OAK RIDGE NATIONAL LABORATORY LIBRARIES
3 4456 0548286 7

CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION

155

[redacted]

ORNL-3663
C-44a – Nuclear Technology – Materials
M-3679 (35th ed.)

OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

JUNE 1964

CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION
LIBRARY LOAN COPY
DO NOT TRANSFER TO ANOTHER PERSON
If you wish someone else to see this document, send in name with document and the library will arrange a loan.

OAK RIDGE NATIONAL LABORATORY
operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

Downgrade to Unclassified After July 7, 1965

[redacted]

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

[redacted]

ORNL-3663

Contract No. W-7405-eng-26

OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

JUNE 1964

DATE ISSUED

July 7, 1964

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

OAK RIDGE NATIONAL LABORATORY LIBRARIES

3 4456 0548286 7

[redacted]

[redacted]

[redacted]

[redacted] [redacted]

CONTENTS

REACTOR DEVELOPMENT..... 1
GAS-COOLED REACTOR PROGRAM..... 1
Steam-Graphite Reaction Experiment..... 1
Coating of Sol-Gel Oxide Particles..... 1
Instantaneous Fission-Gas Release from Coated Particles and Graphite-Matrix Fuels..... 1
Instantaneous Fission-Gas Release from UO_2 (C-1)..... 2
GCR-ORR Loop No. 1..... 2
LITR Capsule Irradiations..... 2
ORR Poolside Capsule Irradiations..... 2
Eight-Ball Experiment..... 3
X-Ray Diffraction Studies on Pyrolytic-Carbon-Coated Particles..... 3
FUNDAMENTAL HEAT TRANSFER AND FLUID MECHANICS..... 3
Critical Heat Fluxes for Natural Convection of Water in Blocked Vertical Channels..... 3
Magnetohydrodynamic Studies of Vortex Flow..... 3
Numerical Calculation of Hydromagnetic Stability..... 4
Potential Vortex Flow near a Stationary Disk..... 4
POWER REACTOR FUEL PROCESSING..... 5
Chemical Analysis of Advanced Reactor Fuels..... 5
THORIUM UTILIZATION PROGRAM..... 5
Separation Chemistry of Protactinium from Thorium in Molten Chloride Salts..... 5
Thorium Alloy Development..... 5
Sol-Encapsulation Studies..... 5
Extension of the Sol-Gel Process to Newer Fuel Types: Thorium Nitride..... 6
Permeability of Vibratorily Compacted Fuels..... 6
Irradiation of Thorium Fuels: Powder-Packed Oxide Fuels..... 6
Irradiation of Thorium Fuels: Thorium Alloy Capsules..... 6
Cleanup of Kilorod Facility..... 6
Thorium-Uranium Fuel Cycle Development Facility (TUFCDF)..... 6
Equipment Development for Thorium-Uranium Fuel Cycle Development Facility..... 7
Coated-Particle Development Laboratory..... 7
Process Development and Evaluation: Study of the Preparation of Sol-Gel Particles of Controlled Porosity..... 7
Pellet Fabrication..... 7
Sol-Gel Microsphere Preparation and Evaluation..... 7
MOLTEN-SALT REACTOR PROGRAM..... 8
Molten-Salt Reactor Experiment..... 8
Reactor Analysis..... 8
Development of Components and Systems..... 9
MSRE Pump Program..... 9
Instrumentation Development..... 10

[redacted] [redacted]

[redacted] [redacted]

NUCLEAR SAFETY..... 10
Melting Experiments in TREAT Facility..... 10
Behavior of High-Temperature Materials Under Irradiation..... 10
Characterization and Control of Accident-Released Fission Products..... 11
Behavior of Fission Products Released During In-Pile Destruction of Reactor Fuels..... 11
Properties of Aerosols Released from Overheated Reactor Fuels..... 13
Nuclear Safety Pilot Plant..... 13
Reactor Containment Handbook..... 15
Nuclear Safety Information Center..... 15
WASTE DISPOSAL..... 15
Clinch River Study..... 15
Mineral Exchange Studies..... 15
Disposal in Deep Wells..... 16
High-Activity-Waste Solidification: Engineering Work..... 16
High-Activity-Waste Solidification: Laboratory Work..... 17
Immediate-Activity Waste..... 17
Low-Activity Waste: Pilot-Plant Work..... 17
Low-Activity Waste: Laboratory Work..... 18
Disposal in Natural Salt Formations..... 19
Engineering, Economic, and Safety Evaluation..... 19
ARMY PACKAGE POWER REACTOR PROGRAM..... 20

PHYSICAL RESEARCH..... 20
PHYSICS AND MATHEMATICS..... 20
High-Energy Electronuclear Machines: The Separated-Orbit Cyclotron..... 20
Medium-Energy Nuclear Physics: 22-Mev Protons..... 21
Medium-Energy Nuclear Physics: Heavy Particles..... 21
Medium-Energy Nuclear Physics: Theory..... 21
Cyclotron Operations: The Isochronous Cyclotron..... 22
High-Voltage Experimental Program: Elastic Scattering of 17- to 21-Mev Neutrons from ^{12}C..... 23
High-Voltage Experimental Program: Alpha-Alpha Correlations in the Reaction $^{7}Li(d,n)2^{4}He$..... 23
Stable Isotopes Development..... 23
Special Separations..... 24
CHEMISTRY..... 24
Analysis of Molten Salts by Electrochemical Methods..... 24
Protactinium Chemistry..... 25
Radiation Chemistry of Organic Materials: High Polymers..... 25
Spectrophotometry of Solutions at High Temperature..... 26
Physical Chemistry of Molten Salts: $HF-H_2O$ Equilibrium with Molten Fluorides..... 26
Physical Chemistry of Molten Salts: Viscosities in the BeF_2-LiF System..... 27
Target Preparation..... 27

[redacted] [redacted]

[redacted]

TRANSURANIUM-ELEMENT PRODUCTION..... 28
Chemical Process Development..... 28
Curium Recovery Facility..... 28
HFIR Target Development: Prototype Target Irradiation..... 28
HFIR Target Development: Plutonium Target Fabrication..... 29
Transuranium Facility..... 29
Corrosion..... 29
Analytical Chemistry Development..... 29
METALLURGY AND MATERIALS..... 30
Preparation of Pure Materials: Single-Crystal LiF..... 30
Preparation of Pure Materials: Single-Crystal Li_2BeF_4..... 30
Reactions at Metal Surfaces..... 30
Deformation of Crystalline Solids..... 31
Spectroscopy of Ionic Media..... 31
Theory of Alloying..... 32
Structure of Metals..... 32
Fundamental Research in X-Ray Diffraction..... 32
Crystal Physics..... 33
Fundamental Investigation of Radiation Damage in Solids: Effect of Lead on the Room-Temperature Colorability of KCl..... 34
Fundamental Investigation of Radiation Damage in Solids: A Capacity Microphone for Ultrasonic Measurements at Megacycle Frequencies..... 34
CONTROLLED THERMONUCLEAR RESEARCH..... 34
Plasma Physics..... 34
Vacuum..... 35

BIOLOGY AND MEDICINE..... 35
MOLECULAR- AND CELLULAR-LEVEL STUDIES..... 35
Physics of Tissue Damage: Measurements of Diffusion-Pump Oil Deposition with Radioactive Tracers..... 35
Physics of Tissue Damage: Optical Properties of Graphite in the Far Ultraviolet Region..... 35
TERRESTRIAL AND FRESH-WATER ECOLOGY..... 35
Analysis of Growth Responses of Two Sedges on White Oak Lake Bed..... 35
Environmental Influences on Responses of Metaphosphate Glass Dosimeters..... 36
Effects of Growth Rate and Ionizing Radiation on Ribonucleic Acids in Mimosa Seedlings..... 37
Analog Computer Simulation of a Five-Compartment Ecosystem..... 37
Evaluation of Fission Product Distribution and Movement in White Oak Drainage System..... 38

ISOTOPE DEVELOPMENT..... 38
Solvent Extraction of Fission Products..... 38
Radioisotope Research and Development..... 39
Radioisotope and Stable Isotope Production..... 39

[redacted]

[redacted]

OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

JUNE 1964

This Status and Progress Report summarizes the unclassified portion of the Laboratory's work. Some of the topics are included every month, but the majority are reported on a bimonthly schedule.

REACTOR DEVELOPMENT

GAS-COOLED REACTOR PROGRAM

Steam-Graphite Reaction Experiment. — An experiment in which the rate of reaction of steam and graphite was determined was terminated after approximately 1000 hr of operation. The cylindrical graphite specimen, which was machined from an EGCR graphite column, was exposed to superheated steam at temperatures ranging from 1300 to 1550°F. Weight measurements indicated that the graphite sleeve, which originally weighed 9994 g, lost 4944 g during the test period.

Visual inspection of the sleeve before it was removed from the test section showed that it was still in the original cylindrical shape; however, the first attempt to remove the sleeve from the test section caused it to crumble. The broken pieces had virtually no compressive strength.

Coating of Sol-Gel Oxide Particles. — Spherical particles of ThO_2–8 wt % UO_2 (uranium highly enriched) derived from the sol-gel microsphere process were successfully coated with pyrolytic carbon by the fluidized-bed process. The nominal diameter of the particles is 200 μ, and the two-layer coating is 100 μ thick. The first layer of pyrolytic carbon (approximately 35 μ) was deposited at 1400°C at an average deposition rate of 84 μ/hr; the second was applied at 1800°C at an average deposition rate of 10 μ/hr. Evaluation of these coated particles showed no evidence of reaction between the oxide and the carbon coating. Portions of this material were encapsulated for irradiation testing.

Instantaneous Fission-Gas Release from Coated Particles and Graphite-Matrix Fuels. — Particles in capsule B9-17 (OR-138 duplex) were irradiated at 2500°F to 16.2 at. % uranium burnup. The fractional fission-gas release rates were from 0.3×10^{-5} to 1.2×10^{-5} at the end of the irradiation. There was a slight increase in fission-gas release with burnup. During the last two days of irradiation, the temperature was cycled between 2200 and 2500°F. No fission-gas release bursts from coating ruptures could be detected during temperature cycling, and the release was temperature dependent. The capsule has been removed from the reactor and opened in the hot cell.

[redacted]

Capsule B9-18, containing 3M-RP-25 duplex coated particles, was installed in the reactor.

Instantaneous Fission-Gas Release from UO_2 (C-1). — Automatic control instruments and equipment were installed in the C-1 facility to allow variation of both the neutron flux and the specimen temperature in a controlled sine wave pattern.

All the new controls have "fail-safe" features, and the facility is designed to operate unattended. The equipment can still be operated in a steady-state manner, but the new controls will allow almost unlimited variations of operating modes. For example, the neutron flux can be varied while the air cooling automatically maintains a constant temperature. By studying the amplitude and phase shift of the fission-gas release, we hope to develop our defect-trap model for fission-gas release and to experimentally establish the parameters in the mathematical form of the model.

Single-crystal UO_2 specimens C1-13 were installed in the facility, and testing of the system is in progress.

GCR-ORR Loop No. 1. — Experimental assembly 13 was removed from the loop on June 1 and transferred to a hot cell for postirradiation examination. The experiment was intended to investigate fission product release to a circulating helium stream from a small hole in a stainless-steel-clad UO_2 fuel element. The fission-gas release rate during the experiment was abnormally low in comparison with that of a previous, similar experiment, indicating that the hole provided in the cladding had become partially or completely blocked.

During the scheduled ORR shutdown which began June 1, both loop compressors were replaced with rebuilt units. A flow-measuring instrument and throttle valve were installed in the loop carbon-trap system for better flow control.

LITR Capsule Irradiations. — Two capsules (L-CP7 and L-CP8) containing pyrolytic-carbon-coated particles in each of four compartments continued operation at indicated fuel central temperatures between 1200 and 2100°F. Both uranium carbide and uranium-thorium carbide mixtures are included in the capsules. Estimated burnups have now reached 9.3 to 18.0 at. % of the heavy metal (U + Th).

ORR Poolside Capsule Irradiations. — Two capsules (03A-6 and 04-8) containing EGCR-vendor fuel materials continued to operate at design conditions. The cladding surface temperatures are 1280 and 1540°F maximum.

Irradiation of one sweep capsule (01A-7) containing 6-cm-diam fueled-graphite spheres was completed June 1, and the capsule was removed for postirradiation examination. This capsule had operated since February at a design central temperature of 2400°F and had reached an estimated burnup (at. % of total heavy metal) of 1.96%. Irradiation of the sweep capsule (05-8) installed in April continued. This capsule also contains 6-cm-diam fueled-graphite spheres. Testing of two new capsules (01-8 and 08-8) was started June 14. Two of the spheres in capsule 01-8 were made by drilling 0.272-in. holes in solid ATJ graphite spheres (6-cm-diam), filling the holes with loose coated particles, and closing them with threaded graphite plugs. The remaining sphere in capsule 01-8 and the three spheres in capsule 08-8 consist of molded graphite-fuel matrices in unfueled shells.

Eight-Ball Experiment. — Irradiation of the fifth eight-ball experimental assembly (F1-8B-5) containing 1-1/2-in.-diam spheres fueled with pyrolytic-carbon-coated UC_2 and $(U,Th)C_2$ particles and specimens of pyrolytic-carbon-coated B_4C continued at design conditions. The indicated central temperature for the top sphere continued to be approximately 2000°F. This assembly is scheduled for irradiation to high burnup.

X-Ray Diffraction Studies on Pyrolytic-Carbon-Coated Particles. — Pyrolytic-carbon coatings, 50 μ thick, deposited on uranium carbide particles at 1400°C at rates of 1.5 to 164 μ/hr were examined by x-ray diffraction for interlayer spacing, C_0; apparent crystallite size, L_c; and degree of preferred orientation. The size, L_c, was determined from the relationship

$$L_c = 0.89\lambda/(\beta \cos \theta) ,$$

where λ is the wavelength, β is the width at half height in 2θ radians of the (00.2) reflection, and θ is the Bragg angle. Values of C_0 and L_c were determined by standard diffractometer methods.

The results indicate that L_c exhibits a minimum at a deposition rate of about 11 μ/hr. The data for coatings applied at 1300 to 1800°C indicate that L_c increases with increasing deposition temperature. However, L_c also appears to be affected by the type of particle used (UC_2 vs UO_2-ThO_2) and parameters other than deposition rate and temperature.

FUNDAMENTAL HEAT TRANSFER AND FLUID MECHANICS

Critical Heat Fluxes for Natural Convection of Water in Blocked Vertical Channels. — A dimensionless correlation was developed for predicting the minimum critical heat flux for natural convection of water in a heated vertical channel closed at the bottom and open at the top to a liquid supply:

$$(\phi_c)_{min} = \frac{\lambda D_e^{3/2} \, [\rho_v g(\rho_\ell - \rho_v)]^{1/2}}{8L_h} ,$$

in which ϕ_c is the critical heat flux, λ is the latent heat of vaporization, D_e is the equivalent diameter of the channel, ρ_ℓ is the liquid density, ρ_v is the vapor density, g is the gravitational acceleration, and L_h is the heated length.

Few experimental data pertinent to this poor heat transfer situation are available, and no predictive techniques were proposed previously. Additional data are being obtained with tubes 0.086 to 0.245 in. in inside diameter and 6 to 48 in. in heated length operated at atmospheric pressure.

Magnetohydrodynamic Studies of Vortex Flow. — Hydrodynamic experiments were performed with a 1.1-in.-diam stainless steel tube in which the vortex was generated by tangential injection through two 0.013-in. slits in an effort to elucidate further the mechanism of transition to

instability on the concave wall. Based on binocular microscopic observation (40X) through plastic end walls of a dye trace injected just downstream of one of the slits, it appears as though the initial phase of transition is an oscillatory two-dimensional expansion and contraction of the wall layer that may be associated with incipient separation of the decelerated jet flow. The fully developed instability, however, is three dimensional; the dye trace rotates in an irregular manner about a circumferential axis. The apparent oscillatory behavior of this phenomenon may be the result of convection of a stationary perturbed state with reference to the fixed point of observation, rather than a truly time-dependent fluctuation. If such is the case, the instability is probably of the Taylor-Göertler type, as assumed in the theoretical analyses. A transparent plastic model is being fabricated that will enable observation in both radial and axial directions, to establish more conclusively the nature of the observed instability.

Numerical Calculation of Hydromagnetic Stability. – Results have been reported (ORNL-3590, pp. 4–5) concerning the effect of an axial magnetic field on the stability of a viscous, electrically conducting fluid confined between concentric conducting cylinders for the type of flow produced by rotating the inner cylinder. These experiments were extended to include outward radial flow superimposed on the primary rotating flow. Such flow might be obtained by making the cylinders of a porous material and pumping the fluid through the inner cylinder, across the annular gap, and out through the outer cylinder.

Radial Reynolds moduli from 0 to 60, Hartmann moduli from 0 to 100, and cylinder radius ratios from 0.25 to 0.8 were considered. Generally, an increase in either the radial Reynolds modulus or the Hartmann modulus tends to stabilize the flow, but the effects are not additive. That is, a high radial Reynolds modulus decreases (but does not eliminate) the stabilizing effect of the magnetic field, and similarly, a high Hartmann modulus decreases the stabilizing effect of the radial flow.

