346

CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION

[redacted]

MARTIN MARIETTA ENERGY SYSTEMS LIBRARIES
3 4456 0361380 0

ORNL-2736
Progress

[redacted]

OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

APRIL 1959

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

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

Downgrade to Unclassified After May 6, 1960

[redacted]

[redacted]

ORNL-2736

Copy No. ____

Contract No. W-7405-eng-26

OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

APRIL 1959

DATE ISSUED

MAY - 7 1959

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

[redacted]

MARTIN MARIETTA ENERGY SYSTEMS LIBRARIES

3 4456 0361380 0

CONTENTS

PROGRAM 2000 — SPECIAL NUCLEAR MATERIALS..... 1
Power Reactor Fuel Reprocessing Pilot Plant..... 1
Dissolver Solution..... 1

PROGRAM 4000 — REACTOR DEVELOPMENT..... 3
HOMOGENEOUS REACTOR PROJECT..... 3
Homogeneous Reactor Test..... 3
Fuel Processing..... 3
Thorium Oxide Slurry Development..... 4
HRP Chemistry: Reactions in Aqueous Solutions..... 4
HRP Chemistry: Heterogeneous Equilibria in Aqueous Systems. 5
HRP Analytical Chemistry..... 5
Slurry Testing and Evaluation..... 6
Slurry Blanket System Development..... 7
Engineering Research..... 7
Core and Blanket Vessel Development..... 7
Centrifugal Circulating Equipment..... 8
Miscellaneous Equipment..... 8
Heat Removal Equipment..... 8
Gas Handling Equipment..... 9
Fuel Systems Development..... 9
Feed Pump Development..... 9
MOLTEN-SALT REACTOR (MSR) PROGRAM..... 9
Chemistry..... 9
Physical Properties..... 10
Fabrication Studies..... 11
INOR-8 Filler Metal Development..... 11
Fuel Penetration into Graphite..... 11
In-Pile Loop Tests..... 12
Pump Development..... 12
Nuclear Calculations..... 13
GAS-COOLED REACTOR PROJECT..... 14
LITR Irradiation Experiments..... 14
ORR Irradiation Experiments..... 14
Closed-Cycle Experiment..... 15
Engineering Research..... 15
Graphite and Coating Studies..... 15
PCTR Material Procurement..... 16
PACKAGE REACTOR DEVELOPMENT..... 16
Army Package Power Reactor (APPR-1)..... 16
GENERAL REACTOR RESEARCH..... 16
Basic Reactor Research..... 16
Fuel Element Development..... 17
Power Reactor Fuel Reprocessing: Alternative Aqueous Process Studies..... 18
Power Reactor Fuel Reprocessing: Zircex Process..... 18
Power Reactor Fuel Reprocessing: Zirflex Process..... 18
Power Reactor Fuel Reprocessing: Solvent Studies..... 18
Power Reactor Fuel Reprocessing: Mechanical Processing..... 19
Power Reactor Fuel Reprocessing: Darex Process..... 19

Power Reactor Fuel Reprocessing: Sulfex Process 20
Waste Disposal Research and Engineering: Disposal of High-Level Radioactive Wastes by Sintering..... 21
Waste Disposal Research and Engineering: Regional Burial Ground Investigations..... 22
Waste Disposal Research and Engineering: Mockup of Field-Scale Salt Experiment..... 22
Waste Disposal Research and Engineering: Laboratory Studies of the Chemical Interaction of Waste and Salt... 23
Waste Disposal Research and Engineering: Design and Construction for Field-Scale Salt Experiment..... 23
Waste Disposal Research and Engineering: Corrosion Studies for Field-Scale Salt Disposal Experiment..... 23
Waste Disposal Research and Engineering: Process Waste Water Treatment Plant..... 24
Waste Disposal Research and Engineering: Maritime Hazards Study..... 24
Waste Disposal Research and Engineering: Tank Hazards..... 24
Waste Treatment..... 25
Radiation Damage: Reactor Flux Measurements..... 26
THERMONUCLEAR PROJECT..... 26
DCX Facility..... 26
Carbon Arc Studies..... 26
Vacuum Studies..... 27

PROGRAM 5000 — PHYSICAL RESEARCH..... 28
ISOTOPE PRODUCTION..... 28
Radioisotope and Stable Isotope Production..... 28
Radioisotope Research and Development..... 28
Stable Isotope Development..... 29
PHYSICS..... 29
Classification of Low-Lying Nuclear Energy Levels..... 29
The 86-Inch Cyclotron: Nuclear Physics..... 30
The 86-Inch Cyclotron: Applied Physics..... 31
Heavy-Particle Physics..... 31
Electronuclear Machines..... 31
Relativistic Isochronous Cyclotron..... 32
Special Separations..... 32
CHEMISTRY..... 33
Druhm Process..... 33
Chemical Engineering Research..... 33
Transuranics..... 33
Feed Materials Processing: Fluorox Process..... 34
Feed Materials Processing: UO_2 Sinterability..... 34
Equipment Decontamination..... 35
Nuclear Chemistry..... 36
High-Temperature and Structural Chemistry..... 36
Molten-Salt Thermodynamics..... 36
Analytical Chemistry Research..... 37
METALLURGY..... 38
Fundamental Investigations of Radiation Damage in Solids... 38

[redacted]

PROGRAM 6000 — BIOLOGY AND MEDICINE 39
BIOPHYSICS 39
Waste Disposal Research: Evaluation of Soil Disposal for Intermediate-Level Liquid Radioactive Wastes 39
Waste Disposal Research: New ORNL Solid-Waste Burial Ground 39
Dosimetry Applications 39
OPEN LITERATURE PUBLICATIONS 41
FOREIGN VISITORS 42

[redacted]

[redacted]

OAK RIDGE NATIONAL LABORATORY
STATUS AND PROGRESS REPORT

April 1959

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

PROGRAM 2000 — SPECIAL NUCLEAR MATERIALS

Power Reactor Fuel Reprocessing Pilot Plant. — The plant was on-stream 15 days during this month, and a total of 4 metric tons of irradiated uranium from BNL and Savannah River reactors was processed. The down time of 15 days was for equipment replacement and plant cleanout between programs.

Gross gamma decontamination factors in solvent extraction were generally lower than expected, and flowsheet changes will be made to improve decontamination. In the co-decontamination cycle, gross gamma decontamination factors decreased from 2200 to 500 during 12 days of operation while Savannah River fuel was being processed. The major fission product contaminants in the product from each cycle were zirconium and niobium. Based on measured losses from the plant, product recoveries for uranium and plutonium were 96 and 98%, respectively. Unusually high losses during this month were caused by plant cleanout and planned high losses to satisfy the requirements of a special program. (AEC Activity 2351)

Dissolver Solution. — Work has continued on the three phases of the dissolver program outlined in the February monthly report (ORNL-2706).

Studies of the effects of 12 fission product elements (I, Ru, Rb, La, Nd, Mo, Nb, Ce, Sb, Ag, Eu, Cs) on the amine-extraction step of the amine-extraction coulometric-titration procedure for the determination of uranium in dissolver solutions were made. Radioactive tracers were used in order to learn what elements carried through the extraction procedure. Molybdenum, ruthenium, lanthanum, and neodymium were found to extract appreciably. Of these elements, molybdenum causes the most interference in the titration procedure.

[redacted]

A method of decreasing the interference of molybdenum was developed. It was found that an additional scrub of the organic phase with an acid-deficient aluminum nitrate solution and the addition of concentrated phosphoric acid into the titration cell permit an accurate determination of uranium. This procedure was applicable when molybdenum-uranium ratios of 1 to 5 were present in the original synthetic dissolver solution. The relative standard deviation was found to be 0.5%.

The controlled-potential coulometric titration of plutonium, which was outlined in the February monthly report, was found to possess relative standard deviations of 0.1 to 0.2% when 1 mg or so of plutonium is titrated, and 0.3 to 0.5% when 200 μg or so is titrated. Preliminary studies indicate that the method is tolerant to very large quantities of uranium and to large amounts of iron, but is subject to serious interference from mercury which is often present as a dissolution catalyst. Some work was devoted to finding a simple method for removal of mercury, but without satisfactory success. Attempts are now being made to couple anion exchange – a very efficient plutonium separation – with the coulometric titration mentioned above. (AEC Activity 2724)

PROGRAM 4000 — REACTOR DEVELOPMENT

HOMOGENEOUS REACTOR PROJECT

Homogeneous Reactor Test. — Studies on the performance of the reactor circulating pumps and on the pressure drops in the main circulating loops established that the core-loop volumetric flow rate and pump head were practically normal. The blanket-loop flow rate was found to be about one-fourth normal, and the pump was delivering less than one-tenth of its required head. Consequently, the blanket pump was replaced. The damaged pump was cut open remotely for examination. A cursory visual inspection revealed that about three-fourths of the titanium impeller and most of the labyrinth seal had been consumed, apparently by combustion.

Remote-maintenance operations, which included replacement of one of the core feed-pump diaphragm heads, are being concluded. Run 20, with investigation of fuel stability as the primary objective, will follow. (AEC Activity 4103.3)

Fuel Processing. — The removal of corrosion-product nickel and manganese from HRT fuel solution by electrolysis with a mercury cathode was found to be feasible in laboratory tests. A final concentration for total nickel and manganese of less than 50 ppm was consistently obtained, with no detectable contamination of the treated fuel with mercury. Before electrolysis, the fuel solution had to be concentrated to about 60 g of U per kg of D_2O to obtain a D_2SO_4 concentration sufficient to prevent precipitation of the tetravalent uranium obtained during electrolysis. After treatment, the residual uranous ion was effectively oxidized back to uranyl by treatment with an O_2-O_3 mixture.

The solubility of nickel, copper, and manganese in the mercury cathode is about 1%. In an HRT-scale test using a glass-lined cell with a platinum anode and with a mercury cathode that was not agitated, an insoluble spongy amalgam rapidly formed on the surface of the mercury and shorted the cell. This indicated that agitation of the mercury is necessary to attain even the 1% concentration. Regeneration of the used mercury was best accomplished by treating it with 0.5 M HNO_3 solution containing 1.5% H_2O_2. (AEC Activity 4103.1)

Thorium Oxide Slurry Development. — The results of irradiation experiments in the ORNL Graphite Reactor with a slurry of mixed thorium-uranium oxide containing 500 g of Th per kg of D_2O indicate that in this system the G value for hydrogen production is 0.8 molecule per 100 ev under stoichiometric D_2-O_2, 0.4 under a 300-psi partial pressure of D_2, and 0.6 under excess oxygen. Pumping a mixed thorium-uranium oxide slurry containing 0.002 m palladium catalyst under a partial pressure of D_2 in an out-of-pile loop for 500 hr at 280°C decreased the catalytic activity for D_2-O_2 combination from 12 moles to 0.1 mole of D_2 per hour per liter at 280°C and a D_2 partial pressure of 100 psi. Continued pumping of this same slurry for an additional 500 hr under excess O_2 did not significantly change the catalytic activity. This marked decrease in the catalytic activity of palladium under reducing conditions contradicts previous autoclave test results and is probably due to the buildup of corrosion products in the pumped material.

Thoria-gel beads, 1 to 125 μ in diameter, were prepared by spraying a thoria sol into isopropyl alcohol, which caused the sol to gel into spherical particles. (AEC Activity 4103.1)

HRP Chemistry: Reactions in Aqueous Solutions. — Uranium peroxide, prepared by precipitation upon the addition of 1.0 M hydrogen peroxide to 0.04 M uranyl sulfate solution, was discovered to occur in two different crystalline forms, corresponding to two different chemical compositions, depending on the temperature of the precipitation. Below 63.5°C a low-temperature form has consistently been produced, with a composition corresponding to $UO_4 \cdot 4H_2O$ and an x-ray diffraction pattern with a 2θ peak at 15.15°; above 64.5°C a high-temperature form has invariably been produced, with a composition corresponding to $UO_4 \cdot 2H_2O$ and an x-ray diffraction pattern having a 2θ peak at 16.92°. The tetrahydrate is easily converted into the dihydrate by heating at 90°C in air for a few hours or by overnight exposure to magnesium perchlorate in an evacuated desiccator; it has not been possible to convert the dihydrate into the tetrahydrate by any unambiguous procedure yet tested. The x-ray diffraction pattern for the tetrahydrate was erroneously attributed to the dihydrate (ORNL-2092). A yellow crystalline precipitate recently recovered from the fuel system

of the North Carolina State Reactor was incorrectly identified as the dihydrate; its x-ray diffraction pattern is identical with that of the low-temperature tetrahydrate mentioned above. (AEC Activity 4103.1)

HRP Chemistry: Heterogeneous Equilibria in Aqueous Systems. — The compound $3CuO \cdot SO_3 \cdot 2H_2O$ (antlerite) was synthesized at low temperature in quantity by refluxing an aqueous $CuCO_3$ slurry with a stoichiometric amount of H_2SO_4 at 100°C. A previously unidentified compound has been formed at high temperatures in the four-component system CuO-UO_3-SO_3-H_2O. This red crystalline solid was synthesized in bulk and identified by chemical analysis as $CuO \cdot 3UO_3$. X-ray diffraction data established that this compound is one of the solids which was formed on metal surfaces in HRE-2 during prepower operation. The binary curve for the two solid phases, $3CuO \cdot SO_3 \cdot 2D_2O$ and $CuO \cdot 3UO_3$, in the four-component system CuO-UO_3-SO_3-D_2O was established as a function of SO_3 concentrations from 0.04 to 0.4 m.

