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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

DECEMBER 1962

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OAK RIDGE NATIONAL LABORATORY

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Oak Ridge, Tennessee
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CONTENTS

REACTOR DEVELOPMENT PROGRAM..... 1
GAS-COOLED REACTOR PROGRAM..... 1
EGCR Components..... 1
Evaluation of PTM Primary Nozzles for the EGCR..... 1
Mechanical and Physical Properties Studies of Graphite..... 1
Instantaneous Fission-Gas Release from UO_2..... 2
Irradiation Testing of Structural Graphite..... 2
Geometry Studies of Cylindrical Bodies..... 2
GCR-ORR Loop No. 1..... 3
GCR-ORR Loop No. 2..... 3
Advanced Fuel Element Mass Transfer and Pressure-Drop Experiments..... 3
Long-Rod-Cluster Pressure-Loss Experiment..... 3
Behavior of Beryllium Oxide Under Irradiation..... 4
Alternate Gas Coolants..... 4
MARITIME REACTORS PROGRAM..... 4
Reactor Compartment Filter Tests..... 4
ARMY POWER REACTORS..... 5
Postirradiation Examination of SM-1 Fuel Element S-81..... 5
Impact Tests on Irradiated SM-1 Pressure-Vessel Steel..... 5
FUNDAMENTAL HEAT TRANSFER AND FLUID DYNAMICS..... 5
Reynolds Stress Measurements..... 5
Transient Heat Transfer..... 6
Two-Phase Hydrodynamic Studies..... 6
Thermal-Conductivity Determinations of Molten Lithium..... 6
Boiling-Potassium Studies..... 7
POWER REACTOR FUEL PROCESSING..... 7
Fluoride Volatility Processing..... 7
Corrosion..... 7
High-Capacity Interim-23 Flowsheet..... 8
Protactinium Adsorption..... 8
Hydrolysis of Uranium Carbides..... 8
Dissolution of PuO_2 Pellets..... 9
Shear-Leach Process..... 9
Chemical Analysis of Advanced Reactor Fuels..... 9
THORIUM UTILIZATION PROGRAM..... 10
Fuel Cycle Development: Chemistry of Solid Fuel Element Materials..... 10
Fuel Cycle Development: Fuel Preparation..... 10
MOLTEN-SALT REACTOR PROGRAM..... 11
Molten-Salt Reactor Experiment..... 11
Reactor Analysis..... 11
Development of Components and Systems..... 12
MSRE Pump Program..... 13
Instrumentation Development..... 13

[redacted] [redacted]

[redacted] [redacted]

NUCLEAR SAFETY..... 14
Release of Fission Products on Out-of-Pile Melting of Reactor Fuels..... 14
Release of Fission Products on In-Pile Melting of Reactor Fuels..... 15
Fission Product Transport Evaluations..... 15
Melting Experiments in TREAT Facility..... 15
Characterization and Control of Accident-Released Fission Products..... 16
Nuclear Safety Pilot Plant..... 16
Reactor Containment Handbook..... 16
WASTE DISPOSAL DEVELOPMENT..... 17
Clinch River Study..... 17
Mineral Exchange Studies..... 17
High-Activity Waste Calcination: Corrosion..... 17
High-Activity Waste Calcination: Fixation of Waste in Glass..... 17
Disposal in Natural Salt Formations..... 18
Engineering, Economic, and Safety Evaluation..... 18
Foam Separation Studies..... 18
PHYSICAL RESEARCH PROGRAM..... 19
PHYSICS AND MATHEMATICS RESEARCH..... 19
High-Energy Physics..... 19
High-Energy Electronuclear Machines: Cyclotron Analogue II..... 19
High-Energy Electronuclear Machines: Proposed Mc^2 Facility..... 19
Basic Studies of Cryogenic Electromagnets..... 20
Multiparticle Medium-Energy Nuclear Physics: The 86-Inch Cyclotron..... 20
Multiparticle Medium-Energy Nuclear Physics: Heavy-Particle Physics..... 20
Physics of Fission..... 21
Scintillation Spectrometry and Instrument Development..... 21
High-Voltage Experimental Program..... 21
Electronuclear Machines: The Isochronous Cyclotron (ORIC)..... 22
Stable Isotopes Development..... 22
Special Separations..... 22
CHEMISTRY RESEARCH..... 23
Thorium Oxide Studies..... 23
Equipment Decontamination..... 23
Spectrophotometric Studies of Solutions at Elevated Temperatures and Pressures..... 24
Protactinium Chemistry..... 24
Surface Chemistry..... 25
Pyrolysis and Radiolysis of Organic Liquids..... 25
Instrumental Methods for Continuous Chemical Analysis... 25
Analysis of Molten Salts by Electrochemical Methods..... 26
Spectrophotometry of Solutions at High Temperature..... 26

[redacted] [redacted]

[redacted]

Radiation Chemistry: Radiolysis of Liquid Oxygen and Oxygen-Nitrogen at 77°K..... 27
Radiation Chemistry: Bromine Oxidizing Fragments Produced in Crystalline Alkali-Metal Bromates by Co^{60} Gamma Rays..... 27
Chemical Physics: Neutron Diffraction..... 28
Corrosion Tests for the TRU Facility..... 28
Target Preparation..... 28
METALLURGY AND MATERIALS RESEARCH..... 29
Preparation of Pure Materials..... 29
Fundamental Research in X-Ray Diffraction..... 29
Fundamental Investigation of Radiation Effects in Solids: Channeling of Energetic Atoms in Solids..... 29
Fundamental Investigation of Radiation Effects in Solids: Influence of Channeling on Displacement Cascade Theory..... 29
Fundamental Investigation of Radiation Effects in Solids: Effects of Neutron Irradiation on Strain-Amplitude-Dependent Internal Friction of Copper..... 30
CONTROLLED THERMONUCLEAR RESEARCH..... 30
Plasma Physics and Arc Research..... 30
DCX-1 Facility..... 30
DCX-2 Facility..... 31
Vacuum..... 31
BIOLOGY AND MEDICINE PROGRAM..... 31
TERRESTRIAL AND FRESH-WATER ECOLOGY..... 31
Evaluation of Fission Product Distribution and Movement in White Oak Drainage System..... 31
Radionuclides in Soils and Plants of Upper White Oak Lake Bed..... 32
Blood Responses of Mammals in Radioactive Waste Areas... 32
Varying Rates of Litter Breakdown in Contrasting Climates..... 33
Cs^{137} Transfers in Forest Floor Litter Layer..... 33
Cs^{137} Movement in Forest Food Chains..... 34
RADIOLOGICAL PHYSICS, HEALTH PHYSICS, AND INSTRUMENTATION.... 34
Aerosol Studies..... 34
ISOTOPE DEVELOPMENT PROGRAM..... 34
Recovery of Fission Products..... 34
Radioisotope Research and Development..... 35
Radioisotope and Stable Isotope Production..... 36
OPEN LITERATURE PUBLICATIONS..... 37
FOREIGN VISITORS..... 39

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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

DECEMBER 1962

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

REACTOR DEVELOPMENT PROGRAM

GAS-COOLED REACTOR PROGRAM

EGCR Components. — Welding of the EGCR main blower casings is proceeding. Static seal tests are scheduled for January. One of the vessel coolant blower casings was damaged in the shop, and the required repairs have necessitated an extension of the delivery schedule.

Manufacture of the TS-2, TS-3, and TL-4 tee sections and bottom closures is proceeding. All lower chase piping for these loops was installed and tested. Preliminary proposals were received on specifications for the emergency-cooling loop compressors.

The control-rod-drive testing facility was completed and is awaiting delivery of the test unit. Procurement and fabrication of equipment for a steam-graphite reaction test are proceeding.

Evaluation of PTM Primary Nozzles for the EGCR. — Pneumatic temperature measurement (PTM) devices, which combine the principles of gas thermodynamics and sonic aerodynamics, will be used to monitor the exit gas temperatures of individual fuel channels in the EGCR. The heart of this temperature-sensing device is a small type 304 stainless steel nozzle through which 1050°F coolant gas is passed at sonic velocity. To prevent erosion of the highly critical nozzle throat, surfaces exposed to the high-velocity gas will be protected by a 0.005-in. nitride coating.

Four samples of type 304 stainless steel rod nitrided in accordance with the specification to be followed for the EGCR nozzles were obtained from the PTM manufacturer. The samples are being used to evaluate the thermal and oxidation stability of the nitrided surfaces. The layers on three specimens were cut back by centerless grinding to expose varying depths of the nitride coating. Each of the specimens is now being subjected to an oxidation test in impure helium at 1050°F. The helium contains approximately 0.01% C, 0.113% CO_2, 0.17% H_2, 0.013% N_2, and 10 ppm H_2O and flows into the reaction chamber at a rate of 40 cm^3(STP)/min. Specimens will be removed after 1000 hr and examined metallographically.

Mechanical and Physical Properties Studies of Graphite. — Specimens taken from an EGCR graphite column (No. 266) were tensile-tested. Seven

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specimens from five axial positions showed that there is no typical section; the strength varies greatly in the axial direction. The average values obtained, however, were not too dissimilar from those obtained previously, except for a lower average Young's modulus value.

Instantaneous Fission-Gas Release from UO_2. — Specimens of thin-plate single-crystal UO_2 are being irradiated to study the fundamental mechanisms of fission-gas release. The fission-gas release from the single crystal decreased by a factor of 4 with burnup until an exposure of about 8×10^{19} neutrons/cm^2 or 9×10^{18} fissions/cm^3. After this burnup, the gas release was apparently stable.

The fission-gas release is temperature-dependent above about 600°C. The activation energy is about 56 kcal/mole. The isotopic composition of the fission gas is the same for both temperature-dependent and temperature-independent gas release. About 45% of the mass-133 and 77% of the mass-135 material that escapes from the specimen escapes as iodine. This proportion is the same as for sintered and fused specimens and is not temperature-dependent.

Measurements of fission-gas escape at different flux levels but at the same temperatures were performed. The non-temperature-dependent gas escape was directly proportional to the flux, but the temperature-dependent fractional gas escape was lower for the higher flux levels than for the lower flux levels, which implies that radiation damage retards the temperature-dependent fission-gas escape from UO_2.

Irradiation Testing of Structural Graphite. — An in-pile experimental assembly for studying graphite creep was inserted in the ORR poolside facility and is operating satisfactorily. In this test, nine parabolic graphite beams are loaded to stress levels from 500 to 1475 psi. Five of the nine transducers for measuring displacement are functioning perfectly, two are yielding intermittent signals, and two are yielding no signal at all. The two transducers that failed were of the variable permeance type, and the successful ones are simply variable resistors. The transducers have a sensitivity of 6×10^{-7} in./in. strain with an indicated accuracy of 1×10^{-5} in./in. strain.

The results for the first 250 hr of testing show that a large primary stage of creep occurs that is similar to the primary creep observed out-of-pile. The zero intercept (or the magnitude of the primary stage) is, however, larger by a factor of 2 to 3. The in-pile data are beginning to indicate second-stage creep, with rates ranging from 4×10^{-7} to 1×10^{-6} in./in. per hour. The zero intercept and the second stage of creep are demonstrating linearity with applied stress. This implies that the stress distribution in the beams retains the original elastic linear stress distribution. At the highest rate given above, 4000 hr of testing will be necessary to obtain the desired 0.5% overall strain.

Geometry Studies of Cylindrical Bodies. — Cylindrical bodies subdivided into sectorial units are often considered for use in moderator structures when the cross-sectional dimensions of the cylinders required are large, as in pebble-bed reactors. The adoption of sectors allows for a reduction in extrusion or molding size over that required for a complete cylinder, and a reduction in maximum stress may also be realized. Hence, studies were made on the effect of fast-neutron exposure on cylinders with

a central channel and on sectors of these cylinders. The fast-neutron exposure distribution used is given by

$$\phi = 1.12 \times 10^4 \, e^{-0.26d} ,$$

where d is inches from the inside surface of the cylinders, and ϕ is exposure in Mwd per adjacent ton per year. Two cylinders with an inside diameter of 30 in. were considered. In the first case, the outside diameter was 54 in., and in the second case it was 90 in.

It was found that the principal in-plane stresses were reduced significantly by subdividing the cylinders. The percentage reductions in stresses between a complete cylinder and a 90° sector were larger for the cylinder with the smaller outside diameter. Because the sectors were assumed to be constrained against warping or bowing as a result of axial stress differences, the reductions in axial stresses were small, and the maximum axial stress could not be less than about 4000 psi for the first cylinder. Thus, sectors must be free to deform to derive full benefit from stress reductions associated with this subdivision.

GCR-ORR Loop No. 1. — A new fuel element having a roughened surface was installed in GCR-ORR loop No. 1 with thermocouples for measuring both the gas inlet and outlet temperatures. This element is performing well, and the gas temperature data tend to confirm the assumption that the No. 8 element operated satisfactorily.

GCR-ORR Loop No. 2. — Design of the fission product deposition portion of the fuel holder assembly for GCR-ORR loop No. 2 is nearly complete. Fabrication is continuing on the first fueled assembly. Loop shakedown tests are continuing.

