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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

DECEMBER 1954

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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

December, 1954

F. T. Howard

and

W. H Sullivan

Date Issued: JAN 7 1955

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December, 1954

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STATUS AND PROGRESS REPORT

December, 1954

This status and progress report presents material on approximately one-half the Laboratory's program, which is covered, with some exceptions, on a bimonthly schedule.

PROGRAM 2000 - SOURCE AND FISSIONABLE MATERIALS

Distribution and Excretion of Uranium in Man - The cooperative study of the metabolism of uranium in humans has continued at the Massachusetts General Hospital. The Hospital needs the data for the evaluation of neutron capture therapy to be employed in the treatment of brain tumor, where U^{235} serves as the capturing isotope, and Health Physics seeks the data on humans to use in the interpretation of the excretion of U^{235} by exposed production employees.

Six patients suffering from brain tumor have been administered single intravenous injections of uranyl nitrate hexahydrate at dose levels ranging from 4 to 60 mgms. All but one of the patients have succumbed to the neoplasm and the samples of hard and soft tissue and body fluid taken at autopsy and biopsy have been chemically analyzed for uranium content.

The distribution and excretion of uranium as a function of the time after a single intravenous injection are to be investigated with the aid of a mathematical model. The application of the model to small animal data has led to the belief that the metabolism of uranium (valence state of 6) can be described by four linear differential equations. The model necessitates that the human excretion data be fitted by a sum of three exponential terms. Difficulty was encountered because the human uranium excretion data are too variable for the application of Householder's modification of Prong's method for the interpolation of exponentials and, therefore, other methods must be investigated. (AEC Activity 2120)

U^{235} Recovery - Results of cost estimates indicate that the annual waste storage savings of the caustic fuel element dissolution system, as compared with acid system, are balanced by higher operating and amortization charges. The acid dissolution flowsheet was, therefore, chosen for the Savannah River U^{235} recovery plant.

A cost comparison indicated that the most economical method of preparing a satisfactory uranium product from the extraction column solvent stream is by stripping the uranium with dilute nitric acid and evaporating the product after each cycle. Other methods considered were (1) citrate-acetate stripping for each cycle, and (2) dilute nitric acid stripping and product evaporation for the first cycle and citrate-acetate stripping for the second and

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third cycles. Corrosion and explosion hazards appeared to be low with the first method. Further laboratory-scale studies showed that the use of 1.0 M sodium acetate--1.25 M sodium citrate instead of the sodium hydroxide--sodium citrate originally proposed would prevent local precipitation of sodium diuranate. The permissible range of reagent concentrations was very narrow, however. Radiation studies on second-cycle waste and product solutions indicated that citrate solutions are stable to the amount of radiation to which they would be exposed in this part of the process.

The 3-in.-diam. 14-in.-long simulated Savannah River tubular fuel elements were cut into 3-in.-long pieces and mechanically compacted before being charged to the 6-in.-diam. continuous dissolver. The packing density in the dissolver was 40%, which is approximately the same as that previously determined for cylindrical slugs. Since the ductility of spent fuel elements is very low, modifications of the procedure may be necessary for use with irradiated elements. (AEC Activity 2560)

PROGRAM 3000 - WEAPONS

Special Electromagnetic Separations - Product assays for the fifth electromagnetic separation of the heavier plutonium isotopes are given below.

	Isotopic Content (atom %)				Product wt.
	Pu 239	Pu 240	Pu 241	Pu 242	(Grams Pu)
Pu 241 Product	23.7	26.9	48.5	0.8	0.101
Pu 240 Product	17.8	81.3	0.88	0.4	0.352

Preparations are being made for a sixth separation of plutonium assaying 12.7% Pu^{240} and 1.7% Pu^{241}. (AEC Activity 3601)

HOMOGENEOUS REACTOR PROJECT

Homogeneous Reactor Test - The contractor's work on the reactor shield tank is 85% complete. Walls have been installed to a height of 20 feet above the tank bottom, side stiffeners were welded in place, and the columns which support the roof plugs are in place. At the present rate, the tank should be completed early in February. The control room area was released to ORNL forces and finishing work is underway.

The fabrication of dump tanks and cold traps has begun; the gas separators and one injection pump are nearing completion. Several components are still being delayed by the delivery of large welded pipe. The fabrication of this pipe is proceeding after numerous difficulties in producing welds acceptable to ORNL. The overall project is approximately two months behind the schedule originally proposed in January, 1954. (AEC Activity 4109.1)

HR Blanket Engineering - The first pump designed specifically for ThO_2 slurries was received from Westinghouse, and is ready for inclusion in a test loop. The ThO_2 blanket mock-up was redesigned to provide for pressurization at the blanket vessel itself, thus insuring better pressure equalization across the core vessel. An 18-inch model of the blanket pressure vessel was tested with water in a circulating system at 150°C; it is now ready for testing with slurry.

Of particular interest in subsequent tests will be the effectiveness of provisions for reslurrying sediment at the bottom of the outer vessel and on the top of the core vessel. Steam heating of the core vessel and measurement of the heat transferred across its wall appear to indicate that the present slurry injection method may be more effective in maintaining a well mixed slurry in the blanket than natural convection caused by non-uniform heat generation within the blanket, assuming that boiling does not occur. (AEC Activity 4109.31)

HRP Chemistry--Fuel Studies - The solubility of uranyl chromate in water was determined from the ice eutectic to 145°C. The stable solid phase below 66.3°C (probably $UO_2CrO_4 \cdot 5\text{-}1/2H_2O$) has a very high positive temperature coefficient, the solubility going from 0.08314 molal at 0.0° to 6.19 molal at 66.3°C. Above 66.3°C the stable solid is probably the monohydrate, and it has essentially a zero temperature coefficient, the solubility remaining at approximately 6.2 molal to at least 145°. All concentrations of solution show hydrolytic precipitation of a basic uranyl chromate at temperatures of from 110°C for very dilute solutions to 140°C for nearly saturated solutions. Efforts to prepare a stable solid uranyl dichromate failed. However, solutions with a chromium to uranium mole ratio of two were investigated at elevated temperatures and are found to be stable to higher temperatures than are the equivalent uranyl chromate solutions. (AEC Activity 4109.31)

HR Blanket Studies--Chemical Development - Approximately 1100 lb of thorium oxide was produced, in 200-lb batches, from mantle-grade thorium nitrate by

treating the nitrate with oxalic acid and calcining the thorium oxalate at 370°C for 5 hr and then at 650°C for 3 hr. The oxide is to be used in preparation of slurry for dynamic tests. Both the thorium nitrate and the oxalic acid contained excessive sulfate, and the thorium oxide product contained 6000 to 7000 ppm of sulfate. There were indications that the sulfate content of the product could be decreased by dissolving the thorium nitrate in acetone, filtering off the acetone-insoluble material, and precipitating thorium oxalate from the acetone solution by sulfate-free oxalic acid. Thirty grams of thorium oxide containing only 17 ppm of sulfate was produced by this method from a sample of thorium nitrate that contained 50 ppm of sulfate. (AEC Activity 4109.31)

HRP Miscellaneous Equipment - Specifications for the Homogeneous Reactor Test valves were completed; contracts were awarded for the 1/2-in., 500-psi valves and for the 1/2-in., 2000-psi inlet and 500-psi outlet valves. The former are now being fabricated, while materials for the latter are being procured by the vendor. Proposals have been received for the remaining valves (1/8-in., 2000 psi) and the contract for these will be awarded January 20. The conceptual drawings for the valves are included in ORNL-1813. Earlier bellows tests demonstrated satisfactory operation for the first two types of valves. Subsequent testing of two 1-1/8-in. Fulton Sylphon bellows at 2000 psi and 250°C with 1/8-in. stroke, showed a life of 40,000 and 50,000 cycles, respectively.

The 1/2-in. prototype valve from Associated Valve and Engineering Company was received, and preliminary checks on leak tightness and flow coefficient are satisfactory. Testing at design conditions (2000 psi and 300°C) will begin upon completion of the valve test loop. The valve test loop components are on hand. Welding on the 50-liter high pressure tank is 75% complete. The assembly of this system is scheduled for January.