An investigation was carried out to establish whether the addition of $LiNO_3$ or $Be(NO_3)_2$ to $Th(NO_3)_4$-H_2O solutions would increase the temperature range of solubility of thorium and reduce the equilibrium concentration of nitrogen oxides in the vapor phase. The experiments did not indicate any substantial improvement (for solutions containing 3 moles or less thorium) by the addition of $LiNO_3$ and $Be(NO_3)_2$ in amounts equivalent to maximum mole ratios of $8LiNO_3$:$1Th(NO_3)_4$ and $2Be(NO_3)_2$:$1Th(NO_3)_4$.

The solubility of $NiSO_4 \cdot H_2O$ at 300°C as a function of H_2SO_4 concentration was shown to be greater in D_2O than in H_2O solvent.

The solubility of $UO_3 \cdot H_2O$ in HNO_3 solution was determined from 0.05 to 2 M HNO_3 and from 150 to 300°C.

The investigation of heavy- and light-liquid-layer compositions in the system UO_3-SO_3-H_2O was extended at 350°C to SO_3/UO_3 ratios of 1.4 for the starting solutions. Results at 350°C show excess free acid in both the heavy and light layers for initial ratios above 1.025. (AEC Activity 4103.1)

HRP Analytical Chemistry. — A radio-frequency titrimeter is being used in the direct titration of SO_4 and free acid in solutions of UO_2SO_4. The determination of SO_4 in a typical HRT fuel solution is accomplished by titrating with a $BaCl_2$ solution, utilizing high-frequency oscillometry

for the detection of the end point. The uranium must be masked with citrate or fluoride; otherwise, no inflection point in the titration, corresponding to the end point, can be noted. The coefficient of variation in the range of 40 to 70 mg of SO_4 is about 2%. The determination of free acid is carried out similarly by titrating the solution with NaOH. The coefficient of variation for the determination of approximately 0.2 meq of free acid is 2%.

A flame-photometric method for the determination of rare-earth elements in thorium was developed. The spectra, in the wavelength region of 380 to 800 mμ, and the limits of detection of all the rare-earth elements were determined after extraction with 2-thenoyltrifluoroacetone in 4-methyl-2-pentanone (hexone). When the organic solution, after extraction, is aspirated into the flame, the emissivity of the rare-earth element is enhanced approximately a hundredfold over that which is observed when aqueous solutions are aspirated. With the exception of lanthanum and ytterbium, the spectral bands of the rare-earth elements overlap and become obscure; therefore, for the determination of each specific element, a further separation is essential. (AEC Activity 4103.1)

Slurry Testing and Evaluation. — During three runs in the 200-gpm loops a mixed thoria-urania slurry proposed for use in an experimental slurry reactor was circulated for 2957 hr. The purpose of the runs was to extend the information on handling characteristics, rheological properties, attack on structural materials, heat transfer, settled-bed properties, and stability of this slurry at operating temperature. The slurry remained essentially Newtonian at concentrations up to 600 g of Th-U per kg of H_2O in the temperature range 150 to 200°C. Heat transfer coefficients for the slurry were not much different from those for water at the same temperature and flow for all concentrations tested. Solids that accumulated in a 14-ft vertical stagnant line for periods of several days could be returned to circulation with the aid of as little as 20 psi pressure above the static head. The uranium content of the slurry appeared to be stable, and there was no appreciable change in particle size. Generalized attack rates were less than 0.2 mil/year. (AEC Activity 4103.1)

Slurry Blanket System Development. — The 300-SM blanket test facility was operated with the new high-pressure vessel set up for re-entrant bottom flow, simulating a single-region reactor. From 93 to 96% of the inventory was in circulation at all flow conditions, and a substantial part of the missing material could be accounted for by holdup in the dead-ended lines and in the pressurizer. Slurry samples drawn from within the vessel indicated the possible existence of slight concentration gradients which were affected by temperature, concentration, and flow rate. There was evidence that sampler-tube orientation influenced the performance of the samplers, and there also was evidence of slightly higher concentrations in the region of the conical wall at all flows. (AEC Activity 4103.1)

Engineering Research. — Slurry dropout velocities were determined in horizontal pipes as a function of thoria concentration, floc diameter, pipe diameter, and slurry yield stress. The data were correlated in terms of the volume fraction of solids and an effective Reynolds number; this Reynolds number was based on the effective viscosity and the fluid velocity at which dropout occurred. Below 8% solids content, the dropout Reynolds number increased with decreasing slurry concentration; above 8% solids, this Reynolds number was about 2100.

Several runs were made in the 30-gpm slurry loop to investigate caking properties of thoria-water slurries. The addition of chromate ion to a normally caking slurry resulted in no cake or sphere formation. In other runs, thoria cakes which had been deposited by a caking slurry were redispersed by subsequent circulation of a noncaking slurry. (AEC Activity 4103.1)

Core and Blanket Vessel Development. — Probes for measuring water velocities to within 0.1 in. of a core wall surface were procured and were applied to the 4-ft re-entrant spherical core model. Existence of a high-velocity stream near the core wall was confirmed. Calibrations of the probes in a 3-in. pipe checked literature velocities to within 3%.

Operation of the 30-in. slurry core vessel was begun at a nominal concentration of 500 g of Th per kg of H_2O. It was shown that consistent sampling bias could be introduced by use of sampling ports of different orientation to the flow stream.

The hindered-settling rate of a mixed thoria-urania slurry was found to be proportional to the 0.5 power of the acceleration, in the range 1 to 20g. This implies a breaking up of floc structure with increasing applied force.

Tests performed on the HRT core model indicated that solids produced by screen removal could be cleaned out with a reverse flush of about 1000 gal of water. (AEC Activity 4103.1)

Centrifugal Circulating Equipment. — The HRT replacement blanket circulating pump was fitted with a stainless steel 230-gpm impeller, and performance data were taken at room temperature over its operating range. The pump was then pretreated and cleaned in consecutive runs by circulating trisodium phosphate solution, dilute nitric acid, and hot oxygenated water. The replacement fuel circulating pump was similarly pretreated.

The 300A slurry pump completed a 2800-hr run in which thoria was circulated at 280°C.

The temperature of the steam which could be circulated by the 20-cfm canned-motor blower was increased from 170°C to 225°C. Overheating of the motor by condensation of steam at the interface between the vapor and the cold water in the rotor cavity has prevented operation at higher temperatures. (AEC Activity 4103.1)

Miscellaneous Equipment. — Development of equipment for inspection and modification of the HRT core was continued. Devices fabricated and tested successfully were a positioner for the core-tank thickness gage, a screen-edge sample cutter, a pit-impression device, a device for mounting over the hole in the core tank a patch that can be sealed mechanically, and an improved external-light-source telescope viewer. Preliminary testing and debugging of machines for cutting screens loose and removing them from the core were carried out. (AEC Activity 4103.1)

Heat Removal Equipment. — Design of the heat exchanger test facility is 80% complete. Its pressurizer will be a full-flow type, with the flow entering just above the conical bottom in a downward-directed nozzle. Griscom-Russell Company is proceeding with fabrication of part of the test steam generators.

A model of the HRT heat exchanger headers and of a U-tube bundle was tested, in order to develop procedures for mechanical-hydraulic cleaning

of fouled heat exchangers. The header was cleaned readily with a reverse flush (flow entering the tube side). Tubes were cleaned with the combined imposition of vigorous vibration and hydraulic pressure. (AEC Activity 4103.1)

Gas Handling Equipment. — The natural-circulation high-pressure recombiner was operated routinely at 1500 psi up to a total of 1500 hr. (AEC Activity 4103.1)

Fuel Systems Development. — The HRT mockup was operated for 694 hr with uranyl sulfate fuel at 1500 psi and 280°C in reversed flow in anticipation of reversing the flow through the HRT core. With the conventional stainless steel pressurizer in service, the fuel remained stable at all pressurizer purge rates up to more than 10 gph, a condition which led to a uranium precipitation rate of 24 g/day in previous operation with forward flow. There was no noticeable difference in gas-separator performance with reversed flow, and the high-pressure-system liquid level was controlled at a liquid entrainment of 0.21 gpm in a 13-scfm gas letdown stream. Performance of the system was satisfactory in all respects. (AEC Activity 4103.1)

Feed Pump Development. — Successful operation of two prototype HRT fuel feed pumps continued to a total of 16,300 hr. The double-diaphragm 10 3/4-in. pump and the 12-in. pump continued to operate to a total of 7791 and 3169 hr, respectively. Two slurry heads operated to a total of 4000 hr without failure, although pumping output was reduced as a result of slurry dropout in the pump suction line.

The three-stage contaminated-service oxygen compressor (Pressure Products Industries) was placed on test, compressing 3 scfm of atmospheric air to 2500 psi. Some minor difficulties required modifications, and a diaphragm replacement was necessitated by failure caused by a metal chip. (AEC Activity 4103.1)

MOLTEN-SALT REACTOR (MSR) PROGRAM

Chemistry. — The maximum ThF_4 concentration available at 500°C in a molten-fluoride-salt blanket mixture containing Li^7F occurs at the composition LiF-BeF_2-ThF_4 (67.5-17.5-15 mole %), corresponding to about 1.4 kg/liter of thorium. The main constituents in the solidified melt are

$2LiF \cdot BeF_2$ and a $3LiF \cdot ThF_4$ phase incorporating small amounts of BeF_2 as a solid solution.

Attempts to measure the activities of nickel in nickel-molybdenum alloys have revealed that the currently used annealing procedures are only partially effective in transforming the high-temperature phases α and $\alpha + \delta$ into β, $\beta + \gamma$, and γ, which are the equilibrium phases of interest at reactor temperatures.

Tracer experiments, employing Fe^{59}, have removed any doubt that the reduction of Fe^{++} by Cr^{0} proceeds rapidly to completion at 600°C in NaF-ZrF_4 (53-47 mole %). Because of the difficulty of avoiding contamination with extraneous iron, there had formerly been some uncertainty regarding the completeness of the reaction; even in the case reported here, 125 ppm of Fe was found by wet chemical analysis of a filtrate which contained no detectable Fe^{59}.

Analyses of additional machined cuttings from 1-in.-dia TSF graphite rods, which were impregnated (after an outgassing treatment) with LiF-BeF_2-UF_4 (62-37-1 mole %) by exposure for 48 hr at 1250°F under a pressure of 15 psig of helium, have continued to show that most of the salt is in the outer 1/16-in. layer. However, the uranium concentration in the center region is disproportionately high, both in comparison with intermediate distances and in comparison with the BeF_2 concentration in the center.

The use of steam to precipitate uranium as UO_2 from fuel mixtures such as LiF-BeF_2 (63-37 mole %) containing 5.6 wt % UF_4 and 1.25 wt % CeF_3 has been studied as a possible reprocessing procedure. The uranium in solution was reduced to 100 ppm without a detectable decrease in the concentration of cerium or beryllium in solution. (AEC Activity 4103.1B)

Physical Properties. — The enthalpies and heats of fusion have been experimentally evaluated for two salt mixtures over a temperature range extending from 100 to 800°C.

