[redacted]

MARTIN MARIETTA ENERGY SYSTEMS LIBRARIES
3 4456 0365953 8

150

ORNL-3477
C-44a – Nuclear Technology – Materials
M-3679 (29th ed.)

OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

JUNE 1963

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 5, 1964

[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-3477

Contract No. W-7405-eng-26

OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

JUNE 1963

DATE ISSUED

JUL 11 1963

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

MARTIN MARIETTA ENERGY SYSTEMS LIBRARIES

3 4456 0365953 8

[redacted]

[redacted]

CONTENTS

REACTOR DEVELOPMENT PROGRAM..... 1
GAS-COOLED REACTOR PROGRAM..... 1
Irradiations of Advanced Clad Fuel Elements in GCR-ORR Loop No. 1..... 1
Capsule Irradiations in the LITR..... 1
Coated-Particle Irradiations in ORR Capsules..... 1
Eight-Ball Experiment..... 2
Instrumented Fuel Assemblies..... 2
Fission Product Deposition..... 2
Thermal Stress and Fuel Migration in Coated Particles..... 2
Experimental Studies on Poison Particles for Fueled-Graphite Reactor Applications..... 3
Geometry Studies of Graphite..... 4
ARMY POWER REACTORS..... 4
FUNDAMENTAL HEAT TRANSFER AND FLUID DYNAMICS..... 5
Boiling-Water Studies..... 5
Reynolds Stress Measurements..... 5
Boiling-Potassium Studies..... 6
POWER REACTOR FUEL PROCESSING..... 6
Studies with Irradiated Uranium Carbides..... 6
Dissolution of Sintered UO_2-PuO_2 Pellets..... 7
Shear-Leach Process..... 7
Solvent Extraction..... 7
Protactinium Adsorption..... 7
Chemical Analysis of Advanced Reactor Fuels..... 8
THORIUM UTILIZATION PROGRAM..... 8
Kilorod Program..... 8
Fuel Cycle Development: Fuel Fabrication Studies..... 8
Fuel Cycle Development: Chemistry of Solid Fuel Element Materials..... 9
Preparation of Dense UO_2 by Precipitation Methods..... 9
Preparation of Thorium Microspheres..... 9
MOLTEN-SALT REACTOR PROGRAM..... 9
Molten-Salt Reactor Experiment..... 9
MSR Design..... 10
Reactor Analysis..... 10
Development of Components and Systems..... 10
MSRE Pump Program..... 11
Instrumentation Development..... 11
NUCLEAR SAFETY..... 12
Release of Fission Products on Out-of-Pile Melting of Reactor Fuels..... 12
Attenuation of Fission Product Escape by Intrinsic Processes..... 13
Characterization and Control of Accident-Released Fission Products..... 13
Nuclear Safety Pilot Plant..... 14
Reactor Containment Handbook..... 14
Nuclear Safety Information Center..... 14

[redacted]

[redacted]

WASTE DISPOSAL DEVELOPMENT..... 14
Clinch River Study..... 14
Mineral Exchange Studies..... 15
Disposal in Deep Wells..... 15
Low-Activity Wastes: Scavenging-Precipitation—Ion Exchange..... 16
Low-Activity Wastes: Foam Separation..... 16
High-Activity Waste: Pot Calcination..... 17
High-Activity Waste: Fixation in Glass..... 17
Disposal in Natural Salt Formations..... 18

PHYSICAL RESEARCH PROGRAM..... 19
PHYSICS AND MATHEMATICS RESEARCH..... 19
High-Energy Electronuclear Machines: Superconducting Magnet Studies..... 19
Medium-Energy Nuclear Physics: Heavy Particles..... 19
Medium-Energy Nuclear Physics: The 86-Inch Cyclotron..... 19
Cyclotron Operations: The Isochronous Cyclotron..... 20
High-Voltage Experimental Program: The $O^{18}(\alpha,p)F^{21}$ Reaction and the Beta Decay of F^{21}..... 20
High-Voltage Experimental Program: Total Neutron Cross Sections in Isotopes of Zr, Fe, Cr, and Ca..... 20
Stable Isotopes Development..... 21
Special Separations..... 22
CHEMISTRY RESEARCH..... 22
Thorium Oxide Studies..... 22
Protactinium Chemistry..... 22
Equipment Decontamination..... 23
Pyrolysis and Radiolysis of Organic Liquids..... 23
High-Temperature Aqueous Solution Chemistry..... 24
Analysis of Molten Salts by Electrochemical Methods..... 24
Properties of Aqueous Solutions: Species and Thermodynamic Properties..... 25
Radiation Chemistry..... 25
Target Preparation..... 25
Transuranium Element Processing..... 26
Corrosion Tests for the TRU Facility..... 26
Preparation of PuO_2 for HFIR Target Prototypes..... 26
Separation of Americium from Curium at High Alpha Levels..... 27
Direct, Nondestructive Method for the Determination of Cf^{252}..... 27
TRU Processing Engineering..... 27
METALLURGY AND MATERIALS RESEARCH..... 28
Preparation of Pure Materials..... 28
CONTROLLED THERMONUCLEAR RESEARCH..... 28
Plasma Physics and Arc Research..... 28
DCX-1 Facility..... 28
DCX-2 Facility..... 29
Vacuum..... 29

BIOLOGY AND MEDICINE PROGRAM..... 30

[redacted]

[redacted]

TERRESTRIAL AND FRESH-WATER ECOLOGY..... 30
Evaluation of Fission Product Distribution and Movement in White Oak Drainage System..... 30
RADIOLOGICAL PHYSICS, HEALTH PHYSICS, AND INSTRUMENTATION..... 30
Health Physics Research Reactor Studies..... 30

ISOTOPE DEVELOPMENT PROGRAM..... 31
Nuclear Analysis Research and Development: Use of a 14-Mev Neutron Generator..... 31
Nuclear Analysis Research and Development: Preparation of a Guidebook on Activation Analysis..... 31
Nuclear Analysis Research and Development: Assistance to Law Enforcement Officers in Evaluating Drug Seizures..... 31
Curium-242 Recovery..... 31
Recovery of Fission Products..... 32
Radioisotope Research and Development..... 32
Radioisotope and Stable Isotope Production..... 33

OPEN LITERATURE PUBLICATIONS..... 34

VISITORS..... 35

[redacted]

[redacted]

OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

JUNE 1963

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 PROGRAM

GAS-COOLED REACTOR PROGRAM

Irradiations of Advanced Clad Fuel Elements in GCR-ORR Loop No. 1. — A second roughened-surface element, identical to the first, failed during startup of the reactor. It was withdrawn to a position above the reactor core and has remained there throughout the present ORR operating cycle. Portions of the element exceeded the allowable temperature, for reasons as yet unknown, before the reactor had reached full power. Although this element operated only briefly at low power, the release of fission gases to the loop was greater than for the previous element. No operational difficulties with the loop resulted from the activity release.

Capsule Irradiations in the LITR. — A capsule containing pyrolytic-carbon-coated particles of $(U + Th)C_2$ manufactured by General Atomic was removed from the LITR at a calculated burnup of 6.4 at. % of the heavy metal. During approximately one year of irradiation, the central fuel temperature in compartment a of the capsule drifted from 1935 to 1650°F and that in compartment b drifted from 2115 to 2050°F.

The LITR tank was revised to provide, for the GCR capsules, an access that would not interfere with removal of the cover for refueling. The new access provisions greatly simplify handling of the capsules and operation of the reactor and reduce the likelihood of failure of experimental assemblies as a result of frequent handling, which had been required in the previous arrangement.

Coated-Particle Irradiations in ORR Capsules. — Three sweep capsules are being operated with 1-1/2-in.-diam graphite-matrix spheres containing pyrolytic-carbon-coated uranium carbide particles. The sphere surface temperatures in these capsules vary from 1000 to 1500°F, the fission power densities vary between 10 and 45 w/cm^3, and the ratio of the fission-gas-release rate to the birth rate for Kr^{88} is $\sim 1 \times 10^{-5}$. The release rates have not shown significant variation with operating conditions, after operation to a burnup of ~1% of the total uranium.

[redacted]

Eight-Ball Experiment. – The first experimental capsule containing eight 1-1/2-in.-diam coated-particle-fuel graphite-matrix spheres operated satisfactorily at estimated power densities varying from 33 to 40 w per cm^3 of fueled sphere. Surface temperatures were generally between 1100 and 1250°F. The capsule was removed from the reactor at completion of its scheduled one-cycle irradiation.

Instrumented Fuel Assemblies. – Four instrumented fuel assemblies for testing in the EGCR were completed, except for the shield plug and final installation, which must await completion of components to be delivered by outside vendors. Except for minor design changes for simplification of the assembly, the work progressed in accordance with the original plan.

Fission Product Deposition. – Two sections of piping from GCR-ORR loop No. 2, one from the high-temperature zone, operated at 1000°F, and the other from the low-temperature zone, operated at 520°F, were analyzed for fission product deposition. The following results were obtained (expressed in units of dis min^{-1} $in.^{-2}$):

Fission Product	1000°F Zone	520°F Zone
Ba^{140}	6.5×10^4	1.5×10^5
Ru^{103}	9.0×10^4	
Ru^{106}	5.8×10^3	0
Zr^{95}	6.3×10^4	126
Nb^{95}	4.3×10^4	210
Te^{129m}	1.2×10^4	0
Ce^{144}	1.4×10^4	4.7×10^3

Mathematical analyses of fission product deposition as a function of metal surface temperatures indicated that the governing differential equation for isothermal deposition would also apply to nonisothermal regions if the probability that a particle would deposit on the surface were made temperature dependent. A computer program for solving the differential equation is being prepared.

Thermal Stress and Fuel Migration in Coated Particles. – Samples of pyrolytic-carbon-coated uranium carbide and uranium-thorium carbide particles were subjected to severe thermal-cycling treatments to observe the effect on coating integrity, from the standpoints of mechanical stability and fuel migration. Rapid cooling is of special interest, because it places the coating in tension as it contracts around the carbide core particle.

Four types of coated particles were investigated. These materials represented variations in average coating thickness (44 to 106 μ), coating microstructure (laminar and duplex), and fuel composition (uranium carbide and uranium-thorium carbide with thorium-to-uranium ratio of 0.6).

The samples were held at temperature (2400 to 2900°C) for 120 to 140 sec. Damage to particle coatings resulting from the thermal-cycling treatments was determined by leaching with 8 M HNO_3 and by microradiographic examination.

The amounts of uranium removed by acid leaching indicate that coating thickness is the strongest single variable in determining resistance to severe thermal cycling. A relatively thin (44 μ) laminar coating showed the weakest resistance; cycling to 2400°C exposed 0.1% of the contained uranium, 2500°C exposed 75%, and 2600°C caused total destruction of the particles. For a 97-μ laminar coating deposited at a low temperature (1400°C), 0.2% of the contained uranium was exposed after heating to 2500°C, but after a thermal cycle to 2600°C, virtually 100% of the fuel was exposed. A second relatively thick laminar coating (91 μ) deposited under different conditions required cycling to 2750°C to expose 0.8% of its uranium for leaching, and heating to 2900°C exposed only 5% of the fuel. The duplex coating, which has a thickness of 106 μ, showed strongest resistance to thermal cycling of the four lots examined; cycling to 2600°C exposed 0.005% of the uranium, while heating to an estimated 2900°C exposed only 0.3%. The results indicate that resistance to thermal cycling is not a simple function of coating thickness; such factors as coating structure and method of preparation play important roles.

Microradiographic examination of these samples showed that fuel migration was reasonably consistent with the acid leaching results. The thinly coated and low-temperature laminar samples showed gross movement of the fuel to the outer surface of the coatings after treatment at 2400 to 2600°C. A more restricted movement of fuel was observed in the altered laminar coatings, but severe coating removal occurred during heat treatment of this material. The duplex-coated particles showed the most restricted movement of fuel, while exhibiting no obvious change in coating integrity. None of the coating failures could be attributed to the effects of thermal stress.

Experimental Studies on Poison Particles for Fueled-Graphite Reactor Applications. — Angular particles of Lindsay mix No. 203 (45% Gd_2O_3, 45% Sm_2O_3, 10% other rare-earth oxides) and Eu_2O_3 in the size range −60 +100 mesh were successfully coated at 1400°C with 65 to 100 μ of pyrolytic carbon. Radiography and metallography showed the coatings to be uniform in thickness and apparently well bonded to the particles.

Short-term thermogravimetric tests in vacuum indicated that the europium oxide particles are compatible with pyrolytic-carbon coatings to temperatures of at least 1665°C for a period of 1 hr, and perhaps to higher temperatures, since no weight loss was detected below 1740°C in either of two experiments.

Thermogravimetric measurements on powdered mixtures of rare-earth oxides and graphite heated under the same conditions indicated that reaction begins at approximately 1100°C and that at temperatures between 1400 and 1500°C, only carbon would remain, given sufficient time. The data can be interpreted in terms of the following reactions above 1100°C:

$$R_2O_3(s) + 7C(s) \rightarrow 2R(\ell) + 4C(s) + 3CO(g) ,$$

$$2R(\ell) \rightarrow 2R(g) ,$$

where R denotes the rare earth.

A dense pyrolytic-carbon coating on the rare-earth oxide particles apparently serves to contain any CO formed below 1740°C, so the reaction of rare earth and carbon is suppressed when the equilibrium CO pressure is reached.