Potential Vortex Flow near a Stationary Disk. – In the design and operation of confined vortex systems, the flow in the boundary layers on the stationary surfaces is not satisfactorily understood at present. In jet-driven vortexes of the type proposed for certain advanced energy-conversion applications, this flow amounts to a significant fraction of the total for the system. To aid in understanding this phenomenon, a study is being initiated in which the interaction between a vortical flow and a stationary disk, perpendicular to the axis of the vortex, will be examined. A two-dimensional swirling flow closely approximating a potential vortex will be generated experimentally by passing water radially inward through a rotating drum made of expanded aluminum honeycomb. (A potential vortex is one in which angular momentum is conserved and the tangential velocity varies inversely with the radius.) The flow is exhausted through a cylinder made of fine screen on the axis of the drum. A preliminary system was assembled and is being used to aid in the final design.

In addition to the experimental work, an analytical study will be made. In this study the complete radial and tangential equations of motion will be integrated simultaneously on an analog computer. A number of flow visualization techniques, such as the tellurium dye method, the

use of fluorescein dye under ultraviolet light, and the use of electrochemiluminescent materials, will be investigated in conjunction with the experimental work.

POWER REACTOR FUEL PROCESSING

Chemical Analysis of Advanced Reactor Fuels. — A laboratory gas chromatograph for nonfluoride corrosive samples has been constructed in which the sample measuring system, the partition column, and the detector are made of glass with Teflon fittings. This instrument is being tested for possible construction defects prior to introduction of corrosive samples.

THORIUM UTILIZATION PROGRAM

Separation Chemistry of Protactinium from Thorium in Molten Chloride Salts. — Experiments employing neutron irradiation of mixtures of $AlCl_3$ and $ThCl_4$ to explore the possibility of removing bred ^{233}Pa by volatilization have continued. Mixtures containing 100, 50, 10, and 0.5 mole % $ThCl_4$ in $AlCl_3$ were used. No transfer of ^{233}Pa was found in the mixtures containing the larger proportions (100 and 50 mole %) of $ThCl_4$. The larger quantities of $AlCl_3$ assisted in the transfer of the ^{233}Pa. When the quartz receiver was at a temperature lower (below 100°C) than the heated mixture (160 to 300°C), it was possible to revolatilize the protactinium. When the receiver region was held at a temperature above 350°C, the ^{233}Pa appeared to remain fixed to the quartz. Volatilization of ^{233}Pa was demonstrated when the concentration of $ThCl_4$ in $AlCl_3$ was 0.5 mole %.

Thorium Alloy Development. — A "getter" alloy of 50 at. % Zr—Ti was made in order to purify the argon atmosphere used in creep tests on thorium alloyed with 5% indium. It is thought that an impure atmosphere was responsible for the surprisingly poor creep test results found last month. No tests have been made yet with argon purified by the getter alloy.

Sol-Encapsulation Studies. — Studies of the encapsulation of sol drops in organic polymer coatings and drying to obtain gel particles were continued. When a 6 M Th-U oxide sol was encapsulated with a cellulose acetate butyrate as the coating polymer, the shell walls collapsed somewhat during air drying, producing an irregularly shaped particle; but the shrinkage and collapse were much less than had been experienced in earlier studies with 2 M Th-U oxide sol.

Studies of the dispersion of carbon in order to prepare sols of 8.4 M carbon—2.0 M Th-U oxide were continued. Tests were made with an ultrasonic vibrator (Bronson Sonifier) to disperse four commercial channel-black carbons at various pH's, and with several commercial surfactants to promote wetting of the carbon. The results indicate that the Sonifier disperses the carbon and that nine of the surfactants give nonviscous dispersions, 8 M in carbon. Brookfield viscosities of 6 to 184 centipoises at pH's of 1.6 to 6.7 were obtained. The most effective system found for reducing viscosity and promoting dispersion was Renex 30 surfactant with Excelsior carbon. Samples were prepared for further evaluation at ORNL.

Extension of the Sol-Gel Process to Newer Fuel Types: Thorium Nitride. — Several additional preparations of thorium nitride were made by the reaction of thoria-carbon gel spheres with nitrogen at about 1800°C. The composition of the nitride, as determined by x-ray analysis, varied from batch to batch. In some cases, only ThN lines were found; in others, additional lines attributable to ThO_2 were found.

Permeability of Vibratorily Compacted Fuels. — Permeability measurements with two sample compacts indicate that not all the porosity within the compacts is involved in the flow of fluid through the samples. In displacement experiments with both miscible and immiscible liquid pairs, only part of the liquid in the voids was displaced. Void volumes and water permeabilities were also reported for the same samples.

Calculations were made showing that the gas-diffusion rates reported in GEAP-4397 on ORNL NRX samples A-1, A-2, C-3, and C-4 are consistent with values expected from theory.

Irradiation of Thorium Fuels: Powder-Packed Oxide Fuels. — Rods of essentially the same design as the 19-in.-long rods for the Brookhaven National Laboratory criticality tests are being fabricated in the Kilorod Facility. These rods will undergo irradiation testing in the MTR or in the ETR. Because of faulty operation of the compacting equipment, three of seven rods prepared during the last report period fractured at the bottom weld. During this report period, five more rods were prepared. Of these, three passed the autoclave test for corrosion resistance. The top welds of the other two were badly corroded. Shipment of the three acceptable rods has been postponed pending results of an investigation of the cause of failure of the other two rods.

Irradiation of Thorium Fuels: Thorium Alloy Capsules. — It was reported previously (ORNL-3647) that one of the two remaining thorium-zirconium alloys prepared in capsules by the Advanced Technology Laboratories was removed from the GETR after a few hours in-pile because of anomalous thermocouple behavior. The other has now been removed after 18 days in-pile because of fission-gas release to the reactor cooling water. Of the six capsules originally prepared, all were removed for one reason or another; consequently, this irradiation study has been terminated.

Cleanup of Kilorod Facility. — The Kilorod Program is now completed. The facility is being cleaned out, and final material balances are being made. Mixed 3% UO_2–ThO_2 collected from samples retained during the program and from system cleanout are being dissolved to permit the analyses required for the material balance. Complete dissolution of the mixed oxide requires about 48 hr in boiling (about 118°C) 13 $\underline{M}$ HNO_3–0.04 $\underline{M}$ NaF–0.04 $\underline{M}$ Al^{3+}–0.0025 $\underline{M}$ Hg^{2+}.

Thorium-Uranium Fuel Cycle Development Facility (TUFCDF). — Title II design work by the architect-engineer was authorized April 3, 1964; project completion is scheduled for early 1967.

It is estimated that title II design is 15% complete. Eleven drawins have been approved. Details of design are being developed for the in-cell fire-protection system, and numerous other design details were developed and sent to the architect-engineer.

The design of the sol-gel and oxide fabrication equipment is proceeding somewhat slower than indicated by the early-start schedule. Major

items receiving attention during this report period were the vibratory compaction device, the fuel-rod cleaning device, the rod-capping machine, and fuel-rod completion devices.

Equipment Development for Thorium-Uranium Fuel Cycle Development Facility. — Plans call for testing radiant-heating panels for use in the sol drying step in TUFCDF. Sol drying in the Kilorod Facility was carried out in trays in a conventional oven. Use of radiant-heating panels should greatly increase the drying rate attainable. Gel fragments prepared by radiant-heat sol drying were cross sectioned and examined. The examination showed the fragments to be dense, with, perhaps, a very slight increase in porosity in fragments produced at the highest drying rate tested. As a test of their suitability, fragments prepared at the design drying rate proposed for the TUFCDF will be sized and vibratorily packed.

Coated-Particle Development Laboratory. — The design of a facility for the investigation of microsphere production and remote pyrolytic-carbon coating on a pilot-plant scale is now complete. A laboratory in Building 4508 will be modified to accept equipment for this work. In addition, the design of several items of equipment is nearly complete. The facility will be used for the collection of engineering data that will be applied to the design of remotely operated equipment for the fabrication of fueled-graphite elements.

Process Development and Evaluation: Study of the Preparation of Sol-Gel Particles of Controlled Porosity. — A small percentage of well-controlled and -distributed porosity may be desirable in fuel particles. Such porosity could provide traps for fission product gases and for gases that may be incorporated in the fuel during fuel preparation. A study of ways to incorporate such porosity in thorium oxide—base fuels was started. In one method, a high-fired oxalate-derived ThO_2 was added to thoria sol prepared by the basic sol-gel method. Since the porosity of the ThO_2 was known, it was postulated that measurement of the quantity added could be used to predict the change in porosity of the final material. However, the added ThO_2 segregated from the sol in spite of agitation, giving a nonuniform distribution of porosity. In another method, carbon was added to the thoria sol. Porosity was produced when the carbon was removed by oxidation. However, the amount and distribution of porosity were not well controlled.

Pellet Fabrication. — Sintering tests have been completed on the pellets to be used in flux-measurement rods for the criticality tests at Brookhaven National Laboratory. Production runs are now in progress. In the first production run, 59 of 60 pellets were acceptable. Production of the 0.425-in.-OD × 0.300-in.-long (Th—3 wt % $^{233}U)O_2$ pellets (approximately 300 in all) will require another two or three weeks to complete. In addition to the pellets, 100 wafers (0.425 in. OD × 0.050 in. long) are required. Sintering tests for the wafers are being conducted along with pellet production in order that wafer fabrication may begin immediately after pellet production.

Sol-Gel Microsphere Preparation and Evaluation. — Evaluation of the chemical and physical properties of the 150- to 300-μ-diam ThO_2—8% $^{235}UO_2$ microspheres prepared for irradiation tests has been completed. Part of the microspheres were calcined in air and part in Ar—4% H_2. Densities,

porosities (open and closed), specific surface areas, oxygen-to-uranium ratios, carbon contents, and total gas evolved at 1200°C in vacuum were determined. The oxygen-to-uranium ratio, evolved gas, and carbon content were notably different in samples calcined in air from those calcined in Ar–4% H_2. The oxygen-to-uranium ratio (calculated by assuming the thorium to be present as ThO_2) was 2.52 in the air-calcined sample and 2.005 in the sample calcined in Ar–4% H_2. The evolved gas was 0.645 and 0.017 std cm^3/g in samples calcined in air and in Ar–4% H_2 respectively. Nearly 90% of the evolved gas was oxygen. The carbon content of the air-calcined microspheres was 80 ppm; the content of those calcined in Ar–4% H_2 was 410 ppm.

Studies of the design of columns for preparing the microspheres have continued. When scale-up of the tapered column from 1.5 in. to 3 in. minimum internal diameter was attempted, low flow velocity near the walls permitted undried sol droplets to fall out of the fluidized region, and excessive sol-droplet impingement on flow distributors caused droplet coalescence. The addition of a tapered core to give a linear velocity distribution over a 4-ft height of column appears to eliminate most of the problems associated with the coalescence of droplets on the flow distributors. Tangential solvent feeding to the column appears to eliminate the problem of undried sol-droplet fallout at the walls. Additional column configurations with tangential solvent feeding will be studied.

When gel microspheres are formed from sol droplets in the columns, water accumulates in the fluidizing-drying solvent at a rate such that about 1 liter of solvent is expended in producing about 40 g of microspheres. To avoid large inventories of solvent, and the attendant handling problems, a distillation system will be installed to remove water from a side stream on the solvent column.

MOLTEN-SALT REACTOR PROGRAM

Molten-Salt Reactor Experiment. — The MSRE reactor vessel and fuel pump were placed on the assembly jig, and the connecting piping and freeze flanges were welded to both units. The fuel-pump rotary element and the pump drive motor were installed.

Modifications to the heat exchanger shell were completed, and reassembly of the heat exchanger was started on the assembly jig.

Installation of the vent-house piping and installation of the south wall of the high-bay enclosure were started by the H. K. Ferguson Company. Progress continued on the installation of instrumentation and electrical systems.

The critical-path schedule showed the project to be 94.6% complete on June 1.

Reactor Analysis. — Studies of MSRE drain tank criticality factors were extended to include the possibility of uranium oxide precipitation during fuel storage. Precipitated solids would be expected to accumulate on metal surfaces; hence the entire deposit was assumed to occur on the coolant thimbles in order to provide the most favorable geometry for criticality. All the zirconium and uranium in the fuel salt were assigned to the deposit. The subcritical multiplication for this configuration

was found to be below that of a drain tank containing frozen homogeneous salt at 20°C. It was concluded that no criticality hazard should exist if UO_2 precipitates in the MSRE drain tanks.

Development of Components and Systems. — Fabrication of three control rods for use in the MSRE was completed, and a final test of the rods at 1200°F was started. A hardened steel worm and worm wheel in the control-rod drive mechanism operated through over 28,600 full-stroke cycles without difficulty. The weights of the worm and wheel were measured periodically, and after 21,000 cycles there was a loss of only 0.04 g from each. While the unit was regreased after each weighing, there was one sustained run of over 10,000 cycles without regreasing. We believe that previous failures of similar gears resulted from momentary overloading caused by malfunctions of other parts of the system and that elimination of these sources of overloading will also eliminate the gear failure.

Tests were conducted to determine the effect on freeze time of flow through the reactor drain valve during the freeze cycle. Salt flows of 0 to 200 in.^{3}/hr had little effect on the time required to freeze, which averaged 18 min. At flows of 320 in.^{3}/hr, the time required to freeze was only 32 min. No important difficulty is expected in freezing the drain valve in the MSRE.

The two INOR-8 prototype bayonets for the drain tank cooler have operated through over 1400 rapid quenches from 1200 to 220°F without difficulty. While some warping of the thimbles has occurred, there has been no evidence of cracking.

The problem of tube vibration in the MSRE heat exchanger was solved by installing a flow baffle at the inlet and lacing bars at each flow-baffle plate and at the U-bends in the tubes. The exchanger shell was lengthened, a 7 × 5 in. conical reducer was installed at the outlet, and the four outermost tubes were removed to reduce the overall pressure drop to an acceptable 27.4 psig at 1200 gpm.

Some of the procedures for maintaining the MSRE after power operation were tried in the drain tank cell. The crane was operated remotely, from the shielded control room, to handle the shield beams, the shield blocks, and the drain tank cooler assembly. The portable maintenance shield was used in demonstrating removal of a helium valve, handling of an auxiliary piping spool piece, and operation of four types of disconnects. Minor difficulties were encountered, but generally the maintenance system testing was successful.

MSRE Pump Program. — Preparation of the fuel and coolant pumps for the MSRE continued. The rotary element and drive motor combination for the fuel and coolant pumps were operated in the prototype pump hot-test facility for approximately 100 hr, and both combinations performed satisfactorily. No indications of the shaft rubbing previously reported were noted. The fuel pump combination was delivered to the MSRE site. The coolant pump combination is being prepared for delivery to the MSRE. Each of the pump rotary elements was operated in the cold shakedown stand before and after the hot tests to check the performance of the shaft, bearings, and seals.

Preliminary tests were made to calibrate the densitometer on the water mockup of the MK-II fuel pump tank preparatory to measuring the concentration of undissolved gas in the circulating water.

One of the two drive motors for the spare pumps was completed and delivered by the vendor.

Instrumentation Development. — We have initiated a program of assistance in the design, fabrication, and testing of an ultrasonic level probe. This probe, which is being developed by Aeroprojects, Inc., under contract to the AEC, will provide a single-point indication of molten-salt levels inside closed, weld-sealed vessels. The method of penetration of the vessel is of particular interest, since it permits ultrasonic energy to be transmitted through the walls of a vessel from a remotely located transducer. Fabrication of the probe assembly was started, and modifications of a level-test facility are in progress.

NUCLEAR SAFETY

Melting Experiments in TREAT Facility. — The third and fourth experiments of a series being performed in the TREAT reactor to study the release of fission products from fuel melted under transient heating conditions were completed. The miniature fuel specimens employed in these experiments contained 30 g of 10%-enriched high-density UO_2 clad with 35-mil-thick type 347 stainless steel. The fuel specimens were given a preliminary irradiation of 2×10^{16} neutrons/cm^2 (17 times the irradiation received during the transient) in order to study the behavior of longer-lived fission products rather than that of their short-lived precursors. The latter situation would prevail if all the fissions occurred during the transient, as in the first two experiments of the series. Specimens were preheated electrically to 800°C in air-steam atmospheres and exposed to similar reactor transients. The atmosphere in experiment 3 contained 3% steam by volume, and a 378-Mwsec transient at a 74-msec period was employed. In experiment 4 the atmosphere contained approximately equal volumes of steam and air, and the specimen was exposed to a 363-Mwsec transient at a period of 77 msec.

When the experimental equipment was disassembled, the fuel specimens were found to have a similar appearance. Approximately 75% of the UO_2 melted in each experiment. The melted UO_2 became porous or foamy, and approximately half the UO_2 in experiment 3 ran out of the unmelted pellet shell, while about 30% of the UO_2 ran out in experiment 4. The bulk density of the porous UO_2 was found to be 5 g/cm^3.