Fabrication of the HRT mock-up loop is about 80% complete; the startup is expected in January. Installation of the high-pressure boiler and air compressor is complete, and the high-pressure circulating system is in the final stage of fabrication.

The apparatus for the first phase of the HRT dump test is complete and is being calibrated. It withstood a 3000 psi hydrostatic test. Actual testing will be underway by January 1. (AEC Activity 4109.4)

Core and Blanket Systems Development Studies - Fabrication of the Zircaloy-2 core vessel for the HRT, at the Newport News Shipbuilding and Dry Dock Company, is approximately 50% complete.

One of two hemispherical stainless-clad pressure vessel heads manufactured by Lukens Steel Company was found to be too thin in the center; consequently, a new one will be made.

Tests of three aluminum prototypes of the core vessel were completed and this program was terminated. The aluminum vessel with 1/4-in. thick spherical and 3/8-in. thick conical sections collapsed under 435 psi external pressure and burst under 970 psi internal pressure.

A bolted joint between stainless steel and Zircaloy-2, containing a Zircaloy-2 gasket and Armco 17-4 PH bolts, was still satisfactory after 100 cycles of heating and cooling between 200°F and 572°F.

A stainless steel nozzle welded into a 4-in. carbon steel plate with 0.4-in. thick stainless cladding has been cycles 2000 times between 200°F and 500°F, and no failure due to thermal stress has been detected. (AEC Activity 4109.4)

HRP Gas Handling Equipment - The high-pressure recombiner loop has been operated at 1000 psi and full operating flow; the platinized-pellet catalyst was as efficient as at low pressures. An explosion, probably caused by some platinum getting into the electrolytic gas supply, resulted in a shutdown. The necessary maintenance has been completed, and further tests will be run to find a metal-based catalyst which will be less likely to chip or break than the alumina. (AEC Activity 4109.4)

HRP Fuel Circulating Equipment - Two model 400A pumps, one 300A, and one 200A were received from Westinghouse. The volute casing of one 400A unit was returned because of defective material. One 400A pump has operated approximately 110 hours on HRT fuel mixture and 100 hours on water. During this time several hundred start-stop operations were performed and several slow startup (flow at 150 gpm) tests were run. One thrust bearing failed during a fuel run; wear was excessive, with chunks gouged out in some areas. The cause of failure has not yet been determined. The 300A pump will be used in the HRT fuel mock-up and the 200A pump on a slurry loop.

Work has started at Westinghouse on fabrication of the 400 gpm and 230 gpm pumps for the HRT. Material samples were received at ORNL; all of the stainless steel used in the pump will be subjected to Huey corrosion tests at ORNL.

After a run on the injection pump slurry loop with Lindsay No. 8 slurry in which the letdown valve trim was destroyed in 50 hours, the loop was charged with oxide prepared at the Laboratory. To date this slurry has been run 650 hours and the valve is still throttling at 1000 psi.

After 290 hours operation of the 4000-gpm loop on water, it was dismantled to recheck leak tightness of the ring-joint flanges and to prepare for operation on UO_2SO_4 solution. No appreciable increase in leak rate of the flanges could be observed on the helium leak detector.

The Scott and Williams drive units for the HRT injector pumps are on order and the ORNL shops have all material for fabrication of the diaphragm heads.

The ORNL 5-gpm pump, which had been on an endurance test to determine life expectancy of the various parts, suffered a stator failure after a total of 4850 hours running time on the pump and 6750 hours on the stator. The pump circulated water at 250°C. (AEC Activity 4109.5)

HR Heat Removal Equipment - Agreement between Foster Wheeler and ORNL on detail design of the HRT fuel and blanket heat exchangers has been almost completed. Fabrication of some parts has begun. Tubes, tube plates, and channel heads have been received, or are expected shortly by Foster Wheeler. (AEC Activity 4109.5)

Homogeneous Reactor Instrumentation - The main HRT instrumentation effort is centered about detail design of components, their interconnections, and the distribution of required electrical and pneumatic services. Component testing completed satisfactorily includes seals for electrical connections, controls for pump flows, performance of valve bellows (life tests), and designs for valve flanges. The status of the present committed expenditure for HRT instrumentation is $75,000 or approximately 40% of the total estimate. (AEC Activity 4109.6)

HR Fuel Reprocessing - In studies on thermal breeder fuel solution processing, $Nd_2(SO_4)_3$ precipitated from 0.02 m UO_2SO_4--0.005 m H_2SO_4 onto hot surfaces, and no significant amount of precipitate remained suspended in the liquid phase. The solubility of $Nd_2(SO_4)_3$ in this solution was about 0.06 g/kg H_2O at 260°C when mixed fission products were present. This represents less than 0.25% poison for the core system of an aqueous homogeneous thermal breeder reactor. Rare earth sulfate crystal growth at 250-325°C on titanium pins immersed in simulated fuel solution was more pronounced than on Zircaloy-2, and more pronounced at higher temperatures than at lower.

More than 99% of the iodine was removed from solutions with an initial iodine concentration of 10^{-5} mole/kg H_2O by contacting the vapor phase above the solution with metallic silver at 260-315°C. With oxygen present in the system, the initial oxidation state of the iodine was immaterial.

The optimum length of a 0.25-in.-diam. unit hydroclone in the liquid-solid separation system was shown to be 0.9 in. The system will use a recycle holdup tank for the hydroclone underflow, which eliminates the need for a continuous pressure let-down system.

In studies on processing the blanket of the plutonium producer reactor, the PuO_2 precipitation rate was so rapid that the plutonium in the blanket under steady-state conditions should exceed its equilibrium solubility by only 0.3 mg/kg D_2O. This will result in only a slight increase of Pu^{240} in the product. When Pu(VI) was added to simulated fuel solution, it was very rapidly reduced to Pu(IV) and the PuO_2 precipitation rate was not affected. The equilibrium adsorption of plutonium on zirconium at 250°C and 160 psi O_2--160 psi H_2 appeared to be about 0.05 mg/cm^2. Non-equilibrium values of 0.92 and 0.83 mg/cm^2 were obtained for titanium and of 0.22 and 0.60 mg/cm^2 for stainless steel.

In scouting studies on possible methods of processing the thermal breeder reactor blanket, ThO_2 that had been irradiated for 62 hr in the ORNL graphite reactor was refluxed with 12 N H_2SO_4 for 4 hr; 70% of the Pa^{233} and only 0.2% of the thorium were dissolved. Complete solution of non-irradiated ThO_2 was obtained by refluxing it for 1 hr with 8.0 N HNO_3 containing 0.04 N HF. The product was 1.4 M $Th(NO_3)_4$--2.5 N HNO_3. A slurry of ThF_4 at 250°C under 175 psi O_2 corroded Zircaloy-2 at a rate of 440 mils/year in a static test. The presence of up to 40% ThO_2 in a $ThF_4 \cdot ThO_2$ slurry did not markedly inhibit the attack on Zircaloy-2. (AEC Activity 4109.81)

PACKAGE REACTOR DEVELOPMENT

Army Package Power Reactor (APPR) - On November 19, 1954, seventeen firms submitted lump-sum contract proposals to the AEC for the design, construction, and performance testing of the APPR. The proposal prices ranged from $2,237,000 to $6,900,000. Several firms submitted bids for fabrication of the core, including control rods, at a cost of substantially less than $140,000. The proposal of the American Locomotive Company was selected and a contract was signed with the AEC on December 10, 1954.