Spheroidal rare-earth carbide particles could not be formed by the process of reacting granules of powdered rare earth and carbon mixtures, because of liquid phase formation at 1700°C, which is well below the temperature (2300°C) at which the ultimate and more thermodynamically stable form of the carbide (dicarbide) is produced. A two-stage approach for forming spheroidal particles by reheating arc-melted rare-earth carbide particles shows promise. When separated by powdered graphite and heated to 2500°C for 30 min in argon, the angularly shaped starting granules show a definite change toward a spheroidal shape.

Geometry Studies of Graphite. — The effects of fast-neutron-induced stresses developed in fully constrained cylindrical sectors were reported in December (ORNL-3402). Since that time additional studies were made to evaluate the influence of various constraints. In general, constraint conditions can be fully specified by defining the axial strain and modifying the equation for axial stress accordingly.

Three constraint conditions were studied: (1) complete axial constraint, in which case the axial strain is zero, (2) constraint against bowing or warping, in which case the axial force is zero but parallel plane cross sections remain plane and parallel after loading, and (3) no axial constraint, in which case the force and moments associated with the axial stress are zero. There is not sufficient freedom allowed by the constraint equations to satisfy exactly and everywhere the equilibrium conditions for the body; however, with the aid of St. Venant's principle, a good estimate of the stresses can be obtained, except near the ends of the body.

The data obtained showed that the constraint conditions greatly influenced the axial stress. The maximum axial and in-plane stresses were always located at the intersection of the sector angle bisector and the inner surface, whereas the minimum axial stress was along a radial edge. The minimum in-plane stress was either inside the body or on the inner surface.

ARMY POWER REACTORS

Previously reported work showed that magnetite can remove most of the activity from samples of water withdrawn from the ORR pressurized-water in-pile loop. In a recent test a side stream of loop water was diverted from the loop pump outlet through a magnetite cartridge and then returned to the pump inlet. The temperature of the magnetite column varied between 217 and 345°F. The adsorption efficiency of the column was between 65 and 94%, but the results are somewhat uncertain because of variations in the activity of the loop water during the test. At the end of the test the concentration of activity on the column was found to decrease in the direction of flow by more than a factor of 10.

During the test the permeability of the column decreased and some magnetite was observed in samples of the filtrate, indicating that the sintered particles of magnetite were breaking down into fines. The magnetite used in the test had been prepared by sintering Mapico Black (synthetic magnetite of 0.2 to 0.8 μ particle size) at 1000°C, grinding, and sieving to give a −30 +50 mesh sample. A sample of this material was also found to break down into fines after being irradiated in the ORR and then treated with water at 260°C. When the sintering temperature was increased to 1400–1440°C, the resulting material was found to have better resistance to mechanical degradation. Some of this magnetite was irradiated and is being used for determining the solubility of Fe_3O_4 in water at 260°C.

FUNDAMENTAL HEAT TRANSFER AND FLUID DYNAMICS

Boiling-Water Studies. – Test sections were designed and fabricated for use in two-phase swirl-flow boiling-water investigations. These sections are 1/4- and 1/2-in.-ID "A" nickel tubes, 22 in. long and fitted internally with twisted tapes of 0.015-in.-thick Inconel. The initial objective of the study is the determination of critical heat fluxes for a vertically oriented test section over the following ranges of variables: exit pressure, 15 to 30 psia; inlet water velocity, 1 to 3 fps; tape twist ratios, 2.5 to 8.0 inside diameters per 180° twist; inlet subcooling, 0 to 30°F; exit quality, 30 to 80 wt %. Three heat exchangers, that is, a steam preheater, a condenser, and a liquid-return cooler, were designed and ordered for use in this program.

The temperature dependence of the electrical resistivity of seamless 70–30 brass tubing was experimentally determined over the temperature range 70 to 500°C. The results obtained with two test specimens of different lengths were in good agreement. The mean values of resistivity are:

T, °C	100	200	300	400	500
ρ_e, microhm-cm	7.39	8.29	9.37	10.58	11.75

Reynolds Stress Measurements. – Three sets of measurements of Reynolds stresses as a function of radial position in a pipe were completed. The Reynolds numbers used in these tests were 2.2×10^5, 1.8×10^5, and 2.5×10^5. Analysis of the first set of data showed that the magnitudes of the three normal stresses were of the same order as those reported by Laufer for air at a somewhat lower Reynolds number. In addition, the values of two of the shear components, $\overline{v_x v_\theta}$ and $\overline{v_r v_\theta}$, were nearly, if not actually, zero. These two components are normally assumed to be zero in pipe flow; however, they have never actually been measured to be so. The scatter of the first set of data was greater than desirable for detailed analysis but was reduced markedly in the second and third tests by decreasing the hot-wire current. The resulting decrease in sensitivity was compensated for by the increased range in wire orientations.

Boiling-Potassium Studies. — The experimental study of forced-convection saturation boiling with liquid potassium was temporarily delayed as the result of extensive damage to the new high-flux boiler prior to installation. Salvage and rebuilding of this unit are proceeding. During this delay, the design and construction of an alternate boiler that will be capable of fluxes of the order of 100,000 Btu hr^{-1} ft^{-2} at the boiling surface were initiated. This test section will be constructed from an 0.84-in.-OD, 0.29-in.-ID, type 347 stainless steel tube. Pressure taps will be located at five positions along the tube. Gunbarrel-type thermocouples will be inserted radially into the wall at two axial levels near the boiler exit. By locating several of these couples at various depths within the wall at each measuring station, it is hoped to gain information on the heat flux and to increase the precision to which the wall temperature is known. Heat will be supplied radiantly to the boiler from a set of surrounding clamshell heaters. The power input to these heaters will provide an additional measure of the heat flux. Losses from these primary heaters will be minimized by a second jacketing set of clamshell heaters.

In an effort to determine the effect of surface conditions on the boiling of potassium, a capsule was designed to measure the wall superheat required to initiate boiling from a surface of controlled roughness with a predetermined cavity size and shape. With this apparatus it is hoped to substantiate, with potassium, as Grifith and Wallis have done with water, ethanol, and methanol, the equation of Ellion relating the wall superheat required to maintain boiling from a surface of a given cavity size to the physical properties of the fluid.

POWER REACTOR FUEL PROCESSING

Studies with Irradiated Uranium Carbides. — Low-level irradiation of uranium monocarbide to about 500 Mwd/ton in a niobium can with an estimated surface temperature of 800°C produced a marked change in the hydrolysis properties of the material. The irradiated carbide, when reacted with water at 80°C, yielded 96 ml (STP) of gas per gram of sample; the gas was 28% hydrogen, 67% methane, 3% ethane, 1.0% higher alkanes, and 0.8% unsaturates. In contrast, unirradiated pellets of uranium monocarbide from the same rods as the irradiated specimens yielded 90 ml (STP) of gas per gram of carbide; the gas was 9% hydrogen, 88% methane, 2% ethane, 0.6% higher alkanes, and 0.2% unsaturates. Only 86% of the carbon present in the specimens before irradiation was found in the gas phase, compared with 98% for the irradiated sample. The nonvolatile residue dissolved completely in 6 N HCl, yielding a solution of tetravalent uranium and fission products. Heating unirradiated specimens in a niobium can for three weeks (the length of the irradiation period) at 800°C had no effect on the hydrolysis products, nor did heating other samples of UC to 2000°C. This indicates that the effect observed with the irradiated specimens is not thermally induced.

Dissolution of Sintered UO_2-PuO_2 Pellets. — Unirradiated fuel pellets containing 80% UO_2—20% PuO_2 (81% of theoretical density, prepared by mechanical blending, fired at 1600°C for 48 hr in an N_2—6% H_2 atmosphere) were dissolved completely in 14, 7, and 3 N HNO_3 in 25, 100, and 180 min respectively. Similar material, 92% of theoretical density, required 360 min to dissolve in 10 N HNO_3. This high-density pellet contained 82.2% UO_2—17.8% PuO_2 and was coprecipitated and fired under similar conditions for 16 hr. In each experiment the ratio of uranium to plutonium in the solution was constant throughout the dissolution.

Shear-Leach Process. — Bench-scale leaching tests on sheared stainless-steel-clad UO_2 and ThO_2-UO_2 were continued. In one run, 1-in. sheared lengths of stainless-steel-clad ThO_2-UO_2 suspended in a basket in a recirculating leacher but without sparging were 90% leached in the same length of time (7 hr) as required for 1/2-in. sheared lengths in a sparged system. This result was unexpected, since a much greater surface area was available in the latter system.

A UO_2 batch-leaching run in which 1-1/2-in. sheared lengths of stainless-steel-clad UO_2 were leached in a 10-liter recirculating batch of 7 M HNO_3 showed that UO_2 dissolution was complete in only 1.3 hr. No effort was made to air sparge the UO_2 fines that escaped the perforated basket and collected in the bottom of the equipment.

Solvent Extraction. — The addition of fluoride to a nitric acid solution of protactinium greatly decreased the extraction of the protactinium with tributyl phosphate. The distribution coefficient for the extraction of protactinium tracer from 5 M HNO_3 by 30% TBP was 4.0 ± 0.2, with (0.1 ± 0.6)% unextractable. The addition of 0.001, 0.01, and 0.1 M HF decreased the distribution coefficient to 3.3 ± 0.1, 0.45 ± 0.45, and 0.003 ± 0.0004 respectively; the corresponding amounts of unextractable protactinium were 1.3 ± 0.7, 36 ± 74, and 63 ± 14% of the total protactinium originally present.

Countercurrent batch-extraction experiments with protactinium tracer resulted in the simultaneous extraction of all the uranium and thorium and about 90% of the protactinium with 30% TBP in n-dodecane from a system with only nitric acid as the salting agent. Decontamination factors were 7, 50, and 250, respectively, from niobium, ruthenium, and rare-earth fission products. There was no separation from zirconium.

Protactinium Adsorption. — The distribution coefficient, γ, for the adsorption of tracer Pa^{233} by unfired Vycor from solutions containing 4 to 100 g of thorium per liter and 0.5 to 12 M HNO_3 varied directly with the nitric acid concentration. For adsorption from solutions containing 4 g of thorium per liter and X M HNO_3, $\gamma = (388 \pm 10)X + 260 \pm 75$; at 50 g of thorium per liter, $\gamma = (318 \pm 15)X + 342 \pm 106$; and at 100 g of thorium per liter, $\gamma = (259 \pm 5)X + 233 \pm 36$. The amount of unadsorbable protactinium ranged from a maximum of about 4% in 1 M HNO_3 to a minimum of about 0.6% in 12 M HNO_3; from 6 to 12 M HNO_3, the percentage of unadsorbable protactinium increased with increasing thorium concentration but varied only from 1.2 to 0.6%.

Chemical Analysis of Advanced Reactor Fuels. — Studies were continued on the controlled-potential coulometric titration of uranium(VI) at the microgram level. Analyses were performed with 2 ml of mercury as the cathode and 2 ml of sample. In order to minimize the tendency for the reaction $2M + O_2 + 2H_2SO_4 \rightleftharpoons 2MSO_4 + 2H_2O$ to occur, where M represents mercury or impurities in the mercury, the sample was purged with argon for 5 min before the mercury was added to the cell. This practice consistently yielded lower readout potentials for the prereduction step.

The results tabulated below were obtained by prereducing the sample at 0 mv vs the S.C.E. and reducing at −250 mv vs the S.C.E. The cutoff was made at a background current of 0.5 μa. Corrections were made for this background and also for a blank equivalent to 0.25 μg of uranium. The titration time was about 10 min for the 48-μg sample and about 6 min for the 2-μg sample. This does not include the time for purging the solution with argon or for prereducing the sample.

Titration of Microgram Quantities of Uranium(VI)

Number of Determinations	Uranium(VI) (μg)		Relative Error (%)	Relative Standard Deviation (%)
	Taken	Found		
11	48.32	48.34	< +0.1	0.2
10	2.48	2.47	< −0.5	0.8

THORIUM UTILIZATION PROGRAM

Kilorod Program. — Fuel rods containing thorium and U^{238} were fabricated to establish the success of the equipment development effort. Difficulties were encountered with the bearings and seals in the ball mill. Teflon was being abraded and introduced into the fine fraction of the oxide fuel. Oilite bearings were substituted for Teflon. Problems encountered in the operation of the pneumatic vibrator were traced to the manner in which the stationary anvil was mounted. In the course of finding the source of the trouble, the chuck system originally designed was replaced by a Swagelok coupling. Standards for the gamma scanner were developed, using lead-tin alloys, so that precise values of density could be derived from the recorder trace.

After the various difficulties had been corrected, production runs were begun, using U^{233}. Fuel rods are being produced with densities in the range 89.5 to 90.5% of theoretical. Uniformity along the axis has been excellent.

Fuel Cycle Development: Fuel Fabrication Studies. — Calculations were completed and a data package was submitted for an irradiation test of a thorium-uranium-zirconium alloy. Two capsules were inserted in the General Electric Test Reactor early in June.

Fuel Cycle Development: Chemistry of Solid Fuel Element Materials. — Urania-thoria solid solutions containing 40 and 60 mole % thoria were shown, by x-ray powder diffraction, to be a single cubic phase after extended equilibration at 1340°C in air. The 40 mole % solution reached an oxygen-to-metal ratio of 2.272, in close agreement with prior measurements.