Advanced Fuel Element Mass Transfer and Pressure-Drop Experiments. — Fabrication of the test apparatus was completed, and preliminary operation was accomplished. The apparatus consists of seven 0.8-in.-diam rods 72-1/4 in. long mounted with the peripheral rods on a bolt-circle diameter of 2.667 in. in a 4-in.-ID channel. At 14-1/2-in. intervals, spacers (similar to the EGCR midcluster spacers) are attached to the cluster, resulting in five cluster sections. Mass transfer and pressure-drop determinations can be made on the fourth section from the inlet end by using bare rods or various degrees of spiral wire wrapping. Preliminary pressure-drop data for bare rods indicate that the pressure-loss coefficient per section is of the order of one-half for a Reynolds modulus range of 20,000 to 80,000.

Long-Rod-Cluster Pressure-Loss Experiment. — An apparatus was fabricated for studying the effects of surface roughness on pressure-loss coefficients in long rod clusters. The apparatus consists of two seven-rod clusters in series. The rods are 0.75 in. in outside diameter and are mounted on a 2-in. bolt-circle diameter in a 3-in.-ID Plexiglas channel. Each cluster is ~52 in. long, with no midcluster spacers or other obstructions between the end spiders, resulting in a length-to-equivalent-diameter ratio of 85:1 between obstructions (as contrasted to ~22:1 for the ratio between obstructions for the EGCR fuel-element cluster).

One of the clusters is provided with five piezometric rings so that entrance-region effects can be studied. Three types of rod clusters were

fabricated for study: (1) smooth, 0.75-in.-diam rods, (2) 0.75-in.-diam rods spirally wrapped with 0.006-in.-diam wire at a pitch spacing of 0.060 in., and (3) Whitworth-threaded rods at 32 threads/in. with a root diameter of 0.75 in.

Behavior of Beryllium Oxide Under Irradiation. — Assembly of the ORNL-41-10 experimental unit was completed, and the unit was shipped to Idaho for insertion in the ETR at the January 7, 1963, shutdown. This unit contains 1/2- and 1/4-in. BeO cylinders for irradiation at 100°C to an integrated (>1 Mev) dose of 10^{21} fast neutrons/cm^2 and will yield the data needed to correlate the information currently being accumulated at high temperatures. Also included in this unit are MgO compacts and single crystals of BeO, MgO, and Al_2O_3.

Alternate Gas Coolants. — The relative merits of He, H_2, CO_2, SO_2, SiH_4, CF_4, SiF_4, SF_6, and C_2F_6 were compared by using the GCR-3 as the base design. The coolants were evaluated only on the basis of their molecular weight, specific heat, thermal conductivity, and viscosity. The radiation and thermal stability and corrosion effects of the gases or their products were not considered. The hydrocarbons, which are known to be thermally unstable at the temperatures of interest, were not included. Chlorine compounds were omitted because of their high neutron absorption cross sections. It was found that H_2, SiH_4, CF_4, SiF_4, SF_6, and C_2F_6 were superior to helium, with SiH_4, SF_6, and C_2F_6 being the best.

MARITIME REACTORS PROGRAM

Reactor Compartment Filter Tests. — A fourth series of in-place tests was conducted on board the NS "Savannah" to determine the iodine removal efficiency of the reactor-compartment ventilation systems. Both the emergency and main ventilation systems were tested by the I^{127} test procedure. In this procedure nonradioactive iodine (I^{127}) is used, and the iodine content in the sample collectors is determined by activation-analysis techniques. This method was first employed during the third series of tests.

Duplicate tests were conducted in this series on both ventilating systems with the I^{131} tracer method as a check on the nonradioactive iodine test method. The I^{131} tracer method has been employed in each of the four test series conducted to date.

The tests of the emergency ventilation system were 2-hr tests at a system flow rate of 200 cfm. The results were:

	Test 1	Test 2
I^{131} test efficiency, %	99.97	≧99.97
I^{127} test efficiency, %	>99.99	≧99.95

The tests of the main ventilation system were 2-hr tests at a system flow rate of 4000 cfm. The results were:

	Test 1	Test 2
I^{131} test efficiency, %	98.6	
I^{127} test efficiency, %	98.5	99.1

To date, the correspondence between the results of the I^{127} and I^{131} tests has been acceptable. Additional experience is needed, however, to determine whether there is a significant variation in the iodine background with time and location of the ship. In addition, further development of certain phases of the testing procedure and slight modification of the sampling equipment appear to be desirable.

ARMY POWER REACTORS

Postirradiation Examination of SM-1 Fuel Element S-81. — Fuel element S-81, which was removed from the SM-1 (APPR) after 19.5 Mwyr of reactor operation, is being examined. This element is the third in a series of core I SM-1 fuel elements that have been subjected to postirradiation examination. The other two elements were S-72, removed after 10.5 Mwyr, and S-79, removed after 16.4 Mwyr. Preliminary findings on element S-81 are that the outer fuel plates exhibit greater rippling than noted on the previous elements; the thickness measurements of individual fuel plates vary proportionately with the axial fuel burnup distribution, with the maximum thickness increase being 10% (0.003 in. over the nominal dimensions) where the U^{235} depletion was found to be 62%; and no matrix cracks are present in the fuel cermet, but the frequency of clad cracking appears to be significantly greater than noted for elements S-72 or S-79.

Impact Tests on Irradiated SM-1 Pressure-Vessel Steel. — Two capsules containing pressure-vessel-steel surveillance specimens were removed from the SM-1. One was removed after 16.4 Mwyr, and the other after 20.8 Mwyr. These capsules each contain three subsize Izod impact specimens made of ASTM A212, grade B, steel with six notches in each bar. These specimens have been tested, and, at this time, it is thought that notch No. 2 of the impact specimens best represents the dose of the pressure vessel. The Charpy-V transition temperature increase for notch No. 2 after 20.8 Mwyr of reactor exposure was determined to be 92°F.

FUNDAMENTAL HEAT TRANSFER AND FLUID DYNAMICS

Reynolds Stress Measurements. — The constant-current hot-wire anemometer system assembled recently to replace the Lintronic Laboratories' constant-temperature anemometer was put into operation. With this instrument the average current can be held constant to ±0.05%; this has resulted in good reproducibility in the velocity sensitivity of the wire. The direction sensitivity of the wire was found to depend greatly on the existence of a clean, taut wire in the flow stream. Under these conditions the direction sensitivity follows the cosine law over a span of approximately ±60° from the direction of the mean flow.

A parabolic approximation of King's law was found to be an adequate basis for the linearization of the response with velocity for a range of about a factor of 2 in velocity. The first measurements of the components of the Reynolds stress tensor were made. These initial measurements involve only the normal components of the stress; the results have not yet been evaluated.

Transient Heat Transfer. — A hot-film surface probe was developed that enabled direct determination of the instantaneous rate of heat transfer and of the instantaneous coefficient of heat transfer at the surface of a pipe in which water was flowing turbulently. The probe consists of a very thin strip of gold, 0.005 in. wide and 0.250 in. long, vacuum-deposited across a pair of platinum electrodes cast in epoxy resin. The surface of the probe is accurately contoured to fit flush with the inside wall of the test section (2-in. pipe). By utilizing a constant-temperature hot-film anemometer circuit (Lintronic) to power the probe and by analyzing the voltage required to maintain the constant film temperature by means of the power spectral density analyzer previously described (ORNL-3351, p 8), the spectrum of instantaneous surface heat flux was recorded. This is also the spectrum of the instantaneous heat transfer coefficient, since the temperature difference is maintained constant by the electronic circuitry. The preliminary data are currently being reduced.

Two-Phase Hydrodynamic Studies. — A special probe was constructed to explore further the application of the capacitance change technique to two-phase flow studies. The probe consists of a 1/4-in.-diam hemispherical electrode attached by a Teflon bushing to the end of a 1/4-in.-OD tube that forms the ground electrode. The response of this probe to moving droplets of fluids having a wide range of physical and electrical properties was studied, and it was found that the volume of the droplets was the principal variable to which the probe was sensitive. This suggested that the probe might be useful for void-fraction determinations, and, indeed, measurement of the response of the probe to variations in void fraction (obtained by surrounding the probe with plastic washers punched out so as to have a known void fraction) yielded a nearly linear relationship over the void fraction range from 0.25 to 0.70.

Difficulty was experienced in utilizing this probe in an air-water system of low void fraction because of capacitive "shorting" of the probe by water bridging across the insulator, although the probe appears useful in the high-void-fraction region. A parallel-plate capacitance probe which, it is believed, will be less sensitive to the "shorting" effect of water and which may therefore be useful in the low- and medium-void-fraction region is under development.

Thermal-Conductivity Determinations of Molten Lithium. — A comparative, axial-heat-flow apparatus with compensating guard heating was developed for determining the thermal conductivity of molten lithium from 315 to 843°C. Over this temperature range the results can be represented to within ±2.2% rms deviation by the linear least-squares line,

$$k\ [\mathrm{w\ cm^{-1}\ (^\circ C)^{-1}}] = 0.343[1 + (8.851 \times 10^{-4})t]\ ,$$

where t is temperature in °C. The maximum error was estimated to range from ±8 to ±15% from the lower to the higher temperatures.

At the melting point (186°C) the present values, previous results, and predicted values agree to within ±7%. At temperatures above 550°C, however, the present results do not compare well with any previous experimental determinations but do compare well with predicted values and theory, particularly in temperature dependency. The temperature dependency of the present conductivity results is positive, which is contrary to the data for either molten sodium or potassium. This is consistent with the coefficient of volumetric thermal expansion of molten lithium being almost half that of both molten sodium or potassium.

From a broad extrapolation of the present data to the calculated critical conductivity, the conductivity of molten lithium appears to reach a maximum of 0.78 w cm^{-1} $(°C)^{-1}$ at 1700°C. This, of course, is only an estimate and will require more experimental data at high temperatures for verification. By substituting a niobium alloy for type 347 stainless steel, the present apparatus could be used to extend the conductivity determination to the normal boiling point (1317°C).

Boiling-Potassium Studies. — Additional data were obtained in the study of forced-convection saturation boiling of potassium in a vertical, copper-jacketed tube. For heat fluxes in the range 150,000 to 330,000 Btu hr^{-1} ft^{-2} and mass flows from 0.95×10^5 to 1.64×10^5 lb hr^{-1} ft^{-2}, it was found that the derived heat transfer coefficients were ~30% higher than those determined from earlier experiments. Any significance to be associated with this change is obscured by the progressively larger number of thermocouple failures (increasing the uncertainty in the knowledge of the boiling-surface temperature) over the lifetime of the boiler and by the recent separation between the copper blocks and the stainless steel tube in the central region of the boiler (restricting the usable boiler length to the final third of the tube).

The boiler is now being removed from the system and will be metallurgically examined to establish the final state of the copper—stainless steel bonds and the inside surface condition. A modified test section that will allow the indirect measurement of liquid temperatures at three positions along the boiler is nearing completion.

POWER REACTOR FUEL PROCESSING

Fluoride Volatility Processing. — Equipment for conducting differential thermal analysis of AlF_3-based fluoride mixtures was designed, constructed, and calibrated. Determinations are made of solid-state and liquid-solid phase transitions at temperatures as high as 1000°C. Provision was made to stir the molten mixtures. Calibration of the apparatus against a known solid-state transition in SiO_2 showed that the transition could be detected with an accuracy of ±1°C. With this apparatus a solid-state transition in AlF_3 was detected at 448°C.

Corrosion. — Corrosion studies in support of the oxidation—ash-dissolution process for graphite fuels and of the chloride volatility process were continued. After three cycles in air at 800°C followed by treatment in boiling 5 M HNO_3, specimens of Corronel 230 showed either slight weight gains or weight losses corresponding to a corrosion rate of less than 0.1

mil/month. After one cycle, type 304L stainless steel specimens showed weight losses corresponding to corrosion rates of 0.3 to 1.6 mils/month. In flowing 1 M HF–0.06 M H_2O_2 at the boiling point, Corronel 230 showed overall corrosion rates of 13, 8, and 7 mils/month for specimens exposed to the vapor, at the interface, and to the solution respectively; there was local pitting attack in welded areas.

In cyclic tests consisting of 5 hr in oxygen at 800°C followed by 5 hr in Cl_2-CCl_4 at 500°C, Nichrome V corroded at rates which increased from 5 or 6 mils/month for two cycles to 80 to 136 mils/month for 13 cycles. Exposure of Hastelloys B and C, "A" nickel, and Carpenter 20 to waste zirconyl chloride solution at room temperature resulted in corrosion rates of less than 0.1, less than 0.1, 0.4, and 0.6 mil/month during 264 hr of exposure. Types 304L and 309SCb alloys corroded at rates greater than 60 mils/month for 96 hr of exposure. At 50°C both Hastelloys continued to show rates of less than 0.1 mil/month for 96 hr exposure; rates for "A" nickel and Carpenter 20 were 10.5 and 9.0 mils/month respectively.

High-Capacity Interim-23 Flowsheet. — A laboratory experiment with 5% tributyl phosphate as the extractant in an Interim-23 flowsheet (acid) resulted in decontamination factors of 4×10^4, 8×10^3, 3×10^4, and greater than 1×10^5 from gross gamma, ruthenium, zirconium-niobium, and total rare earths, respectively, and a uranium loss of 0.1%. An experiment with 10% TBP resulted in the formation of a third phase. The extraction capacity provided by an Interim-23 flowsheet, using 2.5% TBP, as is normal, resulted in approximately the same capacity for UO_2-ThO_2 fuel elements as that provided by the Acid Thorex process in a plant optimized for the Purex flowsheet. These experiments indicate that 5% may be the maximum TBP concentration that can be used successfully with the Interim-23 process and therefore provide the maximum capacity for thorium processing in a given set of equipment. This capacity is about twice that achieved with the Acid Thorex process. The capacity for the two systems would be the same in equipment optimized for the extraction process used.