There was no shattering or fracturing of the fuel in either experiment. Analysis of gas from the experiment showed insignificant generation of hydrogen from the water—stainless steel reaction. Determinations of the release of fission products and of the transport of particles are in progress.

Behavior of High-Temperature Materials Under Irradiation. — The second or low-dose section of the current high-temperature materials experiment containing MgO, Al_2O_3, and BeO received a fast-neutron dose of 3.9×10^{21} nvt, and it is now undergoing hot-cell disassembly. The high-dose section will be removed from the ETR after a dose of about 7.5×10^{21} nvt has been accumulated.

Assemblies for the low-temperature unfueled carbide irradiation experiments are now being fabricated. Preliminary construction drawings

of the assemblies for the high-temperature carbide irradiation experiments have been completed.

Characterization and Control of Accident-Released Fission Products. — It has been demonstrated that radioiodine in the elemental form can be removed from gases quite efficiently by activated-charcoal adsorbers. However, under certain circumstances a small fraction of the iodine apparently can assume other forms, such as methyl iodide, which are more difficult to remove. Experiments indicate that the mixture of iodine vapor species emanating from an iodine source includes methyl iodide or a similar compound. Penetration of activated-charcoal adsorbers by these nonelemental forms of iodine is under continuing investigation. The effects of moisture and temperature on the adsorption of these forms of iodine by several types of charcoal have been studied. At room temperature and under dry conditions (relative humidity of 3 to 5%), all charcoal types tested are effective in removing these forms of iodine from air. However, at a relative humidity of 65 to 75%, a coke charcoal and a metal-impregnated charcoal are ineffective (efficiency of ~60% under the conditions of these tests), while cocoanut-shell charcoal exhibits an efficiency of 97%. This efficiency may be adequate, since the nonelemental forms comprise only a fraction of the iodine inventory. In similar experiments at 100°C, consistently high efficiencies (99.7 to 99.99+%) were observed under all conditions.

A method for characterizing radioactive aerosols has been developed, which interprets the distribution of ^{65}Zn particles in a series of uniform fiber filters in terms of the three main mechanisms of filtration — diffusion, interception, and inertial impaction. Quantitative analysis of the data in the diffusion regime to yield particle sizes consistent with those obtained from electron photomicrographs and from particle concentration techniques has been previously reported.

Several series of experiments have been performed in which the porosity of the filter mats has been raised from its original value of 0.72 to 0.94. Very thin filters were employed in the last set of experiments to interpret the initial sections of the curves depicting filtration efficiency vs depth of filter. A computer program is being prepared to show whether the data obtained are in conformity with existing theories of fiber filtration and where modifications in the theories will have to be made.

Behavior of Fission Products Released During In-Pile Destruction of Reactor Fuels. — In previous experiments, miniature UO_2 fuel elements have been melted in-pile in helium, air, moist helium, and moist air atmospheres to investigate the behavior of released fission products. In the next series of experiments the fuel will be melted in the presence of steam to simulate conditions in a pressurized-water reactor accident. The use of a steam atmosphere has necessitated considerable modification of the present equipment to provide controlled steam injection, to prevent premature condensation, and to condense the steam at the proper point. In all experiments the gas exit tube has served the dual function of (1) a delivery tube which transports released fission products to the filters, and (2) a diffusion tube which provides analytical information relative to particle size and the activities associated with the particles. Condensation in this tube is prevented by heating it with a steam jacket.

As the steam sweep gas emerges from the heated region of the exit tube, it passes through a condenser where the water is removed before the sweep gas passes through the filters. Separate radioassay of the condenser and filters will provide information on the fate of fission products during condensation. The filter unit has also been modified to include several improvements: (1) increased filter area, (2) provision of a means for testing each filter in place, and (3) changes to facilitate handling in the hot cell.

One of the objectives of these fuel destruction experiments is to determine the extent to which fission products released in a reactor accident will penetrate filters and adsorbers. Distributions of released fission products and uranium found in the off-gas system of fuel meltdown assemblies have been determined for all experiments. The table shows the amounts of material downstream of high-efficiency filters expressed as percent of the total amount of that type of material. The high-efficiency filter material is either Gelman type E glass, HV-70, or Flanders F-700. It is seen that only very small amounts of nonvolatile Ru, Sr, Ba, Zr, Ce, and U and slightly larger amounts of volatile I, Te, and Cs are usually found downstream of a high-efficiency filter. The large amount of

Material Found Downstream of First Intact High-Efficiency Filter
(Type E Glass, HV-70, or F-700)

Experiment[a]	Conditions	I	Te	Cs	Ru	Sr	Ba	Zr	Ce	U
G-1	Burned in air	5.4	0.067	21	25	0.43	0.26	0.31	4.9	0.17
G-3	Burned in air	1.5	0.07	14	1.1	0.47	0.38	0.18	0.7	
2	He atmosphere			0.13	0–0.10	0.008	4	0.005	0.024	
3	He atmosphere	0.8		10	0.002–0.010	0.19	0.019	0.0037	0.011	0.0003
5	He atmosphere	3.5	0.4	19	2.3	0.63	0.22	0.008	0.13	0.008
6	He atmosphere	15	22	12	0.66	0.20	0.18	0.08	0.027	0.012
7	He atmosphere	0.16	0.06	2.9	0.7	0.48	0.32	0.012	0.16	0.0051
9	He atmosphere	2.8	12	4.2	1.3	0.25	0.13	0.013	0.035	0.0033
10	High-flow He	1.3	0.022	2.5	0.0011	0.23	0.23	1.2	0.13	0.008
11	Very-high-flow He	2.9	0.19	2.7	0.022	0.023	0.17	0.0011	0.017	0.0006
12	Moist He	0.81	0.111	1.1	0.06	0.15	0.053	0.0030	0.022	0.005
13	Moist air	45	0.61	1.0	0.02–0.06	0.071	0.11	0.0012	0.019	0.002

[a]"G" prefix indicates experiments with uranium carbide–graphite fuel. Others were with UO_2.

iodine shown for experiment 13 is probably due to its oxidation by the moist air atmosphere. Thus the iodine was deposited to a lesser extent prior to the charcoal, and only 0.7% penetrated the first 2 in. of charcoal. Although the efficiencies of the filters are not very high, the amounts of fission products found downstream represent very small fractions of the total inventory because of the effectiveness of deposition and sorption processes prior to the filters.

Properties of Aerosols Released from Overheated Reactor Fuels. — A comparison of the effect of several meltdown parameters on the properties of the fission product aerosol released from melted UO_2 fuel materials has been made. The form of iodine produced and its behavior in conjunction with vaporized UO_2 were observed, as well as the effect of iodine concentration.

Stainless-steel-clad UO_2 fuel specimens irradiated to tracer level and to a high burnup level (7000 Mwd/ton) were melted to study the effect of fission product iodine concentration on the fraction of iodine in a penetrating form. The effect of trace atmospheric contaminants on iodine behavior was reported previously.

In the latest experiment an attempt was made to produce a realistic aerosol by use of tracer ^{131}I. A sample of carefully prepared molecular iodine having a known quantity of carrier iodine in addition to several millicuries of ^{131}I was sealed in a 1/4-in.-diam quartz ampoule and placed in the melting crucible under a layer of insulating ZrO_2 powder, in an effort to delay the heating of the quartz until after some UO_2 had melted and UO_2 vapor was already entering the tank. Premature release of iodine occurred in this experiment, so that melting of the specimen cladding and of the UO_2 occurred after the iodine vapor entered the containment tank.

Results of four varied experiments, which are compared on page 14, indicate that the UO_2 meltdown with simulated fission product iodine was reasonably successful in duplicating the aerosol from the 7000-Mwd/ton fuel, and the iodine behavior differed from that in the low-iodine-concentration experiments. The amount of iodine penetrating the 1.5-in.-thick charcoal bed was lower with the higher-iodine-concentration runs, C and D. The proportion of iodine in one particle size range (18 to 22 A) was low in the simulation run, and it possibly would have been higher if vaporization of the iodine and UO_2 had occurred simultaneously as planned. The results also show that a larger fraction of the iodine on the stainless steel aerosol tank wall was desorbed by the argon sweep gas in the high-iodine-concentration experiments. The desorption conditions employed in these experiments are not representative of accident conditions, and experiments are in progress to determine the extent of desorption under more realistic conditions.

Nuclear Safety Pilot Plant. — Much of the report period was spent upgrading the containment in order to obtain an acceptable leak rate prior to operation with fission products. The required changes included backwelding threaded connections and replacing unions with flanges. Several iron-body valves in the decontamination system which had undergone catastrophic attack by the decontaminating solution [0.4 $\underline{M}$ $(NH_4)_2C_2O_4$ + 0.2 $\underline{M}$ $NH_4C_2H_3O_2$ + 0.5 $\underline{M}$ H_2O_2, pH = 2.5] were replaced with stainless steel ball valves.

Additional results from the two UO_2 melting runs were received. The aerosol produced appears to consist of ultimate particles (some spherical, some cubic) ranging from 30 to 1000 A. These are highly agglomerated, with agglomerate sizes appearing to range from 0.1 to several microns. (It is often difficult to determine whether a given mass of particles is a single agglomerate or several.) The aerosol appears very much like that produced in other UO_2 melting work. No unusual behavior of the aerosol in the vessel was observed.

Behavior of Iodine in UO_2 Fuel Meltdown Tests

Iodine Disposition	Percent of Iodine Inventory			
	Low Iodine Concentration		High Iodine Concentration	
	A	B	C	D
Total release from fuel	83.8	63.8	87.2	91.6
Amount deposited in furnace chimney	6.3	10.6	0.4	0.4
Total on aerosol tank wall after 2-hr aging and 2.5-hr gas sweep period	46.5	23.8	26.4	34.2
Amount airborne and desorbed from tank wall during 2.5-hr gas sweep period	31.0	29.6	60.4	57.0
Amount in penetrating form[a]	3.7	0.4	0.3	0.5
Loss through 1.5-in. charcoal bed[b]	1.3	0.14	0.01	0.03
Particle Size Distribution in Aged Aerosol (A)	Percent of Airborne Iodine			
8–14 (I_2)	80.2	68.0	78.0	92.0
18–22	14.4	12.4	22.0	8.0
50–100	~5	19.6		0.02
>100	0.4		0.4	

Run A – Tracer-irradiated UO_2 (containing ~1 μg of I) with 20% unfiltered air (~5 μg I/m^3 in the containment tank).

Run B – Tracer-irradiated UO_2 (containing ~1 μg of I) with 100% filtered air (~5 μg I/m^3).

Run C – 7000-Mwd/ton UO_2 (20 g UO_2, containing ~400 μg of I) with filtered air (~3 μg I/m^3).

Run D – Tracer ^{131}I simulation with ~200 μg of I and with filtered air (~1.5 μg I/m^3).

[a]Determined from distribution of iodine activity in charcoal beds after removal of elemental iodine.

[b]This is a part of the penetrating iodine activity.

Two new sampling devices were designed and are being fabricated. One is a fallout sampler that permits collection of 11 time-dependent fallout samples during a run. The other consists of five May-pack samplers with diffusion channels inserted after a different May-pack element in each sample. This arrangement will be used for determining whether unusual iodine compounds exist in the Nuclear Safety Pilot Plant aerosol.

Reactor Containment Handbook. — Review of 9 of the 12 chapters of the handbook have been completed, and review of the remaining 3 will be completed within a few weeks. This review is based upon further ORNL work as well as the external comments which have been received to date. In addition, an index for the entire volume is now over 50% complete. The volume will be assembled and given limited distribution as a Technical Memorandum subject to receipt and resolution of further comments from the Commission.

Nuclear Safety Information Center. — The miniature Nuclear Safety Information Center (NSIC) for the Third International Conference on the Peaceful Uses of Atomic Energy has been completed. Approximately 1300 documents relating to the containment of nuclear facilities were indexed and placed on microfiche for use in the Geneva exhibit.

NSIC continues to collect, evaluate, and store information in its own retrieval system. During the Center's first year of operation, over 2500 sources of information were processed.

During this report period (May–June), NSIC has received and answered ten specific requests for information. In the same period six persons have visited the Center to utilize its services. Of the several "state-of-the-art" reports in process, one, Current Practices in the Release and Monitoring of I^{131} at NRTS, Hanford, Savannah River, and ORNL (ORNL-NSIC-3), was completed, and the report is in reproduction. The use of an IBM 7090 for the preparation of periodic bibliographies and the selective compilation of other bibliographic information is being explored.

WASTE DISPOSAL

Clinch River Study. — One purpose for the coring of Clinch River bottom sediments in 1962 was to inventory the quantity of radionuclides associated with these sediments. During this reporting period, processing procedures of the cores for this inventory have been completed. The processing of about 150 cores included compositing the radioactive portion of the core, and then determining the specific weight and moisture content of each composite and splitting the core into aliquots for analytical work. Aliquots of all cores have been submitted for radionuclide analyses. Aliquots from selected cores were submitted to the U.S. Geological Survey, Quality of Water Branch, Raleigh, North Carolina, for particle size analyses.

Some of the results of these analyses have been returned, but not enough are available to compute the inventory.

Mineral Exchange Studies. — Comparisons were made of the behavior of alumina and Dowex 50 in sorbing trace concentrations of strontium in

the presence of EDTA (ethylenediaminetetraacetic acid). With 3.2 g of α-alumina and 2.5 g of Dowex 50-X4 and the addition of 60 ppm EDTA at pH 8.4, the strontium distribution coefficient (K_d) was decreased from 96 to 0 for Dowex 50 and was increased from 60 to 90 for alumina. The increase in K_d of alumina was suspected to be due to complexing of strontium by EDTA and sorption of the complex by alumina. Pretreatment of alumina with EDTA showed that the K_d of the treated material was increased from 25 to 165. With long contact time the K_d slowly decreased; the decrease is suspected to be caused by dissolution of alumina in the alkaline system and possible complexing of aluminum ions by the EDTA.

Distribution coefficients were measured for two minerals, muscovite and pyrophyllite, as a function of sodium nitrate concentrations. Both materials showed sorption curves which did not correspond to ideal ion exchange. These materials were selected since they represent minerals of relatively low ion exchange capacities; muscovite derives its exchange capacity by broken bonds at the edges of the crystallites and by isomorphic substitution, in contrast to pyrophyllite whose capacity is due only to broken bonds. Responses similar to those of muscovite and pyrophyllite were observed with several soil materials which had not undergone intensive weathering.

Disposal in Deep Wells. — An injection of 150,000 gal of ORNL-type waste was completed on May 28, 1964, at a depth of 890 ft. This injection was the fifth in a series to test the performance of the hydrofracturing plant and mixes. The waste was mixed with 6-1/2 lb of cement-base mix per gallon, and the injection required 10-1/2 hr. Over 5000 curies of activity was injected; no activity was detected in the observation well 150 ft updip from the injection well.

The continuous operation for 10-1/2 hr served to test the ability of the pump to perform without changes in packing and seals. The mix was designed not only to be cheap and to have a long pumping time, but to have improved radionuclide retention characteristics. Drilling operations have begun at the site to recover cores containing the grout sheets from five injections.

High-Activity-Waste Solidification: Engineering Work. — Two tests were made in 16-in.-diam pots to investigate scaleup effects and the performance of induction heating equipment. Simulated Purex waste of a type which may be produced at Hanford in 1965 was used as feed in these tests. The first test was stopped after 95 hr because a hole, 4 in. wide by 6 ft long, developed in the calciner pot. Examination indicated that failure was caused by corrosion and localized overheating. In the second test, with control thermocouples set at 800 to 850°C, a 4 × 6 in. elliptical hole developed in the pot. There were no indicated temperatures in excess of 860°C, but the pot was obviously overheated in the region of the hole.

Both calcination tests were satisfactory from the standpoint of process performance, and pot filling rates following those predicted by the radial-deposition model and applicable to smaller diameter pots. The maximum feed rate was 2.0 liters/min, and there was no indication of excessive entrainment in the off-gas.

Fabrication was started on two 15-in. susceptors, one of stainless steel and the other of Hastelloy X, to be used in further induction-heating tests. These tests will be made with 8-in. pots inside the susceptors and will resemble the proposed mode of heating in the Hanford pilot plant.

A continuous melting unit for producing glassy solids (20 in. in diameter and 40 in. long) was operated for 150 hr before corrosion caused failure of the stainless steel vessel. The maximum steady-state rate for this unit was 40 liters of waste per hour, which is equivalent to waste production from a Purex plant processing 9 tons of uranium a day.

High-Activity-Waste Solidification: Laboratory Work. — A small-scale continuous melter was constructed and tested to evaluate problems in the continuous conversion of aqueous wastes to glassy solids. Problems to be evaluated include processing rate, scale formation, melt segregation, and corrosion. A conventional stainless steel "pot" (4 in. in diameter and 24 in. high) was modified for use as the continuous melter.

The melt depth was maintained at about 1 in. by a fixed overflow outlet in the center of the bottom. Also, a pipe containing a heating element was inserted down the center of the pot to increase the heat transfer surface. Feed rates as high as 5 to 7 liters of waste per hour were obtained. In runs over periods up to 100 hr, corrosion of the stainless steel container was slight with the Purex (FTW-65) lithium aluminum phosphate melt as long as the temperature was kept below 900°C. The process was shut down and restarted a number of times with no apparent difficulty.