For the salt mixture LiF-BeF_2-ThF_4 (71-16-13 mole %), the enthalpy of the solid (100–400°C) can be expressed as

$$H_T - H_{30°C}(cal/g) = -7.7 + 0.209T + (4.58 \times 10^{-5})\ T^2,$$

and the enthalpy of the liquid (550–800°C) can be expressed as

$$H_T - H_{30°C}(cal/g) = -47.4 + 0.473T - (11.90 \times 10^{-5})\ T^2,$$

where T is the temperature in °C. The heat of fusion at 500°C was found to be 74 cal/g.

For the salt mixture $LiF-BeF_2-UF_4$ (70-10-20 mole %), the enthalpy of the solid (100–400°C) can be expressed as

$$H_T - H_{30°C}(cal/g) = -17.8 + 0.289T - (3.50 \times 10^{-5})\ T^2,$$

where T is the temperature in °C. The heat of fusion at 450°C was found to be 53 cal/g. The viscosities of these salt mixtures between the temperatures of 550 and 800°C were found to be

$$\mu\ (\text{centipoises}) = 0.0526e^{4838/T(°K)}$$

for the 13 mole % ThF_4 mixture, and

$$\mu\ (\text{centipoises}) = 0.489e^{4847/T(°K)}$$

for the 20 mole % UF_4 composition. (AEC Activity 4103.1B)

Fabrication Studies. — An experiment has been completed to determine the feasibility of cladding a porous nickel strip with two Inconel tubes so that the porous nickel is between the tubes. Such a composite tube is being considered for heat exchanger application. The porous metal annulus would serve as a leak detection device. Examination of a triplex tube specimen fabricated by drawing (10% reduction in area) and sintering at 2150°F showed good bonding between the core and cladding and no apparent oxide formation at the interface. (AEC Activity 4103.1B)

INOR-8 Filler Metal Development. — A heat of INOR-8 filler metal (ORNL heat MP-9) containing 2% niobium has been melted, deposited as weld metal, and machined into tensile bars. This alloy exhibited an exceptionally high ductility of 30% at 1500°F, as compared with ductilities of 13% and 5% previously obtained for melts ORNL MP-3 and Westinghouse M-5, respectively. (AEC Activity 4103.1B)

Fuel Penetration into Graphite. — A specimen of CCN graphite was evacuated to less than 12 μ and was covered with molten $NaF-ZrF_4-UF_4$ (50-46-4 mole %) under a pressure of 150 psia for 100 hr at a temperature of 1300°F. Radiographic examinations of the graphite specimen before and after the test indicated that the fuel mixture had uniformly penetrated the pore (void) spaces of the graphite. The weight gain of the penetrated

specimen indicated that 29% of the calculated total pore volume of the graphite (17% of the bulk volume of the specimen) was filled with the fuel mixture.

The CCN graphite specimen was then thermally cycled to determine whether expansion of the fuel salt would cause physical damage, such as cracking or spalling. During the thermal cycling, thermocouples located near the outer surface and the central axis of the specimen indicated temperature changes from room temperature to 1436 and 779°F, respectively, within a 3-min period. In the cooling half of the cycle (2 min), the 1436°F temperature was lowered to 842°F and the 779°F temperature to 752°F. Radiographic and macroscopic examinations of the graphite specimen at room temperature did not reveal any damage. The fuel-impregnated graphite specimen will be subjected to additional thermal cycles. (AEC Activity 4103.1B)

In-Pile Loop Tests. — The assembly of the second MSR in-pile loop was completed. The loop was inserted in the MTR during the April 27 reactor shutdown. This loop and the loop controls incorporate modifications expected to overcome the difficulties encountered with the first loop. The prototype of the pump in this loop has completed more than 4000 hr of satisfactory operation. (AEC Activity 4103.1B)

Pump Development. — An INOR-8 bearing was operated with a carburized INOR-8 journal for 272 hr at 1200°F and at a load of 200 lb. Two hundred sixty starts and stops were made. Postrun examination of the bearing and journal showed both parts to be in excellent condition.

An INOR-8 bearing made with two axial grooves spaced 180° apart, instead of the three grooves used in previous tests, was operated for 38 hr and then removed from the tester for inspection because of signs of impending seizure. Score marks were found near the top and bottom of the bearing. It is believed that these may have been induced by a short period of tester operation without an applied load. This test will be repeated.

The MF-F centrifugal pump has accumulated 15,810 hr of continuous operation with molten salt fuel 30 ($NaF-ZrF_4-UF_4$) at 1200°F (or above), at a flow rate of 645 gpm and a speed of 2700 rpm. A similar pump equipped with a Fulton-Sylphon bellows-mounted seal has operated continuously for 13,150 hr with NaK at 1200°F, at a flow rate of 1200 gpm and a speed of 2800 rpm. (AEC Activity 4103.1B)

Nuclear Calculations. — Investigations were made of the nuclear performance over a 20-year lifetime of spherical, heterogeneous, graphite-moderated, one- and two-region, unreflected, molten-salt-fueled thorium-converter reactors. Various rates of continuous fuel processing and various blanket modifications were studied. The results are presented below for a core 14 ft in diameter, a total power of 600 Mw(th), 215 Mw(e), a plant factor of 0.8, a total fuel volume of 900 ft^3, and fuel channels 3.6 in. ID in an 8-in. lattice.

Reactor No.	Fuel	Blanket Thickness (in.)	Core Vessel	Processing Rate (ft^3/day)	Average Specific Power (Mw per kg of fuel)	Average Burnup Rate [g/Mwd(th)]
1	U^{235}	0	1-in. INOR-8	0	0.40	0.45
2	U^{235}	0	1-in. INOR-8	2.5	0.64	0.21
3	U^{235}	0	1-in. INOR-8	30	0.66	0.19
4	U^{235}	30	1/2-in. INOR-8	30	0.69	0.06
5	U^{235}	30	2-in. graphite	30	0.70	0.00
6	U^{233}	30	2-in. graphite	30	0.70	-0.07

It may be seen that the fifth reactor is self-sustaining with respect to fuel requirements, and the fourth, even though it has an INOR core vessel, is nearly so.

In the study of one-region, graphite-moderated, graphite-reflected, molten-salt-fueled thorium converters, regeneration ratios up to 0.84 have been obtained in cores measuring 15 ft high by 15 ft in diameter with reflectors 30 in. thick and initial fuel inventories of 1100 kg of U^{235} at power levels of 760 Mw(th).

A study of two-region, graphite-moderated, molten-salt-fueled thorium-breeder reactors has disclosed that doubling times of 20 years can be

obtained in reactors having 5-ft cores, 2-in.-thick graphite core vessels, and 30-in.-thick blankets, provided a specific power of 2 Mw/kg can be obtained by proper design of the external heat transfer system. The initial inventory was 60 kg of U^{233} for a power of 125 Mw(th). The fuel volume was 40 ft^3. (AEC Activity 4103.1B)

GAS-COOLED REACTOR PROJECT

The AEC has announced that the Experimental Gas-Cooled Reactor (EGCR) will be built at Oak Ridge. The EGCR will be both a power producer and an experimental facility designed primarily for providing test capability for future advanced gas-cooled reactor fuel elements. The EGCR will be an 85-Mw(th) reactor which will produce 25 Mw(e) from the 1050°F helium coolant. The reactor will incorporate, initially, eight through facilities for experimental irradiations. Four through tubes will be 9 in. ID; the other four will be 5 1/4 in. ID. The reactor will be provided with an adequate containment vessel. The experimental loops will be individually compartmentalized and separately contained. A task force consisting of Kaiser-Engineers, ACF Industries, and ORNL personnel was assembled in Oak Ridge during the period April 20–30 to complete preliminary specifications for the EGCR as it will be built on the Oak Ridge site. A preliminary presentation was made to the Advisory Committee on Reactor Safeguards on the EGCR on April 17 in Chicago.

At the request of the AEC, ORNL reviewed the Sanderson and Porter proposal for a Pebble Bed Reactor Experiment. Detailed comments on the proposal were submitted to the AEC. (AEC Activity 4110)

LITR Irradiation Experiments. — Two experimental assemblies (L-2 and L-15) were removed from the reactor during the month, and one assembly (L-17) was installed. Assembly L-17 contained 30% enriched UO_2 pellets with a theoretical density of 95%. A new type of center thermocouple was used in this assembly that consisted of an MgO-insulated Pt, Pt–10% Rh thermocouple in a tantalum sheath. The center temperature, which was initially 2410°F, has progressively decreased in steps of 25 to 80°F per day to 2000°F. (AEC Activity 4110)

ORR Irradiation Experiments. — Installation of the first eight GCPR-ORR capsules was completed. Irradiation of these capsules, which contain

UO_2 pellets, was started at 2:30 PM on April 11, when the ORR returned to 20-Mw power operation for reactor cycle X. At startup the thermal-cycling shutters were in the raised position, and the control gas in all eight capsules was 100% helium. When the reactor reached the 20-Mw level, the surface temperatures of the capsules varied from 700–800°F for the outermost capsules to approximately 900–1000°F for the innermost capsules. When conditions appeared to be steady, the procedures for injecting controlled mixtures of helium and nitrogen gas were started. Although the operation procedures assumed that purges with these gas mixtures could be accomplished in 10 to 15 min each, experience gained thus far indicates that purges of 2 to 3 hr are required to obtain the desired adjustment. (AEC Activity 4110)

Closed-Cycle Experiment. — All components of the closed-cycle experimental loop for testing reactor materials have been checked, including the safety circuit into the reactor control system, and the necessary repairs and adjustments completed. The test-sample temperature control system will be evaluated in actual operating conditions. (AEC Activity 4110)

Engineering Research. — An experimental program has been initiated to determine the transverse temperature gradients induced in the fuel rods of a seven-rod cluster by a known amount of tube bowing. It is planned to reassemble the heat transfer experiment in a larger-diameter Pyrex channel with the fuel rods held at correspondingly larger spacing. A set of electrically nonconducting rod spacers hold the individual rods at any desired spacing from the axial mid-point of the bundle. By using different sets of spacers, any desired degree of rod bowing can be simulated, and the surface temperature profiles can be determined for various Reynolds moduli and power densities. It is believed that this experiment will yield stability criteria for fuel rod bowing. (AEC Activity 4110)

Graphite and Coating Studies. — Two graphite specimens that were coated to reduce permeability to gaseous materials have been removed from the ORR after irradiation at water temperature for approximately 5×10^{20} nvt. One sample showed pits and breaks in the coating, as evidenced by the appearance of bubbles following immersion in CCl_4. Both samples decreased in density about 1%. The weight loss of the pitted sample was

65 mg; the other sample did not change in weight. Following preliminary examination the undamaged specimen was subjected to a continuing high-temperature oxidation test. Up to the present it has withstood 111 hr at 1100°C in air without weight change. (AEC Activity 4110)

PCTR Material Procurement. — The materials required for a critical experiment in the Physical Constants Test Reactor have been procured and shipped to Hanford, with the exception of the UO_2 pellets being fabricated by GE at San Jose. Oxide powder from reprocessed scrap is now being fabricated, with good yield of acceptable pellets. Current inspection reports indicate that the order for pellets will be completed on schedule. (AEC Activity 4110)

PACKAGE REACTOR DEVELOPMENT

Army Package Power Reactor (APPR-1). — During a recent shutdown of the APPR-1, the control-rod neutron absorber sections, as well as the fuel elements, were examined at the reactor site. Swelling at the tip of the boron-bearing absorber section was observed that was particularly serious in the central control-rod sections. These sections were replaced with Eu_2O_3—stainless steel absorbers. Examination of fuel elements revealed significant pitting and erratic corrosion films on the stainless steel cladding. At least one fuel element and two boron-iron absorber sections will be sent to ORNL for postirradiation examination. An unexpectedly large quantity of suspended solids accumulated in the primary water in the reactor vessel during the course of the examination of core components. The examinations were hampered by the solids until cleanup measures were instituted. Another unexpected development during this shutdown was the discovery of two broken reactor vessel studs, which had apparently failed during initial reactor operation.

The reactor has operated for two years with the initial core loading, and the accumulated burnup is 10 Mwyr. After further examination the reactor is to be put back into operation. (AEC Activity 4601)

GENERAL REACTOR RESEARCH

Basic Reactor Research. — A general-purpose Monte Carlo reactor code, the Oak Ridge Random Research Reactor Routine (the O5R), is being written

for the IBM-704 computer, and several phases have been completed. The neutron-history-producing routine, which can be used to calculate the neutron slowing-down density and flux as a function of energy and position, has been checked out for infinite and finite slab geometries and for spherical geometries. In addition, routines to compute the neutron age, the resonance escape probability, and the Fourier transform of the slowing-down distribution have been written. Thermal effects and a more general geometry routine will be added.