Additional efforts to form solid solutions between U_3O_8 and ThO_2 at 900°C in air were unsuccessful and confirm the previous findings that the appearance of U_3O_8 as a second phase from a previously formed solid solution of urania and thoria is a kinetically irreversible process at 900°C.

Urania—10 mole % thoria mixtures equilibrated at 1340°C exhibited two phases: a cubic solid solution and a U_3O_{8-y} phase. The oxygen-to-metal ratio of the final mixture was found to be 2.530, in agreement with previous findings. Urania—30 mole % thoria mixtures equilibrated under identical conditions showed only the cubic solid solution with an oxygen-to-metal ratio of 2.276. This composition had been reported to show two phases under these conditions. In any event, the 30 mole % thoria at 1340°C is very close to the two-phase boundary line.

Preparation of Dense UO_2 by Precipitation Methods. — Work has continued on the preparation of larger (30-g) batches of dense UO_2 by precipitating uranium from $U(NO_3)_4$ solution by addition of NH_4OH, followed by washing, drying, and final calcination at 1100°C. A product having satisfactory characteristics has not yet been produced at this larger scale.

It appears impracticable to continue the precipitation and washing steps in a three-necked flask under an argon blanket. The slow settling of the precipitate results in increased opportunity for uranium oxidation. Therefore, use of the "instantaneous mixing device" for the precipitation and use of centrifugation in the washing step have been resumed to reduce the processing time. Further, all operations are being carried out in a plastic envelope under an argon atmosphere to reduce the possibility of air oxidation. Even with these changes the last UO_2 preparation showed qualitative evidence that sufficient oxidation had occurred to affect the final properties of the material. However, sufficient improvement in the last preparation over the previous ones was noted to indicate that the changes in procedure were in the proper direction.

Preparation of Thorium Microspheres. — Spherical beads of thorium oxide gel in the size range 100 to 1000 μ were prepared by passing a 4.5 M thoria sol through a 0.006-in.-diam orifice into a CCl_4—15 to 17 vol % isopropyl alcohol solution, which fluidized and dried the resultant droplets. The gel beads were further dried and calcined at 1000°C to give thoria spheres having high resistance to crushing and little internal porosity.

MOLTEN-SALT REACTOR PROGRAM

Molten-Salt Reactor Experiment. — Work continued on the installation of auxiliary lines and services in the drain tank and reactor cells.

The fuel system flush-salt tank was assembled on the jig and fitted into the cell. The drain tank furnaces were installed. The fuel system jig was assembled and the heat exchanger was placed on the jig. One drain tank and one steam dome were delivered to the site. Machining was resumed on the fuel and coolant pumps in the Y-12 Shops. The overall project was 73.5% complete as of June 1, 1963.

MSR Design. — Design of an overflow tank to be attached to the MSRE fuel pump was initiated. The tank will replace an overflow line from the pump bowl to the reactor drain line. The overflow line caused several maintenance problems, and flanges that were proposed for use in the line performed unsatisfactorily in tests under simulated reactor conditions.

Reactor Analysis. — A review of the MSRE biological shield was completed. Special attention was given to irregularities in the lateral shielding, and it was found that several areas will require additional localized shielding. Most of this is required to lower the dose rates in the blower house and vicinity. In every case the layout permits use of simple walls of concrete block to obtain adequate protection. With the added lateral shielding, it proved unnecessary to use boron in the thermal shield water to reduce capture gammas from the thermal shield.

The total worth of the control rods (fully inserted) was recalculated, using improved approximations and core properties appropriate for various fuel salt compositions. Values ranged from 5.6 to 7.6% $\delta k/k$, depending on the fuel composition, with the higher value in the core with fully enriched uranium and no thorium. The shim requirement is estimated to be about 3% $\delta k/k$, so that the rods give adequate control and shutdown margin in any case.

Development of Components and Systems. — The engineering test loop has been operated continuously for 2000 hr with 1.5 ft^3 of salt of MSRE fuel composition at 1200°F in contact with 1.1 ft^3 of graphite. During this period the increase in chromium concentration in samples of the salt was greater by a factor of 3 than the increase during similar operation with the flush salt. After an initial increase of 200 ppm, the change was roughly equivalent to a corresponding decrease in the concentration of iron, and the increase in chromium concentration is expected to stop after the iron is depleted.

The helium purification loop was shut down for alterations after operating for 1500 hr with the oxygen removal unit at 1000°F. Tests were run at varying inlet-oxygen concentrations; the results indicated that 1000°F is not hot enough to reduce the oxygen concentration to 1 ppm with inlet-oxygen concentrations above 50 ppm. Additional tests will be run at 1000 and 1200°F to determine the minimum temperature for satisfactory operation.

A modified cell was installed in the oxygen analyzer for evaluation, and initial tests indicate that the background reading is about 10% of the background with the previous cell. The analyzer now operates satisfactorily.

The prototype drain-tank heater has operated 2670 hr at 1300 to 1400°F without difficulty. The thermal gradient on the 8-ft vertical axis of the heater surface was only 200°F after the insulation on the top surface of the test enclosure was increased to a thickness of 8 in.

The drain-tank cooler test was terminated after 2800 cycles when a leak in the water system was indicated. Investigation revealed that two of the 1-in. cooling tubes and the concentric 1/2-in. inlet-water tubes were cracked. The cracking appears to have been caused by severe thermal stressing of the metal in tightly restricted areas. Most severe cracking was found near spacer weldments. These Inconel tubes were constructed from early preliminary drawings. A prototype cooling element of an improved design is being built for further tests.

The removable heaters with reflective insulation for 5-in. pipe have operated continuously at temperatures above 1300°F for over six months. Heat losses per linear foot of heater have increased 15 to 20% due to loss of emissivity of reflective surfaces. This increase is considered acceptable for the conditions of reactor operation.

MSRE Pump Program. — The MSRE prototype fuel pump circulated an MSRE fuel salt for 4700 hr before the test was stopped in order to modify the test stand. An attempt was made to determine the effectiveness of the spray for removal of fission gas by putting Kr^{85} in the pump bowl and measuring its concentration in the circulating fuel through the pipe wall, but the intensity of the Kr^{85} signal was too low. An attempt to measure the effect of flow rate on the concentration of gas entrained in the circulating salt was halted when the variable-frequency motor-generator set failed. The failures were found in the electrical insulation of the generator and the shaft bearings of the magnetic clutch. They were repaired and the set has been satisfactorily tested with a dummy load.

The modifications to the test facility are almost complete. They include installation of the flexible pump support, a float-type level indicator in the pump tank, and a flanged joint in the off-gas line. The rotary element of the pump was reassembled with new preloaded bearings, and measurements of the shaft deflection and the natural vibration frequency indicated that the stiffness of the shaft was increased by about 15%.

The PKP molten-salt pump was reassembled with a new lower shaft seal in which the Graphitar nosepiece was attached to the bellows assembly with a clear baking varnish. The stator and rotor of the drive motor were rewound. The pump test will be restarted after the motor is operated satisfactorily at room temperature.

Inspection of the pump having a molten-salt bearing revealed that the seizure, previously reported, resulted from the loss of two of the four fulcrum pins from the gimbals on which the bearing was mounted. The bearing had moved axially downward toward the impeller until the impeller and the shaft were jammed against the pump volute and the bearing respectively. The bearing and journal were not reusable and are being replaced. The volute was not damaged severely, and it will be reused without repairs.

Instrumentation Development. — Testing of the mercury-switch temperature scanning system on the MSRE engineering test loop was completed. There were some minor troubles, such as failure of a dc amplifier and erratic behavior of an oscilloscope; however, performance was satisfactory after these troubles were corrected, and the system has been accepted for use on the MSRE.

Two differential transformers fabricated for use on the ball-float level transmitter installation on the MSRE pump test loop failed during preliminary testing. The failure was caused by sulfur embrittlement of the nickel wire in the transformer. The sulfur contamination is thought to have resulted from the use of a lubricating oil in fabrication of the lava sleeves in the transformer. Procedures have been revised to prevent reoccurrence of such failures. The component parts were baked at high temperature. The transformers were then reassembled, and they performed satisfactorily. Fabrication and installation of the ball-float level transmitter for the test loop is complete; testing will commence during the next period of loop operation.

Testing of mineral-insulated Inconel-sheathed thermocouples of the type to be used in the MSRE is continuing at a reduced level of effort. The drift of six thermocouples being furnace tested in air continues to be less than 2°F, after 18 months of operation at 1200°F. Testing of thermocouples under field conditions on the MSRE engineering and pump test loops has been temporarily discontinued due to shutdown of the loops. All MSRE-type thermocouples installed on these loops were intact and performed satisfactorily when the loops were shut down. Efforts to develop radiation-resistant cold-end seals for mineral-insulated thermocouples which will be installed in MSRE reactor and drain cells were continued. Excellent seals have been obtained with Physical Science's 0900 glaze compound. Techniques for quantity production of these seals under field conditions are being developed. Initially 40 hr was required to produce each seal. Present techniques show promise that this time may be reduced to 30 min or less per seal.

Two-channel and single-channel stereo TV systems are being evaluated for use in remote maintenance on the MSRE. Both systems produce a definite three-dimensional image when correctly adjusted; however, both have disadvantages. The single-channel system has the inherent disadvantage that the horizontal field of view is restricted and more light is required to obtain a readable picture. The two-channel system requires more equipment and is therefore more subject to failure. Also, the two-channel system being tested appears to be more difficult to adjust and may produce more operator fatigue than the single-channel system.

NUCLEAR SAFETY

Release of Fission Products on Out-of-Pile Melting of Reactor Fuels. — A radioactive mirror-like deposit of material vaporized from highly irradiated Al-U alloy in helium or in a vacuum was found to be almost pure zinc containing radioactive cesium, iodine, and tellurium (the last in trace amounts). In these experiments, very little radioiodine passed through the absolute filters; this is in contrast to the behavior in air, where large fractions of radioiodine pass the filters. Similar but nonradioactive deposits were obtained from unirradiated fuel samples, showing that zinc was a minor component of the fuel material. It is reasonable to suspect that the vaporization and condensation of

zinc or of Zn-Al alloys might be connected with the corresponding decrease in iodine release. However, the problem has not been definitely solved and is being examined further with $I^{127,129}$ tagged with I^{131}.

Attenuation of Fission Product Escape by Intrinsic Processes. — Certain intrinsic processes which tend to limit or attenuate the escape of fission products have not been fully utilized in reducing the assumed consequences of a nuclear reactor accident. Consequently, theoretical analyses of two such processes have been performed to determine whether the attenuation effects are of sufficient magnitude to warrant further investigation or to warrant possible consideration in containment system design.

One analysis was concerned with the decontamination resulting from diffusional deposition of radioparticulates in containment vessel leaks. The calculated decontamination factors were found to be extremely sensitive to both particle size and leak size, the higher decontamination factors being associated with the smaller particles and with the smaller leaks. Considering the single example of a containment vessel with a 1-in. wall and air escaping through leaks under a pressure drop of 7 psig, decontamination factors (ratio of entering and exit concentrations) of 100 or higher were calculated for particles $\leq 0.3\ \mu$ in diameter when the nominal leak dimension was no greater than $5\ \mu$. For larger particles or leaks, the results were much less favorable. The nominal dimension is the diameter of a cylindrical leak or the smallest dimension of a rectangular leak. The overall results indicated that this effect is well worth careful attention.

The other analysis involved particle agglomeration and its effect on the penetration of filter systems in dual containment systems by fission products carried by particulate matter. A simplified model was analyzed in which the particulate matter was assumed to be pure iodine, and all particulates having diameters of less than $0.3\ \mu$ were assumed to penetrate the filters. It was shown that agglomeration would occur rapidly enough that the particles, even though initially much smaller, would reach a $0.3\text{-}\mu$ size in a short time (< 20 min for typical aqueous reactor systems). This time is directly proportional to the containment volume and inversely proportional to the power level. Assuming that the filter efficiency of the secondary-containment sweep gas filtration system is zero for particles less than $0.3\ \mu$ in diameter, the fraction of the core activity released to the atmosphere during that time was calculated for some conditions of interest. For one reactor system treated, less than 10^{-5} of the iodine available would be released. It was shown that conditions existing during a reactor accident could not be as pessimistic as those of this model.

Characterization and Control of Accident-Released Fission Products. — A method for characterizing radioactive aerosols has been developed. The method interprets the distribution of Zn^{65} particles in a series of uniform fiber filters in terms of the three main mechanisms of filtration, that is, diffusion, interception, and inertial impaction. It was previously reported that observed collection efficiencies were qualitatively in accord with the theory over the velocity range employed. Quantitative theoretical analysis of the data corresponding to the diffusion regime

has yielded particle sizes consistent with electron photomicrographs and with estimates based on the amount of activity in the aerosol and the number of particles measured by a condensation nuclei counter.

Nuclear Safety Pilot Plant. — The Nuclear Safety Pilot Plant furnace is completely fabricated and is in shakedown. Design of process equipment and piping is complete, and its installation in the process shell is 50% complete. Electrical drawings are complete and are being reviewed. Instrumentation design is 40% complete. Design of remote-handling tools, shielding, and access to the cell is 50% complete.

Four 8-in. butterfly valves used to control air flow into the model containment vessel did not meet specifications and were returned to the manufacturer for correction; some delay may result.

