neon is uniquely suited for our purposes. A serendipitous attribute of this system is that the structure of the dilute, repulsive

neon in supercritical argon solution can be measured subsequently in the same apparatus.

We have designed a special equilibrium cell and ancillary equipment for these measurements. The cell is stirred by a small magnetically driven turbine and is suitable for both thermodynamic and subsequent scattering experiments. (The thermodynamic experiments are necessary because the Ne-Ar system has not previously been studied at temperatures below the argon triple point.) Phase equilibrium can be determined by observing the frost point of mixtures of known composition; the cell has a window permitting observation by a video system of a gold mirror on the cold end of the cell. The maximum temperature differential is limited to 20 mK. The cell is cooled by a Displex helium refrigeration unit, and temperature is measured and controlled to within ± 20 mK by two Lakeshore platinum resistance thermometers with a BTI conductance bridge and a Linear Research controller. Pressure is measured to within 0.01% accuracy by two Digiquartz transducers and is adjusted by a Ruska hand pump. The components for this apparatus are on hand and are presently being assembled. Detailed design of the cell has been completed, and the cell is awaiting fabrication.

5.3.3 Multicomponent Separations by Continuous Annular Chromatography

Continuous annular chromatography (CAC), a concept for truly continuous, steady-state chromatographic separations, utilizes an annular sorbent bed, which is packed in the space between two concentric cylinders. In isocratic mode, the eluant is uniformly distributed at the top of the bed along the bed circumference while the bed assembly slowly rotates about its axis. The multicomponent feed is continuously introduced at the top of the bed at a fixed position.

The chromatographic elution of the feed components coupled with the bed rotation causes the formation of individual helical component bands which extend to the bottom of the bed, where the separated components are continuously recovered as they exit the bed at different angular positions from the feed point. The background developed at ORNL for the current research is reviewed in a several publications.³⁶⁻³⁹

Significant advances have been made recently, both in terms of improved energy efficiency as well as in terms of new applications of the separation concept. Two improved operating modes, which are common in conventional chromatography, have been implemented for the CAC: step-wise elution, which leads to gradient elution if multiple concentration steps are used, and displacement development. Step-wise elution was shown to allow continuous operation at high throughput with minimal dilution of the separated products; continuous displacement development, on the other hand, was shown to result in a concentration of the separated products at the expense of some product recycle.

Several novel applications of CAC were investigated in depth, including (1) the separation of sugars,^{40,41} (2) the separation and purification of proteins,^{42,43} (3) the separation of iron-chromium mixtures by step-wise elution,⁴⁴ and (4) the separation of mixtures of amino acids and rare earth elements by displacement development.⁴⁵ Both experimental and theoretical studies were carried out for these systems with CAC units ranging in scale from 10.2 to 45.7 cm in diameter and from 30 to 110 cm in depth. Dowex 50W-X8 ion-exchange resin was most often used for these studies, although various molecular sieve zeolites, Sephadex G-15, CM cellulose, and Dowex 50W-X4 resin were used for some of the experiments. Resin sizes varied from ~ 30 to $400 \mu\text{m}$, with the bulk of the experiments being performed with $50\text{-}\mu\text{m}$ material.

Scaleup of Isocratic Chromatography

In considering the fundamental conceptual differences between the CAC and the conventional column chromatograph, annular dispersion in the CAC is the only divergent point. Recently, we performed investigations of hydrodynamic dispersion in the CAC.^{39,40} The experimental study was performed in a 1.27-cm-OD column 60 cm long and a 30.5-cm OD CAC unit, with an annulus width of 1.27 cm and a depth of 60 cm, using Dowex 50W-X8 resin with particle sizes of $45 \mu\text{m}$ and $380 \mu\text{m}$ as the chromatographic media. In these experiments, an aqueous solution of dextran (mol. wt = 300,000) was used as the nonadsorbing feed and deionized distilled water served as the eluant. The results of this study are summarized in Fig. 5.29 in the form of $\epsilon Pe = u d_p / D_c$ vs $Re = u d_p / \nu$, where d_p is the particle diameter and ν is the

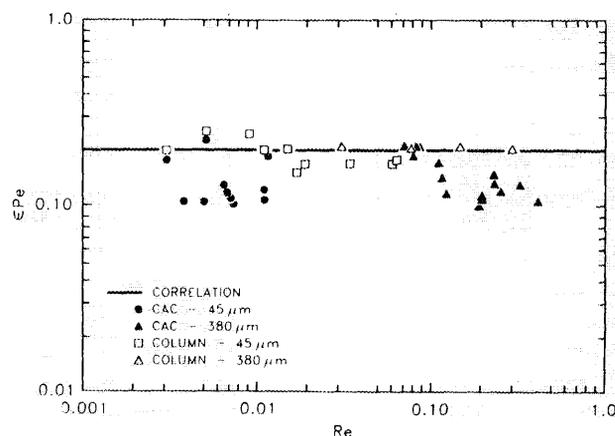


Fig. 5.29. Comparison of Butt⁴⁶ Pelet number-Reynolds number correlation with the experimental dispersion results for a column chromatograph and a CAC.

fluid kinematic viscosity. Also shown are measurements of axial dispersion for a fixed bed packed with the same resin used in the CAC experiments and a line calculated from a correlation by Butt.⁴⁶ The column data compare well with the correlation, while the CAC data are somewhat more scattered, generally tending to fall below the correlation line. On the average, the effective Pelet number for the CAC unit is ~ 0.15 , ~ 0.75 times smaller than the value found for fixed beds. Thus, dispersion in the CAC appears to be 1.5 times larger than in fixed beds and therefore not significant for most industrial scaleup situations in liquid systems.

An example of the application of chromatographic techniques to production-scale operations in the food processing industry is the glucose-fructose separation. A pilot-scale CAC (44.5-cm OD, with a 3.17-cm-wide annulus and packed with Dowex 50W-X8, 45 μm in diameter to a depth of 107 cm) has been developed for scaleup and optimization studies.^{47,48} Based on work with a simple column chromatograph containing the same resin, distribution coefficients, K , and overall solid-phase mass transfer coefficients, $k_o a$, were generated for use in the scaling of the glucose-fructose separation. At high loadings, the feed occupies a significant circumferential sector, and for the case of negligible angular and axial dispersion and of linear equilibrium, the analytic solution obtained by Carta⁴⁹ for periodic applications of finite feed volumes to a chromatographic column may be used

to compute any CAC concentration profile for a component that enters the unit with concentration c_F .

$$\frac{c(z, \theta)}{c_F} = \frac{\theta_F}{\theta_F + \theta_E} + \frac{2}{\pi} \sum_{j=1}^{\infty} \left\{ \frac{1}{j} \exp \left[-\frac{j^2 k_o a z}{(j^2 + r^2) u} \right] \times \sin \left[\frac{j \pi \theta_F}{\theta_F + \theta_E} \right] \right. \\ \left. \times \cos \left[-\frac{j \pi \theta}{\theta_F + \theta_E} + \frac{2j \pi \theta}{\theta_F + \theta_E} - \frac{2j \pi z \omega \epsilon}{u(\theta_F + \theta_E)} - \frac{j r k_o a z}{(j^2 + r^2) u} \right] \right\} \quad (5.4)$$

where θ_F is the arc over which feed is applied to the column, θ_E is the elution arc, and r is given by

$$r = \frac{k_o a (\theta_F + \theta_E)}{2\pi(1 - \epsilon)K\omega} \quad (5.5)$$

Experiments were carried out with the pilot-scale CAC unit, a 55°-wide feed sector. The feed was distributed by means of three feed nozzles, spaced $\sim 18.3^\circ$ apart from each other. To ensure an even flow distribution, each feed nozzle was fed by an individual metering pump. A comparison of the experimental CAC concentration profiles with theoretical predictions based on Eq. (5.4) is shown in Fig. 5.30. The good agreement shows that the linear model can be used even for these overloaded conditions, and we conclude that Eq. (5.4) can be used as an accurate scaling relationship.

The separation of protein mixtures was studied on a bench-scale CAC unit.⁵⁰ S-Sepharose, a strong-

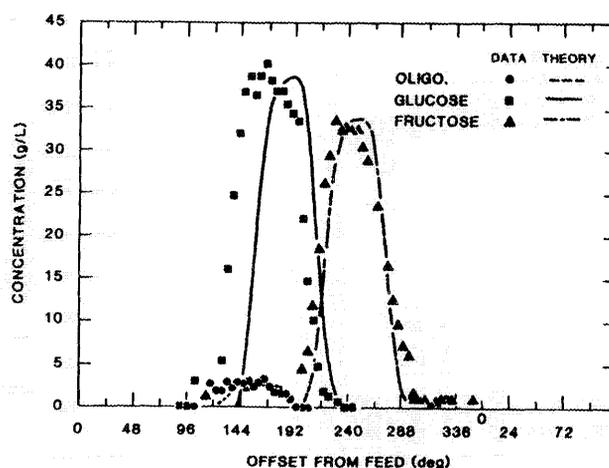


Fig. 5.30. Comparison of experimental 55°-feed-sector run with the theoretical predictions of Eq. (5.4). A diluted (10:1) industrial high-fructose corn syrup was used as the feed. The rotation rate was 144°/h.

acid, porous, cation-exchange resin, is used as the separation medium. Mixtures of albumin, hemoglobin, and cytochrome-C are used as a model separation system. Equilibrium and mass transfer parameters are developed for this system on the basis of fixed-bed chromatograph experiments. As an example of a broader study, a typical separation of albumin-hemoglobin mixtures by the CAC is shown in Fig. 5.31. The solid lines are model calculations that are based on the equilibrium and mass transfer parameters determined from the fixed-bed studies. Equation (5.4) can be used directly for these calculations and the scaling from the fixed-bed case. In the absence of axial dispersion, the equation gives an excellent description of the process. From Fig. 5.31, it is apparent that the steady separation performance of the CAC is quite close to predictions using parameters determined from fixed-bed experiments. This indicates that no significant channeling or other nonidealities take place in the CAC.

Economic and Energy Assessments of Sugar Separations

A program to assess the energy-related and economic advantages of using the concept of continuous chromatography as a replacement

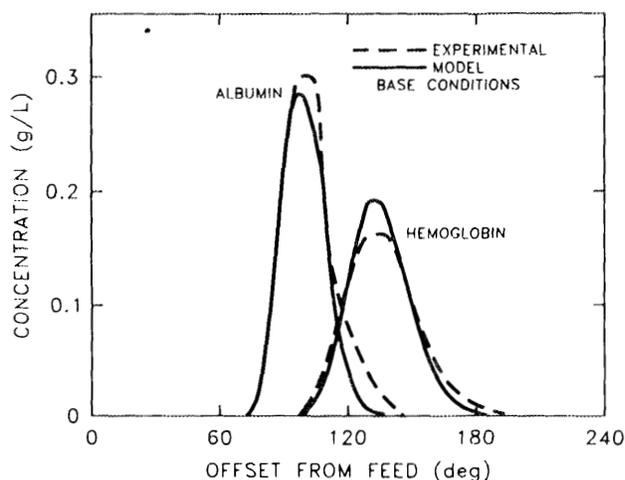


Fig. 5.31. Concentration profiles for the continuous chromatographic separation of an albumin-hemoglobin mixture.

technology for the separation portion of a number of technologies selected the production of high-fructose sugars as a representative of the burgeoning biotechnology industry.⁵¹ The study focused on the replacement of the separation unit, currently based on the SAREX process, with the CAC, a recently developed concept in multicomponent separations.

In establishing a comparison between the two plants, it was decided to consider a typical plant, processing 1270 m³ bushels of corn per day, which yields ~1300 metric tons per day (MTD) of 42% isomerization product, a significant fraction of which is recycled. Involved in this combined plant were recycle glucose streams, which all returned to the saccharification unit. The major energy load on such a plant is evaporation of 2420 MTD of water. Using the CAC as the separation unit, the ability to produce multiple products (separation of recycle into heavy materials and glucose) allows a significant reduction in recycle to the saccharification unit, thus reducing its size by ~10% as compared with the SAREX-based plant (Fig. 5.32). This not only reduced the energy load but the capital costs as well. The CAC was operated rather conservatively, with a 45° feed sector. In this case, 4.2 MTD of a crystalline 99+% fructose would be made in addition to the 368 MTD of 42%

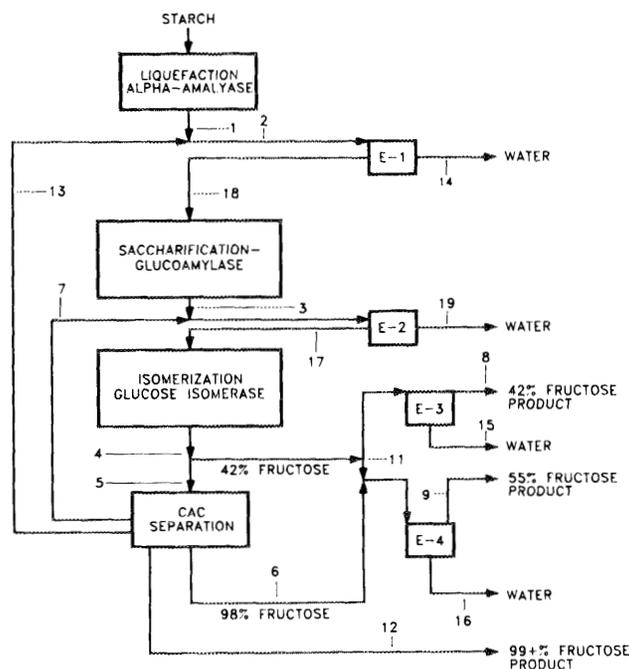


Fig. 5.32. Schematic flowsheet of a typical high-fructose corn syrup plant with a CAC separation unit.

fructose and 372 MTD of 55% fructose. The evaporation load here is 1982 MTD of water, with significant improvement realized if a less conservative 60° feed sector is used.

Assuming that energy consumption by evaporation is the only major difference between the two plants, the energy saving in converting to the CAC is $\sim 4 \times 10^{11}$ British thermal units (BTU)/year. Given a 47.3-lb/capita consumption rate in 1987, this would amount to a total national energy saving, for this product slate alone, of ~ 0.01 quad/year (1 quad = 10^{15} BTU). If the crystalline fructose market grows as projected, this number could easily become 0.1 quad/year. The differences in the capital costs of plants are inconsequential.

During the reporting period, a second economic study on the industrial application of the CAC was published.⁵² In this study, the dump leaching of copper ores, a good case was made for the implementation of the CAC to new installations.

Dilution Elimination

Two advanced techniques were explored with considerable success for the elimination of dilution in the CAC. Gradient elution and displacement development were explored in a variety of chemical separation systems. In the first case, a simplification to a step in the eluant concentration was used as the prototype for a broad selection of separations.

Gradient elution chromatography involves varying one of the properties of the eluant as a function of time (or angular position). In the example selected for study, a dilute mixture (1.0–5.0 g/L) of Fe^{3+} and Cr^{3+} (ferrous and chromic nitrate dissolved in water) is eluted with 0.4 M to 1.5 M ammonium sulfate at constant 0.025 M H_2SO_4 in a step-elution mode.⁵³ Through judicious choice of eluant steps, it is possible to decrease dilution by changing the eluant "strength." The theory for this operation is very similar to the isocratic eluant case, with the important exception that one must account for the local concentration of the eluant throughout the bed. For this reason, the equations become nonlinear and must be solved by computer methods. Orthogonal collocation was used as the method of choice.

In exploring the potential of this method, the experimental result shown in Fig. 5.33 may be given as an example. Finally, an experiment was conducted in which the feed nozzle position was changed from 35° to 90°. At a feed concentration in

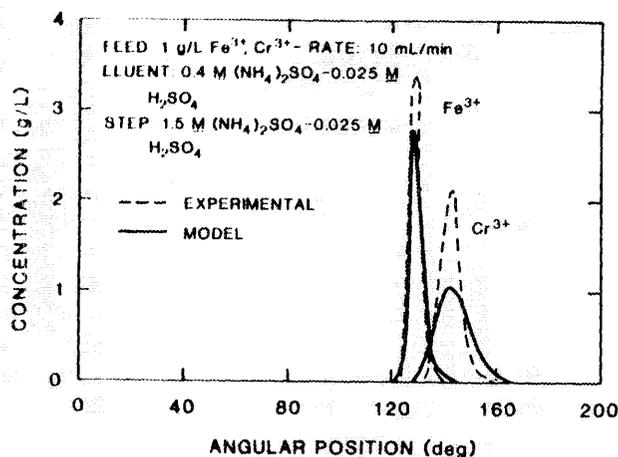


Fig. 5.33. Concentration profiles obtained in step-elution CAC separation of a dilute mixture of ferric and chromic nitrate.

both metals of 1.0 g/L and a feed flow rate of 10 cm^3/min , the step was set at 1.5 M at 90° from the feed location and the rotation rate was set at 180°/h. The model somewhat underestimates the peak concentrations for both the iron and chromium peaks, probably because of slight differences in the eluant concentrations between models. Experimentally, near-complete separation was attained, and the peak concentrations were greater than twice the feed concentration. Average concentrations for iron and chromium were found to be 1.4 and 1.0 g/L, respectively. The separation, therefore, was accomplished with almost complete resolution, a 40% concentration of iron, and no dilution of chromium. This represents a very significant energy saving in the ultimate recovery of the metals. Ideal conditions occur where components are separated in the low concentration eluant, quickly eluted from the resin with the high concentration eluant, and immediately removed from the column to avoid further dispersion.

In displacement chromatography operation, introduction of the feed mixture to the chromatographic bed is followed by elution by a solution containing a component (the displacer) which has affinity for the sorbent greater than that of any component of the feed mixture. The displacer competes for adsorption sites with the feed components and, if present in a sufficiently high concentration, will effectively desorb and concentrate the feed components. If mass transfer resistances are small, the various components will

be distributed along the column length in order of decreasing affinity for the sorbent, with each upstream component acting as a "displacer" for each downstream species. The separated components exit the column as adjacent bands in order of increasing affinity, with the displacer immediately following the feed components. The displacer is then removed from the sorbent with an appropriate regenerant, and the column conditions are restored for the next feed-displacement cycle. The overall result is a separation of a dilute mixture into concentrated fractions. In practice, of course, some overlapping of the eluted peaks will result from dispersion and mass transfer resistance. Impure product fractions are generally recovered and recycled.

Conceptually, displacement chromatography separations can be carried out in a truly continuous manner by arranging the CAC apparatus as shown in Fig. 5.34. In this case, the annular bed is provided with four different solutions: the feed to be separated, the displacer, the regenerant, and the rinse. These four solutions are evenly distributed by an appropriately designed stationary manifold and a flow downward simultaneously through the rotating bed, each occupying a sector of circumference proportional to its flow rate. The solutions exit

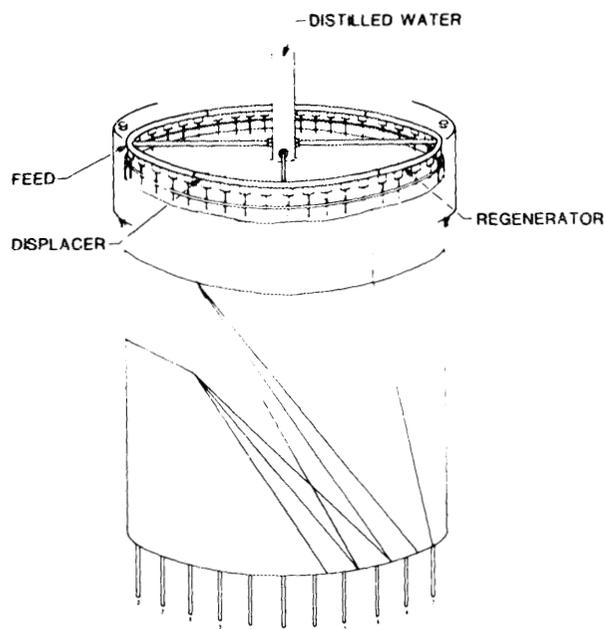


Fig. 5.34. Simplified diagram of a CAC operating in the displacement mode.

through uniformly spaced tubes that rotate together with the annulus. The displacer sector must be placed immediately beside the feed sector and must be sufficiently long (in the θ direction) to completely displace all of the feed components. Following the displacer region, a regeneration region is necessary to regenerate the sorbent material back to the proper form for loading with the feed. Between the regeneration and feed regions, the bed is flushed with a rinse solution to remove excess regenerant from the bed. A fundamental study was made of rare earth separations.⁵⁴

An amino acid mixture of L-glutamic acid, L-valine, and L-leucine was selected as a model separation system.⁵⁵ Dowex 50W-X8 resin ($45 \mu\text{m}$), which was used as the medium, has a total ion-exchange capacity of $5.6 \pm 0.2 \text{ mmol/g}$ dry resin. A typical experimental CAC chromatogram is shown in Fig. 5.35. The focus here is on the feed-loading and displacement processes, since it was determined

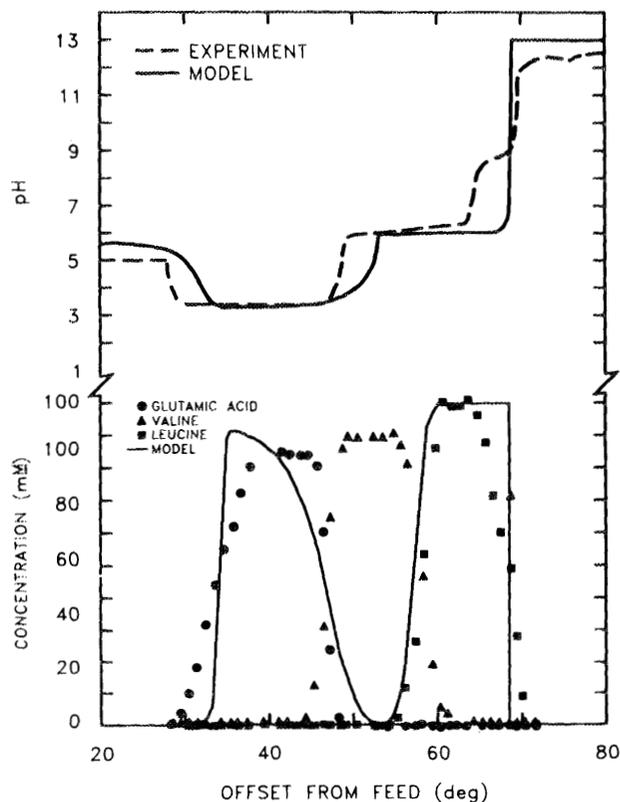


Fig. 5.35. Comparison of experimental and calculated CAC displacement chromatographic separation.

experimentally that the acid regeneration portion of the process completely converted the resin to the hydrogen form. The excess acid was then essentially completely removed from the annular bed by the distilled water rinse. The origin of the angular coordinate in Fig. 5.35 is set at the boundary between the feed sector and the displacement sector.

The quality of the separation is essentially the same as in a fixed-bed experiment that had been previously made, and the plateau concentrations are again in good agreement with Glueckauf's method. The experimental pH profile closely reflects the separation resulting from the buffering effect of the amino acids. Initially, the pH is ~ 7 and reaches a plateau at ~ 3 when glutamic acid is eluted. The pH then rises to values between 5 and 6 during the elution of valine and leucine and makes a final jump to a value between 12 and 13 when the NaOH front breaks through. It is interesting to note that glutamic acid is eluted at concentrations which exceed the solubility limit in water. While the eluted fractions were initially clear, supersaturation, possibly maintained by crystallization kinetics limitations, was relieved by precipitation of crystals when left in a closed, undisturbed container at room temperature for a few days. Of significance is the fact that the exit concentration is 4 to 5 times more concentrated than the feed, and the capacity is very much higher than the typical isocratic separation.

A relatively simple mathematical model, embracing the fundamental features of the system and based on an equilibrium stage model, was developed as a part of this program and is discussed elsewhere.³⁷ Details of the equilibrium relationship and the individual-component mass transfer could be added later at the expense of considerable computer time. Conversely, our model allows an approximate prediction of the concentration levels and of the separation performance achievable requiring only equilibrium data for the separation system. Simple batch chromatography can then be performed to determine the number of equilibrium stages that will provide a reasonable representation of experimental CAC concentration profiles. Therefore, the model serves as a useful guide to selecting optimum operating conditions.

Calculated concentration and pH profiles based upon the equilibrium stage model are also shown in Fig. 5.35. For the purpose of a qualitative comparison, 50 stages were again used in the computations as for the fixed-bed calculations. To

compensate for the slight shift in angular position caused by the neglect of fluid-phase accumulation, the entire computed profiles were also shifted to match the midpoint of the experimental trailing edge of the leucine peak. This correction amounted to $< 6^\circ$ and was in approximate agreement with an estimate of $\epsilon = 0.4$. The general agreement between experimental and calculated profiles is essentially the same as that observed for the fixed-bed run, revealing that the CAC provides little or no additional peak dispersion relative to a corresponding conventional chromatography process.

Thus, it may be concluded that the CAC is ready for commercial application and is indeed proceeding in that direction at several industrial research facilities.

5.4 Materials Research

The DOE Office of Basic Energy Sciences (BES), through the Division of Materials Sciences, supports Chem Tech research programs that are concerned with thermodynamics and kinetics of energy-related materials, as well as basic chemical engineering studies.

5.4.1 Thermodynamics and Kinetics of Energy-Related Materials

During this reporting period, we have been studying the oxygen nonstoichiometry and thermodynamics of the oxide superconductor $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (123). In the first phase of the study, the literature data for the dependence of x in 123 on temperature and oxygen pressure were retrieved to provide a numerical data base. The data base was reviewed, and each data set was fitted to give the general interrelationship of the experimental variables.⁵⁶ Since these data were not internally consistent, an experimental program was started in which the dependence of the nonstoichiometry of 123 was studied over five orders of magnitude in oxygen pressure and from 573 to 1173 K.⁵⁷ These data were used to derive a chemical thermodynamic representation, which is also the only known expression that correlates the experimental variables. These results were also compared with those of

several other investigations to identify the self-consistent sets of data. Hydrogen-reduction methods for determining the absolute oxygen-to-metal ratio were developed.⁵⁷ This work has been presented to several audiences⁵⁸ and was selected as the feature article in the October 1989 issue of the *Journal of the American Ceramic Society*.⁵⁷

It was also noted that carbonate in 123 had the potential for degrading the superconducting properties. Therefore, the solubility of CO₂ in 123 was determined at 1173 K, a common processing temperature. Reports on these results are in the publication process.^{59,60}

An informal cooperative effort was established with ANL to determine the standard enthalpies of formation of compounds in the Y-Ba-Cu-O system. We provided well-characterized compounds for the ANL experiments. Again, the results are in the publication process.⁶¹

Earlier work on actinides has now been published.⁶² The nonstoichiometry of UO_{2-x}N_{3x/4}, the UO₂-based solid solution extending toward U₂N₃, was determined experimentally and represented by an adaptation of solution thermodynamics. This resulted in expressions that accurately reflect the interdependence of x on temperature and the partial pressure of N₂ and O₂. Partial molal Gibbs free energy expressions were derived for UO₂ and U_{4/3}N₂ and are useful for any thermodynamic calculation involving the solution within the range of the data. A description of this and other nuclear fuels research was also presented as a seminar.⁶³

5.4.2 Nucleation, Growth, and Transport Phenomena

Two interdependent research programs in the Chemical Technology Division utilized laser-light-scattering spectroscopy to investigate the phenomena of homogeneous precipitation, controlled aggregation, and particle growth. The first was a series of fundamental studies of nucleation and growth in systems that are of interest as precursor materials in ultrafine processing for a new generation of ceramic materials. The second area of interest was the measurement and theoretical interpretation of transport properties of fluids in the critical region. The transport properties, such as viscosity and diffusion coefficient, provided important clues to the

behavior of the fluid media in which the precipitation occurs.

As a result of the difficulty in forming uniformly mixed oxide particles by homogeneous precipitation techniques, another program, the Electric Dispersion Reactor, was initiated which investigated the use of pulsed dc electric fields in the synthesis of precursor particles for ceramic applications. This program was a collaborative research effort between the Emulsion-Phase Contactor Program and the Nucleation, Growth, and Transport Phenomena Program.

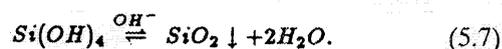
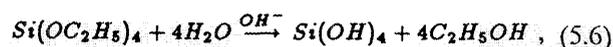
Particle Growth Kinetics in the Synthesis of Pure Component and Mixed Ceramic Powders by Metal Alkoxide Hydrolysis

The synthesis of single-component and mixed oxide ceramic powders that consist of very fine (submicron), monodisperse particles with uniform composition and high purity is desirable in the improvement of existing ceramics and in the development of new ceramic materials. Metal alkoxide hydrolysis is a very attractive method for the synthesis of ultrapure powders at low temperatures by the sol-gel process. This method has been used to synthesize monodisperse particles of silica, titania, zirconia, and composite materials.⁶⁴⁻⁶⁸ The parameters that affect the monodispersity and morphology of the final powder include the reactant concentrations (i.e., water, alkoxide, and catalyst), temperature, solvent, and the type of alkoxide group that is attached to the central metal. Fundamental studies have focused on how these important parameters affect the chemical kinetics and the colloidal stability of the particles. The ultimate goal is to eventually model, from microscopic properties and Brownian coagulation, the formation of submicron particles by metal alkoxide hydrolysis.

During this phase of the program, research efforts have primarily focused on two systems: the synthesis of silica from the hydrolysis of tetraethylorthosilicate (TEOS) and the formation of mixed powders of alumina and titania from the hydrolysis of a mixture of aluminum *sec*-butoxide and titanium 1-butoxide in ethanol and 1-butanol. The synthesis of submicron silica particles by the hydrolysis and condensation of dilute and concentrated solutions of TEOS was studied in low-

molecular-weight alcohols (C_1 - C_4). A base (ammonia) was used to catalyze the reaction. Raman spectroscopy, gas chromatography, and the molybdate method were used to establish the hydrolysis and condensation kinetics. Dynamic and classical light-scattering techniques were employed to monitor particle growth, particle number concentration kinetics, and particle size distribution. These experiments were used to test the theories of homogeneous nucleation and controlled aggregation, which have been proposed as mechanisms that govern the growth of submicron monodisperse silica particles by TEOS hydrolysis.

Understanding the mechanisms that control the formation of monodisperse silica by the base-catalyzed hydrolysis of TEOS (known as the Stober process) required an initial investigation of the chemical behavior of the hydrolysis and condensation reactions.⁶⁵ The overall hydrolysis and condensation reactions are given as follows:



Using Eqs. (5.6) and (5.7) as the reaction mechanisms, which has been proposed by others,^{69,70} and taking into account the presence of colloidal particles after nucleation and the precipitation of silicic acid on the surface of these particles, the following equations were proposed for the hydrolysis and condensation reactions:

$$\frac{d[TEOS]}{dt} = -k_H^*[H_2O]^{1.5}[NH_3]^{0.5}[TEOS], \quad (5.8)$$

$$\frac{d[Si(OH)_4]}{dt} = k_H^*[H_2O]^{1.5}[NH_3]^{0.5}[TEOS] - k_C^*S_p[Si(OH)_4] + k_D^*S_p[H_2O]^2[SiO_2], \quad (5.9)$$

where

- k_H^* = first-order hydrolysis rate constant,
- k_C^* = condensation rate constant,
- k_D^* = dissolution rate constant,
- S_p = total surface area of the colloidal particles.

In practice, the total soluble silica concentration, [TS], was measured by the molybdate method. Therefore, Eq. (5.9) was written in terms of [TS] by using the equation $[TS] = [TEOS] + [Si(OH)_4]$. The [TEOS] was determined by gas

chromatography analysis and by Raman spectroscopy. The results from both methods were in excellent agreement. The quantity S_p was determined by obtaining particle size and particle size distributions by dynamic light scattering, and the particle number concentration was determined from turbidity measurements. Figure 5.36 shows the fit by Eq. (5.7) of the experimental [TS] and $[Si(OH)_4]$ data and the theoretical [TEOS] data for the hydrolysis of 0.05 M TEOS in ethanol (6 M H_2O and 0.7 M NH_3). The data are adequately described by the model. Equation (5.7) was also used to fit the condensation data of 1 M TEOS in ethanol (6 M H_2O and 0.7 M NH_3). The rate of condensation was proportional to the total surface area of the colloidal particles. After careful evaluation of the hydrolysis and condensation chemistry, it was determined that models which try to explain the synthesis of silica particles by the Stober process must include both the controlled aggregation mechanism (which governs the earliest stages of particle growth) and the precipitation of monomer on the surface of the particle (which governs the largest portion of particle growth).

The monitoring of particle size and particle size distribution is an integral part of the program for understanding how submicron particles are formed. Figure 5.37 shows the particle size distribution obtained from dynamic light scattering and transmission electron microscopy (TEM) of silica particles that were formed during the hydrolysis of 0.05 M TEOS and 1 M TEOS in ethanol. The

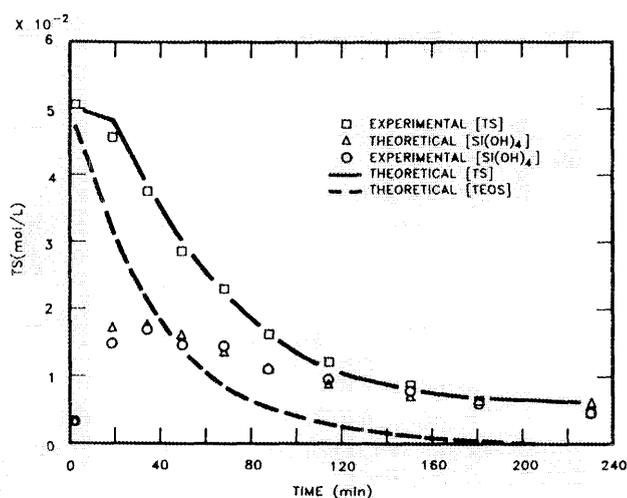


Fig. 5.36. TEOS hydrolysis in ethanol (0.05 M TEOS, 0.7 M NH_4OH , 6.0 M H_2O).

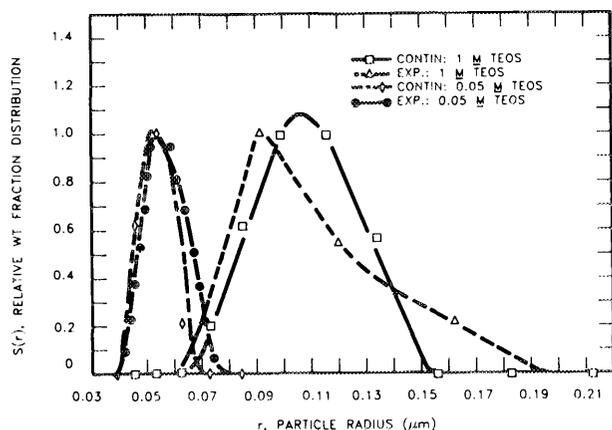


Fig. 5.37. TEOS hydrolysis in ethanol: particle size distribution ($0.7\text{ M NH}_4\text{OH}$, $6.0\text{ M H}_2\text{O}$).

results from both methods were in excellent agreement. Light scattering was more attractive because of speed and because the analysis was done in situ.

In the formation of alumina-titania particles, it was discovered that alumina content affects the rate of precipitation and colloidal stability, which ultimately affects the particle morphology. Earlier studies have shown that monosized titania particles are easily formed in ethanol, 1-propanol, and 1-butanol.⁶⁶ Figure 5.38 shows the particles containing 2:1, 1:1, 1:2, and 0:1 (molar ratio) alumina:titania. As the alumina content is decreased, the particles change from smaller, incoherent, open structures (a) to the formation of larger, more spherical and compact particles (d). It was proposed that the effect of alumina on the shape of the particles is due to an increase in the rate of nucleation caused by the alumina and to an increase in repulsive force (barrier to aggregation) between the particles as the alumina content increases. The zeta potential for titania:alumina particles increases from 10 mV for pure titania particles to ~ 60 mV for alumina particles, thus, indicating that the barrier to aggregation increases with the alumina content in the particles.

It was proposed by Thomas and McCorkle⁷¹ that as the barrier to aggregation increases, the particles have a tendency to form chain aggregates of rather compact clusters. Thus, the final particles formed by the aggregation of these smaller particles will consist of a more open and incoherent structure.

A population balance for Brownian particles was developed and tested to predict the growth

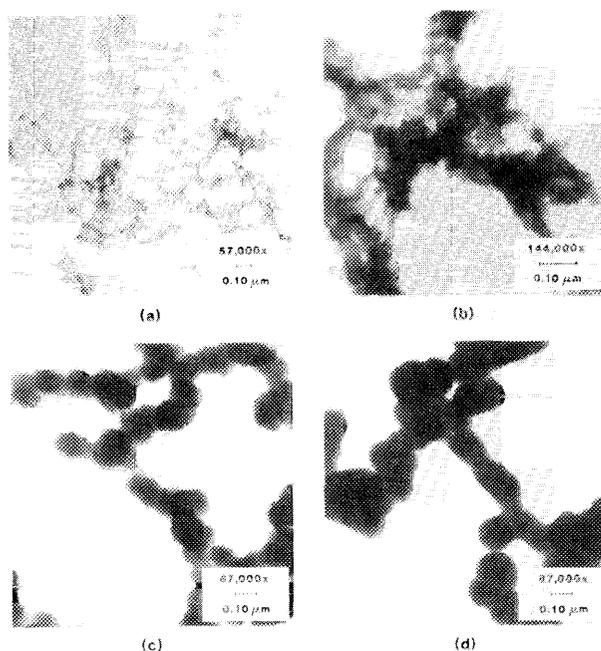


Fig. 5.38. TEM of "alumina"-titania particles produced in ethanol (effect of the initial molar ratio of $[\text{ASB}]:[\text{TIE}]$; $0.8\text{ M H}_2\text{O}$): (a) 0.015 M TIE , 0.023 M ASB ; (b) 0.023 M TIE , 0.019 M ASB ; (c) 0.031 M TIE , 0.012 M ASB ; (d) 0.02 M TIE .

kinetics of particles that are produced by the controlled aggregation model. This model will be applied to the systems where spherical particles are formed in dilute solutions (i.e., silica, titania, and zirconia). To apply the population balance model to systems where aggregates form nonspherical particles, it is necessary to couple the population balance model with Brownian or Stokesian computer simulations where the shape of the particles as well as the particle size distribution is determined.

Electric Dispersion Reactor

The formation of uniformly mixed oxide particles by homogeneous precipitation was found to be difficult and even impossible in some cases. This is especially true as the number of constituents in the particle increases. Therefore, a new concept called the electric dispersion reactor (EDR—see Fig. 5.39) was developed which allows the continuous production of composite oxide ceramic precursor materials, such as superconductors. The basic concept of the EDR involved the creation of a microreactor in the form of a micron-sized droplet

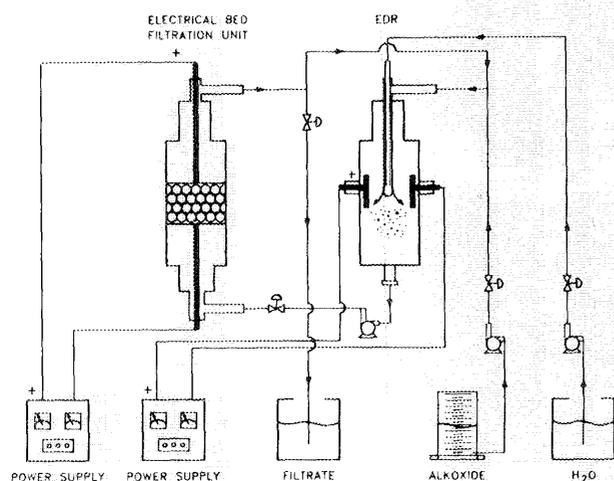


Fig. 5.39. The electric dispersion reactor (EDR) and the electric bed filtration unit.

within a liquid continuous phase. A drop was introduced between electrodes on which was imposed a pulsing dc electric field. Under properly selected field conditions, the drops were shattered creating an emulsion consisting of micron-sized droplets in a close size range. These droplets served as the basis for a microreactor, with some of the reactants in the continuous phase and some in the drops. After forming the solid particles, an electric bed filtration unit could be used to remove the colloidal particles (See Fig. 5.39). Several reactant schemes were tested. The first involved the formation silica particles from the reaction of TEOS with water that contained 0.7 *M* ammonia. The continuous phase was TEOS, and 0.7 *M* ammonia solution was the disperse phase (drops). TEOS diffused into the submicron aqueous droplets and was hydrolyzed to form silicic acid, which then condenses to form silica, which precipitates in the interfacial region. Highly porous silica shells were formed using this scenario (See Fig. 5.40). Denser silica particles were formed when the continuous phase was a mixture of acetic acid in 2-ethyl-1-hexanol and the disperse phase was sodium metasilicate in water (See Fig. 5.41). Finally, spherical particles containing 1:2:3 molar ratio of Y:Ba:Cu were produced when the continuous phase

contained a mixture of ammonia, cyclohexane, ethanol, and 2-ethyl-1-hexanol and the disperse phase consisted of 0.1 *M* yttrium nitrate, 0.2 *M* barium nitrate, and 0.3 *M* cupric chloride in water (Fig. 5.42). A patent application was filed on this invention.

Transport Properties of Fluids

Initial studies were done to measure the mutual diffusion coefficient of methanol in benzene, *tert*-butyl alcohol in water, and TEOS in methanol by low-angle dynamic laser-light scattering (i.e., scattering angles from 5 to 10°). Mutual diffusion coefficients ($8.0 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$) for a 0.5 mol fraction benzene:methanol solution were within 3% of literature values.⁷² The mutual diffusion coefficient for 0.25 mol fraction *tert*-butyl alcohol in water was $1 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$ at $\leq 22^\circ\text{C}$, which compares with the literature value ($1 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$; 0.26 mol fraction *tert*-butyl alcohol in water at 22°C).⁷³ Solutions with 0.1 and 0.5 mol fraction TEOS in methanol yielded mutual diffusion coefficients of 1.1×10^{-5} and 0.96×10^{-5} , respectively. The mutual diffusion coefficients for TEOS in methanol indicated that TEOS has a molecular complexity of one methanol, which is in accordance with the literature value. Low-angle light-scattering measurements will be helpful in determining the molecular complexity of various alkoxydes in various solvents and the average size variation of the "pre"-nucleation species.

5.5 Cold Fusion

The early experiments of Pons and Fleischman⁷⁴ and of Jones⁷⁵ excited the scientific world concerning the possibilities of D-D fusion occurring at ambient temperatures ("cold fusion") during electrolysis of heavy water using a palladium cathode.⁷⁶⁻⁷⁹ Most researchers from laboratories all over the world were subsequently unable to confirm the findings of Pons and Fleischman concerning major quantities of excess energy; a few laboratories, including our own, have observed the generation of small amounts of excess heat (5 to 15%), the production of significant quantities of tritium, and some neutron anomalies.⁸⁰⁻⁸²

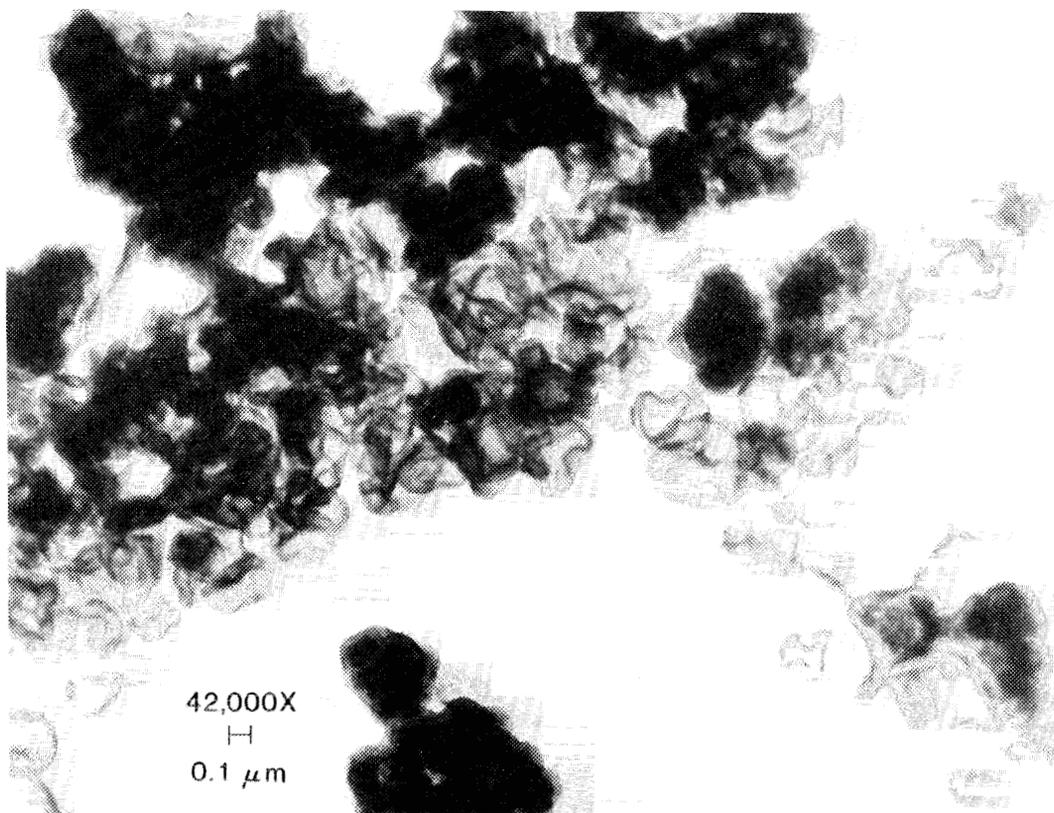


Fig. 5.40. Silica particles produced in the EDR [$0.7\text{ M NH}_4\text{OH}$ (dispersed phase) and TEOS (continuous phase)].

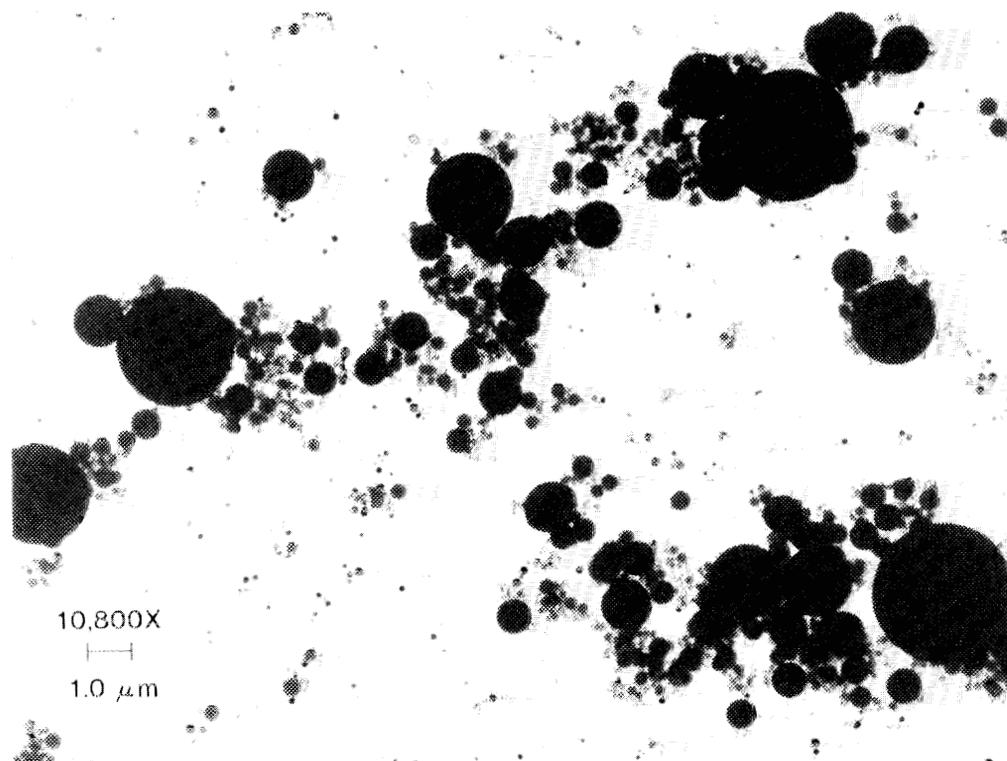


Fig. 5.41. Silica particles produced in the EDR [sodium metasilicate in the water (dispersed phase) and acetic acid in 2-ethyl-1-hexanol (continuous phase)].

