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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

DECEMBER 1963

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Contract No. W-7405-eng-26

OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

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Oak Ridge, Tennessee
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CONTENTS

REACTOR DEVELOPMENT..... 1
GAS-COOLED REACTOR PROGRAM..... 1
Graphite Oxidation Experiment..... 1
GCR-ORR Loop No. 2 Irradiations..... 1
LITR Capsule Irradiations..... 1
ORR Poolside Capsule Irradiations..... 1
Eight-Ball Experiment..... 2
ETR Capsules..... 2
Instantaneous Fission-Gas Release from Pyrolytic-Carbon-Coated Fuel Particles (B9-C1 Experiments)..... 2
Phase Relationships in Thorium Dicarbide..... 2
Self-Diffusion of Beryllium in Unirradiated Beryllium Oxide..... 2
MARITIME REACTOR PROGRAM..... 3
NS "Savannah" Filter Testing..... 3
ARMY PACKAGE POWER REACTOR PROGRAM..... 3
PM Irradiation Test Program..... 3
Corrosion Tests..... 4
Water Chemistry..... 4
APPLIED AND BASIC REACTOR PHYSICS..... 4
Basic Reactor Physics..... 4
REACTOR FUELS AND MATERIALS..... 5
Behavior of High-Temperature Materials Under Irradiation..... 5
FUNDAMENTAL HEAT TRANSFER AND FLUID DYNAMICS..... 5
Boundary Layer Transient Phenomena..... 5
Boiling-Potassium Studies..... 6
Boiling-Water Studies..... 6
POWER REACTOR FUEL PROCESSING..... 7
Chemical Analysis of Advanced Reactor Fuels..... 7
Uranium Carbide Hydrolysis..... 7
Protactinium Adsorption..... 8
Thorex Solvent Extraction..... 8
Uranium Adsorption..... 9
Shear-Leach Process..... 9
Corrosion..... 10
THORIUM UTILIZATION PROGRAM..... 11
U^{233} Purification by Solvent Extraction..... 11
Thoria Preparation..... 11
Mixed Thoria-Urania Preparation in the Fuel-Rod Facility..... 11
Rod Fabrication..... 12
Pellet Fabrication..... 12
Thorium-Uranium Fuel Cycle Development Facility (TUFCDF)..... 12
Sol-Gel Microsphere Preparation..... 12
Irradiation of Thorium Oxide Fuels..... 13
Permeability of Vibratorily Compacted Fuels..... 13
Preparation and Properties of U(IV) Sols and UO_2..... 13

[redacted] [redacted]

Sol-Gel Preparation and Properties of ThC_2 and UC_2-ThC_2 14
Physical and Chemical Properties of Molten Chlorides 14
MOLTEN-SALT REACTOR PROGRAM 14
Molten-Salt Reactor Experiment 14
Development of Components and Systems 15
MSRE Pump Program 15
Instrumentation Development 16
NUCLEAR SAFETY 16
Fuel Perturbation Effects in Pool-Type Reactors 16
Prompt-Neutron Lifetime in Critical Enriched-Uranium Metal Cylinders 17
Melting Experiments in TREAT Facility 17
Innocuous Simulation of Accidents and Hazards 19
Release of Fission Products on Out-of-Pile Melting of Reactor Fuels 20
Release of Fission Products on In-Pile Melting of Reactor Fuels 20
Characterization and Control of Accident-Released Fission Products 21
Nuclear Safety Pilot Plant 22
Reactor Containment Handbook 22
Nuclear Safety Information Center 23
WASTE DISPOSAL 24
Clinch River Study 24
Mineral Exchange Studies 24
Disposal in Deep Wells 24
Foam Separation Studies 25
High-Activity-Waste Calcination 26
Low-Activity Waste: Ion Exchange Studies 27
Low-Activity Waste: Pilot Plant Work 27
Disposal in Natural Salt Formations 27
Engineering, Economic, and Safety Evaluation 28
PHYSICAL RESEARCH 29
PHYSICS AND MATHEMATICS 29
High-Energy Electronuclear Machines: The Separated-Orbit Cyclotron 29
Medium-Energy Nuclear Physics: The 86-Inch Cyclotron 29
Medium-Energy Nuclear Physics: Theory 30
Cyclotron Operations: The Isochronous Cyclotron 30
Stable Isotopes Development 31
Special Separations 31
CHEMISTRY 32
Instrumental Methods for Continuous Chemical Analysis 32
Analysis of Molten Salts by Electrochemical Methods 33
Protactinium Chemistry 33
Properties of Aqueous Solutions: EMF and Solubility Studies 34
Properties of Aqueous Solutions: Species and Thermodynamic Properties 34

[redacted] [redacted]

Spectrophotometry of Solutions at High Temperature..... 34
High-Temperature and Structural Chemistry..... 35
Physical Chemistry of Gaseous Thermodynamic Working Fluids..... 35
Target Preparation..... 35
TRANSURANIUM-ELEMENT PRODUCTION..... 36
Chemical Process Development: Tramex Safety..... 36
Chemical Process Development: Americium-Curium Separation at High Activity Levels..... 36
Chemical Process Development: Separation of Lanthanides from Actinides by the TALSPEAK Process..... 36
Process Equipment Development: Cell Mockup Studies..... 37
METALLURGY AND MATERIALS..... 37
Preparation of Pure Materials..... 37
Reactions at Metal Surfaces..... 37
Deformation of Crystalline Solids..... 38
Spectroscopy of Ionic Media..... 38
Fundamental Research in X-Ray Diffraction..... 39
Crystal Physics..... 39
Fundamental Investigation of Radiation Damage in Solids: Dose Dependence of Dislocation Breakaway Stress in Neutron-Irradiated Copper as Measured by Amplitude-Dependent Internal Friction..... 40
Fundamental Investigation of Radiation Damage in Solids: Model for Radiation Equilibrium Between F and M Centers in KCl..... 40
Fundamental Investigation of Radiation Damage in Solids: Assembly and Use of an Ionized Argon Beam System for Determining the Sputtering Yield of Gold..... 40
Fundamental Investigation of Radiation Damage in Solids: Microstrain in Low-Dislocation-Density Copper Crystals as Investigated with Borrmann X-Ray Transmission..... 41
CONTROLLED THERMONUCLEAR RESEARCH..... 41
Plasma Physics..... 41
DCX-2 Facility..... 42
Vacuum..... 42
Magnetics and Superconductivity..... 42

BIOLOGY AND MEDICINE..... 43
MOLECULAR- AND CELLULAR-LEVEL STUDIES..... 43
Physics of Tissue Damage..... 43
TERRESTRIAL AND FRESH-WATER ECOLOGY..... 43
Radiobotanical Studies on White Oak Lake Bed..... 43
Transport of Radioactive Materials by Mud-Dauber Wasps..... 44
Electrophoretic Analyses of Wild-Mammal Blood..... 45
Incorporation of 5-Iodo-2′-Deoxyuridine-I^{131} into the DNA of Indigenous Mammals..... 45
Ribonucleic Acid Characteristics in Mimosa..... 46
Transfer of Cs^{137} from Tagged Tulip Poplar Trees to the Untagged Understory..... 46

[redacted] [redacted]

[redacted] [redacted]

Detritus in the Aquatic Food Chain..... 47
Evaluation of Fission Product Distribution and Movement in White Oak Drainage System..... 48
RADIOLOGICAL PHYSICS, HEALTH PHYSICS, AND RADIATION INSTRUMENTATION..... 48
Radiation Physics and Dosimetry: Optical Absorption in the Alkali Metals..... 48
Radiation Physics and Dosimetry: Low-Energy End of the Secondary Electron Cascade in Metals..... 48
Radiation Physics and Dosimetry: Interaction of a Stationary Plasma with an Electron Beam..... 49
Radiation Physics and Dosimetry: Plasma-Beam Instability in the Hartree-Appleton Approximation..... 49
Radiation Physics and Dosimetry: Optical Constants of Vacuum-Evaporated Silver Films..... 50
Radiation Physics and Dosimetry: Transition Radiation and Optical Bremsstrahlung from Electron-Bombarded Thin Gold Foils..... 50
Radiation Physics and Dosimetry: Angular and Spectral Distributions of Transition Radiation from Thin Silver Foils..... 50
Radiation Physics and Dosimetry: Interaction of Low-Energy Electrons with Other Polar Molecules..... 51
Radiation Physics and Dosimetry: Time-of-Flight Investigations of Electron Transport in Gases..... 51
Radiation Physics and Dosimetry: Swarm Measurement of Cross Sections for Dissociative Electron Capture in Heavy Water, Chlorobenzene, and Bromobenzene..... 51
Radiation Physics and Dosimetry: Optical Emission from Argon Excited by Alpha Particles..... 52
Health Physics Research Reactor Studies..... 52
ISOTOPE DEVELOPMENT..... 52
Nuclear Analysis Research and Development..... 52
Recovery of Fission Products..... 52
Radioisotope Research and Development..... 53
Radioisotope and Stable Isotope Production..... 53
OPEN LITERATURE PUBLICATIONS..... 54
VISITORS..... 57

[redacted] [redacted]

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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

DECEMBER 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

GAS-COOLED REACTOR PROGRAM

Graphite Oxidation Experiment. — The engineering-scale test for studying problems associated with maintaining constant trace levels of CO and CO_2 in helium in a simulated reactor coolant system and for determining graphite oxidation reaction rates in the system was terminated after 833 hr of operation. Examination of the TSF-grade graphite test piece indicated that the surface exposed to the coolant was roughened along its entire length, with the highest temperature areas suffering the greatest amount of attack. Measurements indicate a weight loss of 0.747 g, which is equivalent to an average reaction rate of 1.1×10^{-5} mg cm^{-2} hr^{-1} per ppm (by volume) of CO_2. Temperature variations along the length of the 12-in. test specimen made it impractical to associate a specific temperature with the reaction rate; however, if it is assumed that the activation energy for this reaction is between 45 and 88 kcal/mole (extreme value found on the Dragon Project for a similar type of graphite), the weighted average temperature is between 1768 and 1778°F.

GCR-ORR Loop No. 2 Irradiations. — Irradiation of the second experimental assembly containing three 6-cm-diam graphite matrix spheres fueled with coated particles of uranium carbide was completed. The fuel was exposed at relatively low temperatures (~1700°F at the sphere center). Fission-gas release was low, with a maximum accumulation of 2000 $\mu\mu c/cm^3$ when the loop was operated without the cleanup system.

LITR Capsule Irradiations. — Two capsules containing pyrolytic-carbon-coated particles in each of four compartments were inserted in the LITR and are operating satisfactorily at indicated fuel central temperatures between 2000 and 2400°F. These capsules contain both uranium carbide and uranium-thorium carbide fuel mixtures supplied by General Atomic, 3M, and National Carbon.

ORR Poolside Capsule Irradiations. — Two capsules containing 6-cm-diam graphite spheres fueled with pyrolytic-carbon-coated uranium carbide particles were removed from the reactor for postirradiation examination. These capsules operated approximately at their design temperatures of 1800°F at the surface and 2500°F at the fuel center, except that the temperature of one capsule repeatedly increased from 2500 to 2800°F during reactor operating periods and returned to the lower temperature after each reactor shutdown. Two new capsules containing similar fueled spheres produced by potential fuel vendors have been installed for irradiation.

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Eight-Ball Experiment. — The third eight-ball experimental assembly containing 1-1/2-in.-diam spheres fueled with pyrolytic-carbon-coated uranium carbide fuel particles completed two cycles of operation and was removed. It has been replaced with a similar assembly designed to operate with sphere surface temperatures of 1800°F.

ETR Capsules. — Six stainless-steel-clad UO_2 capsules from the EGCR fuel irradiation program were removed from the reactor. Three capsules remain for high-burnup irradiations. The capsules removed had achieved accumulated burnups varying between 3400 and 11,500 Mwd per metric ton of UO_2.

Instantaneous Fission-Gas Release from Pyrolytic-Carbon-Coated Fuel Particles (B9-C1 Experiments). — The performance of GA fuel particles triplex coated with pyrolytic carbon was evaluated in two high-temperature experiments. Coated uranium carbide particles were irradiated at 2500°F to 20.6 at. % uranium burnup with no evidence of failed coatings, and coated uranium-thorium carbide (Th/U = 0.6) particles were irradiated at 2050°F to 14.7 at. % uranium burnup with no evidence of failed coatings. The temperature of the uranium-thorium fuel was periodically cycled during irradiation, both below the phase-transformation temperature (1800 to 2050°F) and through the phase-transformation temperature (2050 to 2400°F). The fission-gas release rates showed temperature dependence but no activity bursts. The fractional release rates from the uranium-thorium fuel were 5×10^{-6} at 2050°F and 10^{-5} at 2400°F. The fractional release rate from the UC_2 fuel at 2500°F was 10^{-5}.

Phase Relationships in Thorium Dicarbide. — High-temperature x-ray diffraction studies of ThC_2 yielded a single-phase pattern at 1600°C corresponding to a face-centered cubic structure with a lattice parameter a = 5.78 A. The phase reverted to monoclinic ThC_2 upon cooling to 1400°C, and the transformation was repeated several times by cycling the specimen between the two temperatures. Differential thermal analysis indicated that the transformation occurs at 1415 ± 10°C. Since x-ray patterns free from oxide lines have been attained, and the specimen is in contact with excess graphite, it appears fairly certain that the high-temperature ThC_2 phase and its transformation temperatures have been confirmed.

Self-Diffusion of Beryllium in Unirradiated Beryllium Oxide. — Diffusion of Be^7 through polycrystalline unirradiated specimens of BeO was studied as a preliminary step to a study of diffusion in irradiated material. Beryllium-7 was deposited on one end of cylindrical specimens by exchange and adsorption from aqueous solution. The depth of penetration perpendicular to the surfaces of deposition was determined after the specimens were annealed at definite, predetermined conditions. A simple lapping device of high precision was developed to determine penetration depths.

For the polycrystalline cold-pressed and sintered BeO studied, the diffusion behavior between 1150 and 1800°C (in cm^2/sec) is described by the empirical relation

$$D = 2.49 \times 10^{-3} \exp\left(-62.5 \times 10^3/RT\right) .$$

A diffusion model was proposed based on vacancy migration which suggests identical activation energies for diffusion in both a and c directions in single crystals and predicts an activation energy of the correct order of magnitude. Furthermore, the model predicts that the frequency factor in the direction perpendicular to the c axis should be about 50% higher than that parallel to the c axis.

MARITIME REACTOR PROGRAM

NS "Savannah" Filter Testing. — In-place tests on the reactor compartment ventilation system of the NS "Savannah" are conducted periodically by ORNL to determine whether there has been any significant deterioration in the iodine removal efficiency of the installed equipment. Such tests have been made on four occasions since the shipboard equipment was extensively revised in April 1963 to provide duplicate parallel systems. One system (F-1) has operated continuously since April with rated airflow through the filters; the other system (F-2) has been held in standby except for brief test periods. In the four series of tests, 25 separate efficiency determinations were made with test procedures based on measuring the penetration of iodine vapor introduced into the air stream ahead of the filters.

Overall results from these tests indicate that the reactor compartment ventilation system, as installed, is capable of achieving at least the iodine removal efficiency observed in a series of 14 laboratory tests of 11- by 11-in. Savannah-type charcoal units under accident conditions. These tests, conducted with continuous iodine injection at 96 to 100°C and with 80 to 90% saturated steam in air, have shown the efficiency of the charcoal unit alone to be 99.86 ± 0.07% at the 95% confidence level.

ARMY PACKAGE POWER REACTOR PROGRAM

PM Irradiation Test Program. — A holder containing 2 PM-1 reactor fuel tube sections and 24 flat-plate fuel coupons was removed from the pressurized-water loop at the Oak Ridge Research Reactor at the end of cycle No. 50 after 16 weeks of exposure. The fuel specimens operated satisfactorily during this period. During the intercycle period the upper portion of the holder was disassembled in the south hot cell of the ORR, and an advanced PM fuel tube was placed in the holder with the rest of the fuel specimens. During the time the holder was in the hot cell, the fuel specimens were examined visually. No signs of any radiation effects or any abnormal corrosion effects were noted. The assembly was returned to the A-1 position of the ORR, and at the same time the other pressurized-water loop position, A-2, was loaded with an assembly containing six advanced PM fuel tubes. The power generated by both fuel assemblies in the loop was observed as the reactor was taken to power. The full-power output of the combined experiment is 200 kw.

Corrosion Tests. — The 15 corrosion specimens on the A-2 suspension rod and the 3 corrosion specimens on the A-1 suspension rod were removed from the ORR pressurized-water loop at the end of cycle No. 50. The 12 Armco iron specimens exhibited significant corrosion effects, whereas the 6 stainless steel specimens showed no visual effects. These specimens are now being examined in detail.

Water Chemistry. — During the first six weeks of the irradiation of the PM fuel specimens in the ORR pressurized-water loop, four samples of suspended solids were filtered from the circulating water. The average concentration found was 100 ppb on the first day and 1 to 4 ppb during the rest of the period.

The initial specific activity of Fe^{59} (45-day half-life) was about a factor of 2 less than the estimated saturation specific activity under the maximum flux of the in-pile assembly, indicating that the iron in the solids came almost entirely from the walls of the in-pile section and not from the surfaces of the test fuel elements. A tenfold drop in this specific activity during the rest of the period indicated that the out-of-pile areas were contributing an increasing part of the greatly decreased amount of suspended iron.

The ratio of chromium to iron in the solids was initially about 1; this decreased during the period to 0.1. The specific activity of Cr^{51} (27.8-day half-life) remained within a threefold range during the entire period. This was only about 1% of the level calculated for saturation under the maximum flux. Thus it appears that the in-pile region contributed only a small part of the chromium found in the circulating solids.

APPLIED AND BASIC REACTOR PHYSICS

Basic Reactor Physics. — One of the parameters required for a fundamental understanding of neutron interactions in reactors is the neutron decay constant for the materials used in reactors over the range of temperatures under which reactors must operate. A common crystalline material used as a reactor moderator and reflector is beryllium, and attempts to measure its decay constant were made at ORNL some time ago. The measurements were complicated by the presence of "trapped" (i.e., long-lived) subpopulations of neutrons, however, which resulted in long-term changes in the decay constant. In fact, it could not be determined that an equilibrium condition describable by a decay constant would ever be reached. On the basis of available theoretical studies, it was hypothesized that the trapping effect was caused by the coherent scattering component of the transport cross section of beryllium, but this could not be established without additional experimental data. Strong evidence supporting this theory would be gained if it could be shown that the same effects do not occur in a crystalline material which has a relatively large incoherent scattering cross section. The results from recent experiments with light-water ice bodies should supply the necessary information.

The ice cylinders used in the experiments ranged in size from a minimum of 2-1/4 in. in diameter by 2-1/2 in. high to a maximum of 18 in. in diameter and height. A pulsed-neutron technique was employed, and the neutron decay was observed as a function of waiting time after the neutron burst.