A rough draft of the hazards report is completed. Experiment planning is under way.

Reactor Containment Handbook. — Approximately 70% of the material for the Reactor Containment Handbook is drafted, including: Chapter 2, "Codes, Criteria, and Regulations"; Chapter 4, "Energy Sources"; Chapter 7, "Containment Proof Tests"; Chapter 9, "Design Details"; Chapter 10, "Containment Accessories"; and Chapter 11, "Economics." Chapters 2 and 4 are being reviewed by ORNL personnel and selected external reviewers. Chapters 9, 10, and 11, written by the Bechtel Corporation, have been reviewed and returned to Bechtel with comments to be incorporated in the final draft. Chapter 7 is being put in final form by Bechtel.

Chapter 3, "Radioactivity — Generation and Transport," Chapter 5, "Analytical Techniques," and Chapter 8, "Specific Containment Systems," are essentially complete, with only minor revisions necessary before their drafts are submitted for review.

For Chapter 5, Stanford Research Institute submitted a section on "Penetrability of Shells," and the Armour Research Foundation submitted one on the "Analysis of the Response of Containment Vessels to Internal Loading in the Range Approaching Failure."

Nuclear Safety Information Center. — A Nuclear Safety Information Center (ORNL-3451, p 14) has been established at ORNL to serve as a focal point for nuclear safety information generated throughout the world. Considerable Center effort is being directed to the collection and storage of this information. Ten special study assignments have been received; one of these has been completed and a report has been issued.

WASTE DISPOSAL DEVELOPMENT

Clinch River Study. — A survey of the longitudinal distribution of radioactivity in the bottom sediment of the Clinch River was conducted, during the last week in April, to determine whether the distribution observed during the summer of 1962 had changed during the fall and winter. Observations with "flounders" (G-M tube arrays) were made and samples were collected at sites marked for the annual surveys by the Applied Health Physics Section. Results indicate that the gross-gamma activity

is about the same near Clinch River miles (CRM's) 5 to 7 and 18 to 20, but is about 40% lower near CRM's 14 to 16.

During May, "flounder" observations were made in several sloughs along the banks of the Clinch River. These sloughs were in the reach from CRM 2.7 to CRM 17.0. The gross-gamma activity in situ was higher than background levels in all sloughs. The maximum observed activity in the sloughs was about 50% of the maximum observed in the mainstream of the Clinch River. In most of the sloughs investigated the activity was higher in the deep water.

Mineral Exchange Studies. — Sodium chloride solutions made with tritiated water and tagged with Cl^{36} were passed through hydrobiotite columns pre-equilibrated with an untagged solution of the same composition. In all cases the breakthrough of Cl^{36} precedes that of H^3, the Cl^{36} breakthrough coming relatively earlier with decreasing concentrations of NaCl. The behavior can be explained on the basis of the electrochemical nature of the hydrobiotite. At the external surface of the particle the chloride ion is repelled by the negatively charged surface, while the uncharged tritiated water is not. The effective volume of chloride exclusion is related to the thickness of the electric double layer, which is dependent, in turn, on the ionic strength of the solution. Within the basal spacings of the vermiculite, which are invaded by water, the degree of chloride exclusion can be calculated on the basis of a Donnan equilibrium. Neither chloride ions nor tritium can invade the collapsed biotite lattices of the hydrobiotite.

Magnesium-saturated natural vermiculite was treated with (1) a 1.5 symmetry concentration of cesium nitrate in contact for 24 hr, (2) a 1.5 symmetry concentration of cesium in contact for 212 hr, and (3) a 35 symmetry concentration of cesium for a total of 2.5 hr in a manner similar to the procedure recommended by Jackson (Soil Chemical Analysis, Prentice-Hall, 1958). The vermiculites were washed free of excess cesium and air-dried. X-ray diffraction tests revealed an expanded c spacing for the sample from treatment No. 3, indicating that it had not sorbed as much cesium as those in treatments Nos. 1 and 2. These results cast doubt on the applicability of the normal technique for saturating vermiculite with cesium. On contacting these vermiculite samples in 0.1 N $NaNO_3$ solutions containing different concentrations of cesium, it was found that the sample from treatment No. 3 sorbed more cesium than the natural vermiculite when the cesium concentration was extremely low. Since lattice collapse of vermiculite enhances cesium sorption in the trace concentration levels, it is believed that treatment No. 3 resulted in collapse of the edges of the vermiculite, followed by a slow diffusion of some of the edge cesium into interlayer positions. Diffusion of the cesium then permitted further sorption of cesium in the trace concentration levels.

Disposal in Deep Wells. — The injection well and the observation well have been drilled, logged, cased, and cemented. The monitoring well is being drilled. The logs of the injection and observation wells show that the injection well is inclined about 20 ft southeast and that the observation well is virtually vertical. The logs also show that the identifiable geologic formations occur at about the same depth in each well; the expected 20° upward slope toward the observation well was not found.

The injection well was slotted, and the formation fractured at a wellhead pressure of 3700 psi. Approximately 2000 gal of water was pumped into the fracture and allowed to flow back slowly; 950 gal had been recovered at the end of 70 hr. The flow at this time was down to 1-1/2 gal/hr. An injection of 50,000 gal of water was then made into the same fracture. This water will be allowed to flow back over a period of several weeks, and a careful record of the volume that flows back will be kept. This figure will be compared with a similar figure obtained from a later injection of 50,000 gal of a mixture of water and a fluid loss additive. This comparison will provide some information as to whether and by what mechanism water is taken up by the shale formation.

A preliminary cost evaluation indicated that for future routine waste disposal by hydrofracture at ORNL, the cost of the mix components will be a significant contributor to total costs — as much as 75% of the total if other costs are minimized. Another cost that may be quite significant is the amortized cost of the well. If it is found that only a relatively few grout sheets can be injected through a single well, the cost of the well becomes a quite significant factor. This cost can be reduced considerably, however, if the volume of each injection is made large, thereby reducing the injection frequency and extending the effective well life.

Low-Activity Wastes: Scavenging-Precipitation—Ion Exchange. — The phosphate concentration of ORNL low-level waste was successfully reduced from ≧3 to 0.2 to 0.5 ppm during treatment of more than 8666 volumes of waste with one bed volume of alumina. This treatment permitted a more quantitative precipitation of calcium carbonate in the scavenging-precipitation clarifier (reduction in hardness of clarified product from 70 to less than 10 ppm). A significant but unknown fraction of the ruthenium was also adsorbed on the alumina.

Low-Activity Wastes: Foam Separation. — Initial tests of a two-step process for decontamination of low-level waste, consisting of sludge column precipitation of calcium carbonate and magnesium hydroxide followed by a countercurrent foam column for strontium removal, were performed with Sr^{85}-spiked ORNL tap water and ORNL low-level process waste water in laboratory-scale equipment. With tap water as feed, strontium decontamination factors in the foam column and in the overall process were 2 to >400 and 320 to >15,000, respectively, depending on operating conditions. With process waste water, foam column and overall strontium decontamination factors were 11 to 16 and about 250, respectively, which are close to those obtained with tap water under the same operating conditions. Also tested with simulated and actual ORNL low-level process waste was a process which involves precipitation of calcium as orthophosphate in a small mixer and then feeding solution and solids to a foam column, where flotation and foam separation occur. Strontium decontamination factors with simulated and actual ORNL waste water were, respectively, >1000 and 200 to 300.

Studies were performed on the biological degradation in ORNL process waste water of the alkylbenzenesulfonate already present in the waste water (primarily from Fab, a household detergent), of the alkylbenzenesulfonate in Fab, and of the branched-chain sodium dodecylbenzenesulfonate

of Trepolate F-95. After 2 to 3 weeks the respective amounts of degradation from existing bacteria were 20, 25 to 30, and 60 to 65%.

A 24-in.-diam foam column was operated with liquid feed introduced through tubes on a 5-in. triangular spacing; channeling was observed for 10 to 30 in. below the feed tubes. Twenty-mesh screens across the column cross sections were not effective liquid distributors, since they accumulated liquid, which dumped through periodically. Good drainage of foam was obtained in a horizontal drainage section. The experimentally measured foam densities agreed well with a foam drainage model. Air-operated sonic whistles condensed 0.05 to 0.10 ft^3 of foam per standard cubic foot of operating air. The experimentally observed concentrations for a three-stage surfactant recovery system agreed well with concentrations calculated by using a distribution coefficient of 7×10^{-4} $(mole/cm^2)/(mole/cc)$ for the dodecylbenzenesulfonate.

High-Activity Waste: Pot Calcination. — Three continuous evaporator-pot calcination tests using simulated Purex formaldehyde-treated waste, 1965 (FTW-65 waste) were completed. The average feed rates varied from 31 to 49 liters/hr, and the maximum hourly rates varied from 125 to 190 liters/hr. The average feed rate is a function of the concentration in the evaporator and the degree of filling of the calciner pot. The calcined solids had densities of 1.23 to 1.30. The residual nitrate composition for the calcined solids varied from 100 to 400 ppm and decreased with increase of the length of time that the calcined solid was above 600°C. The sulfate retention in the calciner was high; only 0.3 to 2% of the sulfate was lost. This loss is attributed to entrainment, since similar values were obtained for nonvolatile ions such as those of iron and sodium. A catch tank, tested as a method of isolating mercury, collected only 27% of the mercury. The amount of mercury caught was proportional to the amount in the last fraction of the calciner condensate. A ruthenium decontamination factor of 3×10^3 (using nonradioactive ruthenium) was obtained in test R-72 across the continuous evaporator; an impingement plate de-entrainer was used.

High-Activity Waste: Fixation in Glass. — Simulated Hanford FTW-65 waste was evaporated to 1/4 of original volume without precipitation of solids. Experimental phosphate glassy melts were made by addition of phosphate, lithium, sodium, and calcium to FTW-65 waste. Generally, these melts were too soluble in water or lost SO_3 readily, but a few appeared to be insoluble and were relatively stable at high temperatures. Substitution of lithium for part of the added sodium resulted in solids that melted from 50 to 100°C lower than the corresponding sodium compounds and that were less soluble in water. A semicontinuous, semi-engineering-scale experiment with the lithium glass was successful, although excessive solids entrainment caused an operating problem. The product contained 45% waste oxides and represented a volume reduction factor of 30.3. Sulfate loss was less than 2%, and 24% of the ruthenium was volatilized. An electrical resistance probe for sensing levels of solution, calcine, and melt in the pot performed satisfactorily.

Imposition of a small cathodic current decreased the time necessary for removal of more than 99% of the mercury in a copper column from FTW-65

waste solution from about 25 min to 12 min or less. More current was required than the minimum to remove the mercury in order to prevent dissolution of excess copper.

Exposure of type 304L stainless steel specimens at 300°C in bedded salt (NaCl) containing naturally occurring impurities showed corrosion rates of about 0.03 mil/month. Type 304 stainless steel and titanium 45A, both coupled and uncoupled to each other, showed low rates for exposure in refluxing FTW-65 waste solution and vapors; the maximum rate was on the coupled 304L and amounted to 0.3 mil/month, with no local attack.

The thermal conductivity characteristics of powdered MgO, Al_2O_3, and ZrO_2 were investigated as a function of void space and temperature. Over the temperature range studied (200 to 1500°F), the thermal conductivity increased quadratically with the temperature.

Disposal in Natural Salt Formations. – Heating of the 8-ft-wide by 10-ft-deep by 2-ft-high cavity in salt at approximately 165°C has now continued for 90 days. The cavity is closing at a rate two orders of magnitude higher than that observed before heating. For a cavity of these dimensions, approximately 12 years would be required for complete closure. However, these results are for an isolated cavity and do not represent the behavior to be expected from a series of adjacent openings such as those seen in a mine operation.

The irradiation of sodium chloride with gamma rays produces an oxidizing reagent within the crystals which can be detected upon dissolving the salt in water. The G value for this process is 1.4×10^{-2} at 5×10^5 rads and decreases linearly with dose to give 1.2×10^{-3} at 2.1×10^8 rads. No release of oxidizer to the atmosphere has yet been detected at these levels.

Work is under way on the design of a demonstration of disposal of high-level radioactive waste solids to be carried out in the Lyons, Kansas, mine of the Carey Salt Company. Fourteen irradiated Engineering Test Reactor fuel assemblies will serve as a source of radiation in lieu of actual solidified wastes. The assemblies will be placed in a circular array of holes in the floor, with one can in the center and the other six cans located peripherally, spaced 5 ft on centers. The test will last 2 years, after which the assemblies will be returned to the National Reactor Testing Station for recovery of the unfissioned fuel.

An identical array with electrical heaters instead of fuel assemblies will be operated as a control to determine the effects of radiation and heat on the salt characteristics.

In addition to the radioactive and control arrays, a rib-pillar located between the two arrays will be heated by electrical heaters around its base to produce information on salt flow characteristics at elevated temperatures.

The engineering and scientific objectives are: (1) demonstration of waste-handling equipment and techniques; (2) determination of possible gross effects of radiation on hole closure, floor uplift, salt shattering temperature, etc., in an area where salt temperatures are in the range 100 to 200°C; (3) determination of possible radiolytic production of chlorine; and (4) obtaining information, on creep and plastic flow of salt at elevated temperatures, which can be used later to design an actual disposal facility.