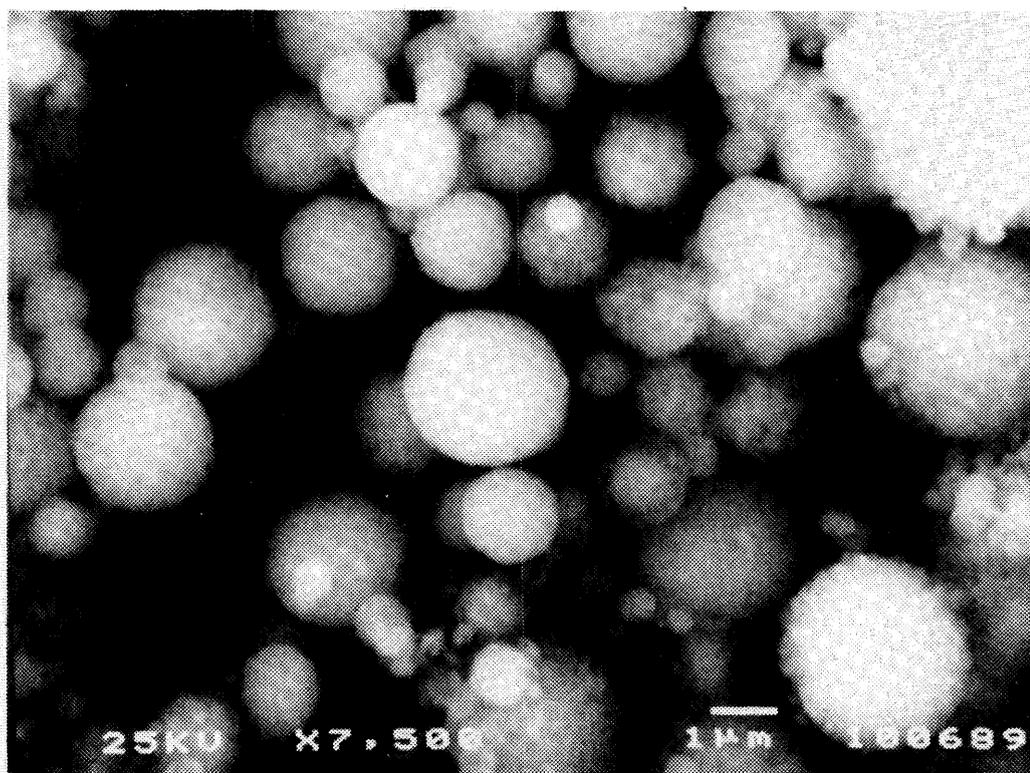


Fig. 5.42. Particles produced in the EDR containing hydrous oxides of Y-Ba-Cu (~1:2:3 molar ratios).

Research in the Chemical Technology Division

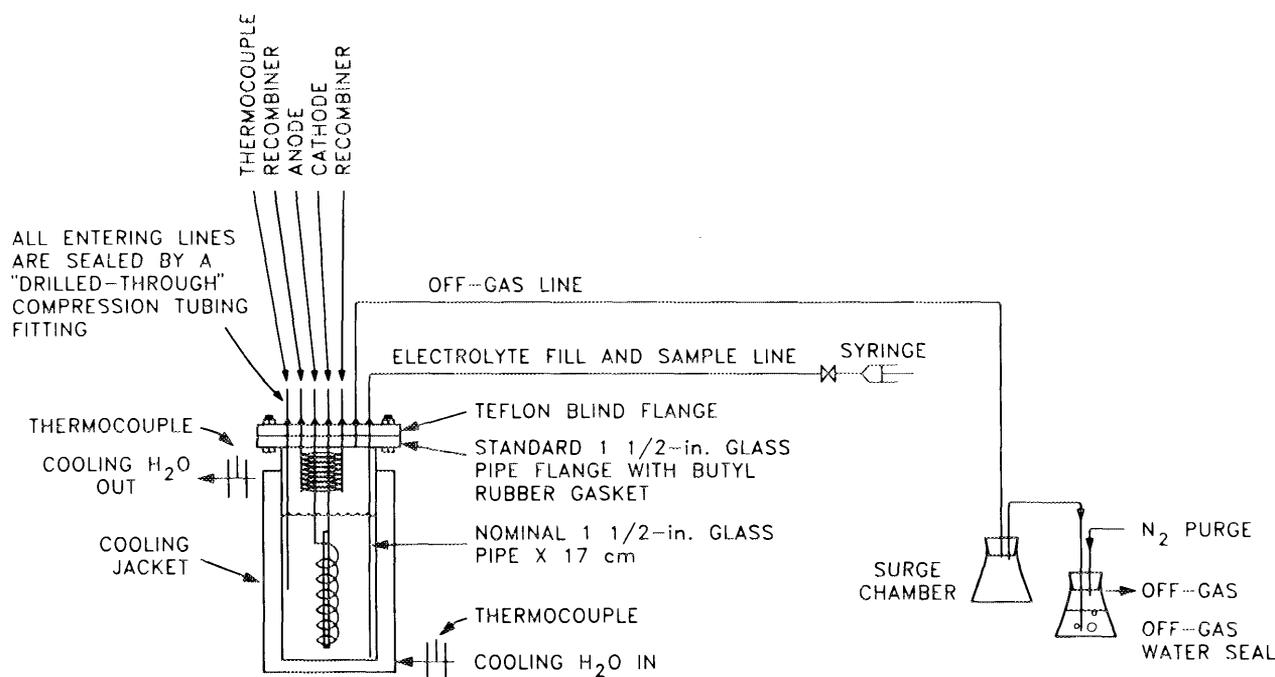
A multidisciplinary group of researchers, including chemists, physicists, and chemical engineers, has conducted a series of experiments on the electrolysis of heavy water using palladium cathodes in an LiOD electrolyte. The group's goal has been to investigate possible thermal or nuclear reactions occurring during the electrolysis process.^{83,84} The experimental system utilized positive heat removal by circulating cooling water, a design concept that is amenable to system scaleup and continuous operation.

Experimental Design

Initial experiments employed an open system in which the electrolyzed D_2 and O_2 exited the system and an inert gas purge was utilized to prevent gas

accumulations. This necessitated correction of the heat balance for the heat of formation of D_2O in the exiting gases (assuming 100% electrical efficiency and no recombination internally within the cell) and the latent heat contained in the exiting gases. These assumptions introduce a degree of uncertainty in the energy balance, especially where small heat excesses are measured. This approach has been used in the vast majority of "positive results" that have been reported from other laboratories using "open systems."

Our most recent experiments have included a proprietary internal D_2-O_2 recombiner, under patent consideration by DOE, which obviated the need for the assumptions employed for the open systems since this electrolysis cell was completely sealed (see Fig. 5.43).⁸⁴ The recombiner proved to be capable of catalyzing the recombination of D_2 and O_2 to D_2O at rates as high as 1.6 g/h of D_2O for many hundreds of hours. This system included a data acquisition system based on a microcomputer that acquired and stored averaged temperature and voltage data every minute.



THE ENTIRE SYSTEM IS INSULATED WITH 4 in. OF FIBERGLASS INSULATION

Fig. 5.43. Closed-system electrolysis cell used in cold fusion tests.

Measurement of Nuclear Particles

All experiments have been monitored for neutrons and gamma rays by means of a NE-213 scintillator with a pulse-shape discriminator placed immediately adjacent to the cell. A separate gamma-ray spectrometer employing a NaI detector was also used for detection of gamma rays. Both the neutron and gamma-ray spectra were periodically recorded by a small computer system. The tritium concentration of the electrolyte was measured regularly by sampling the electrolyte and using an external scintillation system.

Experimental Results

Several different tests have been made with both open and closed electrolysis systems. Two different long-term (hundreds of hours) open-system experiments yielded heat in excess of that provided by joule heating (generally in the range of 5 to 10%). The experimental uncertainty for these experiments was 3 to 5%. In one test, excess energy

of up to 50% was observed for a few hours (see Fig. 5.44). Excess energy of the order of 5 to 8% has also been measured for the "closed system" containing a recombining where the experimental uncertainties were 1 to 3%. Measurements on this latter system are more reliable because fewer assumptions are required due to its sealed-cell configuration.

Background measurements for neutron counts were performed before and after individual experiments using the same counting systems. Neutron count rates were measured by a NE-213 scintillator which was placed immediately adjacent to the insulated cell and which employed pulse-shape discrimination having a threshold of 1.2 MeV. The overall detection efficiency was 1.46×10^{-3} , as determined by a ²⁵²Cf source. Neutron count rates exceeding three standard deviations of background were rare with only about three excursions being observed over several thousand hours of run time. Figure 5.45 illustrates one such excursion after 70 h of operation during our third experiment.

A 25-fold increase in tritium was observed over a 2-d period during the early phases of one test. No

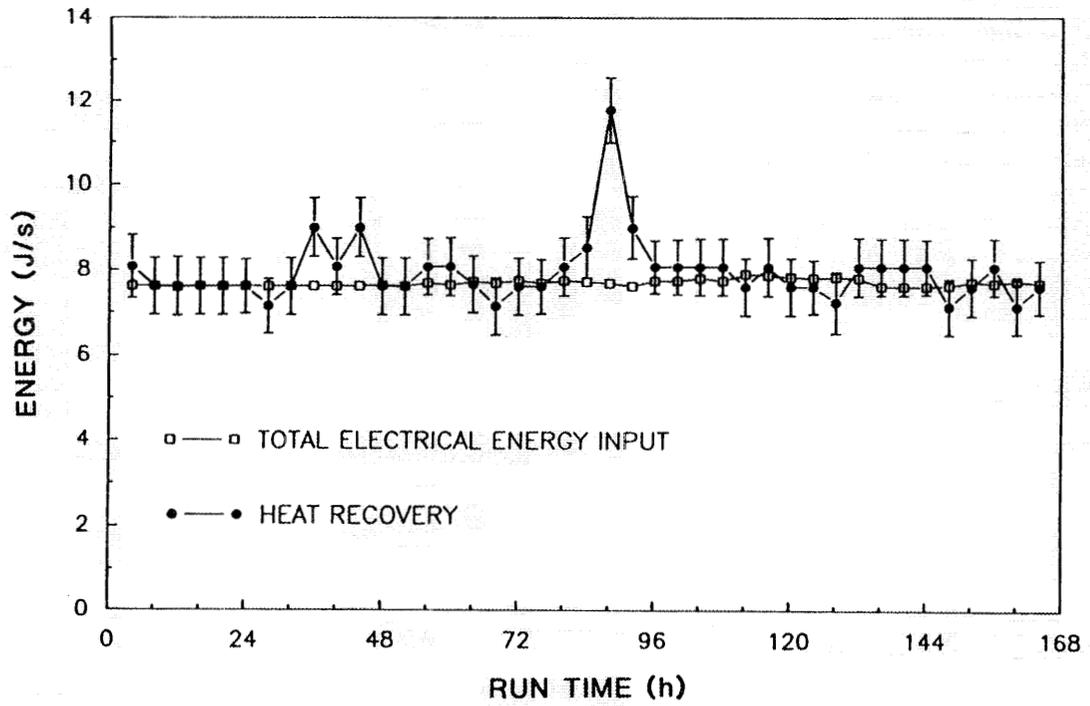


Fig. 5.44. Energy balance for Test CF-2.

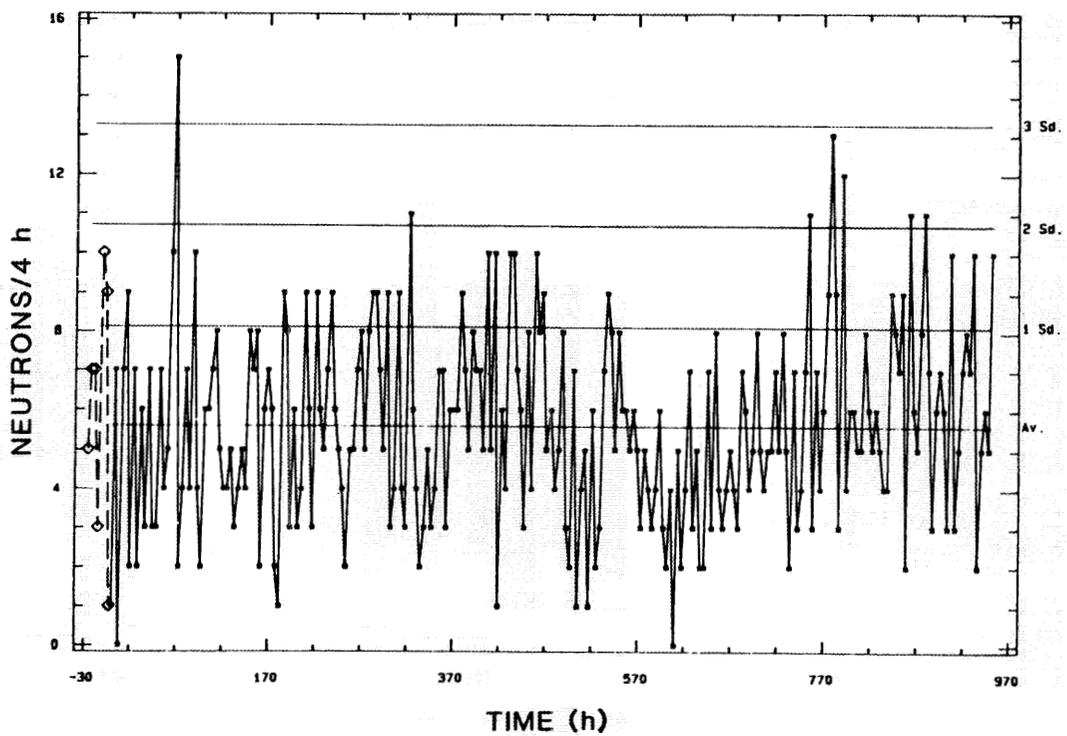


Fig. 5.45. Neutron count rate for Test CF-3.

further increases in tritium were observed during this or any other experiment.

Although several research groups, including our own, have observed apparent excess energy during electrolysis of heavy water, this phenomenon is not understood or predictable. System instabilities or perturbations may have initiated this event, but this also needs to be further studied and defined. There has been no verification of nuclear interactions (cold fusion) that is compatible with current theory

and with the level of excess energy that has been detected. However, except for a very few research groups, including our own, neutron count rates have not been measured simultaneously with the determination of an energy balance. Our capabilities on the design and operation of a closed electrolysis system, coupled with nuclear measurements, appear to offer a unique approach that will allow a more definitive investigation of this interesting phenomenon.

References

1. B. D. Faison, C. A. Woodward, and R. M. Bean, "Microbial Solubilization of a Preoxidized Subbituminous Coal: Product Characterization," *Appl. Biochem. Biotechnol.*, in press (1989).
2. B. H. Davison, D. M. Nicklaus, A. Misra, S. N. Lewis, and B. D. Faison, "Utilization of Microbially Solubilized Coal: Preliminary Studies on Anaerobic Conversion," *Appl. Biochem. Biotechnol.*, in press (1989).
3. C. D. Scott, and C. A. Woodward, "Coal Solubilization by Enzymes in Organic Solvents," *Appl. Biochem. Biotechnol.*, in press (1989).
4. C. D. Scott, B. D. Faison, and C. A. Woodward, "Solubilization of Coal by Biocatalysts in Organic Solvents," presented at the Direct Liquefaction Contractors' Review Meeting, Pittsburgh, PA, October 2-4, 1989.
5. S. G. Reeves and D. O. Hall, *Methods Enzymol.* **69**, 85 (1980).
6. C. D. Scott, C. A. Woodward, and J. E. Thompson, *Enzyme Microb. Technol.* **11**, 258-63 (1989).
7. G. Johansson, and M. Joelsson, *Anal. Biochem.* **158**, 104-10 (1986).
8. R. K. Bajpai, J. E. Thompson, and B. H. Davison, *Appl. Biochem. Biotechnol.* **24-25** (1990, in press).
9. J. F. Richardson and W. N. Zaki, *Trans. Inst. Chem. Eng.*, **32**, 35 (1954).
10. O. A. Basaran, T. C. Scott, and C. H. Byers, "Drop Oscillations in Liquid-Liquid Systems," *AIChE J.*, **35**, 1263 (1989).
11. C. A. Miller, and L. E. Scriven, "The Oscillations of a Fluid Droplet Immersed in Another Fluid," *J. Fluid Mech.*, **32**, 417 (1968).
12. S. V. Subramanyam, "A Note on the Damping and Oscillations of a Fluid Drop Moving in Another Fluid," *J. Fluid Mech.*, **37**, 715 (1969).
13. J. G. Kirkwood and F. P. Buff, "The Statistical Theory of Solutions I.," *J. Chem. Phys.*, **19**, 774 (1951).
14. H. D. Cochran, L. L. Lee, and D. M. Pfund, "Application of the Kirkwood-Buff Theory of Solutions to Dilute Supercritical Solutions," *Fluid Ph. Equilib.*, **34**, 219 (1987).
15. D. M. Pfund, L. L. Lee, and H. D. Cochran, "Application of the Kirkwood-Buff Theory of Solutions to Dilute Supercritical Solutions II. The Excluded Volume and Local Composition Models," *Fluid Ph. Equilib.*, **39**, 161 (1988.)
16. H. D. Cochran, D. M. Pfund, and L. L. Lee, "Theoretical Models of Thermodynamic Properties of Supercritical Solutions," *Sep. Sci. & Technol.*, **23**, 2031 (1988).

17. H. D. Cochran, L. L. Lee, and D. M. Pfund, "Structure and Properties of Supercritical Fluid Mixtures from Kirkwood-Buff Fluctuation Theory and Integral Equation Methods," *Fluctuation Theory of Mixtures*, E. Matteoli and G. A. Mansoori, eds., Taylor and Francis, New York, in press.
18. H. D. Cochran and L. L. Lee, "General Behavior of Dilute Binary Solutions," *AIChE J.*, **33**, 1391 (1987).
19. H. D. Cochran and L. L. Lee, "General Behavior of Dilute Binary Solutions," *AIChE J.*, **34**, 170 (1988).
20. H. D. Cochran, D. M. Pfund, and L. L. Lee, "Study of Fluctuations in Supercritical Solutions by an Integral Equation Method," *Proc. Int. Symp. on Supercritical Fluids*, 245, M. Perrut, ed., Nice (1988).
21. H. D. Cochran and L. L. Lee, "Solvation Structure in Supercritical Fluid Mixtures Based on Molecular Distribution Functions," *Supercritical Fluid Science and Technology*, K. P. Johnston and J. M. L. Penninger, eds., ACS Symp. Ser. 406, 27 (1989).
22. L. L. Lee, P. G. Debenedetti, and H. D. Cochran, "Fluctuation Theory of Supercritical Solutions," *Supercritical Fluid Technology*, T. G. Bruno and J. F. Ely, eds., CRS Press, submitted.
23. R.-S. Wu, L. L. Lee, and H. D. Cochran, "The Structure of Dilute Supercritical Solutions: Clustering of Solvent and Solute Molecules and the Thermodynamic Effects," *I&EC Res.*, submitted.
24. H. D. Cochran, L. L. Lee, and R.-S. Wu, "Molecular Clustering in Supercritical Solutions," *Sep. Sci. & Technol.*, submitted.
25. J. F. Brennecke and C. A. Eckert, "Fluorescence Spectroscopy Studies of Intermolecular Interactions in Supercritical Fluids," *Supercritical Fluids Science and Technology*, K. P. Johnston and J. M. L. Penninger, eds., ACS Symp. Ser. 406, 14 (1989).
26. J. C. Wheeler, "Behavior of a Solute near the Critical Point of an Almost Pure Solvent," *Ber. Bunsenges. Phys. Chem.*, **76**, 308 (1972).
27. P. G. Debenedetti and R. S. Mohamed, "Attractive, Weakly-Attractive, and Repulsive Near-Critical Systems," *J. Chem. Phys.*, **90**, 4528 (1989).
28. I. B. Petsche and P. G. Debenedetti, "Solute-Solvent Interactions in Infinitely Dilute Supercritical Mixtures: a Molecular Dynamics Investigation," *J. Chem. Phys.*, in press (1989).
29. D. R. Biggerstaff and R. H. Wood, "Apparent Molar Volumes of Aqueous Argon, Ethylene, and Xenon from 300 to 716 K," *J. Phys. Chem.*, **92**, 1988 (1989).
30. L. L. Lee and K. S. Shing, "A Test Particle Approach to the Zero Value Theorems of Molecular Correlation Functions," *J. Chem. Phys.*, submitted (1989).
31. D. M. Pfund, L. L. Lee, and H. D. Cochran, "Chemical Potential Prediction in Realistic Fluid Models with Scaled-Particle Theory," *Int. J. Thermophys.*, in press (1989).
32. D. M. Pfund, H. D. Cochran, and L. L. Lee, "Chemical Potentials from Integral Equations," presented at the AIChE Annual Meeting, paper 42e, San Francisco, CA, 1989.
33. T. A. Barber, H. D. Cochran, and P. R. Bienkowski, "Solubility of Solid CCl_4 in Supercritical CF_4 Using Directly Coupled Supercritical Fluid Extraction-Mass Spectrometry," *Sep. Sci. & Technol.*, submitted (1989).
34. R. G. Rubio, J. C. G. Calado, P. Clancy, and W. B. Streett, "A Theoretical and Experimental Study of the Equation of State of Tetrafluoromethane," *J. Phys. Chem.*, **89**, 4637 (1985).
35. V. P. Gilbert, P. R. Bienkowski, and H. D. Cochran, "Proposed Technique for Surface Dynamics Measurements at High pressure," *Sep. Sci. & Technol.*, **23**, 1941 (1988).

36. W. G. Sisson, J. M. Begovich, C. H. Byers, and C. D. Scott, "Continuous Chromatography," *CHEM TECH* (1988).
37. G. Carta and C. H. Byers, "Novel Applications of Annular Chromatography" pp. 167-85 in *New Directions in Sorption Technology*, Keller, G. E., II, and Yang, R. T., eds, Butterworth, Stoneham, MA, 1989.
38. J. P. DeCarli, II, G. Carta, and C. H. Byers, *Advanced Techniques for Energy-Efficient Industrial Scale Continuous Chromatography* ORNL/TM 11282, Oak Ridge National Laboratory, 1989.
39. C. H. Byers, C. D. Scott, and G. Carta, "Recent Research in Continuous Chromatography", presented at the AIChE Annual Meeting, San Francisco, CA, 1989.
40. A. J. Howard, G. Carta, and C. H., Byers, *Separation of Sugars by Continuous Annular Chromatography*, ORNL/TM 10318, Oak Ridge National Laboratory, 1987.
41. A. J. Howard, G. Carta, and C. H. Byers, "Novel Applications of Continuous Annular Chromatography: Separation of Sugars", *Ind. Eng. Chem. Research*, **27**, 1873 (1988).
42. V. T. Taniguchi and C. H. Byers, "Large Scale, Continuous Linear Elution and Displacement Chromatography for Process-Scale Bioseparations Employing A Continuous Annular Chromatograph", presented at PREP '89, Washington, DC, May 9, 1989.
43. C. H. Byers and G. Carta, "The Application of Continuous Annular Chromatography to Biochemical Separations," presented at the Sixth Symposium on Separation Science and Technology for Energy Applications, Knoxville, TN, October 1989 and to be published in *Sep. Sci. Technol.*
44. C. H. Byers, W. G. Sisson, and J. P. DeCarli, II, "The Use of Gradient Elution in Optimizing Continuous Annular Ion Exchange Chromatography with Applications to Metal Separations," in *Ion Exchange for Industry*, M. Streat, ed., Ellis Horwood, Chichester, 242, 1988.
45. J. P. DeCarli II, G. Carta, and C. H. Byers, "The Separation of Amino Acids by Continuous Annular Displacement Chromatography" presented at PREP '89, Washington, DC, May 9, 1989.
46. J. B. Butt, *Reaction Kinetics and Reactor Design*, Prentice-Hall, Englewood Cliffs, N.J., 1980.
47. C. H. Byers, W. G. Sisson, J. P. DeCarli, II, and G. Carta, "Pilot -Scale Studies of Sugar Separations by Continuous Chromatography," *Appl. Biochem. Biotechnol.* **20**, 635 (1989).
48. C. H. Byers, W. G. Sisson, J. P. DeCarli, II, and G. Carta, "Separation of Sugars on a Pilot Scale by Continuous Annular Chromatography," *Biotechnol. Prog.*, in press (1989).
49. G. Carta, "Exact analytic solution of a mathematical model for chromatographic operations," *Chem. Eng. Sci.*, **43**, 2877 (1988).
50. G. T. Bloomingburg, J. S. Bauer, G. Carta, and C. H. Byers, "Preparative-Scale Protein Separations by Continuous Annular Chromatography," submitted to *Sep. Sci. Technol.* (October 1989).
51. J. M. Holmes, and C. H. Byers, *An Economic and Energy Evaluation of the Replacement of Contentional Technology with Continuous Chromatography in the Production of High-Fructose Sugar*, DOE/OR/21400-T352 (DE89010393), October 1988.
52. C. H. Byers, J. M. Begovich, and J. M. Holmes, *An Economic and Energy Evaluation of the Replacement of Contentional Technology with Continuous Chromatography in the Dump Leaching of Copper Ores*, DOE/OR/21400-T357 (DE89009038), February 1989.
53. G. Carta, J. P. DeCarli II, C. H. Byers, and W. G. Sisson, "Separation of Metals by Continuous Annular Chromatography with Step Elution," *Chem. Eng. Comm.*, **79**, 207-27 (1989).
54. V. T. Taniguchi, and C. H. Byers, "Large Scale Chromatographic Separations using Continuous Displacement Chromatography (CDC)" in *Rare Earths, Extractions, Preparation and Applications*, R. G. Bautista and M. M. Wong, Minerals, Metals, and Materials Soc., 1989.

55. J. P. DeCarli, II, G. Carta, and C. H. Byers, "Displacement Separations by Continuous Annular Chromatography," submitted to *AIChE J* (October 1989).
56. T. B. Lindemer and A. L. Sutton, Jr., *Review of Nonstoichiometry in $YBa_2Cu_3O_{7-x}$* , ORNL/TM-10827, Oak Ridge National Laboratory, November 1988.
57. T. B. Lindemer, J. F. Hunley, J. E. Gates, A. L. Sutton, Jr., J. Brynestad, C. R. Hubbard, and P. K. Gallagher, "Experimental and Thermodynamic Study of Nonstoichiometry in $<YBa_2Cu_3O_{7-x}>$," *J. Am. Ceram. Soc.* **72**, 1775–88 (1989). Also, ORNL/TM-10899, Oak Ridge National Laboratory, May 1989.
58. The preceding research was presented to the following audiences:
Seminar, Department of Ceramics, Rutgers, The State University, Piscataway, NJ, February 7, 1989.
91st Annual Meeting of the American Ceramic Society, Indianapolis, IN, April 23–27, 1989.
44th Annual Calorimetry Conference, Oak Ridge, TN, July 30–Aug. 4, 1989.
Seminar, National Institute of Standards and Technology, Gaithersburg, MD, August 10, 1989.
Seminar, School of Materials Engineering, Purdue University, West Lafayette, IN, September 29, 1989.
Annual Meeting of the Metallurgical Society, Indianapolis, IN, October 2–5, 1989.
Basic Energy Sciences Review, ORNL, Oak Ridge, TN, October 16–17, 1989.
59. T. B. Lindemer, C. R. Hubbard, and J. Brynestad, " CO_2 Solubility in $YBa_2Cu_3O_{7-x}$ " to be submitted to *Physica C*.
60. C. R. Hubbard, T. B. Lindemer, J. Brynestad, and D. Kroeger, "Reaction of CO_2 with the High T_c Superconductor $YBa_2Cu_3O_{7-x}$ " presented at the annual meeting of the Am. Cryst. Assn., Seattle, WA, July 23–28, 1989.
61. L. R. Morss, S. P. Dorris, T. B. Lindemer, and N. Naito, "Enthalpies of Formation at 298 K of Complex Oxides Coexisting with $YBa_2Cu_3O_y$: $BaCuO_{2+x}$ and Y_2BaCuO_5 ," to be submitted to the *J. Am. Ceram. Soc.* Also, presented at the 44th Annual Calorimetry Conference, Oak Ridge, TN, July 30–Aug. 4, 1989.
62. T. B. Lindemer, "Chemical Thermodynamic Representation of Nonstoichiometry in $<UO_{2-x}N_{3x/4}>$," *CALPHAD* **13**, 109–13 (1989).
63. T. B. Lindemer, "Fuel and Fission-Product Reactions in HTGR Fuels," presented at the Savannah River Laboratory, April 17, 1989.
64. M. T. Harris, C. H. Byers, and R. R. Brunson, "A Study of Solvent Effects on the Synthesis of Pure Component and Composite Ceramic Powders by Metal Alkoxide Hydrolysis," *Mat. Res. Soc. Symp. Proc.*, **121**, 287 (1988).
65. C. H. Byers, M. T. Harris, and D. F. Williams, "Controlled Microcrystalline Growth Studies by Dynamic Laser-Light Scattering Methods," *I&EC Research*, **26**, 1916 (1987).
66. M. T. Harris and C. H. Byers, "Effect of Solvent on the Homogeneous Precipitation of Titania by Titanium Ethoxide Hydrolysis," *J. Noncryst. Solids*, **103**, 49 (1988).
67. C. H. Byers, R. R. Brunson, M. T. Harris, and D. F. Williams, *Controlled Nucleation and Growth Studies in Metal Oxide and Alkoxide Systems by Dynamic Laser-Light-Scattering Methods*, ORNL/TM-10285, Oak Ridge National Laboratory, April 1987.
68. M. T. Harris, C. H. Byers, and R. R. Brunson, "The Effects of Aluminum Alkoxides on the Synthesis of Composite Powders of Alumina and Titania," to be published in the 1898 Material Research Society Symposium Proceedings, Spring 1989 Meeting.
69. M. R. Harris, R. R. Brunson, C. H. Byers, "The Base Catalyzed Hydrolysis and Condensation Reactions of Dilute and Concentrated TEOS Solutions," to be published in *J. NonCryst. Solids*.
70. K. D. Keefer, "The Effects of Hydrolysis Conditions on the structure and Growth of Silicate Polymer," *Mat. Res. Soc. Symp. Proc.*, **32**, 15 (1984).

71. I. L. Thomas, "The Theory of Oriented Flocculation," *J. Colloid Interface Sci.*, **36**, 110 (1971).
72. W. Krahn, G. Schweiger, K. Lucas, "Light Scattering Measurements of Mutual Diffusion Coefficients in Binary Liquid Mixtures," *J. Phys. Chem.*, **87**, 4515 (1983).
73. G. W. Euliss, C. M. Sorensen, "Dynamic Light Scattering Studies of Concentration Fluctuations in Aqueous *t*-Butyl Alcohol Solution," *J. Chem. Phys.*, **80**, 4767 (1984).
74. M. Fleischmann, S. Pons, and M. Hawkins, "Electrochemically Induced Nuclear Fusion of Deuterium," *J. Electroanal. Chem.* **261**, 301-08 (1989).
75. S. E. Jones et al., "Observation of Cold Nuclear Fusion in Condensed Matter," *Nature* **338**, 737-40 (1989).
76. G. A. Wurden et al., "Electrochemical 'Cold Nuclear Fusion' Attempts at IPP," presented at Workshop on COLD FUSION Phenomena, Santa Fe, NM, May 23-25, 1989.
77. D. R. McCracken et al., "In Search of Nuclear Fusion in Electrolytic Cells and Metal/Gas Systems," presented at Workshop on COLD FUSION Phenomena, Santa Fe, NM, May 23-25, 1989.
78. G. Kreysa, G. Marx, and W. Plieth, "A Critical Analysis of Electrochemical Nuclear Fusion Experiments," submitted to *J. Electroanal. Chem.*, 1989.
79. J. F. Ziegler et al., "Electrochemical Experiments in Cold Nuclear Fusion," *Phys. Rev. Letters* **2**, 2929-32 (1989).
80. A. J. Appleby et al., "Evidence for Excess Heat Generation Rates During Electrolysis of D₂O in LiOD Using a Palladium Cathode—A Microcalorimetric Study," presented at Workshop on COLD FUSION Phenomena, Santa Fe, NM, May 23-25, 1989.
81. N. J. C. Packham et al., "Production of Tritium from D₂O Electrolysis at a Palladium Cathode," *J. Electroanal. Chem.* **270**, 451 (1989).
82. P. K. Iyengar, "Cold Fusion Results in BARC Experiments," presented at Fifth International Conference on Emerging Nuclear Energy Systems, Karlsruhe, Federal Republic of Germany, July 3-6, 1989.
83. C. D. Scott et al., "Preliminary Investigation of Possible Low-Temperature Fusion," *J. Fusion Energy* (in press).
84. C. D. Scott et al., *A Preliminary Investigation of Cold Fusion by Electrolysis of Heavy Water*, ORNL/TM-11322, Oak Ridge National Laboratory, November 1989.

6. NRC and EPRI Severe Accident Research Programs

The U.S. Nuclear Regulatory Commission (NRC) and the Electric Power Research Institute (EPRI) currently sponsor programs in Chem Tech related to the safety of light-water reactors (LWRs). These programs emphasize mechanisms and pathways by which fission products can be released to the environment from failed LWR fuel under severe core-melt conditions. The following subsections discuss programs involving investigation of fission product release from LWR fuel, the chemistry and transport of iodine in the reactor containment, and the behavior of aerosols in the reactor containment.

6.1 Fission Product Release from LWR Fuel

The release and behavior of fission products under severe LWR accident conditions are being studied experimentally to provide data needed for the analysis of reactor accidents. This work is sponsored by the NRC to measure the release rates and the chemical forms of the major fission products as functions of the principal variables — temperature, atmosphere (steam and/or hydrogen), fuel type, time, and fraction of fission products remaining in the fuel. Based on reviews by the NRC of the available data and of the needs for additional information, extensive work at ORNL has been conducted and published.¹⁻⁴ The most recent results of these studies will be discussed.

The tests have been conducted in a hot cell facility using an induction furnace capable of operation in steam and/or hydrogen at temperatures up to 2700 K. The released material was collected in three sequentially operated, parallel trains of fission product collectors; each collection train included a thermal gradient tube, graded filters, and both heated and cooled charcoal. Both pretest and posttest inventories of the gamma-emitting fission products in the fuel specimens were determined by gamma spectrometry, and the computer program ORIGEN2 was used to calculate the inventories of other nuclides.⁵ The accumulation of ⁸⁵Kr and ¹³⁷Cs released from the fuel was monitored at 1-min intervals during the test. The deposits on all apparatus components were measured after the test by various methods; direct gamma spectrometry of the long-lived gamma-emitting fission products was the most valuable, and other techniques (weighing of collectors; mass and emission spectrometry of

solid and solution samples; neutron activation analysis for I, Br, and Te; and scanning electron microscopic examination of deposits) provided supplementary data.

The fuel specimen history, test conditions, and release results for three recent tests are summarized in Table 6.1. These tests were conducted in helium-steam atmospheres at temperatures of 2000 to 2700 K for periods of 20 to 60 min at a particular test temperature. As shown in Fig. 6.1, the release rates of ⁸⁵Kr and ¹³⁴Cs as functions of time and temperature were similar throughout test VI-3, which is typical for these tests. (In all cases, ¹³⁴Cs and ¹³⁷Cs behaved identically.) In the temperature range of 2000 to 2300 K, total krypton and cesium

Table 6.1. Conditions and results for fission product release tests

Parameter	Test number		
	VI-1	VI-2	VI-3
Fuel specimen: ^a	Oco	BR3	BR3
Mass (fuel + clad), g	135	103	102
Burnup, MWd/kg	40	44	44
In-pile Kr release, %	0.7	-2	-2
Test conditions:			
Heatup rate, K/s	1	1	0.3
Test temperature, K	2020, 2300 ^b	2300	2000, 2700 ^b
Time at test temperature, min	20, 20	60	20, 20
Effective time at test temperature, min	8.9, 55.6	71.2	64.2, 47.3 ^c
Time at >2000 K, min	54	70	90
Steam flow, L/min	1.5	1.5	1.6
Fractional release, %			
⁸⁵ Kr	57	>31 ^d	>96
^{110m} Ag	33	8.9	
¹²⁵ Sb	28	68	>89
¹³⁷ Cs	65	63	99

^aOco = Oconee, BR3 = BR3 reactor in Belgium.

^bSome tests were conducted in two phases, at two different temperatures.

^cBecause of slow heatup and cooldown rates in test VI-3, the effective times at test temperature were unusually long.

^dIndicated Kr release in test VI-2 was low because of a loss of Kr during posttest transfer.

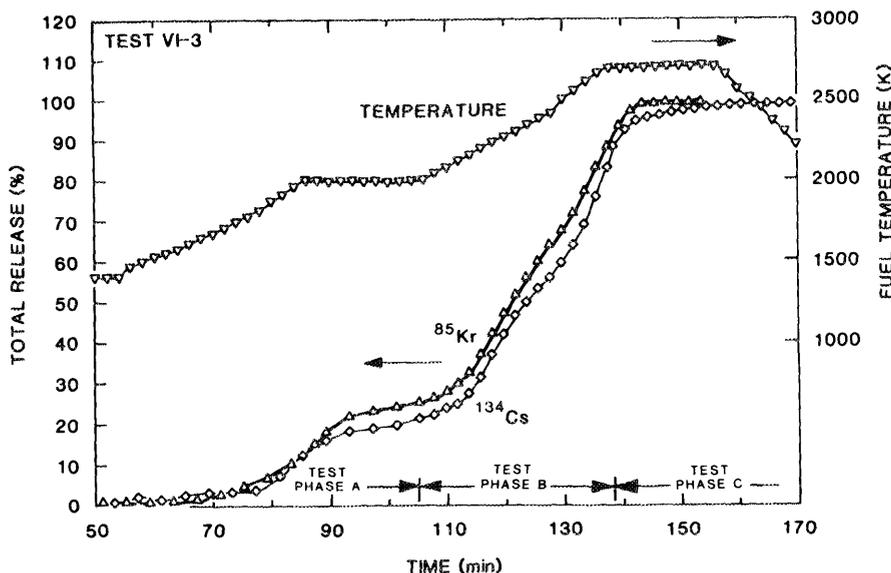


Fig. 6.1. Fission product release as a function of temperature and time in test VI-3.

releases have varied between 20 and 65%. At the highest temperatures (2700 K), essentially all of the fission product krypton and cesium were released within a few minutes, as indicated in Fig. 6.1. Other volatile fission products (I, Te, and Sb) have been shown to be released from the UO_2 at rates similar to, or slightly less than, those of krypton and cesium. Tellurium and antimony, however, tend to combine with metallic Zircaloy and to be retained until the Zircaloy cladding is almost completely oxidized, as occurs in a steam atmosphere.³ In some tests, large fractions (up to ~30%) of the $^{110\text{m}}\text{Ag}$ have been released and transported as aerosols. Predictably, bromine and rubidium have been shown to behave much like their respective chemical analogs iodine and cesium, which pose much greater safety hazards. Significant releases of fission product molybdenum and the cladding constituent, tin, have been measured also; the latter is significant primarily as an aerosol transport vehicle for the hazardous species. In addition, generally small releases of such fission product elements as Sr, Ru, Ba, and Eu have been detected in some tests.

Comparison of the release data from tests in steam vs tests in hydrogen has shown large differences for some elements. Whereas the release of Kr, I, and Cs is similar in both steam and hydrogen atmospheres, the release of many other fission product elements is strongly dependent on

atmosphere. As noted previously, antimony and tellurium are retained in the Zircaloy cladding (and possibly in other structural metals) as long as it remains metallic. Consequently, the release of antimony and tellurium from the fuel is delayed or prevented altogether under reducing conditions.

The total fractional release of cesium for our tests was compared with the CORSOR-M model, which is the current NRC standard, in Fig. 6.2.⁶ With one exception, all data from our tests (data points) fall significantly below the CORSOR-M curve, indicating that it is unduly conservative (high) for cesium. Except for very low temperatures (<1200 K), where krypton release may exceed cesium release, all results from our tests show almost identical behavior for krypton and cesium. Though fewer data are available for other volatile fission product elements, the comparisons indicate similar conclusions.

6.2 Containment Fission Product Behavior

Assessment of the chemistry and mass transport of iodine in containment during severe accidents in LWRs is an important component in determining the "source term" or fission product release from containment.

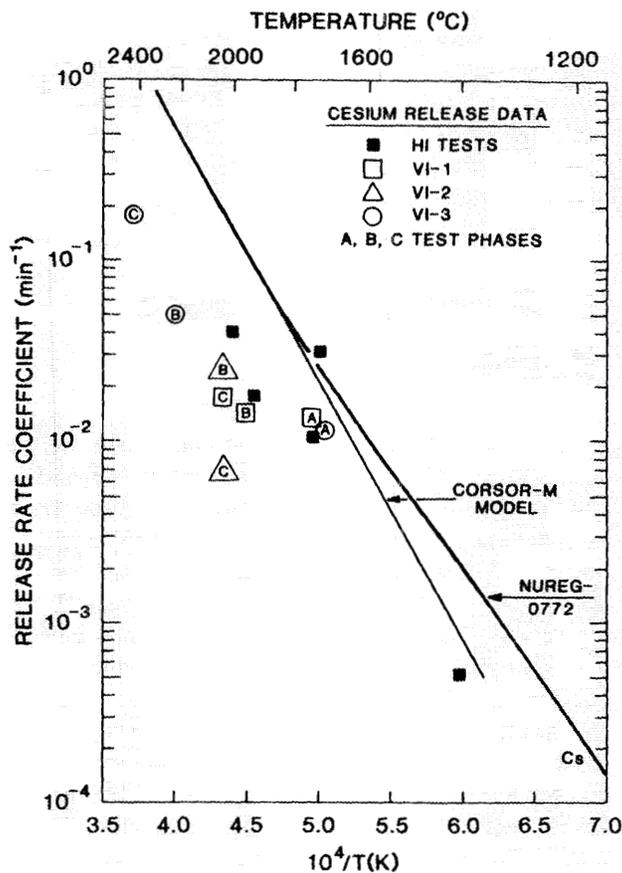


Fig. 6.2. Variation of measured cesium release rate coefficients with temperature, compared to CORSOR-M models.

The TRENDS code for calculation of iodine behavior and release in containment is a system of FORTRAN routines whose objective is the quantitative determination of fission product transport and retention characteristics during severe reactor accidents. It represents a best estimate of nuclide behavior under the hypothetical conditions of various severe accident sequences.

Every attempt has been made to account for all significant processes. Modeling of iodine hydrolysis has been carried out for two formations by a mathematical optimization procedure. In addition, an experimental effort has resulted in over 130 data points taken at a range of pHs between 2.8 and 9.0 and at temperatures of 25, 50, and 90°C. The optimization procedure is applied to yield rate constants at 25°C for both models and activation energies for the ten-step model. Using the optimized values, the rate equations were solved to obtain time-dependent iodine species inventories.

These calculated results compare favorably with actual data, indicating that the models are useful for simulating both forward (hydrolysis) and the reverse (Dushman) reaction.

Tables 6.2 and 6.3 give the reactions and rate constants for the six- and ten-step reaction models, respectively. Both models give a good representation of I₂ hydrolysis and its reverse; however, the ten-step reaction model gave a better approximation of the experimental data (i.e., a lower error residual). Furthermore, for calculation of radiation effects on iodine speciation, it may be essential to use the ten-step reaction model because it includes all oxidation numbers of iodine from -1 to +5.

Table 6.2. Six-step reaction model

Reaction	Optimized rate constants K(m ⁻¹ s ⁻¹) at 25°C
I ₂ + OH ⁻ → I ⁻ + HOI	7.072 × 10 ²
I ⁻ + HOI → I ₂ + OH ⁻	4.394 × 10 ^{4a}
2HOI ⇌ I ⁻ + HIO ₂ + H ⁺	2.408 × 10 ⁵ forward 3.086 × 10 ⁴ reverse ^a
HOI + HIO ₂ ⇌ I ⁻ + HIO ₃ + H ⁺	1.180 × 10 ⁴ forward 8.989 × 10 ⁶ reverse ^a

^aUnits are m⁻² s⁻¹.

Table 6.3. Ten-step reaction model

Reaction	Optimized rate constants K(m ⁻¹ s ⁻¹) at 25°C
I ₂ + OH ⁻ → I ⁻ + HOI	1.883 × 10 ³
2HOI → ½I ₂ + OI + H ₂ O	1.58 × 10 ²
2OI + H ₂ O → HOI + HIO ₂	1.62 × 10 ¹⁰
2HIO ₂ → OI + IO ₂ + H ₂ O	1.62 × 10 ¹⁰
2IO ₂ + OH ⁻ → HIO ₂ + IO ₃ ⁻	4.42 × 10 ^{10a}
H ⁺ + I ⁻ + HIO ₃ → ½I ₂ + IO ₂ + H ₂ O	7.61 × 10 ^{4a}
H ⁺ + I ⁻ + IO ₂ → ½I ₂ + HIO ₂	6.25 × 10 ^{3a}
H ⁺ + I ⁻ + HIO ₂ → ½I ₂ + OI + H ₂ O	1.81 × 10 ^{7a}
H ⁺ + I ⁻ + OI → ½I ₂ + HOI	1.81 × 10 ^{7a}
H ⁺ + I ⁻ + HOI → I ₂ + H ₂ O	5.75 × 10 ^{8a}

^aUnits are m⁻² s⁻¹.

6.3 ACE "Phase-B" Iodine Chemistry Research

The Advanced Containment Experiment (ACE) is a multiphase reactor safety experimental program investigating the anticipated chemistry and physics in a reactor containment in the course of a severe accident. A serious concern in severe accident analysis is the potential release of gaseous radioiodines to the atmosphere. To address this concern, "Phase B" of ACE will examine the retention of iodine species in the reactor containment system during postulated severe accidents. The effort at ORNL in support of the ACE Phase B Project consists of the following subtasks: (1) small-scale laboratory tests to investigate hygroscopic aerosol/iodine chemistry; (2) small-scale laboratory tests to investigate iodine chemistry issues associated with hydrogen burns; and (3) code-coordination activities associated with computer code modeling of the ACE Phase B intermediate-scale tests, to be performed at Whiteshell Nuclear Research Establishment (WNRE) in Canada, and the large-scale experiments to be performed at the Battelle Pacific Northwest Laboratory (PNL).

6.3.1 Aerosol Adsorption Studies

Tests of I_2 adsorption have been completed. The materials evaluated in the tests include Cs_2CO_3 , two-phased $Cs_2CO_3 \cdot (0.5H_2O + 1.5H_2O)$, Cs_2CO_3 + saturated solution, Cs_2O , $CsOH$ + saturated solution, $Cs_2B_4O_7$, HBO_2 , $CsNO_3$, and MnO . The temperature range of the tests was 30 to 100°C with iodine concentrations of $\sim 5 \times 10^{-8}$ to $\sim 1 \times 10^{-5}$ mol/L.

The tests showed that only basic powders (or solutions) reacted with the I_2 . This means that there was no reaction with $CsNO_3$, HBO_2 , or MnO . The reaction with basic materials was irreversible and formed iodide and iodate in a 5:1 ratio.

A test run with a mixed aerosol made up of Cs_2O , Sn, Cd, B_2O_3 , and In_2O_3 gave a result that was consistent with the Cs_2O content.