Preliminary corrosion testing of the APPR fuel plates to be irradiated in the STR was successfully completed at the Bettis Field Laboratory (Westinghouse). Final assembly of fuel plates into an integral unit is now underway at the ORNL Metallurgy Division. Delivery of the assembly to the STR at Arco is scheduled for January 15, 1955. (AEC Activity 4250)

AIRCRAFT NUCLEAR PROPULSION PROJECT

Aircraft Reactor Experiment - After completion of the operation of the ARE the fuel was dumped, a sample of fuel was taken from the dump tank for analysis, and the fuel was then allowed to freeze in the dump tank. The shielding blocks were removed from the tops of the pits and radiation measurements are being made daily to ascertain decay rates. A list of the equipment to be salvaged and a salvage schedule was prepared. (AEC Activity 4401.1)

Aircraft Reactor Test - A preliminary summary was prepared of the considerations of hazards associated with the operation of the Aircraft Reactor Test (ART), formerly known as the Circulating-Fuel Reactor Experiment (CFRE). In the report, which will be discussed with the AEC Reactor Safeguard Committee in February, it is proposed to construct the ART in an addition to the ARE building. (AEC Activity 4401.1)

ANP Experimental Engineering--Basic Developments - The fourth sodium-beryllium-Inconel mass transfer test assembly has operated over 300 hr of a scheduled 1000 hr with a sodium flow rate of 4 gpm, a Reynolds number > 400,000 inside the beryllium tube insert, a Reynolds number < 100 between the beryllium insert and the Inconel container, a temperature of 1300°F at the beryllium,

and a temperature of 1000°F at the coldest portion of the circuit. Since the previous tests had provided evidence of a relationship between mass transfer and the thickness of the almost stagnant layer of sodium between the beryllium and the Inconel, this test assembly was designed to provide definitive information on the relationship.

A high-velocity, high-temperature-differential Inconel test loop was terminated after 800 hr of operation because a small leak developed. This test system consisted of a pump, a heating section, and a cooling section. The circulated fluid was $NaF-ZrF_4-UF_4$ (53.5-40-6.5 mole %). Operation was continuous at a maximum Reynolds number of 10,000, maximum and minimum fluid temperatures of 1500° and 1300°F, and a maximum Inconel temperature of about 1740°F. Samples of the Inconel were submitted for metallurgical examination (see Corrosion Studies). A similar test assembly is being operated with a maximum Reynolds number of about 10,000, maximum and minimum fluid temperatures of 1500° and 1200°F, and a maximum Inconel temperature of about 1600°F. (AEC Activity 4401.1)

ANP Experimental Engineering--In-Pile Components - Sections of two proposed in-pile loop heat exchangers were tested to obtain design information. A 30-in.-long finned tube section with a fluoride salt flowing through it at a temperature of 1500°F and forced air flowing over it at a rate of 0.1 to 0.5 lb/sec gave a heat transfer rate of 25 Btu/sec. However, the air pressure drop of 70 psi was excessive. The conditions for the second test were the same except that the tube was unfinned. A heat transfer rate of about 20 Btu/sec was obtained with a 30-psi pressure drop. The heat exchanger for the in-pile loop can be as much as 5 ft in length.

A pump designed for the horizontal in-pile loop for insertion in the MTR was tested with water and then welded in a loop for testing at design conditions. The water tests indicated satisfactory performance. (AEC Activity 4401.1)

ANP Critical Experiments - The second in the current series of critical assemblies of the reflector-moderated circulating-fuel reactor was constructed and the required measurements were completed. A loading of 4.66 kg of U^{235} completely filled the fuel region to a density of 0.092 g of U^{235}/cm^3 and resulted in 0.9% excess reactivity in the assembly; thus this loading corresponds to a "clean" critical mass of approximately 4.35 kg of U^{235}. This value differs significantly from the calculated value of 7.16 kg. Neutron-flux and fission-rate distributions were measured in the assembly, and the reactivity values of several materials were determined. (AEC Activity 4401.2)

ANP Radiation Damage - A circulating-fuel loop has been operating in hole HB-2 of the LITR at full power since December 14. The fuel mixture, $NaF-ZrF_4-UF_4$ (62.5-12.5-25 mole %), which contains approximately 2 g/cm^3 of U^{235} is circulated by means of a sump pump external to the reactor. The maximum fuel temperature is 1500°F. Flow at the in-pile end of the loop corresponds to a

Reynolds number of 5000. The exact amount of fission power produced has not yet been determined, but it appears to be of the order of 2 kw. Therefore, considerable electrical heating is necessary to maintain the entire loop at an elevated temperature. The maximum power density will be determined when the experiment is terminated; it has been estimated to be of the order of 1000 w/cm^3. The loop is scheduled to operate for 500 hr.

Preliminary tests have been made of a miniature in-pile loop to be operated vertically in a LITR fuel-element position. Isothermal operation at 780°C with a pump speed of 4000 rpm gave a salt velocity of 3.3 to 3.8 fps in a 200-mil-dia tube and a Reynolds number of 3000. The loop was subjected to four freezing and melting cycles and was operated a total of 260 hr. (AEC Activity 4401.4)

ANP Reactor Chemistry - The stockpiling of processed fluoride mixtures has continued, and approximately 6000 lb will be available by January 1. An inventory of compositions and quantities available will be distributed to interested personnel so that requests for these materials can be based on availability.

Previous results which indicated that CrF_2 disproportionated in NaF-KF-LiF (11.5-42.0-46.5 mole %) at 600° and 800°C were confirmed. A preliminary estimate of the equilibrium constant K_x (concentrations expressed as mole fractions) for the reaction $3CrF_2 = 2CrF_3 + Cr^o$ indicates that it is of the order of 10^3 at 600° and 800°C. The value of K_a (calculated from the standard free energy of formation of CrF_2 and CrF_3) for this reaction at 800°C is about 10^{-3}.

Preliminary experiments on the electrolytic treatment of fluoride melts under H_2, for which nickel cathodes and graphite anodes with low current densities ($\sim$0.3 amp/cm^2) were used, showed very poor efficiency for overall reduction when UF_4 was present. Additional experiments at higher cathode current densities (2 amp/cm^2) gave much better efficiencies. When reduction of UF_4 at the cathode is carried out faster than the reduced product can dissolve or react with reducible compounds in the melt, efficiencies of the order of 50% are obtained. In this way UF_3 has been electrodeposited from ZrF_4-base mixtures and U^o from alkali fluoride mixtures. Attempts are being made to utilize this behavior for the rapid preparation of UF_4-UF_3 bearing melts with a controlled amount of reduction. (AEC Activity 4401.5)

ANP Corrosion Studies - A preliminary examination of the first high-velocity, high-temperature-differential loop after it had circulated the fluoride fuel mixture NaF-ZrF_4-UF_4 for 800 hr with a Reynolds number of $\sim$10,000, a temperature drop of 200°F, and a maximum fuel temperature of 1500°F revealed subsurface void attack with a maximum penetration of 18 mils. To achieve the 1500°F fuel temperature, the maximum tube wall temperature was 1740°F.

Metal crystals, which indicated mass transfer, were found in a Hastelloy B thermal-convection loop which had circulated sodium for 500 hr at 1500°F. Metallographic examination of another Hastelloy B loop, which had plugged after 1010 hr of operation, showed intergranular attack to 1.5 mil in all sections, and the hot-leg surface was very rough. There were scattered depressions that were as deep as 4 mils. No deposit was found on the cold-leg wall.

The dissimilar metal transfer characteristics of zirconium and type 304 stainless steel with sodium as a coolant were investigated. Metallographic examination made after tests at 540° and 810°C for 100 hr showed that there was a slight amount of transfer of zirconium to the steel and that the transfer increased with increasing temperature. (AEC Activity 4401.5)

ANP Metallurgy--Fabrication - Thirty-four control rods were fabricated for the GE-ANP Project. The rods were type-304 stainless steel tube mechanically filled with a 50-50 mixture of B_4C and aluminum. After the tubes were filled, the assembly was evacuated, closed, and swaged from 5/8 to 1/2 in. in diameter.

Solid fuel elements of Al-UO_2 with 52S aluminum cladding, for strengthening the fuel element, were fabricated for use in a Lid Tank Shielding Facility mockup. The fuel plates were made by hot rolling to establish the bond and finished by cold rolling with reductions of 20, 30, and 40% to increase the strength of the plates. (AEC Activity 4401.5)

ANP Metallurgy--Welding and Brazing - Several small radiator sections were fabricated of Hastelloy B. The tube-to-header welds were made by using the automatic welding machines available in the Welding Laboratory, and the type-310 stainless steel clad copper fins were brazed to the tubes with Coast Metals No. 52 brazing alloy. These radiators were then cyclically heated between room temperature and 1500°F. One radiator has been tested for 500 hr at 1500°F with intermittent cooling to room temperature 100 times, and another radiator was held 500 hr at 1500°F with intermittent water quenches. Both radiator sections were still leak tight after these drastic tests.