Protactinium Adsorption. — A series of 24-hr batch adsorptions on various amounts of 100- to 200-mesh unfired Vycor in contact with an 11 M HNO_3 solution of thorium (116 g/liter), uranium (5 g/liter), and protactinium (3.24×10^6 counts min^{-1} ml^{-1}) showed that the distribution coefficient of the protactinium was 3100; 5% of the protactinium was unadsorbable. Solutions containing 0.04 M F^- in addition to the other constituents gave coefficients of about 200, with none of the protactinium unadsorbable. The addition of 0.1 M Al^{3+} to complex the fluoride increased the coefficient to 2900, with 6% of the protactinium unadsorbable. When zirconium (0.01 g/liter) and niobium (about 0.01 g/liter) were added to the solutions, in addition to the fluoride and aluminum, the coefficient decreased to 1400, but the percentage of unadsorbable protactinium remained the same.

Hydrolysis of Uranium Carbides. — Basic chemical studies on the hydrolysis of uranium and thorium carbides in water and mineral acids were continued. Although the products of the hydrolysis at 80°C of as-cast uranium carbides of compositions between UC and $UC_{1.86}$ showed a systematic decrease in the methane concentration and an increase in the amounts of higher hydrocarbons, free hydrogen, and wax produced as a result of increasing carbon concentration in the alloy, the amount of methane evolved

was considerably less than expected from the calculated amount of UC present in a $UC-UC_{1.86}$ or a $UC-UC_{2.00}$ binary mixture. Since both x-ray and metallographic evidence indicated that the as-cast specimens having compositions between UC and $UC_{1.86}$ were binary mixtures of the mono- and dicarbide, it would appear that the carbon units in the monocarbide crystal react preferentially with hydrogen to produce methane during hydrolysis, but if dicarbide is also present in the solid, it reacts with other carbon units to produce higher hydrocarbons, and with hydrogen to produce methane.

Dissolution of PuO_2 Pellets. — The instantaneous dissolution rate of PuO_2 pellets (90% of theoretical density) in boiling mixtures of HNO_3-HF-$Al(NO_3)_3$ at an Al/F mole ratio of 3 is proportional to the 0.45 power of the hydrofluoric acid concentration.

Shear-Leach Process. — The UO_2-ThO_2 particles smaller than 9520 μ dislodged from 0.5- and 1.0-in. sheared lengths were 90.7 and 39.1 wt %, respectively, of the original core material in the assembly, compared with 84.5 and 35.4 wt % for UO_2. Particle-size measurements from 0.5- and 1.0-in. sheared lengths of stainless-steel-clad unirradiated ThO_2-UO_2 ORNL Mark I fuel assembly show that there are averages of 3.5 and 2.6 wt %, respectively, more core material found in the particle-size range less than 9520 μ to less than 44 μ than was observed on the shearing of UO_2-filled assemblies. Similarly, the amount of stainless steel cladding reduced to less than 1000 μ in size was 0.23 and 0.05 wt %, and 0.38 and 0.10% for ThO_2-UO_2 and UO_2 respectively.

The conveyor-feeder and leacher were decontaminated (from natural uranium) and dismantled preparatory to the installation of larger-diameter feed chutes on each unit and a steam jacket on the inner drum of the leacher. The ram of the shear was removed and was machined to larger tolerances to allow better adjustment of clearance between the fixed and moving blade with the wedge-shaped gibs. Inspection of shear components following 13 chop-leach runs with stainless-steel-clad UO_2 assemblies showed no evidence of wear on the stepped blade, gibs, liners, or connecting rod. The shear had made about 1600 cuts on UO_2-filled Mark I prototype fuel assemblies containing 36 tubes per assembly during the 13 runs.

Chemical Analysis of Advanced Reactor Fuels. — A method was developed for the determination of uranium by extraction into cyclohexane solutions of tri-n-octylphosphine oxide, followed by back-extraction, and subsequent coulometric titration at a controlled potential. This procedure has been applied to the determination of uranium in the following types of dissolver solutions:

U—Al, Zr	F, HNO_3
U—Al, Th	F, HNO_3
U—Al, Hg	HNO_3
U—stainless steel	HNO_3, HCl
U—Fe, Cr, Ni	HNO_3, HCl
U—Fe, Mo	HNO_3
U—Mo	HNO_3
U—Nb	HNO_3

The method has proved to be widely applicable, accurate, and precise.

Some studies are in progress in an attempt to determine if NaF can be used successfully as a supporting electrolyte for the controlled-potential coulometric titration of uranium and if conditions can be established whereby U(VI) can be titrated in the presence of Fe(III) in a solution of NaF. The effects of various concentrations of NaF were determined, and optimum coulometric titration potentials were established.

Several analyses were made of standard U(VI) solutions in both 0.5 M and 0.75 M NaF at a pH of 7; the reduction of U(VI) to U(IV) was made at −1.0 v vs the S.C.E. The results are given below:

NaF Conc (M)	Initial Current (ma)	Titration Time (min)	Amount of Uranium (mg)		Bias (%)	S* (%)
			Taken	Found		
0.5	23	25	5.45	5.46	+0.2	0.4
0.75	26	22	5.45	5.45	0.0	0.2

*S = standard deviation.
Note: Each value is the average of 11 determinations.

THORIUM UTILIZATION PROGRAM

Fuel Cycle Development: Chemistry of Solid Fuel Element Materials. — In a continuing study of the determination of the chemical composition and crystal structure of ThO_2-UO_{2+x} equilibrium phases (ORNL-3223, p 4; ORNL-3242, p 4; ORNL-3269, p 6; ORNL-3286, p 5; and ORNL-3321, p 6), 26 thoria-urania solid-solution specimens containing 10 to 80 mole % ThO_2 were equilibrated in air at constant temperatures between 331 and 543°C for three months, after which they were rapidly cooled and examined for identification of the number and identity of phases present. Results of metallographic examination indicated that in the temperature range 421 to 508°C, the phase boundary limiting the fluorite cubic phase occurs at ~40 mole % ThO_2. Compositions containing 50 to 80 mole % ThO_2 consist of a single phase; those containing 10 to 30 mole % ThO_2 consist of two solid phases.

Efforts to determine the oxygen dissociation pressure of ThO_2-UO_{2+x} solids in the temperature range 800 to 1400°C have disclosed several experimental difficulties. Observations indicate that the oxygen partial pressures are near or beyond the lower limit of detection. Mass analysis of gas samples indicates a slow but measurable outgassing in the system over extended periods of time. The contaminant gases are primarily CO_2 and CO. It has not been established whether the source of the impurity gases is in the ThO_2-UO_2 sample or other parts of the vacuum system or both. The quartz tube used in the hot region of the system has recently been replaced with a double-walled mullite tube, and a new set of experiments has begun.

Fuel Cycle Development: Fuel Preparation. — Pure ThO_2 and 3 at. % UO_2—ThO_2 particles, 100 to 200 μ in diameter, were prepared by dispersion

of the oxide aquasols in carbon tetrachloride, then "setting" the particles with isopropyl alcohol and drying to gel. The particles retained their rounded-gel shapes when fired to 1150°C in air.

Steam denitration of uranyl nitrate was investigated as a method for the preparation of active UO_3 hydrate. A carrier gas (air, bubbled through water at controlled temperatures) was passed over the uranyl nitrate in a rotary calciner, giving various hydrated UO_3 products at temperatures from 175 to 300°C in about 5 hr. Products had crystallite sizes of 166 to 850 A. The most active oxide was the β-$UO_3 \cdot H_2O$ prepared at 175°C.

In six successive batches (a total of about 35 kg) of 3 mole % UO_2–ThO_2 prepared by the Kilorod sol-gel procedure, the uranium was distributed within the specified 3 ± 0.03 mole % for all particle-size fractions. The addition of 4-1/2% U as UO_3 or $(NH_4)_2U_2O_7$ to a mixed urania-thoria sol gave a 7-1/2% UO_2–ThO_2 sol-gel product without excessive uranium segregation. Calcination in Ar–4% H_2 gave products having O/U ratios of 2.019 to 2.026, and in pure nitrogen or argon, 2.099 to 2.31. Residual gases released at 1200°C in vacuum were 0.004 to 0.018 cc/g for the coarse fractions and 0.067 to 0.130 cc/g for the fines after calcination. Grinding and sizing increased the amount of evolved gases to 0.012 to 0.021 cc/g for the coarse and 0.170 to 0.320 cc/g for the fine fractions.

A product prepared with 6% enriched uranium for irradiation specimens was analyzed for 24 trace elements after crushing and sizing. The macroscopic cross section of total impurities was equivalent to 5 ppm of boron, with iron, sodium, and nickel as the principal impurities. Two 22.5-in.-long irradiation capsules were sent to the MTR for irradiation of the 5 wt % enriched uranium material (sol-gel UO_2-ThO_2) to 120,000 Mwd per metric ton of thorium.

MOLTEN-SALT REACTOR PROGRAM

Molten-Salt Reactor Experiment. – MSRE design work was completed except for some instrumentation and controls and heating-system design.

Fabrication of major components continued in the Y-12 shops. The steam domes for the drain-tank cooling system were completed. Most of the tube-to-tube-sheet welds on the heat exchanger were completed.

Construction work at Building 7503 was essentially completed. A few contractor employees continued to do minor cleanup.

The estimated cost for the design, procurement, construction, and installation of the MSRE was revised. The new estimate is $8,585,000 plus a contingency of $779,000, which totals $9,364,000.

Reactor Analysis. – The nuclear characteristics of the MSRE with fuel containing highly enriched uranium and no thorium were explored further. Density coefficients of reactivity were computed for use in estimating the effects of uncertainties in fuel and graphite densities upon reactivity. Calculated values for the ratio of reactivity change to fractional density change were 0.53 for the graphite and 0.34 for the fuel. The reactivity coefficient for Xe^{135} was also obtained; with xenon distributed uniformly in the core, the reactivity associated with Xe^{135} was 0.64 of the xenon fraction poison (neutrons absorbed in xenon relative to those absorbed in U^{235}).

A fuel fill accident involving the addition of highly concentrated fuel was analyzed to determine the effectiveness of corrective action in limiting core temperatures. Inserting the control rods after the power reached 15 Mw limited the mean fuel temperature rise to about 10°F during the initial excursion. To prevent a second excursion, the fill must be stopped after the rods are inserted. The exigency of the situation can be relieved by limiting the fill rate, without incurring core fill times greater than 3 to 4 hr.

Preliminary physics computations were made for fuels containing up to 1 mole % uranium of appropriate enrichment. Resonance integrals calculated for U^{238} ranged from the infinite-dilution value of 280 barns for highly enriched fuel to 150 barns for fuel containing 1 mole % $U^{238}F_4$.

Development of Components and Systems. — The engineering test loop was drained of salt after 1500 hr of operation at 1200°F with graphite in the 8-in.-diam container. Two graphite elements plus miscellaneous graphite samples were removed by means of a dry-box facility for metallographic examination.

The graphite access joint of the above container was operated with a free liquid surface instead of a frozen-salt seal such as was used previously. The higher temperature permitted at the liquid-gas interface reduced the cooling requirement in the joint. A dark-colored magnetic deposit was removed from the joint in the region of the liquid-gas interface. Chemical analyses indicated that corrosion products and cold-trapped zirconium oxide with 22% nickel and 52.5% zirconium were the main constituents.

The salt is being treated with HF in the drain tank for oxide removal to verify oxygen analysis of salt samples taken during operation.

Fabrication of a mockup of the MSRE sampler-enricher is complete, and installation at the engineering test loop is in progress.

Testing of the oxygen removal section of the MSRE helium supply system encountered problems with impurities produced in the helium stream by volatilization of an organic binder from the internal thermal insulation and with failure of an internal heater. A prefired insulating material and a modified heater design were incorporated, and testing was resumed.

Two design concepts of the MSRE control rod, using different types of flexible metal hose as part of the rod, have been cycled through a 52-in. stroke in a heated reactor thimble mockup. The flexible hose is used to supply cooling air to the rod as it moves through two 45° bends in the thimble. High friction at the bends and excessive stretching of the flexible hose were experienced with both types.

Bearings are being incorporated in the thimble bends, and cycling will be continued.

Thermal-shock cycling of the drain-tank cooler test has continued through a total of 1650 cycles in the 200 to 1400°F range. Only five of the nine reactor-grade thermocouples installed in this test for evaluating attachment methods have failed; this represents an improvement in performance over previous attachment methods.

Remote operation of freeze flanges in the MSRE maintenance mockup was successfully demonstrated under simulated reactor conditions. Detailed drawings of the special tooling for FF-100 were completed. This location required special tools because of interferences with piping and structure. The heater-insulation carriage and pipe support structure for a portion of line 200 was installed in the mockup for testing.