Two Monte Carlo codes have been written to compute the probability of neutron escape from solids. One code, Lump I, treats parallelepipeds, cylinders, spheres, and ellipsoids with sources distributed uniformly throughout the solid. Lump II treats disks with the sources distributed axially as $e^{-z/\lambda}$. The radial distribution is one which results from a collimator and is roughly Gaussian in shape. These codes have been used to calculate the fast-neutron effect for U^{233} in a manganese-bath experiment now being carried out to determine the value of η of U^{233}.

Additional routines have been written and debugged for the Corn Pone reactor code which will (1) allow the insertion of initial sources other than flat sources, (2) punch the converged source on paper tape if desired, and (3) reserve the converged source for possible use in the next case. The initial source options are: flat source, $\sin(ax + \delta)$, $[\sin(ar + \delta)]/r$, $e^{-(ax + \delta)}$, arbitrary source loaded into the machine from paper tape, and converged source from the previous case. (AEC Activity 4202)

Fuel Element Development. — The chemical determination of boron in stainless steel was shown to be accurate to within 6% of the intended analysis, with a precision of ±1.5% at a 95% confidence level. Analytical results on full-size fuel plates containing UO_2 and B_4C dispersed in stainless steel showed that the average boron loss in conventional fabrication by sintering in hydrogen and rolling into plate was 22.5%. However, the data were very scattered, the losses ranging from 0 to 50%.

A full-size ORR test element containing 20% enriched uranium as U_3O_8 dispersed in aluminum has been irradiated to an estimated 50% burnup of the U^{235} atoms. Visual examination of the element in the reactor tank showed no deleterious effect from this extensive irradiation. (AEC Activity 4203)

Power Reactor Fuel Reprocessing: Alternative Aqueous Process Studies. — Processing problems arising from the presence of lead as the bonding agent in BORAX-IV fuel were studied. In dissolving the BORAX-IV Al-Ni alloy cladding in nitric acid containing mercuric ion as a catalyst, the rates were very low when lead was present unless the mercury concentration was about 0.5 M. If not removed during decladding, lead seriously limited the uranium and thorium concentrations that could be attained by dissolution of the core in 13 M HNO_3—0.04 M NaF—0.04 M $Al(NO_3)_3$ because of the very low solubility of lead nitrate in concentrated nitric acid. (AEC Activity 4301)

Power Reactor Fuel Reprocessing: Zircex Process. — In the original Zircex process, the uranium in the hydrochlorination residue could never be completely dissolved in nitric acid. Losses ranged from 1% with the EBWR (low-enriched U^{235}) fuel to 6% with the STR (highly enriched U^{235}) fuels. In recent tests in which both the nitric acid—leached and unleached residues were chlorinated at 550 to 600°C with carbon tetrachloride, the uranium loss was less than 0.01% for both fuels. Secondary advantages of the use of carbon tetrachloride as a final chlorination agent are reduced corrosion and elimination of possible explosive reaction of uranium-zirconium alloys with nitric acid. (AEC Activity 4301)

Power Reactor Fuel Reprocessing: Zirflex Process. — In corrosion tests the addition of 0.05 M borate and 0.05 M silicate lowered the corrosion rates of Ni-o-nel and type 309 stainless steel in boiling 6 M NH_4F—1.0 M NH_4NO_3 from 12 to 6 and from 7.2 to 4.0 mils/month, respectively. (AEC Activity 4301)

Power Reactor Fuel Reprocessing: Solvent Studies. — In countercurrent batch extractions of synthetic Consolidated Edison fuel solution, first-cycle decontamination factors were 3.5×10^5 from gross gamma, 2.9×10^4 from ruthenium gamma, 1.8×10^5 from zirconium-niobium gamma, and 1.2×10^6 from total rare-earth beta; second-cycle factors were 7.6×10^4 from gross gamma, 3×10^4 from ruthenium gamma, 6.3×10^4 from zirconium-niobium gamma, and 1.4×10^5 from total rare-earth beta. Passage of the feed through silica gel at 50°C gave a decontamination factor from zirconium plus niobium of 37. The use of cadmium as a neutron poison will not

have to be restricted because of its extraction by 5% tributyl phosphate; the distribution coefficients were less than 0.001 under a wide variety of conditions.

In engineering-scale tests in 2-in.-dia glass-packed columns, the flooding rate for a solvent extraction flowsheet of the Darex-Redox type was 330 $gal \cdot hr^{-1} \cdot ft^{-2}$ when the feed contained 0.015 M Na_2SiO_3. Addition of 100 ppm of Mistron (an insoluble calcium-magnesium silicate powder) increased the column capacity by a factor of 2.5.

Radiolysis studies were made under subcontract by Stanford Research Institute on solvent consisting of 30 g of tributyl phosphate diluted to 100 cc with each of the following solutions: (1) 5 and 10 vol % isopropylbiphenyl in Amsco 125-82; (2) 5 and 10 vol % naphthalene in Amsco. Each solution was saturated with 2 N HNO_3 before irradiation. Neither aromatic compound appreciably affected the amount of gas or of monobutylphosphoric acid (MBPA) produced by a radiation dose of 200 or 400 whr/liter. The G values of 0.42 to 0.80 for MBPA (molecules of MBPA per 100 ev of energy absorbed) support previous conclusions that nitric acid increases the rate of formation of MBPA by hydrolysis of dibutylphosphoric acid, which is the principal acid formed by radiolysis of tributyl phosphate. (AEC Activity 4301)

Power Reactor Fuel Reprocessing: Mechanical Processing. — The replacement cost for saw blades for removing the inert end adapters of fuel assemblies, based on experimental wear ratios, was calculated as varying from 12¢ per element for the ORNL Gas-Cooled Reactor fuel to a maximum of $1.20 for the Consolidated Edison Reactor fuel.

In core leaching studies, unirradiated 0.4-in.-dia prototype fuel elements containing UO_2 and clad with stainless steel were sheared into 0.5-, 1-, 2-, and 2.75-in. lengths. The uranium was leached out completely by batch treatment with 10 M HNO_3 in 30, 45, 60, and 75 min, respectively. In all tests the ratio of the acid volume to the initial weight of UO_2 was 2.5 ml/g (150% excess). (AEC Activity 4301)

Power Reactor Fuel Reprocessing: Darex Process. — Results of tests on the removal of chloride from APPR-Darex dissolver product which were made under flowsheet conditions indicated that sparging with air assists

in chloride removal. Removal by sparging is complete in less than 6 hr, and the HNO_3 concentration required to remove chloride to 50 ppm, with or without sparging, is at least 9 M. Before the chloride was removed, the silica was apparently in a completely soluble form, and the solution was filtered through a 4-in. sand bed at a 6-in. solution head at an average rate of 51 $ml \cdot cm^{-2} \cdot min^{-1}$. During chloride removal, highly hydrated silica precipitated, and after volume adjustment, the average filtration rate of the chloride-free solution was 0.09 $ml \cdot cm^{-2} \cdot min^{-1}$. Metal filter elements (pore size, 20 μ) in which filter aid had not been used were satisfactorily regenerated with boiling 25% NaOH solution even when they were dried for four days before cleaning.

An over-all heat transfer coefficient of 450 to 500 $Btu \cdot hr^{-1} \cdot ft^{-2} \cdot (°F)^{-1}$ was measured in a titanium tank of the design proposed for the feed adjustment tank of the APPR pilot plant when the temperature difference between the steam and the boiling water was 40 to 55°F.

In corrosion tests, titanium tubes exposed to boiling 2 M HCl–5 M HNO_3 at 135°C for 2000 hr corroded at maximum rates of 1.5 to 2 mils/month. Stressed and welded specimens of types 304 ELC and 347 stainless steel in boiling 3 M HNO_3 containing 60 g/liter of dissolved stainless steel showed severe intergranular attack, though the maximum over-all rates were about 5 mils/month. This attack appears to be independent of the chloride concentration up to 800 ppm.

In tests on the decladding of prototype Consolidated Edison (stainless-steel-clad UO_2-ThO_2) fuel pins with 5 M HNO_3–2 M HCl, uranium losses were 0.3 to 0.4% in a 2-hr run.

In preliminary tests on titanium as a material of construction for a Darex-Thorex head-end process, welded titanium in boiling 13 M HNO_3–0.04 M HF was corroded at a maximum rate of 11.8 mils/month in a 24-hr exposure. In the presence of varying amounts of Al(III) and Th(IV), maximum rates were as low as 0.5 mil/month in 240-hr exposures. (AEC Activity 4301)

Power Reactor Fuel Reprocessing: Sulfex Process. — Two prototype Yankee fuel pins (stainless-steel-clad UO_2) irradiated to 500 and 775 Mwd/ton, respectively, were declad with 200% excess boiling 6 M H_2SO_4. The maximum uranium and plutonium losses to the decladding solution, determined in the presence of undissolved stainless steel end caps, were 0.012

and 0.05%, respectively. Passivation of the stainless steel cladding was more difficult to overcome than on autoclaved unirradiated specimens, but when the irradiated pins were wrapped with iron wire they dissolved in 4 hr in fresh 6 M H_2SO_4. This procedure is unacceptable in a process. Other work has indicated that dissolution of stainless steel would not start if the H_2SO_4 contained more than a few grams per liter of dissolved stainless steel or more than 0.01 M nitrate. Dissolution of the declad UO_2 in boiling 10 M HNO_3 was incomplete in 4 hr.

In tests with Consolidated Edison fuel, type 304L stainless steel cladding was passivated in boiling 6 M H_2SO_4 containing HNO_3 at a concentration higher than 0.015 M. In 3 M H_2SO_4–0.1 M HNO_3, passivation did not occur, but the dissolution rate was lower. In 3 and 6 M H_2SO_4 containing 0.2 M HNO_3, the passivation was easily broken by steel wool in contact with the specimen. In 15 tests on the decladding of unirradiated prototype Consolidated Edison fuel with 6 M H_2SO_4, the uranium loss to the solution varied from 0.02 to 0.24%, averaging 0.08%; with 80% of the samples the loss was 0.10% or less. The losses reported are probably not representative of losses from specification-grade fuel, since the uranium oxide in the prototype fuel analyzed approximately $UO_{2.10}$ and the tube had been seal-welded in air rather than in helium.

Continuing corrosion tests on the Sulfex-Thorex system indicate that long boiling of 13 M HNO_3–0.04 M HF in Ni-o-nel results in oxidation of chromium to Cr(VI) and in intergranular attack on the Ni-o-nel. Over-all rates after 1000 hr were as high as 6.2 mils/month, as compared with 2.1 mils/month when all the chromium was in the Cr(III) state. In 3.5 M H_2SO_4 containing 50 g/liter of dissolved stainless steel, Ni-o-nel was corroded at a maximum rate of 1.1 mils/month. (AEC Activity 4301)

Waste Disposal Research and Engineering: Disposal of High-Level Radioactive Wastes by Sintering. – Before its use as a base exchange material for sorbing cesium from neutralized Purex waste, Wyoming bentonite was heated to temperatures ranging from 20 to 800°C. The results show that the clay's ability to sorb cesium is not affected until a temperature is reached at which irreversible lattice collapse occurs; then the sorption of cesium is increased. However, if this critical temperature is exceeded,

the layer lattice structure is destroyed and the sorption of cesium is greatly reduced. The highest sorption was obtained with potassium-saturated bentonite heated to 600°C (0.3 mg of cesium per gram of clay); the greatest increase in sorption was obtained with calcium-saturated bentonite heated to 600°C (50% increase). These results are as expected from considerations of the structural concepts of cesium sorption by layer silicates, or irreversible ion exchange. (AEC Activity 4352)

Waste Disposal Research and Engineering: Regional Burial Ground Investigations. — Geologic and hydrologic studies, in cooperation with the U.S. Geological Survey, are being made in order to determine the suitability of the Bear Creek valley for use as a regional burial ground for solid low-level radioactive waste. Seven thousand feet of access roads, to permit entry of the churn and core drilling equipment, has been constructed. Stream gaging stations have been established at three sections on Bear Creek and at two tributary springs. (AEC Activity 4352)

Waste Disposal Research and Engineering: Mockup of Field-Scale Salt Experiment. — A one-tenth-scale mockup of the field-scale salt experiment was operated in a 1-m cube of Hutchinson salt for a period of 35 days. The experiment was carried out in a cavity 10 in. square by 13 in. deep which was filled with 7 M Purex solution saturated with salt and was provided with a titanium cover similar in design to that used in the field-scale experiment.