PHYSICAL RESEARCH PROGRAM

PHYSICS AND MATHEMATICS RESEARCH

High-Energy Electronuclear Machines: Superconducting Magnet Studies. — A superconducting magnet built in nine separate sections was completed and is being used to study the so-called "degradation effect" in superconducting magnets. The nine sections are assembled, as a three-layer stack of three concentric solenoids, to provide a usable inner volume 2.1 in. in inside diameter and 5.1 in. long. All coils are bifilar wound of Nb-Zr wire and copper wire and potted in epoxy (the fabrication method developed at ORNL for study magnets); the copper provides a secondary circuit which protects the coil during a transition.

The nine sections of this magnet are being test operated with various series and parallel arrangements and at various current and self-field configurations in an attempt to determine the critical factors which limit the performance of the magnet to much less than should be expected from tests of straight short lengths of the same superconducting wire. Thus far, these magnet tests have demonstrated that the transition from superconducting to normal is not initiated in an inner section, which is in the highest magnetic field. For example, when all coils in series are at just below their critical current (14 amp, or 23.5 kilogauss), the current in the smallest inner coil can be increased to 25 amp before the transition occurs. Preliminary attempts to maximize the central field by using a nonuniform current distribution in the windings have thus far produced a maximum field of 31.7 kilogauss, whereas when all coils in series carry 14 amp the central field is only 23.5 kilogauss.

Medium-Energy Nuclear Physics: Heavy Particles. — During a two-week run on the Tandem Van de Graaff, excitation functions and angular distributions were obtained for the reaction $C^{12} + O^{16} \rightarrow Mg^{24} + \alpha$. Measurements were made at 19 Mev and at 100-kev steps in the energy region from 29.6 to 32.7 Mev (lab). Angular distributions were taken every 2.5° with an array of eight solid-state counters. At high energies the angular distributions display a very pronounced $\ell = 8$ pattern.

A study of nuclear reactions induced by N^{14} ions on Li^6 and Li^7 targets was initiated. A search is also in progress for a possible alpha activity of Eu^{148} produced by 22-Mev protons in the 86-Inch Cyclotron.

Medium-Energy Nuclear Physics: The 86-Inch Cyclotron. — A new 1.2-hr half-life was observed in a separated source of Ir^{190} prepared for the internal-conversion electron spectrograph. The Ir^{190} was produced by irradiating enriched Os^{190} with 10-Mev protons. The characteristic radiation is of 26-kev energy, and the differences in energy of the $L_I L_{II} L_{III}$ and $M_I M_{III} M_V$ conversion lines are uniquely those of iridium. The intensity data for the transition are consistent with an M3 multipole order assignment. It is proposed that the M3 transition proceeds between the 26.3-kev state and the 12-day ground state in Ir^{190}. As Ir^{190} is known also to have a 3.2-hr isomeric state, this constitutes a rare case of triple isomerism.

Cyclotron Operations: The Isochronous Cyclotron. — Test operation of the ORIC for the development of the beam deflection system and for a better understanding and control of the conditions at the cyclotron center was continued. The "hot-wire" technique was used for determining the configuration of the beam at the deflection radius; the septum and the deflector were then shaped to fit. About 20% of the 75-Mev alpha-particle beam arriving at the entrance to the deflector was deflected onto an internal probe. When tests were made to determine the effective beam center and the amplitude of the betatron oscillation, it became apparent that a dependable means of controlling the beam geometry at the center of the machine was desirable. A 120° "spectrometer" slit for defining the beam at the source was installed as part of the accelerating electrode. With a narrow probe the first ten separate turns, or orbits, of the beam have been distinguished. Tests are being made to determine the beam center and the betatron oscillation of the beam as the accelerating slit is moved in a controlled manner around the geometric center and when the harmonic coils are energized in various combinations.

High-Voltage Experimental Program: The $O^{18}(\alpha,p)F^{21}$ Reaction and the Beta Decay of F^{21}. — The reaction $O^{18}(\alpha,p)F^{21}$, with alpha-particle bombarding energies of 8 to 10 Mev, has been studied by observing the betas following F^{21} decay. Doubly ionized helium ions were produced in the ORNL 5.5-Mv Van de Graaff, and the beta rays were detected in approximately 2π geometry by a 1.25-in.-diam, 1-in.-long anthracene crystal in conjunction with an electrostatic beam deflector and electronic timer. Thin targets were prepared by anodizing tantalum blanks with water enriched to 97% in O^{18}. The relative cross section for the $O^{18}(\alpha,p)F^{21}$ reaction, obtained as a function of energy by measuring the yield of betas with energy greater than 1 Mev, shows a complex structure corresponding to excited states in Ne^{22} between 16.21 and 17.85 Mev. The resonance structure observed is, in general, wider than the experimental resolution and has broad peaks at incident-particle laboratory energies of about 8.36, 8.95, 9.18, 9.45, and 9.80 Mev. An average half-life value of 4.35 ± 0.03 sec for F^{21} beta decay was determined from several measurements with varying pulse-height biases.

High-Voltage Experimental Program: Total Neutron Cross Sections in Isotopes of Zr, Fe, Cr, and Ca. — Total cross-section measurements continue to be made with the pulsed Van de Graaff on separated isotopes for which the amount of material is small (~1/20 mole) and the level spacing is 100 v or more. Measurements have been made with 1 to 2% energy resolution in the region 2 to 30 kev on the following nuclides: Zr^{90}, Zr^{91}, Zr^{92}, Zr^{94}, Zr^{96}, Ca^{43}, Cr^{53}, and Fe^{57}. Following are the principal resonances observed in kev units of energy:

Zr^{90}: 3.8, 7.4, 13.5, 17.4, 18.0, 26.5, 41.3

Zr^{91}: 6.7

Zr^{92}: 6.7, 15.4, 17.1, 22.7

Zr^{94}: 5.7, 6.8, 10.7, 12.8, 15.8, 17.8, 19.5

Zr^{96}: 2.62, 2.82, 3.7, 4.2, 5.25, 6.7, 29.3

Ca^{43}: 4.3, 5.2, 8.8, 13.8, 19.2, 22.5, 26.6, 31.4, 37.0

Cr^{53}: 19.3, 26.3, 34.7, 39.0, 28.8, 37.0

No levels other than those already known at 3.90 and 6.42 kev could be detected up to 30 kev with the Fe^{57} sample employed. Other levels, either small or overlapping so as to make energy assignment uncertain, were observed but not listed. Interpretation of the data is as yet incomplete.

Stable Isotopes Development. — The conversion of two beta calutrons to 255° isotope separators was completed. Water-cooled liners for design testing are being constructed of copper. Permanent water-cooled liners will be fabricated from stainless steel. Some baffling was added to produce maximum isolation of the receiver region from the source. This is expected to reduce receiver contamination by thermal vapors and sideband beams from the source.

Initial runs with argon beams indicated that the source was operating normally and that no serious electron-drain problems existed. In tantalum separations using Ta_2O_5 internally chlorinated with CCl_4, assay results showed enrichment factors for Ta^{180} to be double those of previous beta calutron series.

Receiver faceplate erosion patterns cut by the beam during satisfactory operations show that the corrected set of shims is focusing the ion beam in agreement with orbits calculated from magnetic-field measurement data. Attempts will be made to obtain Cd^{111}, Cd^{113}, and Cd^{114} of > 99% isotopic purity in a one-pass separation.

Ion receiver experiments are continuing, using the Ca^{46} ion beam striking on graphite, copper, and crystalline copper with the 110 or 112 crystal face perpendicular to the incident ion beam. A periodically rotating holder, on which four different targets can be mounted, was constructed. Its use in the comparison of receiver materials provides data that are independent of operating conditions, which vary from run to run.

A sample of Ca^{46} collected on a 110 copper crystal assayed 50.0% Ca^{46}, 2.51% Ca^{44}. The highest isotopic purity previously achieved in a normal receiver was 43.4% Ca^{46}, 4.1% Ca^{44}.

An attempt is being made to establish a correlation between arc chamber geometry and the extent of source modulation transmitted to the primary beam in the calutron. This type of modulation generally is detrimental to the separation process and difficult to eliminate.

Movable sections placed within the arc chamber permitted observation of beam response to various arc chamber dimensions. Material reduction in the amplitude of modulations in an ion source was demonstrated through the use of specifically dimensioned arc chambers.

A straight image or focal line at the calutron receiver would simplify the construction of collectors. Magnetic-field measurements were made on a set of shims, which might produce such an image. These shims consist of two soft iron plates, each 1/2 in. thick and ~5 in. long, shaped like a football and fastened to the calutron tank walls at the 90° position of the beam.

Experiments with Ca^+ beams show that the magnetic field produced by the shims, coupled with the strong azimuthal variations in the base magnetic field due to irregularities in the tank walls, gives very nearly a straight image. Orbits calculated from the magnetic-field measurements indicate a lack of perfect focusing, even in the magnetic midplane. The assumption is made that if the "football" shim shape were changed to focus the beam in the midplane, the image would remain straight and serious

aberrations would not be present in the image outside the midplane. If the assumption becomes reality, these shims could replace the present linear shims without other modification.

Special Separations. – Estimated weights (based on monitored ion currents) of separated plutonium isotopes and ranges of enrichment achieved are as follows:

Isotope	Estimated Weight (g)		Purity Range (%)
	For Period	Cumulative	
Pu^{239}	14.7440	49.2037	a
Pu^{240}	26.8696	85.9896	98.17–99.30
Pu^{241}	6.7880	21.7891	89.99–95.88
Pu^{242}	3.4823	10.9482	86.23

[a]No assays for period; previous assays 94.88–94.96%.

CHEMISTRY RESEARCH

Thorium Oxide Studies. – Stable sols of crystalline (calcined at 500 to 850°C) thorium oxide suspended in dilute nitric acid were studied by photomicrography, x-ray, and a variety of physical chemistry methods. The colloidal particles of these sols appear to have three levels of organization: single crystallites; tightly bonded low-number multiples of crystallites; and elongated, weakly associated agglomerates of crystallites. The single crystallites are nearly centrosymmetric and range in size from 70 to 250 A, depending on calcination temperature. Tightly bonded multiples of crystallites are produced by the higher calcination temperatures and have axial ratios near 2. High-dilution viscometry shows the hydrodynamically effective particles to have axial ratios of 6.5 to 8.5 at pH 2.00, eccentricity decreasing with particle concentration at constant pH. This behavior, as well as exaggerated non-Newtonian flow properties at solids volume fractions exceeding 0.05, is believed to be associated with reversible agglomerate structures rather than with the crystallites themselves.

Electrophoretic measurements for thoria in pH 2.00 nitric acid showed a three-day aging effect and a high value of the apparent zeta potential, which increased to 90 mv over the three-day period. This is higher than results found in published data. However, first-order electroviscous effects are expected to be negligible at the high particle eccentricity and high ionic strength used in the present experiments.

Protactinium Chemistry. – Additional measurements of protactinium distribution between 5 N sulfuric acid and 0.03 N trilauryl amine in diethylbenzene confirmed that there is a change in distribution behavior as

the protactinium concentration is increased. For aqueous-phase Pa^{231} concentrations less than approximately 2×10^5 alpha counts min^{-1} ml^{-1} the concentration in the organic solvent was proportional to the 1.3 power of the aqueous-phase concentration. At higher concentrations the protactinium concentration in the solvent was proportional to the 3.2 power of the aqueous-phase concentration. This behavior is similar to that observed in concentration dependence between 5 N sulfuric acid and Dowex 1 resin, except that the change was even larger in the case of the resin.

These observations are generally consistent with a reversible polymerization of protactinium in one or both phases, with both the polymer and the simpler species being extracted, but the polymer being more extractable. The polymer is small, perhaps a hexamer, and is formed reversibly with respect to changes in the concentration of both protactinium and sulfuric acid. Polymer formation is inhibited by sulfuric acid, with the dependence on acid concentration being second power.

Equipment Decontamination. — A solution of 0.5 M oxalic acid with 0.1 M fluoride and 0.2 M hydrogen peroxide at 95°C, which was an effective decontaminant for stainless steels contaminated in helium at 500 to 650°C, produced no stress corrosion cracking on U-bends of types 304, 316, 321, and 347 stainless steels in 22.5 hr. The solution strength was restored every hour by the addition of $H_2C_2O_4$ and H_2O_2.

Several samples of contaminated stainless steel from loop 2 of the ORR were successfully decontaminated. Barium-140 was a prominent contaminant of all samples. Those samples which had been exposed to contaminated helium at 540 to 600°C were also contaminated with Ru, Zr, Nb, and Ce. One such sample had a decontamination factor (DF) of only 7 in 3 hr at 95°C in the noncorrosive pH 4 solution of oxalate-citrate-peroxide. In three additional hours at 95°C in a more corrosive solution of 0.5 M $H_2C_2O_4$–0.1 M NaF–0.2 M H_2O_2, the total DF was 54. The DF was 84 on another sample, contaminated chiefly with Ba^{140} at 270°C, in 1 hr at 95°C in the pH 4 solution. In 3 N HNO_3 in 1 hr at 95°C the DF was 128. A graphite sample exposed to helium at 600°C resisted decontamination, probably due to its porosity (DF = 1.6 in 1 hr at 95°C in pH 4 solution).