MSRE Pump Program. — The Inconel centrifugal pump (PKP type) has accumulated 8186 hr of operation in an endurance test with molten-salt $LiF-BeF_2-ThF_4-UF_4$ (65-30-4-1 mole %) at 1225°F, 1950 rpm, and 510 gpm. The test pump with a molten-salt journal bearing was placed in operation at 1225°F, 1200 rpm, and 100 gpm, circulating molten-salt $LiF-BeF_2-UF_4$ (62-37-1 mole %). After 58 hr of operation the pump seized. The pump rotary element will be removed and inspected for cause of the seizure as soon as practicable.

Operation of the prototype fuel pump continued; it is circulating molten-salt $LiF-BeF_2-ZrF_4-ThF_4-UF_4$ (70-23-5-1-1 mole %) at 1200°F. The total operating time on the current run is 1523 hr. Preliminary back-diffusion tests were made with Kr^{85}, during which it was determined that the instrumentation and the gas-flow system required maintenance.

Fabrication of the lubrication stands (including piping, reservoirs, valves, and support structures) was completed; the instrumentation remains to be installed.

Fabrication of the pump tanks was completed to the point of making the final girth welds. Delivery of two bearing housings by an outside vendor is expected in January 1963.

Fabrication of the drive motors was continued but has not progressed as expected. Defects have been found in the cylindrical sections of the vessels; a new delivery date has not been established.

Instrumentation Development. — Testing and evaluation of sealing and potting compounds for use in sealing electrical and thermocouple lead wires at penetrations of the reactor containment were continued. No satisfactory seals have been obtained with any of the inorganic compounds which set and cure at room temperature. Excellent seals were obtained, however, with several epoxy compounds. The epoxy seals will be satisfactory for use in areas outside the reactor shield. Preliminary test results indicate that satisfactory seals could be obtained with inorganic compounds which cure and set at elevated temperatures. A glass-to-metal solder seal appears promising as an alternative to the potted seal. Results of tests to date indicate that the cost and effort required to fabricate the high-temperature-setting potted seals, under field conditions, would be significantly greater than that required to make the glass-to-metal solder seals.

Further tests of a heat-conducting ceramic cement (Thermon X-63) revealed that it is mildly corrosive to INOR-8 at 1200°F. This material will not, therefore, be used to bond thermocouples to the MSRE radiator tubes as previously reported.

Testing of thermocouples of the type specified for use in the MSRE is continuing. Six thermocouples being furnace-tested at 1200°F continue to exhibit stability of calibration after 7500 hr of operation. Eight of ten thermocouples were still functioning after nine weeks of cycling between 300 and 1400°F.

A developmental ball-float-type molten-salt level transmitter for use on the MSRE continued to operate satisfactorily after 11 months at 1200°F. Preparations for further tests of this transmitter under simulated reactor conditions on the molten-salt pump test facility are nearing completion.

Testing of a developmental temperature-scanning system was temporarily discontinued when the electrical noise generated in the switch became excessive. The noise level increased suddenly after changing the nitrogen bottle supplying sweep gas to the mercury switch. Examination of the switch revealed a black precipitate in the mercury sump. An effort is being made to determine the composition of the precipitate and the cause of the switch failure. The switch had operated more than 2500 hr when the failure occurred.

An optical device for use in obtaining three-dimensional "stereo" television pictures with a single-channel closed-circuit television system was purchased from the Stereo-Tronic Company.

NUCLEAR SAFETY

Release of Fission Products on Out-of-Pile Melting of Reactor Fuels. — Two new apparatus for melting irradiated UO_2 have been devised and tested. A tungsten resistor which can be surrounded by cored UO_2 pellets permits melting of about 40-g samples; a tungsten crucible with rf heating will also accommodate large samples (30 g) and permits, in addition, studies of fission product release as a function of time in the molten state. Melts with the tungsten resistor have shown (1) fractional releases of iodine, tellurium, and cesium about 25% below previous values where 40-mg samples of UO_2 were melted; (2) release of fractions of a percent of ruthenium, strontium, and barium when melting occurred under pure helium; and (3) markedly smaller release of tellurium from Zircaloy-clad UO_2 than from stainless-steel-clad material. Melts in the tungsten crucible give generally similar data and (see the table below) apparently show an appreciable effect of time in the molten state on fractional release of rare gases, iodine, tellurium, and cesium. Of these materials, however, only cesium is still being released in appreciable quantities after 2.5 min.

Fission Product Release from UO_2 Melted in Helium by the Tungsten-Crucible Method

Samples: 29 g of tracer-irradiated UO_2 (PWR type)
Atmosphere: purified helium flowing at a rate of 700 cc/min

Run No.	Time (min)		UO_2 Vaporized (%)	Gross Gamma Release (%)	Percent Release							
	Preheat	Molten			Rare Gas	I	Te	Cs	Ru	Sr	Ba	Ce
10-24-62	4.75	1.0	0.097	22.8	92.9	76.9	89.9	62.5	0.45	0.33	4.8	0.05
10-31-62	5.0	1.5	0.157	15.1[a]	98.0	98.3	97.6	66.0	0.05	0.47	2.57	0.07
11-7-62	4.75	2.0	0.156	21.2	99.3	98.8	99.1	59.6	0.32	0.41	2.95	0.17
11-12-62	4.5	2.5	0.246	12.6[a]	99.0	95.0	98.9	72.0	0.33	0.53	2.4	0.13
11-15-62	4.5	1.5		13.7[a]	98.7							
11-19-62	4.5	2.5		12.6[a]	99.3							

[a]Decayed 4 to 7 days longer than the fuel in the previous experiment.

Release of Fission Products on In-Pile Melting of Reactor Fuels. — The ninth miniature UO_2 fuel element was melted in the ORR under conditions similar to those employed for the fourth and fifth specimens. Preliminary examination in the hot cells revealed that the fuel melted completely and that the appearance of the residue is similar to those from the fourth and fifth specimens.

Radiochemical analyses were completed for specimen No. 8. Release data from this specimen, 79% of which was melted, are as follows:

Material Released by the Eighth In-Pile UO_2 Melting Experiment (%)

	Sr^{89}	Zr^{95}	Ru^{106}	I^{131}	Te^{132}	Cs^{137}	Ba^{140}	Ce^{144}	UO_2
Release from fuel	40.0	58.4	58.5	98.8	97.7	98.0	70.0	44.5	
Release from high-temperature zone of the furnace	1.4	0.05	1.7	95.3	81.7	87.6	0.82	0.24	0.02

An experimental burning of a miniature fuel element composed of a graphite matrix containing spheroidal uranium carbide particles coated with pyrolytic carbon was performed; this experiment, funded by the Gas-Cooled Reactor Program, was described previously (ORNL-3351, p 4; ORNL-3368, p 4).

Fission Product Transport Evaluations. — Iodine labeled with I^{131} has been used to test sample methods for determining heats of vaporization in a search for a technique to identify the chemical form of released fission product species. One method, which depended on the rate of transfer of iodine by helium flowing at a constant rate, gave values for the heat of vaporization of iodine ranging from 13 to 15 kcal/mole. The average value, 14.2 kcal, agrees reasonably well with the literature value of 14.9 for solid iodine. A completely sealed glass system, consisting of two small bulbs connected by a long capillary tube, comprised the sample container employed in the second method. The change in concentration of iodine vapor in one bulb, which was in equilibrium with solid or liquid iodine in the other bulb, was measured over a range of temperatures by means of a crystal detector. Early results obtained by this method varied over a wider range than those obtained by the transport method, but recent data indicate that this may be a useful technique, and experiments are in progress to obtain a better understanding of iodine behavior in this apparatus. Evidences of absorption of iodine on glass or quartz walls were observed in both types of experiments.

Melting Experiments in TREAT Facility. — Release of fission products when UO_2 is melted under reactor transient conditions is to be studied in the TREAT facility at the NRTS; a preliminary proposal has been reviewed

with TREAT management to establish a mutually satisfactory design for the experimental assembly. In the experiments, fully clad 10%-enriched UO_2 should be melted in an argon-filled autoclave subjected to fast transients of varying magnitude. The samples will be preheated to 1200°C; the released gases and smoke will be sampled promptly by admission through a filter into an evacuated autoclave. Heat transfer determinations and leakage testing of the autoclaves have been satisfactorily completed, and a hazards analysis is being prepared. The first experimental assembly should be inserted in the reactor during early 1963.

Characterization and Control of Accident-Released Fission Products. — Removal of extremely fine radioactive particles from air streams by columns of foam was examined; previously described diffusional deposition techniques were used to determine the efficiency with which the particles are removed. In preliminary experiments, particles of Al_2O_3 carrying radioactive iodine were dispersed in air and passed at 5 fpm through aqueous detergent solutions which produced the desired height of foam. A 5-in. height of pure water used as a control showed negligible removal of 0.0065-μ particles, while a 12-in. column of foam removed about 85% of such particles. Surprisingly, the initial tests show the same removal efficiency for a 42-in. column of the foam.

The distribution vs depth for radioactive aerosols in fibrous filters has been examined under carefully controlled conditions. Initial tests have used filters of very uniform 11-μ-diam Dacron fibers and aerosols containing particles (40 to 300 A in diameter) of $Zn^{65}O$ flowing at 0.7 to 9.8 fpm (linear). The radioactive deposit in the filter was decreased by factors of 10^3 to 10^4 over a filter depth which provided 10^5 cm of fiber per square centimeter of filter area. At the lowest linear velocity, where diffusion would be controlling, collection efficiency was increased by decreased velocity, as expected. At the higher flow rates, where the primary mechanism of filtration is by interception for particles of this size, the efficiency appears to be independent of velocity, in accord with theory. This method will characterize aerosols in terms of their interaction with fibers and perhaps extend the theory of filter performance to conditions for which little information is now available.

Nuclear Safety Pilot Plant. — The cell in which equipment for the Nuclear Safety Pilot Plant is to be installed is being used for a test of the foam suppression of activity in air in cooperation with personnel from the Harvard Air Cleaning Laboratory. To date, one of a series of six planned tests has been completed. In this test 10 curies of elemental I^{131} was vaporized into the 6000-ft^3 cell volume, and the concentration of the activity in the air was measured as a function of time after release. Tests with and without the introduction of foam demonstrated the efficiency of the foam for suppression of activity.

Reactor Containment Handbook. — Chapter II, "Codes, Criteria, and Regulations," and Chapter IV, "Energy Sources," are being edited for reproduction. Chapter IV is scheduled for distribution in mid-January and Chapter II by February 1, 1963.

Literature surveys are being completed for most of the chapters under subcontract to Bechtel Corporation. These include Chapter VII, "Containment Proof Test"; Chapter IX, "Material Properties"; Chapter X, "Design Details"; and Chapter XI, "Economics." Completion in draft form is scheduled for March 1, 1963.

WASTE DISPOSAL DEVELOPMENT

Clinch River Study. — Assuming that present methods of releasing radioactive materials from White Oak Lake are continued, a reservoir of radioactivity will be formed in the Clinch River during periods of no release from Melton Hill Reservoir. During periods of release from the reservoir, the radioactivity will be swept downstream in a cloud. The dispersion effects of the power wave caused by the release from Melton Hill Reservoir on this cloud were found in preliminary evaluations of flume tests to be similar to those caused by steady-flow conditions.

Gross gamma and gamma spectrum analyses of Clinch River bottom sediment cores have been initiated. The device being used is a specially designed "core scanner" which consists of a 3 × 3 in. NaI scintillation crystal and matched phototube enclosed in a 4-in.-thick lead shield with a 2 × 2 × 4 in. collimator slit, and a hoist which automatically moves the core vertically past the collimator in 2-in. increments.

A procedure for identifying and adjusting unreasonable radiochemical analytical results was developed for use in computing radionuclide loads in Clinch River water. The method utilizes a comparison of analyses of water collected at four Clinch River Study sampling stations. Radionuclide mass balance computations, which take into account the adjusted analytical values, show that transfer of radionuclides from the water phase to the river-bottom sediments involves only a small percentage of the total radionuclide load transported by the river between the upstream and downstream sampling points.

Mineral Exchange Studies. — Strontium removals by four different sorbents were compared by use of column techniques. Tests were performed with a 0.5 M $NaNO_3$ solution adjusted to pH 10 and containing 10^{-5} M $Sr(NO_3)_2$; the flow rate was 3 ml min^{-1} cm^{-2}. Commercial-grade activated alumina was most efficient; the 50% breakthrough volume for 10 g of material was approximately 5500 cc. The same weight of Dowex 50-X12 treated 2600 cc for the same breakthrough percentage. Both vermiculite and clinoptilolite were ineffective under these test conditions; in both cases breakthrough occurred when the throughput volume was less than 150 cc. Intermittent checks showed that the effluent pH from the Dowex column had dropped to 9.7, whereas the pH from the alumina column showed readings generally between 8 and 9, with some readings as low as 7.5.

High-Activity Waste Calcination: Corrosion. — Specimens of types 304L and 347 stainless steels corroded at rates of less than 0.1 mil/month in the superheated vapor above refluxing simulated Purex waste solution, compared with rates of 2.6 and 3.3 mils/month, respectively, for specimens in the condensing vapor. Type 304L stainless steel underwent elongation of between 9.2×10^{-5} and 5.4×10^{-5} in./in. while subjected to a Purex evaporation-fixation cycle followed by about 165 hr at 1050°C. Welded specimens of various stainless steels exposed for 31 days at about 575°C in the Carey salt mine, Hutchinson, Kansas, corroded at rates of 0.45 to 1.3 mils/month.