The cover, which exhibited no apparent corrosion, was effective in preventing the condensate from dissolving salt in the upper portion of the cavity. However, the cavity was deeply undercut by dissolution at the liquid-vapor interface because of inadequate mixing of the condensate and the brine. This suggests the need for modifying the design of the field-scale cover to allow for the return of condensate to the liquor at some point well below the liquid-vapor interface where thorough mixing will result.

Electrolytic heating of the liquor with graphite electrodes was successful, although a slight mechanical erosion of the graphite was evident at the liquid-vapor interface. There was no loss of liquid during the test, indicating again that salt is impervious. (AEC Activity 4352)

Waste Disposal Research and Engineering: Laboratory Studies of the Chemical Interaction of Waste and Salt. — Studies of the gases produced by the chemical interaction of acid Purex waste with sodium chloride show that NO_2Cl and Cl_2 are produced initially. Since both NO_2Cl and Cl_2 react with water and other materials, by-products due to decomposition and hydrolysis are also present in the gas stream. The maximum yields of Cl_2 and NO_2Cl were determined by sweeping the reaction vessel with air at 1 liter/min and collecting the gases in a cold trap at -80°C. Separation by fractional distillation and subsequent absorption and volumetric analysis showed that 5.0 ml of liquid chlorine and 1.2 ml of liquid NO_2Cl were produced by 250 ml of a mixture of waste and salt.

Chemical analysis of the cores taken from the experimental cavities in Hutchinson showed that extensive deposits of shale and anhydrite ($CaSO_4$) are present in this particular formation. Preliminary experiments indicate that some of the shale decomposes in acid Purex waste to form CO_2. (AEC Activity 4352)

Waste Disposal Research and Engineering: Design and Construction for Field-Scale Salt Experiment. — A cost-plus-fixed-fee subcontract has been signed with the Carey Salt Company, Hutchinson, Kansas, to allow the use of mined-out space for field-scale experiments on the disposal of liquid wastes in rock salt. The excavation of two cavities 7.5 ft square by 10 ft deep by the Carey Salt Company has been completed.

Equipment design is completed with the exception of the off-gas system. A cobalt-chromium-tungsten alloy will be used for the fabrication of the covers sealing the cavities.

Power supplies have been procured which are capable of supplying up to 3000 amp at 6 to 20 v and 1500 amp at 12 to 40 v. Design of the power supply installation, including instruments, controls, and bus bars, is completed.

All equipment will be prefabricated at ORNL and shipped to Hutchinson, Kansas, for final assembly and pretesting in the mine. (AEC Activity 4352)

Waste Disposal Research and Engineering: Corrosion Studies for Field-Scale Salt Disposal Experiment. — Haynes alloy No. 25 (Aeronautical Material Specification 5537A, a cobalt-chromium-tungsten alloy) and commercially pure titanium were the most corrosion-resistant of several materials tested

in hot (80°C) acid Purex waste saturated with sodium chloride. The corrosion rate at 80°C for coupons cut from butt-welded Haynes alloy No. 25 pipe was 0.001 in./year for unwelded samples and 0.003 in./year for specimens containing welds.

The corrosion rates for Haynes alloy No. 25 welded at ORNL were 0.012 in./year for Heliarc welding with backup gas, 0.013 in./year for Heliarc welding without backup gas, and 0.014 in./year for metallic arc welding. The samples were tested half submerged in the solution. An unwelded sample in the vapor phase at 110°C corroded at a rate of 0.007 in./year.

An unwelded titanium sample, half submerged in the solution at 80°C, corroded at a rate of 0.001 in./year. A welded sample in the vapor phase of the 80° solution also corroded at a rate of 0.001 in./year.

Teflon and normal impact unplasticized polyvinyl chloride were also tested. Teflon was not affected. Polyvinyl chloride sheet did not corrode, but a marked swelling was observed in the pipe samples. (AEC Activity 4352)

Waste Disposal Research and Engineering: Process Waste Water Treatment Plant. — From November 1958 through March 1959, the regular addition of 150 ppm of clay (Grundite) to the water, supplementing the treatment with lime and soda ash, resulted in an average removal of 84% of the Cs^{137}. During the same period, removals of radiostrontium averaged 85%, of total rare earths 89%, and of gross radioactivity 86%. (AEC Activity 4352)

Waste Disposal Research and Engineering: Maritime Hazards Study. — The water-borne contamination resulting from an accident to the NS "Savannah" during startup tests at Camden, New Jersey, is being evaluated on the basis of model tests performed at the Army Corps of Engineers Experiment Station, Vicksburg, Mississippi. One thousand curies released at dockside during average (12,350 cfs) and low (3000 cfs) river flow at Trenton would produce maximum concentrations at the Benjamin Franklin Bridge of 4.5×10^{-6} and 6.7×10^{-6} μc/ml, respectively. If the accident caused 1000 curies of radioactive material to be released during each tidal cycle, then the maximum concentration at the Benjamin Franklin Bridge for average flow and low flow would be 2×10^{-5} and 9.4×10^{-5} μc/ml, respectively. (AEC Activity 4352)

Waste Disposal Research and Engineering: Tank Hazards. — A study is being made of the hazards associated with storage in a hypothetical tank

farm at a site within the Ohio River valley of high-level liquid wastes from a central processing plant. A concurrent economic study specified the waste types and quantities, as well as the fuel cycle for the reactors serviced by the plant. Allowing for a 120-day cooling period before processing, it was found that storage tanks having a capacity of 4×10^5 gal would contain 4×10^9 curies at the end of a filling time of 5.2 years.

Geologic investigations of the site showed that the limestone shale bedrock is overlain by glacial till up to 50 ft in thickness. Movements of fission products through this soil have been calculated for two limiting cases. These calculations were based on a measured cationic exchange capacity of 12.3 meq per 100 g and selectivity coefficients of 85 for cesium and 10 for strontium.

Basic hydraulic data to evaluate the dispersion in the Ohio River are not available. The discharge into the Ohio River will be continuous, and simple dilution coefficients may be used. The average flow at the plant site is 1.05×10^5 cfs. At Vicksburg, Mississippi, there is a further dilution factor of only 5. (AEC Activity 4352)

Waste Treatment. — Conversion of high-level liquid wastes to a more readily disposable solid form by evaporation and calcination is under study. Phase compositions were determined as a function of temperature for the evaporation of TBP-25 [$Al(NO_3)_3$-HNO_3] wastes to 146°C. Little nitrate decomposition occurred below 146°C, but the final solution was very viscous. The condensate volume and weight were about 80 and 70%, respectively, of those of the original waste. The condensate contained about 55% of the total nitrate and 100% of the original free nitric acid. The density of the final solid product after calcination at 650°C was 0.5 g/cc, as compared with a density of 1.0 to 2.0 obtained with ferric oxide solids in earlier experiments.

The thermal conductivity and thermal diffusivity were determined as a function of temperature at the center of an 8-in.-dia cake of Darex waste calcined at about 900°C. The thermal conductivity increased from 0.042 to 0.11 $Btu \cdot hr^{-1} \cdot ft^{-1} \cdot (°F)^{-1}$ when the average temperature increased from 120 to 480°F. For the same temperature range the thermal diffusivity increased from 0.0037 to 0.0061 ft^2/hr. These results show that values assumed in previous theoretical predictions of temperature maxima in stored calcined waste were conservative. (AEC Activity 4352)

Radiation Damage: Reactor Flux Measurements. — Calorimetric measurements of the energy absorbed in nylon and in graphite were made in Hole 51N of the ORNL Graphite Reactor and compared with those in Hole 19. The data were analyzed to show the energy absorbed from fast neutrons in hydrogen and from gamma rays in carbon. In terms of these units, the ratio of fast neutrons to gamma rays for Hole 51N was half that for Hole 19. Since Hole 51N has a uranium converter and Hole 19 does not, it was concluded that moderating material preferentially attenuates the energy from gamma rays. The loss in energy of the neutrons in the moderating material is compensated by the larger scattering cross section of hydrogen at low energies. (AEC Activity 4540)

THERMONUCLEAR PROJECT

DCX Facility. — The DCX machine was dismantled and was reassembled with the following modifications:

1. The anode for the carbon arc was restored to the location at which it had previously been found to give an H_2^+ breakup efficiency of 20 to 30%. This should improve by a factor of perhaps 5 the rate of capture of H^+ ions and achieve a trapping rate of about 1 ma, which will be the highest yet obtained.
2. A diffusion pump was attached to the liner to assist in degassing during bakeout.
3. A modified collector for the undissociated H_2^+ beam was installed which is intended to lead ultimately to an effective removal of this beam and its collector as a source of gas.
4. A new accelerating tube was installed.
5. Sundry minor alterations were made.

These modifications should increase the chances for obtaining burnout. (AEC Activity 4851)

Carbon Arc Studies. — A hollow carbon arc from a 4 1/2-in.-dia cathode has been run successfully for rather short intervals. The arc swells to a diameter of 7 1/2 in. at the mid-plane of the mirror magnetic field. The current is about 2000 amp. An end window is arranged in the anode mount so that a person can look axially into the interior.

Mass spectrometric studies of the ions emerging from a carbon arc show the presence of an apparently dominating population of C^{3+} and C^{4+} ions halfway between electrodes in the long solenoid facility. The measurements are still in a qualitative stage.

Ultraviolet vacuum spectroscopic observations of the carbon arc in the long solenoid have been made as a function of arc current, up to 1500 amp. In the 1500-amp arc, the relative intensities of the 459-Å line (excitation potential, 33.3 ev) and the 977-Å line (12.6 ev) of C^{++} indicate that the mean electron temperature is lower than it is in the case of arcs running at lower currents. (AEC Activity 4851)

Vacuum Studies. — The performance characteristics of the 14-in. Leybold diffusion pump have been evaluated. Between 10^{-4} and 10^{-6} mm Hg, the pumping speeds for air and H_2 range between 1300 and 3000 liters/sec, depending upon heater power and type of baffling. Octoil S was used; the ultimate vacuum was 3–4 x 10^{-7} mm Hg; oil back-streaming was 0.054 cc/hr. (AEC Activity 4851)

PROGRAM 5000 — PHYSICAL RESEARCH

ISOTOPE PRODUCTION

Radioisotope and Stable Isotope Production. — Approximately 20,000 curies of Cs^{137} was produced from the Fission Product Pilot Plant, bringing the total to date to ~50,000 curies. During this period, 316 Cs^{137} sources, totaling 4200 curies, were fabricated, and 310 sources, totaling 552 curies, were shipped.

Approximately 16,000 curies of Pm^{147} is now in the process of being recovered by ion exchange.

Calutron separations of the isotopes of Rb, Bi-Tl, Hg, and Sr were processed during this period. The Rb and Bi-Tl collections were completed, while those of Hg and Sr are still in progress.

Calcium and strontium metals, rubidium bromide, rubidium chloride, and mercuric sulfide were prepared for calutron feed. The RbBr was prepared from RbCl by an ion exchange method.

The extraction of small quantities (200 ppm) of radiogenic Sr^{87} from lepidolite has been resumed. A base-exchange process, in which the finely ground ore is heated with sodium acetate, is being examined.

Eight new lots of gadolinium and ruthenium isotopes and one returned lot of Se^{74} were refined. New lots of Ca^{43}, Cu^{63}, $Gd^{156,160}$, and $Lu^{175,176}$, and three returned lots of iridium and potassium were prepared for inventory following chemical refinement and spectrographic and mass analyses. In addition, 28 samples were recovered from collector pockets, homogenized, partially refined, and sampled for mass analysis. Seventeen of these were samples of mercury, while the remainder comprised Ca, Mg, Rb, and Sr.

During this period, 24 conversions and 7 targets were made.

Radioisotope Research and Development. — A special Cs^{137} source consisting of a doubly sealed hollow cylinder, 1 in. by 4 in. ID and with a 0.33-in. annulus, was made. The capsule was charged with 100 curies of Cs^{137} and is now undergoing a shelf test.