The decontamination of Zircaloy-2 should be very rapid and complete in solutions of oxalic acid containing fluoride and hydrogen peroxide. The uniform dissolution rate in 0.5 M $H_2C_2O_4$–0.1 M NaF–0.2 M H_2O_2 at 25°C was 3.4 mils/hr, without pitting or gas formation. With 0.05 M $H_2C_2O_4$–0.05 M NaF–0.07 M H_2O_2, the rate was 1.1 mils/hr. The Zircaloy surfaces were very bright and clean.

Pyrolysis and Radiolysis of Organic Liquids. — Biphenyl was pyrolyzed in the liquid phase at 425°C for extended periods (up to 48 hr but still with less than 0.5% decomposition). The yields of both benzene and hydrogen showed, to a first approximation, a linear relationship with time. In continuation of the studies of surface effect (ORNL-3451, p 23), the quantity of hydrogen, as well as methane, produced during pyrolysis for 12 hr at 425°C was measured. Much larger amounts of both gases were formed in the presence of stainless steel wool and stainless steel wool plus borosilicate glass wool than were formed when quartz wool and borosilicate were used alone, although the surface area of added material was the same in all cases. However, there were only minor differences in the small benzene yield; the mixture of stainless steel wool and borosilicate glass wool produced the least benzene.

High-Temperature Aqueous Solution Chemistry. — A new optical cell has been developed in which aqueous solutions have been subjected to hydrostatic pressures as high as 30,000 psi at temperatures up to 425°C. Phase and color changes could be observed visually. Previously, the upper limit of pressure for such observations had been 4500 psi (ORNL-3331, p 24). The new cell has been used to determine the effect of pressure on the liquid-liquid immiscibility of UO_2SO_4-H_2O solutions up to the extremes of temperature. Values of ($\Delta t/\Delta p$) were found to be essentially independent of both pressure and concentration for UO_2SO_4 solutions varying from 0.071 to 4.54 m, the average value being +0.074°/bar. Thus, by applying 20,000 psi to a UO_2SO_4 solution showing immiscibility at 300°, this temperature could be raised to 400°. It is believed that the effect may be directly related to the dielectric constant of the solvent, which is strongly dependent on the density and, thus, the pressure.

Temperatures for the appearance of liquid-liquid immiscibility and of critical phenomena (where liquid ≡ vapor) were determined in the condensed system UO_3-Li_2O-SO_3-D_2O at concentrations of SO_3 from 0.05 to 1.0 m, at molal ratios of total metallic oxides to SO_3 from 0 to 1.0, and at molal ratios of UO_3:Li_2O of 3:1, 1:1, and 1:3. Based on these data and on information obtained previously on the systems UO_3-SO_3-D_2O and Li_2SO_4-D_2SO_4-D_2O, isotherms were drawn to describe boundary limits of liquid-liquid immiscibility, critical phenomena, and appearance of solids in the system UO_3-Li_2O-SO_3-D_2O.

The study of the electrical conductance of K_2SO_4 in H_2O solution at several concentrations, at temperatures from 25 to 800°C, and at pressures from 1 to 60,000 psi was completed (ORNL-3331, p 25). From these results, K_2SO_4 was found to behave as a relatively strong electrolyte at solution densities between 0.8 and 1.0 g/cm^3. As the density decreased to low values, extensive association and hydrolysis appeared to occur. By the use of existing theories of conductivity, developed originally for use at 25 to 100°C, values of Λ_0 were calculated. With these values, and with further use of existing theory, thermodynamic dissociation constants for the dissociation $KSO_4^- \rightleftharpoons K^+ + SO_4^{2-}$ were roughly estimated (0.06 at 100°, 1.0 g/cm^3; and 0.01 at 200°, 1.0 g/cm^3 or 0.90 g/cm^3).

Analysis of Molten Salts by Electrochemical Methods. — Preliminary tests revealed that a pyrolytic graphite electrode (1/8 in. in diameter), encased in boron nitride such that only the impervious surface of the graphite is exposed to the fluoride melt, might be an excellent indicator electrode for voltammetric studies in fused salts. The compatibility of the boron nitride with the molten fluoride environment and the subsequent useful life of the electrode over an extended period of time, however, have not yet been determined.

An ORNL model Q-1988 controlled-potential polarograph was modified so that current-voltage curves could be recorded at voltage scan rates up to 10 v/min. The effect of scan rate on the current-voltage curves for the reduction of iron(II), to the metal, at a pyrolytic graphite electrode was then ascertained. From 50 to 500 mv/min, the curves were S-shaped. From 500 mv/min to 10 v/min, curves with a peaked shape were obtained. The occurrence of the peak is in agreement with the theory for diffusion-controlled reversible processes at stationary microelectrodes under conditions of continuously changing potential. The relationship between peak current and scan rate at 500°C is given by

$$i_p = 2.29 \times 10^5 \times n^{3/2}CAD^{1/2}v^{1/2} ,$$

where i_p is the peak current, n is the electron change, C is the concentration of electroactive species, A is the electrode area, D is the diffusion coefficient of the electroactive species, and v is the rate of voltage scan. A plot of i_p vs $v^{1/2}$ resulted in a straight line, and from the slope of the line a value of D of approximately 1×10^{-6} cm^2/sec was calculated. This is believed to be one of the first reported values for the diffusion coefficient of iron(II) in molten fluoride salt systems at 500°C.

Properties of Aqueous Solutions: Species and Thermodynamic Properties. — The nature of the aggregated species (polymolybdates) formed on addition of acid to alkali molybdates is yet greatly in dispute, despite extensive studies in many laboratories. Equilibrium ultracentrifugation has indicated that the average degree of polymerization is probably in the range 7 to 9 for p = 1 to 1.5, where p is the average number of hydrogen ions bound per molybdate, and that higher aggregates form above p = 1.5. Acidity measurements on solutions covering a range of p and molybdate concentrations could be explained by postulating species in this range of molecular weight, but were no more conclusive in arriving at a detailed description; that is, the experimental titration curves agreed equally with computed curves based on monomer-heptamer, monomer-octamer, or monomer-heptamer-octamer equilibria. With the help of Raman spectra, however, a more definitive choice appears possible. On the basis of structural studies, others have proposed that certain solid polymolybdates are arranged in heptameric and octameric units; these proposals are consistent with analytical alkali/molybdate ratios. Comparison of the Raman spectra of such solids with the Raman spectra of solutions of corresponding p indicates that both heptameric and octameric species are found in solution.

Radiation Chemistry. — There is some evidence from studies of the radiolysis of inorganic salts that thermal effects in alpha-particle tracks may be an important factor in the radiolysis. To test this hypothesis further, a comparison was made of the yields and the effect of postirradiation annealing on these yields for the decomposition of sodium chlorate by gamma rays and by alpha particles. The 100-ev yield, $G(Cl^-)$, for sodium chlorate irradiated with gamma rays at 25°C was 1.50 ± 0.07. Postirradiation annealing at 185°C increased the yield to 1.92 ± 0.04. The yield was still higher (2.30 ± 0.04) for salt irradiated at 185°C. The yield of chloride for decomposition by alpha particles (3.3 Mev) at 25°C was 2.12 ± 0.05, about 40% higher than for gamma rays. Also, there was no effect of postirradiation annealing at 185°C. The results are consistent with the previous postulate that thermal effects in alpha-particle tracks are a significant factor in the radiolysis.

Target Preparation. — Self-supporting germanium films were prepared by evaporation of the isotopically enriched element onto a glass substrate coated with a vapor-condensed layer of sodium chloride. Earlier experiments with this element had yielded friable films which deteriorated within a few days, apparently because of an improper choice of parting agent. Most poor films contained pinholes which made them less valuable for nu-

clear experiments. Germanium films now can be prepared in the 100- to 1000-$\mu g/cm^2$ thickness range.

Self-supporting thin films of antimony and silicon were prepared without pinholes and gross deviations in film thickness. Both elements were evaporated from a tantalum filament and condensed on a sodium chloride film (used as the parting agent). Glass was found to be preferable as the substrate for silicon, whereas polished stainless steel was found to be more suitable for antimony. In the case of antimony films, ruptures or cracks were observed on the substrate unless it was heated to ~100°C. The heating apparently enhances migration of condensed molecules on the sodium chloride surface. Silicon films thicker than 300 $\mu g/cm^2$ tended to roll when stripped and were therefore difficult to mount. Since annealing techniques to eliminate this strained condition have proved unsuccessful, the thickness range for silicon is relatively narrow, 100 to 300 $\mu g/cm^2$. Antimony films between 200 and 1000 $\mu g/cm^2$ thick have been prepared successfully.

To further improve evaporation characteristics of many elements, boron nitride is being studied as a high-temperature crucible material. This is expected to be an improvement over metal crucibles or filaments, since alloying will be prevented, and evaporation efficiency will be increased. Only metals and compounds which are nonreactive with nitrogen at high temperatures will evaporate satisfactorily from these crucibles.

Transuranium Element Processing

Corrosion Tests for the TRU Facility. — The corrosion rate for grade 11 zirconium was very slightly accelerated from a 1000-hr rate of 1.7 mils/month to a 1000-hr rate of 2.1 mils/month when 50 ppm of ferric ions as $FeCl_3 \cdot H_2O$ were added to boiling and oxygenated 6 M HCl solution. The hydrogen content of the specimen in the absence of ferric ions was 790 ppm; this value increased to 1300 ppm when ferric ions were present. Both specimens underwent brittle fracture during bending upon completion of the tests.

Solution and vapor specimens of Hastelloy C corroded at constant rates of 0.3 and 1.0 mil/month, respectively, after a 1000-hr exposure to oxygenated 10 M LiCl–0.1 M HCl solution at 43°C. Both specimens experienced preferential attack in heat-affected zones adjacent to welds, with the heavier attack, up to 10 mils in depth, occurring on the vapor-exposed specimens.

Preparation of PuO_2 for HFIR Target Prototypes. — Eighty-five grams of high-fired PuO_2 containing about 75 g of Pu^{239} was prepared for use in prototype HFIR targets. Plutonium hydroxide was precipitated and dried at 150°C, the hydroxide was ground through a 70-mesh screen, and the +325-mesh $Pu(OH)_4$ was fired in air at 1200°C to produce glassy solid particles with a density of 10.99 (96% of theoretical). Particle size of the fired oxide is as follows:

Screen Size	wt %
−70 +140	43.5
−140 +170	16.3
−170 +200	8.6
−200 +325	24.3
−325	7.3

The oxide will be mixed with aluminum powder (−325 mesh) and pressed into cermets. With PuO_2 particles of this size the aluminum phase should be continuous, and conductivity will be satisfactory for irradiation in high neutron fluxes.

Separation of Americium from Curium at High Alpha Levels. — Separation of americium from curium by chromatographic elution from a cation exchange column with ammonium α-hydroxyisobutyrate at activity levels of about 50 w/liter was attempted. Four milligrams of Cm^{242} was loaded on a 30-ml column of Dowex 50-8X (−400 mesh) resin at 80°C from a solution containing americium and curium. The loaded column was eluted with 0.4 M ammonium α-hydroxyisobutyrate at pH 4.2 at a flow rate of 0.4 ml cm^{-2} min^{-1}. Extensive gassing problems were encountered, with gas pockets up to 1 in. long developing. However, in one of the runs a 70% curium recovery with a 14% americium carry-over was obtained. The column was modified with a spring-loaded device that kept the resin bed compacted. Gas pockets and channeling were prevented despite large amounts of radiolytic gas, and a curium recovery of 90% with a 10% americium carry-over was achieved. A separation factor of 1.4 was obtained, and the system behaved well chemically.

Direct, Nondestructive Method for the Determination of Cf^{252}. — The prompt gamma rays accompanying the spontaneous fission of Cf^{252} offer the interesting possibility of a direct, nondestructive method for its determination. Californium-252 has an alpha half-life of 2.55 yr and a spontaneous fission half-life of 66 yr. The gamma-ray spectrum in coincidence with the spontaneous fission extends to about 5 Mev. Each fission event yields an average of 10 to 12 gamma rays. Discriminatory gamma scintillation counting has been found to provide a sensitivity of at least 0.01 nanogram of Cf^{252}. The method is applicable to the direct determination of Cf^{252} in the presence of other actinide elements and gross fission products.

TRU Processing Engineering. — Performance of a manipulator-booting-seal extractor was satisfactory; however, application of talcum powder to the rubber seal surfaces before installation was necessary to prevent excessive friction during extraction. From tests in the cell mockup, alterations in the cell lighting system were recommended to give more uniform intensities and easier remote maintenance. A tantalum-to-Hastelloy rolled joint was tested with temperature cycling from ambient to 285°C. The joint appears to be acceptable for the intended uses, with no detectable leakage for 15 psig below 200°C.

METALLURGY AND MATERIALS RESEARCH

Preparation of Pure Materials. — A bulk sample of the fourth LiF crystal (ORNL-3451, p 27) was analyzed by the thermal-neutron activation method. The results, though in generally good agreement with those obtained with the emission spectroscopic method, indicate that even greater purity may have been achieved than was indicated by the emission spectroscopy results. Concentrations of the contaminants, Na, K, and Mn, were found to be 0.01, $\leq$0.2, and 0.349 ppm respectively. There was no evidence of the presence of chlorine, aluminum, or vanadium (indicating concentrations of < 1 ppm).

A 290-g charge of high-purity LiF containing 79.28 at. % Li^7 was dehydrated for the production of the sixth single crystal, which is now being grown.