High-Activity Waste Calcination: Fixation of Waste in Glass. — Melts containing 39 to 43% waste oxides from simulated high-sulfate Purex waste retained about 95% of the original sulfate when heated to 100°C above the softening points for 100 min. Ruthenium volatilized to the extent of

about 24% from semiengineering-scale fixation-in-glass experiments with simulated TBP-25 wastes. Also, about 77% of the mercury volatilized.

Disposal in Natural Salt Formations. — In the tests at the Carey Salt Company mine at Hutchinson, Kansas, both wall and floor heater holes showed an apparent decrease in diameter of about 1 in. at a depth of 6 ft. Maximum expansion of the wall and of the floor occurred at 2 ft from the heater holes and was 0.5 and 0.8 in. for the wall and floor respectively. Unrestrained thermal expansion would have resulted in an expansion of 1.14 in. There was essentially no recovery of the expansion on cooling.

Engineering, Economic, and Safety Evaluation. — Man may be exposed to radiation through a number of avenues as a result of the discharge of radioactive fluids to the Clinch River. Calculations of dose received or dose rate by the various avenues are in progress. Of the organs exposed to radiation by ingestion of contaminated water, the estimated exposure to the bone was largest, attaining 0.51 of the MPC_W limit in 1954 at Clinch River mile 14.5. By immersion in contaminated water, a maximum dose rate of 0.043 millirad per day of exposure (0.032 of beta and 0.01 of gamma) is calculated at mile 14.5 during 1960. Contaminated bottom sediments can also contribute to the total dose received by man. Based on sediment analyses and the assumption of a uniform, infinite source, the largest bottom sediment dose rate (at contact) of 16 millirads per day of exposure would have occurred in 1959 on the Clinch River; the dose rate is about equally divided between beta and gamma radiation. The beta dose rate is eliminated by about 1 cm of water shielding; the gamma dose rate is reduced by an average factor of 18 after attenuation through 3 ft of water.

Foam Separation Studies. — The effects of column diameter, sludge height, and flow rates on the precipitation of calcium and magnesium from ORNL low-activity waste were studied. Column diameters varied from 2 to 9 in., and flow rates from 7.4 to 60 gal ft^{-2} hr^{-1}. Within this range the hardness of the waste water was reduced from about 100 ppm to 2–4 ppm (as $CaCO_3$) when precipitation was induced by making the water 0.005 M each in NaOH and Na_2CO_3. An apparatus was tested for the removal by foaming of the surface-active agent sodium dodecylbenzene sulfonate; 90% was separated from a solution containing the above-listed chemicals plus 50 ppm of the surfactant.

Engineering studies were continued in 6-in.-ID columns, with dodecylbenzene sulfonate (DBS) as the surfactant. One liquid-feed distributor gave HTU_x values of less than 2 cm for flows of 120 gal ft^{-2} hr^{-1} or less; the HTU_x values were 2 to 20 cm for other distributors or higher flow rates. The difference in HTU_x values for the spinnerette gas spargers with 50- and 80-μ-diam holes was small, variable, and probably not significant. Recovery of DBS by simple foaming could be applied to give effluents of about 5 ppm of surfactant for 0.02 N Na^+ and about 20 ppm for 0.001 N Na^+. Process water was used to simulate low-activity waste. The calcium and magnesium were precipitated, and the slurries were used as feed for the foam separation column. Decontamination factors of 81 to 162 for strontium obtained with $CaCO_3$-$Mg(OH)_2$ slurry feed were reduced to less than 3 when 10 ppm of Fab household detergent was added to simulate the effect of such detergents in low-activity wastes.

PHYSICAL RESEARCH PROGRAM

PHYSICS AND MATHEMATICS RESEARCH

High-Energy Physics. — A precision measurement of the Λ° lifetime was recently completed in conjunction with Northwestern University, Syracuse University, and the University of Bologna. The K^- mesons stopped in the Northwestern helium bubble chamber yielded 2239 Λ°'s with a mean momentum of ~250 Mev/c and satisfied rigorous selection criteria designed to minimize biases. The mean lifetime was found to be $\tau = (2.36 \pm 0.06) \times 10^{-10}$ sec. Analysis of the interaction $\Sigma^- + He^4 \rightarrow \Lambda^\circ + n + H^3$ is continuing, approximately 40 rolls of film having been measured. A 7090 computer program for the automatic analysis of these interactions was completed. The program incorporates the existing HeGuts program into a one-pass system which kinematically fits the decay of the hyperon and uses this fit to make all possible three-body fits.

High-Energy Electronuclear Machines: Cyclotron Analogue II. — A large fraction of the beam (to date, 75%) has been extracted in the Analogue, an electron cyclotron which duplicates the ion-orbit behavior anticipated for 810-Mev protons in the Mc^2 Cyclotron. Extraction of the beam is difficult because of the close spacing of the orbits. Extraction occurs at a radius of 13.4 in., where the spacing between successive turns is normally only 0.003 in. This orbit spacing is greatly increased by exploiting a natural instability in the radial motion of the electrons which occurs at extraction radius. The addition of a small (0.5%) doubly periodic bump in the magnetic field causes the spacing between turns to increase rapidly in successive revolutions. Most of the beam can then enter an extraction channel. In the Analogue the channel consists of two thin electrodes with a potential of 2000 v between them to create an electric field which deflects the electrons away from the circulating beam. Adjustments are provided so that the channel can be shaped to fit the beam path.

In the Mc^2 Cyclotron a magnetic channel operating in an analogous manner will replace the electrostatic system used in the Analogue. The results with the Analogue are of considerable importance. They predict that a high beam current (>100 μa) can be directed to the experiment rooms and that the inner parts of the cyclotron will not be unreasonably activated by wasted high-energy beam.

High-Energy Electronuclear Machines: Proposed Mc^2 Facility. — A conference on "Meson and Nuclear Research Below 1 Bev," sponsored by the ORNL-ORINS Nuclear Facilities Group, was held in Gatlinburg on November 12 and 13. About 135 participants from 40 institutions in the United States and abroad took part. Papers were presented on pion physics, weak interactions, nuclear structure, space research, biology, and medicine. It was abundantly clear from the conference that the region below 1 Bev is a fertile area of research open to high-intensity accelerators such as the Mc^2 machine. In almost every paper the speaker deplored the lack of intensity available for the experiments. The feeling was that a genuine frontier exists at high intensity, as it does at high energy.

Basic Studies of Cryogenic Electromagnets. — Three inner sections for the nine-section, 2-in.-ID, 30- to 50-kilogauss superconducting magnet were built and tested. Each section has the following dimensions: a 2.3-in. ID, a 3.75-in. OD, and a 1.5-in. length. Maximum currents of 34 to 38 amp were achieved, with each section tested separately; this produced up to 14 kilogauss at the center.

Tests in the High-Field Solenoid (100 kilogauss) with longitudinal Nb-Zr samples show that critical-current ratios at 30 kilogauss are roughly 1:2 for samples 4 in. and 2 in. long; that is, when the pressed-copper joints are in the high-field region, the current capacity is higher.

Multiparticle Medium-Energy Nuclear Physics: The 86-Inch Cyclotron. — In the interpretation of the elastic scattering of protons from Ni^{64} and Zn^{64} in terms of an $(N - Z)/A$ dependence of the optical-model potential, an ambiguity arises; the difference in deformation in the two nuclei might affect the elastic scattering as much as the $(N - Z)/A$ term in the potential, obscuring the interpretation. To clarify this point the isobar pair Fe^{58}-Ni^{58} was studied. The experiment was performed on the variable-energy cyclotron at the Lawrence Radiation Laboratory, Livermore, California. Iron-58 has the larger deformation and the larger $(N - Z)/A$ of the $A = 58$ pair, while Ni^{64} has the smaller deformation and the larger $(N - Z)/A$ of the $A = 64$ pair. The angular shift in the experimentally measured scattering pattern is toward smaller angles for the larger $(N - Z)/A$ in both cases. This is consistent with a dependence of the real potential well depth on $(N - Z)/A$.

Differential cross sections for (p,d), (p,t), and (p,α) pickup reactions for selected cases leading to the same final states in Zr^{90} and in Fe^{56} were measured and compared. The cases were chosen so that the spectroscopic factors for each reaction should be near unity. Thus the data are characteristic of the reaction and are uncomplicated by nuclear structure effects.

The state in Zr^{89} at 0.6 Mev excited in the reaction $Zr^{90}(p,d)Zr^{89}$ has been interpreted as the p 1/2 hole state. This state is closely related to a state at 8 Mev found in the reaction $Y^{89}(p,n)Zr^{89}$ [Phys. Rev. 126, 2170 (1962)]. The interpretation has been made that the 0.6-Mev state is characterized by $T = 9/2$, $T_z = 9/2$, and the 8-Mev state by $T = 11/2$, $T_z = 9/2$.

Multiparticle Medium-Energy Nuclear Physics: Heavy-Particle Physics. — During the shutdown of the tandem Van de Graaff, the 63-Inch Cyclotron is being reactivated on a part-time basis for heavy-ion experiments. The ratio of Sc^{44m}/Sc^{44g} from the nitrogen bombardment of phosphorus and sulfur was measured from 20 to 27 Mev. The importance of angular momentum in the compound nucleus is clearly brought out. An extensive computer analysis of angular distributions of alpha particles emitted in heavy-ion reactions was completed. Data were obtained on the spin cutoff parameter, and the moment of inertia of some light nuclei was calculated to be about 0.7 of the rigid moment. Proton transfer reactions of nitrogen ions on various light elements leading to 2-min O^{15} are being investigated.

Physics of Fission. – The response of silicon surface-barrier and diffused-junction detectors to Br^{79}, Br^{81}, and I^{127} ions was studied. The ions were accelerated in the tandem Van de Graaff to energies in the range 15 to 110 Mev; the ions were then magnetically analyzed and allowed to impinge on the detectors. Pulse-height responses as functions of ion energy, incident angle, detector voltage, and detector resistivity were obtained for both types of detectors. Data were obtained for detector voltages below the saturated pulse-height condition, through the plateau region, and into the "multiplication" region. It is found, for the usual operating conditions, that a relationship of the form $E = a \times PH + \delta$, where a and δ are constants, describes the result over most of the ion energy range. The energy resolution for these detectors was found to be approximately 1 Mev. Differences are observed between the PH-vs-E relationships for bromide and iodide ions.

Scintillation Spectrometry and Instrument Development. – The nuclear Zeeman splitting utilizing the Mössbauer effect for Fe^{57} in a rhodium host lattice was measured. Near 50 at. % Fe–Rh the alloy may exist as an ordered system with a CsCl crystal structure. The alloy exhibits a ferromagnetic-antiferromagnetic transition near room temperature, but the exact transition temperature region depends critically on the excess rhodium in the alloy. The temperature dependence of the hyperfine splitting has been measured in both the ferro- and antiferromagnetic regions. The high-temperature ferromagnetic region shows a decrease in the internal magnetic field with increasing temperature and follows the usual Weiss curve with a Curie temperature at 670°K. Below the transition temperature the data points follow another curve indicating a slightly lower internal magnetic field on the low-temperature or antiferromagnetic side of the transition. At room temperature and in the ferromagnetic state, an internal magnetic field of 267 kilogauss is obtained. This is somewhat smaller than one might expect if the atomic moment on the iron atom were 3.0 Bohr magnetons.

High-Voltage Experimental Program. – Neutron capture gamma-ray spectra are being studied as a function of neutron energy in a manner previously described. Resonances are observed in fluorine at 27 and 49 kev. These are known to be p-wave in character, and, indeed, the spectra show marked differences from the spectrum at thermal energy. Specifically, the supposedly 2^+ state of F^{20} should strongly depress capture to ground in the case of thermal capture and permit the p-state capture to ground via electric dipole radiation. Just the opposite experimental situation seems to prevail. The resonant capture spectrum observed in calcium contains substantially the same lines as does thermal capture, but the relative line intensities are completely different. The capture is attributed to small resonances at about 20 and 50 kev not previously observed in transmission experiments. The rather frequent appearance of relative transition intensities which seemingly contradict assignments of spins and parities obtained from other types of experiments makes it of interest to study as many cases as possible, especially those in which the character of the capturing state is known.

Electronuclear Machines: The Isochronous Cyclotron (ORIC). — Installation of the full-power rf system is essentially complete; the system is being checked in preparation for test operation of the cyclotron at full power. During December the supply for the power amplifier (6949) plate was checked out, and all interlocks and radiation protection systems were tested. Meters and probes were calibrated, and servos and controls were checked. The rf system was first tuned up on December 14; power was achieved on the dee without vacuum on December 18. The system was operated under vacuum for the first time on December 20; 50 kv was achieved on the dee without any difficulty. Further power tests will be performed after an examination of the entire system to check for possible sources of trouble at high power levels.

From magnetic-field measurements made earlier in the year, an extensive catalog of calculated operating conditions was prepared for the guidance of cyclotron operators; this includes a tabulation of the various coil currents required for the acceleration of protons, alpha particles, and nitrogen ions to a wide range of energies.

Stable Isotopes Development. — A definite relationship was established between oscilloscope patterns of beams being collected and source conditions which are needed to produce improvement in beam quality. More precise selection and better control of operating parameters (arc voltage, current, and pressure) should improve isotopic purity of separated samples. The technique of oscilloscope monitoring is applicable to all separations and is now in use.