Corrosion tests have shown the Ni-GE, Ni-Ge-Cr, and Ni-Ge-Cr-Si alloys to be the most promising brazing alloys for use as back-up brazes for intermediate heat exchangers. It has been difficult to produce large quantities of these brazing alloys because of slag entrapment during melting; however, the technique of prebright annealing of loose powder compacts of the alloys prior to arc melting has resulted in production of higher-purity ingots of the brazing alloys.

A careful analysis of the phases present in Hastelloy B at various temperatures was undertaken in order to obtain a better understanding of the welding and mechanical problems involved in the use of Hastelloy B. Preliminary results indicate that the rate of cooling from a temperature of 1700°F or higher greatly affects the rate of precipitation of the second phase on

annealing at 1300 and 1500°F. The microstructure of Hastelloy B varies greatly with the length of aging time, and thus it appears that equilibrium conditions are not reached even after 1000 hr of aging.

Preliminary observations indicate that there are at least three phases present in the microstructure of Hastelloy B. Studies are under way for correlating the microstructure with prior mechanical and thermal treatment so that the behavior of the alloy can be predicted more accurately. (AEC Activity 4401.5)

ANP Heat Transfer Research - Preliminary forced-convection heat-transfer experiments with the fuel mixture $NaF-ZrF_4-UF_4$ (53.5-40-6.5 mole %) in an Inconel tube yielded heat-transfer coefficients which differed from expected values by only 24% after 115 hr of operation at about 1300°F; no corrosion deposits were observed on the inner wall of the tube at the end of the experiment. Some preliminary fluid flow information on a proposed ART core was obtained, and some unfavorable flow characteristics were found. The question as to whether or not the electric currents which generate heat in the circulating fluids of experimental volume-heat-source systems affect the fluid flow characteristics was investigated. It was found that the hydrodynamic structure was not influenced by the presence of the electric currents. (AEC Activity 4401.5)

ANP Physical Properties Research - The enthalpies and heat capacities of $NaF-ZrF_4-UF_4$ (53-43-4 mole %) were determined; the heat capacity in the solid state over the temperature range 70° to 525°C was found to be 0.18 cal/g-°C, and the heat capacity in the liquid state over the temperature range 570° to 885°C was found to be 0.26 cal/g-°C. The enthalpies and heat capacities of $LiF-KF-UF_4$ (48-48-4 mole %) were also obtained; the heat capacity in the solid state over the temperature range 125° to 465°C was found to be $0.234 + 0.95 \times 10^{-4}$ T (cal/g-°C); and in the liquid state over the temperature range 565° to 880°C, it was found to be $0.657 - 3.93 \times 10^{-4}$T (cal/g-°C). The thermal conductivity of molten $NaF-ZrF_4-UF_4$ (53.5-40.0-6.5 mole %) was determined to be 1.2 Btu/hr-ft-°F. (AEC Activity 4401.5)

ANP Analytical Studies - Evaluation of the data obtained for the determination of U(III) in fluoride fuels by the methylene blue, one-step, oxidation method has shown that the coefficient of variation for NaF-KF-LiF base materials is 2%. The coefficient of variation obtained by the hydrogen evolution method for the determination of U(III) in similar materials, including $NaZrF_5$-base eutectics, is 4%. The agreement of the results obtained by the two methods is, in general, satisfactory. However, the procedure which involves oxidation with methylene blue permits the making of simultaneous determinations of U(III) and total uranium on the same sample.

Further studies were carried out to improve the method which is used for the determination of uranium metal in fluoride fuels by converting the metal to the hydride and measuring the quantity of hydrogen which is evolved by the

thermal decomposition of UH_3. Although results of satisfactory precision and accuracy can be obtained by the procedure in which the decomposition of the hydride is carried out under an atmosphere of carbon dioxide, it was found that for some samples, periods in excess of 8 hr were required for the complete evolution of hydrogen. In addition, no completely dependable method has been found for removing the carbon monoxide which is formed when carbon dioxide is reduced by uranium metal and trivalent uranium. In order to eliminate the possibility of interference by carbon monoxide and to reduce the ignition time, procedures were tested in which the decomposition of the hydride was carried out under atmospheres of hydrogen chloride and ammonia. In the presence of each of the above gases the volumes of hydrogen which were obtained were not reproducible and were less than predicted by the postulated reactions

$$UH_3 + 3HCl \longrightarrow UCl_3 + 3H_2$$

$$\text{and} \quad 2UH_3 + 4NH_3 \longrightarrow 2UN_2 + 9H_2.$$

A procedure is now being tested in which the sample is ignited in a stream of oxygen and the effluent gases are passed over hot copper oxide to ensure the conversion of hydrogen to water, which is then measured volumetrically at reduced pressure. (AEC Activity 4401.5)

ANP Shielding Analysis - A method was devised for calculating the induced heat distribution from the scattering and absorption of gamma rays in slab shields. The general procedure was coded for the ORACLE. Initial results were obtained for a 9-in. slab of beryllium with incident gamma rays having energies up to 2.5 Mev. Modifications to the code are being made to include gamma rays with energies up to 10 Mev. Analysis of the neutron air-scattering problem for the divided shield was completed, and the problem is to be coded for the ORACLE. The energy flux of singly and multiply air-scattered neutrons at the crew compartment will be calculated by using the code. Calculations of the penetration of composite slab shields by slant incident monoenergetic gamma rays were completed; a survey of the various thicknesses of hydrogeneous material and lead to be expected in the crew compartment of the divided shield was included. (AEC Activity 4401.7)

ANP Shielding--Bulk Shielding Facility (BSF) - Measurements of gamma-ray dose and thermal-neutron flux were extended to appreciably greater attenuations than had previously been possible. The reactor was operated at 1 Mw for the greatest attenuations. The gamma-ray data will be particularly valuable as design data for the top water shields of reactors such as the ORR. The thermal-neutron flux at large distances checked rather closely with calculations of the expected photoneutron flux produced in the 0.016% deuterium content of the water shield. A preliminary evaluation of the Hornyak Button as a pseudo fast-neutron dosimeter in water by comparison with a Hurst dosimeter indicated identical attenuations from about 50 to 150 cm. Three inches of lead was inserted between the reactor and the detectors to reduce the gamma-ray level so that the Hurst dosimeter could be operated.

Previously reported experimental work at the BSF (Report ORNL-1760) showed that the visible light intensity to be expected around an operating aircraft reactor is proportional to the gamma-ray dose in the air. A 200-Mw reactor in a GE R-1 aircraft reactor shield was estimated from further experiments to produce light equivalent to that from a 30-watt tungsten-filament light bulb. (AEC Activity 4401.7)

ANP Shielding--Tower Shielding Facility (TSF) - Most of the measurements in the detector tank for the GE-ANP R-1 mockup experiment were completed. These measurements included the fast-neutron dose rate distributions in plain and borated water, which indicated that an appreciable quantity of capture gammas were present in the plain water, i.e., the gamma-ray dose in the center of the tank was reduced by a factor of 1.7 by the addition of 9.85 wt % boron. The fast-neutrons relaxation lengths which were observed at the side and front of the tank are 5.2 and 4.2 cm, respectively.

The reactor was removed from the mockup and installed in a 28-in.-dia tank in order to conduct an experiment which will contribute to the Los Alamos Weapons Testing Program. (AEC Activity 4401.7)

GENERAL REACTOR RESEARCH

General Reactor Radiation Damage - Three natural-beryllium-bearing crystals which were bombarded with fast neutrons reveal widely variable results. The crystals were beryl, $Be_3Al_2Si_6O_{18}$, phenakite, Be_2SiO_4, and chrysoberyl, $BeAl_2O_4$. Bombardment with an integrated flux of 3.6×10^{20} neutrons/cm^2 completely disorders beryl and renders the crystal amorphous. The other two crystals, however, retain a high degree of crystal perfection despite the fact that as many as 2 to 3% of the atoms are displaced from their normal sites, as seen from lattice-expansion measurements. Although the expansion effects are structure dependent, the disordering effects seem to be more closely related to the binding and the ability to reform ruptured bonds.

