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STATUS AND PROGRESS REPORT

APRIL 1958

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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

April 1958

Date Issued: MAY 7 1958

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
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UNION CARBIDE CORPORATION
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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

April 1958

This Status and Progress Report summarizes somewhat more than one-half the activities of the Laboratory. Some of the topics are included every month, but many of them are reported on a bimonthly schedule. Program 4700 is covered in a separate report.

PROGRAM 2000 — SPECIAL NUCLEAR MATERIALS

Distribution and Excretion of Uranium in Man. — Five additional dogs have been made to inhale uranium metal fume to determine the initial retention patterns and to evaluate recent modifications of the equipment. Four of these animals were sacrificed at 1 hr, and the remaining animal was sacrificed at 24 hr postinhalation.

Six dogs have been injected intravenously in pairs with three dose levels of enriched uranyl nitrate: 10 μg, 100 μg, and 1 mg of U per kg of body weight. The animals will be serially sacrificed to determine the rate of elimination from the kidney as a function of dose. Radiochemical analysis of excreta has begun.

A study of the distribution of Mn^{54} in mice has begun. Fifty DBA mice are being exposed to continuous ad libitum ingestion of 0.1 μc of Mn^{54} (in the form of the chloride) per ml of water.

A feasibility study has been concluded on the determination of enriched uranium body burden by fissioning a small fraction of the uranium and counting the fast neutrons. It was seen that the fission body counter offers very little promise for use at this time as an analytical tool. (AEC Activity 2120)

Fluorox Process. — In the Fluorox process, UF_6 is produced from compounds of lesser fluoride content without the use of elemental fluorine. The process currently under study is the green-salt—oxygen reaction, $2UF_4 + O_2 \rightarrow UO_2F_2 + UF_6$. The reaction is carried out at temperatures of approximately 800°C in a fluidized bed of UO_2F_2. Green salt is continuously fed to the reactor, and UF_6 is continuously removed from the off-gas stream by means of a cold trap.

In two fluidized-bed runs at 800°C of 12 and 5.5 hr, respectively, the UF_6 recovery was 67.3 and 64.4% of theoretical. In the first run, more than 2 kg of UF_6 was isolated, and in the second run more than 1 kg was isolated. The 3-in.-dia reactor used for these two runs is being replaced by a 4-in.-dia reactor of improved design. (AEC Activity 2702)

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Chemical Plant Criticality Studies. — A program of critical experiments utilizing homogeneous, hydrogen-moderated uranium enriched to 2% in U^{235} has been inaugurated to determine quantities of interest in fundamental reactor physics and to establish bases for nuclear safety in production and fuel-element processing operations. Measurements are being made of the critical parameters and the nuclear characteristics of arrays of various shapes with and without a neutron reflector. From an extensive study of materials, a well-homogenized mixture of UF_4 and paraffin, which can be formed into blocks of relatively high density, was selected as the core material, and paraffin and a methacrylate plastic were selected as reflectors. The mixture for the first experiments contains 8 wt % paraffin, about the minimum quantity necessary to "wet" the densified UF_4, equivalent to an $H:U^{235}$ atomic ratio of 199. The uranium density in the blocks is 3.12 g/cc. In the course of the experiments with this material, several geometric shapes have been made critical with an effectively infinite paraffin (or plastic) reflector. (The current inventory, 25 kg of U^{235}, is insufficient to become critical in unreflected cylindrical geometry at this moderation.) Some typical values of critical dimensions and the corresponding U^{235} masses for parallelepipeds are as follows: 36.2 x 36.2 x 17.5 in., 23.1 kg; 29.2 x 29.2 x 21.7 in., 18.6 kg; and 28.2 x 28.2 x 23.0 in., 18.4 kg. Corresponding values for pseudocylinders are 36-in. dia x 18.7 in., 19.4 kg; 31-in. dia x 22.2 in., 17.4 kg; and 29-in. dia x 25.2 in., 16.9 kg. The present uncertainty in these data is ±5%. (AEC Activity 2724)

PROGRAM 3000 — WEAPONS

Special Separations. — Runs were made in order to collect two 45-mg samples of plutonium enriched in Pu^{240} to 99.5 and 99.8%. Collections were also made in order to accumulate an inventory of uranium enriched in U^{235} to 99%.

Following the Pu^{240} second-stage separation, 1 g of feed was recovered which represented 50% of the original 2-g charge. The material has been converted to $PuCl_3$ in preparation for a second run expected to yield approximately 40 mg of Pu^{240} above 99%.

Uranium-236 recycle has been completed through extraction to the oxide.

One 50-g $PuCl_3$ charge is being held for calutron separation.

Twelve shipments have been made from research pool material: two of U^{234}, two of U^{235}, one of U^{236}, four of U^{238}, and three of Pu^{240}. (AEC Activity 3610)

HOMOGENEOUS REACTOR PROJECT

Homogeneous Reactor Test. — On March 28, the HRT was charged with 5813 g of U^{235} and sufficient copper, as $CuSO_4$, to recombine all the radiolytic gas at 5 Mw. The reactor was made critical at 250°C and 1500 psi on March 30, and the power was raised to approximately 1 Mw an hour later. The power was raised to 2 Mw briefly on the following day; however, a loss of reactivity resulted, as evidenced by a 10°C loss in the average temperature of the core, and the power was again lowered to 1 Mw. Subsequent investigation of power level vs fuel temperature showed that stable operation could be achieved at core average temperatures above 260°C; the reactor power was accordingly raised stepwise over a four-day period, to a maximum estimated at 6.3 Mw (core and blanket) on April 4. This power was maintained for 26 min, at which time a sudden drop of the level in the fuel pressurizer was observed. When normal corrective measures failed to restore the level, further investigations were made which revealed that an interconnection existed between the fuel and blanket liquid circulating streams. The reactor was automatically diluted by control-circuit action upon loss of the pressurizer level. A high-pressure blanket sample withdrawn during the shutdown confirmed the presence of fuel in the blanket system.

After the core and blanket loops were rinsed with condensate, measurements were undertaken to locate the elevation of the leak in the core wall. Several checks indicated the leak to be in the vicinity of the welded peripheral joint between the 30-deg cone and 90-deg cone entrance sections near the bottom of the Zircaloy core vessel, and flow measurements indicated a probable open area of the order of 1/2 in.^{2}.

Examination of the core interior with periscopes failed to reveal the exact shape or location of the leak, since the point of failure was obscured from overhead observation by several of the core diffuser screens. External examination of the core through the blanket access port confirmed that the leak was in the hemisphere inaccessible to direct observation; attempts to view the back half of the core with a periscope and remote auxiliary mirror have not yet succeeded. (AEC Activity 4103.3)

Fuel Processing. — The behavior of iodine in the HRT was studied for the two power runs. Analysis of 18 samples taken during the last run showed 2 to 3% of the I^{131} activity to be expected if all the iodine that was produced remained in solution. Comparison of I^{133}/I^{131} ratios showed that some iodine which reached the low-pressure system was returned to the high-pressure system. Laboratory studies of iodine absorption by silvered stainless steel Yorkmesh showed that the rate of removal of iodine from an oxygen-steam-iodine mixture of fixed composition and flow rate was eight times faster at 150°C than at 110 to 120°C. Attack of silvered Yorkmesh by UO_2SO_4 solutions containing either 10 or 40 g of U per kg of H_2O and 50 mole % excess sulfuric acid at 100°C was negligible. At 110°C, however, silvered Yorkmesh was rapidly attacked by such solutions, so that the silver was loosened from the supporting steel surface after 24 to 48 hr and eventually dissolved in the solution.

Radiation from a Co^{60} source apparently accelerated the reaction. No such behavior has been observed in a gas phase containing oxygen and steam at a temperature of 120°C after several hundred hours. (AEC Activity 4103.1)

Uranyl Sulfate Blanket Processing. — Experiments in the P-1 loop to determine the behavior of plutonium in circulating 1.4 m UO_2SO_4 at 250°C under 350 to 400 psi of O_2 were completed. During the final run, 8.5 g of plutonium as dissolved plutonium sulfate was injected into circulating uranyl sulfate solution at the rate of about 1 g of plutonium every 8 hr. The results were similar to those previously obtained. Although enough plutonium had been added to give a concentration of 645 mg per kg of H_2O, only 26 mg per kg of H_2O remained in solution. Essentially all the rest of the plutonium was adsorbed on the walls of the stainless steel loop. The adsorption of plutonium was as much as 2.2 mg/cm^2, with an average of 1.1 mg/cm^2 for the entire loop.

The corrosion scale and adsorbed plutonium were completely removed from the loop by three successive passes of 0.4 M $CrSO_4$ in 0.7 M H_2SO_4. (AEC Activity 4103.1)

Thorium Oxide Slurry Development. — In development studies of thorium oxide preparation, oxide products (1- to 5-μ particle size) produced with a draft tube around the precipitator agitator were as uniform as those produced without its use and had as small an oversize fraction after firing at 1600°C. In thorium oxide pressing studies, a pressure of 20,000 psi broke up hard lumps of oxide, producing a material suitable for classification in the usual way; a pressure of 50,000 psi broke up microscopic agglomerates, dispersing the oxide into the individual particles of the oxalate relic structure.

Studies of the precipitation of thorium oxalate showed that digestion parameters, including the presence of organic amines, could be changed to give oxalate particles of varying sizes and shapes even though the mixing and initial precipitation steps were completed within seconds. Oxide products from carbon-reflected flame calcination had surface areas of 2.3 to 2.8 m^2/g and average diameters of 1.0 to 1.5 μ.

A thoria-urania sol submitted by the Davison Chemical Company for evaluation was irradiated for 145 hr at 280°C without agitation. When the irradiation bomb was opened, the sol was found to have broken into a dilute slurry of gray material and a hard yellow cake.

In out-of-pile studies of gas recombination, a slurry of thorium-uranium oxide with a U/Th ratio of 0.05 showed little catalytic activity with MoO_3 concentrations up to 0.3 m. Heating a slurry of thorium-uranium oxide (U/Th = 0.005) containing 1000 g of Th per kg of H_2O and 0.023 m MoO_3 at 200 to 285°C for 543 hr under an oxygen overpressure decreased the catalytic activity for gas recombination (in the absence of hydrogen activation) by 20%.