All data have been taken, but the analysis is not yet complete.

REACTOR FUELS AND MATERIALS

Behavior of High-Temperature Materials Under Irradiation. — The status of the high-temperature materials irradiation program is summarized below:

Samples	Temperature (°C)	Dose (neutrons/cm^2)	Status
MgO, Al_2O_3, BeO, and pyrolytic graphite	200	~1×10^{21}	Irradiation complete
MgO, Al_2O_3, BeO	200	~2×10^{21}	Irradiation complete
MgO, Al_2O_3, BeO, and single crystals	800 and 1100	~6×10^{21}	Irradiation in progress
MgO, Al_2O_3, BeO, and single crystals	800 and 1100	~3×10^{21}	Assembled and shipped to ETR

Dosimetry analyses for BeO irradiation experiments 41-8, 41-9, and 41-10 were completed. The maximum fast-neutron dose in experiment 41-8, which is now being disassembled in the ORNL hot cells, was 8.2×10^{21} neutrons/cm^2; the maximum fast flux was 5.5×10^{14} neutrons cm^{-2} sec^{-1}.

FUNDAMENTAL HEAT TRANSFER AND FLUID DYNAMICS

Boundary Layer Transient Phenomena. — Work continued on the installation of apparatus for studying heat transfer. The Chromalox heaters were satisfactorily brazed to the solid stainless steel test section, the "gun-barrel" thermocouples for instantaneous surface-temperature measurement and the thermal-flux probes were installed, and the inside wall of the test section was carefully honed and polished. Vacuum deposition of a thin film of nickel to complete the thermocouple junction at the inner surface is the only remaining step in fabrication.

Determinations of the instantaneous film coefficient of heat transfer inside a 2-in.-ID pipe were made with the hot-film probe and constant-temperature circuit previously described (ORNL-3402, p. 6). Water was

supplied by the 200-gpm vacuum deaerator recently put into operation. With the hot-film probe it was possible to detect the onset of turbulence, which appeared as intermittent fluctuations in the instantaneous film coefficient at a Reynolds modulus of about 2800. At higher values of the Reynolds modulus, the characteristic pattern of continuous random fluctuations was observed, the total amplitude of which was frequently as high as 20% of the mean film coefficient.

Boiling-Potassium Studies. — A 20- by 20-in. natural-circulation loop was operated to obtain a quantitative description of the superheat phenomenon in potassium that is believed to be a cause of the large-amplitude thermal oscillations observed in previous boiling heat-transfer experiments. The loop was constructed of 1/2-in. sched-40 stainless steel pipe, which was radiantly heated. It accommodated test sections consisting of an as-received commercial surface and a surface obtained by coating the inside of the pipe with an 8- to 20-mil-thick layer of sintered stainless steel particles. The particles were about 45 μ in average diameter and formed pores varying in radius from about 10^{-4} to 6×10^{-4} in. During operation in the range of heat flux from 16,000 to 37,000 Btu hr^{-1} ft^{-2}, it was found that, with the smooth, as-received commercial surface, the tube wall temperature fluctuated continuously with a total amplitude of about 140°F at a frequency of about 1.5 cycles/min. The corresponding superheat (uncorrected for wall temperature drop) was about 210 to 240°F. In contrast with this behavior, the porous surface exhibited occasional large-amplitude transients, associated with which were uncorrected superheats of 140 to 180°F. These transients were separated by periods believed to correspond to stable local boiling with low-amplitude relatively-high-frequency thermal oscillations and superheats in the region of 25 to 30°F, consistent with predictions based on the thermodynamics of bubble formation and the estimated pore size of the sintered coating. Occasional periods of higher amplitude and lower frequency fluctuations were also observed. The average cavity size for the smooth, as-received surface, as estimated from the superheat data, is about 9×10^{-6} in.

Boiling-Water Studies. — Twenty-one series of swirl-flow tests were conducted in which all the heating took place in the twisted-tape swirl generator. This is in contrast to past ORNL swirl-flow tests, in which approximately 99% of the heat was generated in the tube wall. In the subject study, water from a constant-head tank flowed by gravity through a 0.27-in.-ID glass tube approximately 12 in. long, in which was positioned a resistance-heated 15-mil-thick "A"-nickel tape. Various twist ratios ($y = 2.7$ to ∞ tape widths per 180° twist), heat fluxes (0.2×10^6 to 1.2×10^6 Btu hr^{-1} ft^{-2}), and inlet water temperatures (63 to 173°F) were used. The present results indicate that critical heat fluxes for twisted tapes fall between 93 and 122% of those for flat tapes. The power density of a swirl-flow tube may therefore be significantly increased by generating heat in the twisted tape as well as in the tube wall. Further increases in power density may be realized by cooling the outside wall with coolant flowing axially through an annular channel.

POWER REACTOR FUEL PROCESSING

Chemical Analysis of Advanced Reactor Fuels. — The electrochemical behavior of some of the halides may be elucidated by voltammetric investigation with the pyrolytic-graphite electrode. The iodine system has been studied as an example. Iodide concentration can be followed by the anodic oxidation to iodine, and the iodine concentration by the converse process. Iodate may also be determined from the reduction current. The half-wave potentials for these systems were determined, and other parameters affecting the electrochemical behavior of these ions were also studied. The voltammetric method of determination is extremely sensitive and does not rely on the solubility product as a governing principle, such as is the case with the silver-ion titration commonly used for the determination of halide.

Coulometry with a rotating-disk pyrolytic-graphite electrode is of interest in the analysis of aqueous solutions of reactor fuels and in the analysis of molten salts. A rugged rotating-disk electrode has been built and is now being evaluated. We are also planning to investigate the use of the rotating-disk electrode as a means of increasing the sensitivity of the voltammetric methods.

Uranium Carbide Hydrolysis. — The recently discovered "passivity" of irradiated uranium monocarbide toward hydrolysis in water at 80 to 100°C has prompted the evaluation of high-temperature steam combustion as a means of converting the uranium monocarbide to an oxide readily soluble in nitric acid. A preliminary step in such a process for chopped, stainless-steel-clad, sodium-bonded carbide fuel is low-temperature steaming to convert the sodium to relatively harmless sodium hydroxide. The process is also designed to remove the carbon as oxides before dissolution of the resulting uranium oxide in nitric acid, in contrast to the alternative process of direct dissolution of uranium monocarbide in nitric acid, which generates soluble organic acids. The latter are troublesome and difficult to destroy or remove.

Laboratory studies of the reaction of steam with unirradiated samples of stoichiometric monocarbide at 650 to 800°C in the absence of air shows that consistently high purity UO_2 may be produced at a controlled rate. With 5-g batches of 5-mm fragments of the monocarbide, complete conversion to UO_2 is accomplished in 45 min at 750°C in steam flowing at 0.45 g/min. Oxygen-to-uranium ratios in the oxide products varied from 2.03 to 2.07, with a maximum of 0.7% of the original carbon remaining.

Off-gas analyses indicate that at least two consecutive reactions occur. The uranium carbide is initially converted by steam to uranium dioxide and free carbon, with the evolution of hydrogen. The free carbon then reacts with additional steam to produce carbon dioxide, carbon monoxide, and hydrogen. In the overall reaction, about 5 moles of off-gas is produced per mole of uranium carbide consumed.

Five experiments on high-temperature steam combustion have been conducted in the hot cell with uranium monocarbide irradiated to 7500 and 16,000 Mwd/metric ton. The 4- to 7-g specimens were steamed with air present for about 5 hr at 700 to 800°C. Air could not be excluded from

the reactor, and, consequently, the higher oxide (U_3O_8) was produced. The condensed steam from the off-gas contained from 1.1% (700°C) to 42% (800°C) of the total cesium in the irradiated fuel samples. Cesium was the principal fission product volatilized.

Dissolution of the resulting U_3O_8 in nitric acid was complete in 30 min, producing a uranyl nitrate–nitric acid solution and a small (unweighable) carbonaceous residue. The residual carbon content of the oxide product could not be determined due to lack of special analytical facilities.

Solvent extraction performance with this feed, which contained some carbonaceous residue, was highly satisfactory under simulated Purex process conditions. In exhaustive batch extractions with 30% tributyl phosphate in Adakane, and subsequent batch strippings of the solvent with 0.01 M HNO_3, recovery of uranium and plutonium was virtually quantitative. Normal phase-coalescence times of 20 to 30 sec were observed.

Protactinium Adsorption. – Processes have been investigated for the separation of protactinium from Th-U^{233} fuel solutions in order to preclude long delay times or to avoid rerunning the protactinium-containing waste streams to effect complete recovery of the U^{233}. Laboratory studies showed that unfired Vycor, a leached borosilicate glass manufactured by Corning Glass Works, will preferentially adsorb protactinium from simulated acid Thorex dissolver solutions. The effect of several solution variables has been determined by measuring the total amount of protactinium which can be adsorbed and its distribution with 60- to 80-mesh unfired Vycor in Thorex dissolver solutions that contained 100 g of thorium per liter and 1 million counts of Pa^{233} per minute per milliliter. These solutions were 0.5 to 12 M in HNO_3, 0 to 0.04 M in HF, and 0 to 0.4 M in aluminum. The distribution coefficient (DC) varied directly with the nitric acid concentration in solutions containing only thorium nitrate–nitric acid and could be calculated from the equation

$$DC = (259 \pm 5)\ M\ HNO_3 + 233 \pm 36\ .$$

The addition of fluoride produced little change in the distribution coefficients in solutions containing less than 4 to 5 M HNO_3. At higher acidities the distribution decreased as the fluoride or the acid concentration increased. This decrease could be partially canceled by the addition of aluminum to complex the fluoride. For example, coefficients were 1500, 2300, and 2800, respectively, in 5, 8, and 10 M HNO_3 without fluoride; 1200, 800, and 300 with 0.04 M HF present; and 1350, 1400, and 1500 with 0.04 M HF, 0.4 M Al present.

The amount of unadsorbable protactinium in thorium nitrate–nitric acid solutions decreased from about 4% in 0.5 M HNO_3 to 1% in 12 M HNO_3. The addition of fluoride produced little change when the acid was below 4 M HNO_3. At higher acidities the percent unadsorbable decreased with increasing fluoride to about 0% in 10 M HNO_3, and the addition of 10 moles of aluminum per mole of fluoride eliminated this fluoride effect.

Thorex Solvent Extraction. – Uranium, thorium, and 90% of the protactinium were extracted from a solution obtained from dissolution of ThO_2 pellets which had been irradiated to produce protactinium tracer. Less

than 5% of the fission products with high neutron cross sections remained with the solvent-extraction product solution. In this experiment, the ThO_2 was dissolved in 14 M HNO_3, 0.04 M NaF, and 0.1 M Al to give 100 g of thorium per liter, and then irradiated U^{235} was added to provide fission product tracer. Excess acid was removed by evaporating to a thorium concentration of 250 g/liter and then diluting to 100 g/liter. Solvent-extraction flowsheet conditions for laboratory countercurrent batch extraction experiments were:

Feed: Thorium, 100 g/liter; uranium, 4.48 g/liter; 4.4 M in HNO_3; 1 vol

Scrub: 4 M HNO_3, 0.1 M $Al(NO_3)_3$; 0.7 vol

Extractant: 30% tributyl phosphate in n-dodecane; 5 vol

Stages: five extraction, three scrub

The decontamination factors were 25, 4000, 40, 2, and 1.5, respectively, from ruthenium, rare-earth elements, niobium, zirconium, and gross gamma. Product recovery was the same, but decontamination was less with a scrub solution 1 M in HNO_3 and 0.5 M in $Al(NO_3)_3$.

Uranium Adsorption. — Scouting tests were made in order to explore the feasibility of recovering uranium from nitrate solutions by direct sorption on sol-gel thoria. Thoria powder sorbed 0.5 to 2.9 mg of uranium per gram of ThO_2 from thorium nitrate—uranium nitrate solutions partially neutralized with ammonium hydroxide. These solutions contained 80 mg of thorium per ml and 4 mg of uranium per ml, at a pH of 2.58 to 3.85, and were contacted for 24 hr with a thorium oxide slurry at a concentration of 100 g/liter. Although the amounts adsorbed were too low for present applications, the principle remains an interesting possibility.

An analytical procedure was developed to determine "free" fluoride in a nitrate-fluoride system containing aluminum or niobium. The analysis is based on a conductimetric titration. Analyses indicate that four fluorides are "combined" with each niobium, and two fluorides with each aluminum. Solvent extraction experiments showed that only the "free" fluoride effects uranium extraction by tributyl phosphate or dibutyl butyl phosphonate. By correcting the total fluoride concentration for that "combined" with the complexing agents, the equations previously used in calculating distribution coefficients in the nitric acid—hydrofluoric acid system may also be used for systems containing aluminum or niobium.

Shear-Leach Process. — Ceramic—stainless steel reactor fuels may be sheared to 1/2- to 2-in. lengths into a perforated basket, which is suspended in a batch dissolver containing recirculating acid for leaching ThO_2-UO_2 or UO_2. Variables being studied for their effect upon leaching rate are basket design, sheared length of fuel, and air sparging. Refractory sol-gel-derived ThO_2-UO_2 is being studied in batch leachings by the "heel" technique to determine heel size at steady state.

The shortest ThO_2-UO_2 leaching time observed to date was 6.7 hr for 99.9% recovery, for 1/2-in. sheared lengths in a 3-in.-diam, 6-in.-high basket having 0.015- by 0.343-in. slots; agitation was by air sparging at 0.3 to 1.4 scfm/ft^2. In comparison, a range of recovery times of about 9 to 13 hr was obtained for a variety of basket designs and sheared lengths, under sparged and nonsparged conditions.

Packing densities of sheared stainless-steel-clad UO_2 pieces (1/2-in. OD) for 1-, 1-1/2-, and 2-in. lengths in a 6-in.-diam cylinder were 4.77, 4.70, and 4.30 g/cm^3 respectively. Corresponding void fractions were calculated to be 0.50, 0.50, and 0.55.

Eight successive 20-hr batch dissolutions of representative sol-gel ThO_2-UO_2 resulted in dissolution of 85, 95, 99, 101, 96, 98, 101, and 102%, respectively, of the 1400 g of fuel charged each time. A maximum ThO_2-UO_2 heel of 25% (356 g) occurred at the end of the sixth cycle. Product solution was nominally 0.5 M Th–0.015 M U–10 M HNO_3–0.04 M F^-–0.10 M Al.

Batch leaching of sheared lengths of stainless steel tubing loaded with sol-gel-derived ThO_2-UO_2 showed a pronounced advantage of 1/2-in. cuts over 1-in. cuts. Leach times for 99.9% core dissolutions were 32 and 51 hr respectively.

Stainless steel equipment corrosion rate for eight sol-gel dissolutions was 2.9 ± 0.5 mils/month.

A bundle of 156 loose porcelain-filled Consolidated Edison core B tubes (0.300-in. OD × 0.011-in. wall × 12 in. long) inserted in the Mark I envelope (3-3/4 in. square) was sheared, and the resulting terminal pieces were ≤1-1/2 in. long; however, in each of three tests, one tube varying in length from 3 to 5 in. was bypassed by the pusher head after the pusher head had entered the outer gag.

An ORNL Mark I prototype fuel assembly filled with porcelain was sheared into 1/2-in. lengths to measure the temperature rise of the moving blade of the shear. Continuous (automatic) shearing at 4 strokes/min and a speed of 1.2 in./sec occurred over a period of about 38 min for a total of 164 cuts. A thermocouple placed between the moving blade and the blade holder registered an increase in blade temperature from 29°C to 62°C within 10.5 min (50 cuts). The temperature remained at about 62°C for 3 min (12 cuts), followed by a gradual decrease in temperature to about 50°C during the next 14 min (63 cuts), with no further change in temperature for the remainder of the shearing (10.5 min, 39 cuts). It was concluded that overheating of the moving blade is not possible at the shear speed tested.

Corrosion. – Corrosion tests were performed that were designed to reproduce the conditions expected in a reactor in which graphite-UC fuel is oxidized and the "ashes" are chlorinated to remove volatile chlorides from the uranium chloride. In this process the uranium chloride is dissolved in an aqueous acid.

Welded specimens of "A" nickel, Nichrome V, and Corronel 230 were corrosion tested in a cycle consisting of (1) air saturated (at room temperature) with carbon tetrachloride at 600°C for 5 hr, (2) oxygen at 800°C for 5 hr, and (3) 5 M HNO_3 at room temperature for 1 hr. The three alloys corroded at maximum rates of 72.8 mils/month (3 cycles), 104

mils/month (1 cycle), and 256 mils/month (2 cycles) respectively. These rates are 3 to 10 times higher than those previously observed in a cycle in which the O_2 exposure preceded the air-CCl_4 part of the cycle.

In cyclic tests consisting of exposure to O_2 at 800°C for 5 hr, exposure to N_2-CCl_4 at 500°C for 5 hr, and ash dissolution in 6 M HCl at room temperature for 15 min, welded specimens of Corronel 230, Nichrome V, Hastelloy B, Inconel, and Hastelloy N corroded at maximum rates, for 3 cycles, of 22.1, 23.1, 46.6, 18.3, and 23.6 mils/month respectively.

No construction material has yet been found which will satisfactorily contain any of these three reprocessing cycles.

THORIUM UTILIZATION PROGRAM

U^{233} Purification by Solvent Extraction. — An additional 5 kg of U^{233} for use in fabricating fuel rods for zero-power criticality experiments at Brookhaven National Laboratory was purified by solvent extraction from nitrate solution with di(sec-butyl)phenylphosphonate in diethylbenzene. The uranyl nitrate product met all sol-gel specifications: U^{233} concentration, 114.4 g/liter (specification, 100 to 125); gross gamma decontamination factor, 300 (100); thorium decontamination factor, 3×10^3 (1×10^3); NO_3/U ratio, 2.20 (less than 2.5); aluminum concentration, ppm based on U^{233}, 26 (less than 30). The total U^{233} loss in the extraction column raffinate was 0.14% of that in the feed.

A total of 38 kg of U^{233} has been purified to date: 25 kg for use in sol-gel operations and 13 kg for fabrication into pellets for use in the Large Power Reactor Physics U^{233} Program at Bettis Atomic Power Division, Westinghouse Electric Corporation. Part of the 13 kg will be used to fabricate ThO_2—1 wt % $U^{233}O_2$ blanket pellets, and part to fabricate ZrO_2—26 wt % $U^{233}O_2$ seed pellets to be clad in Zircaloy-2.

Thoria Preparation. — Sufficient ThO_2 has been prepared by the pyrohydrolytic denitration of thorium nitrate at 500°C to complete the Brookhaven National Laboratory Kilorod program. The denitrator operations will continue to prepare additional sol-gel-grade ThO_2 for other customers.

The only significant equipment failure so far in the thoria preparation occurred during this report period. A failure of the steam supply caused the steam superheater to burn out. Operations will be resumed when a replacement is received.