Continuous melt processing was found to require a somewhat more fluid melt than is necessary for semicontinuous fixation by the rising-liquid-level method. The addition of more lithium and phosphate, reducing waste oxide content from 44.5% to 39.6%, produced a melt that was easily handled (mp, 700°C; pouring point, about 850°C). A program of computer coding was started to assist in correlating compositional data with the physical and chemical properties of the melts developed.

Intermediate-Activity Waste. — The incorporation of evaporator concentrates and sludges in emulsified asphalt is being studied as a method of preparing these wastes for disposal. The waste slurries or solids are first mixed with asphalt, and the temperature of the mixture is raised to 160°C to volatilize the water, leaving the solids encased in asphalt. The leach rate (in distilled water at 25°C) of ^{137}Cs from an asphalt product containing 20 wt % solids from simulated ORNL waste-evaporator bottoms was 7.4×10^{-5} g cm^{-2} day^{-1} at the end of five weeks, while the rate from a product containing 62 wt % solids was 2.4×10^{-4} at the end of five weeks. The leach rate of ^{106}Ru from a product containing 62 wt % solids was 6.9×10^{-7} at the end of five weeks. Workers at Mol, Belgium, reported a somewhat lower rate for cesium for a product containing 20% waste solids in asphalt, but their ruthenium leach rate is roughly equivalent to that reported here.

Experiments thus far indicate that the leach rate is affected more by the percentage of solids and by the elements being leached (Cs and Na > Ru and Sr) than by the type of leach test (i.e., flowing water vs stagnant water).

Low-Activity Waste: Pilot-Plant Work. — Several modifications made in the scavenging-precipitation ion exchange pilot plant resulted in improved process performance during three runs with ORNL low-level waste. A fluidized bed of activated alumina, added as an initial process step to reduce phosphate to 1 ppm or below, reduced phosphate in 500,000 gal (15,000 column volumes) of process waste water from 0.5–2.0 ppm to 0.1–0.7 ppm. More than 50% of the phosphate in the low-level waste was removed

in the first 10,000 bed volumes treated, which confirmed earlier laboratory results. A new 4-ft-diam, agitated clarifier operated at a rate of 48 gal min^{-1} ft^{-2}, compared with 27 for an older unit, yielded effluent having an average total hardness of 7 ppm (as equivalent $CaCO_3$). Filtration through a deep-bed anthracite filter further reduced the hardness in the clarifier effluent to 4 ppm. In addition, the new clarifier eliminated premature hardness breakthrough caused by channeling of the influent through the sludge blanket.

Grundite clay was added to the ion exchange regenerant solution to remove cesium, and this stream was then recycled to the process feed stream. This causes all the radionuclides separated from the waste to leave the process in the solid sludge and eliminates a regenerant evaporator.

The overall decontamination factors for the three runs averaged 17, 11, 1200, 1400, 2.6, and 580 for gross beta, gross gamma, cesium, strontium, ruthenium, and rare earths, respectively, which reduced the concentration of radioactive constituents in the treated waste water to less than 2% of the 168-hr occupational MPC_w values. Further treatment with an anion exchange resin column increased the ruthenium and cesium decontamination factors to 14 and 11 respectively. These changes were estimated to reduce the cost of waste treatment by this process from 78¢ to about 66¢ per 1000 gal; if the alumina column is not required, the cost is 60¢ per 1000 gal; if strontium is limiting instead of cesium, the cost is 56¢ per 1000 gal.

Low-Activity Waste: Laboratory Work. — Modifications to the scavenging-precipitation ion exchange process for the decontamination of ORNL low-level radioactive waste are being evaluated on a laboratory scale. They include (1) the use of activated alumina to reduce the concentration of phosphates in raw untreated waste (pH 7 to 8) to about 1 ppm or lower in order to prevent abnormally high residual hardness after the scavenging step; (2) the consolidation of the activity retained in the process into a solid form by adding Grundite clay to the regenerant waste of the cation exchange column to sorb cesium activity and the recycle of this slurry into the scavenging step; (3) the possible substitution of another phenolic-carboxylic resin, Ionac C-265, in place of the Duolite CS-100 resin now in use; and (4) the possible use of an anion exchanger to remove ruthenium and cobalt activity remaining after cation exchange treatment at pH 11.

Column experiments indicated that activated alumina does not sorb hexametaphosphate and thus provides a possible explanation for the observed incomplete sorption (only 50% or better) of phosphates contained in untreated waste. The capacity of alumina for sorbable phosphates was estimated to be 0.4 meq of PO_4^{3-} per ml of Al_2O_3 (equivalent to removing 1 ppm of phosphate from 13,000 volumes of waste per volume of alumina), which is numerically equivalent to 60 meq of Al^{3+} per meq of PO_4^{3-}. The stated equivalence ratio together with work reported in the literature indicates that phosphate sorption on alumina is a surface-dependent phenomenon.

Micro-pilot-plant tests demonstrated that the addition of Grundite clay not only consolidated the cesium and strontium activity into a solid form but also increased the factor for waste volume reduction (to air-dried cake) by 1.6 — that is to say, from 3400 without recycle to 5500 with recycle.

Ionac C-265 resin was expected to have up to twice the capacity for strontium that Duolite CS-100 resin has, while retaining the same capacity for cesium. However, the sorption of cesium was nil. Incomplete analyses show that the decontamination factor (DF) for strontium was greater than 1500 after 2700 of the 6000 volumes of waste treated. This compares with a DF of 1200 for 2000 volumes, using CS-100 resin.

Illco TAD-1 resin showed a DF of 5 and 1.6, respectively, for ruthenium and cobalt for 300 volumes of waste processed, but decreased to 1.5 and 1.0 for 1500 volumes.

Disposal in Natural Salt Formations. — Specifications for the 19.1-in.-ID waste-charging shaft for the demonstration experiment were prepared and put out for bid. The successful low bidder was the Sterling Drilling Company of Sterling, Kansas. The contractor has set up his rig and is currently drilling at a 300-ft depth.

Delivery of the cast uranium shield plugs for the fuel element canisters, the surface carrier internal rack, electrical hole mockup liners, and the underground transporter were made during the month. Modification of the surface carrier and shipping rack is in progress.

In order to check out operating techniques and design, a prototype test using one hole liner with its electrical heater assembly is being operated in the same room in which the radioactive floor array is to be located. Heated operation began on June 2, and results have been about as expected. Approximately 1 liter of water (released from the shale interbedded with the salt in the floor) has been collected in the off-gas condensate trap. The need for minor engineering changes in certain parts of the equipment has become apparent.

Engineering, Economic, and Safety Evaluation. — Although it is believed that the most responsible method of managing very radioactive liquid wastes from the processing of power reactor fuels will consist in converting them to solids and disposing of them permanently in salt mines, a possible alternative is long-term storage as liquids in tanks. Analyses are in progress to compare the costs and safety of tank storage with the costs of the preferred, more conservative approach.

The basis for the tank-storage analyses is the same as has been used in previous economic studies of waste management; a hypothetical 6-ton/day fuel processing plant is assumed which handles a combination of uranium and thorium converter fuels irradiated to 10,000—20,000 Mwd/ton. Storage of wastes under both acid and alkaline conditions is considered. In each case the volumes are adjusted to the smallest values consistent with efficient removal of decay heat. As acid solutions, Purex wastes are stored at a rate of 100 gal per ton of fuel processed, and Thorex wastes are stored at 200 gal/ton. Under alkaline conditions the wastes are stored at 600 and 1200 gal/ton respectively.

The tanks, with a capacity of 1 to 1.25 million gallons, are enclosed in steel-lined concrete vaults for secondary containment and are similar to those at Savannah River. Decay heat is removed by circulating water through vertical and horizontal banks of coils submerged in the waste.

In the safety analysis the consequences of failure of these tanks by a variety of mechanisms will be considered. The cost of "perpetual" storage will be estimated by using the concept of a sinking fund, set up to draw interest at a rate that will accumulate sufficient money to replace the tanks at the end of their life expectancies.

ARMY PACKAGE POWER REACTOR PROGRAM

PM fuel specimen assemblies A-1 and A-2 continued to operate satisfactorily in the pressurized-water loop in the ORR during ORR cycle 53 (4-10-64 to 5-31-64) and are now operating in cycle 54, which began June 13, 1964.

On May 25, 1964, a routine water sample was removed from the loop, and analysis with a multichannel gamma-ray spectrometer showed the presence of fission products ^{88}Kr, ^{133}Xe, and ^{135}Xe. The water sample analyzed radiochemically on May 26, 1964, contained a considerably higher amount of fission products than previous water samples removed from the loop since the start of the PM fuel irradiation program, August 30, 1963. The May 26 results along with some typical results obtained since the beginning of the PM tests are listed below:

Fission Product Nuclide Activities (dis sec^{-1} $liter^{-1}$) in ORR Pressurized-Water-Loop Coolant

Date	^{91}Sr (9.67 hr)	^{92}Sr (2.7 hr)	^{132}I (2.33 hr)	^{133}I (20.8 hr)
9-18-63	209	505	1852	292
2-4-64	640	932	893	300
4-2-64	301	426	735	168
5-26-64	632	786	5610	7660

After considerable study of this problem, it was decided that the fission product activity level was not serious enough to warrant removing the specimens. Day-to-day monitoring of the activity in the loop coolant has continued. Since May 26, the fission product activity levels have gone down, but with considerable fluctuation. It is believed that the activity is due to a small defect which has developed in the cladding of one of the fuel specimens. Unless the fission product level rises significantly, it is planned to leave the specimens in the loop until the end of ORR cycle 54 (7-26-64), at which time the regularly scheduled interim hot-cell examination will be performed.

PHYSICAL RESEARCH

PHYSICS AND MATHEMATICS

High-Energy Electronuclear Machines: The Separated-Orbit Cyclotron. – It now appears practicable to design an 800-Mev separated-orbit cyclotron with all orbits lying in a single plane. This would make it convenient to deflect the beam from various orbits (at various energies) into a fixed external ion-optics system. Model magnet measurements demonstrate that the desired magnetic field gradients can be maintained with an orbit turn separation as small as 4 in.

A separated-orbit cyclotron to accelerate protons from 200 to 800 Mev in a 25-turn spiral of alternating-gradient magnet elements is being evaluated. The guide-field pole tips are disposed on 31 magnet yokes, or sectors. Since each yoke is driven by a single pair of large coils, the total magnet power requirement is reduced to only about 0.5 Mw. Radio-frequency cavities between the magnet yokes provide unusually high energy gain per turn, about 25 Mev, but require less than 10 Mw of rf power for their excitation. Operating at a frequency of 50 Mc/sec, the rf system uses commercially available equipment and is expected to cost somewhat less than the 200-Mc/sec systems previously studied.

Theoretical evaluation of cavity design for the separated-orbit cyclotron has been extended to determine the cavity shape required to achieve a constant voltage, or a specified voltage variation, along the acceleration gap. The method used is an extension of that used in the design of optimally shaped drift tubes for linear accelerators.

Medium-Energy Nuclear Physics: 22-Mev Protons. — The information about the (p,n) residual interaction between a $1g_{9/2}$ proton and a $2d_{5/2}$ neutron obtained by studying the $^{93}Nb(p,d)^{92}Nb$ reaction has now been employed in calculating spectra for many elements in the mass region A = 90. The magnetic analysis system at the 86-Inch Cyclotron was used to obtain more experimental information on odd-odd nuclei to explore further the nature of the (p,n) residual interaction. The reaction $^{59}Co(p,d)^{58}Co$ was studied, and the four low-lying states arising from the shell-model configuration $(\pi f_{7/2})^{-1}(\nu p_{3/2})^{-1}$ were identified. In addition to the previously known 2^+ ground state and 5^+ level at 24 kev (these are unresolved in this experiment), we have found the 3^+ level to be at 340 kev and the 4^+ level to be at 430 kev.

The new scattering chamber was installed in the small experiment room at the ORIC, and a test run was made with protons. The spectrum of protons scattered from a polystyrene target provides the beam energy measurement. The elastic and the inelastic scattering from ^{12}C provide two reference energies. The position of the proton-proton scattering peak on this scale determines the beam energy.

Medium-Energy Nuclear Physics: Heavy Particles. — The zero-degree cross section for the reaction $^{12}C(^{16}O,\alpha)^{24}Mg$ was measured from 25.6 to 35.5 Mev excitation energy in the ^{28}Si compound nucleus for the ground-state and five excited-state alpha-particle groups. Results show that the compound-nucleus mechanism is dominant and that the lifetime of the compound nucleus decreases slowly with increasing excitation energy.

Alignment of the polarized proton experiment on the ORIC was completed. With 10 μa of alpha particles on the primary hydrogen target, about 2×10^7 polarized protons were measured in the second scattering chamber. This is precisely the intensity we expected with the ion optics designed for the purpose.

Medium-Energy Nuclear Physics: Theory. — The spectroscopy of nuclei in the mass region A = 90 is expected to be determined to a large extent by the residual interactions between nucleons outside the semiclosed shell core of ^{90}Zr. In particular, the low-lying energy levels of these nuclei will arise primarily from configurations in which protons are filling the $1g_{9/2}$ shell while the neutrons are filling the $2d_{5/2}$ shell. Hence the energy of these levels will be determined by the residual interactions

between two protons in $1g_{9/2}$ orbits, between two neutrons in $2d_{5/2}$ orbits, and the interaction of a $1g_{9/2}$ proton with a $2d_{5/2}$ neutron.

The (n,n) effective interaction is well known from the spectrum of ^{92}Zr. The recent determination of the energies of the 0^+, 2^+, 4^+, 6^+, and 8^+ states in ^{92}Mo, and the spectrum of 2^+, 3^+, 4^+, 5^+, 6^+, and 7^+ states in ^{92}Nb completely defines the set of effective (n,n), (p,p), and (p,n) residual interactions necessary for the calculation of the structure of the low-lying states of nuclei in this region.

Although the ^{90}Zr core is known to contain proton pair excitation, calculations on the basis of a simple single-configuration model will serve as a basis for interpreting the origin of experimentally observed levels. The effect of this pair excitation on the calculated energy spectra is expected to be partially accounted for by the use of experimentally determined two-body matrix elements. Deviations from the results of the simple model should serve as an indication of the importance of other configurations in this region of nuclei.

Low-lying energy levels in isotopes of niobium, molybdenum, and technetium arising from the coupling of protons in the $1g_{9/2}$ level and neutrons in the $2d_{5/2}$ level were calculated. A comparison with available experimental data shows reasonably good agreement and indicates that the consideration of ^{90}Zr as a core state is a reasonable assumption. The simple model serves as a good basis for interpretation of the levels observed in these isotopes and as a good guide in judging the importance of other configurations. The intensity with which states in ^{92}Zr other than the 0^+, 2^+, and 4^+ are excited as a result of the pickup of the $1g_{9/2}$ proton in the reaction $^{93}Nb(d,^3He)^{92}Zr$ would provide experimental evidence on the extent of configuration mixing.

Interesting evidence was seen in the results of neutron stripping experiments for admixing of higher single-particle neutron states with the calculated levels. The evidence for the excitations of the protons to higher orbits is only indicated by the log ft values for the β transitions. Proton stripping experiments like (^3He,d) would be very useful in identifying proton excited states.

A more general calculation including the effect of the higher configuration of the protons and neutrons would require information regarding the interactions between the two nucleons in the higher states. Experiments such as $^{90}Zr(t,p)^{92}Zr$, $^{91}Zr(d,p)^{92}Zr$, $^{90}Zr(^3He,p)^{92}Nb$, and $^{91}Zr(^3He,d)^{92}Nb$, done with sufficient energy resolution, can provide such information and would be very desirable.

Cyclotron Operations: The Isochronous Cyclotron. — Operation of the ORIC has continued on a schedule of seven days a week, 16 hr per day. Since most of the external ion-optics equipment for transmitting the beam into the small experiment room is now installed, about 60% of the 112-hr operations week can be scheduled for nuclear research work. The continued installation of various components associated with the beam optics control circuitry requires about 20% of the total time. The remaining 20% is scheduled for ion dynamics and other development studies for the improvement of cyclotron operation.

During the hours allotted for nuclear bombardments, the cyclotron is now "on beam" about 75% of the time. The "outage" time is for routine operation maintenance needs, such as filament changes in the ion source,

target setup, and changing or readjusting power supplies for different operating conditions. The experimental bombardments have included beams of ~35-Mev protons, ~75-Mev alpha particles, and ~37-Mev deuterons. These beams were supplied to the various reaction target chambers located in the small experiment room. The reaction chambers were aligned with the ion beams, and the irradiations were successfully completed.