Studies to delineate the chemistry of molybdenum oxide in slurries of thorium oxide and thorium-uranium oxide indicate that, when molybdenum oxide

is autoclaved at 280°C in slurries of thorium oxide or thorium-uranium oxide, a part of the molybdenum oxide is converted to acid-soluble species and another part is either strongly adsorbed on the thoria or converted to a form insoluble in both base and acid. (AEC Activity 4103.1)

HRP Chemistry: Fuel Studies. — Phase-equilibrium data for the system $NiO-SO_3-H_2O$ have been obtained from 150 to 300°C and from 1 to 10^{-4} m SO_3. These data indicate that $NiSO_4 \cdot H_2O$ is the stable phase at high concentrations of SO_3 and at the higher temperatures, whereas several basic nickel sulfates appear to exist as stable species in lower concentration ranges.

A differential manometer apparatus has been constructed to accurately measure small vapor-pressure lowerings in aqueous systems at high temperatures. The vapor-pressure lowering of a 0.25 m Na_2SO_4 solution has been measured from 175 to 320°C in preliminary tests of the apparatus. This program is directed toward the measurement of vapor-pressure lowerings for $UO_2SO_4-H_2O$ solutions and aqueous systems in general.

Laboratory measurements were made of the rates of decomposition of peroxide in heavy-water solutions of uranyl sulfate over the temperature range 50 to 90°C. These studies were analogous to measurements made in light-water systems and reported by Silverman, Watson, and McDuffie (Ind. Eng. Chem. 48, 1238, 1956). The temperature dependence of the decomposition rate in a solution representing the fuel solution used in the HRT (containing 0.043 m uranyl sulfate, 0.001 m copper sulfate, and 0.022 m sulfuric acid in 99% D_2O) was found to be identical with that previously reported for a sample of inactive fuel solution from HRE-1. Direct comparison of the decomposition rates for 0.043 m uranyl sulfate solutions in light and heavy water at 60°C has been made; two rectifications of the heavy-water solvent have reduced the rate of decomposition of the heavy-water solutions from 32% higher than the rate in light water to less than 13% higher. Thus there do not seem to be any complications attributable to peroxide decomposition introduced by the substitution of heavy for light water in aqueous homogeneous reactor fuel solutions. (AEC Activity 4103.1)

HRP Analytical Chemistry. — A polarographic method has been applied to the simultaneous determination of mercury and copper in solutions of uranyl sulfate. By use of a supporting electrolyte of 0.9 M NaCl (pH 9) which is 0.25 M with respect to either of two complexing agents, ethylenediaminetetraacetate (EDTA) or nitrilotriacetate (NTA), the half-wave potentials of mercury and copper, respectively, are -0.1 and -0.5 v versus the S.C.E. The reduction waves of the two elements are readily resolved, and concentrations as low as 4 μg of either element per milliliter can be estimated. Iron(III) interferes with the determination of mercury; however, this interference can be eliminated by the use of a mixture of citrate and carbonate as a complexing agent. In this medium, the half-wave potential of mercury is -0.06 v, while that of iron is -0.55 v versus the S.C.E.

It has been found that microgram and milligram quantities of ruthenium as the metal or as RuO_2 on stainless steel and in oxide scale can be removed

by fusion and pyrolysis. In this method the sample in a platinum boat is overlaid with $NaBiO_3$, an oxidant and flux, and then pyrolyzed in the presence of moist oxygen at 800 to 1000°C. The ruthenium is volatilized as RuO_4, absorbed, and subsequently estimated spectrophotometrically. The RuO_2 can also be removed by dissolving it in warm 0.1 M HCl saturated with SO_2, after which the ruthenium metal is removed by pyrolysis and determined separately, and the ruthenium that was present as RuO_2 may be estimated by analyzing the acid solution in which it is dissolved. (AEC Activity 4103.1)

Slurry Component Development and Slurry Testing. — Run 200A-15, for studying the degradation characteristics of 1600°C fired thorium oxide, is still continuing. The oxide concentration was reduced after 400 hr to bring down the yield stress to the selected design maximum of 0.1 lb/ft^2. To study the effect of uranium oxide on the slurry yield stress, 1/2 wt % UO_3 was added after 1544 hr. The average mean particle size had decreased from 1.78 ± 0.16 μ to 1.63 ± 0.09 μ after 1350 hr of circulation. The generalized attack rate was 1.5 mils/year at a concentration of 1500 g of Th per kg of H_2O, decreasing to 0.6 mil/year at 700 g of Th per kg of H_2O.

Five runs in 100-gpm loops were made as a continuation of the study of the effects of additives on caking and sphere formation. The addition of 3000 ppm of H_2SO_4 to a slurry of 1600°C fired oxide produced a Newtonian fluid, but the attack rate was unacceptably high. Reducing the sulfate to 300 ppm lowered the attack rate to normal values. (AEC Activity 4103.1)

Slurry Blanket System Development. — The 300-SM blanket test facility was shut down for maintenance and alterations. Changes include replacement of the 1-1/2-in. inlet nozzles with the original 2-in. nozzles, modification of the blanket exit, installation of a continuous density bridge, addition of gamma-transmission survey facilities on the pressurizer, and alterations of the pressurizer to permit gas pressurization.

Operation of the 24-in. dump tank revealed that slurry density gradients of 10% or less could be maintained with 1 lb/hr of open steam and 16 lb/hr of auxiliary heating steam.

A contract was signed with Combustion Engineering for the fabrication of an experimental blanket pressure vessel for operation at 2000 psi in the 300-SM loop. The new vessel will permit three different flow patterns:

1. Entrance at the bottom, flow up between the core and a surrounding flow guide, and flow down outside the flow guide.

2. Entrance at the top and flow down and out the bottom (in this case, the flow guide is removed).

3. Entrance through two tangential nozzles 180 deg apart on the equator and exit at the top and bottom (flow guide removed).

(AEC Activity 4103.1)

Blanket Engineering Research. — It was found that thoria-water slurries have a yield stress proportional to the cube of the thoria concentration and inversely proportional to the square of the mean particle diameter of the thoria.

The 30-gpm loop for investigation of thoria-caking problems was completed and pressure-tested.

Studies were made of treatment methods which would make thoria spheres more resistant to degradation. Spheres were treated first with $Th(NO_3)_4$ and then with NH_4OH, followed by drying and then by calcination at 1200°C. The resulting material had greater resistance to degradation than the original material, as determined by tests with a Waring Blendor.

A capillary-tube viscometer was designed for use in the routine determination of shear-stress diagrams for thoria-water slurries in the temperature range 30 to 85°C.

The fluid-flow patterns in three small-scale models of some HRE-3 blanket configurations were studied. Visual and photographic techniques were used to evaluate the flow patterns. The results indicated a change from laminar to turbulent flow when the equivalent Reynolds number was between 1000 and 1500. (AEC Activity 4103.1)

Core and Blanket Vessel Development. — Fabrication of the 30-in. low-pressure blanket-vessel model is 50% complete. Preliminary flow experiments with a plastic model indicate that desirable flow patterns are generated in the annular volume between shroud and core.

Tests of a glass 6-in.-dia vertical dump tank indicate that slurry can be kept in uniform suspension by steam-heating. Boiling heat transfer coefficients were the same for both water and thorium oxide slurry.

Additional heat transfer measurements on a 4-ft-dia spherical re-entrant type of core vessel indicated a 15% variation in heat transfer coefficient with position. The effect of changes in fluid viscosity was in agreement with generally accepted correlations. (AEC Activity 4103.1)

Centrifugal Circulating Equipment. — The 400A-2 pump, with a revised motor cooling arrangement used by Westinghouse in their most recent pumps, completed satisfactorily a 1000-hr run in which fuel solution was circulated. Cast Zircaloy hydraulic parts were ordered for conversion of this pump to a 200-gpm slurry pump. Design of loop revisions in order that the rebuilt pump could be accomodated is 50% complete.

The 300A slurry loop was fabricated, and shakedown runs with water were started.

The detailed design of the 6000-gpm HRE-3 prototype fuel circulating pump was started. Agreement was reached with the manufacturer on a program for

developing and testing Belleville spring gaskets for the thermal barrier of this pump.

Agreement was reached with Franklin Institute for a one-year program in which they will determine the wear of bearings in water and in thorium oxide slurries at various loads. (AEC Activity 4103.1)

Miscellaneous Equipment. — Samples of bolting materials which may possess desirable strength, corrosion resistance, and nongalling properties were placed in various service environments. Hardened stainless steel thread inserts appear to be quite promising in permitting the use of bolting which otherwise galls.

Filtration and sedimentation were found to be promising methods of separating water from thorium oxide slurry. Continuous centrifuging did not appear to be satisfactory for high-concentration slurries.

The titanium-lined heat exchanger manufactured by the Pfaudler Company was received. The Crane Company experienced difficulties in welding the titanium-lined pipe and notified ORNL that they wished to withdraw from the contract because of these problems. The titanium 110AT pipe for application in a boiling uranyl sulfate pressurizer was received. (AEC Activity 4103.1)

Heat Removal Equipment. — The Griscom-Russell Company has started detailed design of test-model heat exchangers incorporating advanced features. One of the most promising ideas they will develop is the application of Inconel—stainless-steel composite tubing to a steam generator model.

Construction of a loop for studying slurry flow problems in heat exchangers is 90% complete.

The thermal-cycle test of five small tube bundles in boiler water containing chloride and oxygen was terminated after a maximum exposure of 5000 hr. The units will be subjected to metallographic examination.

The Babcock and Wilcox Company completed the fabrication of the HRT replacement heat exchanger and is preparing it for shipment. The REED thermal-cycle facility has been prepared for testing the exchanger. (AEC Activity 4103.1)

Gas Handling Equipment. — The fabrication of Inconel replacement parts for the high-pressure recombiner loop was completed.

The solubility of oxygen in light- and heavy-water solutions of uranyl sulfate—copper sulfate was measured at 250°C. The results were very close to the solubilities reported by Battelle for oxygen in water. The same equipment has been prepared for measuring the solubility of stoichiometric mixtures of hydrogen and oxygen during recombination. (AEC Activity 4103.1)

Fuel Systems Development. — The HRT mockup has operated for 1753 hr on 0.042 m UO_2SO_4 solution containing 0.005 to 0.035 m $CuSO_4$ and 0.021 to 0.025 m excess acid. The operation has been at 1400 psi and 280°C.