Mixed Thoria-Urania Preparation in the Fuel-Rod Facility. — Five hundred seventy kilograms of mixed ThO_2-$U^{233}O_2$ was prepared for use in fabricating fuel rods for the zero-power criticality experiments at Brookhaven National Laboratory. Equipment performance was satisfactory, yielding an on-stream factor of 97% for the past six months. The only significant equipment failures were the two reported in October on the calciner elements.

Close examination of the element installed as a replacement and which failed immediately showed that the failure occurred from fracture rather than burnout. This indicates poor fabrication. The heating element now in the calciner has performed satisfactorily through seven cycles.

Rod Fabrication. – On November 15, 1963, the 480th rod passed all inspections, thus completing the rods for the fourth shipment to Brookhaven National Laboratory.

The system was shut down (after processing sol-gel batch 53) in order to perform the required criticality cleanup and to evaluate the performance of the facility over campaign 4. The material balance at the end of the campaign again showed a net loss of powder of 0.15%. The operation over the campaign was routine, with no major equipment failures.

The facility has produced 570 rods to date – 138 rods in this reporting period. Ninety rods are ready in the fifth shipment to BNL.

Pellet Fabrication. – Fifty fuel rods made with pellets are to be fabricated for use in flux distribution measurements during the zero-power criticality tests of sol-gel ThO_2-$U^{233}O_2$ at Brookhaven National Laboratory. Two pellet fabrication methods are being studied: one based on wet blending ThO_2 and U_3O_8 and the other based on direct fabrication of pellets from sol-gel material. In studies using sol-gel material, it has been found that the standard technique of cold forming in steel dies followed by sintering is not well suited for use with powders prepared from this material. It can be done, but the technique required is not practical for remote fabrication. Efforts are being concentrated now on a simplified fabrication technique employing isostatic pressing. Initial tests show considerable promise from the points of simplicity of technique and the elimination of cracking and laminating during sintering. To date, the densities obtained by this technique have not been higher than those obtainable with the cold-press and sinter technique (maximum of 9.2 g/cm^3).

Thorium-Uranium Fuel Cycle Development Facility (TUFCDF). – Design of the TUFCDF, a facility to be used in the development of processes for remotely processing and fabricating Th-U^{233} fuel assemblies, has been in progress since January 3, 1963.

The title I design effort is complete, and the bound report was issued on December 16. The total estimated cost is $6,682,000, leaving a contingency of $593,000 within a project budget of $7,275,000. Title II design is scheduled for completion in November 1964, and facility construction is scheduled for completion in December 1966. A status review was held for the benefit of local and Washington AEC personnel responsible for guidance of the program. It was indicated that authority would probably be granted soon to proceed with title II design, using funds already available. Design of components for test in a facility mockup, and scheduling of fabrication and test of these items, are proceeding. The items include a 3-in. vertical-tube furnace, for which a requisition has been issued; a continuous ball mill, which is being tested; vibratory compaction equipment; a gamma-ray scanner for determining fuel-rod-density variations; and a fuel-rod cleaning and transfer device.

Sol-Gel Microsphere Preparation. – Procedures for preparing sol droplets of controlled diameter (particularly about 500 μ) in a drying solvent were tested. Sol discharged into the drying solvent through nozzles broke up by several mechanisms, depending on the sol properties and flow rates, the nozzle dimensions, and the properties and agitation of the drying solvent. Conditions were determined for successful fluidization and drying of sol drops to give thoria, thoria-urania, or thoria-urania-carbon

gel microspheres 50 to 1000 μ in diameter. Different additives are necessary to prevent coalescence, glass wetting, or flocculation, depending on the composition of the sol and the diameter of the drop.

Irradiation of Thorium Oxide Fuels. – All the MTR-III and ETR-I vibratorily packed sol-gel ThO_2-UO_2 rods have now been inserted in the reactors and are operating successfully. Postirradiation examination of the two high-burnup ThO_2–4.4% UO_2 pellet rods irradiated in the MTR indicated excellent compatibility between fuel and clad, with no marked changes in microstructure.

Operational problems still prevent making meaningful measurements on the capsules containing Th-U-Zr alloy.

Permeability of Vibratorily Compacted Fuels. – The permeability of Kilorod fuels and the relationship of this permeability to failure through waterlogging are under study. Early results on the permeabilities of compacts indicate that the values observed for room-temperature water are much lower than those for the same compact when nonpolar liquids and/or gases are used. This will be studied at higher temperatures. Sufficient results are not yet available to correlate the associated effects of surface area and theoretical density on the permeability of the compact.

Preparation and Properties of U(IV) Sols and UO_2. – Development of a sol-gel process for the production of dense urania fuels from uranyl nitrate solutions would significantly extend the usefulness of the sol-gel technique. Studies are in progress on the preparation of U(IV) sols by electrolytic reduction and dialysis and by precipitation and subsequent dispersion by peptization of the hydrous oxide. The electrolytic work has been carried out in a two-compartment cell with a platinum anode and a mercury cathode, the compartments being separated by a cation exchange membrane. During the past month, further studies were carried out on U(IV) sols stabilized by formate, acetate, and formate-acetate. Sufficiently large amounts of sol were prepared to make possible evaluation of fired products.

In the formate-stabilized sols, uranium concentrations ranged from about 0.036 to 0.16 M, and pH's ranged from 3.35 to 4.02. Although large fragments of nearly theoretically dense UO_2 were obtained upon firing in a reducing atmosphere, the required formate concentrations for good sol formation were generally about the same as the uranium concentrations. It is desirable to find a system requiring less stabilizing ion per mole of uranium sol. For example, it should be noted that in the nitrate-stabilized ThO_2 sol system the nitrate-to-ThO_2 ratio is less than 0.1.

Acetate-stabilized sols of 0.06 to nearly 0.08 M were prepared, and one was fired in a reducing atmosphere to produce UO_2 of medium gloss; the particles were about 1 mm in diameter. Acetate concentrations were about half the uranium sol concentrations or less. The sols broke and precipitated when reduced in volume by evaporation, even when nitrate was added prior to evaporation. These sols and final oxide products showed no advantages over formate-stabilized sols. The formate-acetate-stabilized sols were no better.

In the precipitation work, a stable sol prepared last month (0.4 M in uranium, with nitrate/uranium and ammonium/uranium mole ratios of 0.03

and 0.06 respectively), which had been dried to a gel by heating in argon at 95 to 110°C, was calcined to UO_2 in a reducing atmosphere. The final product was composed of several shiny, blue-black fragments with sponge-like surfaces and a few cracks. Fragment edges were rounded, rather than jagged or sharp. The spongy appearance was attributed to voids produced by the steam formed while drying at 110°C. After being crushed and passed through a 30-mesh screen, the density of the oxide determined by toluene displacement was a very encouraging 10.80 g/cc, or about 98.5% of the theoretical value (10.96 g/cc).

Sol-Gel Preparation and Properties of ThC_2 and UC_2-ThC_2. — Spheroids of ThO_2-carbon and $(Th,U)O_2$-carbon gels were prepared by methods completely parallel to those used for making $(Th,U)O_2$ gels. Efforts are being made to confirm a tentative flowsheet for a process for making ThC_2 and ThC_2-UC_2 microspheres.

A method of preparing carbon-containing sols that can consistently be formed into spheres has not yet been found. The problem appears to be in the variability of the carbon blacks used. There was also evidence of nonuniform carbon distribution in several carbide products, probably because of nonuniform distribution of carbon in the original sol. In a series of firings of gel microspheres to form $(U,Th)C_2$, some sintering was found upon firing at 2200°C, and incomplete conversion to carbide was observed. Excessively high firing temperature probably caused sintering, and too short a reaction time led to the incomplete conversion.

Physical and Chemical Properties of Molten Chlorides. — It was previously reported here that $AlCl_3$ and $ThCl_4$ apparently do not show mutual solubility when contacted for short times in the proportions 0.9 to 0.1 mole fraction. These results were obtained by observing salt mixtures in 1-mm-ID quartz capsules, heated in a fused-salt bath. The materials were viewed directly as the temperature was varied from 140 to 350°C. Additional observations were made by using thermal analysis and differential thermal analysis. When heating and cooling these mixtures, it was noted that temperature halts occurred and that they corresponded approximately to the normal melting points of the individual salts. No other halts are noteworthy. Apparently very little mutual solubility occurs, or whatever solubility might have occurred was not observable as a depression in the melting point during the time period of the experiment.

MOLTEN-SALT REACTOR PROGRAM

Molten-Salt Reactor Experiment. — Installation of the 5-in.-OD coolant-salt piping was completed in both the coolant and reactor cells. The graphite for the MSRE core was received, and the core assembly was 80% complete. The H. K. Ferguson Company completed the pulling of electrical and thermocouple cables and started the connection of these cables to terminals in the instrument panels. The Ferguson portion of the work is ~65% complete. The overall project was ~89% complete on December 1, 1963.

Development of Components and Systems. — Testing of the modified 1-1/2-in. freeze valves with the reactor-grade module control system and thermocouples was completed. Freeze times from an initial valve temperature of 1200°F were about 30 min; melt times were from 5 to 12 min. It was demonstrated that, for a power failure condition, the heat capacity of the valve heater box was sufficient to melt out the frozen valve and hold the pipe temperature above 850°F for 45 min.

Testing of the drain-tank heater unit was continued after a burned-out heater lead wire was repaired. Faulty welding of the electrical junction was the probable cause of failure, and a revised inspection method was used after the repairs. The INOR-8 tubes of the drain-tank cooler test were operated through a total of 159 thermal shock cycles from 1300 to 200°F. At the completion of each 50 cycles, the tubes were examined by dye check, visual, and pressure methods. No damage to the 1/2-in. tubes or 1-in. pipe was noted. Test is continuing.

Testing of the Vard Corporation control-rod drive was continued with a brass worm wheel replacing the bronze gear which had failed. The front bearing of the drive motor failed after 23,900 full-stroke cycles, and the motor was replaced.

The oxygen removal unit of the helium purification system was modified to improve reliability and then returned to test. It was found that, when operated at a temperature of 1160°F and a 10-liter/min inlet flow of helium containing 220 ppm of oxygen, the outlet concentration was reduced to less than 0.1 ppm of oxygen through 120 hr of operation.

The engineering test loop continued in operation for testing of the sampler-enricher mockup. A total of five additions of uranium-lithium fluoride have been made, increasing the uranium concentration from 0.1 mole % to approximately 0.15 mole %. Uranium-bearing salt has circulated through the graphite in the loop for a total of 5300 hr, and 30 samples have been removed routinely through the sampler-enricher mockup. These samples were processed through the analytical hot-cell facility mockup for the development of analytical processing.

The portable maintenance shield was received, and a typical assembly was set up to try out all components. Lubrication provisions, wiring, limit switches, and hand tools for manual operation were added or requisitioned. The time required for this first partial assembly indicated that setting up this equipment will take considerably less time than the original estimate of four men for 4 hr.

Detailed procedures for the removal of drain-cell components were prepared, and, as a result, the design of devices for handling spool pieces of various sizes and shapes was started. An adjustable, portable television camera stand was designed and is being built for testing with the television equipment from the MSR maintenance demonstration facility.

MSRE Pump Program. — The MSRE prototype fuel pump was operated for 400 hr, circulating molten salt $LiF-BeF_2-ZrF_4-ThF_4-UF_4$ (66.4-27.3-4.7-0.9-0.7 mole %) at temperatures of 1100 to 1300°F.

The MSRE fuel-pump lubrication package was proof-tested in conjunction with the operation of the prototype pump and was delivered to the MSRE. The coolant-pump lubrication package is undergoing the same proof-testing.

Bench tests indicate that the individual fuel-pump supports can provide the desired flexibility. A final proof test of the supports will be made in the test facility for the prototype fuel pump.

Dimensional inspection after final machining was completed on one of the vessels for the MSRE pump drive motors prior to final leak testing. Cooling coils were attached to two other vessels and will be attached to the fourth vessel.

Instrumentation Development. — A device has been developed which permits the insertion of a test signal into the input of reactor temperature safety instrument channels without disconnecting wires and which permits the required isolation of circuits to be maintained. The device consists of a heated thermocouple assembly of the type used to measure rf current, and an associated isolation transformer and push-button switch. Closing the switch applies power to the heater, thereby heating the thermocouple and causing an emf to be developed in series with the thermocouple. Either temperature increase or decrease can be simulated, depending on the polarity of connection. Tests to date indicate that the device will not insert errors when not in use and will have a satisfactory response.

Radiation tests are presently being performed on a multiconductor thermocouple cable and disconnect assembly of the type being installed in the MSRE reactor and drain cells. The assembly is being irradiated with 10^6 r/hr of gamma radiation from a Co^{60} source. There was no measurable change in insulation resistance or thermocouple signal after two weeks of exposure; however, pressure began building up inside the cable sheath as soon as exposure began, indicating that materials within the sheath are off-gassing. Gas samples have been taken for analysis.

NUCLEAR SAFETY

Fuel Perturbation Effects in Pool-Type Reactors. — One of the various mechanisms affecting the reactivity of a reactor during a power transient is the motion of fuel, such as that due to buckling of fuel plates from pressure or temperature gradients. The magnitudes and directions of these effects are in general unknown. In order to investigate these effects for a pool-type reactor, a program was initiated in which perturbations in experiments were to be simulated by adding and removing fuel in a reactor core in such a way as to keep the total amount of fuel in the core constant. Measured reactivity effects would be compared with those determined by calculations. In the initial experiments with the Bulk Shielding Reactor I, the measurements indicated that negative reactivities resulted from all perturbations that decreased the homogeneity of the core except for those perturbations that added fuel to high-importance regions, such as those adjacent to interior water volumes. However, not enough experimental evidence was obtained to establish this supposition as a conclusion, and further, the calculational methods used could not cope with the small-dimension, large-magnitude perturbations represented by the experiments. The only codes available which adequately treat the transport of neutrons across region boundaries are one-dimensional codes that consider variations in fluxes or reactor composition

in one direction only. Therefore it became obvious that experiments and calculations could be correlated only by performing a "one-dimensional" experiment, that is, using a parallelepipedal critical assembly which is effectively infinite in two dimensions and which varies in composition in the third dimension only.

Initial critical experiments have now been performed to set the parameters of the "one-dimensional" assembly. It consists of U-Al fuel elements (10 wt % U, 93% U^{235}), 23.6 in. high, 24 in. wide, and 0.058 in. thick, spaced in water 200 mils apart center to center to a total slab thickness of approximately 6 in. The assembly is water reflected on the 23.6- by 24-in. faces only, thus fulfilling the requirement that variations in composition be in one direction only. Plastic strips between the elements prevent a shifting of fuel during each experiment. The assembly is now ready for the perturbation experiments in which the fuel position within the slab will be varied.

Prompt-Neutron Lifetime in Critical Enriched-Uranium Metal Cylinders. — In the design of reactors, especially fast or unmoderated reactors, the kinetics, or time dependence of fission rate, must be well known if safe reactor operation is to be effected. A parameter basic to this problem is the lifetime of neutrons produced directly in fission, or prompt neutrons, in the reactor core.

In the present measurement the Rossi-α technique has been employed to measure the prompt-neutron decay constant of five unreflected and unmoderated delayed-critical cylinders of enriched uranium metal (93.15 wt % U^{235}), ranging in height from 7 to 15 in. and in height-to-diameter ratios from 0.2 to 0.7. The prompt-neutron lifetime was computed from the measured prompt-neutron decay constant and the effective delayed-neutron fraction calculated by transport theory.

The value of the prompt-neutron lifetime obtained in these measurements, $(6.25 \pm 0.04) \times 10^{-9}$ sec, is independent of the critical cylinder dimensions (within the experimental error) and is in agreement with the experimental value obtained at Los Alamos in work with the Godiva reactor [$6.24 \pm 0.11) \times 10^{-9}$ sec]. A calculation of the prompt-neutron lifetimes based upon the relation given by Gross and Marable [Nucl. Sci. Eng. 7, 281 (1960)] gave slightly lower values, ranging from 5.43×10^{-9} to 5.50×10^{-9} sec. The disagreement is comparable to the disagreement between measured and computed values of the decay constant for uranium spheres given in the literature.

Melting Experiments in TREAT Facility. — Two experiments have been performed in the TREAT reactor to study the release of fission products from fuel melted under transient heating conditions. Previously unirradiated, stainless-steel-clad UO_2 specimens were heated in an argon atmosphere in both experiments. A description of the experimental assembly and preliminary results of the experiments were given in previous progress reports (ORNL-3483, -3508, and -3532). Determinations of the release of fission products have been completed for both experiments, and total release values are as follows:

Experiment No.		Release (%)[a]								
		Sr^{89} (Br+Kr)	Cs^{137} (I+Xe)	Ba^{140} (Xe+Cs)	I^{131} (Sn+Sb)	Te^{129} (Sn+Sb)	Ce^{144} (Ba+La)	Zr^{95} (Rb+Sr)	Ru^{103} (Tc)	UO_2
1[b]	Total release from UO_2	23	26	8.2	23[d]	21	7.0	0.21	0.10	0.005
	Release from high-temperature zone	19	16	2.4	1.1[d]	0.3	0.05	0.09	0.04	0.0002
2[c]	Total release from UO_2	58	53	42	30	21	8.8	1.5	0.60	0.75
	Release from high-temperature zone	46	38	15	1.1	1.3	0.32	0.13	0.06	0.06

[a]The isotopes listed were radiochemically analyzed after approximately 50 days of decay. The elements in parentheses were the principal precursors present during the first few seconds after transient.

[b]Reactor integrated power, 320 Mw; period, 108 msec.

[c]Reactor integrated power, 328 Mw; period, 87 msec.

[d]Iodine-131 released in experiment 1 is based on the calculated amount formed. All other release values are based on the total amount found by radiochemical analysis.

These data were obtained with essentially zero burnup fuel, and they show the extent of release of fission products formed during the reactor transient. These experiments thus simulated the type of transient power excursion with unirradiated fuel that could occur during startup of a new reactor or in SPERT-type experiments. The release of fission products from fuel with significant burnup would be quite different from the behavior shown in the tabulation, and experiments will be performed with previously irradiated fuel when facilities become available for handling the high-activity materials.

The comparatively greater release from the fuel in experiment 2 was apparently due to the more rapid rate of heating, which resulted in less heat loss and a higher maximum temperature in the fuel. Sixty-five percent of the UO_2 specimen in experiment 2 "melted" or "foamed," whereas the fuel material employed in experiment 1 did not melt or deform significantly. The release of most isotopes from the high-temperature zone, except those with rare-gas precursors, was quite low under the conditions of these experiments, and the values were approximately proportional to the release from the fuel in both experiments.