A correlation has been made of radiation stability with rigidity of organic polymers. The effects of the reaction of high-energy radiation with polymers is to cause crosslinking or chain cleavage. The rate of change in physical property is governed by the rate of crosslinking and chain cleavage, and also by which reaction is most predominant. The reaction rate is governed by the chemical structure of the polymer and by the hardness or rigidity. Whether it is the crosslinking reaction or the cleavage reaction that predominates is also governed by both the chemical structure and the rigidity. The dependence of crosslinking on the rigidity (which is governed by the strength of the Van der Waals force) can be explained in the following way. The bonding together of molecular chains must be accompanied by a decrease in specific volume. This volume change can be thought of as a diffusion of holes out of the material. For very rigid materials diffusion is very slow and it is this rather than the chemical structure which is rate controlling for the cross-

linking process. For non-rigid materials, although still a rate determining factor, the diffusion is more rapid and may not be the controlling factor.

The volume change for cleavage is much smaller than the volume change for crosslinking and therefore the rigidity dependence of the cleavage reaction is much smaller than for the crosslinking reaction. Only for extremely rigid materials is the cleavage reaction dependent on rigidity.

Dissolution Studies - The ratio of $UO_2.\underline{x}H_2O$ to UAl_3 in the caustic dissolver product was shown to depend on the feed caustic concentration. Caustic digestion readily converted all the uranium solids to UO_2 hydrate. On a laboratory scale, separation of the uranium solids by filtration of the caustic dissolver solution through nickel-plated screens with 10- to 30-μ pores was feasible but slow, and hydroclones showed no promise of effecting complete uranium separation. Scavenging with inorganic solids was not practical in either centrifuge or hydroclone experiments. Large-scale continuous centrifugation and filtration gave unsatisfactorily high uranium losses (0.3 to 0.5%).

Caustic dissolution of uranium was carried out in the 5.5-in.-diam. continuous dissolver at a satisfactory rate with both sodium hydroxide and sodium hydroxide--sodium nitrate feeds. The rate was of the order of 10 kg per day, varying somewhat with caustic concentration and flow rate. Under comparable conditions the rate in the presence of sodium nitrate was about 1.6 times as rapid as with caustic alone. However, even in the presence of sodium nitrate the hydrogen evolved was about 72% of the total dissolver off-gas, and the final uranium product was sodium diuranate, which is as difficult to separate from the caustic dissolver solution as the uranium oxide formed with caustic alone.

A wafer from a 9-months-cooled Hanford J slug was successfully batch-dissolved in 6 M sodium hydroxide. The ratio of the amount of fission products accompanying the solid to that remaining in the solution when the solid and liquid were separated was, with the exception of cesium and iodine, greater than 1000. With cesium and iodine the ratio was 0.001 or less. (AEC Activity 4581)

Sanitary Engineering and Development - The evaporation study (ORNL-1763, p. 15) to determine the watertightness of the asphalt-tamped clay liner for a disposal pit was completed after a 44-day observation period. Data analysis implies that under the conditions to which the liner was subjected it is leaking at a mean rate of about 0.02 ± 0.03 gallon per sq. ft. per day, amounting to a mean loss of 8.3 ± 13.1 gallons per day. Nonuniformity of the asphalt liner thickness and puncturing of the liner with tamped shale are deemed responsible for its non-watertightness and the decrease in its retentive qualities by a factor of approximately 10^3. (AEC Activity 4583)

PROGRAM 5000 - PHYSICAL RESEARCH

ISOTOPE PRODUCTION

Stable Isotope Production - An estimated 560 grams of iron isotopes (masses 54, 56, 57, 58) were collected in the interim July 18 to November 8, 1954. During the separation, emphasis was placed on obtaining high-purity Fe^{58} to be used for producing 45 d Fe^{59} in the reactor. A total of 1.4 grams of Fe^{58} was collected. Collection of Fe^{56} (for cross-section measurements) in a "second-pass" separation from charge feed already enriched to 99.7% Fe^{56} was completed. The estimated collection was 1.44 grams, and the purity is expected to be 99.99+% Fe^{56}.

Refinement of the ion source and operating technique has increased the average calcium ion beams at the collector from 100 ma to 150 ma.

New lots of K^{41}, Cr^{50}, Fe^{56}, Ni^{64}, and Sm^{154} were added to the inventory. Charge materials were prepared for the separation of isotopes of calcium, iron, rubidium, and tantalum. A search is under way for natural Ta^{180}.

Fifty shipments were made, consisting of 42 different isotopes of 16 elements. (AEC Activity 5121)

PHYSICS

Scintillation Spectrometry - A gamma-gamma coincidence system which uses the 3" x 3" NaI scintillation spectrometers was improved by new and better anti-Compton scattering shields; this improvement makes possible the observation of weaker transitions in the presence of strong ones. The study of the decay of 8 d I^{131} was recommenced with this system and some additional weak lines have been found. New gamma rays of about 80 kev, 175 kev, 275 kev, and 331 kev were observed. The line near 80 kev is in coincidence with only the 638-kev gamma ray and appears to be the gamma ray from the 724-kev level to the 638-kev level. This gamma ray was previously found at this laboratory, but the random-coincident sum line at 648-kev made its existence doubtful, whereas it now appears certain. The 275-kev gamma ray is in coincidence with the 364-kev line but not the 638- or 724-kev gamma rays. Hence, it appears to be the transition between the 638-kev and 364-kev levels. The weak gamma ray at 506-kev which has long been known although it is quite weak seems to be a transition to the gound state; the 175-kev and 331-kev gamma rays which are in coincidence with each other probably come from this level. The sum of their energies is the correct value and neither of them is in coincidence with any other gamma ray. A determination of the intensities of these various weak gamma rays is now in progress. (AEC Activity 5200)

High Voltage Program - The energy of the proton beam from the 5.5-Mev Van de Graaff is determined by deflection of the beam in a magnetic field whose strength is determined by means of a nuclear resonance device. A new calibration has recently been obtained by the use of known p-n thresholds, using

both mass one and mass two beams. In addition, some of the previously used $F^{19}(p,\alpha\gamma)O^{16}$ resonances and the $Al^{27}(p,\gamma)Si^{28}$ resonance at 993.3 kev (mass one and mass two) were remeasured. The consistency of these data was excellent except in the case of the $F^{19}(p,\alpha\gamma)O^{16}$ resonances at 1355 and 1381 kev. New values obtained for these resonances are 1347 and 1374 kev with an error of less than two kev. These values are in agreement with recent measurements made at California Institute of Technology (private communication).

Evidence was obtained for a new isomer in Zr^{90} (N = 50, Z = 40) with a half-life of 0.83 $\pm$ 0.03 sec and a gamma energy of 2.30 $\pm$ 0.02 Mev. The isomer is found in pile-irradiated electromagnetically enriched (98.7%) Zr^{90}. Gamma measurements were made with a NaI spectrometer and multi-channel analyzer. The isomer was also produced by monoenergetic neutrons from the T(p,n) reaction. An electronic timer and an electrostatic proton beam deflector provided a repetitive cycle for short neutron bombardment and gamma counting. The yield curve exhibits a threshold at 2.3 Mev neutron energy. This is taken as evidence that the isomer is produced by inelastic neutron scattering, i.e. $Zr^{90}(n,n')Zr^{90m}$. The complicated structure of the yield curve, which was measured up to 4.4 Mev, gives evidence on the existence of excited states of Zr^{90} which feed the isomeric level. The energy and half-life are consistent with an E5 assignment for the isomeric transition. It is suggested that the 5- level might arise from the promotion of a $p_{1/2}$ proton into a $g_{9/2}$ state. (AEC Activity 5200)

<u>86-Inch Cyclotron, Applied Physics</u> - Preliminary bombardments of normal calcium in the capsule target produced Ca^{47} at the rate of 15 microcuries per hour. This indicates that if enriched Ca^{48} (50%, presently available) were used in the target, Ca^{47} could be produced at the rate of 3.7 mc/hr. The 4.8 d Ca^{47} produced would be relatively free from 154 d Ca^{45}, and would be very attractive for use as a tracer in biological studies. Suitable Ca^{47} is not readily produced by neutron-induced reactions. (AEC Activity 5200)