CONTROLLED THERMONUCLEAR RESEARCH

Plasma Physics and Arc Research. — A deuterium plasma with an electron temperature greater than 100 kev and a density of about 10^{11} electrons/cm^3 can be formed between magnetic mirrors by the beam-plasma interaction of the pressure-gradient arc. A measurement of the change in magnetic field by the voltage induced in a loop around the plasma indicates an average plasma β ($8\pi nkTB^{-2}$) of 0.1, in agreement with the value calculated from the electron temperature and density. The plasma decay after electron beam turnoff has been observed by the change in magnetic field and by the x-ray flux. Decay constants as long as 70 msec have been measured. This is in rough agreement with the calculated decay constant from scattering losses through the mirrors.

The velocity of ionic sound waves in a plasma has now been measured directly by a time-of-flight technique, instead of being inferred from the frequency and wavelength of a standing-wave system. The agreement between the experimentally measured velocities and the theoretically computed velocities is quite good, having an rms scatter of about ±25%.

DCX-1 Facility. — Experimental emphasis in the DCX-1 has again been placed on the production of plasmas by Lorentz dissociation of H_2^+ beams formed from H_3^+ injected into a gas cell. In one series of experiments the H_3^+ current was a constant 2.5 ma, and the gas cell pressure was varied to change the injected H_2^+ current. The plasma density was found to increase linearly (to within 20%) with the H_2^+ current in the range from 20 to 200 μa (the latter corresponding to 8% conversion of H_3^+ to H_2^+). These data showed that the fraction of the H_2^+ beam trapped by Lorentz dissociation was not a pronounced function of gas cell pressure in the range investigated. The plasma containment time during these experiments was 2 sec, and the trapped proton density with 200 μa of injected H_2^+ was about 2×10^8 protons/cm^3.

Another series of experiments investigated the effects on base pressure of an "inner liner" which was placed inside the usual liner so as to reduce the temperature of the walls surrounding the plasma to more nearly

that of liquid nitrogen. With the inner liner present, charge-exchange decay times of 60 sec were easily obtained. This liner outlined the construction practices necessary for future liner assemblies but was itself too small to permit extensive plasma property studies.

So far the most dense plasma (5×10^8 protons/cm^3) was attained with 200 μa of injected H_2^+ and trapped proton containment times of 7 sec. The source has recently been realigned, and 4.5 ma of H_3^+ has been extracted through the conversion cell. With this gain in injected current and the gain of 10 that should be realizable through pressure reduction, the maximum density would be about 10^{10} protons/cm^3. Experiments designed to attain this density are in progress.

DCX-2 Facility. — After rechecking magnetic-field uniformity and repairing some faults in the DCX-2 apparatus, several attempts were made to establish the type of time correlation which exists between fast ion current bursts as a function of z or θ. Although a gross, qualitative correlation exists and can be observed in time-integrated signals from two probes displayed on an X-Y oscilloscope, a correlation in detail cannot be seen in the signals to probes, even when they are located adjacent to each other in the machine. An explanation is not yet available for these observations. An electron beam, 7 amp and 4 kv, was injected through one mirror in a 3.5-in. ring about the axis of the DCX-2. It was hoped to modify the onset of the Harris instability by virtue of the large flux of electrons flowing through the machine. The intensity of the ion cyclotron frequency radiation was only slightly affected, and a large increase in radiation was observed over the entire measurable frequency spectrum (10 to 100 Mc). Further, the ion energies were observed to spread to energies as high as 1 Mev. The observed density was not greatly affected. Additional experiments with the backstream from the lithium arc confirmed the earlier observation of ion-burst suppression, but the trapped density did not increase as previously observed. It was noted that the magnetic-field uniformity had been disturbed, and a coil short was found in one of the inboard coils. Following an argon-arc bakeout, the pressure did not appear to fall as expected, and the machine was disassembled for thorough cleaning.

Vacuum. — The energy of desorption of hydrogen from a molybdenum surface was measured with the 15-ft^3 ultrahigh-vacuum test facility and was found to be 2.2 kcal/mole from an Arrhenius plot of the pressure-temperature data. In order to allow the development of these data for application to the large experimental devices, a series of experimental design modifications was begun. The vacuum test facility was modified to minimize the surface area of materials which are not representative of the experimental surface and to minimize the area of those surfaces at temperatures other than the experimentally controllable temperature. A facility was added to allow the gas conductance from the inner (liner) region to the intermediate vacuum region to be varied. A baffle was designed and constructed which will allow trapping of condensable species to be effective, even when the inner liner is at its lowest operating temperature. The modified unit has not yet been tested.

The bakeable flange test on 0.086-in.-diam aluminum wire gaskets was concluded in this period. Leak-tight sealing of this type of gasket between plain flange surfaces was found to be closely related to the mechanical properties of the alloy (in this case 1100-0 aluminum). Although

the gasket was limited to a finite number of thermal cycles because of creep, it was possible to maintain vacuum closure for more than 18 cycles between room temperature and 250°C with 4 hr per baking cycle at the elevated temperature. The maximum loading force necessary for the gasket to remain in the plastic sealing region was found to be about 8-1/2 tons/in.^{2} or about 3000 lb per linear inch of gasket. Initial sealing of this gasket was achieved with a gasket deformation of approximately 0.028 in. and a force of 1500 lb per linear inch. A study of commercially available silver-plated stainless steel hollow O-rings in appropriately designed flanges was initiated. After four baking cycles at 400°C, closure quality was unimpaired.

BIOLOGY AND MEDICINE PROGRAM

TERRESTRIAL AND FRESH-WATER ECOLOGY

Evaluation of Fission Product Distribution and Movement in White Oak Drainage System. — Analyses of a series of 250 core samples taken in the bed of former White Oak Lake showed that as of December 1962 the soil contained 1038 ± 88 curies of Ru^{106}, 704 ± 34 curies of Cs^{137}, 152 ± 9 curies of Co^{60}, 16.4 ± 3.3 curies of the rare earths (exclusive of Y^{90}), and 14.6 ± 1.1 curies of Sr^{90}. The percent distributions of the various radionuclides were:

Layer of Lake-Bed Soil	Radionuclide Distribution (%)				
	Ru^{106}	Cs^{137}	Co^{60}	Rare Earths	Sr^{90}
0 to 6 in.	57	67	78	79	68
6 to 12 in.	27	29	16	15	24
12 to 18 in.	11	4	5	6	7
18 to 24 in.	5	<1	1	<1	1

RADIOLOGICAL PHYSICS, HEALTH PHYSICS, AND INSTRUMENTATION

Health Physics Research Reactor Studies. — The Health Physics Research Reactor (HPRR) attained criticality in its permanent facilities on May 31, 1963, at 7:23 PM. Although the reactor performed well during its operation on the BREN tower in 1962, the control system was simplified and more instrumentation was added. Also, a modification was made to facilitate removal of various components in order to minimize personnel exposures during routine and nonroutine servicing and inspection. There are now two count-rate channels for startup (formerly one) and five temperature channels (formerly two).

ISOTOPE DEVELOPMENT PROGRAM

Nuclear Analysis Research and Development: Use of a 14-Mev Neutron Generator. — The effects of the plastic rabbit assembly of the 14-Mev neutron generator (Texas Nuclear Corporation) on fast-neutron irradiations have been studied. The decrease in induced activity inside the rabbit caused by scattering of the neutrons is somewhat compensated for by what is believed to be a hardening of the flux in the forward direction. The reduction in induced activity caused by the plastic varies from zero for the reaction $Cu^{63}(n,2n)Cu^{62}$ to 47% for the reaction $Si^{28}(n,p)Al^{28}$.

Nuclear Analysis Research and Development: Preparation of a Guidebook on Activation Analysis. — Material has been written, assembled, edited, and forwarded to the Division of Isotopes Development (AEC) for a guidebook on activation analysis. The compilation contains a comprehensive survey of background information necessary for an understanding of activation analysis, together with detailed descriptions of techniques utilized in the application of nuclear analysis methods.

Nuclear Analysis Research and Development: Assistance to Law Enforcement Officers in Evaluating Drug Seizures. — Assistance has been given to the police department of New York City in determining the similarity of a number of samples of heroin. Attempts to match a minute sample of the seized drug with material from a large capture were only partially successful. Qualitatively, some agreement was noted as to the presence of certain trace elements, but exact matching was not observed. In similar work in the past for the Bureau of Narcotics, it was conclusively demonstrated that another sample was not from the same source as that of an earlier seizure.

Curium-242 Recovery. — About 20 mg of Cm^{242} was recovered from an irradiated Am^{241} target in the Building 3019 analytical hot cells by the LiCl anion exchange method. Curium product analysis by gamma-ray spectrometry showed Ru^{103} to be the only gamma-emitting fission product present. Gross fission product decontamination was greater than 10^4. The 20 mg of Cm^{242} product emitted 2.5 r/hr of gamma radiation; however, this activity was completely attenuated with 1 in. of steel.

The recovered Cm^{242} is being used in Building 3508 for the evaluation of the extent of acid depletion by radiolysis of chloride solutions, separation processes, and the physical handling of 10- to 20-w/liter solutions.

The principal effect of alpha radiation in Tramex feed is a loss of acid by radiolysis. Attempts were made to stabilize the acidity of the feed solution by the addition of materials that liberate HCl by hydrolysis. Hydrazine dihydrochloride and hydroxylamine hydrochloride were investigated. Both hydrolyze to give approximately the required concentration of HCl. However, in the presence of high alpha activity they liberate acid at a faster rate than acid is destroyed, and a net hydrogen ion increase results. Therefore, addition of an excess of these materials does not appear to be a satisfactory way of stabilizing the acidity of Tramex feed for long-term storage.

Bubbling dry helium gas containing small amounts of HCl through Tramex feed was evaluated as a possible method of acid adjustment. Equilibrium acid values were greater than 0.1 M; therefore, this procedure appears to be of dubious value because acid must be less than 0.1 M for satisfactory actinide extraction, and the HCl partial pressure cannot be reduced appreciably below that used (23 mm) and still replenish acid lost by radiolysis at a reasonable rate of gas flow.

Another method that makes use of an electrochemical pH-Stat (pH adjuster) developed at ORNL was investigated for replenishing acid. In radioactive solution the electrode was seriously poisoned. It is doubtful whether electrodes satisfactory for service in Tramex feed at 10 w/liter can be found.

Recovery of Fission Products. — In a continuous countercurrent demonstration run in miniature mixer-settler equipment, more than 99.3% of the cesium was recovered from tartrate-complexed simulated Purex 1WW waste (pH 12.7) with 1 M 4-sec-butyl-2-(α-methylbenzyl)phenol (BAMBP) in Amsco 125-82. The extract was scrubbed with 0.002 M sodium tartrate and stripped with 0.1 M HNO_3. Flow ratios, aqueous feed/organic/scrub/strip, were 2/1/0.2/0.05. No apparent change in performance was noted through 26 hr of operation, equivalent to 23 complete cycles of the solvent through the system.

Effective extraction of technetium from simulated Hanford tank-waste supernatant was obtained with quaternary amine extractants. The extraction coefficient (1/1 phase ratio) with 0.17 M Aliquat 336 in 94% diisopropylbenzene—6% tridecanol was 75 at pH 9 and 130 at pH 13.

Radioisotope Research and Development. — The feasibility of I^{125} production with < 1% I^{126} has been investigated. Analog computer calculations indicate that a loop with normal xenon or a short batch irradiation of either normal or enriched Xe^{124} with early separation of the undecayed Xe^{125} should be considered. The most feasible method appears to be batch irradiation for a production rate of less than 5 curies/week. The capital expense involved in the use of a loop appears to be prohibitive at the ~5-curie/week level.

The preparation of Tc^{99} as a source to be used in reducing scar tissue formation which occurs as a result of eye surgery is being investigated. The presently used 28-yr Sr^{90} offers disadvantages because of the 2.27-Mev Y^{90} beta particle resulting from the 0.54-Mev beta decay of Sr^{90}. The 2.1×10^5-yr Tc^{99} has only a 0.29-Mev beta particle associated with its decay. Technetium-99 metal has been plated on copper foil from a solution of 0.65 M ammonium oxalate, using sulfuric acid to adjust the pH to 2.0. A 0.002-in.-thick plating, which weighed 5.9 mg, was deposited in 30 min with a current of 1.0 amp and 4.0 v. A smoother and more adherent deposit was obtained at a pH of 1.0 to 1.5; with less-acid solutions a flaky oxide was obtained. The Tc^{99} is malleable when plated on copper foil and may be formed into a 0.5-in.-diam cylinder without cracking.

The thermal power of a 3934-g Pm^{147} sample, with a radiometrically determined specific activity of 641 mc/g, was calorimetrically measured and found to be 1.02 ± 0.01 w or 0.404 w/kc. The calculated value, based on the beta decay energy obtained from the literature, is 0.38 w/kc.

Radioisotope and Stable Isotope Production. — Ion currents equivalant to 2450 g of separated isotopes were monitored during May and June. Isotopes of Ca, Si, Pb, and Ba were separated and collected, using 28 of the calutrons associated with the program.

Special feed materials have been supplied for charge evaluation studies associated with the current silicon separation series. Several runs have been made with $SiCl_4$, SiS_2, and SiS_2 mixed with silicon metal. Use of both the SiS_2 and the mixture resulted in more stable operating conditions, doubled the ion outputs achieved with previous charge materials, and permitted the simultaneous collection of silicon and sulfur isotopes.