The rate of I_2 reaction with Cs_2CO_3 , two-phased $Cs_2CO_3 \cdot (0.5H_2O + 1.5H_2O)$, Cs_2CO_3 + saturated solution, and $CsOH$ + saturated solution was

limited only by the rate of gas-phase transport to the surface.

Tests of the interaction of organic iodides with the same aerosol materials have begun. Initial tests run with the adsorption of ethyl iodide on Cs_2CO_3 show that some I^- is formed on the powder. However, the extent of reaction is very small compared to I_2 adsorption.

6.3.2 Hydrogen Burn Studies

Oxidation of cesium iodide in a hydrogen flame occurs to a significant degree, producing molecular iodine and iodates in varying proportions. Oxidation in the bulk of the flame, the interconal region, which is >4 mm above the burner used in these tests, gave $\sim 30\%$ conversion of volatile CsI to I_2 , regardless of flame temperature or gas composition. Iodate production varied from 5 to 15%, with lower conversion in the leaner hydrogen flame mixtures. Oxidation of CsI closer to the leading edge of the flame in the initial reaction zone, <4 mm above this burner, gave considerably greater quantities of both I_2 and IO_3^- and greater oxidation ($I_2 = 30$ to 55%, $IO_3^- = 10$ to 20%). This is consistent with OH radical oxidation of CsI in the hydrogen flame.

Burning KIO_3 in a hydrogen flame redistributed the iodine species into $I_2 = 50\%$, $IO_3^- = 30\%$, and $I^- = 20\%$. The iodate decomposes more readily than the iodide and probably gave rise to much higher local concentrations of iodine atoms in the flame. This would account for the higher levels of I_2 .

Even higher local concentrations of iodine can be produced by inserting I_2 crystals into a hydrogen flame. Ninety percent of the recovered iodine is I_2 , 10% is iodate, and $<1\%$ is I^- . Once again, however, significant amounts of oxidation to iodate occur once the solid sample has been volatilized.

6.3.3 ACE Code-Comparison Coordination

The objective of this task is to coordinate iodine chemistry and transport code-comparison activities associated with the performance of the Radioiodine Test Facility (RTF) experiments to be done at

WNRE in Pinawa, Manitoba, Canada. In these tests, iodine species will be injected into a water pool within a 386-L vessel; the behavior of aqueous and gaseous iodine species will be measured. Test variables will include the iodine species injected into the pool, water-pool pH, and the radiation source to the water pool.

A series of pretest iodine code calculations were performed to model the RTF tests. The comprehensive results from these calculations have been summarized in a project report that is currently under review. Five iodine chemistry and transport codes were used in this study; these codes are listed in Table 6.4. A major difference in the codes was the approach taken to model iodine chemistry and speciation within the water pool.

Table 6.4. Summary of iodine codes used for the RTF pretest calculations

Code	Country using code
IMPAIR-2	Switzerland
INSPECT	United Kingdom
IODE	France
LIRIC	Canada
TRENDS	United States

Although the results from the calculations are still under evaluation, they illustrate large differences in calculated iodine behavior. The results also illustrate that water-pool evaporation effects and gas-and-liquid surface deposition modeling effects had an important influence on the calculated results.

6.4 LACE Aerosol Code-Comparison Studies

A significant area of uncertainty in the evaluation of radioactive releases from severe LWR accidents is the level of validation of the computer codes used to calculate aerosol releases from the containment building. The LWR Aerosol Containment Experiments (LACE) were performed

at the Westinghouse Hanford Engineering Development Laboratory to address this uncertainty. These tests investigated, on a large scale, the aerosol retention behavior in containment under simulated severe LWR accident conditions. In addition, a LACE aerosol code-comparison exercise was conducted, under ORNL/Chem Tech guidance, to provide information on the state of validity of the aerosol transport codes.

Pretest and blind posttest aerosol transport code calculations were performed — by analysts from 6 countries using 14 computer codes — to model results from LACE tests LA1, LA2, LA3, LA4, and LA6. The aerosol transport computer codes used and the countries that used them are listed in Table 6.5. Tests LA1 and LA3 investigated the transport of CsOH and MnO aerosols through a pipe (the pipe included six 90° bends) for flow velocities ranging from 20 to 200 m/s. Tests LA2, LA4, and LA6 investigated the behavior of CsOH and MnO aerosols injected into a 852-m³ test vessel under condensing steam-atmosphere conditions for varied aerosol leakage rates and times.

Numerous improvements to the aerosol-transport code models resulted from the code-comparison efforts. These include (1) improvements in code aerosol-deposition models, including models for turbulent aerosol deposition in pipe flows and aerosol deposition in bends; (2) the development of new models to treat the hygroscopic nature of CsOH aerosols in the presence of steam; (3) methods of eliminating "numerical deagglomeration,"

Table 6.5. Aerosol codes used in LACE code-comparison studies

Aerosol code	Countries using code
AEROSIM-M	United Kingdom
CONTAIN	United Kingdom, United States
HAA4	United States
MAAP.3	Sweden
MCT-2	United States
NAUA-4	United States
NAUA4-HYGROS	Finland, United States
NAUA-5	Finland, Italy
QUICK-M	United States
RAFT	United States
REMOVAL/2G	Japan
RETAIN-2C	Finland
SWNAUA-HYGROS	United States
TRAP-MELT2	Italy, Japan, United Kingdom, United States

or fictitious breakup of aerosols due to the numerical solution methods used in the codes; and (4) improved methods for calculating aerosol size-distribution parameters. In addition, the code analysts also improved their abilities to perform calculations using the available codes. Important

areas for further study that resulted from these efforts included (1) the need for improved multicomponent aerosol transport modeling and (2) the addition of aerosol resuspension models to the codes used to calculate aerosol transport in pipe flows.

References

1. R. A. Lorenz, J. L. Collins, and A. P. Malinauskas, "Fission Product Source Terms for the Light Water Reactor Loss-of-Coolant Accident," *Nucl. Technol.* **46**, 404-10 (1979).
2. M. F. Osborne, J. L. Collins, and R. A. Lorenz, "Experimental Studies of Fission Product Release from Commercial LWR Fuel Under Accident Conditions," *Nucl. Technol.* **78**(2), 157-69 (August 1987).
3. J. L. Collins, M. F. Osborne, and R. A. Lorenz, "Fission Product Tellurium Release Behavior Under Severe Light Water Reactor Accident Conditions," *Nucl. Technol.* **77**(1), 18-31 (April 1987).
4. J. L. Collins et al., "Fission Product Iodine and Cesium Release Behavior Under Severe LWR Accident Conditions," *Nucl. Technol.* **81**(10), 78-94 (1987).
5. A. G. Croff, *ORIGEN2 — A Revised and Updated Version of the Oak Ridge Isotope Generation and Depletion Code*, ORNL-5621, Oak Ridge National Laboratory, July 1980.
6. M. R. Kuhlman, D. J. Lehmicke, and R. O. Meyer, *CORSOR User's Manual*, NUREG/CR-4173 (BMI-2122), Battelle Memorial Institute, March 1985.

7. Office of Safety and Operational Readiness

The Chemical Technology Division Office of Safety and Operational Readiness (OSOR) was created in mid-FY 1989 to integrate the functions of quality assurance, safety, radiation control, environmental protection, hazards communication, training, crisis management, operational readiness, and the Comprehensive (and continuing) Self-Assessment and Upgrade Program (CSAUP) for the division. Interlaced throughout all of these functional areas are the management and information systems that serve to track the division's commitments and prioritized schedules for the action items; provide the reports and other communications tools; and build, maintain, and manipulate the data bases of information about the division and its facilities. OSOR will also serve as one repository for the documentation and records that are so important to the external reviews and audits. Each of these aspects will be described briefly in the following sections.

The office has a broader mission as well. It serves as the interface, under the guidance of the Division Director, for these functions between the division and other organizations at ORNL, within Martin Marietta Energy Systems, Inc., and at DOE. As an example of these less obvious functions, OSOR prepared the responses for the Laboratory to the findings and observations about hot cell facilities resulting from the DOE/ORO and DOE/HQ Technical Safety Appraisals (TSAs) (held in calendar year 1989). OSOR is working with the ORNL Quality Department to track each Chem Tech action item to closure.

Another mission of OSOR is the continuing, deliberate assessment of Chem Tech's performance, analysis of trends, and preparation of corrective actions to continually improve Chem Tech's safety, effectiveness, and efficiency in its operations.

At this time, OSOR is seeking to staff its permanent positions for safety and radiation control, environmental protection and hazards communication, and training and certification.

7.1 Health, Safety, and Environmental Protection

Chem Tech has received the highest rating ("superior") for safety in each of the last five consecutive years. This is the result of the dedication and safety consciousness of the staff. Section and division safety meetings are held regularly with good attendance. The division sessions are videotaped to allow shift workers and absent employees to make up any safety meetings that were missed. A significant amount of time has been invested by employees working on safety in Chem Tech (i.e., serving on safety committees, facility emergency squads, and safety inspection teams). Follow-up activities have addressed the significant safety issues. Furthermore, several special safety concerns were addressed: (1) an audit of welding and burning activities for safety, (2) an investigation and report on the safety of the isotopes area ventilation system, and (3) a study of asbestos fibers found in division areas.

Several reports were issued: "Safety Action Plan," "CTD Safety Report," and revised safety manuals for the Chemical Development and Isotopes sections. Several safety suggestions were submitted to the Industrial Safety Department by division employees. General safety indoctrination for new employees, both payroll and nonpayroll, was held. Other special safety and orientation training was held for employees needing these courses. And finally, the division has gone to special efforts in the Spring and Fall Cleanup and Fire Prevention campaigns. There were several indications that improvements are needed in Chem Tech's operations. Fortunately, most of these were items with safety implications rather than accidents or incidents. Many of the deficiencies stemmed from assessments of Chem Tech's operations using the new DOE technical safety requirements. They were brought out as findings from our own CSAUP (see Sect. 7.4) and numerous external reviews and assessments. On the negative side, there were ten Unusual Occurrence reports filed in this period and one Recordable Injury and Illness report; one

employee received the "Gold Buckle" award for wearing a seat belt in a motor vehicle accident. Each of these categories has provided the stimulus for self-improvement actions.

The newly formed OSOR is charged with reducing the frequency and the severity of such occurrences through (1) more reliance on procedures, (2) more vigor and formality in operations, (3) more visible management oversight and auditability, and (4) trend analysis. OSOR represents a major commitment of division resources to safety in Chem Tech.

7.2 Quality Assurance

The main thrust of the Chem Tech Division QA effort has been in converting the CTD QA Program to ANSI/ASME NQA-1 Standards. All QA plans have been rewritten to meet these requirements, and implementation is well under way. A vigorous QA surveillance program has been initiated to assist Chem Tech line managers with implementation of their new QA plans. Operating procedures are being rewritten to incorporate the systems defined in NQA-1, and this conversion is expected to be completed in FY 1990.

7.3 Training

7.3.1 Chem Tech Training Staff

The training staff in Chem Tech consists of four part-time and three full-time section training officers in five of the sections in the division and a part-time division training coordinator. This staff is required to know the current Laboratory training requirements as promulgated in a variety of sources, including ORNL procedural, industrial hygiene, health physics, and environmental protection manuals. At present, there is no central ORNL training function which coordinates all requirements. Chem Tech's training manual summarizes many of the requirements from these sources.

The division training staff members report to the various section heads. Not all sections have

training officers, and some training officers serve multiple sections. Communications of new training requirements flow through the division training coordinator to the officers.

Responsibilities of the training officers include (1) maintaining training records (including test scores) of operating staff in all experimental facilities, (2) scheduling training for new employees and retraining for existing employees, (3) conducting some training modules, and (4) arranging instruction by internal and external instructors.

Topics that are presented by internal instructors (either the training officers, facility supervisors, or other subject-matter experts) include general facility orientation and safety training for facility users and occupants, "dos and don'ts" (facility access requirements), local emergency procedures, waste handling operations for laboratories, evacuation procedures, location of emergency equipment, contamination control procedures, procedures for specific facility operations and maintenance, and Operational Safety Requirement (OSR) and Final Safety Analysis Report (FSAR) training both for Chem Tech operating and service staffs.

Topics that are presented to Chem Tech operating staff by external ORNL instructors include general employee training (GET), contact-handled transuranic (CH-TRU) waste handling, solid low-level waste handling, hazardous and mixed waste generator training, wastes governed by the Resource Conservation and Recovery Act (RCRA), cardiopulmonary resuscitation (CPR), ORNL respirator program, vacuum technology, X-ray training, Scott Air-Pak training, fire equipment training, and criticality safety training (basic and advanced).

Specific training for the Isotope Distribution Office is provided for its license verification program and for Department of Transportation (DOT) hazardous materials transportation.

The training staff has been involved in special activities, such as preparation of the restart training program for Bldg. 3517 (Fission Product Development Laboratory), conducting Training Accreditation Workshops and Instructors' Skills Workshops, and self-evaluation of training programs against Chem Tech's performance objectives and criteria (POC) and against the training accreditation criteria. An effort is now being made to recruit a Division Training Coordinator who will be assigned to OSOR.

7.3.2 Performance-Based Training and Training Accreditation

In anticipation of forthcoming DOE performance-based training requirements, ORNL moved (in April 1989) to centralize the Laboratory training functions in conjunction with similar Martin Marietta Energy Systems' actions. A Laboratory training manager was named and given the responsibility for ORNL's meeting the requirements of the order. A Training Accreditation Steering Committee (TASC) was formed, reporting to the Laboratory Director and consisting of members of the management of divisions which have training accreditation responsibilities in HFIR and REDC. Chem Tech is included on this committee.

An early responsibility of the ORNL training manager is to prepare a training manual that consolidates all of the Laboratory's training requirements into a single document. Chem Tech has a review function for this document.

7.4 Operational Readiness

The CSAUP has been patterned on a similar activity performed at HFIR. Using the Draft DOE POC for TSAs (May 1987)¹ as a starting point, the Chem Tech Issues Evaluation Committee (IEC) determined that 14 functional areas for evaluation, listed in the report, were suitable for Chem Tech use. Six additional functional areas were added for completeness, since Chem Tech has a broader set of missions than a reactor facility (Table 7.1).

The IEC modified the POC, for each functional area in the above-mentioned DOE TSA document, to reflect the characteristics of various Chem Tech operations. A policy statement was written to serve as an overall guide, and the performance objectives were written to give clear indication of the materials covered in each category. The new functional areas added by Chem Tech were derived in a similar fashion. For each objective, a set of criteria was derived to provide measures of how well the objectives were attained. Each POC was approved by an IEC composed of senior managers and representatives of safety and quality organizations in

Table 7.1. Safety-related functional areas specified in Chem Tech POC

Original functional areas and order

1. Organization and administration	(OA)
2. Operations ^a	(OP)
3. Maintenance	(MA)
4. Training and certification	(TC)
5. Auxiliary systems	(AX)
6. Emergency readiness	(ER)
7. Technical support	(TS)
8. Security/safety interface	(SS)
9. Experimental activities ^a	(EA)
10. Facility safety review	(FR)
11. Nuclear criticality safety	(CS)
12. Radiological protection	(RP)
13. Personnel protection	(PP)
14. Fire protection	(FP)

New functional areas

15. Transportation and packaging	(TP)
16. Configuration management	(CM)
17. Design adequacy	(DA)
18. Quality assurance	(QA)
19. Control and use of radioactive and hazardous products	(CU)
20. Environmental protection	(EP)

^aThese two were combined into operations and experimental activities for Chem Tech.

Chem Tech. This process is described in more detail in the Progress Report² for the period January 1 to June 30, 1989.

Following this approval process, an assessment was made, both internally and by external review teams, comparing Chem Tech's current practice with our objectives and policy statements. From these evaluations, a set of 242 action items was identified to bring Chem Tech's practice into line with the focus on continuing improvement. The action items are in the process of being prioritized by the IEC committee. A final round of approvals will be obtained at the division level prior to seeking funding and resources and scheduling and performing the corrective actions.

This procedure, based on the lessons learned in the nuclear industry, will enhance Chem Tech's operational performance in some important ways, while maintaining the special factors that have allowed the Chem Tech staff to be creative and successful in their R&D activities.

7.5 OSOR Management and Information Systems

7.5.1 Management Systems

Effort in the area of management systems has consisted of working with the IEC to define a set of procedures by which Chem Tech will do business. Several draft procedures have been prepared for review, and the committee has approved some procedures for interim use, but the overall structure of the system is not yet defined. It is anticipated that the definition of the system and preparation of division-level procedures will be major activities in the months to come.

Resource management for safety and operational readiness activities, which will be a major activity in this area, has just been initiated.

7.5.2 Information Systems

Significant progress has been made in the area of information management. A commitment tracking and reporting system is nearly complete. The mission is to inform division management of the status of action items, including unusual occurrence reports, quality event reports, quality incident reports, corrective action reports, responses to findings from spot inspections and technical safety appraisals (including Chem Tech's own CSAUP),

findings by the ORNL Radioactive Operations Committee, and Award Fee milestones. The data base for these items has been completed, and periodic reports will be issued to the Division Director, Section Heads, and safety and quality specialists, providing information about completed actions, actions due in the near future, and newly assigned actions.

Data bases are being prepared to assist in training, safety, and facility management. The Training Data Base (~35% complete) will provide the central training record for individuals in the division. It will be used to assist management in determining whether training requirements for work assignments are fulfilled. We expect it will also provide information about personnel for possible future assignments and career development. The Safety Data Base will assist in determining that personnel are receiving the mandatory safety training. The preliminary data have been input to the Facility Data Base and are out for comment. This initial data set includes information about each of the facilities and lists the responsible managers and owners. Eventually, the structure of this data base will be expanded to include information about special equipment or capabilities and the hazards associated with each facility.

The use of a badge reader for recording attendance at meetings is being investigated and could result in very significant savings with minimal investment. The major saving is expected to be realized in the time required for analyzing attendance records and crediting attendance to each individual. (This is now being done manually.)

References

1. U.S. Department of Energy, *Performance Objectives and Criteria* (Draft), Washington, DC, May 1987.
2. K. H. Lin, P. Standifer, and V. C. A. Vaughn, *Comprehensive Self-Assessment and Upgrade Program Progress Report for the Period January 1 to June 30, 1989*, ORNL/TM-11288/R1, November, 1989.

8. Administrative Resources and Facilities

Chem Tech's facilities span three plant sites in Oak Ridge and include numerous buildings as well as advanced equipment. Although some of these facilities have been improved as a result of capital projects, many remain in need of maintenance, upgrades, and renovations because these activities have not kept pace with normal deterioration and changes in regulating requirements and technological advances.

8.1 Facilities

Chem Tech, as one of the largest and most diverse divisions of ORNL, operates research programs in ~40 buildings. While most of these facilities consist of general-purpose laboratory and office space, some are unique or rare. The Radiochemical Engineering Development Center (REDC) contains some of the most advanced radiochemical hot cells capable of containing alpha activity while providing shielding from gamma rays.

The division operates two user facilities used for tests by scientists and engineers from other government, industrial, and academic organizations. In the Drop Test Facility, the ability of shielded shipment containers to survive transportation accidents can be tested, and in the Bioprocessing User Facility, a unique variety of bioprocessing equipment has been assembled.

Most of the buildings are more than 35 years old, and some of them have undergone one or more stages of modification or renovation. Some of the facilities no longer meet the needs of the division, and efforts are being made to decommission them or convert them to types needed for current programs. Others will need significant upgrades in order to comply with DOE Orders and the state and federal environmental rules and regulations.

Approximately half of the Chem Tech facilities are involved in the production and processing of radioisotopes. Several of these buildings are scattered around the ORNL site; most of them are located in the "Isotope Circle," which is in the center of the main ORNL plant. One of the major challenges facing Chem Tech in the foreseeable future will be to systematically evaluate the various facilities, identify their current and future uses, make short- and long-term plans for satisfactory resolution of individual facility shortfalls, seek appropriate funding sources, and implement these strategic plans for revitalizing or decommissioning the buildings. The overall goal is to consolidate operations into fewer buildings for a more efficient radiochemical operation that provides adequate

containment to protect both the environment and personnel.

8.2 Capital Projects

During the report period, Chem Tech participated in nine General Plant Projects (GPPs), which provide capital improvements or new additions to the division's physical facilities (Table 8.1). Chem Tech also participated in four line items affecting major projects in many of the operational areas and facilities in which the division has interest. The progress made on both of these types of capital projects advanced through various stages consistent with the engineering procedures by which the projects are conducted, varying from feasibility studies, conceptual design, systems requirements and scope, design criteria, and design on through various stages of construction. The amount of capital funds allocated for all the GPPs is ~\$7M. About \$350M in capital funds is allocated for the four line items currently affecting Chem Tech and requiring planning and operational information input from divisional personnel. Only a portion of those funds will be directly expended on Chem Tech facilities; the bulk of expenditures will be allocated for the three line items directed toward upgrading the low-level waste (LLW) collection and transport facilities at ORNL.

Those projects completed during this time period were accomplished safely; within budget; in accordance with all relevant environmental, health, and quality considerations and procedures; in agreement with initial conceptual designs and systems requirements documents; and, for the most part, in an acceptable time frame. Services and facilities to be provided by these capital projects include renovation and expansion of normal and emergency electrical circuitry, upgrade of ventilation systems and filtration systems, replacement of LLW drain systems, and upgrade of vital oil treatment systems (Table 8.1). These activities are being conducted in a variety of facilities, ranging from areas utilized for basic and applied research to hot

Table 8.1. Capital projects and line items for Chem Tech

Fiscal year	Project title	Cost of construction		Completion date	
		Estimate (\$10 ³)	Actual (\$10 ³)	Scheduled	Actual
Capital projects					
1988	Upgrade HVAC ^a and operational areas, Building 4501, high bay	850	In design	5-91	In progress
1987	Upgrade utilities, Laboratory 16, Building 4501	141	~ 141	12-87	8-89
1988	Upgrade HVAC, Building 9204-3	740	Ready for construction	5-91	In progress
1988	Upgrade east cells, Building 3525	1100	In design at 90%	3-91	In progress
1989	Upgrade north cells, Building 3525	1050	In design at 90%	3-91	In progress
1988	Upgrade cell off-gas filtration system, Building 3517	630	Construction at 90%	1-90	1-90 estimate
1989	Upgrade in-cell filtration system, cell D, Building 3047	135	On hold	On hold	
1988	Construct filter house over cell off-gas filter pit, Building 3019	200	On hold	On hold	
1990	Upgrade Z-oil line area of 9204-3	717		12-91	In progress
1990	Upgrade cells 13 and 15, Building 3517	546		5-92	In progress
Line Items					
1991	Bethel Valley area low-level liquid wastes (LLLW) collection and transport system upgrade	35	In progress	1994-95	In progress
1992	Isotopes area LLLW collection and transport system upgrade	41	In progress	1995-96	In progress
1992	Melton Valley LLLW collection and transport system upgrade	25	In progress	1995-96	In progress
1993	Waste Handling and Packaging Plant	242	In progress at 90%	1996-97	In progress

^aHVAC = heating, ventilating, and air conditioning.

cells in critical facilities, such as the Metallurgical Examination Hot Cells and the REDC. During the same time frame, the Central Engineering Organization continued its efforts to process and complete numerous work orders (~10 to 20) that required the work of various engineering disciplines (civil and structural, electrical, hydraulic, mechanical, theoretical stress and strain analysis, etc.) for expense-funded projects for Chem Tech, totaling between \$2M and \$4M (design plus construction).

Traditionally, Chem Tech has enjoyed a unique relationship with Central Engineering. For many

years, a staff engineer was assigned as a liaison to Central Engineering to directly represent Chem Tech or a project manager on whatever capital and expense projects were under way. This expertise has always been sought to ensure that the engineering projects undertaken by Chem Tech are conducted in an appropriate manner and that all activities such as estimates, conceptual design, design, design review, fabrication, procurement, installation, and construction are conducted according to relevant DOE and ORNL health, safety, environmental, and quality regulations and design criteria.

In 1988, Chem Tech absorbed the activities of the metallurgical examination and Isotopes Production Facilities of ORNL, thereby assuming responsibility for almost all hot cell operations at ORNL (except Building 2026 and a few other areas). These new responsibilities increased Chem Tech's number of capital and expense projects so that one liaison project manager could no longer manage the interactions with Central Engineering (the number of project engineers now assisting on Chem Tech projects is about ten). Nonetheless, in future work, Chem Tech intends to maintain the closest possible contact, cooperation, and communication with the project management of Central Engineering at ORNL. The responsibility for maintaining the traditional unique relationship will be vested in a new position of Engineering Project Coordinator.

Acquisition of expertise from Central Engineering becomes increasingly important in the contemporary atmosphere of "critical facilities" and the "new culture" in which engineering projects and facility operations must be conducted. In addition, all activities must be performed in compliance with increasingly stringent nuclear quality assurance regulations, contemporary configuration management directives, and current DOE engineering design criteria to avoid potential health, safety, or environmental insults.

8.3 Engineering Design and Facilities Development

Approximately 80% of this report period was dedicated to the Radiation Sterilization Inc. (RSI) Recovery Program, and the remaining effort was on facilities upgrade, operational support, and work for others.

The RSI effort spans three phases:

1. July 1988 through October 1988, inspection and leak testing of the cesium capsules at the Decatur, Georgia, facility.
2. July 1988 to mid-December 1988, assisting in the packaging and shipping of suspect and failed capsules.
3. January 1988 through mid-June 1989, assisting in the nondestructive examination of two

confirmed failed capsules and preparing for their destructive examination.

The inspection and testing phase consisted of designing and fabricating hardware, establishing procedures, and performing the test at the RSI facility. Two types of testing devices were used; a confined isolation of the most suspect capsules with sample monitoring of the container did not locate the failed capsule(s), so a second system added the capability of pressure cycling the capsule container as well as inverting and repeating the test. Two failed capsules were identified during this test.

Participation in the packaging and shipping involved procedures, redesign of the shipping overpack, and a system of drying the overpacks after loading in the RSI pool. Three shipments were made, with a total of three suspect capsules and two confirmed failures. The procedures and modified overpacks were supplied to RSI for use at their Ohio facility.

The nondestructive examination of the two failed capsules required many test and inspection fixtures. Fixtures for metrology, dye penetrant, visual, and pressure testing were developed and used. Assistance in operating special devices was furnished.

Tooling for the cutting at Building 3517 of the outer capsule, for inspecting and testing of the inner capsule, and segmenting and sampling of the salt was designed and tested, and the operators were trained. Shutdown of Building 3517 has this phase on hold.

Design reviews and conceptual designs were provided to the capital-funded facilities upgrade programs. Miscellaneous engineering support in maintenance and operational enhancement was provided to Buildings 3525 and 3517.

The design and installation procedures for a dosimetry experiment to be installed in the Portland General Electric Company Trojan Nuclear Plant was provided to the Nuclear Regulatory Commission Heavy Section Steel Technology Program. This experiment is to provide neutron fluence data near the reactor pressure vessel supports and is to be installed during April or May of 1990. Chem Tech assistance in the installation has been requested for this program that is managed by ORNL's Engineering Technology Division, with participation by the National Institute of Standards and Testing for the dosimetry and by the Computing and Telecommunications Division for the neutronics.

Administrative Summary

Publications and Oral Presentations

Aaron, W. S.

Aaron, W. S. *Report of Foreign Travel by W. S. Aaron, September 4-16, 1988*, ORNL/FTR-3052, September, 1988

Aaron, W. S.; Petek, M.; Zevenbergen, L. A.; Gibson, J. R. "Development and Preparation of Thin, Supported Targets and Stripper Foils," presented at 14th World Conference International Nuclear Target Development Society, Darmstadt, Federal Republic of Germany, September 5-9, 1988, and published in *Nucl. Instrum. Methods Phys. Res. A*, **282**, 147-52 (1989)

Kobisk, E. H.; Ramey, D. W.; Aaron, W. S.; Tompkins, J. A.; Haff, K. W.; DeVore, J. R.; Adair, H. L. "Tritium-Processing Operations at the Oak Ridge National Laboratory with Emphasis on Safe-Handling Practices," presented at Int. Nucl. Target Dev. Soc. and IAEA-INDC Conf., Darmstadt, Federal Republic of Germany, September 5-9, 1988, and published in *Nucl. Instrum. Methods Phys. Res. A* **282**, 329-40 (1989)

Adair, H. L.

Adair, H. L. "Possible Solutions for Difficult Accelerator Material and/or Target Requirements," presented at the Tenth Conference on the Application of Accelerators in Research and Industry, Denton, Texas, November 7-9, 1988, and published in *Application of Accelerators in Research and Industry '88*, Part II, J. L. Duggan and I. L. Morgan, eds., pp. 1132-1141, 1989

Kobisk, E. H.; Ramey, D. W.; Aaron, W. S.; Tompkins, J. A.; Haff, K. W.; DeVore, J. R.; Adair, H. L. "Tritium-Processing Operations at the Oak Ridge National Laboratory with Emphasis on Safe-Handling Practices," presented at Int. Nucl. Target Dev. Soc. and IAEA-INDC Conf., Darmstadt, Federal Republic of Germany, September 5-9, 1988, and published in *Nucl. Instrum. Methods Phys. Res. A* **282**(1) 329-40 (1989)

Alexander, C. W.

Bigelow, J. E.; Alexander, C. W.; Aramayo, G. A.; Carley, T. G.; Feldman, M. R.; Hammond, C. R.; Insalaco, J. W.; McCauley, V. S.; Theiss, T. J. *Safety Analysis Report for Packaging (SARP) for the Oak Ridge National Laboratory (ORNL) Californium Shipping Container*, ORNL/M-900, August 16, 1989

Macklin, R. L.; Alexander, C. W. *Neutron Absorption Cross Section of Uranium-236*, ORNL/TM-10999, November, 1988

Ashline, R. C.

Hermann, O. W.; Parks, C. V.; Renier, J. P.; Roddy, J. W.; Ashline, R. C.; Wilson, W. B.; LaBauve, R. J. *Multicode Comparison of Selected Source-Term Computer Codes*, ORNL/CSD/TM-251, April, 1989

Attaway, C. R.

Attaway, C. R. *Transportation Operations System Cask Maintenance Facility: Systems Requirements and Description*, ORNL/TM-10855, August, 1988

Basaran, O. A.

Perona, J. J.; Basaran, O. A.; Byers, C. H. "The Effect of Fluid Properties on Electric-Field-Enhanced Drop Formation," AICHE Annual Meeting, Washington, DC, December 1, 1988

Basaran, O. A. "Equilibrium Shapes and Stability of Pendant and Sessile Drops in an Electric Field," presented at 3rd Int. Colloq. on Drops and Bubbles, Monterey, CA, September 18-21, 1988

Basaran, O. A.; Harris, M. T. "Asymptotic Analysis of Performance of Membrane Modules for Gas Separations," submitted to *Sep. Sci. Technol.*

Basaran, O. A.; Auvil, S. R. "Asymptotic Analysis of Gas Separation by a Membrane Module," *AICHE J.*, **34**, 1726, 1988

Basaran, O. A.; Burban, P. M.; Auvil, S. R. "Facilitated Transport with Unequal Carrier and Complex Diffusivities," *I&EC Res.* **28**, 108, 1989

Basaran, O. A.; Scriven, L. E. "Axisymmetric Shapes and Stability of Isolated Charged Drops," *Phys. Fluids A*, **1**, 795, 1989

Basaran, O. A.; Scriven, L. E. "Axisymmetric Shapes and Stability of Charged Drops in an External Electric Field," *Phys. Fluids A*, **1**, 799, 1989

Basaran, O. A.; Scott, T. C.; Byers, C. H. "Theoretical and Experimental Analysis of the Oscillations of a Fluid Drop Immersed in Another Fluid," presented at Am. Inst. Chem. Eng. Meet. on Fundamental Research in Viscous Flows Session, Washington, DC, November 27-December 2, 1988

Basaran, O. A.; Scott, T. C.; Byers, C. H. "Drop Oscillations in Liquid-Liquid Systems," presented at 3rd Int. Conf. on Separations Technology, Davos, Switzerland, May 14-19, 1989, and published in *AICHE J.* **35**, 1263-70 (1989)

Basaran, O. A.; Scott, T. C.; Kim, S. "Drop-Drop Interactions and Coalescence in an Electric Field," presented at 3rd Int. Conf. on Separations Technology, Davos, Switzerland, May 14-19, 1989

Basaran, O. A.; Scriven, L. E. "The Taylor Pump: Viscous-Free Surface Flow Driven by Electric Shear Stress," *Chem. Eng. Commun.* **67**, 259-73 (1988)

Scott, T. C.; Basaran, O. A. "Effects of Interfacial Distribution Behavior and Droplet-Continuum Hydrodynamics on Transient Separations in Liquid-Liquid Solvent Extraction," presented at 3rd Int. Conf. on Separations Technology, Davos, Switzerland, May 14-19, 1989

Beahm, E. C.

Beahm, E. C. *Actinide Thermophysical Properties: A Preliminary Assessment*, ORNL/M-729, March, 1989

Beahm, E. C.; Brown, M. L.; Shockley, W. E. "ACE Phase B: ORNL Aerosol and Hydrogen Burn Studies," presented at Advanced Containment Experiment Technical Advisory Committee Review, ANL, Argonne, IL, May 10, 1989

Beahm, E. C.; Daish, S. R.; Shockley, W. E.; Hopenfeld, J. "Iodine Partitioning in PWR Steam Generator Accidents," submitted to *Nucl. Technol.*

Kress, T. S.; Beahm, E. C.; Wright, A. L. "Containment Transport Behavior," presented at ICHMT Semin. on Fission Product Transport Processes in Reactor Accidents, Dubrovnik, Yugoslavia, May 22-26, 1989

Beahm, E. C.; Weber, C. F.; Kress, T. S.; Shockley, W. E.; Daish, S. R. "Chemistry and Mass Transport of Iodine in Containment," in *Proc. 2nd CSNI Workshop on Iodine Chemistry in Reactor Safety*, Toronto, Canada, June 2-3, 1988 (1989), pp. 251-66

Daish, S. R.; Beahm, E. C.; Shockley, W. E. "Aqueous Iodine at Low Concentrations and High Temperatures," in *Proc. 2nd Workshop on Chemistry of Iodine in Reactor Safety*, Toronto, June 2-3, 1988 (1989), pp. 74-94

Kress, T. S.; Beahm, E. C.; Shockley, W. C.; Weber, C. F. "Trends Status," presented at Severe Accident Research Partners Meet., Washington, DC, October 17-21, 1988

Weber, C. F.; Beahm, E. C.; Kress, T. S.; Daish, S. R.; Shockley, W. E. "TRENDS: A Code for Modeling Iodine Behavior in Containment During Severe Accidents," presented at ICHMT Int. Semin. on Heat and Mass Transfer Aspects of Fission Product Releases, Dubrovnik, Yugoslavia, May 22-26, 1989, and to be published in *Proc. ICHMT Int. Semin. on Heat and Mass Transfer Aspects of Fission Product Releases*, Dubrovnik, Yugoslavia, May 22-26, 1989

Wright, A. L.; Fish, B.; Beahm, E. C.; Weber, C. F. "The Chemistry and Behavior of Iodine-Vapor Species in Nuclear Plant Air-Monitoring Sampling Lines," presented at 20th DOE/NRC Nuclear Air Cleaning Conf., Boston, August 22-25, 1988, and to be published in *Proc. 20th DOE/NRC Nuclear Air Cleaning Conf.*, Boston, Aug. 22-25, 1988

Beaver, J. E.

Cochran, D. K.; Beaver, J. E. "Production of Thallium-201 for Nuclear Medicine Cardiac Scans," a poster for use in the Isotope Program

Begovich, J. M.

Robinson, S. M.; Begovich, J. M.; Scott, C. B. "Low-Activity-Level Process Wastewaters: Treatment by Chemical Precipitation and Ion Exchange," *Water Air Soil Pollut.* **60**, 2120-27 (1988)

Sisson, W. G.; Begovich, J. M.; Byers, C. H.; Scott, C. D. "Continuous Chromatography," *CHEMTECH* **18**, 498-502 (1988)

Bell, J. T.

Bell, J. T.; Watson, J. S. "Fifth Symposium on Separation Science and Technology for Energy Applications," *Sep. Sci. Technol.*, **23** (12, 13) (1988)

Toth, L. M.; Bell, J. T.; Friedman, H. A. "The Disproportionation of Pu⁴⁺ in Nitric Acid Solutions," submitted to *Radiochim. Acta*

Berry, J. B.

Berry, J. B.; Homan, F. J. "Changing Generators for Waste Management Costs," presented at SPECTRUM '88, Nuclear and Hazardous Waste Management International Topical Meeting, Washington, D.C., September 11-15, 1988

Brown, C. H., Jr.; Berry, J. B.; Turner, D. W. "An Overview of the ORNL Waste Handling and Packaging Plant, a Major Processing Facility for Remote-Handled Transuranic Waste," in *Proc. Waste Management '89*, Tucson, AZ, February 26-March 2, 1989, Library of Congress No. 86-644175, 1989, v. 1, pp. 217-27

McNeese, L. E.; Berry, J. B.; Butterworth, G. E., III; Collins, E. D.; Monk, T. H.; Patton, B. D.; Snider, J. W. *Overall Strategy and Program Plan for Management of Radioactively Contaminated Liquid Wastes and Transuranic Sludges at the Oak Ridge National Laboratory*, ORNL/TM-10757, December, 1988

White, T. L.; Berry, J. B. "Microwave Processing of Radioactive Materials," presented at IAEA Division Winter Symp. Am. Chem. Soc., Atlanta, May 1-4, 1989, and to be published in *Proc. IAEA Division Winter Symp. Am. Chem. Soc.*, Atlanta, May 1-4, 1989; also in *Proc. 197th Am. Chem. Soc. Symp. on Innovative Waste Management Technologies*, Dallas, April 9-14, 1989 (1989), v. 29, pp. 127-34

Berry, J. B.; Robinson, S. M. "Improved Treatment of Slightly Radioactive Process Waste: Reduced Generation of Waste," presented at Spectrum '88, Nuclear and Hazardous Waste Management Int. Top. Meet., Pasco, WA, September 11-15, 1988, and published in *Proceedings of the Ninth Annual DOE Low-Level Waste Management Conference*, vol. 6, pp. 36-43, 1988

Berry, J. B.; Brown, C. H. Jr.; Fowler, V. L.; Robinson, S. M. "Treatment of ORNL Liquid Low-Level Waste," presented to the Division of Environmental Chemistry, American Chemical Society, Toronto, Canada, June 5-11, 1988

Bigelow, J. E.

Bigelow, J. E.; Alexander, C. W.; Aramayo, G. A.; Carley, T. G.; Feldman, M. R.; Hammond, C. R.; Insalaco, J. W.; McCauley, V. S.; Theiss, T. J. *Safety Analysis Report for Packaging (SARP) for the Oak Ridge National Laboratory (ORNL) Californium Shipping Container*, ORNL/M-900, August 16, 1989

Bigelow, J. E.; Cagle, E. B.; Knauer, J. B. "Fabrication of 50-mg ²⁵²Cf Neutron Sources for the FDA Activation Analysis Facility," *J. Radioanal. Nucl. Chem.*, Articles, 123 (2) 471-89 (1988)

Collins, E. D.; Bigelow, J. E.; Ottinger, C. L.; Burney, G. A. "Recovery of Selected Elements from Irradiated Targets," to be published in *Nuclear Waste Management Technology*, vol. 2, Harwood Academic Publishers

Blankinship, S. L.

Scott, C. D.; Woodward, C. A.; Thompson, J. E.; Blankinship, S. L. "Coal Solubilization by Enhanced Enzyme Activity in Organic Solvents," presented at 11th Symp. on Biotechnology for Fuels and Chemicals, Colorado Springs, CO, May 8-12, 1989, and published in *Appl. Biochem. Biotechnol.*

Bolla, L. L.

Donaldson, T. L.; Strandberg, G. W.; Bolla, L. L. "Bioremediation of TCE-Contaminated Groundwater," presented at Meet. American Inst. of Chemical Engineers, Houston, April 2-6, 1989, and to be published in *Proc. Meet. American Inst. of Chemical Engineers*, Houston, Apr. 2-6, 1989

Pitt, W. W.; Strandberg, G. W.; Donaldson, T. L.; Bolla, L. L. "Biodegradation of Trichloroethylene Contaminated Groundwater," presented at Conf. on Biotechnology Applications in Hazardous Waste Treatment, Longboat Key, FL, October 30-November 4, 1988, and to be published in *Proc. Conf. on Biotechnology Applications in Hazardous Waste Treatment*, Longboat Key, FL, Oct.30-Nov.4, 1988

Strandberg, G. W.; Donaldson, T. L.; Bolla, L. L.; Palumbo, A. V.; Eng, W. "Degradation of Trichloroethylene and *Trans*-1,2-Dichloroethylene by a Methanotrophic Consortia in a Trickle-Type Bioreactor," to be published in *Proc. DOE Model Conf.*, Oak Ridge, TN, Oct. 3-7, 1988

Bond, W. D.

Bond, W. D.; Becher, P. F. "Synthesis of Alumina-Zirconia Powders by Sol-Gel Processing," *Ultrastructure Processing of Advanced Ceramics*, John Wiley and Sons, 1988, pp. 443-52

Bond, W. D. "The Thorex Process," *The Science and Technology of Tributyl Phosphate*, vol. 3, CRC Press, 1990, pp. 225-247

Box, W. D.

Collins, E. D.; Box, W. D.; Godbee, H. W.; Scott, T. C. "Analysis of Data from Leaching Concrete Samples Taken from the TMI-2 Reactor Building Basement," presented at Am. Nucl. Soc. Top. Meet. on TMI-2 Accident Materials Behavior and Plant Recovery Technology, Washington, DC, October 31-November 4, 1988, published in *Trans. Am. Nucl. Soc.* **57**, 525-27 (1988), and to be published in *Nucl. Technol.*

Brown, C. H., Jr.

Berry, J. B.; Brown, C. H., Jr.; Fowler, V. L.; Robinson, S. M. "Treatment of ORNL Process Waste," presented at Spectrum '88, Int. Top. Meet. on Nuclear and Hazardous Waste Management, Pasco, WA, September 11-15, 1988, and published in *Proc. Spectrum '88*, 1988, pp. 416-20

Brown, C. H., Jr.; Berry, J. B.; Turner, D. W. "An Overview of the ORNL Waste Handling and Packaging Plant, a Major Processing Facility for Remote-Handled Transuranic Waste," presented at Waste Management '89, Tucson, AZ, February 26-March 2, 1989; in *Proc. Waste Management '89*, Tucson, AZ, February 26-March 2; and published in 1989, Library of Congress No. 86-644175, 1989, v. 1, pp. 217-27

Berry, J. B.; Brown, C. H. Jr.; Fowler, V. L.; Robinson, S. M. "Treatment of ORNL Liquid Low-Level Waste," presented to the Division of Environmental Chemistry, American Chemical Society, Toronto, Canada, June 5-11, 1988

Hall, R.; Brown, C. H., Jr.; Robinson, S. M.; Hewitt, J. D. "Operation of a Mobile Pilot-Scale Continuous Countercurrent Ion Exchange System for Treatment of Low-Level Radioactive Wastewater," presented at HAZTECH Int. '89 Conf., San Francisco, September 26-October 1, 1989, and published in *Proc. HAZTECH Int. '89 Conf.*, 1989, pp. 91-102

Walker, J. F.; Brown, C. H., Jr.; Wilson, J. H. "Treatment of Chromium-Contaminated Plating Shop Rinsewater Streams by Reverse Osmosis," presented at Water Pollution Control Federation Conf., Dallas, October 2-6, 1988, and published in *Proc. of the Industrial Waste Symp.*, 61st Annu. WPCF Conf., Dallas, October 3-6, 1988, Water Pollution Control Federation, 1988

Brown, M. L.

Beahm, E. C.; Brown, M. L.; Shockley, W. E. "ACE Phase B: ORNL Aerosol and Hydrogen Burn Studies," presented at Advanced Containment Experiment Technical Advisory Committee Review, ANL, Argonne, IL, May 10, 1989

Brunson, R. R.

Brunson, R. R.; Byers, C. H. "Viscosities of Alcohol-Hydrocarbon Systems in the Critical Region: A Dynamic Laser Light Scattering Approach," *J. Chem. Eng. Data* **34**, 46-52 (1989)

Harris, M. T.; Byers, C. H.; Brunson, R. R. "The Effects of Aluminum Alkoxides on the Synthesis of Composite Powders of Alumina and Titania," presented at Mater. Res. Soc. Symp. on Processing Science of Advanced Ceramics, San Diego, April 24-29, 1989, and to be published in *Proc. Mater. Res. Soc. Symp. on Processing Science of Advanced Ceramics*, San Diego, Apr. 24-29, 1989

Harris, M. T.; Byers, C. H.; Brunson, R. R. "The Application of Laser-Light-Scattering Techniques in the Study of Ceramic Powders Synthesis by the Hydrolysis of Metal Alkoxides," presented at Workshop on Advanced Laser Technology for Chemical Measurements, Argonne, IL, May 9-11, 1989, and published in *Proc. Workshop on Advanced Laser Technology for Chemical Measurements*, Argonne, IL, May 9-11, 1989 (1989), pp. 82-86

Harris, M. T.; Byers, C. H.; Brunson, R. R. "The Base-Catalyzed Hydrolysis and Condensation Reactions of Dilute and Concentrated TEOS Solutions," presented at 5th Int. Workshop on Glasses and Ceramics from Gels, Rio de Janeiro, August 6-10, 1989; to be published in *Proc. 5th Int. Workshop on Glasses and Ceramics from Gels*; and submitted to *J. Non-Cryst. Solids*

Harris, M. T.; Byers, C. H.; Brunson, R. R. "A Study of Solvent Effects on the Synthesis of Pure Component and Composite Ceramic Powders by Metal Alkoxide Hydrolysis," in *Proc. Spring Meet. Mater. Res. Soc.*, Reno, NV, April 5-9, 1988, Mater. Res. Soc., 1988, v. 121, pp. 287-92

Burtis, C. A.

Adams, S. M.; Beauchamp, J. J.; Burtis, C. A. "A Multivariate Approach for Evaluating the Integrated Response of Fish to Chronic Pollutant Stress," *Mar. Environ. Res.* **24**, 223-26 (1988)

Burtis, C. A. "Recent Technological Developments and Their Impact on Laboratory Automation," to be published in *Proc. 3rd Int. Congress on Automation and New Technology in the Clinical Laboratory*, Kobe, Japan, Sept. 5-8, 1988

Burtis, C. A. "Introduction—Robotics Symposium," presented at 3rd Int. Congress on Automation and New Technology in the Clinical Laboratory, Kobe, Japan, September 5-8, 1988

Burtis, C. A. "Recent Technological Developments and Their Impact on Laboratory Automation," presented at 3rd Int. Congress on Automation and New Technology in the Clinical Laboratory, Kobe, Japan, September 5-8, 1988

Burtis, C. A. "Advanced Technology and Its Impact on the Clinical Laboratory," presented at Joint Meeting AACC-Ohio Valley Section/Central Indiana Clinical Biochemistry Forum, Indianapolis, October 22, 1988

Burtis, C. A.; Painter, P. C. "Analytical Systems for the Clinical Laboratory, Part 1," *Am. Clin. Lab.* **8**, 14-19 (1989)

Burtis, C. A.; Painter, P. C. "Analytical Systems for the Clinical Laboratory," *American Clinical Laboratory* **8**, 43-49 (1989)

Haeckel, R.; Geary, T. D.; Burtis, C. A. "The Term "Random Access" is Inappropriate as a Descriptor for Clinical-Analysis Systems—Letter to the Editor," *Clin. Chem.* **34**, 1520 (1988)

Butterworth, G. E., III

Butterworth, G. E.; Morris, M. I.; Lee, D. W. *Radioactive Waste Management Implementation Plan for Paducah Gaseous Diffusion Plant*, ES/ESH-9, KY/H-86, May, 1989

Butterworth, G. E.; Morris, M. I.; Lee, D. W. *Radioactive Waste Management Implementation Plan for Portsmouth Gaseous Diffusion Plant*, ES/ESH-10, POE F-2011, May, 1989

Butterworth, G. E., III "Overview of the Low-Level Waste Disposal Development and Demonstration Program and the Experiences Associated with Its Development and Implementation," presented at Discussion of Low-Level Waste Disposal Development and Demonstration Activities with Westinghouse, Hanford, WA, May 17, 1989

Butterworth, G. E., III; Lee, D. W. "Pathways Analysis as a Management Tool for Low-Level Radioactive Waste Programs," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988

McNeese, L. E.; Berry, J. B.; Butterworth, G. E., III; Collins, E. D.; Monk, T. H.; Patton, B. D.; Snider, J. W. *Overall Strategy and Program Plan for Management of Radioactively Contaminated Liquid Wastes and Transuranic Sludges at the Oak Ridge National Laboratory*, ORNL/TM-10757, December, 1988

Rivera, A. L.; Butterworth, G. E., III "Overview of the Waste Facility Performance Assessment Model (WFPAM) Project for the Low-Level Waste Disposal Development and Demonstration (LLWDDD) Program— Systems Analysis Framework for Model Development and Refinement," presented at 11th Annu. DOE Low-Level Waste Conf., Pittsburgh, August 22-24, 1989

Byers, C. H.