High-Voltage Experimental Program: Elastic Scattering of 17- to 21-Mev Neutrons from ^{12}C. — A search for a resonance in the yield of elastically scattered 17- to 21-Mev neutrons from ^{12}C has been made. The differential cross section has been measured for laboratory angles of 36, 51, 60, 86, 123.5, and 139°. The total cross section was also measured for the same range of neutron energies. In the differential cross sections measured at 36, 51, and 60°, a broad resonance was present at a laboratory neutron energy of 19.5 ± 0.2 Mev. Its total width at half maximum was approximately 1.1 Mev, and its peak height was between 14 and 21% of the value of the continuum. Within the 4.5% statistical error of our measurements, the resonance does not appear at 86°. At 123.5 and 139°, the cross section decreases monotonically by a factor of 3 between 17.5 and 20.5 Mev. The total cross section is consistent with a weak resonance at 19.6 ± 0.2 Mev which has a total width at half maximum of about 1.2 Mev. The resonance, which presumably results from a state or states in the compound nucleus ^{13}C, appears similar to the known resonance in the yield of elastically scattered 22.5-Mev protons from ^{12}C.

High-Voltage Experimental Program: Alpha-Alpha Correlations in the Reaction $^{7}Li(d,n)2^{4}He$. — The reaction $^{7}Li(d,n)2^{4}He$ has been studied at two deuteron bombarding energies, 4 and 7 Mev, by observing the two emitted alpha particles in coincidence. A total of 15 three-dimensional correlation functions were measured with different combinations of detector angles. The pronounced structure observed in these spectra is consistent with the assumption that the reaction proceeds primarily through a sequential mechanism via the intermediate nuclei ^{5}He and ^{8}Be.

Stable Isotopes Development. — The combination of an RuF_5 charge and an internally valved feed system fabricated entirely of nickel has greatly improved the separation of ruthenium. The new approach yields outputs averaging 6 ma, results in a substantial increase in isotopic purity, and increases process efficiency by a factor of 5. Previous ruthenium processing required the use of metallic ruthenium charges which were vaporized by electron bombardment. The operation was erratic and outputs were low, averaging only 2.5 ma total current.

The feasibility of the new type of separation was demonstrated in the 255° separator system. The success achieved was due to the design of a feed system compatible with both the operating pressure and the requirements for chemical stability of RuF_5. The all-nickel construction minimized chemical decomposition of the feed, and the internally valved feed system provided controlled conductance in various pressure regions and further reduced the possibility of charge dissociation. The charge container was operated at the equilibrium pressure of RuF_5, and laminar flow was through a tube of small diameter to an all-metal leak valve. At this point in the system, a sharp transition in pressure occurred. Low-pressure flow continued to the arc region through a specifically

sized tube providing conductance for the required amount of vapor with only a small pressure differential between the ends of the tube.

This concept in equipment design makes practical use of thermally unstable feeds, and the technique may be applied in the separation of other elements in the platinum metals group, in which volatile but relatively unstable compounds exist.

One 48-in.-radius calutron was fitted with an additional diffusion pump to provide a means of studying the characteristics of the pumping system. A 10-in., 4000-liter/sec pump was placed directly under the ion source to supplement the four 20-in., 5000-liter/sec pumps normally used. A calibrated leak introduced through the ion-exit slit of the source produced a pressure rise in the system. The single 10-in. pump below the source provided ~40% of the effective pumping speed for the source-receiver region that the four 20-in. pumps provide as they are now positioned. Further testing showed that equivalent pressures could be maintained at various gas loads by the single 10-in. pump or by three of the original 20-in. pumps.

The addition of this small pump almost doubles pumping speed in the region midway between the source and the receiver, and practical applications of such an increase are being determined in selenium separations. A 20% increase in ion output is being achieved in the system which has increased pumping capacity.

Special Separations. — A one-year separation of plutonium isotopes from 652 g of available feed was completed last September. Since that time, recovery operations needed to process feed residues have been completed, and a material balance is available. The 652 g of feed was recycled to provide 2458 g loaded into the system, and 1800 g was vaporized. Total recovery amounted to 540 g or 82.8% of the starting material. Unrecoverable feed losses to the system are approximately 5% per cycle. The material actually recovered consists of 231 g of separated isotopes and 309 g of available charge material. A loss of 3.9 g was allocated to decay to americium, and 8.4 g was buried in salvage which was uneconomical to process. The calutron system contains ~100 g of plutonium which was not removed by recovery techniques applied in equipment cleaning.

Recycle recovery has been completed for uranium separation series "c"; unrecoverable losses to the system total $<4\%$ of the starting feed. A total of 30.3 g of ^{233}U with ^{232}U content ranging from 4 to 12 ppb resulted from this series. A total of 1300 g of ^{238}U with ^{235}U content ranging from 1.5 to 20 ppm was collected in uranium series "b." Recovery of unresolved feed material has not been completed for this series.

CHEMISTRY

Analysis of Molten Salts by Electrochemical Methods. — A molten mixture of $LiF-BeF_2$ (66-34 mole %), the coolant salt for the MSRE, was prepared for voltametric studies by the Reactor Chemistry Division. This mixture had been treated with beryllium metal and then filtered to remove traces of reducible impurities. Subsequent current-voltage curves recorded at a pyrolytic graphite indicator electrode revealed the absence of reduction waves which could be attributed to electroactive impurities

such as iron or nickel. A small rather ill-defined wave was observed at ~1.1 v vs the platinum quasi-reference electrode. This is believed to be due to the reduction of zirconium, since a small amount (700 ppm) was found by chemical analysis. It is planned to study this wave further after making standard additions of ZrF_4 to the melt.

The potential at which the cell current becomes too large to measure (potential limit of the melt) occurs at approximately −1.8 v. At this potential, it is believed that the beryllium is being reduced at the pyrolytic graphite indicator electrode. The potential limit of the LiF-BeF_2 melt is ~0.2 v more negative than that observed for the LiF-BeF_2-ZrF_4 eutectic, a fact which suggests that beryllium metal is a stronger reductant, in the melt, than zirconium metal.

It appears that the platinum quasi-reference electrode may not be as well poised in the LiF-BeF_2 melt as it seemed to be in an LiF-NaF-KF mixture. This fact is borne out by a peculiar shift of 0.1 to 0.2 v in the potential limit of the molten mixture, depending on whether the cover gas is static or flowing through the cell. The reason for this phenomenon is as yet not known. Work is in progress, however, to prepare Ni-NiO reference electrodes similar to the one used by Steinmetz (UNC-5032, Aug. 24, 1962). If this electrode functions properly, it should then be possible to observe whether the potential of the platinum quasi-reference electrode shifts.

Protactinium Chemistry. — The purification of the ^{231}Pa borrowed from England is nearly complete; the oxide has been packaged and is awaiting shipment. The material recovered contains an estimated 38.2 g of ^{231}Pa, with an uncertainty of about 1 g. In addition, approximately 200 mg of protactinium has been recovered and purified from various solid and liquid wastes removed from the hot cell used for processing the above material. The usual purification procedure, based on anion exchange of a chloride complex, was unsuccessful for this last operation because of interference by very fine solids suspended in the solutions. A solvent extraction method utilizing Alamine 336, a commercial mixture of tertiary amines, was successful.

Arrangements are being made to recover from the burial ground the equipment used for the ^{231}Pa-^{232}U processing done about two years ago. There is a possibility that a small amount of ^{231}Pa may have been accidentally discarded with the equipment. If so, an effort will be made to recover it.

Radiation Chemistry of Organic Materials: High Polymers. — Solubility measurements have been made on polystyrene irradiated in a reactor and in cobalt gamma sources. The results have been treated in a manner which reveals the rates of both molecular cross-linking and scission. The ratio of scission to cross-linking was low, about 0.14. Significantly, the ratio is the same for both reactor neutrons [LET (linear energy transfer) ~3 ev/A] and for gamma radiation (LET = 0.02 ev/A), although the absolute rate for either process is 2.8 times as high for neutrons as for gamma radiation. (The scission yields are 0.0094 ± 0.0005 per 100 ev for gamma radiation and 0.027 ± 0.004 for neutrons.)

The two processes would not be expected to have the same dependence on LET, since the simplest mechanism for scission would be first order, while cross-linking would be second order. The observed dependence of

both processes on LET indicates either that the kinetics of both processes are complex or that the small amounts of scission observed were oxidative degradation with an LET dependence similar to that of the cross-linking.

Spectrophotometry of Solutions at High Temperature. — We have reported (ORNL-3628) measuring the absorption spectrum of CO_2 in the condensed states between 1.19 and 1.66 μ as a function of temperature from −80°C and 13 psia to 65°C and about 2000 psia. The $(3\nu_3)$, $(2\nu_1 + 2\nu_2 + \nu_3)$, and $(\nu_1 + 4\nu_2 + \nu_3)$ bands are sufficiently intense to allow a reasonably precise study of the effect of temperature on band contours and other parameters. The third harmonic asymmetric stretch band exhibits a slight asymmetry due to the presence of one or more unresolved components centered on the low-frequency side of the band, and we have not yet attempted its analysis. However, we have found that the $(2\nu_1 + 2\nu_2 + \nu_3)$ and $(\nu_1 + 4\nu_2 + \nu_3)$ bands can be fitted to a good approximation over the entire fluid range studied (from the freezing point, −56.6°, to 65°, 34° above the critical point) by a Lorenz function which assumes a statistical distribution of perturbations of each molecule by its neighbors.

Physical Chemistry of Molten Salts: HF-H_2O Equilibrium with Molten Fluorides. — In continuing studies of the equilibrium

$$2HF(g) + BeO(cryst) \rightleftharpoons BeF_2(soln) + H_2O(g)$$

in LiF-BeF_2 melts saturated with BeO, the variation of the equilibrium pressure quotient P^2_{HF}/P_{H_2O} with melt composition in the range $2LiF \cdot BeF_2$ to BeF_2 has been determined. As is seen from the expression of the equilibrium constant for the above reaction,

$$K = \frac{P_{H_2O}}{P^2_{HF}} a_{BeF_2} ,$$

the increase of P^2_{HF}/P_{H_2O} with BeF_2 content of the melt should be proportional to the increase in the activity of BeF_2, a_{BeF_2}. If this activity is expressed as the product of an activity coefficient γ_{BeF_2} and the mole fraction X_{BeF_2} [≡moles BeF_2/(moles BeF_2 + moles LiF)], then the quantity γ_{BeF_2}/K is given by $(P^2_{HF}/P_{H_2O})/X_{BeF_2}$. The following are results of experiments at 600°C:

X_{BeF_2}	0.33	0.4	0.5	0.6	0.7	0.8
P^2_{HF}/P_{H_2O}(atm)	0.0057	0.0145	0.0320	0.0490	0.0590	0.0660
γ_{BeF_2}/K	0.0171	0.036	0.064	0.082	0.084	0.0825

If, instead, the components are redefined as Li_2BeF_4 and BeF_2, a different mole fraction X'_{BeF_2} results, and hence a different activity coefficient γ'_{BeF_2} may be computed by an analogous calculation from the same experimental data:

X'_{BeF_2}	0	0.25	0.50	0.667	0.786	0.875
γ'_{BeF_2}/K		0.058	0.064	0.073	0.075	0.075

It is seen that, in terms of the components Li_2BeF_4 and BeF_2, the resulting activity coefficient is more nearly constant over a wider composition range, suggesting that such LiF-BeF_2 mixtures may, to a good approximation, be considered ideal mixtures of Li_2BeF_4 and BeF_2 when BeF_2 exceeds 40 mole %. Similar results were obtained at other temperatures in the range 500 to 700°C.

Physical Chemistry of Molten Salts: Viscosities in the BeF_2-LiF System. — The viscosity of fused media containing BeF_2 is interesting because of (1) the importance of liquids containing BeF_2 in nuclear technology (e.g., in molten-salt reactors) and (2) the structural similarity of BeF_2 to SiO_2 (therefore BeF_2 can serve as a low-temperature research analog in the study of SiO_2 glasses). A continuing program of study of the viscosity of the BeF_2-LiF system is indicating how viscosity changes with composition, how these changes are correlated with structural parameters of the individual components, and how this system compares with the analogous SiO_2-MgO system.

Measurements were obtained with a commercial rotational viscometer on 13 mixtures with compositions varying between pure BeF_2 and 34 mole % BeF_2—66 mole % LiF. In this composition range the viscosity changed by at least five orders of magnitude (e.g., at 725°C the viscosity of pure BeF_2 was found to be $\sim 10^6$ centipoises, while the viscosity of the 60-40 mole % BeF_2-LiF mixture was 30 centipoises; the viscosity of the 34-66 mole % mixture at 725°C would have been even lower). The energy of activation for viscous flow (E_η) decreased from ~60 kcal for the pure BeF_2 to ~17 kcal for 60-40 mole % BeF_2-LiF and to ~11 for the 34-66 mole % mixture.

The change of the energy of activation with composition in the BeF_2-LiF system parallels that in the SiO_2-MgO system. In other words, the ratio of E_η(pure BeF_2)/E_η(x-y mole % BeF_2-LiF) is approximately equal to E_η(pure SiO_2)/E_η(x-y mole % SiO_2-MgO).

The pronounced lowering of viscosity and energy of activation with increasing LiF concentration is believed to be associated with the gradual breakup of the continuous, random network of Be-F tetrahedra. This network is presently thought to account for the glassy properties of pure BeF_2.

Target Preparation. — Milligram quantities of isotopically enriched rare-earth metals, excluding lanthanum and cerium, were produced from their respective oxides by electron bombardment heating. Because of the porosity of the metal derived in this manner, all of these materials can be cold rolled into foils ~1 mg/cm^2 thick. Similar techniques are now being applied to chromium oxide and other oxides for which a suitable, low-vapor-pressure, metal reductant can be used.

Zone-refining techniques applied to isotopically normal tungsten and molybdenum bar stock produced metals which were ductile enough to be cold rolled. Extrapolation of this technique to the electron beam melting of

small quantities of these metals has been used to obtain molybdenum isotopes in ductile form. These beads have been rolled to thicknesses of ~1 mg/cm^2. A similar technique for tungsten is being developed.

High-temperature vapor sources and substrate rotation devices have been developed which show promise in obtaining highly uniform thin-film deposits. Using one combination of these devices, two $^{241}PuO_2$ targets were prepared. The film thickness of each target was 2 mg/cm^2, to an estimated uniformity of ±5%.

TRANSURANIUM-ELEMENT PRODUCTION

Chemical Process Development. — Studies have been initiated to evaluate ethylenediaminetetraacetic acid (EDTA) as an eluant for californium and einsteinium from cation exchange resin and to study the behavior of californium and einsteinium on anion exchange resins in the EDTA form.

Attempts are being made to produce plutonium sols with the lowest possible nitrate:plutonium mole ratio. In connection with this work, methods for accurately determining the nitrate concentration of plutonium sols are being developed.

Various procedures are being investigated for producing sols of lanthanides as a stand-in for americium and curium. Although sols have been produced by several methods, a completely satisfactory procedure has not yet been found.

Curium Recovery Facility. — Cells 3 and 4 in Building 4507 are being equipped as an integrated facility to test transuranium process chemistry at full-scale activity levels and to recover gram quantities of ^{242}Cm and ^{244}Cm. Startup with tracer-level runs is scheduled for July. Operations during this reporting period include continued equipment testing and modification and flowsheet testing.

Polyethylene service lines have continued to degrade during cold testing operations, and we have decided to replace nearly all the in-cell polyethylene lines with metal tubing. The first Zircaloy-2 mixer-settler was received, and it was tested hydraulically with the major chemical flowsheets. Satisfactory operation was obtained. Fabrication of the rack containing two such Zircaloy-2 mixer-settlers, which will replace the present Homalite units prior to startup, is progressing satisfactorily.

In laboratory testing, Adogen 364 (a tertiary amine substitute) appeared superior to the previously used Alamine 336, showing no tendency to form emulsions in mixer-settler operations.

HFIR Target Development: Prototype Target Irradiation. — The initial phases of the postirradiation examination on the first HFIR prototype target were completed at Vallecitos. The spacing between the tubing and the hex can, the diameter and length of the capsule, and the capsule to hex-can attachments were unchanged. All surfaces of the aluminum were covered with the typical thin, off-white oxide.

A gamma scan of the element indicated almost uniform flux distribution over the entire length, with slight peaking occurring about 4-1/2 in. from the top of the full column. Cobalt-activation analysis of the central aluminum fin indicates that the flux level was essentially as intended, to produce 40 at. % burnup with an average heat rating of 700,000 Btu hr^{-1} ft^{-2}. Burnup analysis will be made to confirm this.

Two of three punctures for gas collection have been made. The quantities of gas collected from the voids at the ends of the element were nearly equivalent to the helium originally in these areas, indicating little diffusion of fission gases from the pellets to the void spaces. A third puncture (into one of the central pellets) and analysis of all the gas samples will be made. Sectioning and metallographic examinations will be done during July.

HFIR Target Development: Plutonium Target Fabrication. — Hardware procurement for the HFIR target fluid-flow test bundle has continued. A new system for handling liquid nitrogen and gas has been installed on the welding glove box.

Transuranium Facility. — Construction progress on the Transuranium Processing Plant included pouring the concrete floor of the decontamination room and the normal concrete portion of the north-south cell partition walls. The intercell conveyor housing was received and located in the cell bank. Procurement of 17 valves from the Velan Valve Company will be delayed beyond the July 1 contract date because of vendor problems in meeting our specifications.