Special Separations. — Eleven plutonium runs were completed, and, based on monitored ion currents, it is estimated that the following quantities of plutonium isotopes were collected:

Pu^{242}	1.4351 g
Pu^{241}	2.9851 g
Pu^{240}	11.6112 g
Pu^{239}	6.9387 g

Twenty-one receiver pockets have been transferred to the plutonium laboratory; ~11.61 g of Pu^{240} from five Pu^{240} pockets and ~2.985 g of Pu^{241} from six Pu^{241} pockets were recovered. Isotopic analysis shows 93.65% Pu^{241} and 98.48% Pu^{240}.

A total of 116 g of purified plutonium was recovered from wash solutions used to clean one liner and two sources, and an additional 148 g of plutonium was recovered from original feed solutions. Insoluble materials have not yet been recovered.

The following sources were prepared:

12 U^{232} sources electroplated on platinum

30 nickel tubes electroplated with 3 mg of U^{235}

5 nickel assemblies electroplated with ~30 mg of Np^{237}

1 nickel cylinder plated with 40 mg of normal uranium

110 Am^{241} sources

CHEMISTRY RESEARCH

Thorium Oxide Studies. — Examination of the thorium oxide preparations irradiated in the multiple-sample, 12-month LITR irradiation experiment is continuing (ORNL-3377, p 25). Code P-82 thoria pellets, when irradiated dry, lost less than 0.05% of their weight; those irradiated under D_2O lost less than 0.4%; and those irradiated in the D_2O vapor lost 6.2%.

Wear rates in spouting-bed tests of the dry- and D_2O-irradiated pellets were only about two-thirds those of the original or D_2O-control pellets throughout 6-hr tests. Pictures of the irradiated pellets after the wear tests showed that, with the exception of an occasional pit, the wear rate was uniform and indicated little radiation-induced structural damage.

Impact fracture tests were carried out on the cold and the irradiated P-82 pellets. The original pellets, those from the 12-month D_2O-control experiment, and those from the 3- and 12-month irradiations (dry and in D_2O) showed impact fracture energies between 0.651 and 0.945 in.-lb. The pellets that had been irradiated for 12 months in D_2O vapor had a fracture energy of 0.342 in.-lb. There was further evidence of the structural weakening that resulted from the vapor-phase irradiation: Unlike the response of pellets irradiated under D_2O, stirring of the vapor-phase-irradiated pellets in water resulted in some breakage.

The interiors of the 12-month liquid-phase-irradiated pellets were similar in color to those of the unirradiated pellets, while those of the dry-irradiated pellets were black. Heating at 400°C for 1 hr in air failed to change the color, but heating at 900°C overnight restored the original color. No weight increase was noted as a result of the 900°C heating, and it is postulated that the blackening resulted from a very slight oxygen deficiency.

Definitive pore-size-distribution data (according to the nitrogen adsorption method) for pellets that had been irradiated for three months in D_2O and for some irradiated for an extended time in 260°C water indicated that irradiation in D_2O decreased the nitrogen-measured void volume from a value of 0.6% to 0.4%. It produced a large number of small pores (of about 30 A radius), probably associated with individual fission events, and removed larger pores of about 100 to 400 A radius. Since both the irradiated and unirradiated pellets had densities less than 97% of theoretical, most of the surface-connected void volume must be associated with pores of greater than 1100 A radius.

Equipment Decontamination. — The steam-jet decontamination of stainless and carbon steels that had been baked in helium at 500°C, simulating severe exposures in gas-cooled reactors, was very satisfactory when oxalate-peroxide reagents were aspirated into the jet at a proportion of about 16% of the steam weight. In 30 min, carbon-steel decontamination factors for strips that were 2 in. from the jet were 1.2×10^4 for Ba-La^{140}, 5.4×10^3 for Ru-Rh^{106}, and 635 for Zr-Nb^{95}. At 24 in. from the jet the factors were 39, 285, and 180 respectively.

The corrosion rates on carbon steel were only 0.006 and 0.002 mil/hr in the two positions so long as sufficient hydrogen peroxide was used to

maintain metal passivity (0.5 M or over in the feed), but the rates increased sharply at lower peroxide concentrations. The decontamination of stainless steel was less effective than that of carbon steel, but after 2 hr in the steam jet with several oxalate-peroxide reagents of low corrosivity, the Zr-Nb^{95} decontamination factor averaged 90 on eight stainless steel coupons spaced 2 to 24 in. from the jet nozzle.

One type of cold-rolled carbon steel was brilliantly polished and decontaminated by a factor of 170 from 500°C-baked Ce-Pr^{144} by a 5-min immersion in 0.5 M oxalate–0.01 M F^-–0.53 M H_2O_2 at pH 3.0, at 60 to 70°C, with a removal of 1.5 mils of surface. The steel had previously been treated with other decontaminating solutions. The polished surface was rust-resistant for several days in water at room temperature. The solution did not polish case-hardened steel, however. Sulfate solutions showed little promise as decontamination solutions for carbon steel because of their high corrosivity.

Spectrophotometric Studies of Solutions at Elevated Temperatures and Pressures. – A method was devised for measuring the densities of aqueous solutions at accurately measured temperatures and pressures up to the solution critical point (about 372°C).

A prototype high-pressure autoclave was designed and was built entirely of pure titanium. It can be operated up to 4000 psi at 400°C, and it consists of a bulb section of about 4 ml containing an internal thermocouple well, a uniform expansion section about 12 in. long, and a high-pressure closure head with a fitting that contains a titanium capillary that leads to a pressure transducer. The densities may be determined at 370 to 371°C, within a maximum error of ±0.6%. (At somewhat lower temperatures the error will be less.)

A special furnace and the necessary control instrumentation were designed and constructed to contain the autoclave in an essentially isothermal state over its entire length of about 18 in., up to temperatures to at least 375°C. The furnace is also slotted so that unobstructed x-ray photographs can easily be obtained. A motor-driven x-ray film cassette and support system has also been designed and constructed. The x-ray film can be driven past the slit in the system behind the autoclave from a rate of about 0.05 in./min up to about 3 in./min. The equipment is being installed and will be used with a 300-kv Norelco x-ray system.

Protactinium Chemistry. – Further studies of protactinium distribution between sulfuric acid solutions and Dowex 1 have verified that apparent distribution coefficients are dependent on the amount of protactinium present. As aqueous protactinium concentrations were increased above a critical value, nearly all the added protactinium went into the Dowex 1 resin phase. However, as the loading of protactinium in the resin increased, the concentration in the aqueous phase in equilibrium also increased, but relatively very much more slowly. Stated differently, above a critical protactinium concentration the apparent distribution coefficient increases very rapidly as more protactinium is added. The critical concentration in the aqueous phase increases with the square of the sulfuric acid concentration, from 2.3×10^4 alpha counts min^{-1} ml^{-1} in 1.5 N sulfuric acid to 1.1×10^6 alpha counts min^{-1} ml^{-1} in 10 N acid.

Surface Chemistry. — Continued calorimetric studies of the heat of immersion of ThO_2 in water have yielded unexpected results; in most cases not all of the heat was released immediately on immersion of the sample. Original firing temperatures of the four samples investigated were 650, 800, 1000, and 1200°C.

Within the sensitivity of the experimental apparatus, no slow heat evolution was ever observed from the sample fired at 1200°C. Of the three samples which did exhibit a slow heat of immersion, the sample fired at 800°C gave the largest quantity (on a unit area basis), while the sample fired at 1000°C gave the smallest quantity of slow heat. The amount of slow heat obtained from any given sample increased with increasing outgassing temperature. The effect of outgassing temperature was most pronounced in the case of the samples fired at 650 and 800°C. The fraction of the total heat which appeared as slow heat varied from about 2 to 20%, depending on the sample and the outgassing temperature. Actual values of the slow heat of immersion ranged from about 10 to 200 $ergs/cm^2$.

The half-lives for the decay of the slow heat were in the same order as the magnitudes. These were 7.2, 10.7, and 5.3 min for the samples fired at 650, 800, and 1000°C respectively. Within experimental uncertainty the half-life of the slow heat was independent of the outgassing temperature.

Pyrolysis and Radiolysis of Organic Liquids. — The reproducibility of the pyrolysis experiments was determined by heating three 3-g samples of biphenyl simultaneously in a furnace for 12 hr at 425°C. The variation of results for the gaseous products is as follows: ±4.2% for hydrogen, ±10% for methane, ±5.4% for ethane, ±12.5% for propane, and ±14.4% for acetylene. The decrease in precision is directly related to the increase in retention time for the gases, as is often the case in gas chromatographic procedures. By the technique employed, all of the gas from a given pyrolytic sample was used for a single analysis. However, the amount of intermediate boiler formed during pyrolysis was sufficiently great to permit the use of aliquots for the analysis of benzene. These data have a variation of ±4.2%, although the precision for the analytical procedure is known to be better than ±2%.

Instrumental Methods for Continuous Chemical Analysis. — A high-sensitivity, controlled-potential, or constant-current, coulometric titrator has been designed for use in the titration of small quantities of substances. The maximum electrical sensitivity of this instrument is 500 times that of the ORNL model Q-2005 titrator and 10 times that of the ORNL model Q-2005-X50 titrator. The full output of the current-integrating amplifier is attained with 1, 10, 50, or 100 μeq of substance titrated. At the highest sensitivity, this titrator is more stable and requires less frequent adjustment than the earlier models. The maximum available cell current is approximately ±8 ma. The titrator has an improved control potential supply and built-in calibration resistors. Provision is made for recording the current integral during the titration to enable better interpretation of the titration and better correction for background currents at high sensitivity. Automatic cutoff is also provided. The prototype instrument has been evaluated for controlled-

potential titration, and the design and performance are considered satisfactory. In this mode of operation, the counter electrode is forced to such a potential that, by current feedback through the titration cell, the potential of the controlled electrode with respect to a reference electrode is held at the control potential. For the titration of about 4 μeq or more of material, the accuracy and precision are similar to those obtained with the models Q-2005 and Q-2005-X50 titrators. For titrations of 0.01 to 4 μeq of material, the performance of this titrator is better than that of the older models. An instrument of this design is being fabricated for analyzing MSRE samples.

Analysis of Molten Salts by Electrochemical Methods. — A study is in progress on the voltammetry of iron(II) in molten LiF-NaF-KF (FLINAK) at 495°C. Since iron is a corrosion product associated with reactor fuels of the molten-salt type, it is of interest here from the standpoint of applying electrochemical methods to the analysis of molten-salt systems. The apparatus utilized was described previously (ORNL-3377, p 27).

Known amounts of iron(II) were added to the melt by constant-current electrolysis, utilizing an anode of pure iron. Current-voltage curves were recorded on this melt at 495°C, utilizing a controlled-potential polarograph and three platinum electrodes, one of which was a quasi-reference electrode. These curves revealed two waves: one anodic wave with an $E_{1/2} \cong +0.05$ v vs the quasi-reference electrode and a cathodic wave with $E_{1/2} \cong -0.5$ v. The electrode reactions responsible for these waves are believed to be the oxidation of iron(II) to iron(III) and the reduction of iron(II) to the metal respectively. The limiting currents (wave heights) are in the approximate ratio of 1:2, and the latter wave appears to be analytically useful. From initial observations, the limiting current of this wave is proportional to the concentration of iron, approximately 20 to 200 ppm, in the melt. Additional measurements are necessary, however, before a definite conclusion can be drawn with regard to the reproducibility of the limiting current.

Further studies on the voltammetry of iron will include the evaluating of this method as a possible analytical technique, ascertaining the reversibility of the electrode reactions, and determining the temperature coefficients of the limiting current processes.

Spectrophotometry of Solutions at High Temperature. — Thermal stability (to 250°C and 600 psia) and optical transparency of the following systems were studied in the ranges 0.25 to 1.2 μ for H_2O and 0.25 to 1.8 μ for D_2O: $DClO_4$-D_2O-H_2O (0.01 to 1.0 f), H_2SO_4-H_2O (0.1 to 0.4 f), and HNO_3-H_2O (0.07 to 4.0 f). The titanium body and sapphire windows of the spectrophotometer cell react measurably with all acids at comparable rates which increase with acidity and temperature. Only perchloric acid at concentrations above 0.1 f etches the windows progressively until they become virtually unusable after several days at 250°C; solutions 0.01 to 1.0 f, however, appear to be stable with respect to reduction products (i.e., no Cl_2, HOCl, $HClO_2$, or $HClO_3$ was detected). Solutions 0.1 to 0.2 f in H_2SO_4 begin to absorb below 0.35 μ (presumably due to titanium species); if they are heated, the absorption disappears between 40 and 70°C. Solutions 0.4 f in H_2SO_4 remain transparent to 0.25 μ at 25°C but develop an absorption band when held at 250°C. Higher concentrations of H_2SO_4 cause gross attack of the titanium even at lower

temperatures. Solutions of HNO_3 heated to 250°C tend to absorb in the 0.35- to 0.8-μ region, probably due to HNO_2 and NO_2. The equilibrium between NO_2 and HNO_3 becomes significant only above 200°C and for $HNO_3 > 0.1$ f; it is reversible on cooling. Solutions 0.1 to 1.0 f in HNO_3, after runs to 250°C for several days in the absence of an overpressure of O_2, show, on cooling to 25°C, 0.01 to 0.5% HNO_2. Addition of O_2 shifts the equilibrium in favor of HNO_3.