The generalized corrosion rate for the system with a titanium letdown heat exchanger has been 0.82 mil/year, as compared with 1.5 mils/year for comparable operating conditions except with a stainless steel letdown heat exchanger in the system.

During operation with a pressurizer purge flow of 10 gph, from 15 to 30 g of uranium per day was precipitated between the pressurizer and the main loop; the uranium redissolved when the purge flow rate was reduced to 3 gph. (AEC Activity 4103.1)

Feed Pump Development. — Feed-pump heads have been operated in endurance testing for 9300, 8800, 8800, 3800, and 2200 hr. Purge-pump heads have been operated for 7000, 7000, and 4800 hr. Testing of the double-diaphragm head is being continued at 2.7 gpm. Testing of the head containing 20-μ Poroloy screening continued successfully.

The remote-leg slurry feed pump has been operated successfully for 1400 hr and appears to be a reliable device. A method of continuously bleeding water from the top of the head to prevent gas-binding was developed. A second slurry diaphragm head, in which slurry will be allowed to contact the diaphragm, was fabricated.

Aluminum oxide check valves appear to pump slurry satisfactorily, with a wear rate of 45 mils/year; this compares with a wear rate of 19 to 33 mils/year for operation in water. Further tests of the many commercial grades of aluminum oxide are planned, as well as additional tests of other ceramics. (AEC Activity 4103.1)

Gaseous-Fission-Product Disposal. — Dynamic studies of the adsorption of krypton from a flowing stream of oxygen showed that the order of effectiveness of Linde molecular sieve materials after being dried is 5A > 13X > 10X > 4A. This is the same as the order of effectiveness observed in static equilibrium determinations. Of various methods tested for drying charcoal, the most effective was to pass dry oxygen through the bed at 100 to 150°C. Adsorption isotherms for xenon on various inorganic materials showed higher adsorption coefficients for xenon than for krypton at the pressures and temperatures studied. The increased capacity was 11.5 times for charcoal and molecular sieve 5A, and 4.5 times for one brand of silica gel, with other materials showing behavior intermediate between these two values. (AEC Activity 4103.1A)

MOLTEN-SALT REACTOR (MSR) PROGRAM

Molten-Salt Reactor: Design. — A conceptual design was completed of a homogeneous molten-salt reactor that would produce 260 Mw of electrical power. In advanced studies, results of machine calculations of two-region

molten-salt reactors fueled with U^{233} and with plutonium were analyzed. With U^{233} fuel, an 8-ft-dia core, 4 mole % ThF_4 in the fuel, and a fuel inventory of 556 kg, the initial regeneration ratio was 1.02. After 15 years of operation at 600 Mw, the regeneration ratio had decreased to 0.955, and the fuel inventory had increased to 840 kg if the fuel was reprocessed so that the content of Np^{235} and fission products equaled the annual production. For an unoptimized 600-Mw reactor with PuF_3 fuel in an 8-ft-dia core, the initial critical mass was 13.7 kg (0.013 mole % PuF_3), and the regeneration ratio in the ThF_4-containing blanket salt was 0.35. The initial fuel inventory was estimated to be 31.2 kg of Pu^{239}.

A preliminary design was completed which indicates that the helium purge gas of the off-gas system can be recirculated. The Kr^{85} can be separated from the purge gas and recovered by simple distillation. (AEC Activity 4103.1B)

Molten-Salt Reactor: Heat Transfer and Physical Properties. — The phosphorescent-particle flow-visualization technique was used to obtain qualitative information on flow in a glass model of a spherical core with straight-through flow (diametrically opposite entrance and exit). It was observed that the main flow channeled directly from the entrance to the exit, with the jet retaining closely the diameter of the inlet pipe. The remainder of the core was filled with a large, slowly rotating eddy. Velocities at the core wall were extremely low, and phosphor settling was detected along the wall in the lower hemisphere. Some flow instability was noted in the eddy region.

Preliminary values of heat capacity and viscosity were obtained for the fused mixture of $LiF-BeF_2-UF_4$ (62-37-1 mole %). Over the temperature range from 1000 to 1300°F, the viscosity was found to range downward from 19 to 7 centipoises. Between 900 and 1400°F the heat capacity of this mixture is 0.52 ± 0.02 $Btu \cdot lb^{-1} \cdot (^{\circ}F)^{-1}$. (AEC Activity 4103.1B)

Molten-Salt Reactor: Fuel Reprocessing. — Processing of the molten-salt reactor fuel ($LiF-BeF_2-UF_4-ThF_4$) by dissolution in aqueous HF appears to be practical. The solubility of LiF in aqueous HF decreases as the HF concentration decreases from 100 g/kg in anhydrous HF to 25 g/kg in 80 wt % HF. Beryllium fluoride is insoluble in anhydrous HF, but its solubility increases as the HF concentration decreases. The solubility of LiF is somewhat greater in the presence of BeF_2, and the BeF_2 solubility is considerably greater in the presence of LiF. In nearly anhydrous HF saturated with LiF, the solubility of BeF_2 is 25 to 30 g/kg, but it increases to about 70 g/kg in 80 wt % HF saturated with LiF. The $LiF-BeF_2$ solubility of greater than 150 g/kg obtained in some experiments with 90 wt % HF is higher than required for processing. In dissolutions of $LiF-BeF_2$ salt containing 0.2 mole % rare-earth fluorides and trace fission products, the rare earths dissolved in the order of only 0.0002 mole %. This makes possible greater decontamination from fission products than is required. (AEC Activity 4103.1B)

Molten-Salt Reactor: Experimental Engineering. — In the development of mechanical joints to facilitate remote maintenance, the freeze-flange joint was shown to be satisfactory for use on 1/2-in.-dia pipe, and therefore similar flanged joints are being fabricated for testing in a 4-in.-dia pipe system. Further tests showed cast-seal joints to be unsatisfactory. Additional tests of indented-seal joints are under way. Remote welding is being investigated with a standard Heliarc torch and a filler rod manipulated with a pair of Argonne model 8 manipulator arms. Welds that pass x-ray inspection have been made on butt-welded coupons. Thus far the operation has been viewed directly; indirect viewing by means of closed-circuit stereoscopic television is now being developed. The layout for the remote demonstration facility was completed.

The design of a heater-insulation unit for installation and removal by remote manipulation was completed. Units for testing on 4-in.-dia pipe are being fabricated. A facility for testing expansion joints with molten salts and liquid metals at high temperatures was designed, and components are being obtained.

A radiator is being fabricated for salt-to-air heat transfer coefficient measurements. Modifications of a facility for the required tests are under way.

Parametric studies of gas-cooled primary heat exchangers for a molten-salt reactor system were completed. Heat exchanger characteristics were evaluated for helium at 150 and 300 psig and for hydrogen at 300 psig.

At the beginning of March 1958, seven forced-circulation corrosion-testing loops were operating with molten salts as the circulated fluids. During March, two more loops began operation, bringing the total to nine. Of these nine loops, six were fabricated of Inconel and three were INOR-8.

On April 6, a faulty electrical circuit breaker opened and caused the failure of four of these loops. Three of the loops that failed were Inconel and one was INOR-8. These loops are now being repaired or rebuilt.

Twelve Inconel and eleven INOR-8 thermal-convection loops were operating at the beginning of March, and 31 loops (the capacity of the convection loop facility) are now operating. Seventeen of the 31 loops are constructed of Inconel, and 14 are constructed of INOR-8.

A 1000-hr test of a Dowtherm A—lubricated seal and hydrodynamic bearing operating in the bearing housing of a pump circulating $NaF-ZrF_4-UF_4$ (50-46-4 mole %) at 1200°F was satisfactorily terminated. Inspection revealed that the surfaces of the test journal bearing (aluminum bearing, Inconel journal) and the antifriction bearing were in good condition and satisfactory for extended operation. Leakage of Dowtherm A through the upper seal was too small to be measurable, and the lower-seal leakage averaged from 1 to 2 cm^3/day. The test bearing housing is being reassembled for further testing.

Operation of a NaK pump with a modified "split purge" gas flow seal (labyrinth) was terminated after 3378 hr when the maximum gas supply pressure (approximately 55 psig) was insufficient to maintain the required minimum pump tank pressure of 9 psig. Inspection revealed that the lower mechanical face seal had not been contaminated with NaK but that the gas passages in the labyrinth seal were plugged with a hard, black-to-gray-colored deposit containing Na, K, C, and possibly oxides of Na and K. (AEC Activity 4301.1B)

Molten-Salt Reactor: Chemistry. — The quaternary system $LiF-BeF_2-ThF_4-UF_4$ with 1 mole % ThF_4 and 1 to 4 mole % UF_4 is being studied. Liquidus temperatures are below 500°C if the BeF_2 concentration is from 35 to more than 60 mole %.

The solubility of CeF_3 in $NaF-BeF_2$ mixtures was determined as a function of temperature and solvent composition. As was the case when $LiF-BeF_2$ served as the solvent, the solubility of CeF_3 goes through a minimum at about 63 mole % NaF. Cerium fluoride is, however, less soluble by about a factor of 2 in $NaF-BeF_2$ than in $LiF-BeF_2$ mixtures. A mixture containing 63 mole % NaF dissolves about 0.10 mole % CeF_3 at 500°C.

Precipitation of zirconium oxide from a molten solution of ZrF_4 in LiF-KF eutectic by sparging with CO_2 was not successful. No observable reaction occurred. Preliminary attempts to precipitate similar oxides by the addition of SiO_2 (as reasonably coarse sand) were likewise unsuccessful; it is possible that the available surface area of the SiO_2 was too small to obtain reasonable reaction rates.