The fission products are grouped in the tabulation roughly according to the characteristics of the principal precursors that were present during the transient heating period and the first few seconds thereafter. The three groups are: (1) volatile materials, (2) metals which alloy readily with stainless steel, and (3) nonvolatile materials. The high release values observed for Sr^{89} and Cs^{137} resulted from their noble-gas precursors, which were also responsible for these isotopes being found throughout the experimental system. Most of the released I^{131} and Te^{129} was found in the stainless steel cladding, probably as a result of alloying between the metal precursors and stainless steel. The release of nonvolatile materials was small.

The high release values of the volatile and metal-group isotopes observed in experiment 1 are rather surprising, since, as mentioned above, the fuel apparently did not melt in this experiment. This is interpreted as an indication of very rapid diffusion rates of the precursor isotopes at temperatures near the melting point of UO_2.

Metallographic analyses of the fuel residues and particle-size determinations on particles collected by filters and in diffusion tubes are continuing.

The results of the first two experiments demonstrated the basic validity of the experimental design for obtaining fission product release and behavior data under transient reactor conditions. Construction of assemblies for experiments 3 and 4 (dry-air and steam-air atmospheres respectively) is in progress.

Innocuous Simulation of Accidents and Hazards. — Study is continuing to develop ways of simulating the radioactive materials which would be released during a reactor accident by making use of test materials which are not hazardous themselves. Since radioiodine is generally considered to be the most significant of the volatile fission products from a hazards standpoint, the behavior of radioiodine and its volatile compounds is being given first consideration. Sources of supposedly pure iodine crystals have been shown by diffusion tube measurements and mass spectrometer analysis to contain small amounts of one or more compounds of iodine. In addition

to I_2, the compounds CH_3I, HI, and ICl and others with higher molecular weight have been observed. It is important that these compounds be identified and their behavior fully characterized so that proper simulation under test conditions can be accomplished. As more understanding of iodine behavior is obtained, the application of nonradioactive iodine vapor and compounds for accident simulations will become more acceptable. More sensitive and faster methods of analysis of natural iodine, other than neutron activation, are being investigated.

Release of Fission Products on Out-of-Pile Melting of Reactor Fuels. — The Hot-Cell Confinement Mockup Facility has been used to test a filter-charcoal-trap combination and to observe the plate-out of fission products released by melting high-burnup uranium-aluminum alloy in air and in a steam-air mixture. Data on the distribution of different forms of radioiodine were obtained by use of diffusion tubes before and after the first absolute filter.

Deposition data obtained in dry air (40% relative humidity) revealed a relatively high iodine plate-out rate in the 180-liter stainless steel confinement vessel (82% in about 1.5 hr), while only 17% of the cesium plated out. When the inside walls of the vessel were coated with condensate in the moist-air experiment, only 48% of the iodine plated out in 1.5 hr, while 29.4% of the cesium was retained.

The chemical state of the released iodine remaining in the aerosol was inferred from (1) the ratio of the filter-retained (particulate) fraction to the unretained fraction adsorbed in the charcoal bed and (2) the ratio of molecular iodine (7 to 8 A) plated out in the silver-coated copper diffusion tube to that of particulate iodine (30 A and 100 to 200 A). In dry air about 60% of the iodine not plated out in the confinement tank was in the 7- to 8-A (molecular) size. In moist air the fraction in molecular size was about 75%.

Retention of radioiodine and radiocesium in the test filter-charcoal combination (roughing filter, "absolute" filter, 1-1/2-in.-deep charcoal bed, and another "absolute" filter) was the same under both the dry and the moist condition: 99.97% for radioiodine and more than 99.99% for radiocesium. Penetration of a small fraction of the iodine through the 1-1/2-in. bed length of charcoal set a lower limit to the presence of an alkyl iodide or other low-retention forms of iodine of less than 0.1% in the dry-air experiment and 0.3% in the moist-air experiment.

The apparently independent plate-out of a large fraction of airborne radioiodine and radiocesium in the diffusion tubes indicated that relatively little chemical combination of the two released fission products had occurred.

Release of Fission Products on In-Pile Melting of Reactor Fuels. — Diffusion tube data for the eleventh experiment on the in-pile melting of UO_2 have been analyzed. This is the experiment having a high rate of helium flow. The calculated diffusion coefficients are considerably larger than those reported for previous experiments (ORNL-3532, pp. 20–21) and indicate that the fission products were of vapor form rather than associated with small particles. Theoretical estimates of the diffusion coefficients of the various elemental fission products and their oxides have been made and are consistent with the observed values within a factor of

2; however, it was not possible to distinguish between the elemental and oxide forms of the fission product vapors. The amounts of fission products transported as vapors ranged from 1 to 50% of the amount released from the high-temperature zone of the furnace and were somewhat greater than the amounts associated with fine particles in previous experiments.

The particle size distribution of material collected by the filters is being studied by means of electron microscopy. Because most of the filters have been too radioactive for study, data on only a few filters have been obtained. The results of this study show that the particles which pass through a Gelman HV-70 filter but are caught on membrane filters have diameters of about 0.05 μ. Frequently, the number of such particles is too small for accurate particle size distribution measurements, and often these particles are highly agglomerated.

The irradiation facility at the ORR is being modified to permit the use of a monitor which will measure both neutron and gamma flux before and after an experiment. This monitor will assist in controlling the intensity of heating, which has been found to be the most important variable affecting fission product release.

Characterization and Control of Accident-Released Fission Products. – The use of the diffusional deposition technique to obtain information regarding the compounds of iodine which occur along with the elemental form has been continued. In the earlier experiments, relatively high iodine concentrations, $\sim 10^5$ $\mu g/m^3$, were involved. The iodine was generated by evolution from a melt of potassium dichromate containing NaI^{131} and KI^{127}. With either helium or air as the carrier gas for the diffusion tube and also as the gas passing over the melt, about 99% of the gaseous iodine produced was in the elemental form; the iodine compounds corresponding to the remainder exhibited diffusion coefficients in air of 0.05 and 0.10 cm^2/sec.

Recently, similar experiments have been performed at much lower iodine concentrations, <1 $\mu g/m^3$. At these concentrations, the relative proportion of iodine compounds, as measured by radioactivity and diffusion tube deposition, was considerably increased (e.g., to 30%) in comparison with the roughly 1% indicated above for the higher iodine concentrations. Even when pure helium was the associated gas, extensive compound formation was indicated; the compound formation possibly resulted from reaction of I_2 with unknown components in the melt, with surfaces of the apparatus, or with gaseous impurities carried by the helium after passage over surfaces. Deposition of I_2 from helium on type 321 stainless steel at 25°C appeared quite similar to that observed for a silver surface. Diffusion coefficients for deposition of the compounds from air indicated that the compounds might have been, in part at least, the same as observed earlier.

Further work on the identification of iodine compounds is planned. One approach being considered is measurement of diffusion coefficients for known iodine compounds, and another is gas-liquid chromatography with apparatus constructed from inert materials in conjunction with a scintillation counter as the detector.

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 – diffusion, interception, and inertial impaction. Quantitative analysis of the data in the diffusion regime to yield particle sizes consistent with

those from electron microscopy photomicrographs and particle concentration techniques has been previously reported.

Analyses of the data in the high-velocity range (linear flow 10 to 90 fpm) have shown that rough particle sizes may be obtained and that further refinements in theory are necessary to define the particle size more accurately. High values of α (volume fraction of solids in the fiber mats) used in the initial series of experiments result in overlapping data in the interceptional and inertial flow regions. Further experiments to separate the filtration mechanisms more sharply in the high-velocity range and therefore to delineate the particle sizes more clearly are being performed with higher-level radioactive zinc sources and lower values of α for the filters.

Nuclear Safety Pilot Plant. – Design of the Nuclear Safety Pilot Plant is complete, and construction is in the final stages. All major equipment and piping have been installed, electrical conduit work is complete, and the panel board has been assembled and installed in the control room. The remaining work principally comprises mounting instruments, installing wiring in the conduit, and running instrument air tubing inside the working cell.

Fabrication of the furnace is complete, and a report covering its design and development has been issued (ORNL-TM-687). A few experimental runs were made at a temporary out-of-cell installation; included were tests on the support of the fuel element and the design of the flow guide.

Several experimental runs were performed with instrumented zirconia pellets to establish the approximate temperature distribution along the pellets during meltdown with the plasma torch. The temperature distribution inside the pellets was measured as a function of time and distance from the plasma flame. These measurements indicate that more than half the solid pellet which is first exposed to the flame will be melted before the central region of the following pellet reaches temperatures at which the vapor pressure of the iodine becomes significant.

Plasma instability is experienced when the torch is operated at a pressure above approximately 8 psig; this causes significant amounts of copper to be vaporized from the anode and to be deposited in the furnace.

The initial experimental program as now proposed consists of a number of runs with each of the following: stable iodine, mixtures of I^{131} and I^{127}, and a series using specially irradiated fuel pins. In the stable iodine runs, primarily for shakedown, neutron activation techniques will be used to measure transport phenomena.

Reactor Containment Handbook. – A chapter outline of the handbook, together with estimated completion dates, is as follows:

		First Draft Completion	Final Draft Completion
I.	Introduction	est. 1-64	est. 4-64
II.	Codes and Criteria	4-63	12-63
III.	Radioactivity – Generation and Transport	est. 2-64	est. 5-64
IV.	Energy Sources	4-63	est. 2-64
V.	Analytical Techniques	est. 4-63	est. 6-64
VI.	Credible Accidents	est. 2-63	est. 5-64
VII.	Containment Proof Testing (Bechtel)	4-63	est. 1-64
VIII.	Specific Containment Systems	est. 3-63	est. 4-64
IX.	Design Details (Bechtel)	4-63	est. 1-64
X.	Containment Accessories (Bechtel)	4-63	est. 2-64
XI.	Economics (Bechtel)	4-63	est. 2-64
XII.	Containment Research	est. 2-63	est. 5-64

All 12 chapters will be prepared for publication before the end of the fiscal year. It is expected that the handbook will be published during the summer of 1964. It will be bound in such a way that periodic updating and revision can be accomplished.

Nuclear Safety Information Center. – The major emphasis of the newly established Nuclear Safety Information Center continues to be directed toward the collection, evaluation, and storage of the backlog of pertinent information. Over 750 sources of information have been extracted and indexed.

The Center also has several studies nearing completion, including (1) radioiodine release and monitoring at four major AEC facilities, (2) safety problems in fast gas-cooled reactors, and (3) iodine release and transport phenomena within containment structures. Several studies initiated more recently include reactivity effects of fuel element bowing and effectiveness of safety injection systems for emergency reactivity control. In addition to these studies, the Center has reviewed for the AEC an experimental program proposal.

In November and December the Center received and answered ten specific requests for information and has been requested to add seven additional names to the distribution list for reports prepared by NSIC. The staff of the Center was augmented by the addition of a second typist. The report period was marked by the preparation of a brochure, several hundred copies of which have already been distributed.

WASTE DISPOSAL

Clinch River Study. – The pulsed releases of water from Melton Hill Dam which are expected to occur in summertime were simulated by flow through the gates during August 1963. The purpose of the experiment was to determine the dilution in the Clinch River under the changed flow regime. Rhodamine B dye was used to simulate radioactive releases. The minimum dilution was found to be 50, in comparison with the previous median dilution of 570.

Cores taken from the bottom sediments of the Clinch River in 1962 were analyzed; they showed that the major portion of the contaminated sediments was deposited on the sides of the original stream channel.

Desorption of the nuclides from bottom sediments shows that acidified water is more effective than highly salted solutions in removing Sr^{90} from the sediments.

Analyses of fish taken from the Clinch River show that a maximum of 2.5% of the maximum permissible intake will be received from eating 37 lb of fish per year from East Tennessee rivers. Radiation levels near filter beds in water plants upstream and downstream of the point of entry of White Oak Creek into the Clinch River are at background.

Mineral Exchange Studies. – Explanations previously offered for the different cesium sorption curves obtained for clay minerals were based on the different structures of the clays. Questions were raised regarding this explanation, since different materials had been used. Cesium sorption curves, similar to those obtained for the different clay minerals, were obtained for the mineral vermiculite under saturation by different cations. The response is consistent with the known structural characteristics of this mineral under saturation by different cations. When the saturating cation was magnesium, the distribution coefficient (K_d) for cesium increased with increasing cesium concentration in the solution; the increase was due to the collapse of the interlayer space induced by cesium, with rejection of other cations (which do not induce collapse) present in the solution. With prior saturation with stable cesium ions, which produced collapse, the K_d for cesium remained constant over a wide range of cesium concentration, as expected for normal isotopic ion exchange without concomitant structural change. The latter curve was similar to those obtained for kaolinites and montmorillonites, which also undergo ion exchange reactions without structural changes. With prior potassium saturation, the K_d decreased sharply with increasing cesium concentration, but the numerical value of the K_d remained above that obtained for the cesium-saturated sample. The higher K_d is due to the greater exchange of potassium ions by the cesium; this type of curve was also obtained for illite, muscovite, and heated montmorillonites. These results with vermiculite not only support the structural interpretations of cesium sorption by clay minerals, but offer a method whereby vermiculite impurities may be detected in mixed mineral systems.

Disposal in Deep Wells. – Construction of the injection plant is proceeding, but has been slowed somewhat by bad weather. The cell walls are

virtually completed, much of the air piping has been installed, and installation of process piping has begun. The estimated completion date is the middle of January 1964.

A hazards report for the first four injections is being written.

On the last water injection test the sand erosion technique for slotting the casing of the injection well failed to cut the casing on the first attempt. Numerous theories have been proposed to explain this failure, but none have been convincing. For this reason there has been considerable interest in viewing the slot that was finally cut in the well to see if the appearance of the slot would provide a useful clue. Remote TV cameras and borehole cameras were investigated for this application. The TV cameras available at the Laboratory would need extensive modification to operate under the existing conditions and, in any case, would not operate at the distance (1000 ft) required. A commercially available TV camera was located, but the rental and mileage charge seemed excessive ($2500). A borehole camera was located, but required that the well be dry, a nearly impossible condition. An impression packer (a tool that will make an impression of the slot) remains about the only feasible way to "view" the existing slot in situ; the use of such a tool is being considered. Since the existing slot cannot be conveniently viewed, it is planned to have Halliburton run a series of slotting tests at Duncan, Oklahoma, to determine why the one slotting operation failed and what the slotting pressure, slotting time, and other variables should be for our conditions.

It has been found that the addition of a small quantity of tributyl phosphate to the synthetic waste solution will virtually eliminate the foaming problem. This discovery will be tested under field conditions at Duncan to verify that the densometer can, in fact, be used with the mix presently proposed for injection No. 2.

The main emphasis on mix development during this period has been on cheap mixes that, although not as strong as previously developed mixes, would be pumpable, would set, and would have no phase separation. It has been found that the mineral attapulgite 150 is a far superior suspender to bentonite for solutions with a high ionic content. Several retarders have been investigated, and glucono-d-lactone (CFR-1) seems particularly effective.

Foam Separation Studies. — The foam separation process for decontaminating low-radioactivity-level process waste water consists in (1) precipitation of calcium (as the carbonate), magnesium (as the hydroxide), and other hardness by making the water 0.005 M each in NaOH and Na_2CO_3 in the presence of 2 to 10 ppm of Fe^{3+} (as $FeCl_3$) coagulant; (2) removal of the resulting solids by filtration in a gently stirred upflow sludge column; and (3) foam separation in a countercurrent column with sodium dodecylbenzenesulfonate or other surfactant.

Past experiments showed that cesium could be removed in the sludge bed by the addition of Grundite (an illite clay) to the feed stream prior to the caustic-carbonate precipitation step. The performance of −230-mesh Grundite was significantly improved by baking the clay at 600°C for 15 min, as evidenced by the cesium decontamination factor of 14.5 obtained with 0.38 lb of clay per 1000 gal, compared with a value of 8 achieved under similar conditions with 0.6 lb of untreated clay per 1000 gal.

At high pH (about 12), there are many substituted phenols that display selectivity for complexing cesium in both ion exchange and solvent

extraction systems containing sodium salts. This approach to the removal of cesium in a foam column was investigated in tests of surfactants having active phenolic groups. Of ten phenols screened for solubility and foamability, two (dichlorophene and hexachlorophene) were chosen for further testing in single-stage foam column tracer runs. Since the foam generated in a solution containing up to 500 ppm of either of these phenolic surfactants was unstable, 20 ppm of dodecylbenzenesulfonate was added. Results of these runs showed no significant enrichment of Cs^{137} in the foam phase.

The solids concentration (sludge density) of sludge beds was determined at water flow rates of 20, 47, and 60 gal ft^{-2} hr^{-1}. The feed solution in these experiments was tap water made 0.01 M in NaOH (no sodium carbonate) and containing 7 to 9 ppm of Fe^{3+}. The top 1/2-in. layer of sludge contained 3350, 2000, and 1400 ppm of total hardness (as $CaCO_3$) at these three flow rates.

At the same flow rates, soluble total-hardness values (measured as ppm of $CaCO_3$) were 1.45 to 2.5, 4.0 to 10.25, and 8.5 to 13.25 in filtered effluent samples. Turbidity (hardness as entrained solids) at water flow rates of 47 and 60 gal ft^{-2} hr^{-1} was high: 7 to 35.5 ppm (as $CaCO_3$). These showed no apparent correlation with feed flow rate.

Results of experiments with caustic and caustic-carbonate systems may be summarized as follows: (1) At low flow rates (20 gal ft^{-2} hr^{-1}) there is no significant difference between these reagents with respect to residual soluble hardness. (2) At high flow rates (47 or 60 gal ft^{-2} hr^{-1}) the caustic-carbonate process produces water having significantly lower dissolved hardness (less than 5 ppm, compared with 4 to 13 ppm) and significantly less turbidity (less than 5 ppm, compared with 7 to 35 ppm) than the water produced by the caustic system.

Foam separation equipment is currently under design for installation in the low-level-waste pilot plant. The equipment will include a countercurrent foam column (2 ft square and 14 ft high) for the removal of activity from ORNL low-level waste, three recovery columns in series for the removal and recycle of surfactant from the waste effluent before discharge, and two types of foam-breaking equipment. The feed rate for the system will be about 5 gpm.

High-Activity-Waste Calcination. — In high-temperature processes for converting high-activity aqueous wastes to thermally stable solids for permanent disposal, provision must be made for volatile constituents such as mercury, sodium, and sulfate. The present approach is to remove mercury from the wastes prior to calcination by reaction with copper or aluminum metal, and to retain sodium and sulfate in the solids by chemical adjustment of the composition to form nonvolatile compounds.

Ninety-nine percent of the mercury was removed from a simulated Purex waste (simulating a waste expected to be produced at Hanford in 1965) by passage through columns packed with either copper shot or aluminum turnings. When this waste was concentrated by a factor of 3, however, the copper was attacked too rapidly to be of use, but the aluminum was still effective. Both copper and aluminum were attacked excessively in simulated solutions of current Hanford Purex waste.

Addition of aluminum to a Purex waste containing more than a 2:1 stoichiometric ratio of sodium to sulfate prevented volatilization of alkali, which did occur when the waste was calcined without any additive.