Byers, C. H.; Watson, J. S. "Effects of Particle Shape and Size Distribution on Sorption and Flow Performance in Electrically Stabilized Expanded Beds," *Sep. Sci. Technol.* **23**, 1869 (1988)

Perona, J. J.; Byers, C. H. "Drop Formation from an Orifice in an Electric Field," *AIChE J.* **34**, 1577 (1988)

Holmes, J. M.; Begovich, J. M.; Byers, C. H. *An Economic and Energy Evaluation of the Replacement of Conventional Technology with Continuous Chromatography in the Dump Leaching of Copper Ores*, DOE/OR/21400-T357 (DE89009038), February, 1989

Tawiguchi, V. T.; Byers, C. H. "Large Scale Chromatographic Separations Using Continuous Displacement Chromatography (CDC)," *Rare Earths, Extractions, Preparation and Applications*, R. G. Bautista and M. M. Wong, Minerals, Metals, and Materials Soc. (1988)

Holmes, J. M. and Byers, C. H. *An Economic and Energy Evaluation of the Replacement of Conventional Technology with Continuous Chromatography in the Production of High-Fructose Sugar*, DOE/OR/21400-T352 (DE89010393), October 1988

Byers, C. H.; Scott, C. D.; Carta, J. G. "Recent Research in Continuous Chromatography," presented at the AIChE Annual Meeting, San Francisco, California, 1989

Scott, T. C.; Byers, C. H. "A Model for Mass Transfer in Oscillating-Circulating Liquid Drops," *Chem. Eng. Commun.* **77**, 67 (1989)

Byers, T. C.; Scott, T. C. "An Electric-Field Induced Dispersion and Coalescence Technique in Multiphase Liquid Systems," *The Liquid State and Its Electrical Properties*, E. E. Kunhardt et al., eds., NATO ASI Series: Physics 193, Plenum Press, New York (1988)

Byers, C. H. "An Aspen Simulation of Fuel Production by Hydrolysis of Woody Biomass," *Appl. Biochem. Biotech.* **18**, 143 (1988)

Lee, D. D.; Bienkowski, P.; Byers, C. H. "Evaluation of Separation and Purification Processes in the Antibiotic Industry," *Appl. Biochem. Biotech.* **18**, 261 (1988)

Lee, D. D.; Bienkowski, P.; Byers, C. H. *Expedient Antibiotics Production*, ORNL/TM-6355, June, 1988

Morris, J. S.; Byers, C. H. *Critical-Region Vapor-Liquid Equilibrium of the CH₄-CO₂-H₂S System*, ORNL/TM-10806, 1989

Basaran, O. A.; Scott, T. C.; Byers, C. H. "Theoretical and Experimental Analysis of the Oscillations of a Fluid Drop Immersed in Another Fluid," presented at Am. Inst. Chem. Eng. Meet. on Fundamental Research in Viscous Flows Session, Washington, DC, November 27-December 2, 1988

Perona, J. J.; Basaran, O. A.; Byers, C. H. "The Effect of Fluid Properties on Electric-Field-Enhanced Drop Formation," AIChE Annual Meeting, Washington, DC, December 1, 1988

Basaran, O. A.; Scott, T. C.; Byers, C. H. "Drop Oscillations in Liquid-Liquid Systems," presented at 3rd Int. Conf. on Separations Technology, Davos, Switzerland, May 14-19, 1989, and published in *AIChE J.* **35**, 1263-70 (1989)

Brunson, R. R.; Byers, C. H. "Viscosities of Alcohol-Hydrocarbon Systems in the Critical Region: A Dynamic Laser Light Scattering Approach," *J. Chem. Eng. Data* **34**, 46-52 (1989)

Byers, C. H.; DeCarli, J. P.; Carta, G. "Continuous Gradient Elution Chromatography: Separation of Metals by Cationic Exchange," to be published in *Natl. Meet. Am. Chem. Soc., Toronto, Ontario, Canada, June 5-11, 1988*

Byers, C. H.; Harris, M. T. "A Microcrystalline Growth Study of Solvent Effects in Alkoxides by Dynamic Laser Light Scattering," *Ultrastructure Processing of Advanced Ceramics*, Wiley Interscience, 1988, pp. 843-54

Byers, C. H.; Perona, J. J. "Drop Formation from an Orifice in an Electric Field," *AIChE J.* **34**, 1577-80 (1988)

Byers, C. H.; Sisson, W. G.; DeCarli, J. P., II; Carta, G. "Sugar Separations on a Pilot Scale by Continuous Ion-Exchange Chromatography," presented at Annu. Meet. Am. Inst. Chem. Eng., Washington, DC, November 27-December 2, 1988, and submitted to *Biotechnol. Prog.*

Byers, C. H.; Sisson, W. G.; DeCarli, J. P., II; Carta, G. "Pilot-Scale Studies of Sugar Separations by Continuous Chromatography," *Appl. Biochem. Biotechnol.* **20/21**, 635-54 (1988)

Byers, C. H.; Sisson, W. G.; DeCarli, J. P., II "The Use of Gradient Elution in Optimizing Continuous Annular Ion Exchange Chromatography with Applications to Metal Separations," presented at IEX'88,

Cambridge, Great Britain, July 17-22, 1988, and published in *Ion Exchange for Industry, Proc. IEX '88*, 1989, pp. 424-42

DeCarli, J. P., II; Carta, G.; Byers, C. H. "The Separation of Amino Acids by Continuous Displacement Chromatography," presented at 6th Int. Symp. on Preparative Chromatography, Washington, DC, May 8-10, 1989

Harris, M. T.; Byers, C. H. "The Effect of Solvent on the Homogeneous Precipitation of Titania by Titanium Ethoxide Hydrolysis," *J. Non-Cryst. Solids* **103**, 49-64 (1988)

Harris, M. T.; Byers, C. H. *An Orthogonal Collocation Approach to Modeling Multicomponent Adsorption in Carbon Beds*, ORNL/TM-10735, May, 1989

Harris, M. T.; Byers, C. H.; Brunson, R. R. "The Effects of Aluminum Alkoxides on the Synthesis of Composite Powders of Alumina and Titania," presented at Mater. Res. Soc. Symp. on Processing Science of Advanced Ceramics, San Diego, April 24-29, 1989, and to be published in *Proc. Mater. Res. Soc. Symp. on Processing Science of Advanced Ceramics*, San Diego, Apr. 24-29, 1989

Harris, M. T.; Byers, C. H.; Brunson, R. R. "The Application of Laser-Light-Scattering Techniques in the Study of Ceramic Powders Synthesis by the Hydrolysis of Metal Alkoxides," presented at Workshop on Advanced Laser Technology for Chemical Measurements, Argonne, IL, May 9-11, 1989, and published in *Proc. Workshop on Advanced Laser Technology for Chemical Measurements*, 1989, pp. 82-86

Harris, M. T.; Byers, C. H.; Brunson, R. R. "The Base-Catalyzed Hydrolysis and Condensation Reactions of Dilute and Concentrated TEOS Solutions," presented at 5th Int. Workshop on Glasses and Ceramics from Gels, Rio de Janeiro, August 6-10, 1989

Harris, M. T.; Byers, C. H.; Brunson, R. R. "A Study of Solvent Effects on the Synthesis of Pure Component and Composite Ceramic Powders by Metal Alkoxide Hydrolysis," in *Proc. Spring Meet. Mater. Res. Soc.*, Reno, NV, April 5-9, 1988, Mater. Res. Soc., 1988, v. 121, pp. 287-92

Harris, M. T.; Scott, T. C.; Byers, C. H. "An Emulsion Dispersion Reactor for Producing Stoichiometrically-Controlled Submicron Superconductor Precursors," submitted to *AIChE J.*

Holmes, J. M.; Byers, C. H. *Countermeasures to Hazardous Chemicals*, ORNL-6492, April, 1989

Howard, A. J.; Carta, G.; Byers, C. H. "Novel Applications of Continuous Annular Chromatography: Separation of Sugars," *New Directions in Sorption Technology*, Butterworth, 1989, pp. 342-49

Perona, J. J.; Gibson, M. R.; Byers, C. H. "Vacuum Sorption Pumping at Cryogenic Temperatures of Argon and Oxygen on Molecular Sieves," presented at NATO ASI on Adsorption: Science and Technology, Vimeiro, Portugal, July 17-20, 1988, and submitted to *Sep. Sci. Technol.*

Sisson, W. G.; Begovich, J. M.; Byers, C. H.; Scott, C. D. "Continuous Chromatography," *CHEMTECH* **18**, 498-502 (1988)

Cagle, E. B.

Bigelow, J. E.; Cagle, E. B.; Knauer, J. B. "Fabrication of 50-mg ²⁵²Cf Neutron Sources for the FDA Activation Analysis Facility," *J. Radioanal. Nucl. Chem.*, Articles, **123** (2) 471-89 (1988)

Campbell, D. O.

Campbell, D. O. "Cesium Removal from Low-Level Liquid Wastes with Hexacyanoferrate(II) Compounds," submitted to *Trans. Am. Nucl. Soc.* **60**, 131-132 (1989)

Campbell, D. O. "Annual Report for IAEA Research Agreement No. 4682/CF, Development of Processes Using Inorganic Sorbents for Liquid Waste Treatment (Attachment to Correspondence)"

Campbell, D. O. "Characterization Studies of Solids from Reactor Coolant System Water and Test Filter Media from Three Mile Island Unit 2," submitted to *Nucl. Technol.*

Campbell, D. O. "Characterization of Solids in TMI-2 Defueling System Water," presented at The TMI Accident: Materials Behavior and Plant Recovery Technology, Am. Nucl. Soc. Top. Meet., Washington, DC, October 31-November 4, 1988, and published in *Trans. Am. Nucl. Soc.* **57**, 527-28 (1988)

Campbell, D. O.; Burch, W. D. "The Chemistry of Fuel Reprocessing: Present Practices, Future Trends," submitted to *J. Radioanal. Nucl. Chem.*

Campbell, D. O.; Collins, E. D. "Use of Inorganic Sorbents for Waste Treatment at Oak Ridge National Laboratory," presented at IAEA Research Coordination Meet. on Use of Inorganic Sorbents for Liquid Waste Treatment and Backfill for Underground Repositories, Mol, Belgium, September 5-9, 1988

Cates, M. R.

Cates, M. R. "Measuring Temperature with Lasers—Newspaper Article," *The Oak Ridger*, Oak Ridge, TN, December 4, 1988

Cline, R. L.

Johnson, C. L.; Cline, R. L.; Ferren, D. M.; Tracy, J. G.; Haff, K. W.; Keller, O. L.; Genung, R. K.; Hoffman, D. C.; Moody, D. "ISOTOPICS Newsletter, No. 17, October 1988," (1988).

Cochran, D. K.

Cochran, D. K.; Beaver, J. E. "Production of Thallium-201 for Nuclear Medicine Cardiac Scans," to be used as a poster in the Isotope Program

Cochran, H. D.

Cochran, H. D.; Lee, L. L. "Solvation Structure in Supercritical Fluid Mixtures Based on Molecular Distribution Functions," presented at Annu. Meet. American Inst. of Chemical Engineers, Washington, DC, November 28-December 2, 1988, and published in *Supercritical Fluid Science and Technology, ACS Symposium Series No. 406*, 1989, pp. 27-38

Barber, T. A.; Cochran, H. D.; Bienkowski, P. R.; "Solubility of Solid CCl_4 in Supercritical CF_4 Using Directly Coupled Supercritical Fluid Extraction—Mass Spectrometry," to be published in *Sep. Sci. Technol.*

Cochran, H. D.; Wu, R.-S.; Lee, L. L. "Molecular Clustering in Super-critical Solutions," to be published in *Sep. Sci. Technol.*

Davis, W., Jr.; Cochran, H. D. *Modeling Vapor-Liquid Equilibrium of UF₆ and Common Impurities*, K/ITP-170, March, 1989

Davis, W., Jr.; Cochran, H. D.; Leitmaker, J. M. Liquid-Phase Compositions from Vapor-Phase Analyses: VI. Preliminary Comparisons of Calculations with Experimental Data on ClO₂F, CClF₂, SF₆, and CO₂, K/ITP-181, September, 1989

Cochran, H. D.; Lee, L. L.; Pfund, D. M. "Structure and Properties of Supercritical Fluid Mixtures from Kirkwood-Buff Fluctuation Theory and Integral Equation Methods," to be published in *Fluctuation Theory of Mixtures and Materials*, Taylor and Francis, New York, in press

Cochran, H. D.; Pfund, D. M.; Lee, L. L. "Study of Fluctuations in Supercritical Solutions by an Integral Equation Method," presented at Symp. on Supercritical Fluids, Nice, France, October 17-20, 1988, and published in *Proc. Symp. on Supercritical Fluids*, 1988, pp. 245-52

Cochran, H. D.; Pfund, D. M.; Lee, L. L. "Theoretical Models of Thermodynamic Properties of Supercritical Solutions," *Sep. Sci. Technol.* **23**, 2031-47 (1988)

Gilbert, V. P.; Bienkowski, P. R.; Cochran, H. D. "Proposed Technique for Surface Dynamics Measurements at High Pressure," *Sep. Sci. Technol.* **23**, 1941-48 (1988)

Lee, L. L.; Debenedetti, P. G.; Cochran, H. D. "Fluctuation Theory of Supercritical Solutions," to be published in *Supercritical Fluid Technology*, CRC Press, Cleveland, in press

Pfund, D. M.; Lee, L. L.; Cochran, H. D. "Application of the Kirkwood-Buff Theory of Solutions to Dilute Supercritical Mixtures. 2. The Excluded Volume and Local Composition Models," *Fluid Phase Equilibria* **39**, 161-92 (1988)

Wu, R. -S.; Lee, L. L.; Cochran, H. D. "The Structure of Dilute Supercritical Solutions: Clustering of Solvent and Solute Molecules and the Thermodynamic Effects," submitted to *Ind. Eng. Chem. Res.*

Collins, E. D.

Osborne, M. F.; Collins, J. L.; Lorenz, R. A.; Travis, J. R.; Webster, C. S. *Data Summary Report for Fission Product Release Test VI-2*, ORNL/TM-11105, September, 1989

Collins, J. L.; Pattison, W. L.; Kelmers, A. D. "Thirty-Gallon-Scale of Uranium from Production or Trash Type Radioactive Wastes from the Y-12 Plant," poster session at the Chemical Technology Division Information Meeting, December 8, 1988

Kelmers, A. D.; Collins, J. L.; Pattison, W. L. "Thirty-Gallon-Scale Leaching of Uranium from Production—or Trash Type Radioactive Wastes," presented at the Chemical Technology Division Information Meeting, December 8, 1988

Clepp, R. B.; Francis, C. W.; Kelmers, A. D.; Cline, J. E.; Collins, J. L.; Jones, L. S. *Trash Plan for the Uranium Lysimeter Project*, Y/TS-404, April 22, 1988

Campbell, D. O.; Collins, E. D. "Use of Inorganic Sorbents for Waste Treatment at Oak Ridge National Laboratory," presented at IAEA Research Coordination Meet. on Use of Inorganic Sorbents for Liquid Waste Treatment and Backfill for Underground Repositories, Mol, Belgium, September 5-9, 1988

Collins, E. D. "Nuclear Fuel Reprocessing," presented at Technical Aspects of Nuclear Weapons Proliferation Workshop, ORNL, Oak Ridge, TN, February 14-17, 1989, and at Nuclear Non-Proliferation Workshop, ORNL, Oak Ridge, TN, May 9-12, 1989

Collins, E. D.; Bigelow, J. E.; Ottinger, C. L.; Burney, G. A. "Recovery of Selected Elements from Irradiated Targets," to be published in *Nuclear Waste Management Technology*, Vol. 2, Harwood Academic Publ.

Collins, E. D.; Box, W. D.; Godbee, H. W.; Scott, T. C. "Analysis of Data from Leaching Concrete Samples Taken from the TMI-2 Reactor Building Basement," presented at Am. Nucl. Soc. Top. Meet. on TMI-2 Accident Materials Behavior and Plant Recovery Technology, Washington, DC, October 31-November 4, 1988, published in *Trans. Am. Nucl. Soc.* 57, 525-27 (1988); and to be published in *Nucl. Technol.*

McNeese, L. E.; Berry, J. B.; Butterworth, G. E., III; Collins, E. D.; Monk, T. H.; Patton, B. D.; Snider, J. W. *Overall Strategy and Program Plan for Management of Radioactively Contaminated Liquid Wastes and Transuranic Sludges at the Oak Ridge National Laboratory*, ORNL/TM-10757, December, 1988

Collins, J. L.

Collins, J. L.; Pattison, W. L.; Kelmers, A. D. "Thirty-Gallon-Scale Leaching of Uranium from Production- or Trash-Type Low-Level Radioactive Wastes," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988, and at Pu/U Recovery Operations Conf., Idaho Falls, ID, October 4-6, 1988

Osborne, M. F.; Collins, J. L.; Lorenz, R. A.; Travis, J. R.; Webster, C. S.; Yamashita, T. *Data Summary Report for Fission Product Release Test VI-1*, ORNL/TM-11104, June, 1989

Osborne, M. F.; Lorenz, R. A.; Collins, J. L.; Nakamura, T. "Time-Dependent Release of Fission Products from LWR Fuel Under Severe Accident Conditions," presented at Int. Eur. Nucl. Soc./Am. Nucl. Soc. Conf. on Thermal Reactor Safety, Avignon, France, October 3-7, 1988, and published in *Proc. Int. Eur. Nucl. Soc./Am. Nucl. Soc. Conf. on Thermal Reactor Safety*, Societe Francaise d'Energie Nucleaire, 1988, v. 4, pp. 1292-302

Osborne, M. F.; Lorenz, R. A.; Collins, J. L.; Travis, J. R.; Webster, C. S. "Fission Product Release at Severe Accident Conditions," presented at Project Review for Visitors from the Japan Inst. of Nuclear Safety, ORNL, Oak Ridge, TN, February 21, 1989

Walker, B. A., Editor; Saylor, R. E., Editor; Kettle, R. H.; Cada, G. F.; Clapp, R. B.; Collins, J. L.; Daniels, K. L.; Griest, W. H.; Kelmers, A. D.; Kroodsmas, R. L.; Lee, S. Y.; Lee, R. R.; McMahan, J. M.; Parr, P. D.; Pattison, W. L.; Tomkins, B. A. *Data Package for the Low-Level Waste Disposal Development and Demonstration Program Environmental Impact Statement*, sects. 1-7, appendixes A-O, ORNL/TM-10939/V1 and 2, September, 1988

Counce, R. M.

Crawford, D. B.; Counce, R. M. "The Depletion of Aqueous Nitrous Acid in Packed Towers," *Sep. Sci. Technol.* **23**, 1573-594 (1988)

Lucero, A. J.; Wilson, J. H.; Counce, R. M.; Villiers-Fisher, J. F.; Singh, S. P.; Ashworth, R. A. "Air Stripping of Volatile Organic Compounds from Groundwater Using Towers Filled with Standard and High-Efficiency Packing," presented at Summer Natl. Meet. Am. Inst. Chem. Eng., Philadelphia, August 20-23, 1989

Robinson, S. M.; Counce, R. M.; Smith, G. V. "An Experimental Study of Important Parameters for the Operation of a Continuous-Flow Fluidic Pump," *J. Fluid Control* **19**, 7-21 (1988)

Singh, S. P.; Counce, R. M. *Removal of Volatile Organic Compounds from Groundwater: A Survey of the Technologies*, ORNL/TM-10724, May, 1989

Singh, S. P. N.; Wilson, J. H.; Counce, R. M.; Villiers-Fisher, J. F.; Jennings, H. L.; Lucero, A. J.; Reed, G. D.; Ashworth, R. A.; Elliott, M. G. "Removal of Volatile Organic Compounds from Groundwater Using a Rotary Air Stripper," presented at Summer Natl. Meet. Am. Inst. Chem. Eng., Philadelphia, August 20-23, 1989

Croff, A. G.

Croff, A. G. "Waste Management," presented at Technical Aspects of Nuclear Weapons Proliferation Workshop, ORNL, Oak Ridge, TN, February 14-17 and May 9-12, 1989

Kocher, D. C.; Croff, A. G. "A Proposed Classification System for High-Level and Other Radioactive Wastes," *Radioact. Waste Manage. Nucl. Fuel Cycle* **11**, 227-268 (1988)

Davison, B. H.

Bajpai, R.; Su, H.; Davison, B. H. "Gas Hold-Up in Three-Phase Immobilized Cell Bioreactors," presented at 11th Symp. on Biotechnology for Fuels and Chemicals, Colorado Springs, CO, May 8-12, 1989, and submitted to *Appl. Biochem. Biotechnol.*

Davison, B. H. "Phase Holdup and Dispersion in a Three-Phase Fluidized-Bed Bioreactor with Low Density Gel Beads," presented at Biochemical Engineering VI, Engineering Foundation Conf., Santa Barbara, CA, October 2-7, 1988, and to be published in *Ann. NY Acad. of Sci.*

Davison, B. H. "Modeling of a Three-Phase Fluidized-Bed Bioreactor as a Dispersed Plug Flow Reactor," presented at Engineering Foundation Conf., Biochemical Engineering VI, Santa Barbara, CA, October 2-7, 1988, and at 11th Symp. on Biotechnology for Fuels and Chemicals, Colorado Springs, CO, May 8-12, 1989

Davison, B. H. "Ethanol Production by Immobilized *Zymomonas mobilis* in a Three-Phase Fluidized-Bed Bioreactor," presented at Biochemical Engineering Group, Dep. of Chemical Engineering, Johns Hopkins Univ., Baltimore, November 1, 1988

Davison, B. H. "Dispersion and Holdup in a Three-Phase Fluidized-Bed Bioreactor," *Appl. Biochem. Biotechnol.* **20/21**, 449-60 (1989)

Davison, B. H.; Nicklaus, D. N.; Faison, B. D. "Characterization and Subsequent Utilization of Microbially Solubilized Coal: Preliminary Studies," presented at Biological Processing of Coal and Coal-Derived Substances, Palo Alto, CA, May 16-17, 1989, and in *Biological Processing of Coal and Coal-Derived Substances*, EPRI Report no. ER-6572, 1989

Davison, B. H.; Nicklaus, D. N.; Misra, A.; Lewis, S. N.; Faison, B. D. "Utilization of Microbially Solubilized Coal: Preliminary Studies on Anaerobic Conversion," presented at 11th Symp. on Biotechnology for Fuels and Chemicals, Colorado Springs, CO, May 8-12, 1989, and submitted to *Appl. Biochem. Biotechnol.*

Faison, B. D.; Scott, C. D.; Davison, B. H. "Biosolubilization of Coal in Aqueous and Nonaqueous Media," to be published in *Proc. Int. Symp. on Chemistry of Microbial Coal Conversion and Bioprocessing of Coal*, Am. Chem. Soc. Meet., Los Angeles, Sept. 25-30, 1988

Godia, F.; Adler, H. I.; Davison, B. H.; Scott, C. D. "Use of Immobilized Microbial Membrane Fragments to Reduce Oxygen Content and Enhance the Acetone-Butanol Fermentation," submitted to *Enzyme Microb. Technol.*

Scott, C. D.; Davison, B. H. "Ethanol Production from Industrial Feedstocks by Immobilized *Zymomonas mobilis* in a Fluidized-Bed Bioreactor," presented at 8th Int. Biotechnology Symp., Paris, July 17-22, 1988

Scott, C. D.; Reichle, D. R.; Davison, B. H. *Bioprocessing Research at Oak Ridge National Laboratory*, 1989

Del Cul, G. D.

Del Cul; G. D.; Tallent, O. K.; Sams, T. L. "Monitor for Waste Immobilized in Cement-Based Materials," presented at Technology Transfer Workshop, Oak Ridge, TN, March 23, 1989

Tallent, O. K.; McDaniel, E. W.; Del Cul, G. D.; Dodson, K. E. "Computer Application to Leach Study Data," in *Abstracts of 90th Annu. Meet. Am. Ceram. Soc., Cincinnati*, May 4-5, 1988, p. 342, 1988

Tallent, O. K.; McDaniel, E. W.; Del Cul, G. D.; Dodson, K. E.; Trotter, D. R. *Development of Immobilization Technology for Hanford Double-Shell Slurry Feed Waste*, ORNL/TM-10906, August, 1989

Delzer, D. B.

McDaniel, E. W.; Tallent, O. K.; Sams, T. L.; Delzer, D. B. "Basis for Selecting Cement-Based Waste Forms for Immobilizing Radioactive Waste," presented at 12th Int. Symp. on Scientific Basis for Nuclear Waste Management, Internationales Congress Centrum, Berlin, October 10-13, 1988

McDaniel, E. W., Compiler; Delzer, D. B., Compiler "FUETAP Concrete: A Tailored, Autoclaved Radioactive Waste Host," *Radioactive Waste Forms for the Future*, North-Holland, 1988, pp. 565-88

DePaoli, D. W.

Clark, B. R.; DePaoli, D. W.; McTaggart, D. R.; Patton, B. D. "An On-Line Voltammetric Analyzer for Trace Metals in Wastewater," *Anal. Chim. Acta* **215**, 13-20 (1988)

Elliott, M. G.; DePaoli, D. W. "In Situ Venting of Jet Fuel-Contaminated Soil," presented at 44th Industrial Waste Conference, Purdue University, West Lafayette, Ind., May 8-10, 1989, and to be published in *Proc. 44th Industrial Waste Conf.*, Purdue University, May 8-10, 1989

DePaoli, D. W.; Herbes, S. E.; Elliott, M. G. "Performance of In Situ Soil Venting System at Jet Fuel Spill Site," presented at EPA Soil Vapor Extraction Technology Workshop, Edison, NJ, June 28, 1989, and to be published in *Proc. EPA Soil Vapor Extraction Technology Workshop*

DePaoli, S. M.

DePaoli, S. M.; Wright, T. C.; Rivera, A. L. "Computer-Based HAZWDDD Data Base Demonstration," presented at WMTC 2nd Workshop on Leaching Tests, Oak Ridge, TN, October 25-26, 1988

Kibbey, A. H.; DePaoli, S. M. *A Compilation of the Electricity Generated and Low-Level Radioactive Waste Shipped for Disposal by U.S. Nuclear Power Plants, 1959-1985*, ORNL/TM-10440, December, 1987

McGinnis, C. P.; Eisenhower, B. M.; Reeves, M. E.; DePaoli, S. M.; Stinton, L. H.; Harrington, E. H. *Hazardous Waste Development, Demonstration, and Disposal (HAZWDDD) Program Plan*, ES/ESH-5/V1, February, 1989

Rivera, A. L.; DePaoli, S. M. "Engineered Waste Disposal: Future Prospects and Challenges," presented at 197th Natl. Meet. Am. Chem. Soc., Dallas, April 9-14, 1989, and published in *Reprints of Papers Presented at 197th Am. Chem. Soc. Natl. Meet.*, Dallas, April 9-14, 1989, Am. Chem. Soc., Div. Environ. Chem., 1989, v. 1, pp. 136-37

Rivera, A. L.; Osborne-Lee, I. W.; DePaoli, S. M. "Systems Analysis Support to the Waste Management Technology Center," in *Proc. DOE Model Conf.*, Oak Ridge TN, October 3-7, 1988, CONF-881054, vol.1, 1988, pp. 81-88

Wright, T. C.; Osborne-Lee, I. W.; DePaoli, S. M.; Rivera, A. L. "Computer-Based LLWDDD Data Base Demonstration," presented at WMTC 2nd Workshop on Leaching Tests, Oak Ridge, TN, October 25-26, 1988

DeVore, J. R.

Kobisk, E. H.; Ramey, D. W.; Aaron, W. S.; Tompkins, J. A.; Haff, K. W.; DeVore, J. R.; Adair, H. L. "Tritium-Processing Operations at the Oak Ridge National Laboratory with Emphasis on Safe-Handling Practices," presented at Int. Nucl. Target Dev. Soc. and IAEA-INDC Conf., Darmstadt, Federal Republic of Germany, September 5-9, 1988, and published in *Nucl. Instrum. Methods Phys. Res. A* **282**, 329-40 (1989)

Dickerson, L. S.

Stewart, R. C.; Dickerson, L. S.; Joost, S. F.; Osucha, D. C. *Remote-Handled Transuranic Solid Waste Characterization Study: Oak Ridge National Laboratory*, ORNL/TM-11050, June, 1989

Dinsmore, T. V.

Solomon, D. K.; Haese, R. C.; Dinsmore, T. V.; Kelmers, A. D. *Sampling and Analysis of SWSA 6 Trench Leachates and Groundwaters*, ORNL/TM-10813, December, 1988

Dodson, K. E.

Spence, R. D.; Bostick, W. D.; McDaniel, E. W.; Gilliam, T. M.; Shoemaker, J. L.; Tallent, O. K.; Morgan, I. L.; Evans-Brown, B. S.; Dodson, K. E. "Immobilization of Technetium in Blast Furnace Slag Grouts," presented at 3rd Int. Conf. on Fly Ash, Silica Fume, Slag, and Natural Pozzolans in Concrete, Trondheim, Norway, June 19-26, 1989, and published in *Proc. 3rd Int. Conf. on Fly Ash, Silica Fume, Slag, and Natural Pozzolans in Concrete*, Vol. 2, pp. 1579-1596

Tallent, O. K.; McDaniel, E. W.; Del Cul, G. B.; Dodson, K. E. "Computer Application to Leach Study Data," in p. 342, *Abstracts of 90th Annu. Meet. Am. Ceram. Soc., Cincinnati*, May 4-5, 1988 (1988)

Tallent, O. K.; McDaniel, E. W.; Del Cul, G. D.; Dodson, K. E.; Trotter, D. R. *Development of Immobilization Technology for Hanford Double-Shell Slurry Feed Waste*, ORNL/TM-10906, August, 1989

Dole, L. R.

Mattus, A. J.; Gilliam, T. M.; Dole, L. R. *Review of EPA, DOE, and NRC Regulations on Establishing Solid Waste Performance Criteria*, ORNL/TM-9322, July, 1988

Morris, M. I.; Dole, L. R. "Treatment of Priority Liquid Waste Streams on the Oak Ridge Reservation," presented at Summer Natl. Meet. Am. Inst. Chem. Eng., Denver, August 21-24, 1988, and at 49th Int. Water Conf., Pittsburgh, October 24-26, 1988

Morris, M. I.; Dole, L. R.; Spence, R. D.; Gilliam, T. M. "Immobilization of Volatile Organics in Cement-Based Grouts," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988

Rivera, A. L.; Alsharif, M.; Phung, D. L.; Dole, L. R. "An Integrated Waste Facility Performance Model," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988, and to be published in *Abstract Booklet, DOE Model Conf., Oak Ridge, TN, Oct. 3-7, 1988*

Donaldson, T. L.

Donaldson, T. L. "Biotechnology for Ground Water Cleanup," to be published in *The Oak Ridger*, Oak Ridge, TN

Donaldson, T. L. "The ChE Pipeline—1987," to be published in *Chem. Eng. Prog.*

Donaldson, T. L. "Scale-Up and Process Design," presented at Meet. Am. Soc. Microbiol., Houston, September 15-16, 1989

Donaldson, T. L.; Strandberg, G. W.; Bolla, L. L. "Bioremediation of TCE-Contaminated Groundwater," presented at Meet. American Inst. of Chemical Engineers, Houston, April 2-6, 1989, and to be published in *Proc. Meet. American Inst. of Chemical Engineers*, Houston, Apr. 2-6, 1989

Donaldson, T. L.; Strandberg, G. W.; McGinnis, C. P.; Palumbo, A. V.; White, D. C.; Hill, D. L.; Phelps, T. J.; Hadden, C. T.; Revis, N. W.; Holdsworth, G.; Osborne, T. *Bioremediation of PCB-Contaminated Soil at the Y-12 Plant*, ORNL/TM-10750, September, 1988

Donaldson, T. L.; Strandberg, G. W.; Palumbo, A. V. "Biotreatment of TCE-Contaminated Groundwater" presented at Spring Natl. Meet. Am. Inst. Chem. Eng., Houston, April 2-6, 1989

Hill, D. L.; Phelps, T. J.; Palumbo, A. V.; Strandberg, G. W.; Donaldson, T. L.; White, D. C. "Bioremediation of Polychlorinated Biphenyls: Degradation Capabilities in Field Lysimeters," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988, and to be published in *Program Booklet, DOE Model Conf., Oak Ridge, TN, Oct. 3-7, 1988*; also published in *Appl. Biochem. Biotechnol.* **20/21**, 233-43 (1989)

Pitt, W. W.; Strandberg, G. W.; Donaldson, T. L.; Bolla, L. L. "Biodegradation of Trichloroethylene Contaminated Groundwater," presented at Conf. on Biotechnology Applications in Hazardous Waste Treatment, Longboat Key, FL, October 30-November 4, 1988, and to be published in *Proc. Conf. on Biotechnology Applications in Hazardous Waste Treatment*, Longboat Key, FL, Oct. 30-Nov. 4, 1988

Sayler, G. S.; Blackburn, J. W.; Donaldson, T. L. *Environmental Biotechnology of Hazardous Wastes*, ORNL/TM-10853, August, 1988

Strandberg, G. W.; Donaldson, T. L.; Bolla, L. L.; Palumbo, A. V.; Eng, W. "Degradation of Trichloroethylene and *Trans*-1,2-Dichloroethylene by a Methanotrophic Consortia in a Trickle-Type Bioreactor," to be published in *Proc. DOE Model Conf.*, Oak Ridge, TN, Oct. 3-7, 1988

Strandberg, G. W.; Donaldson, T. L.; Bolla, L. L.; Palumbo, A. V.; Eng, W. "Biodegradation of Trichloroethylene and *Trans*-1,2-Dichloroethylene by Methanotrophs in a Laboratory-Scale Trickle Bioreactor," presented at DOE Model Conf., Oak Ridge, TN, Oct. 3-7, 1988, and to be published in *Abstract Booklet, DOE Model Conf., Oak Ridge, TN, Oct. 3-7, 1988*

Strandberg, G. W.; Donaldson, T. L.; Farr, L. L. "Degradation of Trichloroethylene and *Trans*-1,2-Dichloroethylene by a Methanotrophic Consortium in a Fixed-Film, Packed-Bed Bioreactor," *Environ. Sci. Technol.* **23**, 1422-25 (1989)

Walker, J. F., Jr.; Helfrich, M. V.; Donaldson, T. L. "Biodenitrification of Uranium Refinery Wastewaters," *Environ. Prog.* **8**, 97-101 (1989)

Egan, B. Z.

Egan, B. Z.; Lee, D. D.; McWhirter, D. A. "Response to Comments on Solvent Extraction and Recovery of Ethanol from Aqueous Solutions," submitted to *Ind. Eng. Chem. Res.*

Egan, B. Z.; Singh, S. P. N.; Fain, D. E.; Kidd, G. J. "Gas Separations Using Inorganic Membranes," in *Proc. 8th Annu. Gasification and Gas Stream Cleanup Systems Contractors Review Meet.*, Morgantown, WV, May 10-12, 1988, DOE/METC 88/6092, 1988, v. 2, pp. 519-24, and in *Gas Stream Cleanup Papers from DOE/METC Sponsored Contractors Review Meetings in 1988*, DOE/METC-89/6099, 1988, pp. 287-92

Egan, B. Z.; Singh, S. P. N.; Fain, D. E.; Kidd, G. J.; Roettger, G. E.; White, D. E. "Gas Separations Using Inorganic Membranes," presented at 9th Annu. Gasification and Gas Stream Cleanup Systems Con-

tractors Review Meet., Morgantown, WV, June 27-29, 1989, and to be published in *Proc. 9th Annu. Gasification and Gas Stream Cleanup Systems Contractors Review Meet.*, Morgantown, WV, June 27-29, 1989

Fain, D. E.; Kidd, G. J.; Egan, B. Z.; Singh, S. P. N. "Development and Evaluation of Ceramic Membranes for Gas Separation," presented at Fossil Energy Materials Conf., Oak Ridge, TN, August 30-September 1, 1988

Fain, D. E.; Kidd, G. J.; White, D. E.; Roettger, G. E.; Egan, B. Z.; Singh, S. P. N. "Development and Evaluation of Ceramic Membranes for Gas Separation," presented at Fossil Energy Materials Conf., Oak Ridge, TN, August 8-10, 1989

Jimenez, B. D.; Burtis, L. A.; Ezell, G. H.; Egan, B. Z.; Lee, N. E.; Beauchamp, J. J.; McCarthy, J. F. "The Mixed Function Oxidase System of Bluegill Sunfish *Lepomis macrochirus*: Correlation of Activities Found in Experimental and Feral Fish," *Environ. Toxicol. Chem.* 7, 623-34 (1988)

Evans-Brown, B. S.

Gilliam, T. M.; Spence, R. D.; Evans-Brown, B. S.; Morgan, I. L.; Shoemaker, J. L.; Bostick, W. D. "Performance Testing of Blast Furnace Slag for Immobilization of Technetium in Grout," presented at SPECTRUM '88, Int. Top. Meet. on Nuclear and Hazardous Waste Management, Pasco, WA, September 11-15, 1988, and published in *Proc. SPECTRUM '88*, Int. Top. Meet. on Nuclear and Hazardous Waste Management, Pasco, WA, September 11-15, 1988, American Nuclear Society, Inc., La Grange Park, IL, 1988, pp. 109-11

Spence, R. D.; Bostick, W. D.; McDaniel, E. W.; Gilliam, T. M.; Shoemaker, J. L.; Tallent, O. K.; Morgan, I. L.; Evans-Brown, B. S.; Dodson, K. E. "Immobilization of Technetium in Blast Furnace Slag Grouts," presented at 3rd Int. Conf. on Fly Ash, Silica Fume, Slag, and Natural Pozzolans in Concrete, Trondheim, Norway, June 19-26, 1989, and published in *Proc. 3rd Int. Conf. on Fly Ash, Silica Fume, Slag, and Natural Pozzolans in Concrete*, Vol. 2, pp. 1579-1596

Faison, B. D.

Davison, B. H.; Nicklaus, D. M.; Misra, A.; Lewis, S. N.; Faison, B. D. "Utilization of Microbially Solubilized Coal: Preliminary Studies on Anaerobic Conversion," presented at 11th Symp. on Biotechnology for Fuels and Chemicals, Colorado Springs, CO, May 8-12, 1989, and submitted to *Appl. Biochem. Biotechnol.*

Davison, B. H.; Nicklaus, D. N.; Faison, B. D. "Characterization and Subsequent Utilization of Microbially Solubilized Coal: Preliminary Studies," presented at Biological Processing of Coal and Coal-Derived Substances, Palo Alto, CA, May 16-17, 1989, and in *Biological Processing of Coal and Coal-Derived Substances*, EPRI Report no. ER-6572, 1989

Faison, B. D. "Coal Solubilization by Lower Fungi," presented at Annu. Meet. Am. Soc. Microbiol., New Orleans, May 15-18, 1989

Faison, B. D.; Lewis, S. N. "Microbial Coal Solubilization in Defined Culture Systems: Biochemical and Physiological Studies," presented at DOE Office of Fossil Energy Workshop on Processing of Coals, Tyson's Corner, VA, August 15-18, 1988, and to be published in *Resources, Conservation, and Recycling*

Faison, B. D.; Lewis, S. N.; Cancel, C. A. "Metal Binding by *Micrococcus luteus*: Recovery of Strontium (Sr) from Aqueous Solution," presented at Annu. Meet. Soc. Ind. Microbiol., Chicago, August 7-12, 1988

Faison, B. D.; Lewis, S. N.; Misra, A. K. "Production of Coal Solubilizing Activity by *Paecilomyces* sp. During Submerged Growth in Defined Liquid Media," *Appl. Biochem. Biotechnol.* **20/21**, 743-52 (1989)

Faison, B. D.; Scott, C. D. "Biosolubilization of Coal in Aqueous and Nonaqueous Media," presented at Int. Symp. on Chemistry of Microbial Coal Conversion and Bioprocessing of Coal, Am. Chem. Soc. Meet., Los Angeles, September 25-30, 1988, and to be published in *Proc. Int. Symp. on Chemistry of Microbial Coal Conversion and Bioprocessing of Coal*

Faison, B. D.; Watson, J. S.; Cancel, C. A.; Scott, C. D. "Applications of Biotechnology in Waste Management for the Nuclear Industry," presented at Annu. Meet. Air Waste Management Assoc., Southern Section, Diamondhead, MS, September 20-22, 1989

Faison, B. D.; Woodward, C. A. "Degradation of Heterocyclic Coal Model Compounds by Coal-Solubilizing Fungi," presented at Annu. Meet. Soc. Ind. Microbiol., Seattle, August 14-18, 1989

Faison, B. D.; Woodward, C. A.; Bean, R. M. "Microbial Solubilization of a Preoxidized Subbituminous Coal: Product Characterization," presented at 11th Symp. on Biotechnology for Fuels and Chemicals, Colorado Springs, CO, May 8-12, 1989, and to be published in *Proc. 11th Symp. on Biotechnology for Fuels and Chemicals*, *Appl. Biochem. and Biotechnol.*

Faison, B. D.; Woodward, C. A.; Lewis, S. N.; Bean, R. M. "The Biochemistry of Microbial Coal Solubilization: Model Compound Studies and Product Characterization," presented at 11th Symp. on Biotechnology for Fuels and Chemicals, Colorado Springs, CO, May 8-12, 1989

Scott, C. D.; Faison, B. D. "The Conversion of Coal to Liquids and Gasses by Advanced Bioprocessing Systems," presented at Coal -- Targets of Opportunity Workshop, Washington, DC, July 12-13, 1988, and to be published in *Proc. Workshop on Coal—Targets of Opportunity*, Washington, DC, July 12-13, 1988

Scott, C. D.; Faison, B. D. "Biological Solubilization of Coal in Aqueous and Nonaqueous Media," in *Proc. Direct Liquefaction Contractors' Review Meet.*, Pittsburgh, October 6-8, 1987, DOE, 1988, pp. 324-39

Watson, J. S.; Scott, C. D.; Faison, B. D. "Immobilized *Micrococcus* for Uptake of Strontium from Dilute Solutions," presented at Spring Symp. Am. Chem. Soc., I&EC Division, Atlanta, May 1-4, 1989, and to be published in *ACS Symp. Series*, Am. Chem. Soc.

Watson, J. S.; Scott, C. D.; Faison, B. D. "Adsorption of Sr by Immobilized Microorganisms," *Appl. Biochem. Biotechnol.* **20/21**, 699-709 (1989)

Farr, L. L.

Strandberg, G. W.; Donaldson, T. L.; Farr, L. L. "Degradation of Trichloroethylene and *Trans*-1,2-Dichloroethylene by a Methanotrophic Consortium in a Fixed-Film, Packed-Bed Bioreactor," *Environ. Sci. Technol.* **23**, 1422-25 (1989)

Felker, L. K.

Felker, L. K.; Toth, L. M. "Fluorine Gettering by Activated Charcoal in a Radiation Environment," *Sep. Sci. Technol.* **23**, 1959-68 (1988)

Toth, L. M.; Felker, L. K. "Fluorine Generation by Gamma Radiolysis of a Fluoride Salt Mixture," accepted by *Radiat. Eff.*

Toth, L. M.; Felker, L. K. "Purex Acid-Split Flowsheet Studies," ORNL/TM-10993, April, 1989

Thiyagarjan, P.; Diamond, H.; Soderholm, L.; Horwitz, E. P.; Toth, L. M.; Felker, L. K. "Pu(IV) Polymers in Aqueous and Organic Solutions," *Inorg. Chem.*, in press

Ferrada, J. J.

Ferrada, J. J.; Stark, L.; Rodgers, B. R. "Developing Expert Systems for Hazardous and Low-Level Radioactive Waste Management," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988, and to be published in *Proc. DOE Model Conf.*, Oak Ridge, TN, Oct. 3-7, 1988

Stark, L.; Ferrada, J. J. "Interfaces Between Artificial Intelligence and Conventional Programming Languages with Applications in Waste Control," presented and published in abstracts of *Proc. Spring Meet. Am. Inst. Chem. Eng.*, Houston, Apr. 3, 1989

Ferrada, J. J.; Osborne-Lee, I. W. "Applications of Artificial Intelligence to Chemical Engineering Processes," *Chem. Process.* **52**, 23-33 (1989)

Ferren, D. M.

Johnson, C. L.; Cline, R. L.; Ferren, D. M.; Tracy, J. G.; Haff, K. W.; Keller, O. L.; Genung, R. K.; Hoffman, D. C.; Moody, D. *ISOTOPICS*, no. 17, October 1988

Fisher, P. W.

Schuresko, D. D.; Cole, M. J.; Fisher, P. W. et al. "Simplified Eight-Shot Pneumatic Pellet Injector for Plasma Fueling Applications on the Princeton Beta Experiment and on the Advanced Toroidal Facility," *J. Vac. Sci. Technol.* **A7** 949-54 (1989)

Combs, S. K.; Foster, C. A.; Milora, S. L.; Schuresko, D. D.; Gouge, M. J.; Fisher, P. W.; Argo, B. E.; Barber, G. C.; Fehling, D. T.; Foust, C. R.; Gethers, F. E.; Haselton, H. H.; Ponte, N. S.; Qualls, A. L.; Simmons, D. W.; Sohns, C. W.; Sparks, D. O.; Tsai, C. C.; Whitson, J. C. "Pellet Injector Research at ORNL," in *Fusion Technology 1988, Proc. 15th Symp. on Fusion Technology*, Utrecht, The Netherlands, September 19-23, 1988, Elsevier Science Publ., 1989, pp. 709-14

Fisher, P. W.; Bauer, M. L.; Baylor, L. R.; Combs, S. K.; Deleanu, L. E.; Fehling, D. T.; Foster, C. A.; Foust, C. R.; Gouge, M. J.; Milora, S. L.; Schuresko, D. D.; Sparks, D. O.; Whitson, J. C. "Tritium Pellet Injector Results," presented at Am. Vac. Soc. 35th Natl. Vacuum Symp./Top. Conf., Atlanta, October 3-7, 1988, and published in *J. Vac. Sci. Technol.* **7**, 938-43 (1989)

Fisher, P. W.; Milora, S. L.; Combs, S. K.; Carlson, R. V.; Coffin, D. O. "Tritium Proof-of-Principle Injector Experiment," *Fusion Technol.* **14**, 977-81 (1988)

Milora, S. L.; Combs, S. K.; Foster, C. A.; Schuresko, D. D.; Gouge, M. J.; Fisher, P. W.; Argo, B. E.; Barber, G. C.; Fehling, D. T.; Foust, C. R.; Gethers, F. E.; Ponte, N. S.; Qualls, A. L.; Simmons, D. W.; Sparks, D. O.; Whitson, J. C. "Pneumatic Pellet Injector Research at ORNL," presented at IAEA

Technical Committee Meet. on Pellet Injection and Toroidal Confinement, Gut Ising, Federal Republic of Germany, October 24-26, 1988, and to be published in *Proc. IAEA Technical Committee Meet. on Pellet Injection and Toroidal Confinement*, IAEA-TECDOC-534, 1989

Qualls, A. L.; Fisher, P. W.; Cole, M. J.; England, A. C.; Fehling, D. T.; Gouge, M. J.; Houlberg, W. A.; Jernigan, T. C.; Milora, S. L.; Wilgren, J. B. "Pellet Ablation Studies on ATF," submitted to *Bull. Am. Phys. Soc.*

Wilgen, J. B.; Bell, G. L.; Bell, J. D.; England, A. C.; Fisher, P. W.; Howe, H. C.; Hutchinson, D. P.; Ma, C. H.; Murakami, M.; Qualls, A. L.; Rasmussen, D. A.; Richards, R. K.; Uckan, T.; Wing, W. R. "Pellet Injection into ATF Plasmas," submitted to *Bull. Am. Phys. Soc.*

Forsberg, C. W.