Metallographic examination of Inconel specimens exposed for 90 hr at 600°C to $KCl-ZnCl_2$ (29-71 mole %) and $LiCl-ZnCl_2$ (23-77 mole %) in capsules of silica showed virtually no evidence of attack. However, when pure chromium was exposed under similar conditions, the colorless melt became green and the capsule contained a globule of metal that was shown by x-ray diffraction to be nearly pure zinc. (AEC Activity 4301.1B)

CIVILIAN-POWER ADVANCED GAS-COOLED REACTOR (GCR)

Gas-Cooled Reactor: Design. — A brief study was completed of the economics of power production by small, high-pressure gas-cooled, graphite-moderated reactors. The diameter of the pressure vessel was taken to be 14 ft, and the thickness 7 in. The coolant considered was helium at 1350 psia. The cores studied were 9 ft in diameter and 8 to 14 ft in length. Fuel elements were 19-rod clusters of UO_2 rods in stainless steel tubes, with central holes up to one-half the rod diameter. Maximum heat fluxes upward of 600,000 $Btu \cdot hr^{-1} \cdot ft^{-2}$ and gross thermal power levels up to 450 Mw were obtained. In spite of excellent heat transfer characteristics, these systems appear to have higher power costs than the GCR-2; the lowest cost found was 11.3 mills/kwhr, compared with 10.4 mills/kwhr for GCR-2, which is significantly underrated with respect to power capacity. These costs result in part from the

smaller capacity of the small cores, and in part from the high cost of pressure vessels for high-pressure heat exchangers. In addition, certain engineering problems appear to become more acute with the high-pressure system, for example, blower drives, fuel recharging equipment, etc.; the incremental cost of these is difficult to evaluate.

The question of surface hot spots on rod-cluster fuel elements is being studied both analytically and experimentally. A program was prepared for solving the second-order partial differential equation whose solution is the two-dimensional temperature distribution in the fuel rod cluster and in the cooling channel. Results were obtained for clusters of seven 0.75-in. rods in channels 3.05, 3.25, and 3.45 in. in diameter. Maximum-to-minimum surface temperature differences are about 500, 400, and 900°F, respectively; maximum-to-average differences are about one-half those values. The location of the hot spot is next to the channel wall, for the smallest channel, and on the central rod for the other two cases. The principal conclusions to be reached from these results are that the rods must be located quite precisely within the cooling channel, and that a given fuel rod cluster will perform satisfactorily only in a very limited range of channel diameter.

A program for analyzing the effect of a large number of variables on heat transfer performance and on power cost was completed for the IBM-704 computer. Extensive results have been obtained for the gases H, He, CO_2, and N. (AEC Activity 4119)

Gas-Cooled Reactor: Metallurgy. — Investigations of the creep-buckling characteristics of thin-walled tubes were continued. A test of two 3/4-in.-dia, type 304 stainless steel specimens with 20-mil walls was discontinued at the end of 240 hr because of a crack in a weld of the pressure container. The test was conducted at 1675°F with an external pressure of 300 psi. Diametral clearances between the tube wall and the inserts were 10 and 20 mils. The specimen with the 20-mil diametral clearance buckled in a two-lobe mode, with one lobe running the full length of the specimen and with the other lobe along the central half of the specimen on the opposite side. The specimen with the 10-mil diametral clearance buckled in a one-lobe mode over about four-fifths the length of the specimen.

Similar specimens were tested for 500 hr at 1500°F under a pressure of 300 psi. Diametral clearances between the tube wall and the inserts were 2, 5, 10, and 20 mils. The specimens with the 10- and 20-mil diametral clearances buckled in a one-lobe mode. Even collapse of the tube wall around the inserts occurred in the specimens with the 2- and 5-mil diametral clearances. In a similar test at 1300°F, neither buckling nor collapsing of the specimens occurred, and the test will be continued for an additional 500 hr.

Two short pieces of 1-in.-dia, type 304 stainless steel tubing with 2- and 4-mil wall thicknesses, respectively, were inspected by means of radiography, ultrasound, and fluorescent penetrants. A few small pinholes were detected with penetrants in the 2-mil-thick tube; however, there were no

discontinuities detected which penetrated completely through the wall of either tube. Two small discontinuities were revealed by the ultrasonic inspection in the 4-mil-wall tube. Radiographs of the tubing indicated severely streaked or banded regions in the longitudinal direction. Careful radiographic and densitometric studies revealed the wall thickness to vary from 1.2 to 3 mils on the 2-mil tube, and from 3.6 to 6.1 mils on the 4-mil tube. (AEC Activity 4119)

Gas-Cooled Reactor: Radiation Effects. — Specimens of 15% enriched UO_2, which generate a power density of ~750 w/cm3, have been in the LITR since March 21, 1958. The central temperature of the UO_2 is maintained at 1770°F, and thus the surface temperature is approximately 1500°F. (AEC Activity 4119)

MARITIME SHIP REACTOR PROGRAM

Maritime Ship Reactor. — The Laboratory continued to furnish technical assistance to the Maritime Reactors Branch (AEC) in the program for developing nuclear-powered merchant ships. Additional drawings, specifications, and proposed development programs for the NS "Savannah" reactor system were reviewed, and recommendations were submitted to the Maritime Reactors Branch.

Brazing studies indicated that 0.02 to 0.03 g of Nicrobraz 50 is required to produce ferrule joints of excellent quality. Joints brazed at 1800, 1850, and 1900°F for 1/2 hr showed no significant undercutting or diffusion of phosphorus from the Nicrobraz; localized embrittlement of the fuel tubes does not occur, apparently, even with a 50°F overshoot from the reference brazing temperature.

A schematic flow diagram was prepared for the pressurized water loop which will be installed in the ORR. Engineering studies of piping layout, equipment arrangement, and instrumentation requirements are under way.

Methods for sea disposal of contaminated ion exchange resins are being investigated. Trace fission products were sorbed on a mixed resin bed which was then eluted with sea water. More than 95% of the sorbed activity was removed in 25 column volumes of sea water. Thus, loaded resin would be eluted very rapidly if disposed directly into the ocean.

Development of a health physics monitoring program for the NS "Savannah" was started. Preliminary recommendations for fixed facilities required on the ship were prepared.

Calculations were started for investigating methods of control-rod programing. The Cure program for the IBM-704 computer is being used. A test calculation gave a power profile which was in satisfactory agreement (within 15%) with experimental measurements. Additional calculations on the IBM-650 with a P3 cylindrical code yielded disadvantage factors for a variety of cladding thicknesses, can thicknesses, and metal-to-water ratios. (AEC Activity 4151)

PACKAGE REACTOR DEVELOPMENT

Army Package Power Reactor (APPR-1). — The Laboratory continued to furnish technical support to the Army Reactors Branch (AEC) on the 10-Mw pressurized-water reactor being test-operated at Fort Belvoir, Virginia, by Alco Products, Inc. Accumulated thermal power production is now approximately 5 Mw-years. The core life is presently estimated to be 13 Mw-years.

In order to minimize the transport of Co^{60} activity, a 304L-type stainless steel with reduced cobalt content is being obtained for fabrication of core II for the APPR-1. Analyses of constituents incorporated in the melt indicate a cobalt level of approximately 0.005%. The steel is now being rolled, and delivery is scheduled by mid-May.

The core II specifications call for approximately twice as much boron burnable poison as was incorporated into core I. Metallurgical experiments are under way to evaluate various materials as possible vehicles for incorporating the larger amount of boron in APPR-type fuel. Materials being investigated are BN, ZrB_2, B_4C, and prealloyed 304L stainless steel containing enriched B^{10}. (AEC Activity 4601)

Army Gas-Cooled Reactor Development. — A mockup of the in-pile section of the gas-cooled loop was installed in the ORR in order to study its nuclear characteristics. Inserting the in-pile section into the ORR decreased the reactivity of the reactor by $1.00. Flooding the loop section increased the reactor reactivity by about 3¢. Measurements are now in progress to determine the effect of inserting a fuel assembly into the loop.

Preliminary tests on the two compressors procured for the gas-cooled loop were completed. Six bids were received for the fabrication of the economizer and are now being reviewed.

Absorber foils for use in the critical experiment were developed and are being fabricated. The foils which have been completed are composed of 0.150 g of samarium as Sm_2O_3 in a dispersion of stainless steel rolled to a thickness of approximately 0.006 in.

Discussions between the Army Reactors Branch (AEC) and ORNL resulted in ARB relinquishing its support of the loop project, effective May 1, 1958. It is anticipated that the loop project will continue under the support of the civilian-power gas-cooled reactor project. (AEC Activity 4603)

GENERAL REACTOR RESEARCH

Basic Reactor Research. — An experiment was performed to measure the neutron diffusion parameters of beryllium at room temperature. Eleven blocks of beryllium were used, with bucklings ranging from 8×10^{-3} to 7.2×10^{-2} cm^{-2}. The results yielded a value of 4 ± 1 msec for t_a, the absorption time;

$(1.30 \pm 0.05) \times 10^5$ cm^2/sec for D, the diffusion constant; and 2 ± 1 cm^2 for C, the diffusion cooling constant. These values were based on measurements of the 11 decay constants which were good to within 10%. The value of D is to be compared with a value of 1.21×10^5 cm^2/sec calculated by R. C. Bahndari, and the value of C is to be compared with a value of 1.1 cm^2 calculated by M. Nelkin.

A surplus heat exchanger, which lowers the ~125°F temperature of the BSF pool water sometimes reached during high-power operations, was installed on a temporary basis, and it proved to be very effective.

The new Pool Critical Facility (PCF) at the BSF was taken critical for the first time on March 12, 1958. The initial loading consisted of four BSR-type control rod elements, each containing 70 g of U^{235}, and 18 ORR fuel elements, each containing about 168 g of U^{235} in 19 plates. The Operations Division had requested a 4 x 6 fuel element array, but the final loading contained only 22 elements for a total of 3307 g of U^{235} with an excess reactivity of approximately 1%. The extrapolated critical mass was 3200 g. A second loading with the highly enriched 168-g elements represented an attempt to repeat the original loading of the Geneva Conference Reactor, which used elements enriched to 20%. The critical mass of the PCF loading was about 2640 g of U^{235}, which should be compared with 3500 g required for the Geneva Conference Reactor. Loadings 3 and 4 were both 3 x 7 element arrays with two control rod elements. Moving one control rod one grid space on the long axis (into a lower flux) decreased the critical mass by about 70 g. The pool water temperature during these first four loadings was approximately 40°C. A series of five additional cores was assembled with 19-plate ORR elements containing 140 g of U^{235}. Three of these cores were essentially 5 x 5 arrays, each containing four control rods, and the critical masses varied between 2970 and 3220 g of U^{235}. Two 3 x 9 arrays were attempted, each with two control rods. One was extrapolated to a critical mass of 3975 g, while the other indicated a critical mass of about 3700 g of U^{235}. The pool water temperature during these five critical experiments ranged between 14 and 22°C. Other critical experiments were performed by ORSORT students. (AEC Activity 4202)

Fuel Element Development. — Difficulties were experienced in brazing fuel elements containing composite plates of 48% U—3% Si—bal Al alloy. Measurements of the plate spacings revealed that a number of the spacings deviated significantly beyond the permissible 10% tolerance. The problem may be related to the difference in coefficient of thermal expansion between the 48% U—3% Si—Al alloy and aluminum. Measurements of the coefficients of thermal expansion at 500°C revealed values of 24×10^{-6} per degree C for the highly concentrated fuel alloy and 33×10^{-6} per degree C for aluminum, a difference of 27%. Corrosion tests, in which the 48% U—3% Si—Al alloy was exposed to water at 90°C for six weeks, revealed some attack of the alloy, which was shown by the formation of blisters and cracks.