The ORNL shop fabrication of chemical process equipment is now 15% complete. Schedules are being revised in order to reflect some material-procurement delays. The target date for completion of this equipment is being moved up from August 15, 1965, to July 1, 1965. Fabrication of some piping assemblies by other shops may be required in order to meet this schedule. Design of the equipment is 65% complete and should be finished by February 1965.

Corrosion. — Welded specimens of Zircaloy-2 were immune to hydrogen pickup during 1000-hr tests at 55°C in nonaerated and oxygenated 6 M HCl solutions. Solution-exposed specimens showed final hydrogen contents within the concentration limits initially present in the unexposed alloy, 38 to 53 ppm. The observed corrosion rates in both environments were less than 0.1 mil/month. Similarly, no hydrogen pickup in welded Zircaloy-2 occurred after 1000 hr at 55°C in a nonaerated solution of 11 M LiCl containing 0.02 M HCl. The corrosion rate was negligible. However, a pickup to 150 ppm of hydrogen took place in a welded, solution-exposed Zircaloy-2 specimen after 1000 hr in oxygenated 8 M HCl at 55°C. The unexposed material contained between 38 and 53 ppm initially. This was the first observed case of hydrogen pickup in Zircaloy-2 exposed in TRU-related solutions at temperatures below boiling. A corrosion rate of 0.5 mil/month was obtained in the 8 M HCl solution; the vapor-phase rate was 0.1 mil/month. The strip solution in the flowsheet for lanthanide-actinide separations is 8 M HCl. All other process streams are 6 M or less.

Analytical Chemistry Development. — An extraction method for the determination of radioactive ruthenium has been evaluated. Ruthenium recovery of less than 98% with an alpha decontamination of less than 3×10^5 has been routinely achieved. It is now proposed to use this method, retaining the distillation procedure for a reference method.

A new method for the separation of trivalent actinides from lanthanides was developed. The quaternary amine Aliquat 336-S (95 to 98% tricaprylmethylammonium chloride) dissolved in xylene (30% by weight) extracts the actinides from solutions that are 0.2 N in H_2SO_4 and, most

important, from solutions that are 0.2 to 0.6 M in NH_4CNS. Excellent separation factors are obtained from the lanthanides (e.g., Cf/La is about 10^4, and Bk/Ce is about 1600). These separations are possible only at the dilute concentrations of NH_4CNS; high concentrations lead to common extraction. In the absence of NH_4CNS, the actinides are not extractable from this system. The extraction system has several advantages over the 11 M LiCl–HCl system for this separation, including higher solubilities (greater extraction per unit of reagent) and noncorrosivity. The stability of the thiocyanate under high radiation and other radiation-induced problems will be investigated.

METALLURGY AND MATERIALS

Preparation of Pure Materials: Single-Crystal LiF. — A 325-g crystal of LiF, designated ORNL-8, containing 50.77 at. % ^7Li was produced and was made available for use at the Materials Science Center of Cornell University as a part of the AEC Pure Materials Program. This crystal was prepared by standard techniques developed at ORNL (ORNL-3341) for preparing single crystals of lithium fluoride of <1 ppm impurity level. Crystals ORNL-7 and -8 were found to be sufficiently pure that the proportionality constants of the Rayleigh scattering term could be shown to be dependent upon isotopic composition.

Preparation of Pure Materials: Single-Crystal Li_2BeF_4. — Crystals of pure Li_2BeF_4 were prepared and were used for initiation of the first structural investigation of this important compound. The product, grown by the modified Stockbarger method, was polycrystalline. A melt of the stoichiometric composition, prefused with ammonium bifluoride in a graphite crucible, was used as the charge material. The polycrystalline character of the ingot obtained from the Stockbarger furnace is therefore probably due to nucleation emanating from graphite centers. Optically clear single crystals of maximum size approximately 3 × 4 × 10 mm comprised some 20% of the frozen mass. Preliminary x-ray diffraction data, obtained with these crystals, indicated that they will serve as excellent specimens for crystallographic studies.

Reactions at Metal Surfaces. — Our investigation of diffusion in the refractory metals tantalum and niobium is continuing. Refinements in the technique for isotope depositions have markedly improved the reliability of the extreme near-surface penetration data for both host metals. These improvements have permitted us to take greater advantage of the extreme sensitivity of our anodic-film sectioning technique.

We obtained penetration profiles for the diffusion of ^{95}Nb into both mono- and polycrystalline tantalum specimens at several temperatures. The non-Gaussian behavior observed was interpreted to be a result of the coexistence of both lattice and "short-circuit" diffusion mechanisms. Even in the single-crystal specimens, the contribution of diffusion along diffusion pipes was easily seen. At low temperatures in polycrystalline specimens, these short-circuit diffusion mechanisms predominated, completely controlling the shape of the penetration curves for penetration depths greater than a few thousand angstroms.

This research has demonstrated that the determination of lattice diffusion coefficients at low temperatures in tantalum and niobium (and probably in general) requires the careful interpretation of data obtained by a technique capable of resolving concentration gradients on the submicron level. Our anodic-film sectioning method is such a technique and has proved satisfactory for the determination of lattice diffusion coefficients as low as 10^{-17} cm^2/sec. We anticipate future improvements in the method to increase its range still further.

Deformation of Crystalline Solids. — Knowledge of the increase in energy content of metals and alloys during deformation is important because it provides information on the defects thus created, and this energy provides the driving force for subsequent recovery and recrystallization. This energy can be measured either during the deformation (as the difference between the mechanical work done on the sample and the heat released by the sample) or during the recovery and recrystallization processes. Values obtained by these two methods do not agree satisfactorily, and we may attribute this to the inadequacy of methods to measure the recovery energy. We are building an isothermal calorimeter that we expect will be able to properly follow the recovery process.

The most critical aspect of such an experiment is the ability to rapidly increase the temperature of the sample and the furnace while keeping both at nearly the same temperature and to precisely control the furnace at this higher temperature. This is critical because recovery proceeds with a rate that decreases so rapidly that reliable measurements must be made in very short times. The more difficult part of this problem is heating the sample. Of the several ways that have been tried, the best appears to be the use of a tungsten filament inside the furnace to heat both the sample and furnace. Unfortunately, in our present apparatus, the filament is not in adequate thermal contact with the furnace, so that the filament cools below the furnace temperature. The new apparatus now being constructed should take care of this problem. Nevertheless, the present calorimeter has worked well enough to measure the recrystallization energy of copper in agreement with accepted data.

Spectroscopy of Ionic Media. — As part of our continuing study of the nature of the molecular entities that exist in high-temperature liquid media, we have been investigating the structure of metal complexes by spectroscopic methods. Preliminary measurements of the optical spectra of molten $NiCl_2$-CsCl mixtures indicate that the coordination geometry of Cl^- ions about Ni^{2+} ions changes markedly when the $NiCl_2$ content changes from a very small value to 75 mole %. The measurement temperature, 850°C, was well above the liquidus curve for all mixtures except those near 75 mole % $NiCl_2$. For $NiCl_2$ concentrations up to about 30 mole %, the spectrum changed only in minor ways and appeared to be due predominantly to Ni^{2+} ions surrounded by tetrahedra of Cl^- ions. When the composition reached 40 mole % $NiCl_2$, the relative intensities of the absorption bands were decidedly altered. At concentrations above 40 mole %, these changes became strongly accentuated. Clearly, something quite different replaces the tetrahedral coordination geometry at the higher concentrations. Although we have not yet identified the new coordination geometry, it appears to be quite similar to that which prevails about Ni^{2+} impurity ions in molten $MgCl_2$. It is relevant to note that the $NiCl_2$-CsCl system has

two binary compounds, $NiCl_2 \cdot 3CsCl$ (mp, 547°C), with an approximately tetrahedral arrangement of Cl^- about Ni^{2+}, and $NiCl_2 \cdot CsCl$ (mp, 758°C), with an approximately octahedral arrangement of Cl^- about Ni^{2+}. However, the unidentified melt spectrum does not correspond to octahedral coordination.

Theory of Alloying. – The general study of alloying zirconium with transition elements adjacent in the periodic table has continued with new measurements on Zr-Sc, Zr-Ti, Zr-Nb, and Zr-Mo. We found from measurements of electrical resistivities that no significant long-range ordering occurs in hexagonal Ti-Zr solid solutions that could account for our previously reported maxima in superconductivity transition temperature and density of electronic states. With increasing scandium content in Zr-Sc, the density of electronic states decreases to a minimum at 10% Sc and then rises nearly linearly to a value for pure scandium that is 3.8 times that for pure zirconium. The decrease in electronic density per electron removed by dilute alloying with scandium is equal and opposite to the changes produced by electron addition through alloying zirconium with Ag, Cd, In, Sn, or Sb, in agreement with a rigid band approximation. On the other hand, in dilute Zr-Nb and Zr-Mo alloys, the experimental rise in density of states and superconductivity transition temperature and the drop in Debye temperature with increasing alloy content were greater than would be predicted by this approximation. Measurement of electrical resistivities of Zr-Nb alloys after annealing at 600 to 620°C showed solid solution to 4 at. % Nb and a smaller change in electrical resistivity for niobium than for usual alloying additions to hexagonal zirconium.

Structure of Metals. – One of the current problems in the theory of annealing of metals concerns the origin and nature of nuclei for recrystallization of cold-worked metals. We are studying the annealing behavior of niobium single crystals containing deformation twins that have been introduced by mechanical working. These twins are thin lamellae which are related in structure to the bulk crystal by a plane of symmetry called the coherent twin boundary. At 1000°C the noncoherent twin boundaries were mobile, and the twins could expand or contract through the migration of these boundaries. The coherent twin boundaries were much less mobile; they did not move in 30 days at 1000°C. These niobium crystals also recrystallized at this temperature, but only at twin intersections. The new grains had fixed orientation relationships to the intersecting twins and the matrix. The growth rates of these grains were very anisotropic, and the direction of fastest growth was parallel to the primary twin system.

Fundamental Research in X-Ray Diffraction. – To understand the oxidation behavior of metal crystals, a detailed knowledge of the structure of the surface films is necessary. This knowledge can be obtained by x-ray diffraction techniques. In our continuing study of thin Cu_2O films grown on copper single crystals, we have found some interesting diffraction effects.

We have found and explained a discrepancy in the spacings between 110 planes in the films. In Cu_2O these planes alternate between planes containing copper atoms only and planes containing both copper and oxygen. Our experiments showed that the spacing between the oxygen-containing planes was less than twice the spacing between adjacent unlike planes, and the difference increased with decreasing film thickness. We had

previously shown [Acta Cryst. 13, 542 (1960); Acta Met. 10, 691 (1962)] that the forces between the copper crystal and the oxide film distort the structure of the film, so that the atom spacing decreases with increasing distance from the metal. Since the diffraction method inherently averages the spacing over the film thickness, we can resolve the discrepancy by recognizing that the plane of atoms next to the metal consists only of metal atoms. The spacing between oxygen-containing planes is measured from a diffraction peak that arises only from scattering by oxygen atoms and thus does not involve the first plane. The spacing between adjacent planes, however, involves the much heavier scattering by copper atoms and thus includes the first plane with its greater separation. This effect is surprisingly large, sometimes amounting to several tenths of a degree in the diffraction angle.

Our theory [Acta Cryst. 13, 542 (1960); Acta Met. 10, 691 (1962)], which assumed uniform film thickness, failed to account completely for the shapes of the x-ray diffraction peaks. We have expanded our theory to include the fact that the film thickness is not uniform, and it now agrees with experiment. Also, the expanded theory has resolved a discrepancy between values of the film thickness derived from different features of the diffraction pattern.

Crystal Physics. — Experiments with single crystals aid the fundamental understanding of the properties of many materials. Since suitable single crystals are frequently difficult to obtain, particularly for refractory substances, we are conducting a continuing program, both to provide crystals needed in research and to devise and improve methods for growing crystals. Also, we are investigating select physical properties as they relate to the crystal-growth process or to new compounds.

We are building a split autoclave (working volume is 5/8 in. in diameter by 5-1/2 in. long) to contain 30,000 psi at 500°C. It incorporates our modified Bridgman seal supported by a retainer ring [Rev. Sci. Instr. 34, 1262 (1963)], which has been most effective in other autoclaves.

Thorium dioxide crystals containing Gd, Dy, Er, or Yb at controlled concentrations below 0.1 wt % were grown from molten $Li_2O \cdot 2WO_3$ containing 1 wt % B_2O_3, at 1225 to 1275°C. These crystals are being studied by electron-spin-resonance (ESR) methods. Since our undoped ThO_2 crystals were sufficiently pure and free of thermal defects that they showed no resonance spectrum down to liquid-helium temperature, the spectra observed on doped crystals are due to the dopant and the necessary charge compensation. We have now obtained ESR spectra for Er^{3+} and Yb^{3+}, each of which combines types that correspond to different point symmetries. One was isotropic with rotation of the magnetic field and was attributed to a paramagnetic ion in a cubic site. The other spectrum for ThO_2(Yb), upon rotation of the magnetic field, had axial symmetry about the four [111] directions of the crystal, while that for ThO_2(Er) had tetragonal symmetry about the three [100] directions. The line widths of the cubic spectra, for low concentration of ytterbium or erbium, were less than 1 oersted. This is quite narrow and somewhat indicative of crystals of good quality.

The crystalline mosaic spread of (111) faces of ThO_2 was determined by x-ray double-crystal diffraction methods. The line widths at half maximum were about 0.05°, and the defect densities were of the order of 10^9 per square centimeter. These numbers give us our first quantitative measure of crystal quality.

Fundamental Investigation of Radiation Damage in Solids: Effect of Lead on the Room-Temperature Colorability of KCl. — The introduction of F centers in lead-doped KCl by ionizing radiation has been measured as a function of lead content for different radiation intensities. For the lowest intensity (gamma irradiation), as little as 0.5 ppm impurity caused a marked decrease in the late-stage coloration, and less than 100 ppm completely suppressed the late stage. Lead impurities are less effective in decreasing late-stage colorability when higher irradiation intensities (electron irradiation) are used. These results suggest that previously observed variations in the late-stage coloration characteristics could have been due to trace amounts of impurities of the order of 1 ppm. Furthermore, it is suggested that these trace impurities are not depleted during irradiation, but affect the late-stage colorability by acting as electron or hole traps, or as recombination centers.

Fundamental Investigation of Radiation Damage in Solids: A Capacity Microphone for Ultrasonic Measurements at Megacycle Frequencies. — A transistorized capacity microphone has been developed for use in ultrasonic measurements employing the pulse-echo technique. Briefly, the purpose of such a development has been threefold. With a gap of known dimensions between the sample and the detecting electrode, it is possible to calibrate the output signal in terms of sample displacement, so that an absolute value of sample strain can be obtained. If the conventional quartz crystal detector is used, the strain can only be estimated at best, since the bond efficiency is usually unknown. Second, it is of importance in studying harmonic generation in solids from finite displacements to have absolute measures of the harmonic as well as the fundamental strain amplitudes. This is achieved in the present microphone by virtue of the flatness of electronic response between 10 and 100 Mc/sec. Across this range there is not more than 1% deviation in sensitivity from one extreme to the other. Finally, the microphone uses no bond (in contrast to the conventional quartz detector), so that the effect of bonds may be assessed.

CONTROLLED THERMONUCLEAR RESEARCH

Plasma Physics. — We have been investigating a very simple magnetic probe for studying the properties of a plasma. The probe is merely a bare wire immersed in the plasma. When an electric current of high amperage flows through the wire, the wire is surrounded by a magnetic field which repels charged particles. By measuring the electric current caused by the charged particles which are able to reach the wire, as a function of the magnetic field, we obtain information as to the momentum distribution of the charged particles in the plasma. In this respect, this probe differs from the Langmuir probe [Phys. Rev. 28, 727 (1962)], which measures the energy distribution of the charged particles. Although other workers have used magnetic fields to analyze the momentum of particles emitted from a plasma [Handbuch der Physik, vol. XXII, p. 61; J. Nucl. Energy: Pt. C 2, 69 (1961)], apparently the use of the probe wire itself as the magnet is novel.

Vacuum. — The studies of the behavior of organic compounds in vacuum systems have been continued in the ultrahigh-vacuum test facility. Experiments were conducted on the effect of electron bombardment of a surface during and immediately following admission of the saturated hydrocarbons heptane and dodecane. No evidence of hydrogen evolution was found unless the hydrocarbon pressure was increased to several microns, which presumably allowed a Townsend discharge to occur. An initial attempt to detect the presence of dodecane on the surface at a temperature below its normal boiling point (−10°C) failed when the electron bombardment technique was used. The ionization potential for this compound (about 10 v) is higher than that of bis-octylphthalate (about 6 v), for which the technique proved useful in previous studies. These findings appear to corroborate the conclusions tentatively reached in the earlier experiments that secondary electrons are involved in the decomposition reactions of absorbed compounds subjected to electron bombardment.