A total of 3488 g of rhenium metal has been recovered from an original supply of 3695 g used as feed material, giving a recovery factor of 94% for this series.

OPEN LITERATURE PUBLICATIONS

Allen, K. A., and W. J. McDowell, "The Thorium Sulfate Complexes from Di-n-decylamine Sulfate Extraction Equilibria," J. Phys. Chem. 67, 1138 (1963).

Armento, W. J., and C. E. Larson, "Determination of Sulfate with Chromium-51," Anal. Chem. 35, 918 (1963).

Chandrasekhar, B. S., W. F. Gauster, and J. K. Hulm, "Degradation Factor and Diamagnetic Currents in Supermagnets," Appl. Phys. Letters 2, 228 (1963).

Dworkin, A. S., R. A. Sallach, J. R. Bronstein, M. A. Bredig, and J. D. Corbett, "The Electrical Conductivity of Solutions of Metals in Their Molten Halides. VI. Lanthanum, Cerium, Praseodymium, and Neodymium in Their Molten Iodides," J. Phys. Chem. 67, 1145 (1963).

Godbee, H. W., and W. E. Clark, "Use of Phosphite and Hypophosphite to Fix Ruthenium from High-Activity Wastes in Solid Media," Ind. Eng. Chem., Prod. Res. Develop. 2, 157 (1963).

Hurst, G. S., J. Stockdale, and L. B. O'Kelly, "Interaction of Low-Energy Electrons with Water Vapor and with Other Polar Molecules," J. Chem. Phys. 38, 2572 (1963).

Lehmann, C., and G. Leibfried, "Higher Order Momentum Approximations in Classical Collision Theory," Z. Physik 172, 465 (1963).

Mallard, W. C., A. B. Gardner, R. F. Bass, and L. M. Slifkin, "Self-Diffusion in Silver-Gold Solid Solutions," Phys. Rev. 129, 617 (1963).

Marshall, W. L., and Ruth Slusher, "Aqueous Systems at High Temperature. VIII. The Solubility of UO_3 Hydrates in HNO_3-H_2O Solutions, 25–350°C, 10^{-4}–6 Molal Nitrate," J. Inorg. Nucl. Chem. 25, 283 (1963).

Martin, W. R., and H. E. McCoy, "The Effect of CO_2 on the Strength and Ductility of Type 304 Stainless Steel at Elevated Temperatures," Corrosion 19, 157t (1963).

McDonald, D. L., "Low Temperature Irradiation of Beryllium Oxide," Appl. Phys. Letters 2, 175 (1963).

Miller, F. J., "The Pyrolytic Graphite Electrode as an Indicating Electrode for Potentiometric Titrations," Anal. Chem. 35, 871 (1963).

Mountcastle, W. R., Jr., W. D. Shults, and P. F. Thomason, "Controlled-Potential Coulometric Determination of Indium," Anal. Chem. 35, 871 (1963).

Ross, H. H., and R. E. Yerick, "Quantitative Interpretation of Color Quenching in Liquid Scintillator Systems," Anal. Chem. 35, 794 (1963).

Rush, R. M., and J. S. Johnson, "Hydrolysis of U(VI): Absorption Spectra of Chloride and Perchlorate Solutions," J. Phys. Chem. 67, 821 (1963).

Shults, W. D., and L. B. Dunlap, "Controlled-Potential Coulometric Determination of Uranium(VI) in Uranium-Niobium Alloys," Anal. Chem. 35, 921 (1963).

Snyder, W. S., and M. J. Cook, "The Distribution of Dose Resulting from a Constant Level of Contamination of the Environment by Sr^{90} and Various Other Radionuclides," Health Phys. 9, 417 (1963).

Strickler, T. D., "Ionization by Alpha Particles in Binary Gas Mixtures," J. Phys. Chem. 67, 825 (1963).

Sturm, B. J., and C. W. Sheridan, "Vanadium(III) Fluoride," pp 87-92 in Inorganic Syntheses, vol VII (ed. by J. Kleinberg), McGraw-Hill, New York, 1963.

Tipton, Isabel H., and M. J. Cook, "Trace Elements in Human Tissue. II. Adult Subjects from the United States," Health Phys. 9, 103 (1963).

Tipton, Isabel H., M. J. Cook, R. L. Steiner, C. A. Boye, H. M. Perry, Jr., and H. A. Schroeder, "Trace Elements in Human Tissue. I. Methods," Health Phys. 9, 89 (1963).

VISITORS

UNITED STATES

Advisory Committee for the Electronuclear and Physics Divisions

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

FOREIGN

Name	Affiliation	Citizen of
Barthoux, J.	French CEA	France
Bishop, C. M.	Atomic Energy of Canada	Canada
Bressler, R. G.	University of Tennessee	Germany
Bromley, D. A.	Yale University	Canada
Colomb, J. P.	S. A. Conrad Zcehokke, Ltd.	Switzerland
Croft, J. F.	United Kingdom AEA	United Kingdom
Dalle-Donne, M.	EURATOM	Italy
Dolphin, G. W.	United Kingdom AEA	United Kingdom
Eggleton, A. E. J.	Atomic Energy Research Establishment	United Kingdom
Gleyvod, R.	Florida State University	Switzerland
Grenon, M. A.	EURATOM	France
Guillaume, L. E.	French CEA	France
Heerschap, M.	EURATOM	Holland
Lapid, F. M.	University of Tennessee	Philippines
Lavie, J. M.	French CEA	France
Lindqvist, T.	University of Uppsala	Sweden
Lovell, G. H. B.	South Africa Atomic Energy Board	South Africa
Makino, N.	Japan Atomic Energy Research Institute	Japan

Name	Affiliation	Citizen of
Marcus, F. R.	Eurochemic	Denmark
Martin, J. C.	French CEA	France
Miyashige, S.	Ishikawajima-Harima Heavy Industries	Japan
Nichols, C. M.	United Kingdom AEA	United Kingdom
Nojima, K.	Ishikawajima-Harima Heavy Industries	Japan
Park, J. Y.	North Carolina State College	South Korea
Pomarola, J.	French CEA	France
Potts, E. L. J.	University of Durham	United Kingdom
Rees, A. L. G.	Chemical Research Laboratories	Australia
Robson, D.	Florida State University	Australia
Rubin, E.	Radiation Applications, Inc.	Israel
Sano, T.	Osaka University	Japan
Schroder, I. G.	Columbia University	Chile
Sequin, M.	French Atomic Energy Commission	France
Shenton, L. R.	Virginia Polytechnic Institute	United Kingdom
Skaardal, R. C.	Argonne National Laboratory	Norway
Smith, H.	United Kingdom AEA	United Kingdom
Spahrbier, D. H.	VARTA	Germany
Stevens, J. E.	Atomic Energy of Canada	Canada
Taylor, R. B.	Florida State University	Australia
Vallette, L.	EURATOM	Belgium
Whitkower, A. B.	High Voltage Engineering Corporation	United Kingdom
Yu, S. K.	Georgia Institute of Technology	Formosa

Reports issued in this series during the past year:

June 1962	ORNL-3321
July 1962	ORNL-3331
August 1962	ORNL-3351
September 1962	ORNL-3368
October 1962	ORNL-3377
November 1962	ORNL-3388
December 1962	ORNL-3402
January 1963	ORNL-3410
February 1963	ORNL-3426
March 1963	ORNL-3436
April 1963	ORNL-3451
May 1963	ORNL-3462

ORNL-3477
C-44a – Nuclear Technology – Materials
M-3679 (29th 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. J. S. Drury
39. J. L. Dunlap
40. A. C. England
41. M. F. Fair
42. D. E. Ferguson
43-46. J. L. Fowler
47. J. H. Frye, Jr.
48. J. L. Gabbard (K-25)
49. W. F. Gauster
50. J. H. Gillette
51. W. Y. Gissel
52. H. E. Goeller
53. C. D. Goodman
54. A. T. Gresky
55. J. C. Griess
56. W. R. Grimes
57. P. A. Haas
58. C. S. Harrill
59. J. C. Hart
60. M. R. Hill
61. C. J. Hochanadel
62. A. Hollaender
63. L. B. Holland
64. R. W. Horton
65. F. T. Howard
66. A. S. Householder
67. H. H. Hubbell
68. G. S. Hurst
69. G. H. Jenks
70. R. W. Johnson
71. R. J. Jones
72. W. H. Jordan
73. P. R. Kasten
74. G. W. Keilholtz
75. G. G. Kelley
76. M. T. Kelley
77. E. M. King
78. K. A. Kraus
79. Laboratory Shift Supervisor
80. J. A. Lane
81. D. M. Lang (K-25)
82. C. E. Larson
83. C. F. Leitten, Jr.
84. R. E. Leuze
85. T. A. Lincoln
86. S. C. Lind
87. R. B. Lindauer
88. R. S. Livingston
89. J. T. Long
90. L. O. Love
91. R. N. Lyon
92. H. G. MacPherson
93. F. C. Maienschein
94. W. D. Manly
95. J. A. Martin

96. H. F. McDuffie
97. J. R. McNally
98. R. A. McNees
99. A. J. Miller
100. E. C. Miller
101. K. Z. Morgan
102. W. L. Morgan
103. E. J. Murphy
104. J. P. Murray (K-25)
105. R. V. Neidigh
106. M. L. Nelson
107. G. W. Parker
108. R. B. Parker
109. P. Patriarca
110. J. J. Pinajian
111. M. E. Ramsey
112. A. W. Riikola
113. L. P. Riordan
114. A. F. Rupp
115. A. D. Ryon
116. H. C. Savage
117. H. W. Savage
118. A. W. Savolainen
119. H. E. Seagren
120. C. H. Secoy
121. E. D. Shipley
122. O. Sisman
123. J. R. Sites
124. M. J. Skinner
125. A. H. Snell
126. W. S. Snyder
127. W. L. Stirling
128. R. W. Stoughton
129. H. Stringfield
130. E. G. Struxness
131. J. C. Suddath
132. W. H Sullivan
133. C. D. Susano
134. J. A. Swartout
135. E. H. Taylor
136. R. E. Thoma
137. C. D. Watson
138. B. S. Weaver
139. M. S. Wechsler
140. A. M. Weinberg
141. M. E. Whatley
142. J. C. White
143. E. O. Wollan
144. R. W. Worsham
145. R. G. Wymer
146. Gale Young
147. W. Zobel
148. A. Zucker
149-150. Central Research Library
151-152. ORNL - Y-12 Technical Library Document Reference Section
153. Reactor Division Library
154. Biology Library
155-345. Laboratory Records Department
346. Laboratory Records, ORNL R.C.

EXTERNAL DISTRIBUTION

347. Division of Reactor Development, AEC, Washington (J. M. Simmons)
348-356. Research and Development Division, AEC, ORO
357. Aeroprojects, Inc. (W. B. Tarpley)
358. Allis-Chalmers Manufacturing Co. (W. F. Banks)
359-361. Ames Laboratory, Iowa State College (F. H. Spedding)
362-364. Argonne National Laboratory (A. V. Crewe)
365. Atomics International (H. Pearlman)
366. Babcock and Wilcox Co. (Atomic Energy Division)
367-368. Battelle Memorial Institute (H. W. Russell)
369-371. Brookhaven National Laboratory (M. Goldhaber)
372. Bureau of Mines, Salt Lake City (J. B. Clemmer)
373-380. General Atomic (General Dynamics Corp.) (Manager)
381-382. General Nuclear Engineering Corp. (J. West)
383. Grand Junction Operations Office (W. C. Hutchinson, Jr.)
384-385. Idaho Chemical Processing Plant (R. L. Doan)
386. Kaiser Engineers (P. D. Bush)
387-388. Knolls Atomic Power Laboratory (K. R. Van Tassell)
389-390. Lawrence Radiation Laboratory (H. Fidler)

391-392. Los Alamos Scientific Laboratory (N. E. Bradbury)
393-394. National Bureau of Standards (L. S. Taylor)
395. National Lead Company (C. K. McArthur)
396. Nuclear Metals, Inc. (A. R. Kaufmann)
397-398. Philadelphia Electric Co. (L. R. Gaty)
399. Sanderson and Porter (Atomic Energy Division)
400. Union Carbide Corporation (R. W. McNamee)
401. USAEC, Assistant General Manager (S. G. English)
402. USAEC, Division of Biology and Medicine (C. L. Dunham)
403-405. USAEC, Division of Isotope Development (P. C. Aebersold)
406. USAEC, Division of Production (F. P. Baranowski)
407. USAEC, Division of Raw Materials (J. C. Johnson)
408-422. USAEC, Division of Reactor Development (F. K. Pittman)
423-428. USAEC, Division of Research (P. W. McDaniel)
429. USAEC, Division of Technical Information (Technical Reports Library)
430-431. USAEC, Chicago Operations Office
432. USAEC, Lockland Aircraft Reactors Operations Office (H. H. Gorman)
433-434. USAEC, San Francisco Operations Office (R. Hughey)
435-436. USAEC, Savannah River Operations Office (H. Rahner)
437. Westinghouse Electric Corp. (A. Squire)
438-694. Given distribution as shown in M-3679 (29th ed.) category C-44a and those addresses shown in C-44b, C-44c, and C-44d, which do not receive copies under C-44a