The results of the irradiation of research samples of enriched Ni^{62} targets indicate that Ni^{63} with a specific activity of 1200 mc/g can be produced.

The waste from ~1.5 tons of 14-month-cooled and 3.0 tons of 6-month-cooled Savannah River fuel was processed in the Fission Product Pilot Plant. The presence of both Ni and Al in this Purex waste required the development of a new chemical procedure involving precipitation of the rare earths, Zr-Nb, Al, Ru, and Fe as phosphates, centrifugation and then leaching of the precipitate with caustic to remove the Al, and dissolution of the precipitate in HNO_3. The rare earths were then precipitated as oxalates. The filtrate from the phosphate precipitation contained Tc, Sr, and Cs.

The equipment for the separation of Y^{91} and Ce^{144} by batch solvent extraction has been installed in cell 9 of the Fission Product Pilot Plant.

The mixer-settler cascades have been installed on test stands in order to check mechanical performance and separation factors. Results of stable chemical and radioisotope tracer runs indicate that very pure promethium product with high recovery efficiency can be obtained from this equipment.

The highly purified promethium from Savannah River waste aged for 8 months and for 14 months was analyzed for gamma activities which could be attributed to Pm^{148} produced by the (n,γ) reaction with Pm^{147}. Seven gamma rays were detected, with energies ranging from 290 to 1000 kev. No gamma rays attributable to europium or other fission products were detected. The data indicate that the maximum amount of Pm^{148} gamma activity in 8-month-cooled material will be 3.6 mc of gamma per curie of Pm^{147} beta and that the gamma activity decays with approximately a 43-day half life.

A sample of sulfur was irradiated in the ORR in a 1/2-in.-dia aluminum can. The sulfur was then processed by dissolving the can in caustic, rinsing the sulfur in HNO_3, distilling the sulfur, and dissolving the P^{32} in HCl. The P^{32} solution met product specifications. Only trace amounts of Al were found by spectrographic analysis. (AEC Activity 5100)

Stable Isotope Development. — Receivers of improved design have been placed in use in mercury separations, and the in-run data acquired are being analyzed to determine the major sources of mercury contamination. (AEC Activity 5100)

PHYSICS

Classification of Low-Lying Nuclear Energy Levels. — The study of the decay of 8.3-day Ag^{106} (ORNL-2706) has been continued. Measurements of

the singles spectrum made three weeks apart showed that all the gamma rays decayed with the half life characteristic of Ag^{106}. The gamma spectrum in coincidence with the 1.21-Mev gamma ray was investigated; the intensities of the gamma rays in this spectrum and in the previously observed coincidence spectra were determined. The coincidence spectra revealed several new gamma rays; their energies (and intensities, which are normalized to a value of 100 for the 0.513-Mev gamma ray) are 0.32 (1), 0.701 (13), 0.74 (15), and 0.78 (15) Mev. The singles spectrum above 1.54 Mev was investigated; energies (and intensities) of gamma rays observed are 1.72 (2) and 1.84 (3) Mev. The upper limit of the intensity for any gamma ray with energy greater than 1.84 Mev is 0.2. Additional energy levels in Pd^{106} have been proposed at 1.36, 1.73, 2.29, and 2.34 Mev. (AEC Activity 5220)

The 86-Inch Cyclotron: Nuclear Physics. — Additional data on (p,d) reactions confirm the hypothesis that the proton picks up the neutron from a shell-model state. Angular distributions for the highest energy deuteron groups from Zr^{91} and Zr^{92} show the pickup to be of $d_{5/2}$ neutrons, and the energy difference between the Zr^{91} and Zr^{92} groups is believed to be the $d_{5/2}$ pairing energy. An angular distribution of deuterons from iron shows an $\ell = 1$ and an $\ell = 3$ group, with the $\ell = 1$ group at higher deuteron energy, consistent with the expected sequence of the $2p_{3/2}$ and $1f_{7/2}$ shells. It is now believed that an incoming proton excites most probably only one nucleon in the target, less probably two, still less probably three, and so on. This assumption accounts well for the observed structure in (p,d) and (p,t) energy spectra; the levels which are most strongly excited are those which result from the removal of one and two neutrons, respectively, leaving the rest of the nucleus undisturbed. This same mechanism accounts for some observed structure in inelastic proton scattering on lead isotopes and on bismuth and probably accounts for "anomalous" inelastic proton scattering.

A comparison was made of the elastic scattering of protons from Zn^{64} and from Ni^{64} in order to investigate a possible dependence of the nuclear potential on the parameter (N-Z)/A. No significant dependence on (N-Z)/A was discerned; the only differences in the differential scattering cross section could be accounted for by the difference in Coulomb potential for the two isotopes. (AEC Activity 5220)

The 86-Inch Cyclotron: Applied Physics. — The isotopes Mn^{52}, Ge^{68}, As^{73}, As^{74}, Sr^{85}, and $Y^{87,87m}$ were made in service irradiations for outside customers. Enriched Ge^{74} was used in one bombardment for As^{74} production to eliminate unwanted arsenic activities. Two flat-plate targets fabricated by soft-soldering 1/16-in. copper sheet into a 0.050-in.-deep depression in a copper backplate were tested to failure. The beam current at failure was more than 2000 μa (beam power 38 kw) in both tests. A solid copper target will easily withstand 2500 μa and, with the present beam distribution, would probably fail at currents in excess of 2850 μa. (AEC Activity 5220)

Heavy-Particle Physics. — Measurements of the elastic scattering of nitrogen from carbon have now been completed at three energies: 27.5, 23.5, and 21.5 Mev. Oscillating angular distributions are found at all energies. There is also some evidence that the structure behaves in a systematic way such that the positions of the minima and maxima appear to be proportional to the De Broglie wavelength. The angular region investigated is from 44 to 144° in the center-of-mass system.

Excitation functions were measured for two nitrogen-induced reactions in fluorine, $F^{19}(N^{14},p)P^{32}$ and $F^{19}(N^{14},2p)Si^{31}$. The ratio of the cross sections for these two reactions does not seem to depend on the incident energy and is the same above and below the Coulomb barrier.

Two counters are being investigated for high-resolution energy measurement of heavy ions: one is a germanium barrier counter, and the other a long proportional counter with re-entrant window. (AEC Activity 5220)

Electronuclear Machines. — Air-cored coils are being designed for Cyclotron Analogue II, in which electrons will be accelerated to 450 kev in an eight-sector spiral magnetic field. This is a model for the proposed 850-Mev fixed-frequency proton cyclotron. The latest coil design produces nearly constant axial focusing frequency from near the center of the machine to near the final radius; there the focusing frequency rises rapidly. Further work in the region of the outer orbits is required because of the sensitivity of the focusing properties of the field to small deviations from the desired spiral, $\theta = r^2$; deviations of only 0.2° change the focusing properties as much as 20%. Studies of nonequilibrium orbits in this latest field design indicate, as expected, a high degree of radial stability throughout the machine. (AEC Activity 5240)

Relativistic Isochronous Cyclotron. — Design criteria and review comments for the title I cyclotron building design were completed and forwarded to the architect-engineer, Catalytic Construction Company, which is expected to complete the title I work by May 4.

Quarter-scale r-f models were constructed and are being tested for frequency range and power level. The preliminary tests for a single-dee system indicate that both the frequency range and the power needed for the full-scale system will be approximately as calculated. An approximately one-fourth-scale model magnet for detailed field measurements and beam deflection studies was fabricated and is being assembled. Flowsheets for the vacuum system and water system are complete, and the instrumentation design for these systems is in progress. A schematic layout for the communication system within the building is being prepared. The yoke and pole pieces for the main cyclotron magnet were received from the United States Steel Corporation. The coil bobbins, which were constructed by Cleveland Diesel, were received. The main coils are now being wound in the shops at Y-12. Preparations are being made to install the full-scale magnet temporarily near Building 9204-3; many field measurements and tests can then be made before the final installation in the new ORIC building. (AEC Activity 5240)

Special Separations. — Recovery of U^{233} from a calutron liner and from salvage produced 79.38 g of metal (yield, 95%) from which two buttons were formed. The batch was fired at a high temperature because of the small quantity of UF_4 available.

Feed for a separation of Pu^{240} and Pu^{241}, scheduled to be made in early May, is being prepared from material received from Atomic Energy of Canada Ltd. Approximately 56 g of the available plutonium has been purified thus far.

The collection of Ca^{46} and Ca^{48} was concluded. In 68 runs, estimated ion currents equivalent to 0.582 g of Ca^{48}, 0.034 g of Ca^{46}, and about 475 g of the other calcium isotopes were collected. The over-all process efficiency for this series of runs was 18.3%, as compared with the previous highest process efficiency of 12.7%, or an increase of about 44%. A preliminary assay indicates a product purity of about 15% Ca^{46}. (AEC Activity 5250)

CHEMISTRY

Druhm Process. -- Direct reduction of UF_6 to metal with sodium at operating temperatures as high as 1200°C is being studied. Commercial-grade graphites of various thicknesses (0.25 to 1.0 in.) were not satisfactory as reactor liners; during operation they developed cracks which allowed the reaction products to go into the insulated lower section of the reactor. In one run 544 g of massive uranium (29% of theoretical) was recovered. A 3/4-in.-thick MgO reactor liner used in one reduction developed external surface cracks from thermal stress.

The intermediate reaction of UF_4 and sodium in the presence of NaF is being studied in the laboratory. Few data have been collected, because a satisfactory reactor liner has not been found. Lime-stabilized zirconia and alundum were unsuitable liner materials for the reduction of UF_4 to metal by sodium at 800°C. Zirconia cracked longitudinally and radially at the interface between the sodium and the molten fluoride; alundum disintegrated and was partly reduced to aluminum metal. High-density magnesia was surface-etched by partial reduction to magnesium metal by sodium vapor in the presence of UF_4 and NaF at 950°C. (AEC Activity 5310)

Chemical Engineering Research. -- A method is under development to measure point KCl concentrations in a flowing system for application in a study of corrosion protection by transpiration of inert liquids. A concentration cell with an active electrode surface 1 mm in diameter was constructed and tested with KCl concentrations from 0.1 to 10^{-5} M. The Nernst equation was found to apply over the entire range. An electrode installed in a fritted glass surface was used to measure the KCl concentration, while water was being transpired through the surface into a tank containing 0.1 M KCl that was being stirred. At transpiration rates of 2 cm/min the measured wall concentration was 0.001 M. (AEC Activity 5310)

Transuranics. -- The use of 0.5 M mono-2-ethylhexylphosphoric acid in xylene as a solvent for the separation of americium and curium from rare-earth fission products in a chloride system is being investigated. With 0.1 N HCl, the distribution coefficients for the rare earths and for americium were about 150. With increasing acid concentration the distribution coefficients decreased rapidly to a minimum of about 0.1 at 6 N HCl. Over

this acid range there was no appreciable separation of americium and rare earths; however, as the acid concentration was further increased, the rare-earth distribution coefficients increased while the americium distribution coefficient remained relatively constant. At 13.8 N HCl the separation factors for various rare-earth elements from americium were: Ce, 5.6; Nd, 14; Eu, 28; and Y, 500. (AEC Activity 5310)

Feed Materials Processing: Fluorox Process. -- In three experimental runs in which UF_4 was treated with oxygen in the 4-in. fluidized-bed reactor, the recovery of UF_6 ranged from 70 to 90% of theoretical. The bed temperatures were 775 to 850°C, and UF_4 feed rates varied from 1.2 to 1.6 kg/hr. The yield was about the same but the reaction rate was decreased when dry air was substituted for oxygen. The Inconel reactor was operated continuously for more than 50 hr, one of the longest runs during the experimental program. The apparent corrosion rate of the reactor varied from 0.14 to 0.44 in./year in the temperature range 800 to 825°C.

The reduction of UO_2F_2 with hydrogen at 700 to 850°C was investigated thermogravimetrically. Under all test conditions the rate-controlling factor was bed diffusion and the product was UO_2. In general, the reaction went to completion in fewer than 2 hours (see ORNL-2690).

Feed for the fluidized-bed reactor was satisfactorily prepared from powdered UF_4 in a Chilsonator roll compactor.