In the development of dispersions of U_3O_8 in aluminum, a cheaper grade of oxide has been obtained which places this type of dispersion in a more

favorable economic position. It is estimated that the cost of manufacturing this oxide will be $50 per kilogram, as compared with $300 per kilogram for the oxide prepared from $UO_3 \cdot H_2O$. Full-size composite plates have been successfully fabricated with this material, and tests have been initiated to compare its compatibility in aluminum at 600°C with that of the $UO_3 \cdot H_2O$ grade of U_3O_8. Corrosion tests of samples of U_3O_8-Al dispersions with the fuel exposed have not revealed any measurable attack after six weeks of testing in water at 60°C and at 90°C.

Work has been initiated to evaluate the uranium contamination on surfaces of fabricated composite aluminum plates. The initial data, obtained by neutron activation analysis of shavings taken at discrete depths into the cladding, indicated diffusion of uranium to the cladding surface after 3 hr at 600°C. Analyses of composites containing both the 18% U—Al and the 48% U—Al alloys indicated 6 to 8 ppm of uranium near the cladding surface after the heat treatment. Midway through the cladding thickness, values of 10 to 44 ppm were observed. Prior to heat treatment the analyses showed 0.15 ppm. Additional data are required in order to verify the diffusion trends statistically. (AEC Activity 4203)

Thorex Process. — In laboratory-scale studies the distribution coefficient for 0.5 M dibasic thorium nitrate to 42.5% tributyl phosphate (TBP) in Amsco was 0.1, as compared with 0.6 for thorium nitrate under the same conditions. The addition of an equivalent amount of nitrate as ammonium nitrate only partially compensated for the decrease in extractability.

The factors which determine the decomposition temperature of sulfite in Thorex solutions have not been determined. Synthetic solutions prepared from pilot plant thorium product completely decomposed bisulfite at 82°C, whereas bisulfite in solutions prepared from $Th(NO_3)_4$ crystals was not completely decomposed at 98°C. After the feeds were irradiated for 200 whr/liter, the bisulfite in each decomposed at 80°C. When thorium solutions containing bisulfite were irradiated, the ruthenium decontamination factor increased with time until the bisulfite was completely consumed; the factor then decreased rapidly.

The distribution coefficients of 0.5 M thorium and uranium in dibutyl butylphosphonate (DBBP) were 3.5 times greater than in TBP, whereas that of nitric acid was only 1.8 times larger. In countercurrent batch tests with DBBP and an all-acid flowsheet, extraction was good, but decontamination was lower than under standard Thorex conditions. The addition of phosphate, nitrite, or ferrous sulfamate to the scrub did not improve the decontamination.

The distribution factor of neptunium was sixfold greater than that of thorium when a feed containing 50 g of U per liter, 1.8 M $Al(NO_3)_3$, and 1.3 M HNO_3 with small amounts of thorium and neptunium was extracted with 15% TBP in Amsco. (AEC Activity 4301)

Power Reactor Fuel Reprocessing: Mechanical Processing. — Unirradiated Yankee Atomic and Consolidated Edison fuel rods were rolled with a Mackintosh-Hemphill rod-straightening machine to crush the oxide cores, and the ends of the fuel elements were removed by shearing. Neither the UO_2 core of the Yankee Atomic fuel nor the ThO_2-UO_2 core of the Consolidated Edison fuel was removed sufficiently to allow the process to be used for fuel reprocessing. (AEC Activity 4301)

Power Reactor Fuel Reprocessing: Darex Process. — In the Darex process, fuel elements containing uranium and stainless steel are dissolved in dilute aqua regia, the chloride is distilled off, and the uranium is recovered by solvent extraction. In two engineering-scale continuous dissolution-stripping runs of 32 and 47 hr with type 304 stainless steel and 5 M HNO_3—2 M HCl, in which azeotropic nitric acid vapor was used for stripping, the vapor/liquid ratio in the stripper was maintained at 0.95 to 0.98. Chloride was successfully stripped to a concentration of 50 to 100 ppm. Complete material balances for dissolved stainless steel, NO_3^-, Cl^-, and H_2O were fairly good. Chloride and nitrate losses to the off-gas were, respectively, 0.15 to 0.25 and 0.6 to 1.5 moles per mole of stainless steel dissolved.

A continuous Darex feed adjustment system with a liquid holdup of 3 liters was installed, and three runs were made at an aqueous feed rate of 130 ml/min. Steady-state operation was achieved 2 to 4 hr after startup. The feed was concentrated by a factor of 4 to 8; the product contained 5 to 7.5 N free HNO_3. The HNO_3 concentration of the distillate was 15.0 to 15.3 M.

In laboratory-scale batch dissolution and chloride boil-off tests with simulated Yankee Atomic fuel samples irradiated to 250 Mwd/ton, ruthenium was essentially nonvolatile but Zr-Nb and I_2 volatilized to a large extent. The activity balances for Zr-Nb were poor. Volatilized I_2 followed the HCl-HNO_3 condensate and was thus recycled to the dissolver. Batch equilibrations with solvent extraction feeds produced in the continuous feed adjustment equipment indicated satisfactory settling characteristics and low Cr(VI) extraction.

In previous laboratory tests with irradiated APPR specimens, about 0.012 M Cr(VI) was formed during the chloride removal step. Preliminary tests indicated that this concentration of Cr(VI) will not interfere with the reduction of plutonium to nonextractable Pu(III) by ferrous sulfamate for first-cycle solvent extraction. (AEC Activity 4301)

Power Reactor Fuel Reprocessing: Zircex Process. — In the Zircex process, uranium-zirconium fuel elements are hydrochlorinated to form volatile $ZrCl_4$, the residue being processed for uranium recovery. A 9-lb section of unirradiated STR subassemblies reacted completely with dry HCl gas in 24 hr at 500°C with an HCl utilization of 70%. Uranium losses to the sublimate were 0.07%; 6.3% of the chlorinated uranium in the reactor was insoluble in 10 N HNO_3. About 1.7% of the uranium was trapped in a glass-wool filter at 375°C preceding the desublimer. In a second similar run, 97.8% of the section was hydrochlorinated in 12 hr at a maximum temperature of 800°C and an HCl

utilization of 78%. The uranium loss to the sublimate was 0.02%, and the uranium chloride insoluble in HNO_3 was 1.8%. In neither run was the HCl pretreated to remove moisture. (AEC Activity 4301)

Power Reactor Fuel Reprocessing: Alcohol-Chloride Process. — Studies are being made of anhydrous ethanol-HCl and ethanol-$HgCl_2$ mixtures as dissolvents for zirconium-containing fuels according to the reactions

$$Zr + 4HCl \longrightarrow ZrCl_4 + 2H_2, \quad \Delta H_f^o = -142 \text{ kcal/mole}$$
$$Zr + 2HgCl_2 \longrightarrow ZrCl_4 + 2Hg, \quad \Delta H_f^o = -120 \text{ kcal/mole}$$

When the first reaction was carried out at 30°C in 10 M HCl and with low moisture content, the uranium losses were less than 1% except for a U-Nb alloy. At low HCl concentrations, insoluble ZrH_2 and ZrO_2 formed and the uranium losses were increased. This reaction must be kept at 30°C to prevent loss of HCl and formation of water by the reaction

$$HCl + C_2H_5OH \longrightarrow C_2H_5Cl + H_2O$$

With ethanol-$HgCl_2$ as dissolvent, the reaction was carried out at 80°C at higher dissolution rates than with HCl at 30°C. The amounts of off-gas were less than when HCl was used. The alcohol was removed after either reaction by adding water and distilling. Both the $ZrCl_4$ and the unreacted $HgCl_2$ are water-soluble. In the ethanol-$HgCl_2$ system, the mercury formed during the reaction was coalesced and the residual $HgCl_2$ was decomposed to Hg by cathodic electrolytic reduction. The aqueous phase was successfully decanted from the mercury phase. In a 2.5-hr electrolytic reduction, the amount of $HgCl_2$ in the aqueous phase was reduced to 1.22 mg of Hg per ml. The aqueous $ZrCl_4$-UCl_4 was then diluted with HNO_3 and the HCl was distilled off.

In four preliminary tests in which STR samples were dissolved in ethanol-$HgCl_2$, the uranium loss to the insoluble solids was 0.1 to 0.8% and the mercury loss was 0.05 to 0.16%. The uranium loss in a test with 10% Nb—90% U alloy was 0.068%.

In scouting corrosion tests, Hastelloy C appeared adequate as a material of construction for either ethanol-HCl or ethanol-$HgCl_2$. (AEC Activity 4301)

Power Reactor Fuel Reprocessing: Aqueous Dissolution. — Although 6 M NH_4F satisfactorily removed Zircaloy-2 cladding from oxide-cored fuels, the tin in the alloy did not dissolve. When nitric acid was added to dissolve the UO_2 core, SnO_2 formed and coated the UO_2 particles, resulting in up to 0.7% uranium loss to the insoluble residue. Addition of 1 M NH_4NO_3 or 0.1 M $CuCl_2$ to the NH_4F solubilized the tin in the decladding step and reduced this loss to 0.09%. The uranium loss to the decladding solution was 0.002%. Addition of NH_4Cl, NH_4I, or $(NH_4)_2C_2O_4$ to the NH_4F was ineffective in solubilizing the tin.

In tests on ion exchange recovery of uranium and plutonium from fluoride decladding wastes, Pu(III) sorbed well on Dowex 50; Pu(IV) did not. Decontamination factors of uranium and plutonium from zirconium were 74 and >300, respectively.