<u>The 86-Inch Cyclotron, Operation</u> - The cyclotron was shut down on October 22 to permit the modifications necessary for the installation of the beam deflector system. The dees, liner, and diffusion pumps were removed. Due to accumulated alpha activity from earlier bombardments of bismuth a thorough decontamination was necessary. A large peripheral section of the vacuum tank was then cut away so that a tank extension to house the magnetic deflector channel could be welded in place. The alterations to the tank are now essentially complete, and leak testing is underway. The installation of electrical bus in the pit for the magnetic deflector and for the beam-centering coils is also complete. The major components of the deflector system have been fabricated and are ready for installation and testing. (AEC Activity 5200)

<u>The 63-Inch Cyclotron</u> - Elastic scattering of nitrogen from He, C, N, O, and Ne is being investigated. The deviation from Rutherford scattering can be used to determine nuclear radii of those elements, and preliminary data

indicate a break in the curve $\frac{\sigma \text{ Ruth.}}{\sigma \text{ elast.}}$. The angular distribution of products of nitrogen-induced reactions is also being investigated; for $N^{14} + C^{12} \longrightarrow F^{18} + Be^{8}$ the distribution is apparently isotropic. (AEC Activity 5200)

Spectroscopy Research Laboratory - The Raman spectrum of DCOF was obtained; five fundamental vibrations and one combination band were identified. Atomic energy level classifications of Th IV, III, and II were evaluated and compiled. Electromagnetically separated Ca^{40}, tested for spectrochemical purity and found to be essentially free from Sr contamination, is being incorporated into "bone ash" standards for health physics studies. Preliminary optical spectro-isotopic studies of uranium isotopes in an a-c arc were made by using a 3.4-meter Ebert grating spectrograph at a dispersion of 0.35 A/mm and Doppler and Stark limited resolution of about 250,000 in the XIV order. A direct reading spectrographic method for analysis of critical metals used in reactor and cyclotron work was set up for rapid and precise determinations. A photoelectric scanning Fabry-Perot interferometer was checked for reproducibility; the central spot aperture was chosen to give reproduction of theoretical intensity distribution to within 5% of the half-width. Measurements of the cadmium red line used as primary standard were evaluated for different isotopes and the data are being correlated by the Oracle to correct for different isotopic impurities. (AEC Activity 5200)

Mass Spectrometer Laboratory - The analyses of sea water for uranium content by isotopic dilution was continued with the examination of nine more samples. The data obtained so far has a spread in the uranium content varying from 3.15×10^{-9} to 3.56×10^{-9} g of U/g of water for samples from both the Gulf of Mexico and the North Atlantic Ocean. Mean values have not yet been established but it appears that the data for the Gulf of Mexico will average a little higher than for the North Atlantic Ocean.

A theoretical study was made of the scattering of ions upon collision with residual gases in a magnetic time-of-flight mass spectrometer. Calculations show that such machines should be superior to conventional mass spectrometers for isotopic abundance measurements, especially when the variation in abundance of two adjacent masses is of the order of 10^{4} to 1 or higher.

Techniques for preparing samples for mass spectrometer analyses were developed for three elements. The addition of ammonium iodide to magnesium iodide inhibits hydrolysis and the isotopic analysis can be performed on the MgI^{+} ions. A method of producing boron tribluoride from boric acid on a micro-scale was devised; it consists of first preparing potassium tetrafluoro-borate and then heating the purified salt in a metal vacuum system to liberate boron trifluoride. Barium silica fluoride is an excellent source of silicon tetrafluoride for determining the isotopic abundance of silicon. The $BaSiF_6$ is placed in a Vycor tube and attached to the gas manifold of a mass spectrometer. The tube is first evacuated and heated to about 300°C to drive off moisture and occluded gases. Upon further heating the $BaSiF_6$ is readily converted to silicon tetrafluoride without any isotopic discriminatory effects.

Twenty-six samples were analyzed for the Stable Isotope Division, and 138 samples were analyzed for other divisions. (AEC Activity 5200)

Research and Development, Stable Isotopes - Special preparations of accelerator targets and target materials included (1) reduction of $Si^{28}O_2$ with Mg to give 5 grams of 96-97% Si^{28}; (2) preparation of 0.5 grams of Si^{29}; (3) coating of Al sheet, C strips, and mica with normal Re by electrolysis; (4) coating of mica with normal Ir both by flame-spraying and by thermal decomposition of a special plating mixture; (5) coating of thin layers of Mg on stainless steel and on copper by a modified casting technique.

The evaluation of inert gas collections is hanpered by the relatively large quantities of impurities in the outgassed samples. A Toepler pumping system containing UH_3, a Pt hot wire, and suitable traps produced high-purity argon from ordinary air. The data obtained from inert gas collections in the calutrons now give more reliable information as to collection efficiency and enhancement factors. (AEC Activity 5200)

CHEMISTRY

Inorganic Solution Chemistry - Ultracentrifugations of Th(IV) in 1 M NaCl solutions indicate much more aggregation at a given hydroxyl number n (average number of hydroxides per thorium ion) in chloride than in perchlorate solutions. For 0.015 M Th(IV) in 1 M $NaClO_4$, n = 1.0, a preliminary degree of polymerization N = 2.4 was observed, while in 1 M NaCl at the same n, N = 3.8. The difference was more striking at lower acidities. For n = 1.55 in perchlorate, N = 3 was obtained; for n = 1.45 in chloride, N is about 30. The polymerization of metal ions under hydrolytic conditions may, therefore, be very sensitive to the type of anion present.

Adsorption of Ag(I) by Dowex-1 was studied with tracer Ag^{110} in the range 1 to 12 M HCl. Adsorption was found to decrease from D = ~700 in 1 M HCl to D = ~2 in 12 M HCl. Tl(I) showed negligible adsorption ($D \leqslant 1$) throughout this whole HCl range. Re(VII) shows good adsorption both at high and low HCl concentrations, although it appears to show an adsorption minimum near 10 M HCl. Adsorption of U(IV) was found to be similar, though considerably lower than that of U(VI). It shows rapidly increasing adsorption with M HCl from ~6 M HCl on. (AEC Activity 5300)

Chemical Physics - The reaction between K and HBr was studied by a molecular beam technique, which uses for the detector a surface ionization gauge with both a tungsten and a platinum alloy filament, the former sensitive to KBr as well as to K and the latter essentially to K only. The activation energy is 3.4 ± 0.1 kcal/mol and the probability or steric factor between 1/3 and 1/2. The angular distribution of the KBr suggests that collisions with the H end of the HBr are more often fruitful than other configurations. The total collision cross section is 850×10^{-16} cm^2. (AEC Activity 5300)

Chemistry of the Solid State - In connection with the study of the structural changes occurring in simple salts on melting, the thermal expansion of solid lithium iodide, up to its melting point, was determined by using the neutron spectrometer. The molar volume of the fused salt was found to be 43.25 cc. The volume increase on melting measured 20%; this is the smallest among the lithium halides (the fluoride exhibits the largest one, 30%), but is still twice as large as the corresponding increase for cesium chloride, where no structural change in the first sphere of coordination occurs. The relatively large changes in the mole volume of the lithium halides are believed to be suggestive of radial distributions for the ions of the fused salt quite different from the coordination number 6 of the crystals, including appreciable concentrations of ion pairs or molecules. (AEC Activity 5300)

Radio-Organic Chemistry - The addition of o-tolylmagnesium bromide to benzil was shown to yield diphenyl o-toluyl carbinol rather than α-o-tolyl benzoin as had been reported by previous investigators. When phenyl-C^{14}-magnesium bromide was added to benzil, a rearrangement was found to occur under certain conditions, since the resulting triphenylketol contained carbon-14 in all of its phenyl groups.

The dehydration of cis and trans-2-phenylcyclohexanol-2-C^{14} was shown to yield 1-, 3-, and 4-phenylcyclohexene, and benzylcyclopentene. There is also some evidence for the presence of benzylidenecyclopentane.

The extent of the isotope effect in the following reactions was determined: (a) the chromic acid oxidations of 2-propanol-2-C^{14}, and of benzyl-α-C^{14} alcohol, (b) the mercuration of methyl cinnamate-α-C^{14}, (c) the iodoform reaction of ketones, and (d) the Oppenaur reduction.

A novel rearrangement was detected in the nitric acid oxidation of dimethyl-C_1^{14}-phenylcarbinol.