Radiation Chemistry: Radiolysis of Liquid Oxygen and Oxygen-Nitrogen at 77°K. – The radiolysis of oxygen produces ozone and represents a conversion of radiant to chemical energy. However, the study of gaseous oxygen systems is handicapped by the reactivity of ozone and the consequent loss of product. As a liquid system at 77°K, oxygen and oxygen-nitrogen solutions may be irradiated with gamma rays and the ozone yields measured, in situ, by spectrophotometric analysis. The method allows the assay of ozone at ppm concentrations; the ozone has been stored for hours without decomposition. Yields were linear with dose to the highest dose used, 4×10^{18} ev/g.

In liquid oxygen at 77°K, $G(O_3) = 12.5 \pm 0.4$ molecules/100 ev. This yield is a factor of 2 larger than that reported by others [J. Phys. Chem. 65, 915 (1961); Radiation Res. 13, 452 (1960)]. Other analytical methods have required doses 10^3 to 10^4 times as large.

Ozone yields were measured at 77°K from the radiolysis of oxygen-nitrogen solutions whose oxygen concentration ranged from pure O_2 down to 0.005 mole % O_2. Extensive "energy transfer" or enhanced ozone yields were found. The enhancement is best described by the ratio of yields from an O_2-N_2 solution to that from liquid O_2. In the absence of "energy transfer" this ratio would be equal to the electron fraction of oxygen in the solution.

The observed ratio of yields is neither equal nor directly proportional to the electron fraction of oxygen but shows both a considerable and persistent enhancement of ozone yield. For example, a solution containing 1.5% oxygen produces 46% of the ozone obtained by irradiating pure oxygen. Even when the oxygen concentration is only 0.005%, the ozone yield is still almost 20% of that from pure oxygen; in short, considerable ozone formation occurs in the irradiation of what is nominally liquid nitrogen.

The enhancement of ozone yield passes through a maximum when nitrogen has one oxygen molecule in its shell of nearest neighbors. Detailed explanation of the enhancement and its remarkable persistence will require further developments in the theoretical description of energy deposition and localization processes.

Radiation Chemistry: Bromine Oxidizing Fragments Produced in Crystalline Alkali-Metal Bromates by Co^{60} Gamma Rays. – Bromine oxidation states intermediate between bromate and bromide produced by Co^{60} gamma rays in crystalline alkali-metal bromates increased nonlinearly with the radiation dose and appeared to approach a "steady-state" concentration after long periods. Moreover, the rates of formation and final concentrations of these species were significantly higher for irradiations at −78° than at 60°C. The oxidizing fragments formed and trapped in crystals

at −78° showed thermal annealing reactions: for example, on standing at 25° for approximately 5 hr, approximately 25% were removed; at 100, 150, and 200°C the fractions disappearing were 32, 60, and 90% respectively. Some of the fragments reverted to bromate, but the majority decomposed to yield bromide ions.

A quantitative description of the kinetics of the radiolysis of molecular bromate ion was obtained by assuming a series of parallel and consecutive irreversible decompositions in which one, two, or three bromine-oxygen bonds were broken following the primary ionization or excitation of BrO_3^-. It has not yet been established whether the thermal decompositions and back-reactions observed after radiolysis occurred via an electron capture mechanism or by a mechanism involving the formation, diffusion, and reaction of oxygen atoms in the crystal.

Chemical Physics: Neutron Diffraction. — Analysis of three-dimensional single-crystal neutron diffraction data shows that xenon difluoride contains linear symmetric triatomic molecules with the Xe-F distance of 2.00 A. The space group of the crystal is I4/mmm, with a = 4.315 A, c = 6.990 A.

Corrosion Tests for the TRU Facility. — An investigation was completed to determine the effect of oxidant species on the corrosion of welded Hastelloy C by 6 M HCl at 35°C. Tests were run with (1) normal atmospheric oxygen present in solution, (2) oxygen aeration of the test solution, and (3) continuous addition of 0.05 M H_2O_2 at a rate of approximately 3 ml/hr. The best corrosion resistance was obtained with atmospheric oxygen present. The observed penetration rate was 0.7 mil/month after 1000 hr. When oxygen was bubbled into the solution, the rate increased to 5.2 mils/month during a similar period, and continuous addition of 0.05 M H_2O_2 at a rate of 3 ml/hr produced an attack rate of 4.0 mils/month. All specimens were subject to preferential attack in heat-affected zones adjacent to welds, with the less severe attack occurring in the solution containing atmospheric oxygen only.

Three Du Pont elastomers were exposed in the solution and vapor phases of oxygenated and boiling 6 M HCl solution. Viton (fluorocarbon polymer) and Hypalon (chlorosulfonated polyethylene) underwent appreciable swelling and were generally unsatisfactory after 100 hr. Neoprene (polychloroprene) exhibited minor dimensional and weight changes during a similar period. However, additional exposure produced a decided loss of flexibility of specimens in both solution and vapor phases.

Target Preparation. — Radioisotope targets ranging in thickness from 10 to 150 $\mu g/cm^2$ are being fabricated, with thin films of normal carbon (25 to 50 $\mu g/cm^2$) as the substrate.

Ductile, self-supporting films of chromium metal having a minimum thickness of ~500 $\mu g/cm^2$ were formed by vacuum evaporation and electron-bombardment heating of the parent metal. A substrate of polished stainless steel was coated with a film of CsI as a parting agent and was heated to 250°C while the chromium was being deposited. Migration of chromium atoms or crystallites over the surface of the substrate at 250°C apparently enhances the continuity and uniformity of the film. High vacuum (10^{-6} torr) is required to reduce oxidation at crystal interfaces within the film, since quantities of oxygen in the ppm range are sufficient to embrittle the film.

METALLURGY AND MATERIALS RESEARCH

Preparation of Pure Materials. – A fourth LiF crystal containing 99.99% Li^7 and weighing 349 g was produced. It has been forwarded to the Materials Science Center of Cornell University for cleavage and thermal conductivity studies.

Magnesium oxide crystals ~1.5 mm in diameter and displaying 111 faces were grown from a fluoride melt.

Fundamental Research in X-Ray Diffraction. – An x-ray diffraction study of radiation damage in BeO was begun last year. Diffractometer traces of irradiated powder showed selective line broadening, line asymmetry, and line displacements. These effects were interpreted in terms of a model for the material in which the radiation debris was arranged in atomic planes parallel to the close-packed planes of the crystal.

Recently, good-quality BeO single crystals on the order of several tenths of a millimeter in dimension were grown. Single-crystal diffraction photographs of an irradiated crystal confirm the general interpretation of the powder pattern, but provide, as well, considerably more detailed information about the intensity in reciprocal space. An effort to interpret the distribution in terms of a more refined model is in progress.

Fundamental Investigation of Radiation Effects in Solids: Channeling of Energetic Atoms in Solids. – Detailed studies of the slowing down in crystalline solids of atoms having energies in the 1- to 50-kev region are being carried out with the aid of an IBM 7090 computer. A significant fraction of primary atoms, emitted isotropically from the lattice sites of a face-centered cubic structure, travel exceedingly large distances before coming to rest. These penetrating particles prove to be moving almost exclusively in <110> directions of the lattice, that is, in channels bounded by the close-packed atomic rows of the crystal. If the primary particles are distributed uniformly over low-index surfaces of the crystal, channeling is also observed in <100> directions and, to a much lesser extent, in other directions (<013>, <112>, and <111>). These results provide a satisfying explanation of the experimentally observed dependence on target orientation of the sputtering yield of monocrystalline Cu by Ar^+ ions. They also provide an interpretation of the previously unexplained penetrating component observed in experimental studies of the penetration of ions into aluminum targets.

Fundamental Investigation of Radiation Effects in Solids: Influence of Channeling on Displacement Cascade Theory. – A problem of long standing in radiation-damage theory is the discrepancy between the number of displaced atoms deduced from experimental data and that predicted by the theory of the displacement cascade. Several lines of investigation indicate that the discrepancy increases as the energy of the primary displaced particle increases, that is, that the number of displacements produced is not a linear function of the energy of the primary atom. The channeled atoms discovered in the computer studies lose their energy in such small increments that they produce virtually no displacements and

offer a possible explanation of the existing discrepancy between theory and experiment. The conventional cascade theory has been modified by assuming that every particle in the cascade has an energy-independent probability of being channeled and, hence, of producing no additional displacements. This modification leads to a number of displacements which depend on the energy of the primary particle to a power less than unity. Recent experiments on radiation damage in copper produced by fast and slow neutrons make possible an estimate that about 7% of displaced copper atoms are channeled. This amount is in reasonable agreement with the computer studies.

Fundamental Investigation of Radiation Effects in Solids: Effects of Neutron Irradiation on Strain-Amplitude-Dependent Internal Friction of Copper. – The phenomenon of radiation hardening in metals by neutron bombardment may be separated into two parts: (1) source hardening, that is, an immobilization of dislocation motion by defect pinning action right at the dislocation sources, and (2) lattice hardening due to the presence of neutron-produced defects or defect clusters in the lattice which are encountered by the dislocations as plastic flow proceeds. A series of experiments has been started which is designed to study source hardening in copper, by use of internal-friction techniques. Neutron bombardments from about 10^9 to 10^{14} nvt can be administered to the sample in situ by using a beam from the ORR. After a given dose the internal friction of the sample is measured as a function of strain amplitude of the sample from about 2×10^{-8} to 2×10^{-5}. As dislocation breakaway from the neutron-produced pinning points (presumably vacancies or vacancy aggregates at 100°C) occurs with increasing strain, the internal friction rises rapidly. Preliminary results indicate that the breakaway stress is about the same as that observed by Young from etch-pit observations in copper in the dose range 10^{13} to 10^{14} nvt. It is hoped that information concerning the variation of the interaction force between the pinning points and dislocations as a function of the integrated neutron flux may be obtained from these experiments.

CONTROLLED THERMONUCLEAR RESEARCH

Plasma Physics and Arc Research. – The study of the deuterium plasma formed between magnetic mirrors by the pressure-gradient arc (mode II) has continued. A microwave cavity enclosing the plasma region has permitted an increase in the power (up to 5 kv at 500 ma) in the electron stream while the characteristics of the mode II plasma are retained. An x-ray flux from the plasma region has been observed. Assuming a point source and isotropic distribution, it is estimated that a total of 10^{10} x-ray photons/sec were emitted by the plasma. The peak in the x-ray energy distribution is at about 100 kev, with a maximum energy of 250 kev. The electron temperature determined from the distribution was about 30 kev, and the density was 4×10^{11} electrons/cm^3, assuming a plasma volume of 100 cm^3.

DCX-1 Facility. – Copious data have been accumulated on the coherent rf signals from plasmas formed by gas dissociation at pressures near 10^{-7}

torr. It is believed that these data will permit an unambiguous description of the roles of both the ion and electron components during the onset and initial growth of the electrostatic instability. That the instability can be damped by altering the median-plane H_2^+ beam trajectory has been confirmed in these rf studies, and it has been demonstrated that slight deflections of the trajectory out of the median plane also have a stabilizing effect. In addition, distinct effects of applying a high positive potential (about 2.5 kv) to one of the end walls have been noted.

DCX-2 Facility. – Experiments with plasma probes have provided a better understanding of the performance of the DCX-2. These experiments show that no significant number of hot ions are escaping through the mirrors or striking the liner walls. Langmuir-type probe characteristics obtained with a gridded probe on the axis are such as would be expected from a plasma at a potential of +500 to +900 v with respect to the tank walls, with an electron temperature of several hundred volts and with an ion temperature not over 20 to 30 v. The plasma potential is controlled by electron loss along field lines and can be varied by changing the potential on an electrode beyond the center of the mirror opposite the probe. When +1000 v was placed on small axial electrodes at both ends of the machine, the amplitude of the neutral-particle signal just after beam turnoff increased by a factor of 2.

Vacuum. – Use tests on Dow Corning 705 silicone diffusion pump oil have been initiated. The conditioning time for this oil appears to be as short as for any previously tested oil except the polyphenyl ether. The 705 oil (previously designated XF-4660) is reported to have a room-temperature vapor pressure of about 2×10^{-9} torr.

The test of the experimental variable-oil-level diffusion pump was concluded. For air, there was not found to be a significant change in speed as a function of oil height above the heater element, but for hydrogen, a maximum speed was found for an oil depth of 1 to 1-3/4 in. which varied with heater input. This indicates that surface-boiling elements have as their only probable advantage a minimizing of oil decomposition rates.

A facility for the testing of metal gaskets has been designed and is being constructed. Apparatus has been modified to allow testing of some of the recently developed high-vacuum oil diffusion pumps and system components.

BIOLOGY AND MEDICINE PROGRAM

TERRESTRIAL AND FRESH-WATER ECOLOGY

Evaluation of Fission Product Distribution and Movement in White Oak Drainage System. – The rectangular weir at the gaging station White Oak Creek at ORNL has been replaced by a 4-ft Parshall flume. Low-base flow conditions have prevailed in the streams, except for short-term rises caused by rainfall.

Fall base-flow discharge measurements were made, and quality-of-water samples were collected at the 26 partial-record and continuous-record stations in and near the Oak Ridge Reservation on December 3.

Despite heavy rainfalls this fall, water levels in observation wells in the ORNL area have risen only slightly. The 600-ft-deep well on Melton Hill has shown no response to rainfall, and the water level has continued to decline, reaching 286.6 ft below land surface in the second week of December.