Forsberg, C. W. "An Ocean Island Geological Repository for Disposal of Spent Fuel and High-Level Waste," submitted to *Nucl. Technol.*

Forsberg, C. W. "Identification and Characterization of Passive Safety System and Inherent Safety Feature Building Blocks for Advanced Light-Water Reactors," presented at IAEA Technical Committee Meet. on Passive Safety Features in Current and Future Water-Cooled Reactors, Moscow, March 21-24, 1989, and to be published in *Proc. IAEA Technical Committee Meet. on Passive Safety Features in Current and Future Water-Cooled Reactors*

Forsberg, C. W. "Directions of Nuclear Power Research," presented at Meet. Local Section, Am. Nucl. Soc., Columbia, MO, November 7, 1988

Forsberg, C. W. "A Process Inherent Ultimate Safety Boiling-Water Reactor PIUS/BWR," presented at Dep. of Nuclear Engineering, Univ. of Missouri, Columbia, November 8, 1988

Forsberg, C. W. "Sources, Hazards, and Disposal Methods for Radioactive Waste," presented at Dep. of Nuclear Engineering, Univ. of Michigan, Ann Arbor, February 3, 1989

Forsberg, C. W. et al. *A United States Sponsored Working Paper on Criteria for the Termination of International Atomic Energy Agency Safeguards on Nuclear Materials in Waste*, K/ITP-273, June, 1989

Forsberg, C. W. et al. *AVLIS Technology Significance Study Part 2: Description of Systems*, K/ITP-169 Part 2, 1989

Forsberg, C. W. "New Developments in Reactor Design," presented at Weld. Test. Technol. Conf., Knoxville, TN, February 14-17, 1989

Fowler, V. L.

Berry, J. B.; Brown, C. H. Jr.; Fowler, V. L.; Robinson, S. M. "Treatment of ORNL Liquid Low-Level Waste," presented to the Division of Environmental Chemistry, American Chemical Society, Toronto, Canada, June 5-11, 1988

Berry, J. B.; Brown, C. H., Jr.; Fowler, V. L.; Robinson, S. M. "Treatment of ORNL Process Waste," presented at Spectrum '88, Int. Top. Meet. on Nuclear and Hazardous Waste Management, Pasco, WA, September 11-15, 1988, and published in *Proc. Spectrum '88*, 1988, pp. 416-20

Fowler, V. L.; Hewitt, J. D. *Filtration of Oak Ridge National Laboratory Simulated Liquid Low-Level Waste*, ORNL/TM-10653, August, 1989

Friedman, H. A.

Toth, L. M.; Bell, J. T.; Friedman, H. A. "The Disproportionation of Pu⁴⁺ in Nitric Acid Solutions," submitted to *Radiochim. Acta*

Gambill, W. R.

Copeland, G. L.; Gambill, W. R.; Harrington, R. M.; Johnson, J. A.; Peretz, F. J.; Reutler, H.; Ryskamp, J. M.; Selby, D. L.; West, C. D.; Yoder, G. L. *Advanced Neutron Source Final Preconceptual Reference Core Design*, ORNL/TM-11234, August, 1989

Lienhard, J. H.; Gambill, W. R. "An Upper Bound for the Critical Boiling Heat Flux," technical note in *J. Heat Trans.*, **111**, 3, 815-818 (1989)

Mochizuki, T.; Gambill, W. R. "Advanced Neutron Source Design: Burnout Heat Flux Correlation Development," *A.N.S. Trans.* **57** 298-300 (1988)

Genung, R. K.

Genung, R. K. "Bioprocessing Applications in the Management of Nuclear and Chemical Wastes," presented at Workshop on Bioprocessing Research for Energy Applications, Alexandria, VA, November 2-4, 1988, and to be published in *Workshop Report, Bioprocessing Research for Energy Applications, Alexandria, VA, November 2-4, 1988*

Genung, R. K. "Overview of the Chemical Technology Division, Oak Ridge National Laboratory," presented at Research and Training Opportunities Conf., Oak Ridge, TN, November 29-December 1, 1988

Genung, R. K. *Chemical Technology Division Safety Program: CY 1989*, ORNL/FPO-88/70, December, 1988

Genung, R. K. *Information Meeting 1988, December 7-9, 1988*, ORNL/M-684, December 7, 1988

Genung, R. K. *Chemical Technology Division Progress Report for the Period January 1, 1987, to June 30, 1988*, ORNL-6490, February, 1989

Johnson, C. L.; Cline, R. L.; Ferren, D. M.; Tracy, J. G.; Haff, K. W.; Keller, O. L.; Genung, R. K.; Hoffman, D. C.; Moody, D. *ISOTOPICS*, no. 17, October 1988

Singh, S. P. N.; Genung, R. K.; Jolley, R. L. *Guidance Document for Conducting Waste Management Demonstration Projects*, ORNL/TM-10845, July, 1988

Gibson, M. R.

Perona, J. J.; Gibson, M. R.; Byers, C. H. "Vacuum Sorption Pumping at Cryogenic Temperatures of Argon and Oxygen on Molecular Sieves," presented at NATO ASI on Adsorption: Science and Technology, Vimeiro, Portugal, July 17-20, 1988, and submitted to *Sep. Sci. Technol.*

Gilliam, T. M.

Gilliam, T. M. "Use of Coal Fly Ash in Radioactive Waste Disposal," presented at Conf. on Use of Coal Fly Ash in Waste Stabilization/Solidification Procedures, Covington, KY, September 20, 1989, and to be published in *Proc. Conf. on Use of Coal Fly Ash in Waste Stabilization/Solidification Procedures*, American Coal Ash Assoc., Inc.

Gilliam, T. M.; Morgan, I. L. "Thermal Properties of Cement-Based Grouts," in *Proc. 19th Int. Conf. on Thermal Conductivity*, Cookeville, TN, October 20-23, 1985, Plenum Publ. Corp., 1988, v. 19, pp. 93-108

Gilliam, T. M.; Spence, R. D.; Evans-Brown, B. S.; Morgan, I. L.; Shoemaker, J. L.; Bostick, W. D. "Performance Testing of Blast Furnace Slag for Immobilization of Technetium in Grout," presented at SPECTRUM '88, Int. Top. Meet. on Nuclear and Hazardous Waste Management, Pasco, WA, September 11-15, 1988, and published in *Proc. SPECTRUM '88*, American Nuclear Society, Inc., La Grange Park, IL, 1988, pp. 109-11

Mattus, A. J.; Gilliam, T. M.; Dole, L. R. *Review of EPA, DOE, and NRC Regulations on Establishing Solid Waste Performance Criteria*, ORNL/TM-9322, July, 1988

Morgan, I. L.; Osborne, S. C.; Spence, R. D.; Gilliam, T. M. "Immobilization of Volatile Organic Compounds in a Cement Waste Form: Sample Preparation Procedures," presented at Meet. Am. Chem. Soc., Div. Environ. Chem., Miami Beach, FL, September 10-15, 1989

Sams, T. L.; Gilliam, T. M. "Systematic Approach for the Design of Pumpable, Cement-Based Grouts for Immobilization of Hazardous Wastes," *Environmental Aspects of Stabilization and Solidification of Hazardous and Radioactive Wastes*, ASTM STP 1033, P. L. Cote and T. M. Gilliam, eds., American Society for Testing and Materials, Philadelphia, 1989

Morris, M. I.; Dole, L. R.; Spence, R. D.; Gilliam, T. M. "Immobilization of Volatile Organics in Cement-Based Grouts," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988

Morris, M. I.; Gilliam, T. M.; Kasten, J. L. "Selection and Treatment of Priority Liquid Waste Streams on the Oak Ridge Reservation," presented at 91st Annu. Meet. Am. Ceram. Soc., Indianapolis, April 23-27, 1989, and at 49th Int. Water Conf., Pittsburgh, October 24-26, 1988; published in *Proc. 49th Int. Water Conf.*, 1988, pp. 469-73

Morris, M. I.; Spence, R. D.; Gilliam, T. M.; Dole, L. R. "A Program to Establish In Situ Immobilization as a Remedial Action Option for Wastes Containing Volatile Organic Compounds (VOC)," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988, and published in *Proc. DOE Model Conf.*, CONF-881054, vol. 5, 1988, pp. 1501-08

Mrochek, J. E.; Gilliam, T. M.; McDaniel, E. W. "Grout Formulation for the Immobilization of Mixed Organic-Containing Wastes," in *SPECTRUM '86, Proc. Am. Nucl. Soc. Int. Top. Meet. on Waste Management and Decontamination and Decommissioning*, Niagara Falls, NY, September 14-18, 1986 (1988), v. 1, pp. 55-76

Spence, R. D.; Bostick, W. D.; McDaniel, E. W.; Gilliam, T. M.; Shoemaker, J. L.; Tallent, O. K.; Morgan, I. L.; Evans-Brown, B. S.; Dodson, K. E. "Immobilization of Technetium in Blast Furnace Slag Grouts," presented at 3rd Int. Conf. on Fly Ash, Silica Fume, Slag, and Natural Pozzolans in Concrete, Trondheim,

Norway, June 19-26, 1989, and published in *Proc. 3rd Int. Conf. on Fly Ash, Silica Fume, Slag, and Natural Pozzolans in Concrete*, Vol. 2, pp. 1579-1596

Spence, R. D.; Gilliam, T. M.; Morgan, I. L.; Osborne, S. C. "Immobilization of Volatile Organic Compounds in Commercial Cement-Based Waste Forms," news release for ACS News Service

Godbee, H. W.

Collins, E. D.; Box, W. D.; Godbee, H. W.; Scott, T. C. "Analysis of Data from Leaching Concrete Samples Taken from the TMI-2 Reactor Building Basement," presented at Am. Nucl. Soc. Top. Meet. on TMI-2 Accident Materials Behavior and Plant Recovery Technology, Washington, DC, October 31-November 4, 1988; published in *Trans. Am. Nucl. Soc.* **57**, 525-27 (1988); and to be published in *Nucl. Technol.*

Greenbaum, E.

Greenbaum, E. "Eleventh Symposium on Biotechnology for Fuels and Chemicals, Preregister Attendees, Program and Abstracts, May 8-12, 1989," 1989

Graves, D. A.; Greenbaum, E. "Evidence for the Interaction of Methane with the Photosynthetic Apparatus During Photosynthesis," presented at 11th Symp. on Biotechnology for Fuels and Chemicals, Colorado Springs, CO, May 8-12, 1989, and to be published in *Proc. 11th Symp. on Biotechnology for Fuels and Chemicals*

Graves, D. A.; Greenbaum, E. "In Situ, Automated Screening of Sustained Oxygen and Hydrogen Photoevolution by Individual Algal Colonies in an Anoxic Atmosphere," presented at Poster Session, Annu. Meet. Soc. Plant Physiol., Reno, NV, July 10-14, 1988

Graves, D. A.; Greenbaum, E. "A Simple Apparatus for Screening Absolute Photosynthetic Rates of Single Algal Colonies in an Anoxic Atmosphere," *Plant Physiol.* **90**, 246-50 (1988)

Graves, D. A.; Reeves, M. E.; Greenbaum, E. "Establishment of Control Parameters for In Situ, Automated Screening of Sustained Hydrogen Photoproduction by Individual Algal Colonies," *Plant Physiol.* **87**, 603-08 (1988)

Graves, D. A.; Spradlin, G. M.; Greenbaum, E. "Photoautotrophic Growth by *Chlamydomonas reinhardtii* in an Anoxic Atmosphere," *accepted by Enzyme Microbial. Technol.*

Graves, D. A.; Tevault, C. V.; Greenbaum, E. "Control of Photosynthetic Reductant: The Role of Light and Temperature on Sustained Hydrogen Photoevolution in *Chlamydomonas* sp. in an Anoxic, Carbon Dioxide-Containing Atmosphere," *Photochem. Photobiol.* **50**, 571-76 (1989)

Greenbaum, E. "Kinetic and Mechanistic Studies of Vectorial Photoelectrochemistry of Photosynthesis," presented at 13th DOE Solar Photochemistry Research Conf., Silver Creek, CO, June 12-15, 1989, and to be published in *Proc. 13th DOE Solar Photochemistry Research Conf.*

- Greenbaum, E. "Photosynthetic Water Splitting," presented at 7th Int. Conf. on Photochemical Conversion and Storage of Solar Energy, Evanston, IL, July 31-August 5, 1988, and published in *Photochemical Energy Conversion*, Elsevier, New York, 1989, pp. 184-95
- Greenbaum, E. "Small Molecule Redox Reactions with Photosynthetic Systems," presented at Gas Research Inst. Basic Research Contractor's Review Meet. on Gaseous Fuel Synthesis from Inorganic Resources, Chicago, October 26-27, 1988
- Greenbaum, E. "Biological Molecular Electronics," presented at Inst. Electr. Electron. Eng. Engineering in Medicine and Biology, 10th Annu. Int. Conf., New Orleans, November 4-7, 1988
- Greenbaum, E. "Biophotonic Materials: The Photosynthesis Connection," presented at Meet. Am. Phys. Soc., St. Louis, March 20-24, 1989
- Greenbaum, E. "Electrical Contact of Molecular Components in a Submicron Biological Structure," in *Molecular Electronic Devices, Proc. 3rd Int. Symp.*, Arlington, VA, October 6-8, 1986, Elsevier Science Publ., 1988, pp. 575-82
- Greenbaum, E. "Energetic Efficiency of Hydrogen Photoevolution by Algal Water Splitting," *Biophys. J.* **54**, 365-68 (1988)
- Greenbaum, E. "Hydrogen and Oxygen Production by Algae," *Algae and Human Affairs*, Cambridge Univ. Press, New York, 1988, pp. 283-304
- Greenbaum, E. "Photobioelectronic Studies with Thylakoid Membranes," *Appl. Biochem. Biotechnol.* **20/21**, 813-24 (1989)
- Greenbaum, E. "Biomolecular Electronics: Observation of Oriented Photocurrents by Entrapped Platinized Chloroplasts," *Bioelectrochem. Bioenerg.* **21**, 171-77 (1989)
- Greenbaum, E. "Announcement and Call for Papers, Twelfth Symposium on Biotechnology for Fuels and Chemicals, May 7-11, 1990," 1989
- Greenbaum, E. "Interfacial Photoreactions at the Photosynthetic Membrane Interface: An Upper Limit for the Number of Platinum Atoms Required to Form a Hydrogen-Evolving Platinum Metal Catalyst," *J. Phys. Chem.* **92**, 4571-74 (1988)
- Scott, C. D.; Greenbaum, E.; Michaels, G. E.; Mrochek, J. E.; Newman, E.; Petek, M.; Scott, T. C. "Preliminary Investigation of Possible Low-Temperature Fusion," presented at Workshop on Cold Fusion Phenomena, Santa Fe, NM, May 23-25, 1989; submitted to *J. Fusion Energy*
- Scott, C. D.; Greenbaum, E.; Wyman, C. E. "Introduction to the Proceedings of the Tenth Symposium on Biotechnology for Fuels and Chemicals," *Appl. Biochem. Biotechnol.* **20/21**, 1-3 (1989)
- Woodward, J.; Lewid, B.; Miracle, G.; Greenbaum, E. "Comparative Study of the Photochemistry of Chloroplast Membranes, and Photosystem II Particles," *Appl. Biochem. Biotechnol.* **20**, 259-65 (1989)
- Woodward, J.; Reeves, M.; Greenbaum, E.; Allison, D. "Effect of Exogenously Added Manganese Chloride on the Operational Stability of the Chloroplast-Ferredoxin-Hydrogenase System," *Enzyme Microb. Technol.* **10**, 123-28 (1988)

Grubb, R. G.

Rawl, R. R.; Grubb, R. G.; Ratledge, J. E.; Jones, R. H. "Transportation Operations Experience Involving Suspected Leaking Cesium Capsules," presented at 9th Int. Symp. on Packaging and Transportation of Radioactive Materials (PATRAM 89), Washington, DC, June 11-16, 1989, and published in *Proc. 9th Int. Symp. on Packaging and Transportation Materials (PATRAM '89)*, CONF 890631, vol. 1, pp. 238-246, 1989

Haas, P. A.

Haas, P. A. "Process for Electrolytically Preparing Uranium Metal," U. S. Statutory Invention Registration H659, August 1, 1989

Haas, P. A. "Gel Processes for Preparing Ceramics and Glasses," *Chem. Eng. Prog.* April, 44-52 (1989)

Notz, K. J.; Haas, P. A. "Properties and Thermal Decomposition of the Double Salts of Uranyl Nitrate-Ammonium Nitrate," *Thermochim. Acta* **155**, 283-295 (1989)

Haff, K. W.

Johnson, C. L.; Cline, R. L.; Ferren, D. M.; Tracy, J. G.; Haff, K. W.; Keller, O. L.; Genung, R. K.; Hoffman, D. C.; Moody, D. "ISOTOPICS Newsletter, No. 17, October 1988," (1988).

Kobisk, E. H.; Ramey, D. W.; Aaron, W. S.; Tompkins, J. A.; Haff, K. W.; DeVore, J. R.; Adair, H. L. "Tritium-Processing Operations at the Oak Ridge National Laboratory with Emphasis on Safe-Handling Practices," presented at Int. Nucl. Target Dev. Soc. and IAEA-INDC Conf., Darmstadt, Federal Republic of Germany, September 5-9, 1988, and published in *Nuc. Instrum. Methods Phys. Res. A* **282**, 329-40 (1989)

Hall, R.

Hall, R.; Brown, C. H., Jr.; Robinson, S. M.; Hewitt, J. D. "Operation of a Mobile Pilot-Scale Continuous Countercurrent Ion Exchange System for Treatment of Low-Level Radioactive Wastewater," presented at HAZTECH Int. '89 Conf., San Francisco, September 26-October 1, 1989, and published in *Proc. HAZTECH Int. '89 Conf.*, 1989, pp. 91-102

Harris, M. T.

Basaran, O. A.; Harris, M. T. "Asymptotic Analysis of Performance of Membrane Modules for Gas Separations," submitted to *Sep. Sci. Technol.*

Byers, C. H.; Harris, M. T. "A Microcrystalline Growth Study of Solvent Effects in Alkoxides by Dynamic Laser Light Scattering," *Ultrastructure Processing of Advanced Ceramics*, Wiley Interscience, 1988, pp. 843-54

Harris, M. T.; Byers, C. H. "The Effect of Solvent on the Homogeneous Precipitation of Titania by Titanium Ethoxide Hydrolysis," *J. Non-Cryst. Solids* **103**, 49-64 (1988)

Harris, M. T.; Byers, C. H. *An Orthogonal Collocation Approach to Modeling Multicomponent Adsorption in Carbon Beds*, ORNL/TM-10735, May, 1989

Harris, M. T.; Byers, C. H.; Brunson, R. R. "The Base Catalyzed Hydrolysis and Condensation Reactions of Dilute and Concentrated TEOS Solutions," submitted to *J. Non-Cryst. Solids*

Harris, M. T.; Byers, C. H.; Brunson, R. R. "The Base-Catalyzed Hydrolysis and Condensation Reactions of Dilute and Concentrated TEOS Solutions," presented at 5th Int. Workshop on Glasses and Ceramics from Gels, Rio de Janeiro, August 6-10, 1989; to be published in *Proc. 5th Int. Workshop on Glasses and Ceramics from Gels*; and submitted to *J. Non-Cryst. Solids*

Harris, M. T.; Byers, C. H.; Brunson, R. R. "The Effects of Aluminum Alkoxides on the Synthesis of Composite Powders of Alumina and Titania," presented at Mater. Res. Soc. Symp. on Processing Science of Advanced Ceramics, San Diego, April 24-29, 1989, and to be published in *Proc. Mater. Res. Soc. Symp. on Processing Science of Advanced Ceramics*, San Diego, Apr. 24-29, 1989

Harris, M. T.; Byers, C. H.; Brunson, R. R. "The Application of Laser-Light-Scattering Techniques in the Study of Ceramic Powders Synthesis by the Hydrolysis of Metal Alkoxides," presented at Workshop on Advanced Laser Technology for Chemical Measurements, Argonne, IL, May 9-11, 1989, and published in *Proc. Workshop on Advanced Laser Technology for Chemical Measurements*, 1989, pp. 82-86

Harris, M. T.; Byers, C. H.; Brunson, R. R. "A Study of Solvent Effects on the Synthesis of Pure Component and Composite Ceramic Powders by Metal Alkoxide Hydrolysis," in *Proc. Spring Meet. Mater. Res. Soc.*, Reno, NV, April 5-9, 1988, Mater. Res. Soc., 1988, v. 121, pp. 287-92

Harris, M. T.; Scott, T. C.; Byers, C. H. "An Emulsion Dispersion Reactor for Producing Stoichiometrically-Controlled Submicron Superconductor Precursors," submitted to *AIChE J.*

Helfrich, M. V.

Walker, J. F., Jr.; Helfrich, M. V.; Donaldson, T. L. "Biodenitrification of Uranium Refinery Wastewaters," *Environ. Prog.* **8**, 97-101 (1989)

Hewitt, J. D.

Fowler, V. L.; Hewitt, J. D. *Filtration of Oak Ridge National Laboratory Simulated Liquid Low-Level Waste*, ORNL/TM-10653, August, 1989

Hall, R.; Brown, C. H., Jr.; Robinson, S. M.; Hewitt, J. D. "Operation of a Mobile Pilot-Scale Continuous Countercurrent Ion Exchange System for Treatment of Low-Level Radioactive Wastewater," presented at HAZTECH Int. '89 Conf., San Francisco, September 26-October 1, 1989, and published in *Proc. HAZTECH Int. '89 Conf.*, 1989, pp. 91-102

Hightower, J. R.

Hightower, J. R. "Isotopes Are Big Business at ORNL," *The Oak Ridger*, Oak Ridge, TN, December 11, 1988

Hightower, J. R.; Houston, B. "Chemical Engineering in the FY 1989 Federal Budget," presented at Summer Natl. Meet. Am. Inst. Chem. Eng., Denver, August 21-24, 1988

Hinatsu, Y.

Osborne, M. F.; Lorenz, R. A.; Hinatsu, Y. "ORNL VI-4 Fission Product Release Tests: Results from Tests VI-3 and VI-4 and Future Plans," presented at Severe Accident Research Program Partners Review Meet., Idaho Falls, ID, April 10-14, 1989

Holladay, D. W.

Krichinsky, A. M.; Holladay, D. W.; Johnson, J. K.; Starnes, B. W. "Initial Decommissioning Activities for the Radiochemical Processing Plant at Oak Ridge National Laboratory," presented at Annu. Conf. Am. Glovebox Soc., Denver, August 22-25, 1988

Hollenbeck, P. E.

Jolley, R. L.; Hollenbeck, P. E.; Kennerly, J. M.; Morris, M. I.; Singh, S. P. N. "Evaluation of Selected Emerging Waste Treatment and Disposal Methods," presented at Meet. Am. Chem. Soc., Miami Beach, FL, September 10-15, 1989, and to be published in *Proc. Meet. Am. Chem. Soc.*, Miami Beach, FL, Sept. 10-15, 1989, Am. Chem. Soc.

Padgett, D.; Hollenbeck, P. E. "Sludge Detoxification Demonstration," presented at Incineration Conf., Knoxville, TN, May 1-5, 1989

Hylton, T. D.

Hylton, T. D.; Walker, J. F. *Environmental Studies in Support of the Live Fire Training Facilities Project*, ORNL/TM-11265, August, 1989

Jennings, H. L.

Singh, S. P.; Wilson, J. H.; Counce, R. M.; Villiers-Fisher, J. F.; Jennings, H. L.; Lucero, A. J.; Reed, G. D.; Ashworth, R. A.; Elliott, M. G. "Removal of Volatile Organic Compounds from Groundwater Using a Rotary Air Stripper," presented at Summer Natl. Meet. Am. Inst. Chem. Eng., Philadelphia, August 20-23, 1989

Johnson, C. L.

Johnson, C. L.; Cline, R. L.; Ferren, D. M.; Tracy, J. G.; Haff, K. W.; Keller, O. L.; Genung, R. K.; Hoffman, D. C.; Moody, D. "ISOTOPICS Newsletter, No. 17, October 1988," (1988).

Jolley, R. L.

Gaitan, E.; Cooksey, R. C.; Cruse, J. M.; Lewis, R. E.; Lindsay, R. H.; Jolley, R. L. "The Problem of Goitrogenic Substances in the Presence of Sufficient Iodine Supplementation," presented at 2nd Symp. on Topical Interdisciplinary Problems of Iodine Deficiency, Iodine Prophylaxis, Iodine Excess, and Antithyroid Substances, Karl-Marx-Stadt, Federal Republic of Germany, January 24-26, 1989, and to be published in *Proc. 2nd Symp. on Topical Interdisciplinary Problems of Iodine Deficiency, Iodine Prophylaxis, Iodine Excess, and Antithyroid Substances*

Gaitan, E.; Cooksey, R. C.; Gaitan, D.; Meydrech, E. F.; Cruse, J. M.; Adams, J. G.; Chapman, S. W.; Lindsay, R. H.; Jolley, R. L.; Gaitan, G. S.; Preuss, T. M.; Guzman, R. N.; Astudillo, J.; Duque, E.; Gallo, H. "Goitre and Lymphocytic Autoimmune Thyroiditis in Iodine-Sufficient Areas," submitted to *J. Autoimmunity*

Gaitan, E.; Jolley, R. L.; Lindsay, R. H.; Cooksey, R. C.; Hill, J. B.; Island, D. P. "Resorcinol: Final Goitrogenic Product in Water from a Goitrogenic Well," *Clin. Ecol.* 5(4): 176-184 (1988)

Johnson, J. D.; Jolley, R. L. "Water Chlorination: The Challenge," pp. 21-27 in *Water Chlorination: Chemistry, Environmental Impact and Health Effects*, vol. 6, R. L. Jolley et al., eds., Lewis Publishers, Inc., Chelsea, Michigan, 1990

Jolley, R. L. "Biographical Sketch," p. ix; "Preface (Proceedings of the Sixth Water Chlorination Conference)," pp. v-vii; "Conference Summary and Perspective," pp. 973-975; in *Water Chlorination: Chemistry, Environmental Impact and Health Effects*, vol. 6, R. L. Jolley et al., eds., Lewis Publishers, Inc., Chelsea, Michigan, 1990

Jolley, R. L. "Symposium Description and Biographical Sketch," presented at 197th Meet. Am. Chem. Soc., Dallas, April 9-14, 1989

Jolley, R. L. "Innovative Waste Management Technologies," presented at 197th Natl. Am. Chem. Soc. Meet., Dallas, April 9-14, 1989, and *Preprints of Papers Presented at 197th Am. Chem. Soc. Natl. Meet.*, Dallas, April 9-14, 1989, Am. Chem. Soc., Div. Environ. Chem., 1989, v. 29, pp. 123-24

Jolley, R. L. "Alternative Treatment Technologies Workshop for Superfund Sites, Oak Ridge, TN, November 7-8, 1988," (1988).

Jolley, R. L. "Trace Substances Present as Chlorination By-Products in Drinking and Process Waters," in *Proc. 22nd Annu. Conf. on Trace Substances in Environmental Health*, St. Louis, May 23-26, 1988, Univ. of Missouri, Columbia, 1989, pp. 205-14

Jolley, R. L.; Hollenbeck, P. E.; Kennerly, J. M.; Morris, M. I.; Singh, S. P. N. "Evaluation of Selected Emerging Waste Treatment and Disposal Methods," *Am. Chem. Soc.*, Division of Environmental Chemistry Preprints 29(2): 331 (1989)

Jolley, R. L.; Johnson, J. D. "The Interim: Selected Chemical Research Reported Between Conferences Five and Six," pp. 3-19 in *Water Chlorination: Chemistry, Environmental Impact, and Health Effects*, vol. 6, R. L. Jolley et al., eds., Lewis Publishers, Inc., Chelsea, Michigan, 1990

Jolley, R. L.; Rivera, A. L. "Continuous Monitoring, Automated Analysis, and Sampling Procedures," *Water Pollut. Control Assoc.* 60(6): 799-801(1988)

Jolley, R. L.; Rivera, A. L. "Continuous Monitoring, Automated Analysis, and Sampling Procedures," *Water Pollut. Control Assoc.* 61(6): 782-785(1989)

Jolley, R. L.; Rivera, A. L.; Fox, E. C.; Hyfantis, G. J.; McBrayer, J. F. "Strategic Planning for Waste Management: Characterization of Chemically and Radioactively Hazardous Waste and Treatment, Storage, and Disposal Capabilities for Diverse and Varied Multisite Operations," in *Proc. Waste Management '88*, Tucson, AZ, February 28-March 3, 1988, Univ. of Arizona, Tucson, 1988, v. 1, pp. 951-53

Morris, M. I.; Jolley, R. L.; Singh, S. P. N. "The Waste Management Technology Center's Role in Waste Management Technology Applications for Martin Marietta Energy Systems, Inc.," presented at Weld. Test. Tech. Conf., Knoxville, TN, February 1989

Pojasek, R. B.; Jolley, R. L. "The Developing Role of Chemists in the Field of Solid and Hazardous Waste Management," presented at Meet. Am. Chem. Soc., Los Angeles, September 25-30, 1988

Singh, S. P. N.; Genung, R. K.; Jolley, R. L. *Guidance Document for Conducting Waste Management Demonstration Projects*, ORNL/TM-10845, July, 1988

Waters, L. C.; Schenley, R. L.; Owen, B. A.; Walsh, P. J.; Hsie, A. W.; Jolley, R. L.; Buchanan, M. V.; Condie, L. W. "Biotesting of Waste Water: A Comparative Study Using the Salmonella and CHO Assay Systems," *Environ. Molecular Mutagenesis* **14**, 254-263 (1989)

Jones, K. E.

Moore, R. S.; Jones, K. E.; Moisson, I. R.; Rhyne, B. T.; Notz, K. J. "Characteristics of LWR Spent Fuel," presented at Am. Nucl. Soc. Int. Top. Meet. on Nuclear and Hazardous Waste Management, Tri-Cities, WA, September 11-15, 1988

Joy, D. S.

Joy, D. S.; Nehls, J. R., Jr.; Harrison, I. G.; Miller, C.; Vogel, L. W.; Martin, J. D.; Capone, R. L.; Dougherty, L. "Modeling the Design and Operations of the Civilian Radioactive Waste Processing System," presented at the Waste Management Meeting, Tucson, Arizona, February 26- March 2, 1989, and published in *Waste Management* **89**, vol. 1, p. 173

Andress, D. A.; McLeod, N. B.; Joy, D. S. "Waste Stream Analysis Model," presented at the Waste Management Meeting, Tucson, Arizona, February 26-March 2, 1989, and published in *Waste Management* **89**, vol. 1, p. 375

McLeod, N. B.; Andress, D.; Joy, D. S. *The Use of Spent Fuel Selection Strategies to Control Waste Stream Radiological Properties*, JAI-317, February 1989

Morris, M. I.; Gilliam, T. M.; Kasten, J. L. "Selection and Treatment of Priority Liquid Waste Streams on the Oak Ridge Reservation," presented at 91st Annu. Meet. Am. Ceram. Soc., Indianapolis, April 23-27, 1989; presented at 49th Int. Water Conf., Pittsburgh, October 24-26, 1988; published in *Proc. 49th Int. Water Conf.*, Pittsburgh, October 24-26, 1988 (1988), pp. 469-73

Kelmers, A. D.

Collins, J. L.; Pattison, W. L.; Kelmers, A. D. "Thirty-Gallon-Scale of Uranium from Production or Trash Type Radioactive Wastes from the Y-12 Plant," poster session at the Chemical Technology Division Information Meeting, December 8, 1988

Kelmers, A. D.; Collins, J. L.; Pattison, W. L. "Thirty-Gallon-Scale Leaching of Uranium from Production—or Trash Type Radioactive Wastes," presented at the Chemical Technology Division Information Meeting, December 8, 1988

Clepp, R. B.; Francis, C. W.; Kelmers, A. D.; Cline, J. E.; Collins, J. L.; Jones, L. S. *Trash Plan for the Uranium Lysimeter Project*, Y/TS-404, April 22, 1988

Collins, J. L.; Pattison, W. L.; Kelmers, A. D. "Thirty-Gallon-Scale Leaching of Uranium from Production-or Trash-Type Low-Level Radioactive Wastes," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988, and at Pu/U Recovery Operations Conf., Idaho Falls, ID, October 4-6, 1988

Kelmers, A. D.; Shappert, L. B.; Roddy, J. W.; Hermann, O. W.; Parks, C. V. *Identification and Evaluation of Radionuclide Generation/Depletion Codes for Potential Use by the Department of Energy's Office of Civilian Radioactive Waste Management*, ORNL/TM-10404, February, 1989

Solomon, D. K.; Haese, R. C.; Dinsmore, T. V.; Kelmers, A. D. *Sampling and Analysis of SWSA 6 Trench Leachates and Groundwaters*, ORNL/TM-10813, December, 1988

Walker, B. A., Editor; Saylor, R. E., Editor; Ketelle, R. H.; Cada, G. F.; Clapp, R. B.; Collins, J. L.; Daniels, K. L.; Griest, W. H.; Kelmers, A. D.; Kroodsmas, R. L.; Lee, S. Y.; Lee, R. R.; McMahon, J. M.; Parr, P. D.; Pattison, W. L.; Tomkins, B. A. *Data Package for the Low-Level Waste Disposal Development and Demonstration Program Environmental Impact Statement*, sects. 1-7, appendixes A-O, ORNL/TM-10939/V1 and 2, September, 1988

Kennerly, J. M.

Jolley, R. L.; Hollenbeck, P. E.; Kennerly, J. M.; Morris, M. I.; Singh, S. P. N. "Evaluation of Selected Emerging Waste Treatment and Disposal Methods," presented at Meet. Am. Chem. Soc., Miami Beach, FL, September 10-15, 1989, and to be published in *Proc. Meet. Am. Chem. Soc.*, Miami Beach, FL, Sept. 10-15, 1989, Am. Chem. Soc.

Faulkner, R. L.; Bundy, R. D.; Crump, B. F.; Kennerly, J. M. *Gaseous Diffusion Complex Decontamination and Decommissioning Strategy Paper*, ORO draft, September, 1989

Butterworth, G. E.; McDaugald, E. S.; Kennerly, J. M.; Rivera, A. L.; Van Hoesen, S. D. *Low Level Waste Disposal Development and Demonstration Program, FY 1989 Integrated Implementation Plan for the Oak Ridge Reservation*, ES/ESH-7 draft 0, December 16, 1988

Van Hoesen, S. D.; Kennerly, J. M.; Williams, L. C.; Lingle, W. N.; Peters, M. S.; Darnell, G. R. *Report of Foreign Travel of U. S. Department of Energy Delegation to France*, ORNL/FTR-2901, August 8, 1988

Faulkner, R. L.; Bundy, R. D.; Crump, B. F.; Kennerly, J. M. *Decontamination and Decommissioning Plan for the Oak Ridge Gaseous Diffusion Plant*, K/SS-523, draft, March, 1989

Butterworth, G. E.; Kennerly, J. M. "Overview of the Low-Level Waste Disposal Development and Demonstration Program," presented to the Agence Nationale pour les Gestion des Dechets Radioactif (ANDRA), Paris, France, June 6, 1988

Kennerly, J. M. "Supercompaction/Grouting of RTR Drums at ORNL," videotape

Kibbey, A. H.

Kibbey, A. H.; DePaoli, S. M. *A Compilation of the Electricity Generated and Low-Level Radioactive Waste Shipped for Disposal by U.S. Nuclear Power Plants, 1959-1985*, ORNL/TM-10440, December, 1987

King, L. J.

Kerr, H. T.; King, L. J.; Shappert, L. B.; Unger, W. E.; Worley, B. A.; Wymer, R. G. "Presentations at Technical Aspects of Nuclear Weapons Proliferation Workshop, Oak Ridge National Laboratory, Oak Ridge, Tennessee, December 13-14, 1988," to be published in *Papers Presented at Technical Aspects of Nuclear Weapons Proliferation Workshop, ORNL, Oak Ridge, TN, Dec.13-14, 1988*

King, L. J. "Transuranium Processing Plant," presented at Technical Aspects of Nuclear Weapons Proliferation Workshop, ORNL, Oak Ridge, TN, February 14-17 and May 9-12, 1989

Klein, J. A.

Klein, J. A.; Ashline, R. C.; DePaoli, S. M.; Godbee, H. W.; Kibbey, A. H.; Pearson, R. L.; Storch, S. N. *Integrated Data Base for 1988: Spent Fuel and Radioactive Waste Inventories, Projections, and Characteristics*, DOE/RW-0006, Rev. 4, September, 1988

Klein, J. A. "A Check List of the Reptiles and Amphibians on the Department of Energy Oak Ridge Reservation, Anderson and Roane Counties, Tennessee," *J. Tennessee Acad. Sci.* **64**, 228-30 (1989)

Knauer, J. B.

Bigelow, J. E.; Cagle, E. B.; Knauer, J. B. "Fabrication of 50-mg ^{252}Cf Neutron Sources for the FDA Activation Analysis Facility," *J. Radioanal. Nucl. Chem.*, Articles, **123** (2) 471-89 (1988)

Kobisk, E. H.

Kobisk, E. H.; Ramey, D. W.; Aaron, W. S.; Tompkins, J. A.; Haff, K. W.; DeVore, J. R.; Adair, H. L. "Tritium-Processing Operations at the Oak Ridge National Laboratory with Emphasis on Safe-Handling Practices," presented at Int. Nucl. Target Dev. Soc. and IAEA-INDC Conf., Darmstadt, Federal Republic of Germany, September 5-9, 1988, and published in *Nuc. Instrum. Methods Phys. Res. A* **282**, 329-40 (1989)

Koop, O. C.

Koop, O. C.; Mulligan, P. J.; Reeves, D. K. *A Preliminary Assessment of the Relationship Between the Mineralogy and Diagenetic History of Four Argillaceous Rocks and Their Thermomechanical Properties*, ORNL/TM-10739, November, 1988

Krichinsky, A. M.

Liebetrau, A. M.; Krichinsky, A. M.; Anderson, L. G.; Crawford, J. M.; Goldman, A. S.; Hume, M. W., Jones, F. E.; LeBaron, G. J.; Spraktes, F. W.; Suda, S. C.; Thomas, C. C. "Volume Calibration Techniques for Nuclear Materials Control," American National Standards Institute Standard N15.19-1988, August 4, 1988

Krichinsky, A. M. "NUMATH: A Nuclear Material Holdup Estimator," *J. Inst. Nucl. Mater. Manage.* **16**, 30-32 (1988)

Krichinsky, A. M.; Holladay, D. W.; Johnson, J. K.; Starnes, B. W. "Initial Decommissioning Activities for the Radiochemical Processing Plant at Oak Ridge National Laboratory," presented at Annu. Conf. Am. Glovebox Soc., Denver, August 22-25, 1988

Lee, D. D.

Butterworth, G. E.; Morris, M. I.; Lee, D. W. *Radioactive Waste Management Implementation Plan for Paducah Gaseous Diffusion Plant*, ES/ESH-9, KY/H-86, May, 1989

Butterworth, G. E.; Morris, M. I.; Lee, D. W. *Radioactive Waste Management Implementation Plan for Portsmouth Gaseous Diffusion Plant*, ES/ESH-10, POE F-2011, May, 1989

Bowers, T. A.; Conner, G. D., Gibson, L. V. Jr.; Kirkpatrick, C. R.; Wojtowicz, B. V.; Lee, D. D. *ORGDP Hazardous Waste Development Demonstration and Disposal (HAZWDDD) Implementation Plan*, K/HS-233, September 1988

Kimmitt, R. R.; Fellers, H. L.; Lee, D. D. *HAZWDDD Implementation Plan for the Y-12 Plant*, Y/TS-468, October 1988

Egan, B. Z.; Lee, D. D.; McWhirter, D. A. "Response to Comments on Solvent Extraction and Recovery of Ethanol from Aqueous Solutions," submitted to *Ind. Eng. Chem. Res.*

Lee, N. E.

Jimenez, B. D.; Burtis, L. A.; Ezell, G. H.; Egan, B. Z.; Lee, N. E.; Beauchamp, J. J.; McCarthy, J. F. "The Mixed Function Oxidase System of Bluegill Sunfish *Lepomis macrochirus*: Correlation of Activities Found in Experimental and Feral Fish," *Environ. Toxicol. Chem.* **7**, 623-34 (1988)

Lee, N. E.; Lima, M.; Woodward, J. "Hydrolysis of Cellulose by a Mixture of *Trichoderma reesei* Cellobiohydrolase and *Aspergillus niger* Endoglucanase," *Biochem. Biophys. Acta* **967**, 437-40 (1988)

Lee, N. E.; Woodward, J. "Reversible Inactivation and Fluorescence Changes of Cellobiohydrolase I by Dithiothreitol Treatment," presented at Natl. Meet. Am. Chem. Soc., Miami Beach, FL, September 10-15, 1989, and to be published in *Proc. Natl. Meet. Am. Chem. Soc.*, Miami Beach, FL

Lee, N. E.; Woodward, J. "Kinetics of the Adsorption of *Trichoderma reesei* Cellulase to DEAE-Macrosorb," presented at 196th Natl. Meet. Am. Chem. Soc., Los Angeles, September 25-30, 1988, and published in *J. Biotech.* **11**, 75-82 (1989)

Woodward, J.; Lee, N. E. "Cellulase Recovery: Problems and Potential," presented at Annu. Review Meet. on Ethanol from Biomass, SERI, Golden, CO, January 17-19, 1989

Woodward, J.; Lee, N. E.; Carmichael, J. S. "Denaturation and Renaturation of *Trichoderma reesei* C30 Cellobiohydrolase I," presented at TRICEL 89 Int. Symp. on *Trichoderma* Cellulases, Technical Univ., Vienna, September 14-16, 1989

Woodward, J.; Lee, N. E.; Carmichael, J. S.; McNair, S. L.; Wichert, J. M. "Comparison of the Hydrolytic Activity and Fluorescence of Native, Guanidine Hydrochloride-Treated and Renatured Cellobiohydrolase I from *Trichoderma reesei*," submitted to *Biochem. Biophys. Acta*

Woodward, J.; Lima, M.; Lee, N. E. "The Role of Cellulase Concentration in Determining the Degree of Synergism in the Hydrolysis of Microcrystalline Cellulose," *Biochem. J.* **255**, 895-99 (1988)

Woodward, J.; McNair, S. L.; Lee, N. E. "Utilization of *p*-Nitrophenylcellobioside as a Substrate for Monitoring the Characteristics of Purified Cellobiohydrolase I from *Trichoderma reesei*," presented at 11th Symp. on Biotechnology for Fuels and Chemicals, Colorado Springs, CO, May 8-12, 1989, and to be published in *Book of Abstracts, 11th Symp. on Biotechnology for Fuels and Chemicals, Colorado Springs, CO, May 8-12, 1989*

Lewis, S. N.

Davison, B. H.; Nicklaus, D. M.; Misra, A.; Lewis, S. N.; Faison, B. D. "Utilization of Microbially Solubilized Coal: Preliminary Studies on Anaerobic Conversion," submitted to *Appl. Biochem. Biotechnol.*

Davison, B. H.; Nicklaus, D. M.; Woodward, C. A.; Lewis, S. N.; Faison, B. D. "Characterization and Subsequent Utilization of Microbially Solubilized Coal: Preliminary Studies," presented at Biological Processing of Coal and Coal-Derived Substances, Palo Alto, CA, May 16-17, 1989, and to be published in *Biological Processing of Coal and Coal-Derived Substances, Palo Alto, CA, May 16-17, 1989, EPRI Report*

Davison, B. H.; Nicklaus, D. N.; Misra, A.; Lewis, S. N.; Faison, B. D. "Utilization of Microbially Solubilized Coal: Preliminary Studies on Anaerobic Conversion," presented at 11th Symp. on Biotechnology for Fuels and Chemicals, Colorado Springs, CO, May 8-12, 1989

Faison, B. D.; Lewis, S. N. "Microbial Coal Solubilization in Defined Culture Systems: Biochemical and Physiological Studies," presented at DOE Office of Fossil Energy Workshop on Processing of Coals, Tyson's Corner, VA, August 15-18, 1988, and to be published in *Resources, Conservation, and Recycling*

Faison, B. D.; Lewis, S. N.; Cancel, C. A. "Metal Binding by *Micrococcus luteus*: Recovery of Strontium (Sr) from Aqueous Solution," presented at Annu. Meet. Soc. Ind. Microbiol., Chicago, August 7-12, 1988

Faison, B. D.; Lewis, S. N.; Misra, A. K. "Production of Coal Solubilizing Activity by *Paecilomyces* sp. During Submerged Growth in Defined Liquid Media," *Appl. Biochem. Biotechnol.* **20/21**, 743-52 (1989)

Faison, B. D.; Woodward, C. A.; Lewis, S. N.; Bean, R. M. "The Biochemistry of Microbial Coal Solubilization: Model Compound Studies and Product Characterization," presented at 11th Symp. on Biotechnology for Fuels and Chemicals, Colorado Springs, CO, May 8-12, 1989

Scott, C. D.; Lewis, S. N. "Biological Solubilization of Coal Using Both In Vivo and In Vitro Processes," presented at Semin., Univ. of Kentucky, Lexington, January 19, 1989, and published in *Appl. Biochem. Biotechnol.* **18**, 403-12 (1988)

Strandberg, G. W.; Lewis, S. N. "Factors Affecting Coal Solubilization by the Bacterium *Streptomyces setonii* 75Vi2 and by Alkaline Buffers," *Appl. Biochem. Biotechnol.* **18**, 355-61 (1988)

Lin, K. H.

Lin, K. H.; Standifer, P.; Vaughn, V. C. A. *Comprehensive Self-Assessment and Upgrade Program Progress Report for the Period January 1 to June 30, 1989*, ORNL/TM-11288, September, 1989

Lindemer, T. B.