Low-Activity Waste: Ion Exchange Studies. – Laboratory studies of the scavenging-precipitation ion exchange process for decontamination of low-activity aqueous wastes include the following: consideration of recycling the eluate from the ion exchange columns to the precipitator in order to avoid generating a liquid-waste effluent stream; ways to remove phosphate, which interferes with calcium precipitation; and the efficiency of anion exchange resins for removal of ruthenium.

Cesium was fixed in the precipitate of the process by adding Grundite clay to the eluate from the cation exchange column, neutralizing, and metering (recycling) this solution into the untreated low-level waste feed. Cesium is most effectively sorbed on Grundite at a pH of 11.8, which is also the optimum pH for operation of the ion exchange column. This makes possible the fixation of more than 99% of the cesium in the sludge from the process and removes the necessity of evaporating and storing the effluent from the ion exchange column.

An alumina column removed approximately half the phosphate from 10,000 bed volumes of raw ORNL waste, which was sufficient to permit hardness reduction to 5 to 7 ppm (as $CaCO_3$) by precipitation.

A Dowex 21 anion resin column removed about half the ruthenium that had escaped removal in the precipitation and cation exchange steps of the process.

Low-Activity Waste: Pilot Plant Work. – About 65,000 gal of low-activity-level process waste from the ORNL equalization basin was treated by a fixed-bed cation exchange process that included anthracite-bed filtration of the waste followed by ion exchange with a weak-acid cationic resin (Amberlite IRC-50) and a strong-acid cationic resin (Dowex 50). Operation was continued until the total hardness of the effluent was 50% of the total hardness of the feed. After 33,000 gal (1000 resin-bed volumes), just prior to cesium breakthrough from the Dowex 50 column, average decontamination factors for gross beta, gross gamma, cesium, strontium, cobalt, and total rare earths were 25, 1.7, 28, 3350, 1.2, and 240 respectively. Plant effluent, which was strongly tinted green, contained 0.05, 1.4, 1.5, 0.09, and 0.25% of the 168-hr MPC_w for Cs^{137}, Sr^{90}, $Ru^{103-106}$, $Zr\text{-}Nb^{95}$, and Co^{60}.

Tests made in the 4-ft-diam agitated clarifier in the scavenging-precipitation head-end step, using tap water as feed, copperas addition to produce an Fe^{2+} concentration of 5 to 10 ppm, caustic addition to produce a feed 0.01 N in NaOH, and 5% sludge recycle to the flash mixer, resulted in effluents that were 6 to 9, 8 to 10, and 12 to 16 ppm in total hardness (as equivalent $CaCO_3$) for feed rates of 5, 7.5, and 10 gpm (about 24, 36, and 48 gal ft^{-2} hr^{-1} respectively.

Disposal in Natural Salt Formations. – Topside renovation of the hoisting facilities at the Lyons mine is currently in progress. As of December 1, 1963, the contractor reported that 80% of the concrete had been poured for the shaft collar and headframe foundations and that the majority of the excavation and backfill had been completed. Shop drawings for the headframe steel have been reviewed, as well as drawings for

all related hoisting components. Specifications are being prepared for a new ventilating fan and modifications to the hoist house. Used mining equipment has been located and a deposit placed to hold the equipment until required.

A prototype fuel-element canister has been received, and one more unit is being fabricated. The hole liner design is completed, and a prototype model has been ordered.

Additional plastic flow gages have been ordered from England for installation around the experimental area and will be delivered in time for installation when access to the mine is again available.

Engineering, Economic, and Safety Evaluation. – Estimations were made of the movement of Sr^{90}, Ru^{106}, and Cs^{137} in Conasauga shale following a hypothetical release of reactor fuel-reprocessing waste from a tank. Computations were based on hydrologic and exchange characteristics of the local formation that had been obtained previously. Although the absolute values for radionuclide movement which have been calculated and presented should not be considered to be precise, since the estimates were based on a rather inadequate description of the site, the procedure that has been employed in making these calculations could be applied for any proposed site. The reliability of the estimates could be improved by improving the quantitative description of the formation involved.

In Conasauga shale, Sr^{90} would apparently move quite rapidly in an acid waste system, even upon neutralization of the acid by calcite ($CaCO_3$) in the formation. Movement of Sr^{90} in an alkaline waste solution would be much slower due to formation of insoluble compounds in addition to ion exchange. Ruthenium-106 would be expected to move rapidly in both acidic and neutralized waste systems, but presents little long-term hazard due to its relatively short half-life. Movement of Cs^{137} would be expected to be slow because of its fixation by the illite of the Conasauga shale.

PHYSICAL RESEARCH

PHYSICS AND MATHEMATICS

High-Energy Electronuclear Machines: The Separated-Orbit Cyclotron. — In an evaluation study of the separated-orbit cyclotron (SOC) concept, an approximate analysis was made of the size and complexity of a comparable linear accelerator (linac); for the comparison both types of machines were considered for continuous operation at various energies with beam currents up to 1 ma. The results indicate that for energies above 100 Mev, the SOC is the more practicable machine. A more detailed analysis is required to determine the optimum for energy injection from a linac into an SOC. It is also very important to determine the exact beam aperture required for attainable tolerances of magnet alignment. The phasing of the multicavity rf system is being analyzed, and the shaping of the cavities to reduce rf power is being studied. Results indicate that the power requirement can be reduced to about one-half of the earlier estimate (ORNL-3431) by suitable flaring of the cavity.

Medium-Energy Nuclear Physics: The 86-Inch Cyclotron. — The low-lying levels of the nucleus Nb^{92} were studied with the $Nb^{93}(p,d)Nb^{92}$ reaction; the new magnetic analyis system was used to resolve the levels. Deuteron groups corresponding to levels at 0, 135, 285, 355, and 485 kev were observed. Comparison of these data with results from Florida State University (Sheline et al., private communication) indicates that the group corresponding to the excitation of 485 kev is an unresolved doublet resulting from two states at approximately 475 and 495 kev. The six levels are interpreted as those arising from the different couplings of the g 9/2 proton and a d 5/2 neutron. The experimental results were analyzed by using the proton-neutron residual interaction, the matrix elements of which are determined from the levels of neighboring nuclei. The residual interaction determined in this way yields a ground-state wave function for Nb^{93} which consists of about 90% of the configuration $(\pi g9/2)_{9/2}(\nu d5/2)^2_0$, with the remaining 10% consisting of a mixture of the neutron pair coupled to spin 2 and the neutron pair coupled to spin 4. The spin order in Nb^{92} is 7^+, 2^+, 5^+, and 3^+, with the 4^+ and 6^+ too close to determine the order. An alternative approach to analyzing the data is to use the experimental data from the (p,d) reaction by themselves and to adjust the Nb^{93} ground-state wave function until the experimentally observed intensity distribution is reproduced. This was done by a least-squares fitting procedure; the wave function is in excellent agreement with the wave function determined from the levels in the neighboring nuclei.

A method was devised for representing the wave functions of systems of many fermions in a binary occupation-number notation. This notation was devised for calculating spectroscopic factors for pickup reactions involving one or more picked-up particles. The representation, in addition to being well suited for machine computation, provides diagrams which show the picked-up particles and the remaining group as distinctly separate entities, uncomplicated by requirements of permutations between the

groups. The Pauli exclusion principle is satisfied by using binary occupation numbers and a simple set of rules for determining the signs of the various terms that occur in calculations. This method provides a clearer pictorial representation than does the Slater determinant representation with its permutations. FORTRAN programs were written for calculating spectroscopic factors for the pickup of an arbitrary number of particles and for classifying, according to T and J, the states of mixed or pure configurations in the jj-coupling shell model.

Medium-Energy Nuclear Physics: Theory. — An IBM 7090 computer program is being written to calculate Hamiltonian matrix elements between nuclear shell-model states of mixed configuration (in jj coupling). For each specific calculation, the Hamiltonian will be defined in terms of the numerical values of its matrix elements between two-nucleon states. The method of calculation is based on a second-quantized isotopic-spin formalism developed by J. B. French (University of Rochester). With this formalism, the multishell Hamiltonian is expressible as a sum in which each term is a zero-rank tensor product of several single-shell operators. Each single-shell operator is, in turn, a tensor product of creation and destruction operators for that shell. The matrix element of each Hamiltonian term can then be factored into a product of single-shell matrix elements, multiplied by some Racah and 9j coefficients which involve angular momentum and isotopic-spin quantum numbers (but not any other details of the wave functions), multiplied by the two-nucleon numerical values mentioned above. Machine routines to list and order the single-shell operators, the multishell (zero-rank) operators, and the multishell basic wave functions are being tested now. A calculation of the momentum spectra of the triton, the lambda, and the neutron in the reaction $\Sigma^- + He^4 \rightarrow \Lambda + n + t$ was completed. Fairly good agreement was obtained for the lambda and the neutron spectra. The triton spectrum disagrees with experiment, indicating the importance of the Λ-n interaction. The structure of the ground state of Nb^{93} was calculated and used to predict, with good agreement, the relative intensities with which the levels of Nb^{92} are excited in the reaction $Nb^{93}(p,d)Nb^{92}$.

Cyclotron Operations: The Isochronous Cyclotron. — The first external beam from the cyclotron was achieved on November 16, 1963. Beams of 36-Mev molecular hydrogen, 36-Mev deuterons, and 72-Mev alpha particles were extracted from the machine. Range-energy measurements were made to confirm these particles.

This successful extraction of the beam followed the fabrication, installation, and alignment of a new iron-and-coil magnet channel, which is the last element in the extraction system. Subsequently, magnetic-field measurements were made to evaluate the effect of the magnet channel in the region of the circulating beam. The maximum disturbance was found to be only about 50 gauss, out of 20 kilogauss, in the region close to the channel at 30 in. radius.

Magnetic-field measurements were also made in the central region of the cyclotron to determine the effect of removing two 5-in.-diam iron plugs, located axially in the magnet yoke. The removal of these plugs gives essentially no change in the shape of the field in the central region; a total overall disturbance of 80 gauss was measured at full excitation of the magnet. This disturbance is easily corrected with the

circular trimming coils. Removal of these plugs had no measurable effect on the beam. The plugged axial holes were specifically provided to permit the future installation of an axial ion source, or an injector-type ion source.

About 60% of the beam circulating at the 30-in. radius is now extracted from the machine, with no measurable loss in the iron-and-coil magnet channel. Preliminary measurements indicate a beam spot 0.7 in. in diameter and a divergence of about 1 milliradian. After the installation and alignment of the two vertical positioning magnets and the first quadrupole is completed, further tests of beam quality will be made in conjunction with the continuing installation of the ion optics system.

Stable Isotopes Development. — Cadmium separations in the 255° separators provided samples of higher isotopic purity than those from normal calutron operation; however, the degree of improvement was not as great as expected. In lead separations which followed in the 255° separators, Pb^{208} was enriched to 98%, which is a substantial improvement over the 81% isotopic purity achieved in 48-in.-radius alpha calutrons. In both separations, comparable ion outputs were maintained for each separator.

The cross-sectional area of the ion beam at the halfway position (128°) in the 255° separator is small enough ($<0.5 \times 6$ in.) that installation of a horizontal baffle within the liner and manifold regions permits a fair degree of differential pumping. The baffle arrangement allows the No. 1 diffusion pump to evacuate the source region, while the No. 2 pump operates only to evacuate the receiver region. Conduction of charge-material vapors through the baffle opening at the 128° position is relatively small, and an appreciable reduction in contaminating vapors should result.

Preliminary runs using mercury or mercurous chloride as charge feed have shown improvement following installation of only a limited degree of baffling. If differential pumping should be effective enough to eliminate dry-ice refrigeration previously required for mercury separations, the cost of these separated isotopes would be substantially reduced.

Experiments concerning ionic sputtering and saturation values for ions in substances such as uranium, graphite, copper, and copper of certain crystalline orientations continued. Thorium ions striking on graphite were observed to penetrate much deeper than theoretically predicted, and 51%-enriched Ca^{46}, the highest isotopic purity ever achieved, was recovered from single-crystal copper targets.

Two miniature ion sources, one approximately a half-scale model and the other approximately one-quarter scale, are being tested. Both sources have potential application in either calutron second-pass or sector instrument separations. The half-scale model performs well and provides more than half the ion output achieved with standard ion sources. The quarter-scale model, however, performs erratically. For example, at times the produced beam focuses extremely well, but at other times focus is poor. Current drains of undetermined origin limit the source output to a few milliamperes. The one-quarter-scale model is preferred, and efforts are being applied to control and correct the drain problems.

Special Separations. — Approximately 85% of the separated plutonium isotopes from series "a" were recovered and purified. A total of 27.3 g of enriched Pu^{241}, 89.6 g of Pu^{240}, and 14.1 g of Pu^{242} were processed.

Two kilograms of U_3O_8 containing 99.983% U^{238} was converted to UO_2 for use as charge feed for current uranium separations utilizing internal chlorination with CCl_4 to provide a volatile chloride vapor. Based on monitored ion currents, > 350 g of U^{238} was collected during the two-month period. The U^{238} charge material will be recovered from calutron components. Four tanks in the contained-separation facility are being used for the uranium series.

A total of 220 g of 99.6% U^{233} was dissolved from two metal buttons, processed to low-fired U_3O_8, and made available for charge material in an anticipated U^{233} separation series.

Up to 2.5 mg/cm^2 of uranium was electrodeposited on zinc-coated (~20 μg/cm^2) aluminum backings. Investigations are being made for supplying a uranium beta source for use in a beta-gamma monitor.

CHEMISTRY

Instrumental Methods for Continuous Chemical Analysis. — The feasibility and utility of determining the concentration of complexed metals in solvent extracts have been investigated polarographically. The ORNL model Q-1988-FES controlled-potential and derivative dc polarograph and the 0.5-sec Smoler dropping mercury electrode (D.M.E.) were used. Well-formed, reproducible polarograms were obtained in a number of solvents with several ion-association systems, including extracts of cadmium and of uranium. Less satisfactory polarograms were obtained with chelate complexes.

An evaluation was made of the deviation of the potential of the D.M.E. from the input voltage of the potentiostat as a function of the specific resistance of the solution, ρ. A large part of the total cell resistance R is located in the high-current-density region within a few drop radii of the surface of the D.M.E. Measurements of the average value of this inner resistance R_i are in good agreement with calculated values. For this D.M.E., R_i is about 3ρ. When ρ is greater than about 1400 ohm-cm and the average value of the cell current i is 1 μa or more, the iR_i loss distorts the polarogram excessively unless the tip of the potentiostat reference electrode is placed within 0.1 drop radius of the drop surface. When the tip is in this position, R is compensated for by the potentiostat. This electrode configuration is not difficult to achieve with a Smoler D.M.E. At usual concentrations, the polarograms recorded by this method had an insignificant amount of iR distortion in solutions with ρ as high as 20,000 ohm-cm. Satisfactory polarograms are obtained without locating the tip of the potentiostat reference electrode close to the D.M.E. if iR_i is insignificant, which is usually the case when ρ is less than 1400 ohm-cm.

The behavior of cadmium extracted with tetrabutylammonium iodide as both the extraction agent and supporting electrolyte in 2,2′-dichlorodiethyl ether or in 1,2-dichloroethane is of interest because even though ρ is low, so that species should be available for migration, polarograms are not obtained. After an alcohol is added to the extract, polarograms can be obtained; it is presumed that the alcohol in association with the cadmium complex makes possible the electron-transfer step.

A method was developed for the polarographic determination of uranium. The extractant is 0.1 M tri-n-octylphosphine oxide in cyclohexane. After equilibration and phase separation, an equal volume of 0.2 M $LiClO_4$ in ethanol is added to the extract to provide a supporting electrolyte. Linear relationships are obtained between concentration and diffusion current and between concentration and first-derivative peak height for uranium concentrations in the polarographic cell from 5×10^{-6} to 1×10^{-4} M. By concentration of the uranium during the extraction step, the method can be applied to the determination of at least tenfold lower concentrations of uranium in aqueous solutions. Consequently, this method should also be useful for the determination of uranium in trace quantities in samples such as process waste streams or natural waters.

It is concluded that controlled-potential polarography directly in solvent extracts is a useful analytical technique. The species to be determined can be concentrated selectively during the extraction, a separate stripping step into an aqueous medium is not required, and the noise level is low. There is also the possibility of making a polarographic determination in an organic solvent for species that are not stable in water.

Analysis of Molten Salts by Electrochemical Methods. — The method of anodic stripping voltammetry (ORNL-3532) looks promising as a means of estimating low concentrations (< 20 ppm) of nickel directly in molten LiF-NaF-KF (46.5-11.5-42 mole %). The voltammetry of nickel in this solvent at 500°C was investigated over the range from 20 to 80 ppm. The current-voltage curves recorded at a pyrolytic graphite indicator electrode are well defined. The half-wave potential occurs at approximately −0.2 v vs the platinum quasi-reference electrode. The curves are peak-shaped at scan rates from 1 to 10 v/min and S-shaped at slower scan rates. This characteristic difference in shape indicates that at the faster scan rates, the transport process to the electrode is mainly diffusion-controlled. The current (wave height) is proportional to the concentration of nickel over the range investigated, a fact which demonstrates the analytical utility of voltammetric measurements. The diffusion coefficient (D) for nickel was calculated to be $\sim 1 \times 10^{-6}$ cm^2/sec at 500°C. At 570 and 600°C, the D values are approximately 2.7 and 4.5×10^{-6} cm^2/sec respectively. From a plot of log D vs 1/T, an activation energy of about 18 kcal/mole was calculated for the current-limiting process, which corresponds to the reduction of Ni(II) to the metal at a pyrolytic graphite indicator electrode.

Protactinium Chemistry. — The approximately 50 g of Pa^{231} obtained from England for use in producing U^{232} for nuclear measurements is being purified before its return to England. The purification consists in preparing an HCl-HF solution of the protactinium and using anion exchange to remove the U^{232} and U^{233} still present as impurities. The protactinium, being complexed with fluoride, does not sorb on the anion exchange column. The fluoride is then competitively complexed away from the protactinium by adding boric acid to the solution. The protactinium forms anionic chloride complexes with the hydrochloric acid and sorbs on a second anion exchange column. Further purification from such nonsorbing contaminants as niobium is obtained in this step. Finally, HCl-HF solution is used to elute the protactinium by re-forming the fluoride complex. These operations have now been completed. A remaining operation is that of precipitating the protactinium from the eluate by adding excess ammonium hydroxide.

During operation, the solution was not fully saturated with boric acid, and, as a result, protactinium loading on the second ion exchange column was not satisfactory. This necessitated recycle of most of the solution after addition of excess boric acid. The anion exchange column raffinate now consists of about 22.5 liters of 9 N HCl–0.3 N HF solution, saturated with boric acid. Besides 12 mg of protactinium, it contains very large amounts of Th^{228} and daughters.