BIOLOGY AND MEDICINE

MOLECULAR- AND CELLULAR-LEVEL STUDIES

Physics of Tissue Damage: Measurements of Diffusion-Pump Oil Deposition with Radioactive Tracers. — A radioactive tracer technique is proposed as a quick, simple, nondestructive method for measuring the deposition of diffusion-pump oil. Ten milliliters of DC 704 oil was irradiated in the LITR (1.25×10^{13} neutrons cm^{-2} sec^{-1}) for 2.6 hr. The ^{30}Si, comprizing 0.54% by weight of the oil, becomes activated to ^{31}Si, which has a half-life of 2.6 hr and emits a beta ray of 1.48-Mev end-point energy. The oil was diluted to 225 cm^3 and added to the diffusion pump. After 1 hr of pumping on a bell jar, the jar was removed and the inside was scanned with a Geiger counter. The ratio of counts/cm^2 in the top of the jar to the counts/cm^3 of the bulk oil gave a thickness of 6 A. Use of larger radioactive samples and higher reactor fluxes may increase the sensitivity by more than two orders of magnitude.

Physics of Tissue Damage: Optical Properties of Graphite in the Far Ultraviolet Region. — The optical properties of hot-pressed pyrolytic graphite were studied in the wavelength region from 1100 to 3000 A. The real (n) and imaginary (k) parts of the complex index of refraction were obtained from reflectance measured at 20° and 70° incidence. The complex dielectric constant and the energy-loss function were determined from these n and k values. The energy-loss function, $Im(1/\epsilon)$, was found to exhibit a maximum at a wavelength of 1800 A, corresponding to an energy of 6.9 ev. This peak has been studied in electron energy-loss experiments, which have placed it at 7.5 ev and attributed it to the excitation of the conduction electron plasma.

TERRESTRIAL AND FRESH-WATER ECOLOGY

Analysis of Growth Responses of Two Sedges on White Oak Lake Bed. — An earlier report (ORNL-2903) concerning the lengths of inflorescences of

sedges (Carex vulpinoidea, C. Frankii) growing on White Oak Lake bed showed that inflorescences were longer in areas with higher air dose rates of gamma radiation. Because of variations in such factors as moisture, site fertility, and soil alkalinity, the differences in inflorescence length could not be ascribed to radiation dose with any certainty. To investigate effects of site factors, a further series of sedge inflorescences was measured from four areas on the lake bed and from four uncontaminated sites, and a number of soil factors were measured at each site. A statistical analysis was performed in which the factors soil pH, calcium, active phosphorus, exchangeable potassium, organic matter, and radiation dose rate in air were considered. The linear model

$$Y = \alpha + \sum_{i=1}^{k} \beta_i X_i$$

was examined, in which Y represents inflorescence length, α is a constant, β_i is a regression coefficient for the ith factor, X_i is an amount of the ith factor, and k = 0, 1, 2, 3, 4, 5, or 6. There are 2^6, or 64, possible models of this type, but in one of them (k = 6) the model would pass through the seven averages, leaving no means for assessing its validity. The remaining 63 models were examined with a computer program, which selected the best model with one through five variables. For Carex vulpinoidea the best general model was

$$Y = 63.55 + 0.2091\ R\ ,$$

where Y = inflorescence length (mm), and R = radiation dose rate in air (mr/hr) 1 m above ground. For Carex frankii the best general model was the three-term one:

$$Y = 157.83 + 0.5827\ R - 14.87\ pH + 0.4362\ Ca\ ,$$

where pH = hydrogen ion concentration in soil, and Ca = exchangeable calcium (lb/acre). For each sedge species, radiation dose rate in air provided the best single-term model and appeared as a significant factor in the two- to four-term models. However, it still remains likely that other soil factors associated with inorganic constituents of industrial wastes influence the lengths of inflorescences. An experimental approach (in both laboratory and field) would seem to be needed to resolve whether the observed increase in lengths of inflorescence is a direct response to radiation, an indirect effect, or the consequence of some purely chemical aspect of waste disposal. In any case a radiation response in natural environments is not likely to be entirely independent of other factors.

Environmental Influences on Responses of Metaphosphate Glass Dosimeters. — Application of glass-rod dosimetry is increasing in field radiation ecology studies. To qualify certain of these field uses, a study was made on the influence of temperature and sunlight on radiation response (fluorescence) of Toshiba low-Z rods. In a temperature range 3.5 to 53.5°C, rods showed no change in response to beta radiation from ^{137}Cs solutions. The response to ^{137}Cs gamma rays, however, increased with increasing temperature. A linear expression describing this increase is Y = 4.12 + 0.007 X, where Y is fluorometer units per rad, and X is temperature (°F).

Predosed rods were found to fade (lose fluorescence) exponentially when exposed to sunlight for 30 hr, given in 6-hr increments at about 10^4 ft-c. Rods lost 37 to 46% of the dose during the first 6-hr exposure. Fading was also related to predose, as high-predosed (5500 fluorometer units) rods had a significantly greater ($p < 0.01$) percentage of fading than low-predosed (400 fluorometer units) rods. The effectiveness of several kinds of shields was studied, and nylon plastic shields painted black were found to prevent fading when tested by 20-day exposures on White Oak Lake bed.

Effects of Growth Rate and Ionizing Radiation on Ribonucleic Acids in Mimosa Seedlings. — Mimosa (*Albizzia julibrissin* Durazzini) seedlings were germinated at controlled rates by varying temperature conditions. Total and soluble RNA (ribonucleic acid) extracts were made from each sample, and their concentration ratios were expressed as percentages of soluble RNA/total RNA. Base compositions of both soluble and total RNA were also determined for each sample. A linear correlation exists between base compositions (individual bases of soluble RNA) and growth rates. However, growth rate apparently has little, if any, effect on the soluble RNA/total RNA percentages.

Groups of Mimosa seedlings germinated at one optimal temperature condition are being irradiated in the ^{60}Co source room and will be used for RNA determinations similar to those for the above controls germinated at the same temperatures. Thus far, only one extraction of soluble RNA has been made from irradiated seedlings. The soluble RNA/total RNA ratio (23.69%) varied considerably from that for the controls (average of 13.94%). The base compositions also showed some differences. Guanine (34.2%) and cytosine (30.6%) were both slightly higher than control figures (averages of 32.7 and 29.3% respectively), while adenine (18.0%) and uracil (17.1%) were both slightly lower than control figures (averages of 20.3 and 17.7% respectively). The dose received by the Mimosa seedlings ranged from 500 to 1500 r over a four-day period preceding extraction.

Analog Computer Simulation of a Five-Compartment Ecosystem. — The pattern of coupling between parts of an ecosystem determines how radioisotopes and other substances will be distributed at equilibrium. Several experiments to illustrate this were performed with the following five-compartment system:

$$\dot{X}_1 = \tau_{01}X_0 - \tau_{12}X_1 - \tau_{10}X_1 - \tau_{14}X_1 ,$$
$$\dot{X}_2 = \tau_{12}X_1 + \tau_{42}X_4 \pm \tau_{23}X_2 - \tau_{20}X_2 - \tau_{24}X_2 ,$$
$$\dot{X}_3 = \tau_{23}X_2 - \tau_{30}X_3 - \tau_{34}X_3 ,$$
$$\dot{X}_4 = \tau_{14}X_1 + \tau_{24}X_2 + \tau_{34}X_3 - \tau_{42}X_4 - \tau_{40}X_4 ,$$

where X_i is the quantity of material in compartment i ($i = 0, 1, \ldots, 4$, with 0 denoting the environment, 1 the primary producers, 2 the herbivores, 3 the carnivores, and 4 the decomposers), and the τ_{ij} are instantaneous fractional transfer rates from compartment i to compartment j ($j = 0, 1, \ldots, 4$). The input was a constant signal, $X_0 = 5$ v.

With parameter values of $\tau_{01} = \tau_{12} = \tau_{23} = \tau_{i4} = 0.05$, $\tau_{42} = 0$, $\tau_{10} = 0.1$, $\tau_{20} = 0.2$, $\tau_{30} = 0.3$, and $\tau_{40} = 0.4$, the equilibrium masses

were $X_1 = 1.3$, $X_2 = 0.22$, $X_3 = 0.031$, and $X_4 = 0.19$ v, for a total of

$$X = 1.741 \text{ v} \left(X = \sum_{i=1}^{4} X_i \right).$$

Thus, 74.7% of the total material was in compartment 1, 12.6% in 2, 1.8% in 3, and 10.9% in 4. When τ_{42} was increased from 0 to 0.05 the values were $X_1 = 1.3$, $X_2 = 0.24$, $X_3 = 0.035$, $X_4 = 0.17$, and $X = 1.745$ v. The total equilibrium mass was increased slightly, with compartments 2 and 3 increasing and 4 decreasing. The relative distribution of material between compartments was only slightly altered: 74.5% in 1, 13.8% in 2, 2.0% in 3, and 9.7% in 4. When the parameters were changed to $\tau_{42} = 0$, $\tau_{10} = 0.4$, $\tau_{20} = 0.3$, $\tau_{30} = 0.2$, and $\tau_{40} = 0.1$ (all others as in the first experiment), the equilibrium was $X_1 = 0.5$, $X_2 = 0.06$, $X_3 = 0.001$, $X_4 = 0.861$ v. The total amount of material retained by the system was markedly reduced, and the distribution among compartments was quite different: 58.1% in 1, 7.0% in 2, 0.1% in 3, and 34.8% in 4. When τ_{01} was increased to 0.1 and τ_{12} to 0.075 (other parameters as in experiment 1), the material in the system was increased considerably at equilibrium: $X_1 = 2.2$, $X_2 = 0.55$, $X_3 = 0.08$, $X_4 = 0.36$, and $X = 3.19$ v. The distribution was 69.0% in 1, 17.2% in 2, 2.5% in 3, and 11.3% in 4.

These four experiments are sufficient to demonstrate that ecological systems with the same input of materials will vary greatly in the amounts of material retained at equilibrium as well as in the distribution between compartments, depending upon how their living components are coupled (the values of the τ_{ij} in the model). Therefore, a primary concern of empirical radioecology must be the elucidation of basic organization patterns in important natural ecosystems, for without such information predictions about how such systems will handle nuclides, and even how they will react to radiation stress, cannot be made with reasonable accuracy or precision.

Evaluation of Fission Product Distribution and Movement in White Oak Drainage System. — The sludge in abandoned waste pits 2 and 3 is primarily a mixture of calcium carbonate and illitic clay, with calcium carbonate comprising about 20% of the total weight of the material in pit 2 and 50% of the weight of the sludge in pit 3. Very small quantities of ^{90}Sr were leached from the sludge with water, base, weak acid, and weak neutral salt solution; however, 1 N neutral salt solutions and 1 N acid solutions were effective leaching agents for strontium. The only leaching solution to remove more than trace quantities of ^{137}Cs from the material was 1 N HNO_3. Most of the ^{90}Sr in the sludge is associated with the calcium carbonate, while the ^{137}Cs occupies highly selective exchange sites on the illitic clay fraction of the sludge.

ISOTOPE DEVELOPMENT

Solvent Extraction of Fission Products. — In recovering strontium and rare earths from Purex waste by the di(2-ethylhexyl)phosphoric acid (D2EHPA) extraction process, efficient separation of strontium from calcium was obtained in the second extraction cycle by making the aqueous feed 0.02 M in citrate and operating at a pH of 3.2 rather than at a pH of 2, as specified in the original flowsheet. More than 98% of the calcium was coextracted with about 100% of the rare earths (from which it can

be easily separated in subsequent cycles) in five extraction and three scrub stages, leaving about 98% of the strontium in the raffinate. Feed for the test was simulated first-cycle strip solution; the scrub was 0.01 M HNO_3, and the solvent was 0.3 M D2EHPA–0.15 M TBP–Amsco 125-82.

Radioisotope Research and Development. – A significantly improved generator for ^{99m}Tc has resulted from changes in the elution method. An activated alumina column of 50 to 200 mesh particles equilibrated with 0.1 M HCl adsorbs ^{99}Mo quantitatively from ammonium paramolybdate in 0.1 M HCl solution. Excess water is removed by air drying and methylethyl ketone (MEK) washes. Elution experiments were performed with MEK containing (1) 0.25 to 5.0% H_2O, (2) 0.25 to 5.0% 0.1 M HCl, and (3) 0.25% 0.001 to 0.1 M HCl. It was established that:

1. Increased H_2O concentration in MEK improved the elution of ^{99m}Tc.
2. An increase in HCl content alone did not increase the elution of ^{99m}Tc.
3. The solutions containing HCl gave somewhat poorer elution, since greater volumes of eluant were required to remove the ^{99m}Tc.
4. The 5% H_2O in MEK gave sporadic traces of molybdenum.
5. The 5% 0.01 M HCl in MEK would be a suitable eluant for the generator, since it extracted most of the ^{99m}Tc from the column with a reasonable volume of solution and showed no detectable ^{99}Mo activity. Neither the 5% H_2O in MEK nor the 5% 0.01 M HCl contained dissolved alumina.

This generator uses reactor-produced ^{99}Mo which is free of interfering activities, such as ^{103}Ru present in fission product ^{99}Mo, that could contaminate the ^{99m}Tc product.

A 250-mg enriched ^{58}Ni target, which had been irradiated for 7.65 years in the isotope stringer of the LITR at a flux of 1.15×10^{13} neutrons cm^{-2} sec^{-1}, was processed for ^{58}Co by use of the HCl–Dowex 2 system. The principal reactions, effective cross section at push time, half-lives, and yields are listed below:

Reaction	Effective Cross Section (barns)	Half-Life	Yield (mc)
$^{58}Ni(n,\gamma)^{59}Ni$	3.0	8×10^4 years	0.14
$^{58}Ni(n,p)^{58}Co$	5×10^{-3}	72 days	4.03

A portion of a 2.5-g NaCl target, which has been in the isotope stringer of the ORR for 3.95 years at a flux of $\sim 2.5 \times 10^{14}$ neutrons cm^{-2} sec^{-1}, was processed to determine the yields of ^{32}P and ^{33}P. The yields were favorable enough to warrant processing the entire target for ^{33}P. The main portion of the target had already been used for the separation of ^{36}Cl and ^{35}S. The phosphorus was associated with a relatively large amount of ^{35}S and therefore was recovered in an intermediate processing step during ^{35}S purification. The $^{33}P/^{32}P$ ratio will be 20:1 on July 1, and ~1.1 mc of ^{33}P will be available after the ^{32}P decays to less than 5%.

Radioisotope and Stable Isotope Production. – Ion currents equivalent to 3423 g of separated isotopes were monitored during May and June. Isotopes of Ba, Mg, Ni, Ru, S, Si, Se, and W were separated and collected, using 28 of the calutrons associated with this program.

OPEN LITERATURE PUBLICATIONS

Arakawa, E. T., N. O. Davis, L. C. Emerson, and R. D. Birkhoff, "The Angular and Spectral Distribution of Transition Radiation from Thin Silver Foils," J. Phys. Radium 25, 129 (1964).

Arakawa, E. T., R. J. Herickhoff, and R. D. Birkhoff, "Detection of Plasma Radiation from Electron-Bombarded Al and Mg Foils," Phys. Rev. Letters 12, 319 (1964).

Arsenault, R. J., "Reply to Discussion of 'Comments on the Yield Point in Iron,'" Acta Met. 12, 758 (1964).

Arsenault, R. J., "Low-Temperature Creep in Alpha Iron," Acta Met. 12, 547 (1964).

Aveston, J., E. W. Anacker, and J. S. Johnson, "Hydrolysis of Molybdenum(VI). Ultracentrifugation, Acidity Measurements, and Raman Spectra of Polymolybdates," Inorg. Chem. 3, 735 (1964).

Bair, J. K., C. M. Jones, and H. B. Willard, "Neutrons from the Proton Bombardment of ^{6}Li, ^{7}Li, ^{9}Be, ^{11}B, and ^{18}O," Nucl. Phys. 53, 209 (1964).

Bopp, C. D., "A Vibration Method for the Elastic Constants of Small Plates," Rev. Sci. Instr. 6, 703 (1964).

Carr, W. H., "Pilot Plant Processing of Uranium-Zirconium Alloy Nuclear Fuels by a Molten-Salt Fluoride-Volatility Method," J. Tenn. Acad. Sci. 39(2), 44 (1964).

Carr, W. H., S. Mann, and E. C. Moncrief, "Uranium-Zirconium Alloy Fuel Processing in the Oak Ridge National Laboratory Volatility Pilot Plant," Chem. Eng. Progr. 60, 56 (1964).

Condo, G. T., "On the Absorption of Negative Pions by Liquid Helium," Phys. Letters 9, 65 (1964).

Crossley, D. A., Jr., "Biological Elimination of Radionuclides," Nucl. Safety 5, 265 (1964).

Davies, K. T. R., and G. R. Satchler, "Spin-Spin Interactions and Scattering from Polarized Nuclei," Nucl. Phys. 53, 1 (1964).

Davis, W., Jr., and H. J. de Bruin, "New Activity Coefficients of 0–100 Per Cent Aqueous Nitric Acid," J. Inorg. Nucl. Chem. 26, 100 (1964).