Hubbard, C. R.; Lindemer, T. B.; Brynestad, J.; Kroeger, D. "Reaction of CO₂ Superconductor YBa₂Cu₃O_{7-x}," presented at Annu. Meet. of American Crystallographic Assoc., Seattle, July 23-28, 1989

Lindemer, T. B. "Experimental Investigation and Chemical Thermodynamic Representation of <U_{1-z}Gd_zO_{2±x}>," presented at 7th Int. Symp. on Thermodynamics of Nuclear Materials, Chicago, September 26-30, 1988

Lindemer, T. B. "Chemical Thermodynamic Representation of Nonstoichiometry in <UO_{2-x}N_{3x/4}>," *CALPHAD* 13, 109-113 (1989)

Lindemer, T. B., seminar, Fuel and Fission-Product Reactions in HTGR Fuels, Savannah River Laboratory, Aiken, SC, April 17, 1989

Lindemer, T. B.; Hunley, J. F.; Gates, J. E.; Sutton, A. L., Jr.; Brynestad, J.; Hubbard, C. R.; Gallagher, P. K. "Experimental and Thermodynamic Study of Nonstoichiometry in <YBa₂Cu₃O_{7-x}>," *J. Am. Ceram. Soc.* 72, 1775-88 (1989)

Lindemer, T. B.; Hunley, J. F.; Gates, J. E.; Sutton, A. L., Jr.; Brynestad, J.; Hubbard, C. R.; Gallagher, P. K. *Experimental and Thermodynamic Study of Nonstoichiometry in <YBa₂Cu₃O_{7-x}>*, ORNL/TM-10899, May, 1989

Lindemer, T. B.; Hunley, J. F.; Gates, J. E.; Sutton, A. L., Jr.; Brynestad, J.; Hubbard, C. R.; Gallagher, P. K. "Experimental and Thermodynamic Study of Nonstoichiometry in <YBa₂Cu₃O_{7-x}>," seminar, Department of Ceramics, Rutgers, The State University, Piscataway, NJ, Feb. 7, 1989

Lindemer, T. B.; Hunley, J. F.; Gates, J. E.; Sutton, A. L., Jr.; Brynestad, J.; Hubbard, C. R.; Gallagher, P. K. "Experimental and Thermodynamic Study of Nonstoichiometry in <YBa₂Cu₃O_{7-x}>," seminar, E.I. duPont Experimental Engineering Station, Wilmington, DE, April 5, 1989

Lindemer, T. B.; Hunley, J. F.; Gates, J. E.; Sutton, A. L., Jr.; Brynestad, J.; Hubbard, C. R.; Gallagher, P. K. "Experimental and Thermodynamic Study of Nonstoichiometry in <YBa₂Cu₃O_{7-x}>," seminar, National Institute of Standards and Technology, Gaithersburg, MD, August 10, 1989

Lindemer, T. B.; Hunley, J. F.; Gates, J. E.; Sutton, A. L., Jr.; Brynestad, J.; Hubbard, C. R.; Gallagher, P. K. "Experimental and Thermodynamic Study of Nonstoichiometry in <YBa₂Cu₃O_{7-x}>," seminar, Department of Materials Engineering, Purdue University, W. Lafayette, IN, September 29, 1989

Lindemer, T. B.; Sutton, A. L., Jr. "Experimental and Thermodynamic Study of Nonstoichiometry of YBa₂Cu₃O_{7-x}," presented at Symp. on Superconductivity, 91st Annu. Meet. Am. Ceram. Soc., Indianapolis, April 23-27, 1989

Lindemer, T. B.; Sutton, A. L., Jr. "Experimental and Thermodynamic Study of Nonstoichiometry of YBa₂Cu₃O_{7-x}," presented at 44th Annu. Calorimetry Conf., Oak Ridge, TN, July 30-August 4, 1989

Lindemer, T. B.; Sutton, A. L., Jr. "Study of Nonstoichiometry of <U_{1-z}Gd_zO_{2±x}>," *J. Am. Ceram. Soc.* 71, 553-61 (1988)

Lindemer, T. B.; Sutton, A. L., Jr. *Review of Nonstoichiometry in YBa₂Cu₃O_{7-x}*, ORNL/TM-10827, November, 1988

Morss, L. R.; Jensen, M. P.; Thorn, R. J.; Dorris, S. E. (Argonne National Laboratory); Lindemer, T. B.; and Naito, N. (GTE Labs. Inc., Waltham, MA), "Enthalpies of Formation of 298 K of Complex Oxides Coexisting with $\text{YBa}_2\text{Cu}_3\text{O}_y$; Y_2BaCuO_5 and BaCuO_{2+x} ," presented at the 44th Annual Calorimetry Conf., Oak Ridge, TN, July 30–August 4, 1989

Tiegs, T. N. (Metals and Ceramics) and Lindemer, T. B., Surface Modified Silicon Carbide Whiskers and Method for Preparing Same, ORNL Patent Application 152,276

Lomenick, T. F.

Lomenick, T. F. "Workshop on the Management of Contaminated Soils," Knoxville, TN, November 10, 1988

Lomenick, T. F.; Kasprowicz, J. D. "Disposal of Radioactive Wastes into Clay-Rich Rocks," presented at Clay Microstructure Workshop/Conf., Stennis Space Center, MS, October 4-7, 1988

Lorenz, R. A.

Kress, T. S.; Lorenz, R. A.; Nakamura, T.; Osborne, M. F. "Correlation of Recent Fission Product Release Data," presented at ICHMT Int. Semin. on Fission Product Transport Processes in Reactor Accidents, Dubrounik, Yugoslavia, May 22-26, 1989, and to be published in *Proc. ICHMT Int. Semin. on Fission Product Transport Processes in Reactor Accidents*, Dubrovnik, Yugoslavia, May 22-26, 1989, Hemisphere Publ. Corp.

Osborne, M. F.; Collins, J. L.; Lorenz, R. A.; Travis, J. R.; Webster, C. S. *Data Summary Report for Fission Product Release Test VI-2*, ORNL/TM-11105, September, 1989

Lorenz, R. A. "Fission Product Release Experiments," presented at Am. Nucl. Soc. Workshop on Safety of Uranium-Aluminum Fuel Reactors, Idaho Falls, ID, March 15, 1989

Osborne, M. F.; Collins, J. L.; Lorenz, R. A.; Travis, J. R.; Webster, C. S.; Yamashita, T. *Data Summary Report for Fission Product Release Test VI-1*, ORNL/TM-11104, June, 1989

Osborne, M. F.; Lorenz, R. A.; Collins, J. L.; Nakamura, T. "Time-Dependent Release of Fission Products from LWR Fuel Under Severe Accident Conditions," presented at Int. Eur. Nucl. Soc./Am. Nucl. Soc. Conf. on Thermal Reactor Safety, Avignon, France, October 3-7, 1988, and published in *Proc. Int. Eur. Nucl. Soc./Am. Nucl. Soc. Conf. on Thermal Reactor Safety*, Avignon, France, October 3-7, 1988, Societe Francaise d'Énergie Nucleaire, 1988, v. 4, pp. 1292-302

Osborne, M. F.; Lorenz, R. A.; Collins, J. L.; Travis, J. R.; Webster, C. S. "Fission Product Release at Severe Accident Conditions," presented at Project Review for Visitors from the Japan Inst. of Nuclear Safety, ORNL, Oak Ridge, TN, February 21, 1989

Osborne, M. F.; Lorenz, R. A.; Hinatsu, Y. "ORNL VI-4 Fission Product Release Tests: Results from Tests VI-3 and VI-4 and Future Plans," presented at Severe Accident Research Program Partners Review Meet., Idaho Falls, ID, April 10-14, 1989

Ludwig, S. B.

McBride, A. F.; Ward, W. M.; Roddy, J. W.; Reich, W. J.; Austin, P. N.; McCarn, L. B.; Ludwig, S. B.; Roussin, R. W. *Compendium of Technical Computer Codes Used in Support of the DOE Office of Civilian Radioactive Waste Management*, ORNL/TM-10919, April, 1989

Mailen, J. C.

Mailen, J. C. "Secondary Purex Solvent Cleanup: Laboratory Development," *Nucl. Technol.* **83**, 182-89 (1988)

Mattus, A. J.

Mattus, A. J.; Kaczmarzsky, M. M.; Cofer, C. K. "Leaching and Comprehensive Regulatory Performance Testing of an Extruded Bitumen Containing a Surrogate, Sodium Nitrate-Based, Low-Level Waste," *Environmental Aspects of Stabilization and Solidification of Hazardous and Radioactive Wastes*, ASTM STP 1033, American Society for Testing and Materials, Philadelphia, Pennsylvania, 1989, pp. 28-39

Swindlehurst, D. P.; Doyle, R. D.; Mattus, A. J. "The Use of Bitumen in the Stabilization of Mixed Wastes," *Environmental Aspects of Stabilization and Solidification of Hazardous and Radioactive Wastes*, ASTM STP 1033, American Society of Testing and Materials, Philadelphia, Pennsylvania, 1989, pp. 21-27

Bouniol, P. E.; Peyre, C. H.; Mattus, A. J.; Pitt, W. W. "Preliminary Results of the Immobilization of Highly Salt-Laden Concentrate in Cement-Based Grout: A Joint DOE/CEA Research Effort," presented at Oak Ridge Model Conf., Oak Ridge, TN, October 3-7, 1988

Mattus, A. J. "A Process for the Unification of Polymers with Cement-Based Materials," presented at Int. Congress on Technology and Technology Exchange, New York, June 28-30, 1989, and to be published in *Proc. Int. Congress on Technology and Technology Exchange*, New York, June 28-30, 1989, International Technology Inst., Pittsburgh, pp. 47-49a

Mattus, A. J. "Process for Impregnating a Concrete or Cement Body with a Polymeric Material," to be published in *Handout at 46th Government Laboratory/Industry Technology Transfer Conf., Atlanta, Dec. 6-7, 1989*

Mattus, A. J.; Gilliam, T. M.; Dole, L. R. *Review of EPA, DOE, and NRC Regulations on Establishing Solid Waste Performance Criteria*, ORNL/TM-9322, July, 1988

Mattus, A. J.; Pitt, W. W.; Bouniol, P. E.; Peyre, C. H. "Wasteform Performance Results of Nitrate and Phosphate-Based Wastes Utilizing Different Types of Cement: A Joint DOE/CEA Research Effort," presented at Oak Ridge Model Conf., Oak Ridge, TN, October 3-7, 1988

McDaniel, E. W.

McDaniel, E. W. "Practical Application of the Use of Cement-Based Materials in Waste Management," presented at Pennsylvania State Univ. Semin., State College, December 13, 1988

McDaniel, E. W.; Tallent, O. K.; Sams, T. L.; Bostick, W. D.; Delzer, D. B. "Basis for Selecting Cement-Based Waste Forms for Immobilizing Radioactive Waste," presented at 12th Int. Symp. on Scientific Basis

for Nuclear Waste Management, Internationales Congress Centrum, Berlin, October 10-13, 1988, and published in *Proc. 12th Mater. Res. Soc. Symp. on Scientific Basis for Nuclear Waste Management*, Internationales Congress Centrum (ICC), Berlin, October 10-13, 1988, Mater. Res. Soc., 1989, v. 127, pp. 421-30

Mrochek, J. E.; Gilliam, T. M.; McDaniel, E. W. "Grout Formulation for the Immobilization of Mixed Organic-Containing Wastes," in *SPECTRUM '86, Proc. Am. Nucl. Soc. Int. Top. Meet. on Waste Management and Decontamination and Decommissioning*, Niagara Falls, NY, September 14-18, 1986 (1988), v. 1, pp. 55-76

Sams, T. L.; McDaniel, E. W. "Development of a Cement-Based Grout for Immobilization of a Low-Level Waste Stream Containing Sodium Sulfate," to be published in *Proc. Waste Management '88 Meet.*, Tucson, AZ, Feb.28-Mar.3, 1988

Spence, R. D.; Bostick, W. D.; McDaniel, E. W.; Gilliam, T. M.; Shoemaker, J. L.; Tallent, O. K.; Morgan, I. L.; Evans-Brown, B. S.; Dodson, K. E. "Immobilization of Technetium in Blast Furnace Slag Grouts," presented at 3rd Int. Conf. on Fly Ash, Silica Fume, Slag, and Natural Pozzolans in Concrete, Trondheim, Norway, June 19-26, 1989, and published in *Proc. 3rd Int. Conf. on Fly Ash, Silica Fume, Slag, and Natural Pozzolans in Concrete*, Vol. 2, pp. 1579-1596

Tallent, O. K.; McDaniel, E. W.; Del Cul, G. B.; Dodson, K. E. "Computer Application to Leach Study Data," in p. 342, *Abstracts of 90th Annu. Meet. Am. Ceram. Soc., Cincinnati*, May 4-5, 1988 (1988)

Tallent, O. K.; McDaniel, E. W.; Del Cul, G. D.; Dodson, K. E.; Trotter, D. R. *Development of Immobilization Technology for Hanford Double-Shell Slurry Feed Waste*, ORNL/TM-10906, August, 1989

McDaniel, E. W., Compiler; Delzer, D. B., Compiler "FUETAP Concrete: A Tailored, Autoclaved Radioactive Waste Host," *Radioactive Waste Forms for the Future, North-Holland*, 1988, pp. 565-88

McGinnis, C. P.

McGinnis, C. P. "Hazardous Waste Technology Program Overview," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988

McGinnis, C. P. "Hazardous Waste Development Demonstration and Disposal Program Overview," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988

McGinnis, C. P.; Eisenhower, B. M.; Reeves, M. E.; DePaoli, S. M.; Stinton, L. H.; Harrington, E. H. *Hazardous Waste Development, Demonstration, and Disposal (HAZWDDD) Program Plan*, ES/ESH-5/V1, February, 1989

McGinnis, C. P.; Eisenhower, B. M.; Reeves, M. E.; DePaoli, S. M.; Stinton, L. H.; Harrington, E. H. *Hazardous Waste Development, Demonstration, and Disposal (HAZWDDD) Program Plan, Executive Summary*, ES/ESH-5/V2, February, 1989

McGinnis, C. P.; Jacobus, R. A.; Bell, L. H. "CEUSP: A Large-Scale Conversion and Solidification of Highly Radioactive Liquid Waste," *Nucl. Chem. Waste Manage.* 8, 115-21 (1988)

McGinnis, C. P.; Pechin, W. H. "HAZWDDD—An Exercise in Corporate Planning," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988, and published in *Proc. DOE Model Conf.*, Oak Ridge TN, October 3-7, 1988, CONF-881054, vol. 5, 1988, v. 5, pp. 1429-38

McTaggart, D. R.

Clark, B. R.; DePaoli, D. W.; McTaggart, D. R.; Patton, B. D. "An On-Line Voltammetric Analyzer for Trace Metals in Wastewater," *Anal. Chim. Acta* **215**, 13-20 (1988)

McWhirter, D. A.

Egan, B. Z.; Lee, D. D.; McWhirter, D. A. "Response to Comments on Solvent Extraction and Recovery of Ethanol from Aqueous Solutions," submitted to *Ind. Eng. Chem. Res.*

Michaels, G. E.

Scott, C. D.; Greenbaum, E.; Michaels, G. E.; Mrochek, J. E.; Newman, E.; Petek, M.; Scott, T. C. "Preliminary Investigation of Possible Low-Temperature Fusion," presented at Workshop on Cold Fusion Phenomena, Santa Fe, NM, May 23-25, 1989; submitted to *J. Fusion Energy*

Morgan, I. L.

Gilliam, T. M.; Morgan, I. L. "Thermal Properties of Cement-Based Grouts," in *Proc. 19th Int. Conf. on Thermal Conductivity*, Cookeville, TN, October 20-23, 1985, Plenum Publ. Corp., 1988, v. 19, pp. 93-108

Gilliam, T. M.; Spence, R. D.; Evans-Brown, B. S.; Morgan, I. L.; Shoemaker, J. L.; Bostick, W. D. "Performance Testing of Blast Furnace Slag for Immobilization of Technetium in Grout," presented at SPECTRUM '88, Int. Top. Meet. on Nuclear and Hazardous Waste Management, Pasco, WA, September 11-15, 1988, and published in *Proc. SPECTRUM '88*, American Nuclear Society, Inc., La Grange Park, IL, 1988, pp. 109-11

Morgan, I. L.; Osborne, S. C.; Spence, R. D.; Gilliam, T. M. "Immobilization of Volatile Organic Compounds in a Cement Waste Form: Sample Preparation Procedures," presented at Meet. Am. Chem. Soc., Div. Environ. Chem., Miami Beach, FL, September 10-15, 1989, and published in *Preprints of Papers Presented at the 198th ACS National Meeting*, vol. 29, no. 2

Spence, R. D.; Bostick, W. D.; McDaniel, E. W.; Gilliam, T. M.; Shoemaker, J. L.; Tallent, O. K.; Morgan, I. L.; Evans-Brown, B. S.; Dodson, K. E. "Immobilization of Technetium in Blast Furnace Slag Grouts," presented at 3rd Int. Conf. on Fly Ash, Silica Fume, Slag, and Natural Pozzolans in Concrete, Trondheim, Norway, June 19-26, 1989, and published in *Proc. 3rd Int. Conf. on Fly Ash, Silica Fume, Slag, and Natural Pozzolans in Concrete*, Vol. 2, pp. 1579-1596

Spence, R. D.; Gilliam, T. M.; Morgan, I. L.; Osborne, S. C. "Immobilization of Volatile Organic Compounds in Commercial Cement-Based Waste Forms," news release for ACS News Service

Morris, M. I.

Butterworth, G. E.; Morris, M. I.; Lee, D. W. *Radioactive Waste Management Implementation Plan for Paducah Gaseous Diffusion Plant*, ES/ESH-9, KY/H-86, May, 1989

Butterworth, G. E.; Morris, M. I.; Lee, D. W. *Radioactive Waste Management Implementation Plan for Portsmouth Gaseous Diffusion Plant*, ES/ESH-10, POE F-2011, May, 1989

Morris, M. I. *K-25 Privatization Plan*, K/HS-278, April, 1989

Jolley, R. L.; Hollenbeck, P. E.; Kennerly, J. M.; Morris, M. I.; Singh, S. P. N. "Evaluation of Selected Emerging Waste Treatment and Disposal Methods," presented at Meet. Am. Chem. Soc., Miami Beach, FL, September 10-15, 1989, and to be published in *Proc. Meet. Am. Chem. Soc.*,

Jolley, R. L.; Hollenbeck, P. E.; Kennerly, J. M.; Morris, M. I.; Singh, S. P. N. "Evaluation of Selected Emerging Waste Treatment and Disposal Methods," presented at Meet. Am. Chem. Soc., Miami Beach, FL, September 10-15, 1989

Morris, M. I.; Dole, L. R. "Treatment of Priority Liquid Waste Streams on the Oak Ridge Reservation," presented at Summer Natl. Meet. Am. Inst. Chem. Eng., Denver, August 21-24, 1988, and at 49th Int. Water Conf., Pittsburgh, October 24-26, 1988

Morris, M. I.; Dole, L. R.; Spence, R. D.; Gilliam, T. M. "Immobilization of Volatile Organics in Cement-Based Grouts," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988

Morris, M. I.; Jolley, R. L.; Singh, S. P. N. "The Waste Management Technology Center's Role in Waste Management Technology Applications for Martin Marietta Energy Systems, Inc.," presented at Weld. Test. Tech. Conf., Knoxville, TN, February 1989

Morris, M. I.; Gilliam, T. M.; Kasten, J. L. "Selection and Treatment of Priority Liquid Waste Streams on the Oak Ridge Reservation," presented at 91st Annu. Meet. Am. Ceram. Soc., Indianapolis, April 23-27, 1989, and at 49th Int. Water Conf., Pittsburgh, October 24-26, 1988; published in *Proc. 49th Int. Water Conf.*, Pittsburgh, October 24-26, 1988 (1988), pp. 469-73

Morris, M. I.; Spence, R. D.; Gilliam, T. M.; Dole, L. R. "A Program to Establish In Situ Immobilization as a Remedial Action Option for Wastes Containing Volatile Organic Compounds (VOC)," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988, and published in *Proc. DOE Model Conf.*, CONF-881054, vol. 5, 1988, pp. 1501-08

Mrochek, J. E.

Mrochek, J. E.; Gilliam, T. M.; McDaniel, E. W. "Grout Formulation for the Immobilization of Mixed Organic-Containing Wastes," in *SPECTRUM '86, Proc. Am. Nucl. Soc. Int. Top. Meet. on Waste Management and Decontamination and Decommissioning*, Niagara Falls, NY, September 14-18, 1986 (1988), v. 1, pp. 55-76

Mrochek, J. E.; Krishnan, R. P.; Graves, R. L. "Characterization and Upgrading of Coal Liquids Derived from Mild Gasification Processes," presented at Meet. Am. Chem. Soc., Miami Beach, FL, September 10-15, 1989, and to be published in *Proc. Meet. Am. Chem. Soc.*, Miami Beach, FL, Sept. 10-15, 1989, Am. Chem. Soc., vol. 33, nos. 3-4, pp. 1227-1237

Mrochek, J. E.; Scott, C. D. "Upgrading of Condensibles from Coal Devolatilization," presented at Oil Shale, Tar Sand, and Mild Gasification Contractors Review Meet., Morgantown, WV, July 19-21, 1988, and published in *Proc. 3rd Annu. Oil Shale, Tar Sand, and Mild Gasification Contractors Review Meet.*, Morgantown, WV, July 19-21, 1988, DOE/METC-88/6098, 1988, pp. 114-23

Scott, C. D.; Greenbaum, E.; Michaels, G. E.; Mrochek, J. E.; Newman, E.; Petek, M.; Scott, T. C. "Preliminary Investigation of Possible Low-Temperature Fusion," presented at Workshop on Cold Fusion Phenomena, Santa Fe, NM, May 23-25, 1989; submitted to *J. Fusion Energy*

Nakamura, T.

Osborne, M. F.; Lorenz, R. A.; Collins, J. L.; Nakamura, T. "Time-Dependent Release of Fission Products from LWR Fuel Under Severe Accident Conditions," presented at Int. Eur. Nucl. Soc./Am. Nucl. Soc. Conf. on Thermal Reactor Safety, Avignon, France, October 3-7, 1988

Nehls, J. R., Jr.

Joy, D. S.; Nehls, J. R., Jr.; Harrison, I. G.; Miller, C.; Vogel, L. W.; Martin, J. D.; Capone, R. L.; Dougherty, L. "Modeling the Design and Operations of the Civilian Radioactive Waste Processing System," presented at the Waste Management Meeting, Tucson, Arizona, February 26- March 2, 1989, and published in *Waste Management 89*, vol. 1, p. 173

Newman, E.

Scott, C. D.; Greenbaum, E.; Michaels, G. E.; Mrochek, J. E.; Newman, E.; Petek, M.; Scott, T. C. "Preliminary Investigation of Possible Low-Temperature Fusion," presented at Workshop on Cold Fusion Phenomena, Santa Fe, NM, May 23-25, 1989; to be published in *Proc. Workshop on Cold Fusion Phenomena*; and submitted to *J. Fusion Energy*

Notz, K. J.

Moore, R. S.; Jones, K. E.; Moisson, I. R.; Rhyne, B. T.; Notz, K. J. "Characteristics of LWR Spent Fuel," presented at Am. Nucl. Soc. Int. Top. Meet. on Nuclear and Hazardous Waste Management, Tri-Cities, WA, September 11-15, 1988

Moore, R. S.; Notz, K. J. *Physical Characteristics of GE BWR Fuel Assemblies*, ORNL/TM-10902, June, 1989

Notz, K. J. "Characteristics of Spent Nuclear Fuel," presented at 4th Int. Symp. on Ceramics in Nuclear Waste Management, Indianapolis, April 23-27, 1989, and to be published in *Proc. 4th Int. Symp. on Ceramics in Nuclear Waste Management*, Indianapolis, Apr. 23-27, 1989, Am. Chem. Soc.

Notz, K. J. "Characteristics of Spent Fuel and High-Level Waste," submitted to *Radioact. Waste Manage. Nucl. Fuel Cycle*

Notz, K. J. "Characteristics of Spent Fuel, High-Level Waste, and Other Radioactive Wastes Which May Require Long-Term Isolation," presented at Spectrum '88, Tri-Cities, WA, September 11-15, 1988, and published in *Proc. Spectrum '88*, Tri-Cities, WA, September 11-15, 1988, Am. Nucl. Soc., 1988, pp. 145-48

Notz, K. J.; Burwinkle, T. W. "Decommissioning the Molten Salt Reactor Experiment," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988

Notz, K. J.; Haas, P. A. "Properties and Thermal Decomposition of the Double Salts of Uranyl Nitrate-Ammonium Nitrate," *Thermochim. Acta* 155 283-295 (1989)

Notz, K. J. "Characteristics of Activated Metal Hardware from Light-Water Reactors: A Status Report," Spent Fuel Management Seminar VI, INMM, January 11-13, 1989, Washington, DC

Moore, R. S.; Williamson, D. A.; Notz, K. J. *A Classification Scheme for LWR Fuel Assemblies*, ORNL/TM-10901, November, 1988

Johnson, E. R.; Notz, K. J. *Shipping and Storage Cask Data for Spent Nuclear Fuel*, ORNL/TM-11008, November, 1988

Phung, D. L.; Lawson, C. G.; Notz, K. J. "Characteristics of Defective LWR Spent Fuel," presented at Am. Nucl. Soc. Int. Top. Meet. on Nuclear and Hazardous Wastes Management, Tri-Cities, WA, September 11-15, 1988

Osborne-Lee, I. W.

Ferrada, J. J.; Osborne-Lee, I. W. "Applications of Artificial Intelligence to Chemical Engineering Processes," *Chem. Process.* **52**, 23-33 (1989)

Osborne-Lee, I. W. "Calculation of Hamaker Constants from Extensive Optical Data," *Particles on Surfaces 1: Detection, Adhesion, and Removal*, Plenum Press, New York, 1988, v. 1, pp. 77-90

Osborne-Lee, I. W.; Rodgers, B. R. "A Natural Language Interface to Relational Data Bases for Waste Management," presented at Summer Meet. Am. Inst. Chem. Eng., Denver, August 20-24, 1988

Rivera, A. L.; Osborne-Lee, I. W.; DePaoli, S. M. "Systems Analysis Support to the Waste Management Technology Center," in *Proc. DOE Model Conf.*, Oak Ridge TN, October 3-7, 1988, CONF-881054, vol. 1, 1988, v. 1, pp. 81-88

Wright, T. C.; Osborne-Lee, I. W.; DePaoli, S. M.; Rivera, A. L. "Computer-Based LLWDDD Data Base Demonstration," presented at WMTC 2nd Workshop on Leaching Tests, Oak Ridge, TN, October 25-26, 1988

Osborne, M. F.

Osborne, M. F.; Collins, J. L.; Lorenz, R. A.; Travis, J. R.; Webster, C. S. *Data Summary Report for Fission Product Release Test VI-2*, ORNL/TM-11105, September, 1989

Kress, T. S.; Lorenz, R. A.; Nakamura, T.; Osborne, M. F. "Correlation of Recent Fission Product Release Data," presented at ICHMT Int. Semin. on Fission Product Transport Processes in Reactor Accidents, Dubrounik, Yugoslavia, May 22-26, 1989, and to be published in *Proc. ICHMT Int. Semin. on Fission Product Transport Processes in Reactor Accidents*, Dubrounik, Yugoslavia, May 22-26, 1989, Hemisphere Publ. Corp.

Osborne, M. F.; Collins, J. L.; Lorenz, R. A.; Travis, J. R.; Webster, C. S.; Yamashita, T. *Data Summary Report for Fission Product Release Test VI-1*, ORNL/TM-11104, June, 1989

Osborne, M. F.; Lorenz, R. A.; Collins, J. L.; Nakamura, T. "Time-Dependent Release of Fission Products from LWR Fuel Under Severe Accident Conditions," presented at Int. Eur. Nucl. Soc./Am. Nucl. Soc. Conf. on Thermal Reactor Safety, Avignon, France, October 3-7, 1988, and published in *Proc. Int. Eur. Nucl. Soc./Am. Nucl. Soc. Conf. on Thermal Reactor Safety*, Avignon, France, October 3-7, 1988, Societe Francaise d'Energie Nucleaire, 1988, v. 4, pp. 1292-302

Osborne, M. F.; Lorenz, R. A.; Collins, J. L.; Travis, J. R.; Webster, C. S. "Fission Product Release at Severe Accident Conditions," presented at Project Review for Visitors from the Japan Inst. of Nuclear Safety, ORNL, Oak Ridge, TN, February 21, 1989

Osborne, M. F.; Lorenz, R. A.; Hinatsu, Y. "ORNL VI-4 Fission Product Release Tests: Results from Tests VI-3 and VI-4 and Future Plans," presented at Severe Accident Research Program Partners Review Meet., Idaho Falls, ID, April 10-14, 1989

Webster, C. S.; Osborne, M. F. *The Use of Fiber Optics for Remote Temperature Measurement in Fission Product Release Test*, ORNL/TM-10366, April, 1989

Osborne, S. C.

Morgan, I. L.; Osborne, S. C.; Spence, R. D.; Gilliam, T. M. "Immobilization of Volatile Organic Compounds in a Cement Waste Form: Sample Preparation Procedures," presented at Meet. Am. Chem. Soc., Div. Environ. Chem., Miami Beach, FL, September 10-15, 1989

Spence, R. D.; Gilliam, T. M.; Morgan, I. L.; Osborne, S. C. "Immobilization of Volatile Organic Compounds in Commercial Cement-Based Waste Forms," news release for ACS News Service

Ottinger, C. L.

Collins, E. D.; Bigelow, J. E.; Ottinger, C. L.; Burney, G. A. "Recovery of Selected Elements from Irradiated Targets," to be published in *Nuclear Waste Management Technology*, vol. 2, Harwood Academic Publ.

Ottinger, C. L. "Radioactive Material Shipment," presented at Meet. Health Phys. Soc., East Tennessee Chapter, Knoxville, TN, August 24, 1989

Pattison, W. L.

Collins, J. L.; Pattison, W. L.; Kelmers, A. D. "Thirty-Gallon-Scale of Uranium from Production or Trash Type Radioactive Wastes from the Y-12 Plant," poster session at the Chemical Technology Division Information Meeting, December 8, 1988

Kelmers, A. D.; Collins, J. L.; Pattison, W. L. "Thirty-Gallon-Scale Leaching of Uranium from Production—or Trash Type Radioactive Wastes," presented at the Chemical Technology Division Information Meeting, December 8, 1988

Collins, J. L.; Pattison, W. L.; Kelmers, A. D. "Thirty-Gallon-Scale Leaching of Uranium from Production-or Trash-Type Low-Level Radioactive Wastes," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988, and at Pu/U Recovery Operations Conf., Idaho Falls, ID, October 4-6, 1988

Walker, B. A., Editor; Saylor, R. E., Editor; Ketelle, R. H.; Cada, G. F.; Clapp, R. B.; Collins, J. L.; Daniels, K. L.; Griest, W. H.; Kelmers, A. D.; Kroodsma, R. L.; Lee, S. Y.; Lee, R. R.; McMahon, J. M.; Parr, P. D.; Pattison, W. L.; Tomkins, B. A. *Data Package for the Low-Level Waste Disposal Development and Demonstration Program Environmental Impact Statement*, sects. 1-7, appendixes A-O, ORNL/TM-10939/V1 and 2, September, 1988

Patton, B. D.

Clark, B. R.; DePaoli, D. W.; McTaggart, D. R.; Patton, B. D. "An On-Line Voltammetric Analyzer for Trace Metals in Wastewater," *Anal. Chim. Acta* **215**, 13-20 (1988)

Thompson, W. T.; Patton, B. D. "Treatment of Dilute Process Wastewaters from ORNL," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988

Turner, D. W.; Patton, B. D. "The ORNL Waste Handling and Packaging Plant," presented at Spectrum '88, Am. Nucl. Soc. Int. Top Meet. on Nuclear and Hazardous Waste Management, Pasco, WA, September 11-15, 1988

McNeese, L. E.; Berry, J. B.; Butterworth, G. E., III; Collins, E. D.; Monk, T. H.; Patton, B. D.; Snider, J. W. *Overall Strategy and Program Plan for Management of Radioactively Contaminated Liquid Wastes and Transuranic Sludges at the Oak Ridge National Laboratory*, ORNL/TM-10757, December, 1988

Pechin, W. H.

McGinnis, C. P.; Pechin, W. H. "HAZWDDD—An Exercise in Corporate Planning," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988, and published in *Proc. DOE Model Conf.*, Oak Ridge TN, October 3-7, 1988, CONF-881054, vol. 5, 1988, pp. 1429-38

Perona, J. J.

Perona, J. J.; Basaran, O. A.; Byers, C. H. "The Effect of Fluid Properties on Electric-Field-Enhanced Drop Formation," AICHE Annual Meeting, Washington, DC, December 1, 1988

Counce, R. M.; Hill, B.; Perona, J. J. "Fundamentals of Industrial Gas Absorption and Stripping," presented at AICHE Summer Meeting, Philadelphia, August, 1989

Byers, C. H.; Perona, J. J. "Drop Formation from an Orifice in an Electric Field," *AIChE J.* **34**, 1577-80 (1988)

Perona, J. J.; Gibson, M. R.; Byers, C. H. "Vacuum Sorption Pumping at Cryogenic Temperatures of Argon and Oxygen on Molecular Sieves," presented at NATO ASI on Adsorption: Science and Technology, Vimeiro, Portugal, July 17-20, 1988, and submitted to *Sep. Sci. Technol.*

Phillips, B. P.

Wike, J. S.; Guyer, C. E.; Ramey, D. W.; Phillips, B. P. "Chemistry for Commercial-Scale Productions of Yttrium-90 for Medical Research," presented at the American Chemical Society (Pacific Basin) meeting, Honolulu, Hawaii, December 19, 1989; to be published in *Int. J. Radiat. Appl. Instrum.*

Petek, M.

Scott, C. D.; Greenbaum, E.; Michaels, G. E.; Mrochek, J. E.; Newman, E.; Petek, M.; Scott, T. C. "Preliminary Investigation of Possible Low-Temperature Fusion," presented at Workshop on Cold Fusion Phenomena, Santa Fe, NM, May 23-25, 1989; submitted to *J. Fusion Energy*

Pitt, W. W.

Bouniol, P. E.; Peyre, C. H.; Mattus, A. J.; Pitt, W. W. "Preliminary Results of the Immobilization of Highly Salt-Laden Concentrate in Cement-Based Grout: A Joint DOE/CEA Research Effort," presented at Oak Ridge Model Conf., Oak Ridge, TN, October 3-7, 1988

Mattus, A. J.; Pitt, W. W.; Bouniol, P. E.; Peyre, C. H. "Wasteform Performance Results of Nitrate and Phosphate-Based Wastes Utilizing Different Types of Cement: A Joint DOE/CEA Research Effort," presented at Oak Ridge Model Conf., Oak Ridge, TN, October 3-7, 1988

Pitt, W. W.; Strandberg, G. W.; Donaldson, T. L.; Bolla, L. L. "Biodegradation of Trichloroethylene Contaminated Groundwater," presented at Conf. on Biotechnology Applications in Hazardous Waste Treatment, Longboat Key, FL, October 30-November 4, 1988, and to be published in *Proc. Conf. on Biotechnology Applications in Hazardous Waste Treatment*, Longboat Key, FL, Oct.30-Nov.4, 1988

Pope, R. B.

O'Sullivan, R. A.; Pettersson, B. G.; Pope, R. B. "The Continuous Review and Periodic Revision Process for the International Atomic Energy Agency's Regulations for the Safe Transport of Radioactive Materials—A Status Report," presented at PATRAM '89 Conf., Washington, DC, June 12-16, 1989, and published in *Proceedings of PATRAM '89*, pp. 1829-1836, June, 1989

Pope, R. B.; Rawl, R. R. "Radioactive Materials Packaging Standards and Regulations—Making Sense of It All," presented at Am. Soc. Mech. Eng. Pressure Vessel and Piping Conf., Honolulu, July 23-27, 1989, and published in *Advances in Packaging and Transportation of Radioactive Materials*, ASME, PVP, Vol. 164, pp. 1-8, July, 1989

Pruett, D. J.

Blahe, S. L.; Anderson, C. J.; Choppin, G. R.; Pruett, D. J. "Evaluation of POCl_3 as a Solvent for Complexation Studies," *Inorg. Chim. Acta* **156**, 37-9 (1989)

Ensor, D. D.; Nicks, M.; Pruett, D. J. "Extraction of Uranyl Ions by Synergistic Mixtures of Thenoyltrifluoroacetone and Macrocyclic Donors^{a-b}," *Solvent Extr. Ion Exch.* **6**, 621-29 (1988)

Ensor, D. D.; Pruett, D. J.; Nicks, M. "Synergistic Extraction of Trivalent Actinides and Lanthanides Using HTTA and an Aza-Crown Ether," *Sep. Sci. Technol.* **23**, 1345-53 (1988)

Pruett, D. J. "Extraction Chemistry of Fission Products," *Science Technology of Tributyl Phosphate*, W. W. Schulz et al., eds., vol III., CRC Press, Boca Raton, Florida, 1990, pp. 81-121

Ramey, D. W.

Kobisk, E. H.; Ramey, D. W.; Aaron, W. S.; Tompkins, J. A.; Haff, K. W.; DeVore, J. R.; Adair, H. L. "Tritium-Processing Operations at the Oak Ridge National Laboratory with Emphasis on Safe-Handling Practices," presented at Int. Nucl. Target Dev. Soc. and IAEA-INDC Conf., Darmstadt, Federal Republic of Germany, September 5-9, 1988, and published in *Nuc. Instrum. Methods Phys. Res. A* **282**, 329-40 (1989)

Wike, J. S.; Guyer, C. E.; Ramey, D. W.; Phillips, B. P. "Chemistry for Commercial-Scale Productions of Yttrium-90 for Medical Research," presented at the American Chemical Society (Pacific Basin) meeting, Honolulu, Hawaii, December 19, 1989; to be published in *Int. J. Radiat. Appl. Instrum.*

Ratledge, J. E.

Rawl, R. R.; Grubb, R. G.; Ratledge, J. E.; Jones, R. H. "Transportation Operations Experience Involving Suspected Leaking Cesium Capsules," presented at 9th Int. Symp. on Packaging and Transportation of Radioactive Materials (PATRAM 89), Washington, DC, June 11-16, 1989, and published in *Proc. 9th Int. Symp. on Packaging and Transportation Materials (PATRAM '89)*, CONF-890631, vol. 1, pp. 238-246, 1989

Rawl, R. R.

Pope, R. B.; Rawl, R. R. "Radioactive Materials Packaging Standards and Regulations—Making Sense of It All," presented at Am. Soc. Mech. Eng. Pressure Vessel and Piping Conf., Honolulu, July 23-27, 1989, and to be published in *Proc. Am. Soc. Mech. Eng. Pressure Vessel and Piping Conf.*, Honolulu, July 23-27, 1989, ASME, vol. 164, pp. 1-8

Rawl, R. R.; Grubb, R. G.; Ratledge, J. E.; Jones, R. H. "Transportation Operations Experience Involving Suspected Leaking Cesium Capsules," presented at 9th Int. Symp. on Packaging and Transportation of Radioactive Materials (PATRAM 89), Washington, DC, June 11-16, 1989, and published in *Proc. 9th Int. Symp. on Packaging and Transportation Materials (PATRAM '89)*, CONF-890631, vol. 1, pp. 238-246, 1989

Reeves, M.

Woodward, J.; Reeves, M.; Greenbaum, E.; Allison, D. "Effect of Exogenously Added Manganese Chloride on the Operational Stability of the Chloroplast-Ferredoxin-Hydrogenase System," *Enzyme Microb. Technol.* **10**, 123-28 (1988)

Reeves, M. E.

Graves, D. A.; Reeves, M. E.; Greenbaum, E. "Establishment of Control Parameters for In Situ, Automated Screening of Sustained Hydrogen Photoproduction by Individual Algal Colonies," *Plant Physiol.* **87**, 603-08 (1988)

McGinnis, C. P.; Eisenhower, B. M.; Reeves, M. E.; DePaoli, S. M.; Stinton, L. H.; Harrington, E. H. *Hazardous Waste Development, Demonstration, and Disposal (HAZWDDD) Program Plan, ES/ESH-5/V1*, February, 1989

McGinnis, C. P.; Eisenhower, B. M.; Reeves, M. E.; DePaoli, S. M.; Stinton, L. H.; Harrington, E. H. *Hazardous Waste Development, Demonstration, and Disposal (HAZWDDD) Program Plan, Executive Summary, ES/ESH-5/V2*, February, 1989

Reeves, M. E. "For Cleanup of Toxic PCBs, an Old Approach to a New Problem," *The Oak Ridger*

Rhyne, B. T.

Moore, R. S.; Jones, K. E.; Moisson, I. R.; Rhyne, B. T.; Notz, K. J. "Characteristics of LWR Spent Fuel," presented at Am. Nucl. Soc. Int. Top. Meet. on Nuclear and Hazardous Waste Management, Tri-Cities, WA, September 11-15, 1988

Rivera, A. L.

Chang, S. -Y.; Rivera, A. L. "A System Optimization Model for Low-Level Nuclear Waste Disposal," presented at 7th Int. Conf. on Mathematical and Computer Modelling, Chicago, August 2-5, 1989, and to be published in *Proc. 7th Int. Conf. on Mathematical and Computer Modelling*, Chicago, Aug. 2-5, 1989

DePaoli, S. M.; Wright, T. C.; Rivera, A. L. "Computer-Based HAZWDDD Data Base Demonstration," presented at WMTC 2nd Workshop on Leaching Tests, Oak Ridge, TN, October 25-26, 1988

Jolley, R. L.; Rivera, A. L. "Continuous Monitoring, Automated Analysis, and Sampling Procedures," *Water Pollut. Control Assoc.* vol. 60, no. 6, 1988, pp. 799-801; also vol. 61, no. 6, June 1989, pp. 782-785

Jolley, R. L.; Rivera, A. L.; Fox, E. C.; Hyfantis, G. J.; McBrayer, J. F. "Strategic Planning for Waste Management: Characterization of Chemically and Radioactively Hazardous Waste and Treatment, Storage, and Disposal Capabilities for Diverse and Varied Multisite Operations," in *Proc. Waste Management '88*, Tucson, AZ, February 28-March 3, 1988, Univ. of Arizona, Tucson, 1988, v. 1, pp. 951-53

Rivera, A. L. "Systems Analysis Considerations in Artificial Intelligence/Expert Systems Technology Applications," presented at Summer Natl. Meet. Am. Inst. Chem. Eng., Philadelphia, August 20-23, 1989

Rivera, A. L. *Diffusion of Waste Technologies and Its Potential Impact on Research and Development*, ORNL/TM-10856, July, 1988

Rivera, A. L.; Alsharif, M.; Phung, D. L.; Dole, L. R. "An Integrated Waste Facility Performance Model," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988, and to be published in *Abstract Booklet, DOE Model Conf., Oak Ridge, TN, Oct. 3-7, 1988*

Alsharif, M.; Phung, D. L.; Rivera, A. L.; Dole, L. R. "Survey of Computer Codes Applicable to Waste Facility Performance Evaluations," DOE Model Conference, Oak Ridge, Tennessee, October 3-7, 1988, and published in *Proceedings of the DOE Model Conference*, CONF-881054, vol. 1, 1988, pp. 95-106

Rivera, A. L.; DePaoli, S. M. "Engineered Waste Disposal: Future Prospects and Challenges," presented at 197th Natl. Meet. Am. Chem. Soc., Dallas, April 9-14, 1989, in *Reprints of Papers Presented at 197th Am. Chem. Soc. Natl. Meet.*, Dallas, April 9-14, 1989, Am. Chem. Soc., Div. Environ. Chem., 1989, v. 1, pp. 136-37

Rivera, A. L.; Osborne-Lee, I. W.; DePaoli, S. M. "Systems Analysis Support to the Waste Management Technology Center," in *Proc. DOE Model Conf.*, Oak Ridge TN, October 3-7, 1988, CONF-881054, Vol.1, 1988, v. 1, pp. 81-88

Wright, T. C.; Osborne-Lee, I. W.; DePaoli, S. M.; Rivera, A. L. "Computer-Based LLWDDD Data Base Demonstration," presented at WMTC 2nd Workshop on Leaching Tests, Oak Ridge, TN, October 25-26, 1988

Robinson, S. M.

Berry, J. B.; Brown, C. H. Jr.; Fowler, V. L.; Robinson, S. M. "Treatment of ORNL Liquid Low-Level Waste," presented to the Division of Environmental Chemistry, American Chemical Society, Toronto, Canada, June 5-11, 1988

Berry, J. B.; Brown, C. H., Jr.; Fowler, V. L.; Robinson, S. M. "Treatment of ORNL Process Waste," presented at Spectrum '88, Int. Top. Meet. on Nuclear and Hazardous Waste Management, Pasco, WA, September 11-15, 1988, and published in *Proc. Spectrum '88*, 1988, pp. 416-20

Berry, J. B.; Robinson, S. M. "Improved Treatment of Slightly Radioactive Process Waste: Reduced Generation of Waste," presented at Spectrum '88, Nuclear and Hazardous Waste Management Int. Top. Meet., Pasco, WA, September 11-15, 1988, and published in *Proceedings of the Ninth Annual DOE Low-Level Waste Management Conference*, vol. 6, pp. 36-43, 1988

Hall, R.; Brown, C. H., Jr.; Robinson, S. M.; Hewitt, J. D. "Operation of a Mobile Pilot-Scale Continuous Countercurrent Ion Exchange System for Treatment of Low-Level Radioactive Wastewater," presented at HAZTECH Int. '89 Conf., San Francisco, September 26-October 1, 1989, and to be published in *Proc. HAZTECH Int. '89 Conf.*, San Francisco, Sept. 26-Oct. 1, 1989

Robinson, S. M. "Impacts of DOE Order 5820.2A on Oak Ridge National Laboratory," presented at Environmental Engineering Dep. Meet., Univ. of Tennessee, Oak Ridge, TN, April 10, 1989

Robinson, S. M.; Begovich, J. M.; Scott, C. B. "Low-Activity-Level Process Wastewaters: Treatment by Chemical Precipitation and Ion Exchange," *J. Water Pollut. Control Fed.* 60, 2120-27 (1988)

Robinson, S. M.; Counce, R. M.; Smith, G. V. "An Experimental Study of Important Parameters for the Operation of a Continuous-Flow Fluidic Pump," *J. Fluid Control* 19, 7-21 (1988)

Roddy, J. W.

Hermann, O. W.; Parks, C. V.; Renier, J. P.; Roddy, J. W.; Ashline, R. C.; Wilson, W. B.; LaBauve, R. J. *Multicode Comparison of Selected Source-Term Computer Codes*, ORNL/CSD/TM-251, April, 1989

Kelmers, A. D.; Shappert, L. B.; Roddy, J. W.; Hermann, O. W.; Parks, C. V. *Identification and Evaluation of Radionuclide Generation/Depletion Codes for Potential Use by the Department of Energy's Office of Civilian Radioactive Waste Management*, ORNL/TM-10404, February, 1989

McBride, A. F.; Ward, W. M.; Roddy, J. W.; Reich, W. J.; Austin, P. N.; McCarn, L. B.; Ludwig, S. B.; Roussin, R. W. *Compendium of Technical Computer Codes Used in Support of the DOE Office of Civilian Radioactive Waste Management*, ORNL/TM-10919, April, 1989

Riggle, K. J.; Roddy, J. W. *A Preliminary Assessment of Selected Atmospheric Dispersion, Food-Chain Transport, and Dose-To-Man Computer Codes for Use by the DOE Office of Civilian Radioactive Waste Management*, ORNL/TM-10915, February, 1989

Rodgers, B. R.

Ferrada, J. J.; Mashburn, S. A.; Rodgers, B. R. "A PROLOG Registry of Experts," in *Abstracts, Spring Natl. Meet. American Inst. of Chemical Engineers, New Orleans, March 6-10, 1988* (1988)

Ferrada, J. J.; Stark, L.; Rodgers, B. R. "Developing Expert Systems for Hazardous and Low-Level Radioactive Waste Management," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988, and published in *DOE Model Conf. Proceedings*, vol. 1, pp. 23-32

Osborne-Lee, I. W.; Rodgers, B. R. "A Natural Language Interface to Relational Data Bases for Waste Management," presented and published in *Abstracts, Summer Meet. Am. Inst. Chem. Eng., Denver, August 20-24, 1988*

Rodgers, B. R. "Advanced Power Generation—Progress Report-I," presented at IEA Coal Research Technical Committee Meeting, Chester, England, December 1, 1988

Rodgers, B. R. "Advanced Power Generation—Progress Report-II, presented at IEA Coal Research Technical Committee Meeting, Madrid, Spain, June, 1989

Sams, T. L.