The second ion exchange column was eluted with HCl-HF to recover the protactinium, and 36 g has been recovered in 14 liters of solution (determined by alpha analysis). This material is quite pure with respect to alpha emitters (more than 99% Pa^{231}), but it does contain some Nb^{95}.

Properties of Aqueous Solutions: EMF and Solubility Studies. – The thermodynamic properties of deuterium chloride solutions are being investigated by use of the cell D_2 (Pt) |DCl (m)| AgCl,Ag. From results so far obtained it appears that the E^0 for the Ag,AgCl electrode is lower in the DCl medium than in HCl and that the difference becomes greater as the temperature increases. The measurements are being made in the concentration range 0.01–1.0 m at temperatures from 25 to 250°C.

Properties of Aqueous Solutions: Species and Thermodynamic Properties. – Of the polyanions formed in the hydrolysis of metal ions, those of tantalum(V) have received relatively little attention. Partly this neglect stems from the fact that the pH range in which they are stable is too high for accurate emf measurements of acidity, in contrast to, for example, tungsten(VI) and molybdenum(VI).

The degree of polymerization of $K_8Ta_6O_{19}$ was investigated by equilibrium ultracentrifugation in 1 M KCl and 1 M KOH supporting electrolytes. The results indicate that a single species is predominant and that, as the stoichiometric formula of the solid suggests, the aggregates contain six tantalum atoms. Extensive binding of K^+ counterions is indicated by the low values of charge observed. These conclusions are consistent with the results of a light-scattering study carried out elsewhere.

Lindqvist and Aransson, from an x-ray structural study, have reported hexamers in solid $K_8Ta_6O_{19}\cdot 16H_2O$, and it is plausible to suppose that the species in solution have the same structure. To check this, Raman spectra were obtained for the solid and for concentrated tantalate solutions. Close correspondence between these confirms the identity of species in solution and solid.

Spectrophotometry of Solutions at High Temperature. – The Chemistry Division cell assembly for the standard Cary model 14 spectrophotometer operates routinely from –80 to 250°C and from 0 to 2000 psia. The resolving power and reproducibility of the basic instrument have not been compromised in the adaptation.

Several dozen test runs were made with pure solvents using 3.81-cm cells. The base-line absorbance of D_2O in the sample cell vs D_2O in the reference cell, both cells being identically controlled, was less than ±0.01 from 5 to 250°C over the spectral region from 0.4 to 1.8 μ. Initial spectral measurements of four solvents as a function of temperature and pressure have been made with CCl_4 at room temperature in the reference cell: water, 0.7 to 1.35 μ, 2 to 250°C and 0.01 to 580 psia; D_2O, 0.7 to

1.85 μ, 4 to 250°C and 0.01 to 580 psia; absolute ethanol, 0.7 to 1.4 μ, −80 to 244°C and 0.01 to 980 psia; anhydrous ammonia, 0.3 to 1.46 μ, −72 to 144°C and ~1 to 2000 psia.

The most striking feature of these complex spectra is the contrast in behavior with temperature between ammonia and the other three solvents. All the absorption bands observed in ammonia decrease in intensity with increasing temperature. With the other solvents some bands increase and others decrease.

With a machine capable of temperatures at least from −80 to 250° and pressures from 0 to 2000 psia, it is now easy to study a wide variety of liquids above their critical temperatures. In a quick exploration, ethanol showed no sudden spectral change in the critical region, whereas ammonia exhibited marked scattering, the intensity of which peaked sharply at the critical point.

High-Temperature and Structural Chemistry. — Thermal analysis of the metal—metal-halide systems Ba-BaX_2 (X = Cl, Br, I) produced thermal halts in addition to those corresponding to monotectic and eutectic transitions and to liquid-liquid phase separation. The extra halts were taken as indications of the existence of intermediary compounds. This tentative interpretation was supported by x-ray diffraction patterns showing the occurrence of structures different from those known for the barium dihalides and barium metal.

Physical Chemistry of Gaseous Thermodynamic Working Fluids. — The expression which relates the thermal transpiration effect exhibited by a gas to the translational component of its thermal conductivity also contains the gas viscosity as a parameter. Because of the large thermal gradients which are necessary in the present work in order that the transpiration effect be measurable, and because of the strong dependence of viscosity on temperature, the assignment of a characteristic temperature to which the viscosity refers cannot be made unambiguously. However, the rate of approach of the thermal transpiration experiments to steady-state conditions is likewise viscosity-dependent, and thus both the gas viscosity and the thermal transpiration effect can in principle be determined simultaneously. The present apparatus has therefore been modified to include a sensitive recording differential-pressure device. The experimental work currently in progress is largely directed toward an evaluation of the performance of this pressure-sensing device, along with its calibration.

Target Preparation. — Emphasis has been placed upon development of very high-temperature evaporation sources (operating temperatures between 1000 and 2800°C) using crucible materials of maximum chemical stability. To develop such temperatures, a study was made of electron bombardment techniques, which can generate temperatures in excess of 3000°C. These methods proved successful for evaporation of rare-earth metals, reduction of magnesium oxide with subsequent evaporation of magnesium metal, and deposition of thin films of elemental silicon and germanium.

Study of vapor source designs has continued in which evaporation-condensation efficiencies greater than 50 to 75% might be achieved. New crucible designs with electron bombardment or electrical resistance heating produced efficiency values in this range for some of the materials

that were evaporated. Also, initial quantities of bulk starting materials were reduced by nearly two orders of magnitude from the amount required to obtain equivalent thicknesses of thin films by use of classical filament designs.

TRANSURANIUM-ELEMENT PRODUCTION

Chemical Process Development: Tramex Safety. — Small-scale laboratory tests to determine fire and explosion hazards of Alamine 336·HNO_3 in diethylbenzene (DEB) have been continued. Hazards are decreased when 4% Alamine 336·HNO_3 in DEB is in contact with either 3 M $AlCl_3$ or 10 M LiCl–0.1 M $AlCl_3$. No fires or explosions occurred even though an electric spark was passed through the vapor phase as the mixtures were distilled to pot temperatures as high as 375°C. Hazards are increased when the Alamine 336·HNO_3–DEB solvent is in contact with 2.6 M $Al(NO_3)_3$. With this mixture, mild explosions were obtained by igniting the vapor phase with an electric spark when pot temperatures were as low as 85°C. These results indicate that it is probably safe to adjust Tramex feed to 10 M LiCl by evaporation even if some solvent is present, but precautions should be taken to make certain that no solvent is present if aluminum nitrate solutions must be heated.

Chemical Process Development: Americium-Curium Separation at High Activity Levels. — Methods of separating americium and curium from each other are being investigated at high activity levels in order to determine the feasibility of separating gram quantities of Am^{241} and Cm^{242}. It has been previously reported that good separation was obtained by oxidizing Am^{3+} in 3 M K_2CO_3 with 0.6 M NaOCl to precipitate $KAmO_2CO_3$. The most recent tests show that equally good results can be obtained by oxidizing with persulfate. With $K_2S_2O_8$ as the oxidant, 96% of the americium was precipitated as $KAmO_2CO_3$; 3% of the curium accompanied it. The curium concentration in this solution was 1.05 g/liter, which is equivalent to an alpha activity level of 130 w/liter. Curium precipitated from the filtrate after about 10 hr, presumably as a result of the high activity level.

Chemical Process Development: Separation of Lanthanides from Actinides by the TALSPEAK Process. — The TALSPEAK process for removing the lanthanides from the trivalent actinides by extraction from solutions containing diethylenetriaminepentaacetic acid (DTPA) with di(2-ethylhexyl)-phosphoric acid was successfully tested at element concentrations likely to be encountered in the TRU processing facility. However, in the planned Savannah River Laboratory (SRL) facility for producing Cm^{244}, large amounts of lanthanum are added to the mixed elements as a carrier to increase the curium recovery by precipitation of a mixed double sulfate. After metathesis with sodium hydroxide and dissolution in acid, the feed solution to any group separation process contains about 1 g of curium and about 30 g of lanthanum per liter. The Tramex process can use feed solutions of this concentration with little modification, since only the americium and curium are extracted. In contrast, in the TALSPEAK process

the lanthanides are extracted first, and problems can arise from the necessity of handling the larger concentrations of these elements, concentrations not previously encountered in previous process developments.

Studies are now under way to find combinations of diluent, reagent concentration, relative phase volumes or flow rates, and aqueous compositions which give suitable performance with SRL solutions. First attempts to prepare a feed solution for the TALSPEAK process were not successful, since the solubility of DTPA was low in solutions obtained by dissolving the SRL hydroxide precipitate in nitric acid and adding lactic acid. However, in recent tests, satisfactory feed solutions were prepared by dissolving the precipitate directly in lactic acid.

Process Equipment Development: Cell Mockup Studies. — The feasibility of installing and removing jumper lines from the hot disconnect well to the equipment rack was demonstrated. A mockup of the tube bundle with the hot disconnect well was completed and installed in the mockup. A mockup of the sampler was completed and is ready for testing.

The new Central Research Laboratory model F extended-reach manipulators perform well. The locking feature has proved very helpful in handling the transfer of equipment racks. This feature enables one operator to perform work which would otherwise require two operators.

METALLURGY AND MATERIALS

Preparation of Pure Materials. — Crystals of very pure potassium fluoride were prepared by sublimation from a melt containing UF_4. Crystals of potassium fluoride obtained from commercial sources are ordinarily contaminated with KOH. We added UF_4 to a KF melt to convert the hydroxide into oxide, and precipitated it from the molten solution as UO_2. Crystals of KF sublimed from this melt contained 500 ppm of oxygen as oxide, roughly half that present in the best material previously available. This precipitation-sublimation method of obtaining pure fluoride crystals was applied successfully to obtain single crystals of AlF_3 and ZrF_4 as well as KF.

Single crystals of LiF grown by the Stockbarger method in nickel capsules have shown accumulations of cationic impurities such as Mn, Mg, and Na in the upper part of the crystal. A study of the segregation behavior of these cations in LiF was initiated using a vertical-tube zone-melting technique, in order to provide a guide to methods for obtaining higher-purity LiF. For this purpose a sample of commercial LiF (containing 1000 ppm NaF) doped with 1000 ppm each of MnF_2, MgF_2, and CaF_2 was prepared. Four zone-melting experiments were carried out with LiF under different conditions with respect to the number of passes and the direction of the molten zone. Samples from different portions of the LiF ingots were removed for analysis.

Reactions at Metal Surfaces. — Many technologically important metals owe their usefulness to the protective qualities of the very thin oxide films that form on their surfaces, yet our understanding of the mechanism by which these films are formed is very limited. The investigation of the "thin film stage" of oxidation remains a primary objective of this

research program. We have studied the influence of surface impurities on the oxidation characteristics of copper to correlate oxidation rate with the state of crystalline imperfection, or lattice disregistry, of the oxide. It was concluded that in general an increase in lattice disregistry in the oxide should lead to an increase in the oxidation rate, and the degree of lattice disregistry in the oxide has been identified as an important factor in determining the overall oxidation rate of a metal. We have investigated, by x-ray diffraction, the thin oxide films on several crystallographic planes of copper single crystals. This research indicated that the lattice disregistry in the oxide varied with the orientation of the substrate and suggested that the marked anisotropy of the rate of oxidation of copper may be related to a comparable anisotropy of the degree of lattice disregistry in the oxide.

Deformation of Crystalline Solids. — The mechanisms by which metals deform under stress are incompletely understood. We are using several approaches to investigate these mechanisms.

To investigate the effect of strain rate on yield drops, the yield drops in tantalum, copper-aluminum alloy, and silver-aluminum alloy were measured as functions of strain rate and temperature. In all three cases the yield drops can be attributed to the rapid multiplication of dislocations.

We are studying the stress-induced ordering of carbon atoms that takes place in an iron-carbon alloy as a result of dislocation motion during creep. By a differential temperature technique, we have found that the activation energy shows a maximum of 20 kcal/mole at intermediate strain rates and levels off 5 to 7 kcal/mole lower at higher and lower strain rates. Increasing the dislocation density increases the strain rate corresponding to the maximum activation energy.

Of the mechanisms that have been considered rate-controlling for the deformation of body-centered cubic metals, the nucleation of a double kink predicts a small activation volume, in agreement with experimental measurements. We have completed theoretical calculations of the energy to nucleate a double kink for external stresses ranging from zero to the Peierls (intrinsic lattice) stress. For stresses above 5 kg/mm^2 these values agree very well with experimental values of the activation energy for the rate-controlling mechanism of deformation. However, this may be fortuitous, since the interaction energy was not considered. Calculations of the interaction energy are in progress.

Spectroscopy of Ionic Media. — In nuclear energy and other modern technological fields, increasing use is being made of high-temperature liquids, such as molten salts. However, there is very little fundamental knowledge concerning the molecular entities that exist in these media. We have been systematically studying the geometry and electronic parameters of ion complexes in molten salts for several years by optical spectroscopy. One phase of this program is a study of halonickel(II) complexes dissolved in halide salt melts. In the experimentally difficult investigation of molten fluoride salts, we have measured the spectrum of nickel fluoride dissolved in the molten eutectic salt composed of the fluorides of lithium, sodium, and potassium at 525°C and analyzed the results in terms of ligand-field theory. Our results show that nickel(II) ions are surrounded by an octahedron (or quasi octahedron) of fluoride

ions with a cubic ligand-field strength of $-650\ cm^{-1}$ and that the electron correlation parameter B is $950\ cm^{-1}$. Comparing our measured oscillator strengths of spin-allowed bands with data for crystals at low temperature indicates that the NiF_6^{4-} ion complex in the melt is less disordered than the hexaoxonickel(II) complex in melts of oxo salts.

Fundamental Research in X-Ray Diffraction. — Investigations of mechanisms of radiation damage in crystalline lattices are important both to the development of practical reactor materials and to the understanding of the fundamental physical processes involved. Beryllium oxide is a particularly suitable material for radiation-damage experiments because of its favorable nuclear properties and because of the variety of damage effects that it exhibits. We are continuing to collect x-ray diffraction data from beryllium oxide single crystals irradiated to $1\text{–}6 \times 10^{21}$ neutrons/cm^2 (neutron energy above 1 Mev) at 1075°C. Even at this high temperature of irradiation, we find that the volume increase due to lattice expansion, as reflected in the average lattice parameters, accounts for only about 25 to 30% of the volume increase calculated from the experimentally observed density. Diffuse diffraction effects that persist in patterns from these crystals indicate some remaining defect agglomeration.

Crystals of binary and ternary oxides with interesting magnetic properties have been grown and are reported under "Crystal Physics" (see next topic). We believe that typical formulas of these compounds are $(R.E.)_2MoGeO_8$ and $(R.E.)_2Ti_2O_7$, where R.E. is a heavy lanthanide. Diffraction data suggest that the former compounds have a tetragonal scheelite-type structure in which molybdenum and germanium ions are randomly located on sites occupied by tungsten in $CaWO_4$ (scheelite). The structure of the latter compounds is probably of the cubic pyrochlore type [J. Res. Natl. Bur. Std. 56, 17 (1956)].

Crystal Physics. — Fundamental understanding of the properties of many materials is aided by experiments with single crystals. Suitable single crystals are frequently difficult to obtain, particularly for refractory substances. Therefore, we are conducting a continuing program on crystal growth, both to provide crystals needed in research and to devise and improve methods for growing crystals.

Our preceding report stated that crystals of a new erbium silicate compound were strongly magnetic at room temperature. As a part of the same investigation into the synthesis of rare-earth compounds, we have grown well-formed crystals, more than 2 mm on an edge, of another new strongly magnetic compound. The chemical composition has been determined as Er_2GeMoO_8, and by x-ray analysis this compound was indexed as corresponding to the tetragonal scheelite-type crystal structure. We have observed the magnetocrystalline anisotropy and the magnetic susceptibility as a function of field of this erbium germanomolybdate. Columnar-shaped crystals, having a length-to-diameter ratio of 5, readily rotate in a magnetic field of 900 gauss to align their long axes (their c axes) perpendicular to the field. Thus the susceptibility is significantly less along the c direction. The direction of maximum susceptibility is not known. The magnetic susceptibility of crushed crystals of this compound has been measured as a function of magnetic field. We observed a large ferromagnetic component.

Thus two pieces of evidence, the distinct magnetocrystalline anisotropy and the large ferromagnetic component observed in the susceptibility, suggest that nearly optically clear (pale pink) Er_2GeMoO_8 is spontaneously magnetized at room temperature. However, there are conflicting bits of evidence which must be resolved. Upon theoretical grounds it can be argued that spontaneous magnetization would not be expected in such a dilute magnetic spin system of a rare-earth element at 300°K. Should these crystals prove to be ferro- or ferrimagnetic, it will be a very interesting discovery.

We have also successfully synthesized millimeter-sized single crystals of four other lanthanide germanomolybdates of the general formula R_2GeMoO_8 (R = Gd, Tb, Dy, and Ho).

Fundamental Investigation of Radiation Damage in Solids: Dose Dependence of Dislocation Breakaway Stress in Neutron-Irradiated Copper as Measured by Amplitude-Dependent Internal Friction. — Measurements of the strain-amplitude dependence of the internal friction in copper have been made at 100°C as a function of fast-neutron dose. Although it is found that the Granato-Lücke theory of amplitude-dependent internal friction does not describe the results obtained, the dose dependence of the dislocation breakaway stress can be deduced by other means. After taking into account the variation with neutron dose of the mean free dislocation length, it is found that the breakaway stress is proportional to the cube root of the integrated flux in the dose range from 10^{11} to 10^{14} neutrons/cm^2 (above 0.6 Mev). Extrapolation of the breakaway stress results to doses greater than 10^{19} neutrons/cm^2 shows essential agreement with earlier neutron-hardening results.

Fundamental Investigation of Radiation Damage in Solids: Model for Radiation Equilibrium Between F and M Centers in KCl. — It has been experimentally demonstrated that the M-center concentration in irradiated alkali halides is proportional to the square of the F-center concentration. This is to be expected, since the accepted model for the M center is a pair of coupled F centers. Recent experiments have shown that the relation N_M/N_F^2 (the proportionality constant for the F-M-center relationship) decreases with increasing ionizing intensity. This suggests that the radiation equilibrium between these centers is established according to the following model: F centers capture photoelectrons produced by the radiation field, becoming F' centers. The F' centers are mobile at room temperature and migrate to F centers, forming M centers. The reverse reaction occurs by hole capture by an M center, causing the latter immediately to break up into two isolated anion vacancies, which subsequently capture electrons to become F centers. An order of magnitude check of this model indicates that it is a reasonable one, whereas an alternative model involving a mobile anion vacancy fails to yield agreement within seven orders of magnitude.