In tests with nonirradiated Consolidated Edison fuel, the stainless steel cladding was removed from the ThO_2-UO_2 cores by 4 M H_2SO_4 with maximum uranium and thorium losses of 0.01 and 0.10%, respectively. The ThO_2-UO_2 dissolved in 13 M HNO_3—0.04 M HF. About 95% of the mixed oxide dissolved in 6 to 8 hr; after this, the dissolution rate was very low. Contrary to an earlier assumption, the presence of stainless steel end caps did not inhibit the dissolution of the ThO_2-UO_2 in HNO_3-HF. Type 347 stainless steel dissolved in boiling 2 and 3 M H_2SO_4 at rates of 1.5 and 3.5 $mg \cdot min^{-1} \cdot cm^{-2}$, respectively. From Yankee Atomic fuel specimens irradiated to 250 Mwd/ton, 4 to 6 M H_2SO_4 removed the type 304 stainless steel cladding, with uranium and plutonium losses of 0.02 and 0.06%, respectively. The gamma activity of the off-gas was 0.01% of that of the decladding solution. The small uranium loss to the decladding solution appears to be caused by oxidation of the surface of the UO_2 pellets during or after irradiation to a form soluble in sulfuric acid. The declad UO_2 was successfully dissolved in 8 M HNO_3 to a uranium concentration of 288 g/liter.

Nonirradiated 93.5% U—5% Zr—1.5% Nb EBWR fuel samples (this fuel with cladding included is 20% Zr) dissolved in 13 M HNO_3—0.033 M HF with as little as 0.43% of the uranium precipitating at an $F^-/(Zr + Nb)$ ratio of 2.4. The precipitate was soluble in HF. In actual practice, an $F^-/(Zr + Nb)$ ratio of 4 must be maintained to prevent formation of an explosive surface deposit, with which an even lower uranium loss is expected. When fuel samples were dissolved in 6 M NH_4F, precipitates of NH_4UF_5 and $(NH_4)_3ZrF_7$ formed. In one run, 97.4% of the uranium was recovered by dissolving the $(NH_4)_3ZrF_7$ in cold water and centrifuging. (AEC Activity 4301)

Power Reactor Fuel Reprocessing: Materials Evaluation. — In corrosion tests, stressed and welded titanium specimens showed no sign of cracking or attack in Darex feed adjustment tank and flowing dissolver solutions. Carpenter stainless steel No. 20S was corroded at maximum rates of 2.7, 3.9, and 11.9 mils/month in 13 M HNO_3—0.05 M NaF containing 0.2 M Al^{3+}, 0.1 M Th^{4+}, and 0.2 M Th^{4+}, respectively.

Two vinyl coatings, Nukemite-33 and -40, were not damaged by gamma irradiation to 5×10^8 r except for a darkening in color. Glass rods and glassed mild steel, stainless steel, Hastelloy C, and Inconel were irradiated to 10^{10} r at the MTR gamma facility. The glass rods were darkened but not damaged. No effect on the glassed mild steel and Hastelloy C was observed, but the glass on the stainless steel and Inconel tended to flake off after the exposure. An epoxy-resin-base concrete irradiated to 10^{10} r swelled 10%, cracked, and partially disintegrated. (AEC Activity 4301)

Hermex Process. — In the Hermex process, uranium is decontaminated from fission products by dissolution in mercury. In two series of experiments on the solubility of uranium in mercury in the temperature range 25 to 356°C, the solubilities at the two limiting temperatures were 0.005 and 0.69 wt %, respectively. Values at intermediate temperatures from the first series of runs were very scattered; the analytical results of the second series are not yet available.

The solubility of ruthenium in mercury, also determined by filtration, varied from 0.86×10^{-7} wt % at 25°C to 3.5×10^{-7} wt % at 356°C. (AEC Activity 4303)

Fused Salt—Fluoride Volatility Process. — The laboratory volatility facility was decontaminated by a factor of >1000 by flushing with molten salt and aqueous solutions. A total of 890 g of uranium, irradiated to 600 Mwd/ton, had been processed through the Inconel system in 32 runs, involving 145 hr of hydrofluorination and 49 hr of fluorination. The contamination of the fluorinator and hydrofluorinator was not known accurately but was >10 r/hr. After decontamination, the level varied from 0 to 320 mr/hr. Most of the activity was in the loosely adherent scale and consisted largely of ruthenium in the hydrofluorinator and niobium in the fluorinator. X-ray analysis showed the scale to be mostly nickel metal, apparently deposited from molten salt as the result of galvanic action. The system was flushed several times with molten $NaF \cdot ZrF_4$, the decontamination effects of which were due mainly to dilution, and excess salt was then dissolved in boiling 0.5 M ammonium oxalate. Scale was dissolved in boiling 5% HNO_3—5% $Al(NO_3)_3$. Alkaline peroxide-tartrate (10% sodium hydroxide, 10% sodium tartrate, 2.5% hydrogen peroxide) was used as a decontaminant in the gas-phase region.

In corrosion tests, INOR-1 and INOR-8 were not attacked in 430 and 60 hr exposure, respectively, to $NaF-ZrF_4$ (50-50 mole %) through which HF was bubbled. Both specimens were covered with nickel crystals from corrosion in other parts of the equipment. Monel, after 1000 hr exposure, was attacked at a minimum rate of 24 mils/month.

In dissolution tests, mock STR fuel elements of Zircaloy-3 were completely dissolved with anhydrous HF while totally submerged in molten sodium fluoride—zirconium tetrafluoride. The HF rates were 11 to 45 lb/hr per square foot of dissolver cross section, and the chemical utilizations were 2 to 6%. The dissolution rates ranged from 1.0 to 1.3 $mg \cdot min^{-1} \cdot cm^{-2}$, based on the original total area of the fuel assemblies. When short zirconium elements were made parts of subassemblies composed almost entirely of copper and when the zirconium was kept above the level of the salt, the average dissolution rates were 2.5 to 7 $mg \cdot min^{-1} \cdot cm^{-2}$, and the maximum rates were 15 to 11 $mg \cdot min^{-1} \cdot cm^{-2}$. During the last run there was no salt in the reactor. (AEC Activity 4302)

Waste Treatment and Disposal. — A concentrated Hope-type radioactive waste was mixed with shale and limestone and heated to 850°C for 16 hr to measure the fixation of fission products by sintering. During the heating, 0.1 to 0.2% of the beta and gamma activity was evolved and was collected in caustic bubblers in the off-gas line. Leaching of the sintered product with distilled water removed ~1% of the original beta activity and 14% of the original gamma activity.

In preliminary tests on the removal from the off-gas of radioactive aerosols generated by the sintering of wastes, a dilute solution of fission products was refluxed, and an off-gas stream was passed through a train of caustic bubblers and micropore paper filters. Radioactive particles that penetrated the filters were less than 0.6 μ in diameter.

Electrolysis of a synthetic waste solution, 3.4 M $NaNO_3$—1.4 M NaOH, at 89°C decontaminated it from tracer Ru^{106} by a factor of 300. The cathode on which the ruthenium was plated was stainless steel. The emf was 6.1 v and the current 12 amp.

As little as 1 wt % fuller's earth carried 80 to 90% of the cesium from a synthetic sodium nitrate, aluminum nitrate, or stainless steel sulfate waste.

In studies on the disposal of waste from the processing of fuel from the maritime reactor, fission products sorbed on a mixed resin were eluted with sea water in three peaks: cesium at 5 column volumes of eluant, strontium at 14 column volumes, and total rare earths at 35 column volumes. (AEC Activity 4305)

PROGRAM 5000 — PHYSICAL RESEARCH

ISOTOPE PRODUCTION

Radioisotope Production. — Two hundred curies of Pm^{147} has been separated from the Redox waste.

Radioisotope Development. — A series of tests was run on the ORR scrubber to determine its scrubbing efficiency for I^{131}. A decontamination factor of 10^2 was obtained.

The revisions in the krypton processing equipment were completed.

Leak testing and tank calibration are in progress at the Fission Products Pilot Plant. (AEC Activity 5100)

Stable Isotope Separation. — The separation of the isotopes of vanadium (V^{50} and V^{51}) has been completed. Approximately 140 g of products was obtained; the V^{50} purity was 20 to 45%, corresponding to enhancements of 100 to 325.

Selenium isotope separation was completed and resulted in approximately 85 g of six isotopes. The production rate was approximately 17 mg per tank hour.

The first calutron separation of the eight isotopes of ytterbium was started in the 48-in.-radius machines, the recently designed M-16 ion source being used with a $YbCl_3$ charge. The average total ion current being received is 4.5 ma. The currents monitored to date are equivalent to approximately 40 g of separated material, and collection is on purified graphite. Mass analysis of one sample of Yb^{168} fixed the product purity at 34%, representing an enhancement of 365.

Preparations of VF_3, $VOCl_3$, SeO_2, $YbCl_3$, and AgCl were made as charge material.

Two batches of potassium, both enriched in K^{40} by neutron irradiation, were recovered from solution as KCl. These are to be used as charge material for the next collection of potassium isotopes. Samples of the batches showed a K^{40} content of 0.08 and 0.19%, as compared with 0.0119% in normal material.

Twelve new lots of isotopes were refined, including samples of C, Rb, Er, Se, and Sb. Two returned samples, Sb^{123} and Nd^{150}, were also refined. Fourteen new lots and three returned lots of isotopes were assayed and prepared for inventory.

In addition to the refinement program, six preliminary samples of ytterbium and six preliminary samples of vanadium were recovered, homogenized, and sampled in order to gain "in-run" data.

Fourteen coversions and eight special targets were made.

Stable Isotope Development. — The use of high-purity equipment and techniques was extended to the separations of vanadium and ytterbium. Whereas in previous vanadium separations the maximum V^{50} purity was 23%, the V^{50} purity achieved in this separation reached 45%.

Fabrication of components for a separator of the 190-deg radially inhomogeneous field type has begun. When completed, this machine should be used widely in the field of high-purity separations. (AEC Activity 5100)

PHYSICS

High Voltage Program. — An investigation of the (α,n) cross section in nuclei with $Z < 40$ has been begun to test the validity of the assumption that the gamma rays observed at a given alpha-particle energy E_α in Coulomb excitation studies result solely from Coulomb excitation. Thick-target yields of neutrons have been measured with the graphite sphere detector from threshold to $E_\alpha = 10$ Mev by using separated isotopes of Ni^{60}, Ni^{62}, Cu^{63}, Cu^{65}, Zn^{64}, Zn^{66}, Zn^{68}, and Zn^{70}. The cross sections near threshold are about 1 mb, and at 10 Mev the cross sections range between 80 and 200 mb.