Sams, T. L.; Gilliam, T. M. "Systematic Approach for the Design of Pumpable, Cement-Based Grouts for Immobilization of Hazardous Wastes," *Environmental Aspects of Stabilization and Solidification of Hazardous and Radioactive Wastes*, ASTM STP 1033, P. L. Cote and T. M. Gilliam, eds., American Society for Testing and Materials, Philadelphia, 1989

Del Cul, G. D.; Tallent, O. K.; Sams, T. L. "Monitor for Waste Immobilized in Cement-Based Materials", presented at Technology Transfer Workshop, Oak Ridge, TN, March 23, 1989

McDaniel, E. W.; Tallent, O. K.; Sams, T. L.; Bostick, W. D.; Delzer, D. B. "Basis for Selecting Cement-Based Waste Forms for Immobilizing Radioactive Waste", presented at 12th Int. Symp. on Scientific Basis for Nuclear Waste Management, Internationales Congress Centrum, Berlin, October 10-13, 1988, and published in *Proc. 12th Mater. Res. Soc. Symp. on Scientific Basis for Nuclear Waste Management*, Internationales Congress Centrum (ICC), Berlin, October 10-13, 1988, *Mater. Res. Soc.*, 1989, v. 127, pp. 421-30

Sams, T. L.; McDaniel, E. W. "Development of a Cement-Based Grout for Immobilization of a Low-Level Waste Stream Containing Sodium Sulfate," to be published in *Proc. Waste Management '88 Meet.*, Tucson, AZ, Feb. 28-Mar. 3, 1988

Scott, C. D.

Faison, B. D.; Scott, C. D. "Biosolubilization of Coal in Aqueous and Nonaqueous Media," presented at Int. Symp. on Chemistry of Microbial Coal Conversion and Bioprocessing of Coal, Am. Chem. Soc. Meet., Los Angeles, September 25-30, 1988, and to be published in *Proc. Int. Symp. on Chemistry of Microbial Coal Conversion and Bioprocessing of Coal*

Faison, B. D.; Watson, J. S.; Cancel, C. A.; Scott, C. D. "Applications of Biotechnology in Waste Management for the Nuclear Industry," presented at Annu. Meet. Air Waste Management Assoc., Southern Section, Diamondhead, MS, September 20-22, 1989

Godia, F.; Adler, H. I.; Davison, B. H.; Scott, C. D. "Use of Immobilized Microbial Membrane Fragments to Reduce Oxygen Content and Enhance the Acetone-Butanol Fermentation," submitted to *Enzyme Microb. Technol.*

Mrochek, J. E.; Scott, C. D. "Upgrading of Condensibles from Coal Devolatilization," presented at Oil Shale, Tar Sand, and Mild Gasification Contractors Review Meet., Morgantown, WV, July 19-21, 1988, and published in *Proc. 3rd Annu. Oil Shale, Tar Sand, and Mild Gasification Contractors Review Meet.*, Morgantown, WV, July 19-21, 1988, DOE/METC-88/6098, 1988, pp. 114-23

Scott, C. D. "Bioprocessing Research for Energy Applications: Needs and Opportunities," *Biotechnol. Prog.*, vol. 5, June 1989

Scott, C. D. "Advanced Bioprocessing Concepts for the Production of Fuels and Chemicals," presented at Semin., Chemistry Dep., North Carolina A&T Univ., Greensboro, March 16, 1989; presented at Symp. on Biotechnology for Aerospace Applications, U.S. Air Force Academy, Colorado Springs, CO, March 1-2, 1989; and to be published in *Proc. Symp. on Biotechnology for Aerospace*

- Scott, C. D. "Advanced Columnar Bioreactor Systems," presented at Lehigh Univ., Bethlehem, PA, October 19, 1988
- Scott, C. D. "The Impact of Bioprocessing Concepts on Fossil Energy: Potentials and Speculations," presented at DOE Workshop on Biological Processing of Fossil Fuels, Tyson's Corner, VA, August 8-10, 1989
- Scott, C. D. "Introduction to the Proceedings of the Ninth Symposium on Biotechnology for Fuels and Chemicals," *Appl. Biochem. Biotechnol.* **17**, 1-3 (1988)
- Scott, C. D. "Corn Products as Chemical Feedstocks: The Corn Refinery," in *Proc. 1st Annu. Corn Utilization Conf.*, St. Louis, June 11-12, 1987, Natl. Corn Growers Assoc., 1989, pp. 286-99
- Scott, C. D.; Davison, B. H. "Ethanol Production from Industrial Feedstocks by Immobilized *Zymomonas mobilis* in a Fluidized-Bed Bioreactor," presented at 8th Int. Biotechnology Symp., Paris, July 17-22, 1988
- Scott, C. D.; Faison, B. D. "The Conversion of Coal to Liquids and Gasses by Advanced Bioprocessing Systems," presented at Coal—Targets of Opportunity Workshop, Washington, DC, July 12-13, 1988, and to be published in *Proc. Workshop on Coal—Targets of Opportunity*, Washington, DC, July 12-13, 1988
- Scott, C. D.; Faison, B. D. "Biological Solubilization of Coal in Aqueous and Nonaqueous Media," in *Proc. Direct Liquefaction Contractors' Review Meet.*, Pittsburgh, October 6-8, 1987, DOE, 1988, pp. 324-39
- Scott, C. D.; Gaden, E. L., Jr.; Humphrey, A. E.; Carta, G.; Kirwan, D. J. *Bioprocessing Research for Energy Applications*, ORNL/TM-11054, April, 1989
- Scott, C. D.; Greenbaum, E.; Michaels, G. E.; Mrochek, J. E.; Newman, E.; Petek, M.; Scott, T. C. "Preliminary Investigation of Possible Low-Temperature Fusion," presented at Workshop on Cold Fusion Phenomena, Santa Fe, NM, May 23-25, 1989; submitted to *J. Fusion Energy*
- Scott, C. D.; Greenbaum, E.; Wyman, C. E., eds. "Proceedings of the Tenth Symposium on Biotechnology for Fuels and Chemicals," *Appl. Biochem. Biotechnol.*, **21-21**, 1989
- Scott, C. D.; Lewis, S. N. "Biological Solubilization of Coal Using Both In Vivo and In Vitro Processes," presented at Semin., Univ. of Kentucky, Lexington, January 19, 1989, and published in *Appl. Biochem. Biotechnol.* **18**, 403-12 (1988)
- Scott, C. D.; Reichle, D. R.; Davison, B. H. *Bioprocessing Research at Oak Ridge National Laboratory*, 1989
- Scott, C. D.; Woodward, C. A.; Thompson, J. E. "Solute Diffusion in Biocatalyst Gel Beads Containing Biocatalysts and Other Additives," *Enzyme Microb. Technol.* **11**, 258-63 (1989)
- Scott, C. D.; Woodward, C. A.; Thompson, J. E.; Blankinship, S. L. "Coal Solubilization by Enhanced Enzyme Activity in Organic Solvents," presented at 11th Symp. on Biotechnology for Fuels and Chemicals, Colorado Springs, CO, May 8-12, 1989, and submitted to *Appl. Biochem. Biotechnol.*
- Scott, C. D.; Woodward, C. A.; Thompson, J. E.; Huggins, J. L. "Coal Solubilization by Enzymes in Organic Solvents," presented at 11th Symp. on Biotechnology for Fuels and Chemicals, Colorado Springs, CO, May 8-12, 1989

Sisson, W. G.; Begovich, J. M.; Byers, C. H.; Scott, C. D. "Continuous Chromatography," *CHEMTECH* **18**, 498-502 (1988)

Watson, J. S.; Scott, C. D. "Use of Bioadsorbents for Removing Dissolved Metals from Dilute Solutions," presented at Natl. Meet. Am. Inst. Chem. Eng., Washington, DC, November 28-December 2, 1988

Watson, J. S.; Scott, C. D. "Potential Roles for Bioprocessing in Enhanced Oil Recovery," in *Proc. Symp. on Applications of Microorganisms to Petroleum Technology*, Bartlesville, OK, August 12-13, 1987, NIPER-351, CONF-870858, 1988, pp. V-1--V-7

Watson, J. S.; Scott, C. D.; Faison, B. D. "Immobilized *Micrococcus* for Uptake of Strontium from Dilute Solutions," presented at Spring Symp. Am. Chem. Soc., I&EC Division, Atlanta, May 1-4, 1989, and to be published in *ACS Symp. Series*, Am. Chem. Soc.

Watson, J. S.; Scott, C. D.; Faison, B. D. "Adsorption of Sr by Immobilized Microorganisms," *Appl. Biochem. Biotechnol.* **20/21**, 699-709 (1989)

Scott, C. D.; Mrochek, J. E.; Newman, E.; Scott, T. C.; Michaels, G. E.; Petek, M. *A Preliminary Investigation of Cold Fusion by Electrolysis of Heavy Water*, ORNL/TM-11322, 1989

Davison, B. H.; Scott, C. D. "Operability of Ethanol Production by Immobilized Microorganisms," *Appl. Biochem. Biotechnol.* **18** 19-34 (1988)

Scott, T. C.

Scott, T. C.; Brumfield, T. L. "Use of High-Gradient Magnetic Fields for the Separation of Macromolecules," *Sep. Sci. Technol.* **23**, 1563-1572 (1988)

Scott, T. C.; Hill, C. G., Jr.; Amundson, C. H. "Effect of the Variation of the Level of Lactose Conversion in an Immobilized Lactase Reactor Upon Operating Costs for the Production of Baker's Yeast from Hydrolyzed Permeate Obtained from the Ultrafiltration of Cottage Cheese Whey," *Arch. Biochem. Biophys.* **18**, 187-202 (1988)

Basaran, O. A.; Scott, T. C.; Byers, C. H. "Theoretical and Experimental Analysis of the Oscillations of a Fluid Drop Immersed in Another Fluid," presented at Am. Inst. Chem. Eng. Meet. on Fundamental Research in Viscous Flows Session, Washington, DC, November 27-December 2, 1988

Basaran, O. A.; Scott, T. C.; Byers, C. H. "Drop Oscillations in Liquid-Liquid Systems," presented at 3rd Int. Conf. on Separations Technology, Davos, Switzerland, May 14-19, 1989, and published in *AIChE J.* **35**, 1263-70 (1989)

Basaran, O. A.; Scott, T. C.; Kim, S. "Drop-Drop Interactions and Coalescence in an Electric Field," presented at 3rd Int. Conf. on Separations Technology, Davos, Switzerland, May 14-19, 1989

Collins, E. D.; Box, W. D.; Godbee, H. W.; Scott, T. C. "Analysis of Data from Leaching Concrete Samples Taken from the TMI-2 Reactor Building Basement," presented at Am. Nucl. Soc. Top. Meet. on TMI-2 Accident Materials Behavior and Plant Recovery Technology, Washington, DC, October 31-November 4, 1988, and submitted to *Nucl. Technol.*

Scott, C. D.; Greenbaum, E.; Michaels, G. E.; Mrochek, J. E.; Newman, E.; Petek, M.; Scott, T. C. "Preliminary Investigation of Possible Low-Temperature Fusion," presented at Workshop on Cold Fusion Phenomena, Santa Fe, NM, May 23-25, 1989; submitted to *J. Fusion Energy*

Scott, T. C. "Use of High-Gradient Magnetic Fields for the Separation of Macromolecules," presented at Gordon Conf. on Separation and Purification, New London, NH, August 8-12, 1988, and at Am. Inst. Chem. Eng. Meet., Novel Separations Techniques Session, Washington, DC, November 27-December 2, 1988

Scott, T. C.; Wham, R. M. "An Electrically Driven Multistage Countercurrent Solvent Extraction Device: The Emulsion Phase Contactor," *Ind. Eng. Chem. Res.* **28**, 94-97 (1989)

Scott, T. C. "The Electrically Driven Emulsion-Phase Contactor: An Innovative Approach to Enhanced Efficiency in Solvent Extraction," presented at Am. Inst. Chem. Eng. Meet., Novel Separation Techniques Session, Washington, DC, November 27-December 2, 1988

Scott, T. C. "Use of Electric Fields in Solvent Extraction: A Review and Prospectus," *Sep. Purif. Methods* **18**, 65-109 (1989)

Scott, T. C.; Basaran, O. A. "Effects of Interfacial Distribution Behavior and Droplet-Continuum Hydrodynamics on Transient Separations in Liquid-Liquid Solvent Extraction," presented at 3rd Int. Conf. on Separations Technology, Davos, Switzerland, May 14-19, 1989

Scott, T. C.; Basaran, O. A. "Effects of Interfacial Distribution Behavior and Droplet-Continuum *Enzyme Microb. Technol.* **10**, 123-28 (1988)

Sears, M. B.

Sears, M. B., contributor to *Disposal of Chemical Agents and Munitions Stored at Tooele Army Depot, Tooele, Utah, Final Environmental Impact Statement*, U. S. Department of the Army, Program Manager for Chemical Demilitarization, Aberdeen Proving Ground, Maryland, July, 1989

Shappert, L. B.

Kelmers, A. D.; Shappert, L. B.; Roddy, J. W.; Hermann, O. W.; Parks, C. V. *Identification and Evaluation of Radionuclide Generation/Depletion Codes for Potential Use by the Department of Energy's Office of Civilian Radioactive Waste Management*, ORNL/TM-10404, February, 1989

Kerr, H. T.; King, L. J.; Shappert, L. B.; Unger, W. E.; Worley, B. A.; Wymer, R. G. "Presentations at Technical Aspects of Nuclear Weapons Proliferation Workshop, Oak Ridge National Laboratory, Oak Ridge, Tennessee, December 13-14, 1988," to be published in *Papers presented at Technical Aspects of Nuclear Weapons Proliferation Workshop, ORNL, Oak Ridge, TN, Dec. 13-14, 1988*

Kerr, H. T.; Moses, D. L.; Shappert, L. B.; Wymer, R. G. "Presentations at Nuclear Non-Proliferation Workshop, ORNL, Oak Ridge, Tennessee, April 25-26, 1989," handout to workshop participants

Shappert, L. B. "Cask Drop Testing and Analysis," presented at Am. Soc. Mech. Eng. Pressure Vessel and Piping Conf., Honolulu, Hawaii, July 23-27, 1989, and published in *Advances in Packaging and Transportation of Radioactive Materials*, C. K. Chou and L. B. Shappert, eds., vol. 164, The American Society of Mechanical Engineers, New York, 1989, pp. 13-16

Shappert, L. B. "Transportation of Radioactive Materials," presented at Technical Aspects of Nuclear Weapons Proliferation Workshop, ORNL, Oak Ridge, TN, February 14-17, 1989; presented at Nuclear Non-Proliferation Workshop, ORNL, Oak Ridge, TN, May 9-12, 1989

Shappert, L. B.; Klimas, M. J. "Transportation Operations Functions of the Federal Waste Management System," presented at 9th Int. Symp. on Packaging and Transportation of Radioactive Materials (PATRAM), Washington, DC, June 11-16, 1989, and published in *Proc. 9th Int. Symp. on Packaging and Transportation of Radioactive Materials*, Washington, DC, June 11, 1989, CONF-890631, vol. 1, Washington, DC, June 11, 1989, pp. 906-12

Shappert, L. B.; Meinert, N. M. "Drop Tests of the Quarter-Scale Model of the NAC Light-Water Reactor Transport Cask," presented at 9th Int. Symp. on Packaging and Transportation of Radioactive Materials (PATRAM '89), Washington, DC, June 11-16, 1989, and published in *Proc. 9th Intern. Symp. on Packaging and Transportation of Radioactive Materials (PATRAM '89)*, vol. 1, CONF-890631, Washington, D. C., June 11, 1989, pp. 25-33

Sherrow, S. A.

Sherrow, S. A. "Observation of UF_6^- in Xe Matrices," *J. Chem. Phys.* **90**, 5886-87 (1989)

Sherrow, S. A. *An Evaluation of the Suitability of Laser-Induced Fluorescence for Measurements of Fission-Product Iodine Sorptivity in the MHTGR*, ORNL-6553 (DOE-HTGR-88298), July, 1989

Shockley, W. E.

Kress, T. S.; Beahm, E. C.; Shockley, W. E.; Weber, C. F. "Trends Status," presented at Trends Status, Severe Accident Research Partners Meet., Washington, DC, October 17-21, 1988

Beahm, E. C.; Brown, M. L.; Shockley, W. E. "ACE Phase B: ORNL Aerosol and Hydrogen Burn Studies," presented at Advanced Containment Experiment Technical Advisory Committee Review, ANL, Argonne, IL, May 10, 1989

Beahm, E. C.; Daish, S. R.; Shockley, W. E.; Hopfenfeld, J. "Iodine Partitioning in PWR Steam Generator Accidents," submitted to *Nucl. Technol.*

Beahm, E. C.; Weber, C. F.; Kress, T. S.; Shockley, W. E. "Iodine Behavior in Containment," presented at ICHMT Semin. on Fission Product Transport Processes in Reactor Accidents, Dubrovnik, Yugoslavia, May 22-26, 1989

Beahm, E. C.; Weber, C. F.; Kress, T. S.; Shockley, W. E.; Daish, S. R. "Chemistry and Mass Transport of Iodine in Containment," in *Proc. 2nd CSNI Workshop on Iodine Chemistry in Reactor Safety*, Toronto, Canada, June 2-3, 1988 (1989), pp. 251-66

Daish, S. R.; Beahm, E. C.; Shockley, W. E. "Aqueous Iodine at Low Concentrations and High Temperatures," in *Proc. 2nd Workshop on Chemistry of Iodine in Reactor Safety*, Toronto, June 2-3, 1988 (1989), pp. 74-94

Weber, C. F.; Beahm, E. C.; Kress, T. S.; Daish, S. R.; Shockley, W. E. "TRENDS: A Code for Modeling Iodine Behavior in Containment During Severe Accidents," presented at ICHMT Int. Semin. on Heat and

Mass Transfer Aspects of Fission Product Releases, Dubrovnik, Yugoslavia, May 22-26, 1989, and to be published in *Proc. ICHMT Int. Semin. on Heat and Mass Transfer Aspects of Fission Product Releases*, Dubrovnik, Yugoslavia, May 22-26, 1989

Shor, J. T.

Shor, J. T. *Tritium Effluent Reduction at Oak Ridge National Laboratory*, ORNL/TM-10664, November, 1988

Singh, S. P. N.

Egan, B. Z.; Singh, S. P. N.; Fain, D. E.; Kidd, G. J. "Gas Separations Using Inorganic Membranes," *Gas Stream Cleanup Papers from DOE/METC Sponsored Contractors Review Meetings in 1988*, DOE/METC-89/6099, 1988, pp. 287-92 in *Proc. 8th Annu. Gasification and Gas Stream Cleanup Systems Contractors Review Meet.*, Morgantown, WV, May 10-12, 1988, DOE/METC 88/6092, 1988, v. 2, pp. 519-24

Egan, B. Z.; Singh, S. P. N.; Fain, D. E.; Kidd, G. J.; Roettger, G. E.; White, D. E. "Gas Separations Using Inorganic Membranes," presented at 9th Annu. Gasification and Gas Stream Cleanup Systems Contractors Review Meet., Morgantown, WV, June 27-29, 1989, and to be published in *Proc. 9th Annu. Gasification and Gas Stream Cleanup Systems Contractors Review Meet.*, Morgantown, WV, June 27-29, 1989

Fain, D. E.; Kidd, G. J.; Egan, B. Z.; Singh, S. P. N. "Development and Evaluation of Ceramic Membranes for Gas Separation," presented at Fossil Energy Materials Conf., Oak Ridge, TN, August 30-September 1, 1988 presented at Fossil Energy Materials Conf., Oak Ridge, TN, August 8-10, 1989

Jolley, R. L.; Hollenbeck, P. E.; Kennerly, J. M.; Morris, M. I.; Singh, S. P. N. "Evaluation of Selected Emerging Waste Treatment and Disposal Methods," presented at Meet. Am. Chem. Soc., Miami Beach, FL, September 10-15, 1989 and to be published in *Proc. Meet. Am. Chem. Soc.*, Miami Beach, FL, Sept. 10-15, 1989, Am. Chem. Soc.

Morris, M. I.; Jolley, R. L.; Singh, S. P. N. "The Waste Management Technology Center's Role in Waste Management Technology Applications for Martin Marietta Energy Systems, Inc.," presented at Weld. Test. Tech. Conf., Knoxville, TN, February 1989

Singh, S. P. N. "Workshop on the Off-Site Release Criteria for Contaminated Materials, May 2-3, 1989," 1989

Singh, S. P. N.; Genung, R. K.; Jolley, R. L. *Guidance Document for Conducting Waste Management Demonstration Projects*, ORNL/TM-10845, July, 1988

Sisson, W. G.

Byers, C. H.; Sisson, W. G.; DeCarli, J. P., II; Carta, G. "Sugar Separations on a Pilot Scale by Continuous Ion-Exchange Chromatography," presented at Annu. Meet. Am. Inst. Chem. Eng., Washington, DC, November 27-December 2, 1988, and submitted to *Biotechnol. Prog.*

Byers, C. H.; Sisson, W. G.; DeCarli, J. P., II; Carta, G. "Pilot-Scale Studies of Sugar Separations by Continuous Chromatography," *Appl. Biochem. Biotechnol.* **20/21**, 635-54 (1988)

Scott, T. C.; Sisson, W. G. "Droplet Size Characteristics and Energy Input Requirements of Emulsions Formed Using High-Intensity-Pulsed Electric Fields," *Sep. Sci. Technol.* **23**, 1541-50 (1988)

Sisson, W. G.; Begovich, J. M.; Byers, C. H.; Scott, C. D. "Continuous Chromatography," *CHEMTECH* **18**, 498-502 (1988)

Byers, C. H.; Sisson, W. G.; DeCarli, J. P., II "The Use of Gradient Elution in Optimizing Continuous Annular Ion Exchange Chromatography with Applications to Metal Separations," presented at IEX'88, Cambridge, Great Britain, July 17-22, 1988, and published in *Ion Exchange for Industry, Proc. IEX '88*, 1989, pp. 424-42

Snider, J. W.

McNeese, L. E.; Berry, J. B.; Butterworth, G. E., III; Collins, E. D.; Monk, T. H.; Patton, B. D.; Snider, J. W. *Overall Strategy and Program Plan for Management of Radioactively Contaminated Liquid Wastes and Transuranic Sludges at the Oak Ridge National Laboratory*, ORNL/TM-10757, December, 1988

Spence, R. D.

Gilliam, T. M.; Spence, R. D.; Evans-Brown, B. S.; Morgan, I. L.; Shoemaker, J. L.; Bostick, W. D. "Performance Testing of Blast Furnace Slag for Immobilization of Technetium in Grout," presented at SPECTRUM '88, Int. Top. Meet. on Nuclear and Hazardous Waste Management, Pasco, WA, September 11-15, 1988, and published in *Proc. SPECTRUM '88*, American Nuclear Society, Inc., La Grange Park, IL, 1988, pp. 109-11

Gilliam, T. M.; Spence, R. D.; Evans-Brown, B. S.; Morgan, I. L.; Shoemaker, J. L.; Bostick, W. D. "Performance Testing of Blast Furnace Slag for Immobilization of Technetium in Grout," in *Proc. SPECTRUM '88*, Int. Top. Meet. on Nuclear and Hazardous Waste Management, Pasco, WA, September 11-15, 1988, American Nuclear Society, Inc., La Grange Park, IL, 1988, pp. 109-11

Morgan, I. L.; Osborne, S. C.; Spence, R. D.; Gilliam, T. M. "Immobilization of Volatile Organic Compounds in a Cement Waste Form: Sample Preparation Procedures," presented at Meet. Am. Chem. Soc., Div. Environ. Chem., Miami Beach, FL, September 10-15, 1989

Morris, M. I.; Dole, L. R.; Spence, R. D.; Gilliam, T. M. "Immobilization of Volatile Organics in Cement-Based Grouts," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988

Morris, M. I.; Spence, R. D.; Gilliam, T. M.; Dole, L. R. "A Program to Establish In Situ Immobilization as a Remedial Action Option for Wastes Containing Volatile Organic Compounds (VOC)," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988, and published in *Proc. DOE Model Conf.*, CONF-881054, Vol. 5, 1988, pp. 1501-08

Spence, R. D.; Bostick, W. D.; McDaniel, E. W.; Gilliam, T. M.; Shoemaker, J. L.; Tallent, O. K.; Morgan, I. L.; Evans-Brown, B. S.; Dodson, K. E. "Immobilization of Technetium in Blast Furnace Slag Grouts," presented at 3rd Int. Conf. on Fly Ash, Silica Fume, Slag, and Natural Pozzolans in Concrete, Trondheim, Norway, June 19-26, 1989, and published in *Proc. 3rd Int. Conf. on Fly Ash, Silica Fume, Slag, and Natural Pozzolans in Concrete*, Vol. 2, pp. 1579-1596

Spence, R. D.; Gilliam, T. M.; Morgan, I. L.; Osborne, S. C. "Immobilization of Volatile Organic Compounds in Commercial Cement-Based Waste Forms," news release for ACS News Service

Spence, R. D.; Godbee, H. W.; Tallent, O. K.; Nestor, C. W. Jr. "Interpretation of Leaching Data for Cementitious Waste Forms Using Analytical Solutions Based on Mass Transport Theory and Empiricism," presented at the 197th ACS National Meeting, Dallas, TX, April 9-14, 1989

Spence, R. D.; Tamura, T. "In Situ Grouting of Shallow Landfill Radioactive Waste Trenches," in *Environmental Aspects of Stabilization and Solidification of Hazardous and Radioactive Wastes, Proc. 4th Int. Hazardous Waste Symp.*, Atlanta, May 3-6, 1987, Am. Soc. Test. Mater. Spec. Tech. Publ. 1033, 1989, pp. 418-29

Starnes, B. W.

Krichinsky, A. M.; Holladay, D. W.; Johnson, J. K.; Starnes, B. W. "Initial Decommissioning Activities for the Radiochemical Processing Plant at Oak Ridge National Laboratory," presented at Annu. Conf. Am. Glovebox Soc., Denver, August 22-25, 1988

Storch, S. N.

Klein, J. A.; Ashline, R. C.; DePaoli, S. M.; Godbee, H. W.; Kibbey, A. H.; Pearson, R. L.; Storch, S. N. *Integrated Data Base for 1988: Spent Fuel and Radioactive Waste Inventories, Projections, and Characteristics*, DOE/RW-0006, Rev. 4, September, 1988

Strandberg, G. W.

Donaldson, T. L.; Strandberg, G. W.; Bolla, L. L. "Bioremediation of TCE-Contaminated Groundwater," presented at Meet. American Inst. of Chemical Engineers, Houston, April 2-6, 1989, and to be published in *Proc. Meet. American Inst. of Chemical Engineers*, Houston, Apr. 2-6, 1989

Donaldson, T. L.; Strandberg, G. W.; McGinnis, C. P.; Palumbo, A. V.; White, D. C.; Hill, D. L.; Phelps, T. J.; Hadden, C. T.; Revis, N. W.; Holdsworth, G.; Osborne, T. *Bioremediation of PCB-Contaminated Soil at the Y-12 Plant*, ORNL/TM-10750, September, 1988

Donaldson, T. L.; Strandberg, G. W.; Palumbo, A. V. "Biotreatment of TCE-Contaminated Groundwater," presented at Spring Natl. Meet. Am. Inst. Chem. Eng., Houston, April 2-6, 1989

Hill, D. L.; Phelps, T. J.; Palumbo, A. V.; Strandberg, G. W.; Donaldson, T. L.; White, D. C. "Bioremediation of Polychlorinated Biphenyls: Degradation Capabilities in Field Lysimeters," presented at DOE Model Conf., Oak Ridge, TN, October 3-7, 1988, and to be published in *Program Booklet, DOE Model Conf., Oak Ridge, TN, Oct. 3-7, 1988*; also published in *Appl. Biochem. Biotechnol.* **20/21**, 233-43 (1989)

Pitt, W. W.; Strandberg, G. W.; Donaldson, T. L.; Bolla, L. L. "Biodegradation of Trichloroethylene Contaminated Groundwater," presented at Conf. on Biotechnology Applications in Hazardous Waste Treatment, Longboat Key, FL, October 30-November 4, 1988, and to be published in *Proc. Conf. on Biotechnology Applications in Hazardous Waste Treatment*, Longboat Key, FL, Oct. 30-Nov. 4, 1988

Strandberg, G. W. "The Solubilization of Low-Ranked Coal by Microorganisms," in *Proc. Biochemie und Mikrobiologie roukohle und Kohleinhaltstoffen*, Saalbau-Essen, Federal Republic of Germany, July 9-10, 1987, K2G (Kohlegewinnong Zweite), 1988, pp. 91-99

Strandberg, G. W.; Arnold, W. D., Jr. "Microbial Accumulation of Neptunium," *J. Ind. Microbiol.* **3**, 329-31 (1988)

Strandberg, G. W.; Donaldson, T. L.; Bolla, L. L.; Palumbo, A. V.; Eng, W. "Degradation of Trichloroethylene and *Trans*-1,2-Dichloroethylene by a Methanotrophic Consortia in a Trickle-Type Bioreactor," to be published in *Proc. DOE Model Conf.*, Oak Ridge, TN, Oct. 3-7, 1988

Strandberg, G. W.; Donaldson, T. L.; Bolla, L. L.; Palumbo, A. V.; Eng, W. "Biodegradation of Trichloroethylene and *Trans*-1,2-Dichloroethylene by Methanotrophs in a Laboratory-Scale Trickle Bioreactor," presented at DOE Model Conf., Oak Ridge, TN, Oct. 3-7, 1988, and to be published in *Abstract Booklet, DOE Model Conf., Oak Ridge, TN, Oct. 3-7, 1988*

Strandberg, G. W.; Donaldson, T. L.; Farr, L. L. "Degradation of Trichloroethylene and *Trans*-1,2-Dichloroethylene by a Methanotrophic Consortium in a Fixed-Film, Packed-Bed Bioreactor," *Environ. Sci. Technol.* **23**, 1422-25 (1989)

Strandberg, G. W.; Lewis, S. N. "Factors Affecting Coal Solubilization by the Bacterium *Streptomyces setonii* 75Vi2 and by Alkaline Buffers," *Appl. Biochem. Biotechnol.* **18**, 355-61 (1988)

Sutton, A. L., Jr.

Lindemer, T. B.; Hunley, J. F.; Gates, J. E.; Sutton, A. L., Jr.; Brynestad, J.; Hubbard, C. R.; Gallagher, P. K. "Experimental and Thermodynamic Study of Nonstoichiometry in $\langle YBa_2Cu_3O_{7-x} \rangle$," *J. Am. Ceram. Soc.* **72**, 1775-88 (1989)

Lindemer, T. B.; Hunley, J. F.; Gates, J. E.; Sutton, A. L., Jr.; Brynestad, J.; Hubbard, C. R.; Gallagher, P. K. *Experimental and Thermodynamic Study of Nonstoichiometry in $\langle YBa_2Cu_3O_{7-x} \rangle$* , ORNL/TM-10899, May, 1989

Lindemer, T. B.; Sutton, A. L., Jr. "Study of the Nonstoichiometry of $YBa_2Cu_3O_{7-x}$," presented at Symp. on Superconductivity, 91st Annu. Meet. Am. Ceram. Soc., Indianapolis, April 23-27, 1989

Lindemer, T. B.; Sutton, A. L., Jr. "Experimental and Thermodynamic Study of Nonstoichiometry of $YBa_2Cu_3O_{7-x}$," presented at 44th Annu. Calorimetry Conf., Oak Ridge, TN, July 30-August 4, 1989

Lindemer, T. B.; Sutton, A. L., Jr. "Study of Nonstoichiometry of $\langle U_{1-z}Gd_zO_{2\pm x} \rangle$," *J. Am. Ceram. Soc.* **71**, 553-61 (1988)

Lindemer, T. B.; Sutton, A. L., Jr. *Review of Nonstoichiometry in $YBa_2Cu_3O_{7-x}$* , ORNL/TM-10827, November, 1988

Lindemer, T. B.; Hunley, J. F.; Gates, J. E.; Sutton, A. L., Jr.; Brynestad, J.; Hubbard, C. R.; Gallagher, P. K. "Experimental and Thermodynamic Study of Nonstoichiometry in $\langle YBa_2Cu_3O_{7-x} \rangle$," seminar, Department of Ceramics, Rutgers, The State University, Piscataway, NJ, Feb. 7, 1989.

Lindemer, T. B.; Hunley, J. F.; Gates, J. E.; Sutton, A. L., Jr.; Brynestad, J.; Hubbard, C. R.; Gallagher, P. K. "Experimental and Thermodynamic Study of Nonstoichiometry in $\langle YBa_2Cu_3O_{7-x} \rangle$," seminar, E.I. duPont Experimental Engineering Station, Wilmington, DE, April 5, 1989.

Lindemer, T. B.; Hunley, J. F.; Gates, J. E.; Sutton, A. L., Jr.; Brynestad, J.; Hubbard, C. R.; Gallagher, P. K. "Experimental and Thermodynamic Study of Nonstoichiometry in $\langle YBa_2Cu_3O_{7-x} \rangle$," seminar, National Institute of Standards and Technology, Gaithersburg, MD, August 10, 1989.

Lindemer, T. B.; Hunley, J. F.; Gates, J. E.; Sutton, A. L., Jr.; Brynestad, J.; Hubbard, C. R.; Gallagher, P. K. "Experimental and Thermodynamic Study of Nonstoichiometry in $\langle YBa_2Cu_3O_{7-x} \rangle$," seminar, Department of Materials Engineering, Purdue University, W. Lafayette, IN, September 29, 1989.

Tallent, O. K.

Del Cul, G. D.; Tallent, O. K.; Sams, T. L. "Monitor for Waste Immobilized in Cement-Based Materials," presented at Technology Transfer Workshop, Oak Ridge, TN, March 23, 1989

McDaniel, E. W.; Tallent, O. K.; Sams, T. L.; Bostick, W. D.; Delzer, D. B. "Basis for Selecting Cement-Based Waste Forms for Immobilizing Radioactive Waste," presented at 12th Int. Symp. on Scientific Basis for Nuclear Waste Management, Internationales Congress Centrum, Berlin, October 10-13, 1988, and published in *Proc. 12th Mater. Res. Soc. Symp. on Scientific Basis for Nuclear Waste Management*, Internationales Congress Centrum (ICC), Berlin, October 10-13, 1988, Mater. Res. Soc., 1989, v. 127, pp. 421-30

Tallent, O. K.; McDaniel, E. W.; Del Cul, G. D.; Dodson, K. E.; Trotter, D. R. *Development of Immobilization Technology for Hanford Double-Shell Slurry Feed Waste*, ORNL/TM-10906, August, 1989

Spence, R. D.; Bostick, W. D.; McDaniel, E. W.; Gilliam, T. M.; Shoemaker, J. L.; Tallent, O. K.; Morgan, I. L.; Evans-Brown, B. S.; Dodson, K. E. "Immobilization of Technetium in Blast Furnace Slag Grouts," presented at 3rd Int. Conf. on Fly Ash, Silica Fume, Slag, and Natural Pozzolans in Concrete, Trondheim, Norway, June 19-26, 1989, and published in *Proc. 3rd Int. Conf. on Fly Ash, Silica Fume, Slag, and Natural Pozzolans in Concrete*, Vol. 2, pp. 1579-1596

Tallent, O. K.; McDaniel, E. W.; Del Cul, G. D.; Dodson, K. E. "Computer Application to Leach Study Data," in p. 342, *Abstracts of 90th Annu. Meet. Am. Ceram. Soc., Cincinnati*, May 4-5, 1988 (1988)

Tallent, O. K.; McDaniel, E. W.; Del Cul, G. D.; Dodson, K. E.; Trotter, D. R. *Development of Immobilization Technology for Hanford Double-Shell Slurry Feed Waste*, ORNL/TM-10906, August, 1989

Tevault, C. V.

Graves, D. A.; Tevault, C. V.; Greenbaum, E. "Control of Photosynthetic Reductant: The Role of Light and Temperature on Sustained Hydrogen Photoevolution in *Chlamydomonas* sp. in an Anoxic, Carbon Dioxide-Containing Atmosphere," *Photochem. Photobiol.* **50**, 571-76 (1989)

Thompson, J. E.

Bajpai, R.; Thompson, J. E.; Davison, B. H. "Gas Hold-Up in Three-Phase Immobilized Cell Bioreactors," submitted to *Appl. Biochem. Biotechnol.*

Scott, C. D.; Woodward, C. A.; Thompson, J. E. "Solute Diffusion in Biocatalyst Gel Beads Containing Biocatalysts and Other Additives," *Enzyme Microb. Technol.* **11**, 258-63 (1989)

Scott, C. D.; Woodward, C. A.; Thompson, J. E.; Blankinship, S. L. "Coal Solubilization by Enhanced Enzyme Activity in Organic Solvents," submitted to *Appl. Biochem. Biotechnol.*

Scott, C. D.; Woodward, C. A.; Thompson, J. E.; Blankinship, S. L. "Coal Solubilization by Enhanced Enzyme Activity in Organic Solvents," presented at 11th Symp. on Biotechnology for Fuels and Chemicals, Colorado Springs, CO, May 8-12, 1989

Scott, C. D.; Woodward, C. A.; Thompson, J. E.; Huggins, J. L. "Coal Solubilization by Enzymes in Organic Solvents," presented at 11th Symp. on Biotechnology for Fuels and Chemicals, Colorado Springs, CO, May 8-12, 1989

Tompkins, J. A.

Kobisk, E. H.; Ramey, D. W.; Aaron, W. S.; Tompkins, J. A.; Haff, K. W.; DeVore, J. R.; Adair, H. L. "Tritium-Processing Operations at the Oak Ridge National Laboratory with Emphasis on Safe-Handling Practices," presented at Int. Nucl. Target Dev. Soc. and IAEA-INDC Conf., Darmstadt, Federal Republic of Germany, September 5-9, 1988, and published in *Nucl. Instrum. Methods Phys. Res. A* **282**, 329-40 (1989)

Toth, L. M.

Felker, L. K.; Toth, L. M. "Fluorine Gettering by Activated Charcoal in a Radiation Environment," *Sep. Sci. Technol.* **23**, 1959-68 (1988)

Toth, L. M.; Bell, J. T.; Friedman, H. A. "The Disproportionation of Pu^{4+} in Nitric Acid Solutions," submitted to *Radiochim. Acta*

Toth, L. M.; Felker, L. K. "Fluorine Generation by Gamma Radiolysis of a Fluoride Salt Mixture," submitted to *Radiat. Eff.*

Toth, L. M.; Felker, L. K. "Fluorine Generation by Gamma Radiolysis of a Fluoride Salt Mixture," accepted by *Radiat. Eff.*

Toth, L. M.; Felker, L. K. "Purex Acid-Split Flowsheet Studies," ORNL/TM-10993, April, 1989

Thiyagarjan, P.; Diamond, H.; Soderholm, L.; Horwitz, E. P.; Toth, L. M.; Felker, L. K. "Pu(IV) Polymers in Aqueous and Organic Solutions," *Inorg. Chem.*, in press

Tracy, J. G.

Tracy, J. G. "Isotope Separation Program—Present and Future," presented at 14th World Conf. of Int. Nucl. Target Dev. Soc., Darmstadt, Federal Republic of Germany, September 5-9, 1988, and to be published in *Proc. 14th World Conf. of Int. Nucl. Target Dev. Soc.*, Darmstadt, Federal Republic of Germany, Sept. 5-9, 1988

Johnson, C. L.; Cline, R. L.; Ferren, D. M.; Tracy, J. G.; Haff, K. W.; Keller, O. L.; Genung, R. K.; Hoffman, D. C.; Moody, D. *ISOTOPICS*, no. 17, October 1988

Travis, J. R.

Osborne, M. F.; Collins, J. L.; Lorenz, R. A.; Travis, J. R.; Webster, C. S. *Data Summary Report for Fission Product Release Test VI-2*, ORNL/TM-11105, September, 1989

Osborne, M. F.; Collins, J. L.; Lorenz, R. A.; Travis, J. R.; Webster, C. S.; Yamashita, T. *Data Summary Report for Fission Product Release Test VI-1*, ORNL/TM-11104, June, 1989

Osborne, M. F.; Lorenz, R. A.; Collins, J. L.; Travis, J. R.; Webster, C. S. "Fission Product Release at Severe Accident Conditions," presented at Project Review for Visitors from the Japan Inst. of Nuclear Safety, ORNL, Oak Ridge, TN, February 21, 1989

Trotter, D. R.

Tallent, O. K.; McDaniel, E. W.; Del Cul, G. D.; Dodson, K. E.; Trotter, D. R. *Development of Immobilization Technology for Hanford Double-Shell Slurry Feed Waste*, ORNL/TM-10906, August, 1989

Vaughen, V. C. A.

Lin, K. H.; Standifer, P.; Vaughen, V. C. A. *Comprehensive Self-Assessment and Upgrade Program Progress Report for the Period January 1 to June 30, 1989*, ORNL/TM-11288, September, 1989

Vaughen, V. C. "A New Focus on Ethics in the Workplace: The Role of the Professional Society," presented at Forum Discussion, Sponsored by the Technology and Society Division Board on Professional Practice and Ethics, Am. Soc. Mech. Eng., Chicago, November 29, 1988

Vaughen, V. C. A. "The Fascination and Lure of Ethics," *The Oak Ridger*

Vaughen, V. C. A. "Engineering Ethics," presented at Semin., Chemical Engineering Dep., Univ. of Tennessee, Knoxville, February 9, 1989

Vaughen, V. C. A. "Science, Technology, and Society," presented at Spring '89 ORSERS Students Presentation, ORNL, Oak Ridge, TN, March 1, 1989

Vaughen, V. C. A. "Engineering Ethics in the 1990s," presented at Meet. of Am. Soc. Mech. Eng., Oak Ridge, TN, September 21, 1989

Villiers-Fisher, J. F.

Lucero, A. J.; Wilson, J. H.; Counce, R. M.; Villiers-Fisher, J. F.; Singh, S. P.; Ashworth, R. A. "Air Stripping of Volatile Organic Compounds from Groundwater Using Towers Filled with Standard and High-Efficiency Packing," presented at Summer Natl. Meet. Am. Inst. Chem. Eng., Philadelphia, August 20-23, 1989

Singh, S. P.; Wilson, J. H.; Counce, R. M.; Villiers-Fisher, J. F.; Jennings, H. L.; Lucero, A. J.; Reed, G. D.; Ashworth, R. A.; Elliott, M. G. "Removal of Volatile Organic Compounds from Groundwater Using a Rotary Air Stripper," presented at Summer Natl. Meet. Am. Inst. Chem. Eng., Philadelphia, August 20-23, 1989

Walker, J. F. Jr.

Hylton, T. D.; Walker, J. F., Jr. *Environmental Studies in Support of the Live Fire Training Facilities Project*, ORNL/TM-11265, August, 1989

Walker, J. F.; Brown, C. H., Jr.; Wilson, J. H. "Treatment of Chromium-Contaminated Plating Shop Rinsewater Streams by Reverse Osmosis," presented at Water Pollution Control Federation Conf., Dallas, October 2-6, 1988, and published in *Proc. of the Industrial Waste Symp.*, 61st Annu. WPCF Conf., Dallas, October 3-6, 1988, Water Pollution Control Federation, 1988

Walker, J. F., Jr.; Helfrich, M. V.; Donaldson, T. L. "Biodenitrification of Uranium Refinery Wastewaters," *Environ. Prog.* 8, 97-101 (1989)

Watson, J. S.

Bell, J. T.; Watson, J. S. "Fifth Symposium on Separation Science and Technology for Energy Applications," *Sep. Sci. Technol.*, **23**, nos. 12, 13 (1988)

Bell, J. T.; Watson, J. S. "Call for Papers—Sixth Symposium on Separation Science and Technology for Energy Applications, Knoxville, TN, October 22-27, 1989," (1988).

Faison, B. D.; Watson, J. S.; Cancel, C. A.; Scott, C. D. "Applications of Biotechnology in Waste Management for the Nuclear Industry," presented at Annu. Meet. Air Waste Management Assoc., Southern Section, Diamondhead, MS, September 20-22, 1989

Kirslis, S. J.; Watson, J. S. "Heat Transfer in a Rotary Kiln," presented at Spring Natl. Meet. Am. Inst. Chem. Eng., Houston, April 2-6, 1989

Misra, A. K.; Watson, J. S. "Microbial Enhancement of Oil Recovery," presented at 1st Int. Symp. on Gas, Oil, and Coal Biotechnology, New Orleans, December 5-7, 1988, and to be published in *Proc. 1st Int. Symp. on Gas, Oil, and Coal Biotechnology*, New Orleans, Dec. 5-7, 1988

Watson, J. S. "Adaptability in Chemical Engineering," presented at Workshop on National Priorities and Technological Change: Fostering Flexibility in the Engineering Work Force, Washington, DC, September 29, 1989, and to be published by the National Research Council

Watson, J. S.; Scott, C. D. "Use of Bioadsorbents for Removing Dissolved Metals from Dilute Solutions," presented at Natl. Meet. Am. Inst. Chem. Eng., Washington, DC, November 28-December 2, 1988

Watson, J. S.; Scott, C. D. "Potential Roles for Bioprocessing in Enhanced Oil Recovery," in *Proc. Symp. on Applications of Microorganisms to Petroleum Technology*, Bartlesville, OK, August 12-13, 1987, NIPER-351, CONF-870858, 1988, pp. V-1--V-7

Watson, J. S.; Scott, C. D.; Faison, B. D. "Immobilized *Micrococcus* for Uptake of Strontium from Dilute Solutions," presented at Spring Symp. Am. Chem. Soc., I&EC Division, Atlanta, May 1-4, 1989, and to be published in *ACS Symp. Series*, Am. Chem. Soc.

Watson, J. S.; Scott, C. D.; Faison, B. D. "Adsorption of Sr by Immobilized Microorganisms," *Appl. Biochem. Biotechnol.* **20/21**, 699-709 (1989)

Webster, C. S.

Osborne, M. F.; Collins, J. L.; Lorenz, R. A.; Travis, J. R.; Webster, C. S. *Data Summary Report for Fission Product Release Test VI-2*, NUREG/CR-5340, ORNL/TM-11105, July, 1989

Osborne, M. F.; Lorenz, R. A.; Collins, J. L.; Nakamura, T. "Time-Dependent Release of Fission Products from LWR Fuel Under Severe Accident Conditions," Proceedings of International ENS/ANS Conference on Thermal Reactor Safety, Avignon, France, October 3-7, 1988, and published in October 1988 (vols. 1-4) and March 1989 (vols. 5-6)

Osborne, M. F.; Collins, J. L.; Lorenz, R. A.; Travis, J. R.; Webster, C. S.; Yamashita, T. *Data Summary Report for Fission Product Release Test VI-1*, NUREG/CR-5339, ORNL/TM-11104, June, 1989

Osborne, M. F.; Lorenz, R. A.; Collins, J. L.; Travis, J. R.; Webster, C. S. "Fission Product Release at Severe Accident Conditions," presented at Project Review for Visitors from the Japan Inst. of Nuclear Safety, ORNL, Oak Ridge, TN, February 21, 1989

Webster, C. S.; Osborne, M. F. *The Use of Fiber Optics for Remote Temperature Measurement in Fission Product Release Test*, NUREG/CR-4271, ORNL/TM-10366, April, 1989

Wike, J. S.