Investigations continued on the chemistry of organo-phosphorus compounds, some of which are valuable in solvent extraction. The de-esterification reaction between amines and neutral phosphorus esters was put upon a quantitative basis by taking advantage of the liberation of the product acids by cation-exchange resins in the H+ form. Rate of acid formation is being studied as a function of time, temperature, structure, and ratio of reactants.

Attempts to synthesize compounds of the type, $R_2P(O)CH_2COOH$, that will allow the participation of the strongly coordinating phosphoryl group in chelate formation were so far unsuccessful. (AEC Activity 5300)

Radiation Chemistry - The alpha-ray induced decomposition of CO in the presence and absence of Kr was studied. The velocity constants for these reactions, based on the partial pressure of CO only and corrected to a 2 cm. sphere, are 14.1 for CO and 14.2 for CO + Kr. When the equivalent pressure of Kr is considered, the velocity constant is 10.0. The lack of evidence of

retardation or acceleration would indicate that there is no energy transfer from the CO ions to Kr or from Kr ions to CO. Retardation due to energy transfer from the CO ions to Kr is energetically possible as their ionization potentials are 14.1 ev and 14.0 ev, respectively. Similar absence of energy transfer where retardation is energetically possible had been reported previously for the first time in the CO + Xe reaction.

Because of the possible use of thorium nitrate solution as a blanket material in a two-region power breeder reactor, the decomposition of concentrated solutions by pile radiations and by fission particles (U^{235} added) is being studied. The principle decomposition products are hydrogen and oxygen from decomposition of the water, and nitrogen and oxides of nitrogen from decomposition of the nitrate. The hydrogen yield decreases with increasing thorium concentration, and for fission fragment irradiation, (0.144 molar $U^{235}O_2SO_4$ added) the yield varies from 1.11 H_2/100 ev. in 0.26 molal thorium nitrate solution to 0.33 H_2/100 ev. in 2.73 molal solution. For mixed fast neutron and gamma radiations (graphite pile) the yield varies from 0.26 to 0.05 in the same concentration range. For fission particle irradiation at a dose of 1.4×10^{21} ev/ml the yield of N_2 increases from zero in 0.2 molal solution to 0.06 N_2/100 ev in 2.7 molal solution. N_2O is produced in about one-tenth this amount and NO_2 and nitrite (in solution) are observed in trace quantities. (AEC Activity 5300)

Chemistry of Fused Salts - Hill and Wetmore (Can J. Chem. 32, 864, 1954) conclude from transference and electrical conductivity measurements that silver chloride dissolved in molten silver nitrate is essentially completely complexed to form Ag_2Cl^+. Therefore, experiments were performed to observe migration of ions during electrolysis between silver electrodes in this molten system. These experiments demonstrate that although some of the chloride is complexed as a cation, probably Ag_2Cl^+, this is incomplete and much free Cl^- ion is also present.

Electrical conductance and density measurements as functions of temperature were made on the complex mixtures formed on melting potassium acid sulfate, $KHSO_4$, but this salt loses water so easily that it was impossible to obtain reliable data concerning the stability or electrolytic dissociation of the HSO_4^- ion. However, inflections in both density and conductivity curves were found which correspond to the onset of rapid decomposition of $KHSO_4$ into $K_2S_2O_7$ in the range from 320-400°C. It was also observed that SO_3 gas was evolved from $K_2S_2O_7$ melts above 370°C. (AEC Activity 5300)

Uranium Chemistry of Raw Materials - Pilot plant evaluation of multistage mixer columns and component testing of mixer-settlers was made for the extraction of uranium from acid leach liquors by using Armeen 2-12 in Amsco G as the extractant. Mixer columns do not appear to be suitable for this sytem. Studies in the mixer-settlers show that the process is applicable to liquors containing high concentrations of contaminates; it yields a product within feed plant specifications with greater than 99% recovery of uranium.

A process using primary amines (particularly Primene JMT) in a kerosene diluent for the solvent extraction of thorium from sulfuric acid solutions of monazite sand was developed on a laboratory scale. The extraction products obtained are high grade and should be particularly suitable for a TBP secondary purification cycle. Solvent extraction of the monazite liquor would offer several advantages over the precipitation methods now in use.

A process using di(2-ethylhexyl)phosphoric acid in a kerosens diluent for the solvent extraction of uranium from sulfuric acid leach liquors of Western ores was developed on a laboratory scale. Preliminary studies show this process to be applicable to ore leach slurries as well as clarified liquors. The di(2-ethylhexyl)phosphoric acid is amenable to an alkaline stripping procedure, thus avoiding the handling of highly concentrated hydrochloric acid solutions which are currently employed as the stripping agent in processes wherein the active extractant is a monoalkyl phosphoric acid.

Small-scale batch tests of solvent extraction of uranium from acidified slurries of Florida Leached Zone ore were reported previously. In larger-scale continuous tests in a pulse column, good phase separation was obtained with 0.2M di(2-ethylhexyl)phosphoric acid in kerosene and a desanded slurry of 10% pulp density when the total flowrate was fairly low, i.e., about 300 gal/ft^2-hour, with slurry:organic flow ratio of 4. The amount of organic phase found entrained in the slurry raffinate was low, e.g., 0.3 to 0.7 gal/ton of slurry. (AEC Activity 5300)

Thorex Process Development - The Thorex pilot plant solvent extraction equipment was tested with solvent and water, and three batches of nonirradiated thorium feed were processed under flowsheet conditions. The dissolution, solvent extraction, and solvent recovery equipment operated satisfactorily. Preliminary evaluation of the chemical data obtained indicated that 5.0 kg of U^{233} product may be prepared for Los Alamos by February 1, 1955.

In further studies on process improvement, the following ruthenium decontamination factors were obtained for the thorium nitrate product: 30 to 40 by thorium oxalate precipitation; 100 to 130 by thorium fluoride precipitation; 15 to 20 by solvent extraction; and 510 for the first crop of crystals and 76 for the second crop by recrystallization. The thorium oxalate precipitation is recommended because of its simplicity, the adequate decontamination obtained, the absence of corrosion, and the suitability of the final form for metal production.

In laboratory-scale studies aluminum extraction into the organic phase and stripping into the thorium and U^{233} product streams were negligible. The ruthenium species present in the acid-deficient Thorex feed after the feed preparation cycle disproportionated slowly; the amount of disproportionation depended on the time the feed stood before the extraction was begun. In countercurrent experiments with the same feed and flowsheet conditions, a one-month holdup decreased the extraction column ruthenium decontamination factor from about 500 to 20.

A total of 2,324.4 g of U^{233} was eluted from the Interim-23 pilot plant isolation columns and sent to Los Alamos in twelve product containers, with a total volume of 15.9 liters. (AEC Activity 5300)

METALLURGY

Ceramics Research - BeO tubes, 0.25 in. OD, 0.170 in. ID, and 4 in. long, containing 16 wt. % UO_2 prepared at the request of the Advanced Design Section, GE-ANP, were tested at Lockland. The tubes exfoliated when heated in air because of the volume changes accompanying the phase transformation from UO_2 to U_3O_8. An attempt will be made to "fix" the UO_2 by using a solid solution of ZrO_2-UO_2 as a starting material instead of UO_2.

By means of differential thermal analysis, weight-loss curves were obtained for four samples of UO_2 powder prepared by different techniques. These data show diagnostic characteristics of the various materials and the technique may be useful as a screening test to evaluate such materials prior to fabrication.

ThO_2 (MP 3050°C) crystals were grown from a Li-Th-F melt (MP ~600°C). Specific heat determinations in the range 0-1000°C were made on Gd_2O_3 by using the precision ice calorimeter; the specific heat for Gd_2O_3 in this range is 0.2068 calories per gram per °C.

A technique was developed for growing single crystals of UO_2 in the range 40-80 microns by first growing UO_3 crystals to the desired size and then reducing these crystals in hydrogen to UO_2; 40-micron crystals of UO_2 were prepared by this method.