Preliminary yield curves have been obtained for the reaction $C^{14}(p,n)N^{14}$. Data were taken at zero and at 90 deg to the direction of the incident beam. The bombarding proton energy overlapped previous data on this reaction between 1.9 and 3.4 Mev and extended the yield to 5.8 Mev. Agreement on the five resonances in the overlap region was good. In the region beyond 3.4-Mev bombarding proton energy, six resonances were found, corresponding to excited states in the N^{15} compound nucleus at 13.39, 13.60, 13.83, 14.13, 14.50, and 14.80 Mev. The experimental widths are 80, 25, 50, 100, 150, and 100 kev, respectively. (AEC Activity 5220)

The 86-Inch Cyclotron: Applied Physics. — The isotopes V^{48}, Ge^{68}, and As^{74} were produced in service irradiations for outside customers. A stack of zirconium foils was bombarded in the external beam for the Florida State University. One platinum and one lead target were bombarded for the Fysika Institutionen, Uppsala, Sweden. The isotopes F^{18}, Kr^{79}, and Tc^{95m} were produced for other ORNL divisions. (AEC Activity 5220)

Heavy-Particle Physics. — Continued progress is being made in the investigation of the odd-even effects in the level densities of light nuclei by means of a particle spectrometer which measures the energy distribution of protons, deuterons, tritons, and alpha particles from carbon, nitrogen, and oxygen targets. The differential cross section for the elastic scattering of nitrogen from beryllium was measured from 40 to 140 deg in the center-of-mass system. (AEC Activity 5220)

Electronuclear Machines. — Additional experimental studies with Analogue I indicate a much closer agreement between measured and calculated orbit properties than that previously reported. The radial focusing frequency measured by r-f knockout techniques agrees with the calculated value within 1%. A new measurement of the threshold voltage for acceleration to full energy (190 kev) gave 85 ev per turn, indicating that approximately 2200 turns are made by the electrons and that the magnetic field must be accurate on the average to about 1 part in 10^4. A new electron source for Analogue I was designed and tested. This source, which injects an accurately defined beam at an energy of 3 kev, will permit quantitative study of resonance transmission and beam deflection problems. Development of the new source was made necessary by the presence of large radial oscillation amplitudes in the beam caused by poor starting conditions in the unrealistic central region of the machine when filamentary or similar sources are used.

The Brookhaven Analog, a 10-Mev electron accelerator used in design studies for the 25-Gev proton synchrotron, was disassembled and transferred to ORNL. As program levels permit, it will be used to study proposed schemes for obtaining multiturn injection in strong-focusing synchrotrons. (AEC Activity 5240)

The 48-Inch Cyclotron. — Grid-type ion sources were tested in the temporary installation of the 48-Inch Cyclotron resonant system. Both horizontal- and vertical-grid sources were tested. In these tests the ion beam was colected on the first revolution in order to separate the effects of ion source output from the effects of ion transmission. The horizontal-grid source had six arc apertures 1/16 by 1 in. The ion output was 155 ma at a dee-to-dee voltage of 40 kv. The vertical-grid source had 12 apertures 3/32 by 1/2 in. and produced 155 ma at a dee-to-dee voltage of 30 kv.

Several magnetic field configurations for an AVF (azimuthally varying field) cyclotron were investigated. The parameters which were varied included the number of sectors (3 and 4), the width of the magnet gap, and the amount of spiral in the pole tips. The object of the investigation is to arrive at a magnet design which will provide the field shape required for focusing the particles throughout all their orbits, and at the same time will allow space for the structures that must be provided for acceleration. (AEC Activity 5240)

Mass Spectrometry and Related Techniques. — A mass spectrometric study has shown that nitrogen reacts with negative formate ions, $HCOO^-$, to form cyanide ions, CN^-, and neutral fragments. In a parallel investigation in which a spark coil was used, it was found that hydrogen cyanide was formed in an equimolar mixture of nitrogen and formic acid at 2 mm total pressure.

Tests to obtain optimum focus of the ion beam in the first stage of the two-stage mass spectrometer are essentially complete. Data obtained on the relative abundance of the isotopes of gadolinium are in good agreement with the accepted values. (AEC Activity 5250)

CHEMISTRY

Raw Materials Chemistry. — The extraction of VO^{++} from acidic sulfate solutions by di(2-ethylhexyl)phosphoric acid in kerosene was found to be well described over a considerable range of conditions by the reaction

$$2VO^{++}(aq) + 2(HX)_2(org) \rightleftharpoons (VO)_2X_4(org) + 4H^+(aq)$$

or

$$2VO^{++}(aq) + 2H_2O(aq) + 2(HX)_2(org) \rightleftharpoons [VO(OH)]_2X_4H_2(org) + 4H^+(aq)$$

where HX is the extractant, which is dimeric in solution. The corresponding equilibrium quotient is

$$Q = \frac{[V(IV)]_{org}[H^+]^4_{aq}}{[V(IV)]^2_{aq}[(HX)_2]^2_{org}} = \frac{E^o_a[H^+]^4_{aq}}{[V(IV)]_{aq}[(HX)_2]^2_{org}},$$

where E^o_a is the extraction coefficient. The formation of such a binuclear extraction complex is unique among the cations studied thus far.

In studies on the recovery of thorium from Amex carbonate strip solutions, thorium was more than 99.9% precipitated by acidification to pH 6 with sulfuric acid and digestion for 0.5 hr. The small amount of uranium present in the strip liquor was not precipitated with the thorium.

In preliminary tests, vanadium(V) was recovered from acid liquors by adsorption on a column of activated carbon. The vanadium was eluted effectively with dilute ammonium hydroxide. (AEC Activity 5310)

Chemical Engineering Research. — The (α,n) method of measuring interfacial area, in which a Po^{210} solution is placed in contact with a fluorocarbon, was calibrated with Teflon sheet and quiescent liquid fluorocarbon in systems of known neutron counting geometry. The method was then applied to a pulsed column. The average drop diameter, as determined by the (α,n) method, varied from 1.5 to 2.5 mm as the throughput was increased; the size range was confirmed visually. The pulse-plate hole size was 2.0 mm.

An alternative to the neutron-detection method is being evaluated. A liquid organic scintillator is contacted with a polonium solution, and a photomultiplier detector is used. The alpha activity necessary for a detectable level of interfacial reaction was estimated to be 10^{-6} times that for the (α,n) reaction.

Thermal diffusion in aqueous salt solutions is under investigation. A horizontal countercurrent thermal diffusion column, with a cellophane membrane separating the two chambers, was designed and constructed for the separation of metallic ions. Measurements of Soret coefficients for salt separation in one- and two-region membrane cells and in column tests established that the presence of the membrane reverses the sign of the Soret coefficient obtained by single-region measurement for the system cobalt sulfate—water. (AEC Activity 5310)

Extraction Reagent Performance. — The organic solvent found most resistant to irradiation was dibutyl phenylphosphonate, which showed a G value for gas production of 0.45 under irradiation of 1200 to 1500 whr/liter. Tributyl phosphate and dibutyl butylphosphonate showed G values of 2.44 and 1.80, respectively, under the same conditions. In a 50-50 Amsco solution, the three extraction reagents showed G values for gas formation of 1.04, 2.78, and 2.05, respectively. A radiation dose of 200 to 300 whr/liter had very little effect on the organic-phase settling time of dibutyl phenylphosphonate solutions but increased the settling times of the other two solvents 1.5- to 2-fold.

Irradiated solvent that was treated with 0.5 M NaOH—0.05 M NH_4OH in a single contact to remove radiolytic products retained less uranium when reused than did such solvent treated with 1 M Na_2CO_3. Uranium separation factors from the treated solvents were 2.8 and 3.5, respectively, as compared with 1 for virgin tributyl phosphate. (AEC Activity 5310)

Feed Materials Processing. — Uranium tetrachloride dissolved in diethyl carbonate was reduced to uranium by 3.8 M sodium amalgam with yields of ~15%. The presence of either ethyl alcohol or hydrogen chloride was necessary to keep the UCl_4 in solution in diethyl carbonate, and the presence of these materials prevented high reduction yields.

Mixing UF_6 with cold 3.8 M sodium amalgam, followed by transferring the mixture to a reactor at 356°C, reduced to metal 63% of the uranium transferred.

Excer-product UF_4 was reduced to uranium metal by sodium amalgam in 8% yield when prepared at 95°C and dehydrated at 25°C, but none of the UF_4 prepared or dehydrated at temperatures above 100°C was reduced.

Uranium solutions in C_7F_{16} containing up to 140 g/liter were prepared by bubbling UF_6 vapor through the liquid fluorocarbon at room temperature. The uranium metal yield was only 10% when this solution was contacted with sodium amalgam because of the reaction of the C_7F_{16} with the sodium amalgam.

In studies on the Druhm process (direct reduction of uranium hexafluoride to metal by sodium) a continuous system for feeding gaseous UF_6 and liquid sodium to a ceramic-lined reactor at 1200°C was designed and installed. In a preliminary test, 533 g of UF_6 was fed to the MgO-lined reactor containing 500 g of sodium at 615°C during a 17-min period. A thermocouple near the reaction zone rose to 870°C and remained at that value during the final 9 min of the run. A total of 317 g of massive metal was recovered from the reactor after the run. Two hundred fifty grams of the metal contained an average of 98% U, and 63 g contained 76% U. The recovery of uranium as consolidated metal was 82%. (AEC Activity 5310)

Molten Salt Thermodynamics. — In connection with a continuing study of thermodynamic activities in molten fluoride mixtures, the vapor pressure of the CsF-BeF_2 system from 850 to 1050°C has been measured. A minimum in the total pressure occurs at 30 mole % BeF_2. The results at 950°C are representative; here CsF has a vapor pressure of 43 mm Hg and BeF_2 33.5 mm, while the vapor pressure for the 30% BeF_2 mixture is 11 mm and that of the 80% BeF_2 mixture is 33 mm. Between 80% and 100% BeF_2 the total pressure is virtually independent of composition. (AEC Activity 5330)

Analytical Chemistry Research. — The apparatus which is now undergoing extensive tests for use in the determination of oxygen in metals and salts by a fluorination method was modified by the addition of multiple reaction chambers, permitting operation with a single gas-measuring unit. As a consequence of this modification in one of the two units, six determinations can

now be completed in an 8-hr day, as compared with four by means of two units of the original single-chamber design. In this method, oxygen is liberated by fluorinating the oxide with $KBrF_4$ at about 400°C, after which the oxygen is measured manometrically. Oxygen in yttrium metal, YF_3, BeF_2, and in a mixture of LiF, MgF_2, and YF_3 is now being determined on a test basis. Concentrations within the ranges of 0.05 to 2.5% in YF_3 and of 0.05 to 0.15% in yttrium metal have been determined. The results of replicate determinations on salts and on standards of pure oxides are reproducible to within 5% or less, while the precision is somewhat less in the analysis of metals. It has been observed that the results by the fluorination method are significantly lower than those obtained by the vacuum fusion method. The fluorination compound $KBrF_4$ appears to be suitable for the evaluation of the oxygen content of substances which contain acidic or amphoteric oxides. The applicability of the addition compound $BrF_2 \cdot SbF_6$, which is suitable for the fluorination of basic oxides, is being investigated.