Wike, J. S.; Guyer, C. E.; Ramey, D. W.; Phillips, B. P. "Chemistry for Commercial-Scale Productions of Yttrium-90 for Medical Research," presented at the American Chemical Society (Pacific Basin) meeting, Honolulu, Hawaii, December 19, 1989; to be published in *Int. J. Radiat. Appl. Instrum.*

Wham, R. M.

Scott, T. C.; Wham, R. M. "An Electrically Driven Multistage Countercurrent Solvent Extraction Device: The Emulsion Phase Contactor," *Ind. Eng. Chem. Res.* **28**, 94-97 (1989)

Wilson, J. H.

Lucero, A. J.; Wilson, J. H.; Counce, R. M.; Villiers-Fisher, J. F.; Singh, S. P.; Ashworth, R. A. "Air Stripping of Volatile Organic Compounds from Groundwater Using Towers Filled with Standard and High-Efficiency Packing," presented at Summer Natl. Meet. Am. Inst. Chem. Eng., Philadelphia, August 20-23, 1989

Singh, S. P. N.; Wilson, J. H.; Counce, R. M.; Villiers-Fisher, J. F.; Jennings, H. L.; Lucero, A. J.; Reed, G. D.; Ashworth, R. A.; Elliott, M. G. "Removal of Volatile Organic Compounds from Groundwater Using a Rotary Air Stripper," presented at Summer Natl. Meet. Am. Inst. Chem. Eng., Philadelphia, August 20-23, 1989

Walker, J. F.; Brown, C. H., Jr.; Wilson, J. H. "Treatment of Chromium-Contaminated Plating Shop Rinsewater Streams by Reverse Osmosis," presented at Water Pollution Control Federation Conf., Dallas, October 2-6, 1988, and published in *Proc. of the Industrial Waste Symp.*, 61st Annu. WPCF Conf., Dallas, October 3-6, 1988, Water Pollution Control Federation, 1988

Woodward, C. A.

Davison, B. H.; Nicklaus, D. M.; Woodward, C. A.; Lewis, S. N.; Faison, B. D. "Characterization and Subsequent Utilization of Microbially Solubilized Coal: Preliminary Studies," presented at Biological Processing of Coal and Coal-Derived Substances, Palo Alto, CA, May 16-17, 1989, and to be published in *Biological Processing of Coal and Coal-Derived Substances*, EPRI Report

Faison, B. D.; Woodward, C. A. "Degradation of Heterocyclic Coal Model Compounds by Coal-Solubilizing Fungi," presented at Annu. Meet. Soc. Ind. Microbiol., Seattle, August 14-18, 1989

Faison, B. D.; Woodward, C. A.; Bean, R. M. "Microbial Solubilization of a Preoxidized Subbituminous Coal: Product Characterization," presented at 11th Symp. on Biotechnology for Fuels and Chemicals, Colorado Springs, CO, May 8-12, 1989, and to be published in *Proc. 11th Symp. on Biotechnology for Fuels and Chemicals*, Appl. Biochem. and Biotechnol.

Faison, B. D.; Woodward, C. A.; Lewis, S. N.; Bean, R. M. "The Biochemistry of Microbial Coal Solubilization: Model Compound Studies and Product Characterization," presented at 11th Symp. on Biotechnology for Fuels and Chemicals, Colorado Springs, CO, May 8-12, 1989

Scott, C. D.; Woodward, C. A.; Thompson, J. E. "Solute Diffusion in Biocatalyst Gel Beads Containing Biocatalysts and Other Additives," *Enzyme Microb. Technol.* **11**, 258-63 (1989)

Scott, C. D.; Woodward, C. A.; Thompson, J. E.; Blankinship, S. L. "Coal Solubilization by Enhanced Enzyme Activity in Organic Solvents," presented at 11th Symp. on Biotechnology for Fuels and Chemicals, Colorado Springs, CO, May 8-12, 1989, and submitted to *Appl. Biochem. Biotechnol.*

Scott, C. D.; Woodward, C. A.; Thompson, J. E.; Huggins, J. L. "Coal Solubilization by Enzymes in Organic Solvents," presented at 11th Symp. on Biotechnology for Fuels and Chemicals, Colorado Springs, CO, May 8-12, 1989

Woodward, J.

Lee, N. E.; Lima, M.; Woodward, J. "Hydrolysis of Cellulose by a Mixture of *Trichoderma reesei* Cellobiohydrolase and *Aspergillus niger* Endoglucanase," *Biochem. Biophys. Acta* **967**, 437-40 (1988)

Lee, N. E.; Woodward, J. "Reversible Inactivation and Fluorescence Changes of Cellobiohydrolase I by Dithiothreitol Treatment," presented at Natl. Meet. Am. Chem. Soc., Miami Beach, FL, September 10-15, 1989, and to be published in *Proc. Natl. Meet. Am. Chem. Soc.*, Miami Beach, FL

Lee, N. E.; Woodward, J. "Kinetics of the Adsorption of *Trichoderma reesei* Cellulase to DEAE-Macrosorb," presented at 196th Natl. Meet. Am. Chem. Soc., Los Angeles, September 25-30, 1988, and published in *J. Biotech.* **11**, 75-82 (1989)

Woodward, J. "Synergism in Cellulase Systems," submitted to *Biomass*

Woodward, J. "The Synergistic Mechanism of Enzymatic Cellulose Hydrolysis," presented at Dep. of Biochemistry, Univ. of Tennessee, Knoxville, November 2, 1988

Woodward, J. "Cellulase Biochemistry and Biotechnology," presented at Dep. of Biochemistry, Univ. of Oxford, Oxford, Great Britain, November 23, 1988, and at Bio-Products Div., Eastman Kodak Co., Rochester, NY, December 13, 1988

Woodward, J. "Immobilized Cellulases for Cellulose Utilization," *J. Biotech.* **11**, 299-312 (1989)

Woodward, J. "Methods of Immobilization of Microbial Cells," *J. Microbiol. Methods* **8**, 91-102 (1988)

Woodward, J.; Bales, J. C. "The Adsorption of *Trichoderma reesei* C30 Cellulase Protein to Protein Adsorbents: Application to Cellulase Recovery and Reuse," *Bioproducts and Bioprocesses*, Springer-Verlag, Berlin Heidelberg, 1989, pp. 87-101

Woodward, J.; Lee, N. E. "Cellulase Recovery: Problems and Potential," presented at Annu. Review Meet. on Ethanol from Biomass, SERI, Golden, CO, January 17-19, 1989

Woodward, J.; Lee, N. E.; Carmichael, J. S. "Denaturation and Renaturation of *Trichoderma reesei* C30 Cellobiohydrolase I," presented at TRICEL 89 Int. Symp. on *Trichoderma* Cellulases, Technical Univ., Vienna, September 14-16, 1989

Woodward, J.; Lee, N. E.; Carmichael, J. S.; McNair, S. L.; Wichert, J. M. "Comparison of the Hydrolytic Activity and Fluorescence of Native, Guanidine Hydrochloride-Treated and Renatured Cellobiohydrolase I from *Trichoderma reesei*," submitted to *Biochem. Biophys. Acta*

Woodward, J.; Lewid, B.; Miracle, G.; Greenbaum, E. "Comparative Study of the Photochemistry of Chloroplast Membranes, and Photosystem II Particles," *Appl. Biochem. Biotechnol.* **20**, 259-65 (1989)

Woodward, J.; Lima, M.; Lee, N. E. "The Role of Cellulase Concentration in Determining the Degree of Synergism in the Hydrolysis of Microcrystalline Cellulose," *Biochem. J.* **255**, 895-99 (1988)

Woodward, J.; McNair, S. L.; Lee, N. E. "Utilization of *p*-Nitrophenylcellobioside as a Substrate for Monitoring the Characteristics of Purified Cellobiohydrolase I from *Trichoderma reesei*," presented at 11th Symp. on Biotechnology for Fuels and Chemicals, Colorado Springs, CO, May 8-12, 1989, and to be published in *Book of Abstracts, 11th Symp. on Biotechnology for Fuels and Chemicals, Colorado Springs, CO, May 8-12, 1989*

Woodward, J.; Reeves, M.; Greenbaum, E.; Allison, D. "Effect of Exogenously Added Manganese Chloride on the Operational Stability of the Chloroplast-Ferredoxin-Hydrogenase System," *Enzyme Microb. Technol.* **10**, 123-28 (1988)

Wright, A. L.

Wright, A. L. *Summary of Pretest Iodine Chemistry and Transport Calculations for the Radioiodine Test Facility (RTF) Experiments*, draft report under review, September, 1989

Wright, A. L.; Wilson, J. H.; Arwood, P. C. *Summary of Posttest Aerosol Code-Comparison Results for LWR Aerosol Containment Experiment (LACE) LA6*, LACE TR-068, ORNL/M-660, March, 1989

Wright, A. L. "Aerosol Transport Code Validation: LACE Project Results," presented at Review of ORNL Severe Accident Research, ORNL, Oak Ridge, TN, February 21, 1989

Wright, A. L. "ACE "Phase-B" Code-Comparison Efforts: RTF Pretest," presented at Advanced Containment Experiments Technical Advisory Committee Meet., Chicago, May 9-11, 1989

Wright, A. L.; Fish, B.; Beahm, E. C.; Weber, C. F. "The Chemistry and Behavior of Iodine-Vapor Species in Nuclear Plant Air-Monitoring Sampling Lines," presented at 20th DOE/NRC Nuclear Air Cleaning Conf., Boston, August 22-25, 1988, and to be published in *Proc. 20th DOE/NRC Nuclear Air Cleaning Conf.*, Boston, Aug. 22-25, 1988

Wright, A. L.; Fish, B. R.; Beahm, E. C.; Weber, C. F. "The Chemistry and Behavior of Iodine Vapor Species in Nuclear Plant Air-Monitoring Sampling Lines," presented at 20th DOE/NRC Nuclear Air Cleaning Conf., Boston, August 22-25, 1988

Wymer, R. G.

Wymer, R. G. "Uranium," *Compton's Encyclopedia, Compton's Learning Co., Div. of Encyclopedia Britannica, 1988*

Youngblood, E. L.

Youngblood, E. L.; du Mont, S. P.; Helms, R. E. "Upgrade of the Radioactive Air Emissions Systems at Oak Ridge National Laboratory," presented at 20th DOE/NRC Nuclear Air Cleaning Conf., Boston, August 22-25, 1988, and published in *Proceedings of the 20th DOE/NRC Nuclear Air Cleaning Conference*, NUREG/CP-0098, CONF-880822, vol. 2, The Harvard Air Cleaning Laboratory, May, 1989

Patents

C. A. Burtis and W. F. Johnson

"Device and Method for Automated Separation of a Sample of Whole Blood into Aliquots," U.S. Patent 4,847,205 (July 11, 1989)

C. A. Burtis, W. F. Johnson, and W. A. Walker

"Rotor for Processing Liquids Using Movable Capillary Tubes," U.S. Patent 4,835,106 (May 30, 1989)

E. Greenbaum

"Method and Apparatus for Nondestructive In Vivo Measurement of Photosynthesis," U.S. Patent 4,789,436 (December 6, 1988)

P. A. Haas

"Process for Electrolytically Preparing Uranium Metal," Statutory Invention Registration H659 (August 1, 1989)

A. J. Mattus and R. D. Spence

"Process for Impregnating a Concrete or Cement Body with a Polymeric Material," U.S. Patent 4,828,761 (May 5, 1989)

T. C. Quinby

"Method for Producing Refractory Nitrides," U.S. Patent 4,800,183 (January 24, 1989)

C. D. Scott and G. W. Strandberg

"Fluidized-Bed Bioreactor Process for the Microbial Solubilization of Coal," U.S. Patent 4,846,964 (July 11, 1989)

T. C. Scott and R. M. Wham

"Surface Area Generation and Droplet Size Control in Solvent Extraction Systems Utilizing High Intensity Electric Fields," U.S. Patent 4,767,515 (August 30, 1988)

O. K. Tallent, K. E. Dodson, and E. W. McDaniel

"Method and Composition for Immobilization of Waste in Cement-Based Materials," Statutory Invention Registration H660 (August 1, 1989)

J. Woodward

"Recovery and Reuse of Cellulase Catalyst in an Enzymatic Cellulose Hydrolysis Process," U.S. Patent 4,840,904 (June 20, 1989)

Consultants

T. Beresovski

Affiliation: Independent
Expertise: Chemical Sciences/Quality Assurance
Duration: 50 d
Section/Program: Department of Defense

P. R. Bienkowski

Affiliation: University of Tennessee
Expertise: Chemical Engineering
Duration: 12 months
Section/Program: Energy Research

S. L. Blankinship

Affiliation: Independent
Expertise: Chemical Sciences
Duration: 90 d
Section/Program: Energy Research

J. M. Cece

Affiliation: Independent
Expertise: Transportation
Duration: 30 d
Section/Program: Transportation Technology

C. F. Coleman

Affiliation: ORNL Retiree
Expertise: Chemical Sciences
Duration: 1 year
Section/Program: Chemical Development

R. S. Crouse

Affiliation: Independent
Expertise: Chemical Sciences
Duration: 30 d
Section/Program: Radiochemical Processing

D. D. Ensor

Affiliation: Tennessee Technology University
Expertise: Chemistry
Duration: 20 d
Section/Program: Chemical Development

S. Gonzales

Affiliation: Independent
Expertise: Geology
Duration: 50 d
Section/Program: Waste Management

J. M. Gossett

Affiliation: Cornell University
Expertise: Chemical and Civil Engineering
Duration: 11 d
Section/Program: Department of Defense

H. L. Greene

Affiliation: University of Akron
Expertise: Chemical Engineering
Duration: 12 d
Section/Program: Environmental Control Technology

K. E. Hines

Affiliation: Independent
Expertise: Chemical Sciences
Duration: 25 d
Section/Program: Waste Management

R. H. Jones

Affiliation: Independent
Expertise: Transportation
Duration: 60 d
Section/Program: Waste Management

B. Kanehiro

Affiliation: Independent
Expertise: Chemical Engineering
Duration: 45 d
Section/Program: Waste Management

K. J. Lee

Affiliation: KAIST
Expertise: Nuclear Chemical Engineering
Duration: 1 year
Section/Program: Waste Management

P. N. McCreery

Affiliation: Independent
Expertise: Transportation
Duration: 25 d
Section/Program: Waste Management

S. L. McNair

Affiliation: Independent
Expertise: Chemical Engineering
Duration: 90 d
Section/Program: Waste Solidification

R. Narayan

Affiliation: Purdue University
Expertise: Coal Chemistry
Duration: 4 d
Section/Program: Energy Research

J. A. Phillips

Affiliation: Lehigh University
Expertise: Biotechnology
Duration: 4 d
Section/Program: Energy Research

P. E. Potter

Affiliation: Cincinnati University
Expertise: Geology/Statistics
Duration: 20 d
Section/Program: Waste Management

J. D. Rollins

Affiliation: Independent
Expertise: Nuclear Engineering
Duration: 12 d
Section/Program: Waste Management

H. C. Savage

Affiliation: Independent
Expertise: Quality Assurance
Duration: 10 d
Section/Program: Process Development

K. Slaughter

Affiliation: EA Corporation
Expertise: Chemical Engineering
Duration: 90 d
Section/Program: Environmental Control Technology

E. H. Snider

Affiliation: Independent
Expertise: Hazardous Waste
Duration: 2 d
Section/Program: Engineering Development

O. K. Tallent

Affiliation: Independent
Expertise: Chemist
Duration: 50 d
Section/Program: Waste Management

C. O. Thomas

Affiliation: University of Tennessee
Expertise: Chemical Engineering
Duration: 23.5 d
Section/Program: Environmental Control Technology

D. I. C. Wang

Affiliation: Massachusetts Institute of Technology
Expertise: Chemical Engineering
Duration: 4 d
Section/Program: Energy Research

W. A. Weigand

Affiliation: Illinois Institute of Technology
Expertise: Chemical Engineering
Duration: 4 d
Section/Program: Energy Research

Advisory Committee

Member	Affiliation	Term
L. Dean Eyman	Executive Director, Waste Policy Waste Policy Institute 1900 Kraft Drive Blacksburg, Virginia 24060	1988-1990
Gerhart Friedlander	Five Lorraine Court Smithtown, New York 11787	1988-1991
C. P. Leslie Grady, Jr.	Environmental Systems Engineering 501 Rhodes Research Center Clemson University Clemson, South Carolina 29634-0919	1988-1991
James R. Merriman	Vice President, Applied Technology Oak Ridge Gaseous Diffusion Plant Building K-1001, MS 7133 Oak Ridge, Tennessee 37831	1990-1992
Kenneth J. Richards	Vice President, Technology Division Kerr-McGee Corporation Post Office Box 25861 Oklahoma City, Oklahoma 73125	1986-1990
Martin J. Steindler	Director, Chemical Technology Division Argonne National Laboratory 9700 South Cass Avenue Argonne, Illinois 60439	1988-1991

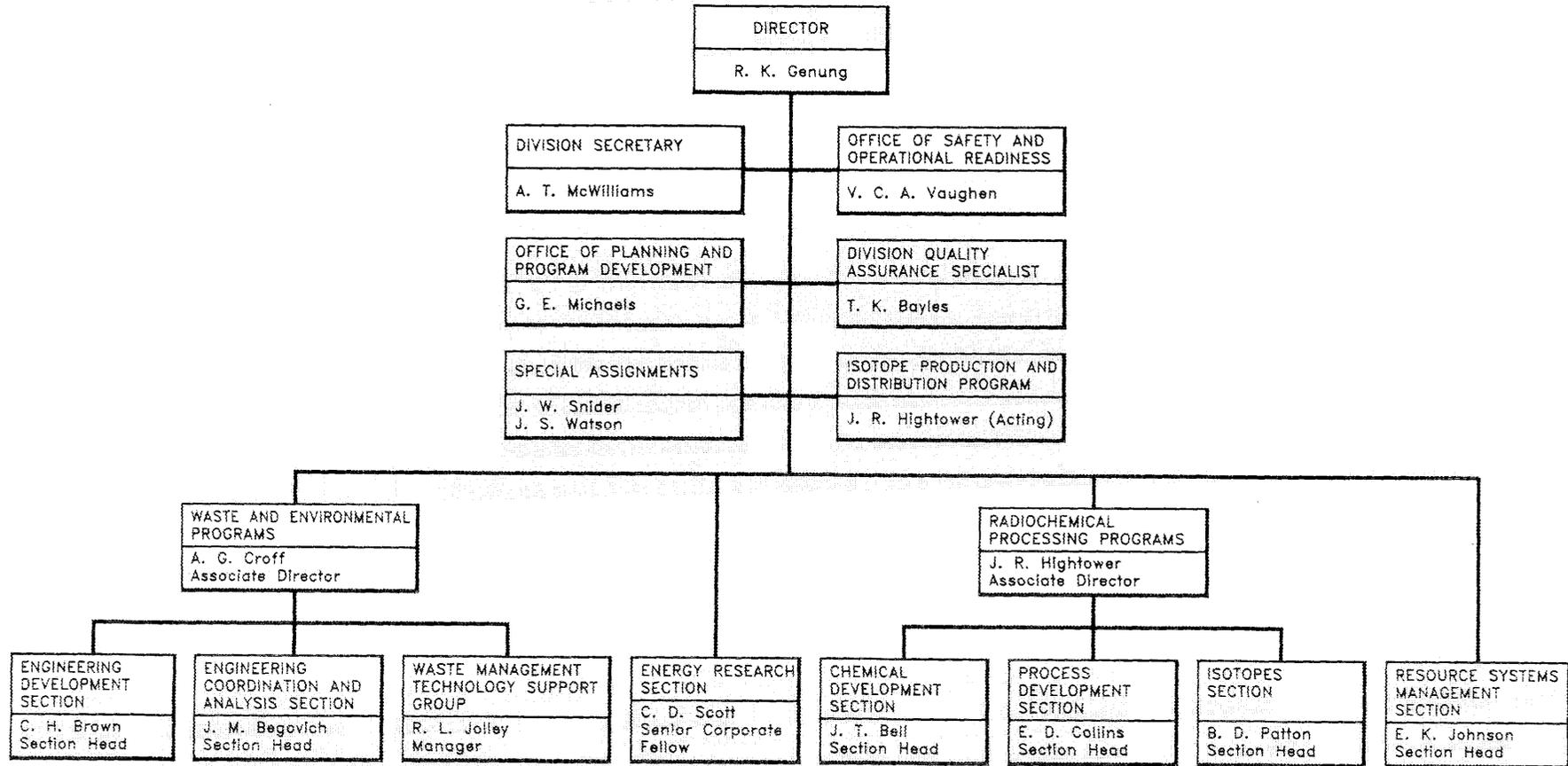
Chemical Technology Division Staffing Level and Financial Summary, FY 1989

	Funding (\$10 ³)	Chem Tech Scientific/Technical ^a (person-years)	Total Scientific/Technical ^b (person-years)
DOE Programs			
Coal	718	4.3	4.3
Gas	0	0.0	0.0
Petroleum	0	0.0	0.0
Breeder Reactor Systems	742	2.2	2.5
Remedial Action	409	1.3	1.3
Nuclear Fuel Cycle	0	0.0	0.0
Defense Waste and By-Products Management	3,893	16.8	18.9
Nuclear Waste Fund	8,034	15.1	16.1
Transportation	0	0.0	0.0
Industrial	156	0.3	0.3
Multisector	305	1.8	1.8
Materials Production	6,466	25.3	26.7
Environmental R&D	394	1.5	1.7
Basic Energy Sciences	10,171	36.7	43.7
Environmental Compliance	<u>1,311</u>	<u>6.1</u>	<u>7.2</u>
Subtotal	32,599	111.4	124.5
DOE-Related Programs			
Nuclear Regulatory Commission	1,012	4.4	4.5
Other DOE and contractors	761	2.9	3.1
DOE-ORO Contractors	49	0.1	0.1
State	7	0.0	0.0
Other Energy Systems plants	<u>1,160</u>	<u>3.5</u>	<u>4.0</u>
Subtotal DOE-related programs	2,989	10.9	11.7
Total DOE programs	35,588	122.3	136.2
Work for Others			
State and local government	320	1.5	1.7
Other federal (excluding NRC)	7	0.0	0.0
Department of Defense (Waste Management)	234	1.0	1.0
Other Department of Defense	<u>2,078</u>	<u>6.0</u>	<u>6.3</u>
Total Work for Others	2,639	8.5	9.0
Total	38,227	130.8	145.2

^aChem Tech research staff supported by the division in FY 1989.

^bTotal research staff supported by Chem Tech funding in FY 1989, including personnel on loan to Chem Tech from other divisions.

CHEMICAL TECHNOLOGY DIVISION



Internal Distribution

1. W. S. Aaron
2. T. J. Abraham
3. H. L. Adair
4. C. W. Alexander
5. W. J. Armento
6. R. J. Andermann
7. T. D. Anderson
8. B. R. Appleton
9. R. C. Ashline
10. S. I. Auerbach
11. D. H. Ault
12. P. A. Balo
13. G. M. Banic
14. R. T. Barnett
15. O. A. Basaran
16. L. Bates
17. T. K. Bayles
18. E. C. Beahm
19. J. E. Beaver
20. J. M. Begovich
- 21-25. J. T. Bell
26. C. E. Benson
27. D. E. Benker
28. J. B. Berry
29. J. E. Bigelow
30. W. D. Bond
31. C. D. Bopp
32. W. D. Box
33. J. Bradbury
34. S. D. Brandel
35. R. W. Brewer
36. W. A. Brooke
37. C. B. Brooks
- 38-42. C. H. Brown
43. J. K. Bryson
44. W. D. Burch
45. C. A. Burtis
46. G. E. Butterworth
47. C. H. Byers
48. B. L. Byrum
49. D. O. Campbell
50. G. D. Campbell
51. K. B. Campbell
52. J. B. Cannon
53. R. L. Cline
54. F. R. Chattin
55. S. D. Clinton
56. H. D. Cochran
- 57-61. E. D. Collins
62. J. L. Collins
63. S. P. Cooper
64. R. M. Counce
65. C. G. Cowart
66. R. Craig
- 67-76. A. G. Croff
77. B. H. Davison
78. D. W. DePaoli
79. S. M. DePaoli
80. J. R. DeVore
81. L. S. Dickerson
82. T. L. Donaldson
83. D. D. Drake
84. B. G. Eads
85. B. Z. Egan
86. B. D. Faison
87. A. J. Farmer
88. R. L. Fellows
89. J. J. Ferrada
90. D. M. Ferren
91. P. W. Fisher
92. C. L. Fitzgerald, Sr.
93. C. W. Forsberg
94. D. Foster, Jr.
95. E. B. Foster
96. E. J. Frederick
97. G. F. Galloway
98. W. R. Gambill
99. A. S. Garrett, Jr.
100. S. E. Gentry
- 101-125. R. K. Genung
126. S. E. Gheesling
127. J. R. Gibson
128. S. M. Gibson
129. T. M. Gilliam
130. R. W. Glass
131. J. C. Glover, Jr.
132. H. A. Glovier, Jr.
133. H. W. Godbee
134. R. M. Gove
135. E. Greenbaum

136. W. S. Groenier
 137. R. G. Grubb
 138. P. A. Haas
 139. S. W. Hadley
 140. K. W. Haff
 141. R. Hall
 142. M. T. Harris
 143–147. J. R. Hightower
 148. D. R. Hines
 149. V. T. Hinkel
 150. D. W. Holladay
 151. P. E. Hollenbeck
 152. F. J. Homan
 153. T. D. Hylton
 154. D. W. Jared
 155. C. L. Johnson
 156–180. E. K. Johnson
 181–185. R. L. Jolley
 186. J. Jones
 187. G. D. O'Kelley
 188. D. S. Joy
 189. R. R. Judkins
 190. J. L. Kasten
 191. S. V. Kaye
 192. O. L. Keller, Jr.
 193. J. M. Kennerly
 194. T. E. Kent
 195. A. D. Kelmers
 196. A. H. Kibbey
 197. S. G. Kimmett
 198. C. D. King
 199. L. J. King
 200. F. G. Kitts
 201. J. A. Klein
 202. J. B. Knauer, Jr.
 203. T. S. Kress
 204. A. M. Krichinsky
 205. E. H. Krieg, Jr.
 206–220. C. E. Lamb
 221. R. R. Laxson
 222. D. D. Lee
 223. N. E. Lee
 224. C. W. Leinart
 225. K. H. Lin
 226. T. B. Lindemer
 227. A. S. Loebel
 228. T. F. Lomenick
 229. R. A. Lorenz
 230. S. B. Ludwig
 231. F. C. Maienschein
 232. J. C. Mailen
 233. A. P. Malinauskas
 234. R. C. Mason
 235. A. J. Mattus
 236. L. E. McBride
 237. V. S. McCauley
 238. W. C. McClain
 239. E. W. McDaniel
 240. C. P. McGinnis
 241. H. O. McNabb
 242. L. E. McNeese
 243. J. R. Merriman
 244. L. J. Mezga
 245. G. E. Michaels
 246. J. M. Miller
 247. H. T. Milton
 248. M. I. Morris
 249. O. B. Morgan
 250. J. E. Mrochek
 251. F. R. Mynatt
 252. T. E. Myrick
 253. J. W. Nehls, Jr.
 254. E. Newman
 255. J. P. Nichols
 256. R. E. Norman
 257. R. P. Norris
 258. K. J. Notz
 259. M. F. Osborne
 260. M. M. Osborne
 261. I. W. Osborne-Lee
 262. C. L. Ottinger
 263. D. B. Owsley
 264. J. H. Paehler, Jr.
 265. G. W. Parker
 266. J. C. Parker
 267. J. R. Parrott, Jr.
 268. W. L. Pattison
 269–273. B. D. Patton
 274. M. Payne
 275. R. L. Pearson
 276. W. H. Pechin
 277. J. D. Pendleton, II
 278. C. E. Pepper
 279. J. J. Perona
 280. M. Petek
 281. J. Petty
 282. H. X. Phillips
 283. W. W. Pitt
 284. R. B. Pope
 285. H. Postma
 286. M. L. Poutsma
 287. D. J. Pruett

288. L. P. Pugh
 289. D. W. Ramey
 290. J. E. Ratledge
 291. R. R. Rawl
 292. M. E. Reeves
 293. W. J. Reich
 294. D. E. Reichle
 295. D. R. Reichle
 296. H. D. Renner
 297. C. R. Richmond
 298. A. L. Rivera
 299. C. S. Robinson
 300. S. M. Robinson
 301. J. W. Roddy
 302. B. R. Rodgers
 303. R. E. Rosenbaum
 304. R. G. Ross
 305. T. H. Row
 306. J. H. Saling
 307. R. Salmon
 308. C. A. Sampson, Jr.
 309. T. L. Sams
 310. F. M. Scheitlin
 311. R. M. Schultz
 312. O. W. Scates
 313-322. C. D. Scott
 323. T. C. Scott
 324. M. B. Sears
 325. L. B. Shappert
 326. S. A. Sherrow
 327. R. C. Shipwash
 328. W. E. Shockley
 329. J. T. Shor
 330. W. D. Shults
 331. R. R. Shoun
 332. S. P. N. Singh
 333. W. G. Sisson
 334. J. W. Snider
 335. R. D. Spence
 336. R. G. Stacy
 337. B. W. Starnes
 338. J. H. Stewart, Jr.
 339-344. M. G. Stewart
 345. J. O. Stiegler
 346. S. N. Storch
 347. G. W. Strandberg
 348. J. H. Swanks
 349. Q. D. Swicegood
 350. D. W. Swindle
 351. P. A. Taylor
 352. R. D. Taylor
 353. J. E. Thompson
 354. J. G. Tracy
 355. L. M. Toth
 356. D. B. Trauger
 357. R. E. Upchurch
 358-362. V. C. A. Vaughen
 363. R. J. Vedder
 364. C. C. Waggoner, Jr.
 365. J. F. Walker
 366. M. W. Wankerl
 367. D. A. Waters
 368. M. J. Welch
 369. T. D. Welch
 370. B. W. Welles
 371. J. S. Watson
 372. R. M. Wham
 373. J. T. Wiggins
 374. J. S. Wike
 375. D. F. Williams
 376. J. H. Wilson
 377. K. Wilson
 378. J. Woodward
 379. A. L. Wright
 380. R. G. Wymer
 381. J. W. York, III
 382. E. L. Youngblood
 383. L. A. Zevenbergen
 384. A. Zucker
 385. Biology Library
 386-387. Central Research Library
 388. Laboratory Shift Supervisor
 389. ORNL Y-12 Technical Library,
 Document Reference Section
 390-391. Laboratory Records Department
 392. Laboratory Records, RC
 393. ORNL Patent Section

External Distribution

394. W. D. Adams, U.S. Department of Energy, Environmental Restoration Division, P.O. Box 2001, Oak Ridge, TN 37831-8540
 395. G. A. Arlotto, Director, Division of Engineering, U.S. Nuclear Regulatory Commission, Washington, DC 20555

396. D. E. Bailey, Director, Division of Fuels and Reprocessing, Office Facilities, Fuel Cycle and Test Program, NE 471, DOE, Washington, DC 20545
397. W. W. Ballard, Jr., Manager, Nuclear Systems and Technology, Battelle Pacific Northwest Laboratories, Battelle Boulevard, Richland, WA 99352
398. C. B. Barlett, Regulatory Analysis Branch, U.S. Nuclear Regulatory Commission, RM N-112, MS NL-007, Washington, DC 20555
399. J. E. Baublitz, Office of Terminal Waste Disposal and Remedial Action, DOE, NE-20, Washington, DC 20545
400. E. S. Beckjord, Director, Office of Nuclear Regulatory Research, U.S. Nuclear Regulatory Commission, Washington, DC 20555
401. D. J. Beecy, Director, Office of Planning and Environment, DOE, FE-52, GTN, Washington, DC 20545
402. M. J. Bell, Chief, Low-Level Waste Branch, Division of Waste Management, MS 623-55, U.S. Nuclear Regulatory Commission, Washington, DC 20555
403. A. K. Benear, Westinghouse Materials Company of Ohio, 7400 Willey Road, Fernald, OH 45030
- 404–405. R. M. Bernero, Deputy Director, Office of Nuclear Material Safety and Safeguards, U.S. Nuclear Regulatory Commission, Washington, DC 20555
406. W. W. Berry, Stearns-Roger Corporation, 4500 Cherry Creek Drive, Box 5888, Denver, CO 80217
407. W. R. Bibb, DOE-ORO, P.O. Box 2001, Oak Ridge, TN 37831
408. F. Block, Research Director, Albany Research Center, P.O. Box 70, Albany, OR 97321
409. E. M. Bowers, Richland Operations Office, Richland, WA 99352
410. D. R. Brown, Energy Programs Division, DOE-ORO, P.O. Box 2001, Oak Ridge, TN 37831-8612
411. C. W. Burger, Energy Programs Division, DOE-ORO
412. J. L. Burnett, Division of Chemical Sciences, Office of Basic Energy Sciences, DOE, MS J-309, Washington, DC 20545
413. J. H. Cavendish, National Lead of Ohio (Fernald), Chemical Department, Cincinnati, OH 45202
414. Richard Chitwood, DOE, 1000 Independence Ave., E-456 (GTN), Washington, DC 20545
415. G. R. Choppin, Florida State University, Department of Chemistry, Tallahassee, FL 32306
416. J. F. Clark, Director, Magnetic Fusion Energy, Office of Energy Research, DOE, Washington, DC 20545
417. F. E. Coffman, I.T. Corporation, Regional Office, 312 Directors Drive, Knoxville, TN 37923
418. J. A. Coleman, Office of Terminal Waste Disposal and Remedial Action, NE-24, DOE, Washington, DC 20545
419. J. W. Cooke, DOE-ORO, P.O. Box 2001, MS-2010, Oak Ridge, TN 37831
420. C. Cooley, U.S. Department of Energy, Forrestal Bldg., EM-50, 1000 Independence Ave., Washington, DC 20545
421. B. L. Crynes, The University of Oklahoma, Room 107, Carson Engineering Center, 202 West Boyd, Norman, OK 73019

422. O. L. Culberson, 12124 Warrior Trail, Knoxville, TN 37922
423. F. L. Culler, Office of the President, Electric Power Research Institute, P.O. Box 10412, Palo Alto, CA 94303
424. W. J. Danker, DOE, RW-321, Forrestal Building, 1000 Independence Ave., Washington, DC 20585
425. K. Debelak, Vanderbilt University, Box 1700, Station B, Nashville, TN 37235
426. J. E. Dieckhoner, Officer of Nuclear Materials Resource Management, DOE, DP-534/GTN, Washington, DC 20545
427. Dr. E. A. Ebadian, Florida International University, College of Engineering and Applied Science, School of Engineering, University Park, FL 33190
428. J. J. Eberhardt, DOE, Energy Conversion and Utilization Technologies Program, CE-121, Washington, DC 20585
429. C. A. Eckert, Department of Chemical Engineering, 113 Roger Adams Laboratory, Box C-3, 1209 W. California Street, Urbana, IL 61801-3791
430. R. L. Egli, Energy Research and Development, DOE-ORO, P.O. Box 2001, Oak Ridge, TN 37831-8600
431. A. E. Evans, Office of Military Applications, DP-242.2, DOE, Washington, DC 20545
432. D. E. Erb, Office of Nuclear Energy, DOE-HQ, NE-40 (GTN), Washington, DC 20545
433. Captain Michael Elliott, Headquarters Air Force Engineering and Services Center, Tyndall Air Force Base, FL 32403-6001
434. David Enegess, Envirogen, Inc., 181 Cherry Valley Rd., Princeton, NJ 08540
435. G. Friedlander, 5 Lorraine Court, Smithtown, NY 11787
436. C. Frank, U.S. Department of Energy, Forrestal Building, EM-50, 1000 Independence Ave., Washington, DC 20585
437. J. W. Gatrell, DOE, Office of Nuclear Energy, NE-22, Washington, DC 20545
438. M. R. Ghate, DOE, Morgantown Energy Technology Center, P.O. Office Box 880, Morgantown, WV 26505
439. J. C. Giddings, Department of Chemistry, Henry Eyring Building, University of Utah, Salt Lake City, UT 84112
440. C. P. L. Grady, Jr., Rhodes Engineering Research Center, Clemson University, Clemson, SC 29634-0919
441. K. V. Gilbert, TWSO, Rocky Flats Area Office, DOE, Box 928, Golden, CO 80401
442. L. E. Graham, Chief, Process Simulation and Analysis Section, Morgantown Energy Technology Center, P.O. Box 880, Morgantown, WV 26505
443. J. M. Haffey, Oak Ridge Associated Universities, Oak Ridge, TN 37830
444. H. J. Hale, Office of Civilian Radioactive Waste Management, DOE, RW-222, Forrestal Building, Room 7F-051, Washington, DC 20585
445. H. D. Harmon, Savannah River Laboratory, E.I. du Pont de Nemours & Co., Aiken, SC 29801
446. M. M. Heiskell, DOE-ORO, P.O. Box 2001, MS-2116, Oak Ridge, TN 37831

447. J. E. Helt, Chemical Technology Division, CMT/205, 9700 South Cass Ave., Argonne, IL 60439-4837
448. E. E. Hoffman, DOE-ORO, P.O. Box 2001, MS-2115, Oak Ridge, TN 37831
449. W. A. Hoffman, Director, Oak Ridge Science Semester, GLCA, Denison University, Main Street, Granville, OH 43023
450. L. C. Ianniello, Director, Division of Materials Sciences, Office of Basic Energy Sciences, DOE, Washington, DC 20545
451. Lisa Jarr, Morgantown Energy Technology Center, P.O. Box 880, Morgantown, WV 26507-0880
452. B. Jinkerson, ORAU, University Programs Division, P.O. Box 117, Oak Ridge, TN 37830
453. E. A. Jordon, Office of Defense Waste and Transportation Management, DP-122, DOE, Washington, DC 20545
454. S. Kasper, Morgantown Energy Technology Center, P.O. Box 880, MS- C04, Morgantown, WV 26507-0880
455. L. Keay, Energy Utilization Research, DOE, CE-121/Forrestal, Washington, DC 20585
456. K. A. Klein, DOE, NP-30, 6B-128/FORS, Washington, DC 20585
457. C. Kouts, Office of Civilian Radioactive Waste Management, DOE, RW-323, 7F-043/FORS, Washington, DC 20585
458. K. O. Laughon, Division of Nuclear Power Development, DOE, NE-30, Washington, DC 20545
459. Captain Ed Marchand, Headquarters Air Force Engineering and Services Center, Tyndall Air Force Base, FL 32403-6001
460. R. S. Marianelli, Director, Division of Chemical Sciences, Office of Basic Energy Sciences, DOE, Washington, DC 20545
461. Michael D. Martin, Product Manager, Votator AMCA International, P.O. Box 35600, Louisville, KY 40232
462. E. F. Mastal, Planning Analysis Group, Office of Nuclear Energy, DOE, NE-53, Washington, DC 20545
463. Connor Matthews, DOE-ORO, P.O. Box 2001, Oak Ridge, TN 37831
464. M. H. McFadden, Waste Management and Transportation Development Division, Albuquerque Operations Office, DOE, Box 5400, Albuquerque, NM 87115
465. G. V. McGurl, DOE, Pittsburgh Energy Technology Center, P.O. Box 10940, Pittsburgh, PA 15236-0940
466. W. H. McVey, Division of Nuclear Power Development, Office of Spent Fuel Management and Reprocessing Systems, DOE, NE-471, Washington, DC 20545
467. R. F. Moorer, Conservation Programs, DOE, Forrestal Bldg., CE- 341, Washington, DC 20585
468. Y. Nakamura, JPL, Systems Analysis Section, California Institute of Technology, 4800 Oak Grove Drive, Pasadena, CA 91103
469. J. H. Nehls, Research Management, ER-122, DOE-ORO, P.O. Box 2001, Oak Ridge, TN 37831-2116
470. R. J. Neuhold, Director of Energy Research Reactor Operations, Office of Facilities, Fuel Cycle, and Test Programs, NE-473, DOE, Washington, DC 20545

471. Col. R. F. Olfenbuttel, Battelle, 505 King Ave., Columbus, OH 43201
472. M. L. Payton, DOE, Office of Civilian Radioactive Waste Management, RW-132, Washington, DC 20585
473. L. K. Price, Technical Services Division, DOE-ORO, P.O. Box 2001, Oak Ridge, TN 37831-2115
474. L. L. Radcliffe, Energy Programs and Support Division, DOE-ORO, MS 2024, Oak Ridge, TN 37831
475. Dr. Nathaniel Revis, Oak Ridge Research Institute, 113 Union Valley Road, Oak Ridge, TN 37830
476. K. J. Richards, Kerr-McGee Corporation, P.O. Box 25861, Oklahoma City, OK 73125
477. Gene L. Rogers, 1005 Southbridge Lane, Chattanooga, TN 37405
478. J. Roberts, DOE, CHO Transportation Programs Office, 9800 S. Cass Ave., Argonne, IL 60439
479. J. Roth, Department of Chemical Engineering, Vanderbilt University, Nashville, TN 37240
480. Clifford M. Rowland, Cheroy-Burrell, P.O. Box 35600, Louisville, KY 40232
481. Dr. Gary Saylor, Center for Environmental Biotechnology, 10515 Research Drive, University of Tennessee, Knoxville, TN 37932
482. A. Schneider, School of Nuclear Engineering, Georgia Institute of Technology, Atlanta, GA 30332
483. P. C. Scott, Office of Fossil Energy, U.S. Department of Energy, FE-14, Washington, DC 20545
484. J. S. Siegel, DOE, Fossil Energy Programs, GTN/FE-20, Washington, DC 20545
485. M. A. Singer, DOE, FE-30, GTN, Washington, DC 20545
486. R. C. Sleeman, U.S. Department of Energy, Environmental Restoration Division, P.O. Box 2001, Oak Ridge, TN 37831-8540
487. B. A. Smith, Chief, Processing Branch, Office of Nuclear Materials Production, DP-133.2, DOE, Washington, DC 20545
488. D. R. Spence, Energy Programs Division, DOE-ORO, P.O. Box 2001, Oak Ridge, TN 37831-8612
489. W. Spindel, Executive Secretary, Office of Chemistry and Chemical Technology, National Research Council, 2101 Constitution Ave. NW, Washington, DC 20418
490. M. J. Steindler, Argonne National Laboratory, 9700 S. Cass Ave., Argonne, IL 60439
491. R. Stein, Office of Civilian Radioactive Waste Management, RW-30, Forrestal Building, DOE, Washington, DC 20585
492. D. K. Stevens, Associate Director, Office of Basic Energy Sciences, DOE, ER-10, GTN, Washington, DC 20545
493. F. D. Stevenson, Processes and Techniques Branch, ER-142, Division of Chemical Sciences, Office of Basic Energy Sciences, DOE, MS J- 209, GTN, Washington, DC 20545
494. J. Stiegler, Transportation Technology Center, Sandia National Laboratories, Department 4550, P.O. Box 5800, Albuquerque, NM 87185
495. P. M. Stone, Magnetic Fusion Energy, Office of Energy Research, ER-6/FORS, DOE, Washington, DC 20585

496. E. Svenson, DOE, Forrestal Bldg., RW-33, 7F-052/FOR, Washington, DC 20585
497. A. L. Taboas, U.S. Department of Energy, Argonne Area Office, 9800 South Cass Ave., Argonne, IL 60439
498. D. W. Tedder, Georgia Institute of Technology, School of Chemical Engineering, Atlanta, GA 30332-0100
499. L. J. Thiboddeaux, Director, Hazardous Waste Research Center, Louisiana State University, Baton Rouge, LA 70803
500. I. L. Thomas, Division of Materials Sciences, Office of Basic Energy Sciences, DOE, GTN/ER13, Washington, DC 20545
501. C. H. Trimble, Director, Process Development, Molycorp, Inc., 461 South Boylston Street, Los Angeles, CA 90017
502. J. E. Van Beek, Westinghouse Hanford Operations, P.O. Box 800, Richland, WA 99352
503. Captain Catherine Vogel, Headquarters Air Force Engineering and Services Center/RDVW, Building 1117, Tyndall Air Force Base, FL 32403-6001
504. S. J. Waddle, Energy Programs, DOE-ORO, P.O. Box 2001, Oak Ridge, TN 37831-2115
505. J. E. Walsh, Jr., DOE, Fossil Energy Programs, GTN/FE-10, Washington, DC 20545
506. D. K. Walter, Biofuels & Municipal Waste Technology, CE-341, Forrestal Bldg., Washington, DC 20585
507. H. L. Watson, Deputy Staff Director, Subcommittee on Energy Development and Applications, Committee on Science and Technology, U.S. House of Representatives, B-374, Rayburn House Office Building, Washington, DC 20515
508. Barry J. Weissman, Pall Power Generation Division, 2200 Northern Blvd., East Hill, NY 11548
509. G. F. Williamson, Westinghouse Hanford Operations, P.O. Box 800, Richland, WA 99352
510. S. J. Wolfe, Energy Programs, DOE-ORO, P.O. Box 2001, Oak Ridge, TN 37831-2067
511. R. W. Wood, Division of Pollutant Characterization and Safety Research, Office of Health and Environmental Research, DOE, MS E-201, Washington, DC 20545
512. L. Yarborough, DOE-ORO
513. John G. Yates, U.S. Department of Energy, ER-42, Room 3F-091, Forrestal Building, 1000 Independence Ave., SW, Washington, DC 20585
514. R. C. Yates, Processing Branch, Office of Nuclear Materials Production, DP 133.2, U.S. Department of Energy, Washington, DC 20545
515. Office of Assistant Manager, Energy Research and Development, DOE-ORO, P.O. Box 2001, Oak Ridge, TN 37831
516. Institute of Energy Analysis, P.O. Box 117, Oak Ridge, TN 37830
517. ORAU Technical Library, MERT Division, P.O. Box 117, Oak Ridge, TN 37830
518. W. R. A. Goossens, Head of the Department of Chemical Engineering, Studiecentrum voor Kernenergie, S.C.K./C.E.N., Boeretang 200, B-2400, Mol, Belgium
- 519-528. Office of Scientific and Technical Information, P.O. Box 62, Oak Ridge, TN 37831

Figure

ORNL drawing or photograph number

2.1	89-1234	5.11	89A-636
2.2	87-1047R	5.12	88-14938
2.3	89A-1180	5.13	88-1356
2.4	89A-1184R	5.14	88-583R
2.5	89A-790	5.15	88-1131
2.6	89A-791	5.16	88-1128
2.7	88A-1141R	5.17	89A-9269
2.8	90A-5920R	5.18	87-17981R
2.9	88A-14883R	5.19	88-1369R
2.10	89A-1246	5.20	89A-1041
2.11	90A-69R3	5.21	89A-963
2.12	89A-1247	5.22	89A-1341
2.13	89A-1242	5.23	89A-1342
2.14	89A-446R	5.24	89A-1343
2.15	90A-114R	5.25	89A-1344
2.16	90-170	5.26	89A-1345
2.17	88-1302	5.27	89A-1346
2.18	89A-1245R2	5.28	89A-1347
2.19	89-473	5.29	89A-1101
2.20	89A-248R	5.30	88-1393R
2.21	89A-640R	5.31	89A-967
3.1	K/G-88-1602	5.32	88-1367
3.2	90A-8	5.33	88-674R2
3.3	7938-88	5.34	88-119
4.1	1281-89	5.35	89A-698
4.2	9867-87	5.36	89A-547
4.3	83-1323	5.37	89A-14766
4.4	KIPH 89-0777	5.38	1839-89
5.1	89-9282	5.39	88-1256
5.2	89A-824	5.40	7145-89
5.3	86-8682R	5.41	7144-89
5.4	89-912	5.42	7143-89
5.5	89-913	5.43	89A-833
5.6	89A-190	5.44	89-18488
5.7	89A-192	5.45	89-18493
5.8	89A-12498	6.1	89-101R
5.9	87-1910R2	6.2	87-1290
5.10	89A-635	Org. chart	90A-201R