DiStefano, J. R., and E. E. Hoffman, "Corrosion Mechanisms in Refractory Metal–Alkali Metal Systems," At. Energy Rev. 2(1), 3 (1964).

Dodd, C. V., "Applications of a Phase-Sensitive Eddy-Current Instrument," Mater. Evaluation 22(6), 260 (1964).

Drisko, R. M., and G. R. Satchler, "'Finite-Range' Effects in the Distorted-Wave Theory of Stripping Reactions," Phys. Letters 9, 342 (1964).

Hancher, C. W., and J. C. Suddath, "Pot Calcination of Simulated Radioactive Waste with Continuous Evaporation," Chem. Eng. Progr. Symp. Ser. 60, 105 (1964).

Horton, A. D., and J. L. Botts, "Gas Chromatography as Applied to Nuclear Technology. Analysis of the Hydrolysis Products of Uranium Carbides and Thorium Carbides," Nucl. Sci. Eng. 18, 97 (1964).

Kelley, M. T., C. Feldman, D. L. Manning, C. D. Susano, D. J. Fisher, R. W. Stelzner, J. S. Eldridge, W. S. Lyon, and P. F. Thomason, "A Résumé of the Seventh Conference on Analytical Chemistry in Nuclear Technology," J. Electroanal. Chem. 7, 165 (1964).

Kelley, M. T., C. D. Susano, J. S. Eldridge, C. Feldman, D. J. Fisher, W. S. Lyon, D. L. Manning, R. W. Stelzner, and P. F. Thomason, "Analytical Chemistry in Nuclear Technology," Science 144, 570 (1964).

Koehler, W. C., H. L. Yakel, E. O. Wollan, and J. W. Cable, "A Note on the Magnetic Structure of Rare Earth Manganese Oxides," Phys. Letters 9, 93 (1964).

Love, G. R., "Dislocation Pipe Diffusion," Acta Met. 12, 731 (1964).

Mahlman, H. A., and T. E. Willmarth, "Radiolytic and Photolytic Reduction of Aqueous Silver Nitrate Solutions," Nature 202, 590 (1964).

Martin, W. R., and J. R. Weir, "Effect of Post-irradiation Heat Treatment on the Elevated Temperature Embrittlement of Irradiated Stainless Steel," Nature 202, 997 (1964).

Moon, R. M., J. W. Cable, and W. C. Koehler, "The Magnetic Structure of Neodymium," J. Appl. Phys. Suppl. 35, 1041 (1964).

Neufeld, Jacob, and Harvel Wright, "Interaction of a Beam of Protons with Plasma," pp. 207–8 in Proceedings of the VIth International Conference on Ionization Phenomena in Gases, North Holland Publishing Co., Amsterdam, 1964.

Olson, J. S., "Gross and Net Production of Terrestrial Vegetation," J. Ecol. 52 (suppl.), 99 (1964).

Penny, S. K., and G. R. Satchler, "Inelastic Effects in Stripping and Other Direct Reactions," Nucl. Phys. 53, 145 (1964).

Perey, C. M., and F. G. Perey, "Optical-Model Analysis of 15-MeV Deuteron Elastic Scattering," Phys. Rev. 134, B-353 (1964).

Perey, F. G., and D. S. Saxon, "The Local Energy Approximation to Nonlocality and Finite Range Effects," Phys. Letters 10, 107 (1964).

Rudolph, P. S., and S. C. Lind, "Energy Transfer from Carbon Dioxide in the Radiation Induced Synthesis of Water," J. Chem. Phys. 40, 2601 (1964).

Silver, E. G., "SPERT Program Status Report," Nucl. Safety 5, 151 (1964).

Stokely, J. R., and F. L. Moore, "Selective Liquid-Liquid Extraction of Radiotin with 2-Thenoyltrifluoroacetone," Anal. Chem. 36, 1203 (1964).

Tamura, Taro, "Coupled-Channel Analysis of the Scattering of Protons by ^{165}Ho and ^{156}Gd," Phys. Letters 9, 334 (1964).

Tamura, Taro, and Takeshi Udagawa, "Higher Random Phase Approximation Applied to the Description of the Spherical Vibrational Nuclei," Nucl. Phys. 53, 33 (1964).

Vandermeer, R. A., and C. J. McHargue, "The Nature of the <001> Fiber Texture Component in Extruded Aluminum Rods," Trans. AIME 230, 667 (1964).

Weeks, R. A., and E. Lell, "Relation Between E′ Centers and Hydroxyl Bonds in Silica," J. Appl. Phys. 35, 1932 (1964).

Willard, H. B., J. K. Bair, and C. M. Jones, "N-D Scattering: A Search for Effects Due to the Dineutron," Phys. Letters 9, 339 (1964).

Yntema, J. L., and G. R. Satchler, "(d,^{3}He) Reaction on ^{40}Ca and the Titanium Isotopes," Phys. Rev. 134, B976 (1964).

Young, F. W., Jr., and L. D. Hulett, "Role of Crystal Imperfections in the Chemical Reactivity of Copper Surfaces," p. 375 in Metal Surfaces, American Society for Metals, Metals Park, Ohio, 1963.

Young, F. W., Jr., and J. R. Savage, "Growth of Copper Crystals of Low Dislocation Density," J. Appl. Phys. 35, 1917 (1964).

VISITORS

UNITED STATES

Advisory Committee for the Electronuclear and Physics Divisions

Clogston, A. M.	Rainwater, L. J.	Wheeler, J. A.
Cohen, B. L.	Judd, D. L.	

FOREIGN

Name	Affiliation	Citizen of
Aalto, E. J.	A B Atomenergi, Sweden	Finland
Barnes, R. S.	United Kingdom AEA	United Kingdom
Bell, I. P.	United Kingdom AEA	United Kingdom
Bogardi, J.	Self	Hungary
Boudry, B.	French CEA	France
Bromley, D. A.	Yale University	Canada
Bush, W. R.	Atomic Energy of Canada, Ltd.	Canada
Cerre, M. P.	French CEA	France
Charlton, J. C.	United Kingdom AEA	Great Britain
Clare, G. W.	United Kingdom AEA	United Kingdom
Cohen, G. G.	French CEA	France
Cohen, Y.	French CEA	France
Davies, C. N.	United Kingdom AEA	United Kingdom
Debiar, A.	French CEA	France
de Calmes, A.	French CEA	France
d'Eye, R. W. M.	United Kingdom AEA	United Kingdom
Dickson, A. N.	Dragon Project	United Kingdom

Name	Affiliation	Citizen of
Dirian, J. M. A.	French CEA	France
Doshi, K. D.	Franklin Institute	India
Dunster, H. J.	United Kingdom AEA	United Kingdom
Estournel, R.	French CEA	France
Farrell, M. S.	Australian AEC	Australia
Fernandez, N.	French CEA	France
Findlay, J. R.	United Kingdom AEA	United Kingdom
Fryer, D. R.	Ministry of Power	United Kingdom
Fujiya, M.	University of Michigan	Japan
Gittus, J. H.	United Kingdom AEA	United Kingdom
Gras, M.	French CEA	France
Harries, D. R.	United Kingdom AEA	United Kingdom
Hogetsu, K.	Tokyo Metropolitan University	Japan
Izawa, M.	National Institute of Radiological Science	Japan
Jones, I. S.	United Kingdom AEA	United Kingdom
Juliano, J. O.	Philippine AEC	Philippines
Kelly, R. G.	Atomic Energy of Canada	Canada
Kim, S. M.	ORINS	Korea
Kistemaker, J.	F. O. M. Institute	Netherlands
Kronberger, H.	British Government	United Kingdom
Lavaud, B. A.	French CEA	France
Lefillatre, G.	French CEA	France
Legge, I. K.	Ministry of Labor	United Kingdom
Leterrier, G. L.	Electricite de France	France
Martin, J. J.	Electricite de France	France
Maushart, R.	Karlsruhe University	Germany
Mawson, C. A.	AECL, Chalk River	Canada
Mestre, E.	French CEA	France
Murray, P.	United Kingdom AEA	United Kingdom
Nicholson, R. L. R.	British Government	United Kingdom
Nishiwaki, Y.	Tokyo Institute	Japan
Oh, K. C.	University of Tennessee	Korea
Perret, R.	Brookhaven National Laboratory	France
Pillai, K. C.	A. E. E. T., India	India
Poedjiardjo, S.	Institute for Atomic Energy, Indonesia	Indonesia
Rebiffe, J.	French CEA	France
Recht, P.	EURATOM	Belgium
Reichenberg, D.	Rutgers University	United Kingdom
Sangster, D. F.	Australian AEC	Australia
Sayers, J. B.	United Kingdom AEA	United Kingdom
Schroeder, R.	University of California, L.A.	Germany
Sebille, J. F. E.	EURATOM	Belgium
Sevin, P. R.	Electricite de France	France
Siegfried, W.	Institute Battelle	Switzerland
Spreadborough, J.	Battelle — Geneva	United Kingdom
Swindell, G. E.	International AEC	Austria
Ting, S. F.	University of Tennessee	China
Tryor, J. G.	United Kingdom AEA	United Kingdom

Name	Affiliation	Citizen of
Van Hoeck, F. F.	EURATOM	Belgium
Vithal, B. P. R.	Osmania University	India
Wait, E.	United Kingdom AEA	United Kingdom
Whitfield, R. T.	Rolls Royce Corp.	United Kingdom
Williams, I. G. K.	United Kingdom AEA	United Kingdom
Wormser, G.	French CEA	France
Wrigley, H. E.	United Kingdom AEA	United Kingdom
Yatsurugi, T.	Japan AERI	Japan
Yoshida, T.	Purdue University	Japan

Reports issued in this series during the past year:

June 1963	ORNL-3477
July 1963	ORNL-3491
August 1963	ORNL-3508
September 1963	ORNL-3517
October 1963	ORNL-3532
November 1963	ORNL-3543
December 1963	ORNL-3561
January 1964	ORNL-3568
February 1964	ORNL-3590
March 1964	ORNL-3614
April 1964	ORNL-3628
May 1964	ORNL-3647

ORNL-3663
C-44a — Nuclear Technology — Materials
M-3679 (35th ed.)

INTERNAL DISTRIBUTION

1. I. Alexeff
2. C. F. Baes
3. E. A. Bagley
4. P. S. Baker
5. C. R. Baldock
6. L. H. Barker
7. S. E. Beall
8. E. E. Beauchamp
9. P. R. Bell
10. D. S. Billington
11. C. A. Blake
12. R. E. Blanco
13. F. F. Blankenship
14. E. P. Blizard
15. J. O. Blomeke
16. A. L. Boch
17. E. G. Bohlmann
18. C. J. Borkowski
19. G. E. Boyd
20. M. A. Bredig
21. J. C. Bresee
22. R. B. Briggs
23. R. E. Brooksbank
24. K. B. Brown
25. F. R. Bruce
26. W. D. Burch
27. T. H. J. Burnett
28. A. D. Callihan
29. A. E. Cameron
30. T. E. Cole
31. C. F. Coleman
32. E. L. Compere
33. D. D. Cowen
34. J. A. Cox
35. F. L. Culler
36. J. E. Cunningham
37. R. A. Dandl
38. Wallace Davis
39. J. S. Drury
40. J. L. Dunlap
41. A. C. England
42. M. F. Fair
43. D. E. Ferguson
44-47. J. L. Fowler
48. J. H. Frye, Jr.
49. J. L. Gabbard (K-25)
50. W. F. Gauster
51. J. H. Gillette
52. W. Y. Gissel
53. H. E. Goeller
54. C. D. Goodman
55. A. T. Gresky
56. J. C. Griess
57. W. R. Grimes
58. P. A. Haas
59. C. S. Harrill
60. J. C. Hart
61. P. N. Haubenreich
62. M. R. Hill
63. C. J. Hochanadel
64. A. Hollaender
65. L. B. Holland
66. R. W. Horton
67. F. T. Howard
68. A. S. Householder
69. H. H. Hubbell
70. G. S. Hurst
71. K. E. Jamison
72. G. H. Jenks
73. R. W. Johnson
74. R. J. Jones
75. W. H. Jordan
76. P. R. Kasten
77. G. W. Keilholtz
78. G. G. Kelley
79. M. T. Kelley
80. E. M. King
81. K. A. Kraus
82. Laboratory Shift Supervisor
83. J. A. Lane
84. D. M. Lang (K-25)
85. C. E. Larson
86. C. F. Leitten, Jr.
87. R. E. Leuze
88. L. F. Lieber
89. T. A. Lincoln
90. S. C. Lind
91. R. B. Lindauer
92. R. S. Livingston
93. J. T. Long
94. J. E. Longendorfer
95. L. O. Love

96. R. N. Lyon
97. H. G. MacPherson
98. F. C. Maienschein
99. J. A. Martin
100. H. F. McDuffie
101. J. R. McNally
102. R. A. McNees
103. A. J. Miller
104. C. E. Miller, Jr.
105. E. C. Miller
106. K. Z. Morgan
107. W. L. Morgan
108. E. J. Murphy
109. R. V. Neidigh
110. M. L. Nelson
111. G. W. Parker
112. R. B. Parker
113. P. Patriarca
114. S. Peterson
115. J. J. Pinajian
116. M. E. Ramsey
117. A. W. Riikola
118. L. P. Riordan
119. A. F. Rupp
120. A. D. Ryon
121. H. C. Savage
122. H. W. Savage
123. A. W. Savolainen
124. H. E. Seagren
125. C. H. Secoy
126. E. D. Shipley
127. O. Sisman
128. J. R. Sites
129. M. J. Skinner
130. A. H. Snell
131. W. S. Snyder
132. W. L. Stirling
133. R. W. Stoughton
134. H. Stringfield
135. E. G. Struxness
136. J. C. Suddath
137. W. H Sullivan
138. C. D. Susano
139. J. A. Swartout
140. E. H. Taylor
141. R. E. Thoma
142. C. D. Watson
143. B. S. Weaver
144. M. S. Wechsler
145. A. M. Weinberg
146. M. E. Whatley
147. J. C. White
148. E. O. Wollan
149. R. W. Worsham
150. R. G. Wymer
151. Gale Young
152. W. Zobel
153. A. Zucker
154-155. Central Research Library
156-157. ORNL — Y-12 Technical Library Document Reference Section
158. Reactor Division Library
159. Biology Library
160-313. Laboratory Records Department
314. Laboratory Records, ORNL R.C.

EXTERNAL DISTRIBUTION

315. Division of Reactor Development, AEC, Washington (J. M. Simmons)
316-325. Research and Development Division, AEC, ORO
326. Aeroprojects, Inc. (W. B. Tarpley)
327. Allis-Chalmers Manufacturing Co. (W. F. Banks)
328-330. Ames Laboratory, Iowa State College (F. H. Spedding)
331-333. Argonne National Laboratory (A. V. Crewe)
334. Atomics International (H. Pearlman)
335. Babcock and Wilcox Co. (Atomic Energy Division)
336-337. Battelle Memorial Institute (H. W. Russell)
338-340. Brookhaven National Laboratory (M. Goldhaber)
341. Bureau of Mines, Salt Lake City (J. B. Clemmer)
342-349. General Atomic (General Dynamics Corp.) (Manager)
350-351. General Nuclear Engineering Corp. (J. West)
352. Grand Junction Operations Office (W. C. Hutchinson, Jr.)
353. Kaiser Engineers (P. D. Bush)
354-355. Knolls Atomic Power Laboratory (K. R. Van Tassell)

356-357. Lawrence Radiation Laboratory (H. Fidler)
358-359. Los Alamos Scientific Laboratory (N. E. Bradbury)
360-361. National Bureau of Standards (L. S. Taylor)
362. National Lead Company (C. K. McArthur)
363. Nuclear Metals, Inc. (A. R. Kaufmann)
364-365. Philadelphia Electric Co. (L. R. Gaty)
366. Phillips Petroleum Company, NRTS (J. P. Lyon)
367. Sanderson and Porter (Atomic Energy Division)
368. Union Carbide Corporation (R. W. McNamee)
369. USAEC, Assistant General Manager (S. G. English)
370. USAEC, Division of Biology and Medicine (C. L. Dunham)
371-373. USAEC, Division of Isotope Development (P. C. Aebersold)
374. USAEC, Division of Production (F. P. Baranowski)
375. USAEC, Division of Raw Materials (J. C. Johnson)
376-390. USAEC, Division of Reactor Development (F. K. Pittman)
391-396. USAEC, Division of Research (P. W. McDaniel)
397. USAEC, Division of Research, Special Projects Branch, Washington (Dorothy Smith)
398-399. USAEC, Division of Technical Information (Technical Reports Library and 1 copy to R. E. Bowman, Room D-403)
400-401. USAEC, Chicago Operations Office
402. USAEC, Lockland Aircraft Reactors Operations Office (H. H. Gorman)
403-404. USAEC, San Francisco Operations Office (R. Hughey)
405-406. USAEC, Savannah River Operations Office (H. Rahner)
407. Westinghouse Electric Corp. (A. Squire)
408-652. Given distribution as shown in M-3679 (35th ed.) category C-44a and those addresses shown in C-44b, C-44c, and C-44d, which do not receive copies under C-44a