Radionuclides in Soils and Plants of Upper White Oak Lake Bed. — More than twice as much Ru^{106} and rare-earth elements per gram were found in the 0 to 1-in. layer of soil than in the 1- to 4-in. layer. In contrast, Cs^{137}, Co^{60}, and Sr^{90} increased in concentration with soil depth. These results can be explained on the basis of the past and present mechanisms of contamination. The Cs^{137}, Co^{60}, and Sr^{90} were deposited with the sediments of the former White Oak Lake, whereas the Ru^{106} and rare-earth elements occur as part of the redistribution by flooding of contamination arising from nearby surface seeps of the liquid-waste pits. Average concentrations of radionuclides ($\mu\mu c/g$) for the 0 to 1-in. and 1- to 4-in. layers of soil are: Ru^{106}, 180,000 vs 87,000; Cs^{137}, 16,000 vs 24,000; Co^{60}, 6500 vs 7100; Sr^{90}, 210 vs 300. Total rare earths (dis min^{-1} g^{-1}) are 890 vs 390.

Relative concentrations of the different radionuclides in plants reflected the values for soil, but the actual concentrations were lower. The most efficient soil-to-plant transfer (based on the 0 to 1-in. layer) was 50% for Sr^{90}. Transfer efficiency for all radionuclides detected in soils and plants was inversely related to the concentration in the soil. Thus the transfer efficiency for Ru^{106} was only 2%, but the concentration was 38 times greater than the concentration of Sr^{90} in plants.

Blood Responses of Mammals in Radioactive Waste Areas. — Interspecific differences of erythrocyte number within the rodent families Cricetidae, Muridae, and Sciuridae revealed that erythrocyte number was inversely related to species body weight, while mean corpuscular volumes are directly related to body weight. Also, the number of erythrocytes and hematocrit percentages are greater in male than in female cotton rats, in contrast to reports by others that these values are not different in the sexes of laboratory rodents.

The first phase of an experiment designed to test the blood responses of adult cotton rats living in pens on the radioactive White Oak Lake bed was completed. The average accumulated absorbed dose estimated with glass-rod microdosimeters was 170 rads in a 10-week period. The mean numbers of erythrocytes at prerelease and at 2-week, 6-week, and 10-week postrelease periods were, respectively, 6.87, 6.39, 6.76, and $6.78 \times 10^6/mm^3$ for the cotton rats on the uncontaminated control area and 7.04, 6.36, 6.74, and $6.47 \times 10^6/mm^3$ for the animals on the contaminated area. The mean numbers of leukocytes for the same periods were 3.64, 5.59, 4.97, and $6.40 \times 10^3/mm^3$ (uncontaminated area) and 4.54, 6.74, 5.29, and $5.31 \times 10^3/mm^3$ (contaminated area). The increases in leukocytes were primarily attributable to increases in the numbers of neutrophils. The percent change in various blood values in 10 weeks for the rats on the uncontaminated and contaminated areas were, respectively, erythrocytes (−1, −8), hematocrit (−9, −13), mean corpuscular volume (−9, −6), total leukocytes (+78, +18), neutrophils (+276, +164), lymphocytes (+32, −28), and eosinophils (+20, +48). The total number of leukocytes and hematocrits was less in the young rats born on the lake bed than in the young rats born on the uncontaminated area.

Numbers of erythrocytes and total leukocytes were also less in unconfined rice rats living on the lake bed than in rice rats living on uncontaminated areas.

Varying Rates of Litter Breakdown in Contrasting Climates. — Two- and three-year comparative experiments, in cooperation with the Botany Department of the University of Tennessee, showed different rates of breakdown of several kinds of deciduous leaves measured in the Oak Ridge Reservation and the Great Smoky Mountains. In all environments, leaf species show large and consistent differences in weight loss of leaves confined in bags of nylon net. Mulberry decayed rapidly to black humus, losing 0.002 to 0.0046 of its total weight per day in different environments. Beech showed slowest visible signs of decay, with fractional weight losses only 0.003 to 0.0012 per day. White oak, shumard red oak, and sugar maple showed intermediate rates of weight loss and visible destruction. For any given leaf species, breakdown was more rapid in deciduous stands than in adjacent conifer stands (white oak vs shortleaf pine at 260 m above sea level; cover hardwood vs hemlock at 1040 m; beech vs spruce at 1600 m). Differences between north and south slopes were small and inconsistent for various leaf species. Published results on first-year breakdown rates in 1959 showed a linear relation of average decomposition rate to elevation. However, 1960–61 results showed relatively slower decay at low elevations (probably due to frequent drying during the warm summers) and relatively faster decay at mid-elevations, where moisture, temperature, and organisms were favorable for decay. The fact that the years differ emphasizes the need for several-year experiments, using closely compared sets of techniques, for drawing generalizations about rates of ecological processes which affect the movement of nutrients or isotopes in contrasting environments.

Cs^{137} Transfers in Forest Floor Litter Layer. — Distribution and availability of Cs^{137} in litter and soil beneath the crowns of tagged tulip poplars were measured as part of the characterization of the environment of the microflora of the forest floor. Before leaf fall, more than 90% of the Cs^{137} of the forest floor (91 $\mu c/m^2$) was found in tree roots. Percentages of cesium leaching from tree crowns were: 50% retained in litter, 40% in the top 1-1/2 cm of humus + soil, and 10% in deeper layers. Leaf fall increased cesium content of the litter layer from 7 to 82 $\mu c/m^2$. Leachability of Cs^{137} from freshly fallen leaves was 6 times higher than from green leaves. Subsequent increased leaching from fresh litter increased the Cs^{137} content of the top 1-1/2 cm of soil + roots from 84 to 104 $\mu c/m^2$. Rocks and old compacted untagged leaves interferred with downward leaching of Cs^{137} by rain and reduced Cs^{137} concentration in the subsoil. Before and after fall of freshly tagged leaves, contamination of litter varied by less than one order of magnitude along a distance of 50 cm. In series of cores taken before leaf fall, Cs^{137} changed by more than two orders of magnitude within distances of 10 cm, and by more than one order of magnitude within only 2.5 mm. High correlation between Cs^{137} in the cores and in the roots taken from the cores ($r = 0.94$) resulted from the large amounts in roots. The percent of Cs^{137} that could easily be leached from the various components

of the forest floor and thus could become available to adjacent microbes increased in the order: soil, litter, leaves. Before leaf fall, these percentages were 0.6, 7.7, and 11.2 respectively. After leaf fall the corresponding percentages had increased to 2.3, 10.3, and 64.0 as a result of decreased turgor of the now fallen leaves and subsequent infiltration of Cs^{137} from these leaves into litter and soil.

Cs^{137} Movement in Forest Food Chains. – Measurements of Cs^{137} concentrations in the biota of a tagged forest plot are being used to evaluate cesium movement through forest food chains. During the summer of 1962, Cs^{137} transfer to herbivorous and predaceous arthropods from green leaves and from leaf litter was estimated from weekly samples. Insects feeding directly on green foliage had Cs^{137} concentrations about 50% as high as concentrations in foliage itself, which suggests a transfer efficiency similar to that estimated for insects living on the contaminated White Oak Lake bed. Cesium-137 also accumulated in organic matter on the forest floor as a result of leaching action of rainwater on the green leaves. Herbivorous and omnivorous arthropods sampled from the forest floor could be separated into three groups: one group with low Cs^{137} activity obtained from the forest floor directly, a group with intermediate activity evidently obtained through partial feeding upon green vegetation, and a group with high activity due to extensive feeding upon foliage. Peaks of Cs^{137} activity in some predaceous arthropods (*Pasimachus*, *Lycosa*) coincided with abundances of particular prey species (geometrid caterpillars). Cesium-137 concentrations in forest-floor animals showed an abrupt increase associated with leaf fall. Collection of these arthropods is being continued throughout the winter.

RADIOLOGICAL PHYSICS, HEALTH PHYSICS, AND INSTRUMENTATION

Aerosol Studies. – Methods of measurement and redispersion of solid particulate surface contamination are being evaluated as a function of surface characteristics. The likelihood of redispersing particulates by air blowing over a given surface is essentially the same for a wide range of surface roughness (e.g., Plexiglas surfaces ranging from 8 to 1000 μin. rms deviation). On the other hand, the apparent hazard, as indicated by dry-smear and adhesive-paper measurements, decreases as surface roughness increases. Other studies have confirmed the fact that surfaces covered with an oily film show little redispersion due to air currents, whereas the apparent hazard as indicated by smears and adhesive-paper samples is the same as for clean surfaces of the same material.

ISOTOPE DEVELOPMENT PROGRAM

Recovery of Fission Products. – Continued studies of substituted phenols as extractants for the recovery of cesium from waste solutions showed that a commercial phenol, 2-benzyl-4-chlorophenol (Monsanto Santophen-1), has the highest cesium extraction power of any phenol tested

so far. In a batch countercurrent test with 1 M Santophen-1 in diisopropylbenzene, 99.8% of the cesium was recovered from simulated Purex 1WW solution (tartrate-complexed and adjusted to pH 12.3) in six extraction and two scrub stages (the scrub solution was 0.01 M NaOH, 0.002 M sodium tartrate) at feed/organic/scrub ratios of 6/3/1. The cesium was quantitatively stripped in a single contact with a 0.05 volume of 0.05 M HNO_3, resulting in an overall cesium concentration of 40 and an overall cesium-from-sodium separation factor of about 6×10^4. In a similar test with 0.6 M Santophen-1, more than 97% of the cesium was recovered at feed/organic/scrub ratios of 5/5/1. As in the previous test, the cesium was quantitatively stripped, resulting in an overall cesium concentration of about 20 and overall separation factors of about 6×10^4 from sodium and about 260 from rubidium.

Further tests with 4-sec-butyl-2(α-methylbenzyl)phenol show that it has excellent selectivity for cesium over sodium, other fission products, and corrosion products and has an almost negligible loss ($\leq$0.4 g/liter) to adjusted 1WW solution.

Preliminary tests showed that certain branched-chain carboxylic acids, for example, neo-tridecanoic acid, extract strontium preferentially to sodium from basic sodium nitrate solution. The strontium extraction increases rapidly above pH 7, reaches a sharp maximum between pH 8.5 to 9.5, and then decreases at higher pH's.

Radioisotope Research and Development. — A new thermal-conductivity type of calorimeter for rapid routine assay of Cs^{137} products in the Fission Products Development Laboratory was designed, fabricated, and calibrated. The calorimeter has a 3/8-in.-thick depleted-uranium head which absorbs 95% of the total decay energy from the Cs^{137}-Ba^{137}. The time required to complete an assay of Cs^{137} was reduced by 50%, and a considerable savings in costs was effected.

Tracer quantities of trivalent americium (10^8 dis/min) have been converted to the sexivalent state by treatment with sodium bismuthate at room temperature in 6 to 8 N HNO_3. Trivalent lanthanides and actinides are scavenged with lanthanum fluoride after the oxidation, leaving sexivalent americium in solution. Since sodium bismuthate is a solid, good agitation is essential for rapid reaction. Solid products from the treatment are easily removed by filtering or centrifugation. The method should provide a reliable analytical procedure which does not require the critical acidity control of the persulfate method and may be applicable to chemical separation of americium from curium.

A preliminary study of some of the stable compounds of mercury was made to determine the feasibility of using these compounds in the ORR for production of Hg^{203}. The substances given consideration are HgF_2, HgS (red), HgS (black), K_2HgCl_4, Au-Hg amalgam, and dental amalgam.

The compounds HgF_2, HgS, and K_2HgCl_4 are quite stable, but in the presence of moisture and radiation they will attack aluminum metal and thus are not considered acceptable as ORR irradiation targets. Future studies will include the investigation of Hg-inorganic exchanger compounds.

A glass containing 43% cesium has been developed as a source material for preparing cesium gamma sources in the shape of molded bars. This glass, $0.15Li_2O \cdot 0.55Cs_2O \cdot 0.3CdO \cdot 2.7SiO_2$, is prepared from a cesium

salt, lithium carbonate, cadmium oxide, and silica. The glass, which melts at 495 ± 10°C, can be molded into bars at 1200 to 1250°C in a graphite mold. The cesium in the glass has a solubility of 0.8 mg cm^{-2} day^{-1} in water, and the leach rate is linear with time. The glass has a density of ~3.0 g/cm^3. The volatility loss at the fusion temperature is ~5 mg of Cs_2O cm^{-2} hr^{-1} at 1200°C and ~10 mg of Cs_2O cm^{-2} hr^{-1} at 1250°C.

An all-glass, laboratory-scale, air-pulsed mixer-settler of five stages was operated under a wide variety of conditions to determine some of its operating characteristics. A number of solvent systems were tested in which the difference in the specific gravity of the phases varied from 0.05 to 0.7. The flow capacity of the unit is directly dependent on the difference in density of the phases and the viscosity of the organic and liquid phases.

Radioisotope and Stable Isotope Production. – Ion currents equivalent to 3220 g of separated isotopes were monitored, and isotopes of Ca, Fe, Te, Br, Ta, Er, Yb, and Si were separated and collected. Recovery of unresolved erbium feed material is estimated at ~80%.

The Yb_2O_3 calutron feed material is being recycled for the current separation.

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June 1962	ORNL-3321
July 1962	ORNL-3331
August 1962	ORNL-3351
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