Two test runs were made with crude UF_4 prepared by reduction and hydrofluorination of ore concentrate by the Allied Chemical and Dye Corporation. A lower feed rate, 0.3 kg/hr, was necessary because the maximum operating temperature of 730°C (required because of the low melting point of the crude feed) resulted in a lower UF_6 production rate than for purified feed at 800 to 850°C. The UF_6 recovery was approximately 20% of theoretical. The very high corrosion rates in these runs (5.0 to 50 in./year) showed that Inconel reactors are not satisfactory with crude UF_4 feed. (AEC Activity 5310)

Feed Materials Processing: UO_2 Sinterability. -- A quick empirical method of estimating the surface area and predicting the sinterability of UO_2 was examined. The method is based on the fact that UO_2 particles in water act as weak acids. Two equivalence points exist, at pH 7 and pH 3, which can be recognized in a potentiometric titration. It is assumed that the number of

milliequivalents of acid required to lower the pH from 7 to 3 is a measure of the total surface area of the UO_2, which is one of several recognized variables affecting the sinterability (higher surface area yields denser compacts). Impurities, particularly anions, interfere; fluoride ion has an equivalence point at pH 5.2. Seven samples of ceramic-grade UO_2 were potentiometrically titrated with HCl to determine the number of milliequivalents required to lower the pH from 7 to 3. After correction for the effects of fluoride, it was found that the acid requirements increased from 0.8×10^{-2} to 10.7×10^{-2} meq as the specific surface area increased from 0.14 to 6.0 m^2/g and as the density of the sintered oxide pellets increased from 76 to 93% of theoretical.

When UO_4 was precipitated at 85°C from uranyl nitrate solutions in which the $UO_2(NO_3)_2:H_2O_2:HNO_3$ mole ratio was 1:2:0.2, high-density hexagonal needles of 1 to 4 μ average particle size were produced; at 50, 27.5, and 5°C, platelets of less than 1 μ average size were obtained. All precipitates filtered rapidly and were decontaminated from uranium daughters by a factor of 125. (AEC Activity 5310)

Equipment Decontamination. — The effectiveness of chromous sulfate as a reagent for decontamination (corrosion scale removal) for aqueous homogeneous reactor components was tested on a canned rotor pump removed from the HRT Chemical Plant. The over-all decontamination factor was 10 when a solution of chromous sulfate and sulfuric acid was passed through the pump at 85°C, followed by water and an acid solution of peroxide. This was sufficient to permit manual disassembly of the pump. Inspection indicated that the reagents had cleaned the surfaces with which they had come in contact and that most of the residual activity was in regions where the solution had not reached the metal surfaces.

The best method found for preparing chromous sulfate decontaminating solution was to treat acidified chromic sulfate solution with mossy zinc at 85°C. The corrosiveness of chromous sulfate solution was considerably decreased with no appreciable loss in effectiveness by decreasing the excess sulfuric acid concentration from the previously recommended 0.5 M to 0.1 M. The oxidation rate of the chromous ion, which is accelerated by bare stainless steel surfaces, was also decreased by a factor of 10.

In laboratory tests, Deoxidine 170 (inhibited phosphoric acid with added detergent) diluted 50% with water decontaminated a section of pipe removed from the HRT by a factor of 10^3. This reagent also gave the lowest corrosion rates obtained to date in a successful decontamination procedure: 0.7 mil/year on type 347 stainless steel and Zircaloy-2 and 42 mils/year on titanium. The Turco 4501 decontamination procedure, made with patented reagents, removed contamination and scale equally well but corroded titanium excessively. (AEC Activity 5310)

Nuclear Chemistry. — Promethium-146 has been reported to decay by emission of a 0.7-Mev beta ray with a half life of about one year. In preliminary work with Pm^{146} an electron-capture decay mode to a 1.2-Mev level in Nd^{146} was discovered. The ratio of electron captures to beta decays was found to depend on the method of production. The half life for electron capture decay appears to be about five years. (AEC Activity 5330)

High-Temperature and Structural Chemistry. — The program of determining the heats of fusion of the alkali halides and other salts was continued. Values obtained for NaI, LiBr, and KCl are 5.62, 4.21, and 6.26 ± 0.05 kcal/mole, respectively. On the basis of earlier measurements the activity of sodium iodide near its melting point in the system NaI-Na had appeared to be ideal, in contrast with the behavior of the systems NaCl-Na, NaBr-Na, and KX-K. With the new heat of fusion of NaI, simultaneous new measurements of the solubility of Na in liquid NaI and of the freezing temperatures of the monotectic liquid and of pure NaI showed a positive deviation from Raoult's law. This result appears to establish that diatomic metal molecules exist in the system NaI-Na, as well as in the other systems. (AEC Activity 5330)

Molten-Salt Thermodynamics. — The lowering of the vapor pressure of CsF at 1000°C by the addition of 20 mole % of various alkaline-earth fluorides has been measured as part of a study of the effect of the size and charge of the cation on the thermodynamic properties of fluoride melts. (The vapor pressure of CsF at 1000°C is 83 mm Hg.) The fluoride of the smallest cation, Mg^{++} (radius 0.78 Å), gives a lowering of 30 mm, that of Ca^{++} (radius 1.06 Å) 26 mm, and that of Ba^{++} (radius 1.43 Å) 18 mm, which was the expected order.

The effects of the alkaline-earth fluorides on the lowering of the freezing point of NaF are being measured. At 2 mole % CaF_2, the freezing-point

depression is equal to that calculated for an ideal solution. In solutions containing between 5 and 20 mole % of alkaline-earth fluoride, the freezing-point depressions are all sufficiently great to indicate that the activity coefficients of NaF in these solutions are less than unity. At equal concentrations (between 5 and 20 mole %) the freezing-point depression produced by MgF_2 is greater than that by CaF_2, and this, in turn, is greater than that due to BaF_2. (AEC Activity 5330)

Analytical Chemistry Research. — Purified tris-2-ethylhexylphosphine oxide (TEHPO) (99.9%), when utilized as a 0.1 M solution in cyclohexane, has an extraction coefficient for uranium (UO_2^{++}) of approximately 100 from 1 M HCl. The corresponding coefficient for thorium is only 0.00002. With less pure TEHPO (about 95%), the coefficient of extraction for thorium is greater by a factor of 100; however, little change in the extraction of uranium was noted. Enhancement in the extraction of thorium due to increasing the HCl concentration is slight; for example, in 5 M HCl the extraction coefficient is 0.001. The corresponding value for uranium is 22. This reagent is one of the best extractants yet discovered for the separation of uranium and thorium.

Procedures for the dissolution of various materials, prior to their determination by spectrochemical methods, have been established. Traces of boron (approx. 10 μg) present as borate in UO_3 can be retained during dissolution in open vessels if the original solvent contains mannitol; this measure prevents the volatilization of boron even in the presence of 1 mg of fluoride, as well as the loss of traces of boron during evaporation of acid borate solutions. It was found that In-Cu-Ag-Sn alloys can be dissolved anodically in dilute nitric acid. Alloys of rare-earth elements and niobium can be made soluble by fusion at 375°C in NH_4HSO_4, and the resulting material can be kept in solution in a mixture containing 5% oxalic acid and 10% nitric acid; this procedure avoids the use of hydrofluoric acid, which tends to form insoluble fluorides.

In the initial work on the spectrographic determination of trace elements in human tissue, it appears that the Stallwood jet source, used in a helium-oxygen atmosphere, possesses good stability.

METALLURGY

Fundamental Investigations of Radiation Damage in Solids. — The E band (7.6 ev) in the ultraviolet absorption spectra of fused silica and crystalline quartz has been observed after irradiations with Co^{60} gamma rays and with fast neutrons. No D band (7.1 ev) was observed, even with very light gamma irradiations, in the very high purity materials available. Impurities seem to play an important role in the development of coloration in the vacuum ultraviolet region, particularly with ionizing radiation and with short neutron bombardments. The E band was shown to be related to a previously described electron spin resonance line by using different materials with different irradiation conditions, followed by annealing and bleaching studies. In addition, an attempt was made to find a correlation between the E band and one or more of the other optical absorbing centers. The C and E bands develop and disappear quite independently of each other. Though electron spin resonance measurements indicate that the C band is due to a trapped electron and the E band to a hole trapped on an interstitial oxygen atom, the results show that there is no simple relation between them. (AEC Activity 5420)

PROGRAM 6000 — BIOLOGY AND MEDICINE

BIOPHYSICS

Waste Disposal Research: Evaluation of Soil Disposal for Intermediate-Level Liquid Radioactive Wastes. — Various materials investigated showed no marked specificity for the sorption of strontium with respect to calcium. Vermiculite, a clay mineral, exhibited the strongest affinity for strontium, the selectivity coefficient being 1.8 ± 0.2 with respect to calcium. Other materials investigated include illite, kaolinite, Wyoming bentonite, Arizona bentonite, limestone, rock phosphate, superphosphate, and Conasauga shale.

The removal of strontium from 1.5 N $NaNO_3$ by Conasauga shale is greatly enhanced by increasing the pH of the solution. This suggests that the removal of strontium is effected primarily by interstitial precipitation rather than by ion exchange. (AEC Activity 6440)

Waste Disposal Research: New ORNL Solid-Waste Burial Ground. — Forty-five auger wells were drilled in Melton Valley (ORNL-2590, p 27) for determining the character of the residual cover, the depth of the water table, and the chemical and radionuclide composition of the ground water. A descriptive geologic map and depth-to-water and water-table contour maps showing seasonal variations were prepared. A 6-in.-dia, 125-ft-deep core well was drilled for determining the occurrence and circulation of ground water at greater depth, the stratigraphy of the Conasauga shale in the area, and the mineralogical composition of the shale. To improve operations and monitoring, a new trench design was adopted. The bottom of the trench, covered with 6 in. of gravel, is sloped to an asphalt-lined sump at one end in which a 6-in. perforated casing is installed. Any liquid entering the trench before or after backfilling will flow through the gravel underdrain to the collecting sump, from which samples can be withdrawn and analyzed. (AEC Activity 6440)

Dosimetry Applications. — Measurements of the attenuation of fast neutrons and gamma rays by slabs of building materials, including those for Japanese-type residential structures, have been completed for the Ichiban Project [Health Phys. 1, 390 (1959)]. Measurements with good and poor slab geometry compared well with theoretical values.

Capsules for correcting the energy dependence of activated metaphosphate gamma dosimeters have been developed for most applications. Further encapsulation in Li^6 is necessary for gamma fields coexistent with thermal neutrons. These lithium capsules have been designed but have not been tested. The response of these dosimeters to fast neutrons has been demonstrated to be less than 1% of that due to gamma rays.

In connection with dosimetry for nuclear accidents, blood serum phantoms have been designed and have performed satisfactorily in preliminary comparisons of sodium activation with that of the sodium in burro blood serum. Current studies include incorporation of these phantoms in burro-sized phantoms for exposure to neutrons from critical assemblies.

Five hundred threshold detector units are being assembled for installations at which nuclear accidents could occur. The work is about 75% completed, but the boron containers will not be ready for approximately ten weeks. (AEC Activity 6600)

OPEN LITERATURE PUBLICATIONS

Biedenharn, L. C., and H. B. Willard, "Eigenphaseshifts in Terms of Wigner's Nuclear Reaction Theory," Proc. Phys. Soc. (London) 72, 874 (1958).

Bredig, M. A., "On the Polymorphism of Calcium Carbide," Z. anorg. u. allgem. Chem. 298, 255 (1959).

Brosi, A. R., B. H. Ketelle, H. C. Thomas, and R. J. Kerr, "Decay Schemes of Sm^{145} and Pm^{145}," Phys. Rev. 113, 239 (1959).

Curtis, C. E., and A. G. Tharp, "Ceramic Properties of Europium Oxide," J. Am. Ceram. Soc. 42(3), 151 (1959).

Fish, B. R., "Applications of an Analog Computer to Analysis of Distribution and Excretion Data," Health Physics 1, 276 (1958).

Gresky, A. T., "On Explanations of Electric and Magnetic Constants and Units," J. Franklin Inst. 267, 201 (1959).

Hurst, G. S., and T. E. Bortner, "Negative Ions of Oxygen," Radiation Research, Suppl. 1, 547 (1959).

Kelley, M. T., H. C. Jones, and D. J. Fisher, "Electronic Controlled-Potential Coulometric Titrator," Anal. Chem. 31, 488 (1959).

Koskela, U., L. G. Farrar, J. E. Attrill, and J. L. Mottern, "Analytical Service for the Homogeneous Reactor Test," Proceedings of the Seventh Hot Laboratories and Equipment Conference (1959 Nuclear Congress), No. 439.