METALLURGY

Ceramics Research. — Experiments in the waste research program involving the simulated Hope reactor waste solution included studies of low- and high-activity mixes, solid-state reactions, mechanism of retention, and flux additions.

Acid-leached and natural kaolins and various commercial grades of attapulgite were made into sinters with the simulated waste solution. The acid-leached material produced the greatest reduction in amount of dry material necessary to fix the radioactivity in the sinters. In the high-level activity tests, there was some retention with both acid-leached and natural kaolin. The leach factor (the ratio of initially introduced activity to the activity in the lixivium) was 2.2×10^5 for the best sinter mix.

In a preliminary study of the solid-state reactions between $Al_2O_3 \cdot 2SiO_2$ and radioisotope oxides SrO, CeO_2, and Cs_2O, x-ray and petrographic analyses showed a reaction with SrO at temperatures as low as 700°C.

The mechanism of retention was investigated by x-ray and petrographic analyses of a mixture of 100 g of shale, 30 g of soda ash, and 30 g of limestone with 250 ml of Hope solution fired at various temperatures. An initial glass formation at 650°C and the identification of mineral phases present were correlated with the radioactivity retention.

The effect of flux additions was studied with sinters made from 50 g of attapulgite and 250 ml of Hope solution plus one of the following: soda ash, limestone, lead monoxide, borax, sodium fluoride, magnesium fluoride, calcium fluoride, magnesium nitrate, sodium nitrate, and lithium carbonate. The lowest fusion point, 1200°C, was obtained with the sodium fluoride.

Experiments involving the simulated Purex waste solution consisted in tests of sinter mixes of alumina, silica, and feldspar; a comparison of sinter mixes of raw and calcined (600°C) talc and kaolin; and a study of leaching rates for these sinter mixes. Only a background count of radioactive material was leached from feldspar and alumina mixes sintered at 870°C. Calcining increased tenfold the retention of the fumed kaolin-Purex mix, but the other mixes were unaffected. The leaching rates over a 12-week period decreased in 2/3 and increased in 1/3 of the 35 mixes studied. Sinters of high initial retention showed the increase. (AEC Activity 5420)

Fundamental Physicometallurgical Research. — The effect of cycling between room temperature and 77°K on the transformations in pure cerium metal has been studied. Earlier work in this laboratory showed no apparent effect for eight cycles or less. A specimen which initially contained only the face-centered cubic structure contained approximately 10% face-centered cubic phase, 40% hexagonal phase, and 50% "collapsed" face-centered cubic phase when cooled to 77°K. After 105 cycles the specimen contained approximately 30% face-centered cubic phase and 70% hexagonal phase at room temperature. No "collapsed" cubic phase was found to form on the 106th cooling to 77°K. Thus it appears that a large number of cooling-heating cycles favors the face-centered cubic → hexagonal transformation and suppresses the cubic → cubic transformation.

A new alloy (Zr + 4% In) has been measured in the study of the low-temperature specific heats of group IVA elements with the following result:

$$C = 2.92 \times 10^4 \times T + 8.75 \times 10^2 \times T^3 \ (\text{ergs} \cdot \text{mole}^{-1} \cdot \text{deg}^{-1}).$$

This corresponds to an increase in the density of states at the Fermi surface upon alloying, a result which is in better agreement with a rigid band model of the alloy than with the Friedel model.

Diffusion rates of cadmium appear to be much smaller in alpha zirconium than in beta zirconium, on the basis of recent observations of vapor pressure equilibrium in this system. (AEC Activity 5420)

Microstresses in Metal Crystals. — An x-ray diffraction study of surface oxidation of copper, in conjunction with the surface chemistry group, has been begun. It has been found that the diffractometer, an instrument of high sensitivity which employs monochromatic radiation, is quite capable of detecting very thin monocrystalline surface oxide layers. To date, the diffraction patterns of films as thin as 190 Å have been measured. On the basis of the intensities obtained from such films, it is estimated that the thinnest detectable copper oxide film is probably less than 80 Å.

Oxide films grown on the 100 and 110 faces of copper single crystals have so far been studied. Interesting deviations from the diffraction pattern obtained from the bulk oxide have been observed. Work is in progress to interpret these changes quantitatively in terms of strain and particle size.

The crystal scintillation detector for the diffractometer has not yet been completed. The instrument continues to be operated with a Geiger counter. (AEC Activity 5420)

Fundamental Investigations of Radiation Damage in Solids. — Fast-neutron-irradiated n- and p-type polycrystalline samples of the semiconducting intermetallic compound InAs have been thermally annealed at temperatures corresponding to those that essentially remove radiation-produced defects from other similar materials. The p-type specimens that were converted to n-type by irradiation revert to p-type under anneal and indicate essentially total recovery to original conditions. The n-type material, on the other hand, does not thermally anneal at the expected temperatures. The production of additional donors in n-type InAs by fast-neutron irradiation and the absence of the expected anneal are as yet not understood.

The carrier concentration of high-purity n-type samples of germanium is being investigated as a function of temperature following Co^{60} gamma irradiation. Some evidence exists for a temperature-sensitive acceptor state that is only effective below 220°K. The position of this state appears almost exactly analogous to one previously observed following fast-neutron irradiation (0.2 ev below the conduction band) and is believed to be the first ionization of the irradiation-produced interstitial atom. Moderate-temperature (100°C) anneals indicate an almost total removal of this temperature-sensitive state. Such results suggest that a preferential migration of interstitial atoms has occurred. (AEC Activity 5430)

PROGRAM 6000 — BIOLOGY AND MEDICINE

BIOPHYSICS

Radiation Dosimetry Programs. — A beta-ray dosimeter of the general size and shape of a wrist watch has been developed to measure the radiation exposure to the hands of workers in the atomic field. The device is rugged, easily and cheaply fabricated, and records the exposure from soft beta radiation, which normally escapes detection, and from x-rays and gamma rays as well. General use of this dosimeter will enable a more careful assessment of beta-ray exposure to be made, which, in turn, will do much to reduce the hazard of beta burns on the hands.

The device is composed of a thick, perforated plastic cover surrounding a thin-walled container which defines the active air volume of the dosimeter. The cover shields the dosimeter to the extent that the dosimeter records about one-half the beta radiation exposure that would be read on a pocket chamber

Minometer. Because the cover does not attenuate x and gamma radiation, the dosimeter has a 100% response to these radiations. Thus in a mixed radiation field the dosimeter automatically takes into account the higher radiation level permitted by Handbook 59 for skin exposure. (AEC Activity 6140)

Internal Dose Studies. — The report of the internal dose committees of the National Committee on Radiation Protection (NCRP) and the International Commission on Radiological Protection (ICRP) has been revised. The basic principles and fundamental philosophy of radiation protection are unchanged. The most important of the several revisions is the reduction of maximum permissible concentration (MCP) of radionuclides in air, water, and food, so that the average RBE dose rate to the total body of the occupational worker will be one-third that formerly permitted. This reduction is intended to lessen the probability of chronic damage, for example, leukemia and premature aging.

The MPC values for bone-seeking radionuclides continue to be based on an accumulation in the bone that is estimated to deliver an RBE dose rate equal to that received from 0.1 μg of Ra^{226} and its daughter products. These values were reduced by 32% because recent data indicate that only 30% (rather than 55%) of the daughter products of Ra^{226} is retained in the body. The new MPC values correspond to an RBE dose rate of 0.56 rem per week to the bone, 0.6 rem per week to the thyroid or skin, 0.1 rem per week to the total body or gonads, and 0.3 rem per week to all other organs of the body.

A major objective in the revision of the ICRP Handbook has been to obtain reliable values for the distribution of radioelements in the human body following many years of exposure. One of the best sources of such data is the normal distribution of stable isotopes of these elements in the human body. For the past six years a study of human tissue has been conducted by the Oak Ridge National Laboratory in cooperation with the University of Tennessee. This study includes a spectrographic analysis for 37 elements in 35 body organs, from over 300 autopsies that were supplied from 15 countries. For elements such as Ca, Fe, Zn, Cu, Mn, etc., which are known to have a role in the body economy, the geographical variation is not large but reflects the state of nutrition of the population. For other elements such as Sr, Cd, Pb, Al, and Cr, which may compete with necessary elements in their normal roles, the geographical variations are striking. Some elements such as Cu and Zn in liver, Al in lung, and Cd in kidney show marked variation with age of subject. Such data are useful not only in calculating the MPC values for the standard man but may serve also as a guide in making refinements to the MPC values when applying them to individuals of a given age, weight, eating habits, etc.

MPC values are given for 86 radionuclides in the 1955 ICRP Handbook and for about 250 radionuclides in the 1958 edition. Also, MPC values are given for insoluble as well as soluble radioactive materials and for several critical body organs, resulting in a sevenfold increase in the number of MPC values. In a few cases (were sufficient data are available) the MPC values are based on a power function rather than on an exponential function of the time of exposure. (AEC Activity 6440)

PROGRAM 8000 — WORK FOR OTHERS

Professional Health Physics Training Program. — An eight-week course in health physics fundamentals has been completed. Ten participants enrolled in this course, which was intended to fill an immediate need in supplying personnel with some understanding of the problems involved in health physics. (AEC Activity 8780)

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OAK RIDGE NATIONAL LABORATORY

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