Fundamental Investigation of Radiation Damage in Solids: Assembly and Use of an Ionized Argon Beam System for Determining the Sputtering Yield of Gold. — As part of a program to study radiation damage in solids, a system was constructed to measure sputtering yields and ejection patterns of crystals when bombarded with a beam of positive argon ions. This beam can be focused on a 3-mm-diam spot with a current density of 100 to

250 $\mu a/cm^2$ and 1000 to 5000 v energy. The system incorporates an rf ionization tube with a dc magnet and an einzel lens for focusing. With this type of lens, ions of different energies are focused by different amounts. Therefore, by wiping the beam and using only the center section, an ion beam was formed with 95% of the beam at the desired energy. This resulted in a small system without magnetic or electrostatic analyzers, therefore having less volume. The vacuum system uses oil diffusion pumps with liquid-nitrogen cold traps. The base pressure of the system was 1.2×10^{-7} torr. During operation, the system pressure was $1\text{-}5 \times 10^{-6}$ torr, the increase being due to argon. The overall electronic system had an accuracy of ±4%, and the overall sputtering efficiency was ±7%.

The sputtering yield (atoms ejected per bombarding ion) was determined by the weight-loss method. The direction of ejection was determined by collecting the sputtered atoms on a glass plate, placed parallel to the crystal surface. These ejection patterns were studied from the three low-index planes of gold.

Fundamental Investigation of Radiation Damage in Solids: Microstrain in Low-Dislocation-Density Copper Crystals as Investigated with Borrmann X-Ray Transmission. — Borrmann transmission of x rays has been used to study the dislocations in copper crystals. Crystal plates $1 \times 1 \times Z$ cm ($0.03 < Z < 0.14$) with the 1×1 cm faces in the (111) plane were prepared from large copper crystals by acid-sawing and polishing. Transmission x-ray topographs were made of these crystals, using the reflected Borrmann beam from the $(\bar{1}11)$, $(1\bar{1}1)$, and $(11\bar{1})$ planes [which intersect (111) at an angle of 70° 32′] with Ag $K\alpha_1$ radiation. The topographs were recorded on Ilford nuclear plates and subsequently enlarged optically to ~20×. The dislocations appear as light streaks on the plate, and for dislocation densities less than about 10^4 per square centimeter, individual dislocations could be studied by this technique. The dislocation-surface intersections on the (111) plane were located by etch pits, and there was agreement between the two methods for detecting dislocations. In order for a dislocation to be observed in the x-ray topograph, its Burgers vector must have a component out of the plane being used for diffraction, and therefore by comparing the topographs from {111}, the Burgers vectors of the dislocations could be determined. Slip traces which resulted from microstrains have been observed and the Burgers vectors of the dislocations determined. If the crystal plates are annealed at high temperature, the dislocations tend to go straight through the crystal, thereby demonstrating the attractive force due to the surface.

CONTROLLED THERMONUCLEAR RESEARCH

Plasma Physics. — The transition between a normal, magnetically supported vacuum arc and the Neidigh mode II pressure-gradient arc has been successfully described by a quite elementary model. The model quantitatively predicts the observed phenomena — the transition from the normal to the pressure-gradient mode of operation occurs at a sharply defined gas feed rate; the pressure-gradient arc emits energetic ions; and ion spectral lines increase in intensity while neutral spectral lines decrease in intensity. The model assumes that in the normal, magnetically supported arc, the electron temperature is limited by excitation processes

in the un-ionized gas. As the gas input is gradually restricted, a limiting gas density is reached in which the excitation processes can no longer dissipate the energy fed into the system. At this gas feed rate, the electron temperature suddenly rises, the gas becomes almost completely ionized, the space potential becomes very positive, and high-energy positive ions are ejected from the arc.

DCX-2 Facility. — Experiments in which a beam is injected into pulsed or dc lithium arcs have been continued. Very large densities of trapped ions have been obtained. Peak densities of $\sim 2 \times 10^{10}$ energetic ions per cubic centimeter with ion mean lifetimes of 10 to 20 msec have been measured. The total trapped population is about 2×10^{15} particles, approximately 10^4 times the number in the DCX-1. No evidence for flutes or other plasma loss mechanisms are observed; essentially all injected ions may be accounted for by charge exchange. The energy spread of the trapped ions is still observed to be quite large and determines the charge-exchange lifetime. Angular scans in the vertical plane by use of the movable energy analyzer have been obtained. These data may be reduced to produce the radial distribution of particles of selected energies in the machine. These measured distributions establish that the plasma produced by arc dissociation does not extend appreciably radially to the snout.

Vacuum. — Mass spectra of two hydrocarbons, n-decane and n-heptane, have been obtained with a time-of-flight spectrometer at both 30 and 70 v electron energy. The absence of metastable mass peaks (due to dissociation) was noted, as was also an enhancement of primary peaks relative to peaks due to fragmentation of primary species (e.g., mass number 43 compared with mass numbers 39–42). The spectra of mechanical pump oils have been obtained; they normally show peaks past mass number 350, with large amounts of methane and other low-carbon-number hydrocarbons.

The ultrahigh-vacuum test facility instrumentation was improved by the incorporation of higher-speed recording equipment for vacuum gages and more stable ion gage controls. Because of the limited compression ratio of a diffusion pump for methane (reportedly $\approx 10^6$) and since methane appears to be the predominant gas during operation of large systems in the 10^{-10}-torr range, an interstage diffusion pump has been added in series with the existing pumps and has been operated.

Tentative procedures have been developed for the testing of block-type vacuum valves for helium leak and permeability rates and for conductance.

Magnetics and Superconductivity. — The field design for the Elmo facility has been completed, and new magnet coils are being built. The following special cases of "absolute B minimum field configurations" were investigated: the magnetic field resulting from the superposition of a quadrupole field ("Ioffe bars") and the regular magnetic field of the DCX-2; three cases of field configurations produced by an axisymmetric coil arrangement suggested by J. Andreoletti; and finally a numerical analysis of a field configuration due to two infinitely long current loops as described by D. A. Baker. Only the Ioffe bar arrangement produces a relatively large increase of the magnitude of B in the field zone of interest.

An extensive systematic study on different models of high-field superconductors was made. Magnetic-moment measurements of bundles of Nb-Zr wires gave very reproducible results for complete magnetization cycles. These measurement values are in fairly good agreement with the theoretical and experimental results of others based on the flux creep theory of Y. B. Kim *et al*.

BIOLOGY AND MEDICINE

MOLECULAR- AND CELLULAR-LEVEL STUDIES

Physics of Tissue Damage. — Measurements of intensities, spectra, and yields of thermoluminescence from irradiated biological materials such as the aromatic amino acids are complicated by difficulties in obtaining a suitable sample environment. A sample holder should be tissue-equivalent for dose measurements, nonfluorescing or -thermoluminescing, electrically and thermally conducting, structurally strong over a wide range of temperature, and capable of holding a high vacuum. After evaluating many materials with regard to the above requirements, a holder was designed and fabricated from boron carbide. Dose measurements in the holder using Fricke dosimeters and Victoreen r meters indicated agreement with air dose values. All other requirements were satisfied by this holder. Thermoluminescence measurements, conducted in vacua ranging from 10^{-6} to 50 torrs, showed that the light intensity depended not on the type but on the pressure of the gas. Studies of the sample temperature revealed that the sole function of the gas was to produce temperature equilibrium between the sample and the bottom of the sample holder, although the sample was almost completely surrounded by walls at the temperature of the sample holder. Study of the radiative heat transfer in the system explains this result and points to a possible source of error in thermoluminescent dosimetry.

TERRESTRIAL AND FRESH-WATER ECOLOGY

Radiobotanical Studies on White Oak Lake Bed. — Three-year-old potted scrub pine (*Pinus virginia* Mill.) trees were exposed to chronic gamma irradiation for 178 days on White Oak Lake bed. These plants were placed on metal racks 3 ft above the soil surface to minimize contamination by uptake or deposition of soil radionuclides. Control plants were placed off the lake bed and 65 yd away from the others. During this period the lake-bed pines received a total gamma radiation dose of 470 rads delivered at an average dose rate of 2.6 rads/day. Absorbed dose was determined by metaphosphate glass rod dosimeters placed on plant shoots. Control plants received 4.5 rads total dose during this time. Neither the high- nor the low-dose (control) group was watered during the course of the experiment.

By the end of the growing season, all pines in the higher-dose group exhibited 100% mortality of needles, while needle mortality of controls

averaged only 10%. Moreover, the length of terminal shoot growth in the higher-dose group (52.3 ± 5.2 mm) (mean ± standard error) was significantly less ($p < 0.01$) than that in the controls (78.4 ± 5.1 mm). Higher-dose pines also exhibited significantly shorter needles and smaller buds.

Compared with other radiation studies on conifers, the growth retardation and lethality produced at this relatively low dose rate and low total absorbed dose seem much greater than would be expected. These enhanced effects are interpreted as being produced by a combination of summer drought and gamma radiation. This study illustrates how an environmental factor may serve to augment or intensify radiation effects in a field situation.

Transport of Radioactive Materials by Mud-Dauber Wasps. — Radioactive nests of four species of mud-dauber wasps have been collected, or surveyed in place, for determination of their level of radioactivity. Several hundred control cells were collected on the X-10 area prior to the first frost, and several hundred others were located but left in place for collection in winter and spring. Nests of *Sceliphron caementarium* (Drury) and *Chalybion californicum* (Saussure) gave readings from slightly above background level of radiation to more than 800 mr/hr of mixed beta and gamma radiation. Although both of these species had radioactive nests in three well-separated areas, the nests of *Trypoxylon politum* Say and *T. clavatum* Say in the same place contained no radioactivity. This is not a sampling phenomenon, because several hundred cells of the species are involved in the observations. Physical, chemical, and organoleptic tests are in progress to determine differences and similarities in the muds utilized by the species with radioactive nests vs the nonradioactive *Trypoxylon* nests. Field observations have established the usual flight range of *Sceliphron* as 150 ft or less, with an inferred maximum of 650 ft. No direct observations of *Trypoxylon* flight range were possible.

The wasp nest building activities with radioactive mud constitute a definite hazard, particularly because of the inclination of the wasps to build nests out of sight within open pipes, behind wired panels, inside small instrument boxes, and in numerous other places where they are not seen and where decontamination becomes virtually impossible. So long as suitable nesting sites are available near the source of their mud, the daubers will probably not contaminate objects farther away. The most satisfactory preventive action is to apply window screening over any opening more than 1/8 in., and this must be a flush fit. If the screened opening is protected from rain or is dark or shaded, it would be advisable to spray the screening with a 5 to 10% DDT preparation because the wasps will otherwise attach nests to the screen itself. Cabinet doors must fit absolutely flush. This is achieved by fitting a gasket of a sponge or foam elastomer so that closure puts a slight pressure on it.

Gamma emitters in the nests were identified as Cs^{137}, Rh^{106}, Co^{60}, Zn^{65}, and Ce^{144}. Activities up to 361 μc/g were obtained among the first 50 nests assayed. Neither live nor dead specimens of *Sceliphron* at the sites of radioactive nests bore radioactive contamination. Spiders stored in highly radioactive nests were nonradioactive, as were larvae growing within radioactive cells. Approximate calculations of the median radiation dose received by various life stages of the *Sceliphron* and *Chalybion*

wasps in radioactive nests are: egg, 3 days, 30 r; larva, 2 weeks, actively eating spiders, 140 r; larva, 4 days, resting and cocoon spinning, 40 r; larva, temporary summer resting stage, 10 days, 100 r; pupa, temporary summer stage, 10 days, 100 r; larva, overwintering form, 9 months, 2700 r; pupa, spring form, 14 days, 140 r; adult, 3 months, estimated at 1 mr/hr, 2 r. An individual wasp will have a total dose of 3022 r. Gross effects of radiation on larvae contained in the radioactive nests have not yet been determined.

Electrophoretic Analyses of Wild-Mammal Blood. — Analyses of blood serum proteins of wild mammals by means of electrophoresis provided data on percent total serum proteins in each fraction, albumin-globulin ratios, and relative mobilities for the various moieties. Sera were analyzed for 115 individuals belonging to 16 species, 6 families, and 4 orders. Certain taxonomic relationships are readily apparent; for instance, the three species of Sciuridae sampled all had serum patterns containing albumin and α_1, α_2, β_1, β_2, and γ globulins, whereas all species of another rodent family, Cricetidae, had the basic pattern of an albumin, a γ globulin, and at least one α and one β globulin. The average albumin percentage of all cricetids was within the range from 50 to 55%, whereas the range for the sciurids was only 31 to 34%.

Average reduction in percent albumin for bot-infested white-footed mice was 19%, and the increase in globulin percentages was nonspecific. The coefficient of correlation between total white cell count and albumin-globulin ratio in this species was -0.360 ($0.057 > P > 0.02$). No changes were detected in the serum proteins of 17 cotton rats irradiated with an acute dose of 250 rads.

Electrophoretic analyses may be a valuable adjunct to other hematological studies in progress, because it appears that certain changes in cell counts, cell types, and hemoglobin percentages may be correlated with changes in serum proteins. Accordingly, electrophoretic measurements have been incorporated into the hematological studies currently proceeding during the seasons of the year to ascertain changes caused by environmental stresses.

Incorporation of 5-Iodo-2′-Deoxyuridine-I^{131} into the DNA of Indigenous Mammals. — The chemical 5-iodo-2′-deoxyuridine-I^{131} (IUDR) is an antimetabolite, which, upon in vivo injection, is incorporated into the DNA (deoxyribonucleic acid) of rapidly proliferating cells and serves as an index to cell proliferation. During the summer of 1963 an experiment was performed with three types of mammals: *Sigmodon hispidis*, cotton rat (12); *Rattus norvegicus*, brown rat (21); and Sprague-Dawley strain laboratory rat (24). The objectives of this study were to establish normal IUDR retention curves for each animal and to determine whether animals with higher metabolism have higher rates of cell proliferation.

The 57 experimental animals were injected with IUDR and counted in a small-mammal whole-body counter. Retention curves were plotted on a semilogarithmic scale from the counter data. Each curve had approximately two major components. The first component had a steep slope that reached a point of inflection at 5 to 10% retention in about 24 hr. This component corresponded to an elimination phase in which most of the unincorporated IUDR was broken down and eliminated. Within 24 hr after injection

the retained IUDR was presumably incorporated into the DNA of a cohort of cells and lost at a constant rate only at cell death. Thus the second component of the curves was a line of less extreme slope and indicated the rate of cell death. Since the animals were apparently maintaining their cell numbers, the curves of cell death were used as an index to cell proliferation.

The first components of the retention curves for the three mammals had about the same slope. However, the point of inflection was higher in cotton rats (mean weight, 89.5 g), intermediate in brown rats (268.0 g), and low in laboratory rats (412.5 g). The degree of slope for the second component of the curves decreased in a corresponding order. It appeared that animals with higher metabolism (lower body weight) incorporated more IUDR, lost it faster, and apparently had higher rates of cell proliferation.

Ribonucleic Acid Characteristics in Mimosa. — Mimosa seedlings (*Albizzia julibrissin* Durazzini) were grown under greenhouse conditions for a period of 12 days. The epicotyls were excised and homogenized for RNA (ribonucleic acid) extraction. Various phenol and NaCl procedures were attempted, with a modified procedure of Ofengand *et al.* yielding the best quantitative results. Total RNA was further separated on a DEAE (diethylaminoethyl) cellulose column in order to prepare a pure sample of SRNA (soluble RNA). This fraction consists of RNA in the lower molecular weight range and is the fraction responsible in vivo for carrying amino acids to the site of protein synthesis within the cell.

Several control samples were run, and the percentage of SRNA to total RNA ranged from 20 to 30%. (Total RNA figures were obtained by using KOH hydrolysis and reacidification.) An ultraviolet light absorption analysis in the range 220 to 290 mμ yielded a ratio of optical density at 290 to optical density at 260 between 0.48 and 0.49 for all samples. This figure indicates a complete lack of protein contamination. A modified diphenylamine test was next used, and all samples yielded a DNA (deoxyribonucleic acid) content below 25% of the total SRNA, which could very likely be caused by NaCl present and not actually DNA. Finally, a Beckman model E analytical ultracentrifuge was used to determine the sedimentation rate of the SRNA. This value was calculated at 3.18, which falls nicely within the range of SRNA sedimentation rate values for animal and bacterial material.

Base ratios were determined by use of HCl hydrolysis and paper chromatography, followed by elution of the nucleotide spots from the paper and determination of nucleotide absorption of ultraviolet light at 260 mμ. Four molar base ratios have been determined from different samples of SRNA, and all have yielded the following ranges of figures based on adenine as 10.0: cytosine, 17.6 to 19.5; guanine, 17.8 to 21.2; and uracil, 9.0 to 10.8. Amino acid acceptor tests will be run on all samples. These results plus all the above-mentioned data for controls will be compared with data for mimosa seedlings of comparable age irradiated with sublethal levels of both gamma rays and fast neutrons.

Transfer of Cs^{137} from Tagged Tulip Poplar Trees to the Untagged Understory. — Study is continuing within the Cs^{137}-tagged tulip poplar forest. One of the main objectives is a quantitative description of the pathways of transfer of the radionuclide. There are two primary paths

for transfer. The most direct is the interception and subsequent foliar uptake by the untagged understory. The second pathway is the transfer of the Cs^{137} from tagged trees to the forest floor (via rain leaching of canopy components, roots, and litter fall) and subsequent root uptake by the understory plants.

A 5-ft red maple (representing the larger woody understory plants) was harvested in March, ten months after tagging, and each plant component was radioassayed for Cs^{137}. The root/shoot ratio, (dis/min per gram of root)/(dis/min per gram of shoot), was 0.339. Literature and collected data suggest that the ratio is 10 to 20 times this value in cases where the uptake is from solutions or soil to roots to tops.

Also, the forest floor area around the harvested plant was sampled to determine the radiocesium concentration of the plant's growing medium (soil). The plant/soil ratio, (dis/min per gram of plant above ground)/(dis/min per gram of soil), for the harvested red maple was 2.50. Shanks and DeSelm reported that the plant uptake of radiocesium was ~0.0077 for White Oak Lake bed, and Witherspoon reported ~0.0085 for oak seedlings on a colluvial dolomitic limestone soil. Even if the plant uptake is greater in this forest than it was in either of the above cases, it is expected that it will remain about 0.01. Comparing the ratios of 2.50 and 0.01 suggests that foliar absorption may be as much as 250 times as important as root uptake after ten months.

Twenty samples (each with an area of 0.1 m^2) were taken in August 1963 to ascertain the amount of transfer of Cs^{137} into the small groundcover plants. Soils within each 0.1-m^2 ring were sampled to obtain the radiocesium concentration of the plant growing medium. Eight species show plant/soil activity ratios ranging from 0.23 to 6.7 and suggest that foliar absorption may be 50 to 400 times root uptake for most undercover species near the end of the second growing season.