Livingston, R., "The Potentialities and Limitations of the Paramagnetic Resonance Method in Radiation Research," Radiation Research, Suppl. 1, 463 (1959).

McGowan, F. K., and P. H. Stelson, "Coulomb Excitation of the Second 2^+ State in Even-Even Nuclei," pp 225, 765, Comptes rendus du congres international de physique Nucleaire, Dunod, Paris, 1959.

Melton, C. E., and P. S. Rudolph, "Mass Spectrum of Acetylene Produced by 5.1-Mev Alpha Particles," J. Chem. Phys. 30, 847 (1959).

Moeller, D. W., G. W. Leddicotte, and S. A. Reynolds, "Radionuclides in Reactor Cooling Water," J. Sanitary Eng. Div. (Proc. Am. Soc. Civil Engrs.) 85, No. SA2 (1959).

Ritchie, R. H., and G. S. Hurst, "Penetration of Weapons Radiation: Application to the Hiroshima-Nagasaki Studies," Health Physics 1, 390 (1959).

Shults, W. D., and P. F. Thomason, "Controlled-Potential Coulometric Determination of Copper and Uranium," Anal. Chem. 31, 492 (1959).

Wilkinson, M. K., J. W. Cable, E. O. Wollan, and W. C. Koehler, "Neutron Diffraction Investigation of the Magnetic Ordering in $FeBr_2$, $CoBr_2$, $FeCl_2$ and $CoCl_2$," Phys. Rev. 113, 497 (1959).

Wittels, M. C., "Evaluation of the ORNL Graphite Reactor," US/UK Graphite Conference Held at St. Giles Court, London, December 16–18, 1957, TID-7565, p 64.

FOREIGN VISITORS

Name	Affiliation	Citizen of
Andresen, E. G.	German Study Team	Germany
Asada, Y.	Japanese AEM Team	Japan
Baron, J. J.	Pechiney	France
Baumgarten, J.	German Study Team	Germany
Bodnarescu, M. V.	LURGI, Germany	Germany
Boehme, G.	Kernreaktor	Germany
Bresesti, M.	Comitato Nazionale Ricerche Nucleari	Italy
Brooks, M.	Austrian Atomic Energy Study Group	Austria
Cherel, G. H.	Saint Gobain Cie	France
Compton, D. O.	International Business Machines	Great Britain
Couture, J.	French Atomic Energy Commission	France
Dobrzynski, F. W.	German Study Team	Germany
Donnert, H. J.	U. S. Army Chemical Warfare Laboratory	Austria
Edeleanu, C.	Tube Investments Ltd.	Great Britain
Germond, P.	CEA, Saclay	France
Goto, M.	Japanese AEM Team	Japan
Harries, D. R.	United Kingdom Atomic Energy Authority	Great Britain
Henry, R.	AEC, France	France
Hessen, V. B.	Pye, Ltd.	Great Britain
Heymons, A. A.	German Study Team	Germany
Hintermayer, F. L.	Austrian Atomic Energy Study Group	Austria
Hovats, W.	Austrian Atomic Energy Study Group	Austria
Huisgen, R.	University of Munich	Germany
Ibe, L. D.	Philippines Reactor Project	Philippines
Imai, M.	Japanese Embassy	Japan
Ino, M.	Japanese AEM Team	Japan
Ishizuka, S.	Japanese AEM Team	Japan
Jacobs, G.	University of Illinois	Belgium
Jaegersberger, J. H.	Austrian Atomic Energy Study Group	Austria
Kadlez, K.	Austrian Atomic Energy Study Group	Austria
Kimbel, K. H.	German Study Team	Germany
Kimura, K.	Kyoto University	Japan
Kohrt, H. U.	LURGI	Germany
Kojima, K.	Osaka University	Japan
Krey, T. H.	AB Bofors	Sweden
Kubishiro, K.	Japanese AEM Team	Japan

Name	Affiliation	Citizen of
Lautenschlager, M.	German Embassy	Germany
Lindhe, J. S.	AB Atomenergi	Sweden
Lunden, A.	Iowa State College & Chalmers University of Technology	Sweden
Luoto, U. A.	Nuclear Power Assocs.	Finland
Mallikarjun, S.	University of Cincinnati	India
Manunapichu, K.	Thailand Atomic Energy	Thailand
Mas, R. R.	French AEC	France
May, H. L.	New Zealand Government	New Zealand
Mestre, E.	French AEC	France
Mialki, W.	German Study Team	Germany
Muenzer, H.	Austrian Atomic Energy Study Group	Austria
Muramatsu, M.	Japanese Embassy	Japan
Nagata, H.	Japanese AEM Team	Japan
Nagoya, K.	Japanese AEM Team	Japan
Neider, R.	German Study Team	Germany
Neuner, L.	Austrian Atomic Energy Study Group	Austria
Niezoldi, O.	German Study Team	Germany
Okamoto, S.	University of Kyoto	Japan
Park, Y. S.	University of Cincinnati	Korea
Peter, H.	German Study Team	Germany
Pomarola, J.	CEA	France
Radebold, R.	German Study Team	Germany
Renner, R.	Austrian Atomic Energy Study Group	Austria
Rose, R. S.	Institutt for Atomenergi	Norway
Rothaler, K.	Austrian Atomic Energy Study Group	Austria
Roy, J.	Editor of Social Welfare, le Monde, Paris	France
Serata, S.	University of Texas	Japan
Starfelt, N. G.	Atomic Energy	Sweden
Tajima, E.	Japan Red Cross	Japan
Takasu, N.	Japanese AEM Team	Japan
Taylor, J. B.	AWRE, Aldermaston	Great Britain
Tayler, R. J.	AERE, Harwell, England	Great Britain
Trouve, S.	French Atomic Energy Commission	France
Tunkelo, E.	Brookhaven National Laboratory	Finland
Uchikoga, T.	Japanese AEM Team	Japan
Uno, H.	Japanese Atomic Energy Research Institute	Japan
Valentin, A. M.	French Atomic Energy Commission	France
Vestergaard, R.	Atomic Energy	Sweden
Vetter, K.	Fritz-Haber-Institut	Germany
Watanabe, K.	Japanese AEM Team	Japan
Werner, O.	German Study Team	Germany

Name	Affiliation	Citizen of
Wheatley, B. M.	CERN, Geneva	Great Britain
Wheeler, D. J.	University of Illinois	Great Britain
White, G. D.	New Zealand Embassy	New Zealand
Wichmann, I.	German Embassy	Germany
Wobrowsky, M.	Austrian Atomic Energy Study Group	Austria
Wormser, G.	CEA	France
Wu, T. Y.	National Res. Council, Canada	China
Yasunaga, K.	Japanese AEM Team	Japan
Yoshida, T.	Japanese AEM Team	Japan

* * * * *

Reports previously issued in this series:

July 1958	ORNL-2578
August 1958	ORNL-2595
September 1958	ORNL-2623
October 1958	ORNL-2636
November 1958	ORNL-2645
December 1958	ORNL-2664
January 1959	ORNL-2691
February 1959	ORNL-2706
March 1959	ORNL-2721

OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

April 1959

Distribution

U.S. Atomic Energy Commission

Oak Ridge Operations Office

1-30. Research and Development Division

External

31.	Aeroprojects, Inc.	W. B. Tarpley
32-34.	Ames Laboratory: Iowa State College	F. H. Spedding
35-37.	Argonne National Laboratory	N. S. Hilberry
38.	Atomics International (N. American Aviation, Inc.)	E. E. Motta
39-40.	Battelle Memorial Institute	H. W. Russell
41-43.	Brookhaven National Laboratory	L. J. Haworth
44.	Bureau of Mines, Salt Lake City	J. B. Clemmer
45-46.	Chicago Operations Office	
47-52.	Hanford Atomic Products Operation	O. H. Greagor
53-54.	Idaho Chemical Processing Plant	R. L. Doan
55.	Kaiser Engineers	P. D. Bush
56-57.	Knolls Atomic Power Laboratory	K. R. Van Tassell
58-59.	Los Alamos Scientific Laboratory	N. E. Bradbury
60-61.	National Bureau of Standards	F. L. Mohler
62.	National Lead Company	C. K. McArthur
63.	Nuclear Metals, Inc.	A. R. Kaufmann
64-66.	Nuclear Products - Erco	E. A. Wick
67.	Union Carbide Corporation	R. W. McNamee
68.	Union Carbide Nuclear Company	C. E. Center
69.	Union Carbide Nuclear Company	J. L. Gabbard
70.	Union Carbide Nuclear Company	D. M. Lang
71.	Union Carbide Nuclear Company	J. P. Murray
72-73.	E. O. Lawrence Radiation Laboratory	D. Cooksey

Internal

74. K. A. Allen
75. E. E. Anderson
76. E. A. Bagley
77. P. S. Baker
78. C. R. Baldock
79. L. H. Barker
80. S. E. Beall
81. E. E. Beauchamp
82. P. R. Bell
83. D. S. Billington
84. R. E. Blanco
85. F. F. Blankenship
86. E. P. Blizard
87. A. L. Boch
88. E. G. Bohlmann
89. C. J. Borkowski
90. G. E. Boyd
91. M. A. Bredig

Distribution (Continued)

92. J. C. Bresee
93. R. B. Briggs
94. R. E. Brooksbank
95. K. B. Brown
96. F. R. Bruce
97. W. D. Burch
98. T. H. J. Burnett
99. A. D. Callihan
100. A. E. Cameron
101. R. A. Charpie
102. C. E. Clifford
103. T. E. Cole
104. C. F. Coleman
105. E. L. Compere
106. J. A. Cox
107. F. L. Culler
108. J. E. Cunningham
109. J. S. Drury
110. D. E. Ferguson
111. J. L. Fowler
112. J. H. Frye
113. W. F. Gauster
114. M. W. Gerrard
115. J. H. Gillette
116. H. E. Goeller
117. C. D. Goodman
118. A. T. Gresky
119. J. C. Griess
120. W. R. Grimes
121. P. A. Haas
122. C. S. Harrill
123. J. C. Hart
124. M. R. Hill
125. C. J. Hochanadel
126. A. Hollaender
127. R. W. Horton
128. F. T. Howard
129. A. S. Householder
130. H. H. Hubbell
131. G. S. Hurst
132. G. H. Jenks
133. R. W. Johnson
134. R. T. Jones
135. W. H. Jordan
136. P. R. Kasten
137. G. W. Keilholtz
138. C. P. Keim
139. M. T. Kelley
140. E. M. King
141. K. A. Kraus
142. Laboratory Shift Supervisor (D. Phillips)
143. E. Lamb
144. J. A. Lane
145. R. E. Leuze
146. W. H. Lewis
147. T. A. Lincoln
148. S. C. Lind
149. R. B. Lindauer
150. R. S. Livingston
151. J. T. Long
152. L. O. Love
153. R. N. Lyon
154. H. G. MacPherson
155. F. C. Maienschein
156. W. D. Manly
157. M. B. Marshall
158. J. A. Martin
159. H. F. McDuffie
160. J. R. McNally
161. A. J. Miller
162. E. C. Miller
163. K. Z. Morgan
164. E. J. Murphy
165. C. E. Normand
166. J. L. Need
167. G. W. Parker
168. M. E. Ramsey
169. P. M. Reyling
170. A. W. Riikola
171. L. P. Riordan
172. G. A. Ropp
173. A. F. Rupp
174. A. D. Ryon
175. H. C. Savage
176. H. W. Savage
177. A. W. Savolainen
178. H. E. Seagren
179. E. D. Shipley
180. A. Simon
181. O. Sisman
182. J. R. Sites
183. M. J. Skinner
184. A. H. Snell
185. R. W. Stoughton
186. H. Stringfield
187. E. G. Struxness
188. W. H Sullivan

Distribution (Continued)

189. C. D. Susano
190. J. A. Swartout
191. E. H. Taylor
192. C. D. Watson
193. B. S. Weaver
194. A. M. Weinberg
195. M. E. Whatley
196. J. C. White
197. C. E. Winters
198. R. E. Worsham
199. H. P. Yockey
200. W. Zobel
201. A. Zucker
202-203. Central Research Library
204. Health Physics Library
205. ORNL - Y-12 Technical Library, Document Reference Section
206. Reactor Experimental Engineering Library
207-398. Laboratory Records Department
399. Laboratory Records, ORNL R.C.