In waste disposal studies a ceramic body was developed from local materials from which Sr^{90} was not leached by tap water after 11 days contact. The body was prepared from a mixture of sodium carbonate, finely ground limestone, and pulverized clay added to a solution, similar in chemical composition to the "Hope Solution", to which Sr^{90} was added. (AEC Activity 5400)

Physical Metallurgy of Reactor Materials - Several one-hundred-gram batches of thorium-amalgam, produced in the Metallex equipment, were reduced to metallic thorium by a combination of pressing and pyrometallurgical treatment. The bulk of the free mercury present can be eliminated by consolidation of the amalgam in a steel die. The residual thorium-mercury complex is broken down by heating at 800 to 100°C under vacuum. Unexplainably large variations in the sintered densities (7.4 to 11.2 g/cc) are observed. The variations do not appear to be consistent with changes in consolidation pressure or sintering temperature and time.

The ductility of the sintered product was demonstrated by a 50% cold reduction in thickness of a sintered button. After annearling, this sample had a Vickers hardness of 35, which compares favorably with the hardness of iodide thorium. (AEC Activity 5400)

Fundamental Physico-Metallurgical Research - Studies in the zirconium-silver alloy system show that increasing the purity of the base zirconium from 99.85 to 99.95% increases the α-solid solubility limit from 0.1 to 0.6 at % silver. With the higher purity zirconium the width of the $(\alpha+\beta)$ region in the zirconium-indium system is substantially reduced; it now agrees with the width expected from the existence of ideal mixing entropy in both phases.

For best representation of the fiber texture type of preferred orientation it is necessary to convert the experimental pole distribution data into fiber axis distribution data. The relationship between the normalized diffraction intensity, $R(\phi)$, and the normalized axis density, $T(\phi,\beta)$, was found to be

$$R(\phi) = \frac{\int_0^{\beta_{max}} T(\phi,\beta)d\beta}{\beta_{max}} = \bar{T}(\phi).$$

These integral equations were rewritten as the series of linear equations $R_i = \frac{1}{\beta_1}(\sum_j \Delta\beta_{ij}T_j)$, and a square matrix of the 190th order was formulated, the inversion of which will give the coefficients in a series of linear equations in which R is the independent variable and T is the dependent variable. The solution of these equations will give directly the value of T at 190 points in the unit stereographic triangle. The coefficients B_{ij} were determined and the matrix will be submitted to the Mathematics Panel for inversion. (AEC Activity 5400)

Fundamental Investigation of Radiation Damage in Solids - Equipment was built which permits the extension of magnetic susceptibility measurements from room temperature to 700°K. Liquid-helium equipment was also designed and is being fabricated. The new high-temperature equipment was used to measure the magnetic susceptibility of a sample of n-type and a sample of p-type InSb (a semiconducting intermetallic compound) having 1.6×10^{16} and 1.1×10^{16} carriers/cm^3 respectively at 77°K. With the onset of intrinsic properties near 200°K, the p-type sample becomes n-type because of the higher mobility of electrons. A preliminary analysis of the magnetic data of the p-type specimen yields a forbidden-energy-gap width of 0.19 ev, a value which is in good agreement with that reported by other investigators. However, degeneracy appears to set in at a lower temperature than that predicted by theory; this indicates that the effective mass of the electrons in this compound is somewhat lower than the values found in the literature. (AEC Activity 5400)

BIOPHYSICS

Ecology (Aquatic and Terrestrial) - Tree-hole molds of *Fagus grandifolia* from southwestern Michigan and of *Tilia americana* from central Illinois were subjected to different doses of hard gamma radiation from a Co^{60} source. The control and experimental samples weighing 50 grams each were then placed in Berlese funnels for 10 days to separate the surviving arthropods. The dosages and immediate lethality are given in the following table:

Dose in roentgens	*F. grandifolia* % Lethality	*T. americana* % Lethality
5×10^4	9.9%	19.8%
7.5×10^4	12.6	29.2
1×10^5	46.6	16.5
1.25×10^5	43.7	41.0
1.5×10^5	36.3	49.1
1.75×10^5	35.4	61.8
2×10^5	74.4	45.2
3.5×10^5	92.5	73.9
5×10^5	98.0	94.4
8×10^5	97.8	---
8.6×10^5	---	98.4

Comparison of these curves with those of the tree-hole molds from the laboratory site and elsewhere indicates that probably any differences are due to biological variance. On the basis of these investigations and fundamental ecologic information, it is suggested that there is a level of radiation dose to which the tree-hole community may be exposed without long-term deleterious effects. If this level is exceeded, however, the community structure will be sufficiently damaged, via mortalities beyond recoverable limits to critical species populations, so that serious long-term effects may result. For descriptive purposes this level of radioactivity is being designated the Manifest Radiation Threshold (MRT), and is defined as that dose of ionizing radiation which, if exceeded, results eventually in the breakdown of the internal structure of the community. It is suggested that the MRT would lie somewhere between 50,000 and 150,000 roentgens of ionizing radiation. This series of experiments concludes the first phase of the ecology program. A report of these results is being prepared for the journal, "Ecology". (AEC Activity 6510)

Evaluating Hazards Associated With Radioactive Wastes - Aerosol penetration through a lead shot column, at two flow rates, was determined as a function of particle size in the 0.1 - 1.0 micron radius range. A column 3.81 cm in diameter and 89 cm long was filled with No. 11 lead shot, radius 0.15 cm. The work was done (1) to provide a rapid method for particle size determinations, and (2) to investigate the applicability of sand-filter theory to ideal columns composed of uniform spherical sand "grains". At a superficial

linear velocity of 45 cm/min upward in the column, aerosol penetration was 2% at a particle radium of 0.13 μ; increasing to 23% for particles of 0.36-μ radius, and then decreasing again to 6% for 0.90-μ particles. When the flow was downward the column penetrations were 2% at 0.13-μ, 7% at 0.36-μ, and 0.5% at 9.90-μ radius. The lower penetrations in this case are due to gravity. Under all test conditions the column showed a size for maximum penetration, confirming previous results with sand columns (ORNL-1781, p.33). Percentage aerosol penetrations through the column may now be used for a rapid determination of particle size in the 0.1 to 1.0 μ-radius range. Only two minutes are needed for a size determination, compared to 15 min to 1 hour for the gravity settling method and about 10 to 20 minutes for the diffusion battery method. (AEC Activity 6510)

RADIOISOTOPE SALES AND COSTS

Type of Transaction	November	FY to Date
Domestic Sales	96,123	472,774
Foreign Sales	2,812	24,452
Project-Cash Sales	- 21,542	28,978
Project-Transfer	11	1,458
Technical Cooperation Program Credits	0	1,510
Plant Credits	731	15,336
Civilian Defense Credits	0	105
Cancer Program Credits	22,914	101,991
*Miscellaneous	0	55
	101,049	646,659
**Income - No Production Costs incurred Subtract from total income	- 28,985	26,959
	130,033	619,700
Radioisotope Costs	90,741	409,042
Radioisotope Shipments	1,091	5,059

*Miscellaneous - Freight and Missing Parts
**Income from H^3, He^3 and B^{10} and miscellaneous items in excess of handling costs.

GROSS OPERATING COSTS

	Cost for November	FY 1955 Cost to Date
Programmatic Operating Cost - Net	2,651,713	12,426,925
Plant and Equipment Cost	296,994	1,262,824
Construction Program "H"	1,625	35,867
Work for Other Parties - Transfers	5,833	143,152
Inventory Changes	4,880*	39,977
Reimbursable Work for Other Parties	177,390	834,210
Deferred Charges	1,866	2,932
Total Laboratory Cost - Net	3,130,541	14,745,887
Estimated Cost for Next Month - Net	3,100,000	17,845,887

*Credit

[redacted]

PERSONNEL SUMMARY

	Number of Employees December, 1954	New Hires December	Terminations December
Administration	61	0	1
Operations*	120	0	1
Engineering, Shops and Mechanical	746	0	4
Laboratory and Research	1869	12	19
Protection	147	0	0
Service	368	0	0
	3311	12	25

*Includes Electrical Distribution and Steam Plant as well as the Operations Division.

A total of 773 Laboratory personnel are located in the Y-12 Area.

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Previous reports in this series:

December 1953	ORNL-1665
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June 1954	ORNL-1748
July 1954	ORNL-1760
August 1954	ORNL-1781
September 1954	ORNL-1793
October 1954	ORNL-1805
November 1954	ORNL-1824

[redacted]