Detritus in the Aquatic Food Chain. — One phase of the study of detritus (particulate organic matter) in the aquatic food chain is concerned with determining the feeding rate of organisms which normally feed on detritus. This can be accomplished with radionuclides when the following information is known: (1) the fraction of an ingested radionuclide which is deposited in the organism, (2) the rate of radioactive decay, (3) the biological turnover of the radionuclide, and (4) the quantity of radionuclide occurring in the organism under natural conditions, for example, such as might occur in organisms living in White Oak Lake or the Clinch River. Datum 4 can be determined by counts of radioactivity of organisms taken from their natural habitat. A laboratory study has been initiated to determine the uptake (1) and the biological half-life (3) of Cs^{134} in carp, *Cyprinus carpio*, as influenced by water temperature.

Two groups of carp, with ten fish in each group, were fed detritus tagged with Cs^{134}. One group of fish was held at 14°C and the other at 20°C. Radioactive cesium was selected as the isotope, since this element enters freshwater fish primarily through ingested food, and because Cs^{137} is one of the radionuclides present in White Oak Lake and, in lesser concentrations, in the Clinch River.

Although the experiment is still in progress, some results are apparent now. Carp held at the colder temperature remove less Cs^{134} from the detritus (6% vs 8% in the warmer water), but retain the food in the

gut for a longer period (18 to 26 hr vs 10 to 20 hr in the warmer water). The biological half-life of Cs^{134} in the fish appears to be about 34 days in the cold water and about 22 days in the warmer water.

Evaluation of Fission Product Distribution and Movement in White Oak Drainage System. — To determine the rates and directions of groundwater movement in the waste-pit area, a tracer test, using tritium, has been initiated. A total of 21 curies of tritiated water was injected into five 40-ft-deep holes, drilled on 1-1/2-ft centers, along a line estimated to be at right angles to the strike of the geologic formation (Conasauga shale) which underlies the area. By periodic sampling and analysis of the water from a series of observation wells, the rates and directions of movement of the tracer are being determined. At the 5-ft-distant observation wells along the strike, tritium was detected within 24 hr after injection; at the 10-ft-distant observation wells, which are located in the same direction, the tritium was first detected after about four days. The concentration of tritium in the groundwater at these points was still increasing eight days after the injection of the tracer. These preliminary data indicate that the maximum rate of travel of groundwater in the pit area is much greater than the average rate of movement; however, it is too early to draw any firm conclusions.

RADIOLOGICAL PHYSICS, HEALTH PHYSICS, AND RADIATION INSTRUMENTATION

Radiation Physics and Dosimetry: Optical Absorption in the Alkali Metals. — Until about a decade ago, absorption of photons in the alkali metals was interpreted chiefly in terms of the same mechanisms giving rise to dc resistivity. It was recognized at an early stage that measured optical absorption is considerably larger than would be predicted on this basis. The contribution of electron-electron collisions to the photon absorption process in a free electron gas has been considered, and it has been shown that the absorption cross section [Phys. Rev. 124, 1297 (1961)] for such a system may be derived by a simple semiclassical argument involving the response of quasi particles to a transverse electromagnetic wave. Absorption due to the anomalous skin effect and to the excitation of surface oscillation has also been considered on this basis. Calculations of surface optical absorptivity made with this approach compare favorably with experimental data on the alkali metals.

Radiation Physics and Dosimetry: Low-Energy End of the Secondary Electron Cascade in Metals. — The processes by which electrons having energies of the order of tens to hundreds of electron volts lose energy in metals are of considerable interest in connection with secondary electron emission and with observations of electron energy distributions in media containing radioactive sources. Interaction cross sections have been used for electrons in a degenerate free electron gas in order to approximate cross sections for conduction-band electrons in real metals. The pertinent cross sections are those appropriate to single-particle excitations and to the excitation of plasma oscillations in the free electron gas. These cross sections have been computed and used in a numerical solution of the slowing-down and cascade spectra resulting from electrons

which are low enough in energy that inner-shell excitations are not possible. These spectra, corrected for reflection at the surfaces, have been compared with experimental data on secondary electron energy distributions from metals.

Radiation Physics and Dosimetry: Interaction of a Stationary Plasma with an Electron Beam. — Investigation has been made of a plasma-beam system in which an electron beam of small intensity moving with velocity βc (c is the velocity of light) interacts with a cold plasma immersed in a magnetic field. It is assumed that the beam and the excited wave resulting from the interaction are aligned along the direction of the magnetic field and that $(1 - \beta)^2 \gg 1/\eta^2$, where η is the ion-to-electron mass ratio. It was found that the frequency $|\tilde{\omega}|$ of the excited wave is $|\tilde{\omega}| = \Omega_i[1 - (1 + 4R)^{1/2}]/2R$, where $R = A^2\beta^2/\eta^2(1 - \beta^2)$, $A = \eta\omega_e^2/\Omega_e^2$, ω_e is the plasma frequency, and Ω_i and Ω_e represent, respectively, the ion and electron gyrofrequencies. An expression was obtained showing explicitly the relationship between the rate of growth $|\mathrm{Im}\ \delta|$ of an excited wave and the velocity β of the beam. It was shown that $|\mathrm{Im}\ \delta|$ attains a maximum for an appropriate value $\beta = \beta_m$ and that β_m increases with A/η. For many typical plasmas, one has $A/\eta \ll 1$, and in such cases the maximum is attained for beam velocity β_m in the relativistic range. One obtains then $|\mathrm{Im}\ \delta|_{max} \sim 0.4\sigma^{1/2}A\Omega_i$, corresponding to a frequency $|\tilde{\omega}| \sim 0.59\Omega_i$ (where σ is the fractional part of the plasma density represented by the density of the beam). When $R \ll 1$, the electron beam excites ion cyclotron waves and in such cases $|\tilde{\omega}| = \Omega_i(1 - R)$; that is, the quantity R represents the departure in frequency from the condition of gyroresonance. The relationship between the rate of growth of an ion cyclotron wave and the beam velocity can be expressed as $|\mathrm{Im}\ \delta| = \sqrt{2}\ \sigma^{1/2}A\Omega_i\beta$.

Radiation Physics and Dosimetry: Plasma-Beam Instability in the Hartree-Appleton Approximation. — The interaction of an ion beam of small intensity moving with velocity $c\beta$ (where c is the velocity of light) through a stationary plasma has been investigated under the assumption that the beam and the waves resulting from the interaction were aligned along the direction of an impressed static magnetic field and that the frequencies of the excited waves were sufficiently high so that the motion of the ions perturbed by these waves could be neglected. It was found that the beam is capable of exciting waves only if $\beta < \beta_{max}$, where $\beta_{max} = [\eta(\eta + 4A^2)]^{1/2}$, η is the ion-to-electron mass ratio, $A^2 = \eta\omega_e^2/\Omega_e^2$, ω_e is the plasma frequency, and Ω_e is the electron gyrofrequency. For each value of $\beta < \beta_{max}$ there are two excited waves having frequencies $\omega_I = \Omega_e(1 + R)/2$ and $\omega_{II} = \Omega_e(1 - R)/2$, where $R = [1 - 4\beta^2A^2/(1 - \beta^2)\eta]^{1/2}$. The rates of growth $|\mathrm{Im}\ \delta_I|$ and $|\mathrm{Im}\ \delta_{II}|$ corresponding to the frequencies ω_I and ω_{II} are

$$|\mathrm{Im}\ \delta_I| = \sigma^{1/2}\omega_e(1 - \beta^2)(1 - R)/2\eta^{1/2}A(1 - R\beta^2)^{1/2}$$

and

$$|\mathrm{Im}\ \delta_{II}| = \sigma^{1/2}\omega_e(1 - \beta^2)(1 + R)/2\eta^{1/2}A(1 + R\beta^2)^{1/2}$$

respectively. Certain characteristic features of the above instabilities were investigated for a "relatively dense" plasma ($A > 1$) and for a "rarefied" plasma ($A \leq 1$). In a "relatively dense" plasma for which $A \ll \eta/4$ a low-velocity ion beam characterized by $(1/A) \ll \beta^2 \ll 1$ excites simultaneously an electron cyclotron wave and a low-frequency wave similar to the one associated with whistling atmospherics. For sufficiently low velocities of the beam, the rate of growth of the low-frequency wave is considerably higher than the rate of growth of the electron cyclotron wave.

Radiation Physics and Dosimetry: Optical Constants of Vacuum-Evaporated Silver Films. — The optical constants of vacuum-evaporated silver films have been determined in the wavelength region 2500 to 4500 A from reflectance measurements made at various angles of incidence. These data were obtained to investigate the discrepancy between the experimental optical emission studies from electron-bombarded thin silver foils and the transition-radiation theory of Ritchie and Eldridge [Phys. Rev. 126, 1935 (1962)] when the optical constants of bulk silver were used as input data. The discrepancy of about 30% in the photon intensity at the peak of emission is removed when the optical constants of evaporated films are used. These optical constants also predict accurately the resonance absorption found in the transmission of thin silver foils [Bull. Am. Phys. Soc. 8, 392 (1963)].

Radiation Physics and Dosimetry: Transition Radiation and Optical Bremsstrahlung from Electron-Bombarded Thin Gold Foils. — Unbacked gold foils 340 and 530 A thick were bombarded by a 1.5-μa electron beam of energy 25 to 100 kev. The light emitted at 30° from the foil normal on the beam exit side was analyzed between 2500 and 5500 A with a vacuum ultraviolet spectrometer and a Glan prism polarizer. The spectral distribution and absolute photon yield in the plane containing the electron beam and photon direction agree well with the transition-radiation theory [Phys. Rev. 126, 1935 (1962)]. Due to optical absorption and refraction, the photon intensity perpendicular to this plane was only 40% of the bremsstrahlung theory [Phys. Rev. 90, 1030 (1953)] for isolated atoms. Calculations of the bremsstrahlung yield which include these effects in a dielectric medium are presently being carried out.

Radiation Physics and Dosimetry: Angular and Spectral Distributions of Transition Radiation from Thin Silver Foils. — The angular and spectral distributions of photons emitted by silver foils 660 and 1980 A in thickness when bombarded by 40-kev electrons have been determined experimentally in the wavelength region from 2500 to 5600 A. The spectral distribution of light polarized in the plane of incidence showed a sharp peak at 3300 ± 12 A, this value being an average over photon directions from 10 to 50° from the foil normal. In addition to the peak, the spectrum showed a continuum which decreased slowly with increasing wavelength and a deep minimum at 3200 A with a rise in the shorter wavelengths. The angular distribution of photons emitted at the peak wavelength showed a maximum at 30° from the foil normal with zero intensity at 0° and near 90°, whereas the photons emitted at other wavelengths in the continuum, for example, 2700 and 4500 A, were most intense at 50° from the foil normal. The absolute photon yield was determined by calibrating the spectrometer, analyzer, and photomultiplier with a tungsten filament lamp

obtained from the U.S. National Bureau of Standards. For photon directions from 10 to 40° the photon yield was found to agree with the theoretical predictions in all respects at all wavelengths except at that of the sharp peak, where the experimental values were about 30% lower.

Radiation Physics and Dosimetry: Interaction of Low-Energy Electrons with Other Polar Molecules. — Electron attachment coefficients and drift velocities have been measured in mixtures of H_2O vapor with N_2, CH_4, C_2H_4, and CO_2. Electron capture takes place only in the H_2O-CO_2 mixture, and even in this case the attachment coefficient is quite small. Thus, low-energy (less than 1 ev) electrons do not, in general, form stable negative ions with H_2O. Electron drift velocities are very sensitive to the addition of small quantities of H_2O vapor in all the mixtures. When certain other polar molecules (acetone, heavy water, methyl alcohol, dimethyl ether, hydrogen sulfide, toluene, and nitrous oxide) were mixed with C_2H_4, it was found that the decreases in drift velocity correlated well with the magnitude of the electrical dipole moments. The drift velocity data were analyzed to obtain the cross sections for momentum transfer. These cross sections are in fair agreement with the theory of Altshuler [Phys. Rev. 107, 114 (1957)], except for H_2O, D_2O, and H_2S, where the experimental values are a factor of 2 larger than the theoretical values. This discrepancy may be due to molecular rotational enhancement of the momentum transfer cross section, or it may be due to the temporary formation of negative ions, an effect which would decrease the observed drift velocity.

Radiation Physics and Dosimetry: Time-of-Flight Investigations of Electron Transport in Gases. — Electron swarm experiments may be arranged to satisfy the boundary conditions applicable to a one-dimensional time-dependent transport equation. Under such conditions it may be shown that the distribution of the times of arrival of individual electrons at a point on a plane which is L cm from another plane releasing electrons at time t = 0 may be interpreted in terms of the diffusion coefficient D and the drift velocity w. The integrals of the distribution functions, that is, the total numbers of electrons arriving before and after the addition of an attaching gas, are directly related to the attachment coefficient β. Since, in such experiments, time is a more accessible independent variable than length, it is not surprising that much more information on the basic character of electron transport in gases is obtained than can be obtained from a Townsend experiment [Phil. Mag. 44, 1033 (1922); Electrons in Gases, Hutchinson's Scientific and Technical Publications, London, 1948]. Results for values of D, and w as a function of E/P, have been obtained for C_2H_4 and CO_2.

Radiation Physics and Dosimetry: Swarm Measurement of Cross Sections for Dissociative Electron Capture in Heavy Water, Chlorobenzene, and Bromobenzene. — One established method for the measurement of the cross section for electron capture involves the introduction of small quantities of the gas under study (sample gas) into a carrier gas, which determines the distribution of electron energies for specified values of E/P. A summary of the available information on the distribution of electron energies in ethylene, nitrogen, and argon shows that for E/P values ranging from 0 to 2.0 v cm^{-1} $torr^{-1}$ the three carrier gases can be used to cover the important range of electron energies between thermal and 10 ev. This

method has been used to study dissociative electron capture in D_2O, C_6H_5Cl, and C_6H_5Br. Measurements show that the values for the energy-integrated dissociative capture cross sections are approximately 4.6×10^{-18} cm^2-ev for D_2O, 1.4×10^{-17} cm^2-ev for C_6H_5Cl, and 5.7×10^{-17} cm^2-ev for C_6H_5Br occurring at electron energies near 6.5, 0.76, and 0.70 ev respectively. That the low-energy processes in C_6H_5Cl and C_6H_5Br correspond to dissociative capture has been confirmed by the appearance of Cl^- and Br^- in a negative-ion time-of-flight mass spectrometer.

Radiation Physics and Dosimetry: Optical Emission from Argon Excited by Alpha Particles. — Optical emission from argon gas, excited with alpha particles, has been studied in connection with the suggestion made in ionization studies [Phys. Rev. 93, 643 (1954); ORNL-3422] that electronic states of long lifetimes are excited by swiftly moving charged particles. A continuum was observed which could be separated into three distinct bands with maximums at 1300, 1800, and 2200 A. The light intensity of the 2200-A band was readily quenched with the addition of about 2% of CH_4, CO_2, O_2, Kr, and N_2. The 1800-A band was quenched more slowly than the 2200-A band by the impurity gases. The results support the notion that alpha-particle excitation of argon at high pressures leaves the argon in long-lived states around 11.5 and 15.5 ev. The observed continua appear to arise mainly from Franck-Condon transitions between two excited states and the ground repulsive state of the argon dimer which forms as a consequence of the long lifetimes of the excited atoms.

Health Physics Research Reactor Studies. — Pulsed operation of the HPRR was initiated in mid-November 1963. Less than 25% of the total kilowatt hours during 1964 is expected to be assigned to pulsed operation. Approximately one-third of all reactor operation has been for radiobiological studies, one-third has been for dosimetry research, and the remaining one-third has been used for numerous types of studies. During 1963, four foreign research groups utilized the HPRR in their studies.

ISOTOPE DEVELOPMENT

Nuclear Analysis Research and Development. — Techniques for the determination of submicrogram amounts of copper in biological materials (particularly freshwater snails) are being studied. A most difficult problem in this analysis is the interference of radioactive Na^{24}, which is produced in large amounts by the neutron irradiation of the sample. We are using an anion exchange method of separation to remove the bulk of the Na^{24}, followed by gamma-ray spectrometry and instrumental subtraction techniques to correct for the interference of the residual Na^{24} in the determination of Cu^{64}. The combined techniques result in a 300-fold increase of the ratio Cu^{64}/Na^{24}, thus bringing about a higher degree of sensitivity and accuracy in the measurement of the copper content of biological materials.

Recovery of Fission Products. — Study of extractions of the rare earths with di(2-ethylhexyl)phosphoric acid (D2EHPA) showed that the slow extractions from tartrate-complexed Purex 1WW solution at pH about 5 are

due to effects of certain metal contaminants in the solution. Iron, aluminum, and especially chromium (contaminants were added one at a time to the solution) decreased the extraction rates of the rare earths, whereas nickel had no effect. Analyses of the solvent phases for the contaminant metals showed that the amounts extracted were not large enough to represent significant competition for the D2EHPA extractant. The magnitude of the effect of each interfering metal was different for the different rare earths, iron depressing the extraction more for europium and neodymium than for cerium, and chromium depressing the extraction more for cerium than for europium. These and previous results suggest that the aqueous phase contains complicated and unextractable complexes (of the rare earths, contaminating cations, and tartrate), which dissociate slowly to yield extractable rare-earth species.

Radioisotope Research and Development. — The half-life of Cs^{137} was measured by three independent methods using $Cs^{137}Cl$ recovered from a 9.5-year-old teletherapy source prepared for ORINS. The values and method of measurement were (1) 30.17 ± 0.96 years by measuring the changes in the ratios Cs^{137}/Cs^{133} and Cs^{137}/Cs^{135}, (2) 29.77 ± 0.09 years by determining the amount of Ba^{137} produced from the decay of Cs^{137}, and (3) 29.84 ± 0.04 years by determining the specific activity of a highly purified cesium salt.

Refinements were made in the cesium tetraoxalate process used in the Fission Products Development Laboratory for Cs^{137} purification and recovery. Cesium-137 recoveries were increased from 60–65% to 88–92% by adding NH_4OH to the cesium solution. The quality of the $Cs^{137}Cl$ product was not altered by the NH_4^+ addition, which resulted in a considerable saving in the cost of recovery and reprocessing of the soluble cesium fraction.

The thermal power of Pm^{147} (2450-curie source) was determined by calorimetric measurement and 4π beta proportional counting to be 0.414 ± 0.005 w/kilocurie. From these data the average beta energy of Pm^{147} was calculated to be 70 ± 2 kev.

An aged sample of Pm^{147} containing $<3.0 \times 10^{-7}$ curie of Pm^{146} and Pm^{148} per curie of Pm^{147} was examined for evidence of a primary gamma ray from the decay of Pm^{147}. A photon was found which has an energy of ~0.124 Mev resulting from 0.018% of the total disintegrations of Pm^{147}.

Radioisotope and Stable Isotope Production. — Ion currents equivalent to 1994 g of separated isotopes were monitored during November and December. Isotopes of Ca, Si, S, Rb, Cr, and Pb were separated and collected by using 28 of the calutrons associated with the program.

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