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STATUS AND PROGRESS REPORT

February 1954

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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

February, 1954

F. T. Howard

and

W. H Sullivan

Date Issued: MAR 4 1954

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OAK RIDGE NATIONAL LABORATORY
STATUS AND PROGRESS REPORT

TABLE OF CONTENTS

	Page
PROGRAM 2000 - SOURCE AND FISSIONABLE MATERIALS	6
Amex Process	6
PROGRAM 3000 - WEAPONS	6
Special Electromagnetic Separations	6
PROGRAM 4000 - REACTOR DEVELOPMENT	7
HOMOGENEOUS REACTOR PROJECT	7
Homogeneous Reactor Experiment	7
Homogeneous Reactor Test	7
Engineering Investigation of Slurries	8
Chemical Investigation of Slurries	8
Solution Chemistry	8
Metallurgy	8
Fuel Reprocessing	9
AIRCRAFT NUCLEAR PROPULSION PROJECT	9
Aircraft Reactor Experiment	9
Experimental Engineering	10
Critical Experiments	10
Radiation Damage	11
ANP Reactor Chemistry	11
Corrosion Studies	12
Metallurgy--Welding	13
Metallurgy--Mechanical Properties	13
Heat Transfer Research	13
Physical Properties Research	14
Analytical Studies	14
Shielding Research	14
GENERAL REACTOR RESEARCH	15
Package Reactor	15
General Reactor Radiation Damage	15
PROGRAM 5000 - PHYSICAL RESEARCH	16
ISOTOPE PRODUCTION	16
Stable Isotope Production	16

[redacted]

[redacted]

TABLE OF CONTENTS (Continued)

Page

Program 5000 (Continued):

PHYSICS 16
High Voltage Physics 16
Scintillation Spectrometry and Instrument Development 17
Nuclear Physics, 86-Inch Cyclotron 18
The 86-Inch Cyclotron 18
Isotope Analysis Methods Laboratory 18
Mass Spectrometer Laboratory 19
The 63-Inch Cyclotron 19
Research and Development, Stable Isotopes 19

CHEMISTRY 20
Inorganic Solution Chemistry 20
Chemical Physics 20
Chemistry of the Solid State 20
Radiation Chemistry 20
Uranium Chemistry of Raw Materials 21
Volatility Studies 22
Thorex Process Development 22
Equipment Decontamination 22

METALLURGY 23
Ceramics Research 23
Physical Metallurgy of Reactor Materials 23
Fundamental Physico-Metallurgical Research 24
Fundamental Investigation of Radiation Damage in Solids . . . 25

PROGRAM 6000 - BIOLOGY AND MEDICINE 25

BIOPHYSICS 25
Waste Disposal Research 25
Radio-Chemical Analysis 26
Experimental Radiation Measurements 26
Radioactive Particulate Problem 26
Consultation and Special Problems 27

RADIOISOTOPE SALES AND COSTS 28

GROSS OPERATING COSTS 28

PERSONNEL SUMMARY 29

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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

February, 1954

The subject material of this report represents approximately half of the Laboratory's program, which is covered with some exceptions on a bimonthly schedule.

PROGRAM 2000 - SOURCE AND FISSIONABLE MATERIALS

Amex Process - The Amex process (see ORNL-1651) for recovering americium from the Hanford plutonium-reduction slag and crucible waste was successfully demonstrated. Laboratory-scale tests were made with both countercurrent batch extractions and 1/2-in. pulse columns. There was no difficulty with flooding or emulsions in the test columns. Calculations based on the results of laboratory-scale tests on sorption of the americium (from the Amex aqueous product solution) on Dowex 50W resin showed that a resin bed 4 in. in diameter and 12 in. high will hold all the americium that will be recovered from the presently accumulated Hanford wastes. (AEC Activity 2344)

PROGRAM 3000 - WEAPONS

Special Electromagnetic Separations - The estimated yields for a plutonium isotope separation run, previously reported (ORNL-1665), were verified by mass analysis of the enriched isotopes. The products were: 7.1 g of 99.39% Pu^{239}, 0.55 g of 62.8% Pu^{240}, and 0.17 g of 14.3% Pu^{241}. The 240/239 enrichment ratio was 32, and the 241/239 ratio 57. The feed material contained 140 g of 700 g/T plutonium. Approximately 6% of the Pu^{239}, which vaporized in the calutron, was collected as product. (AEC Activity 3601)

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PROGRAM 4000 - REACTOR DEVELOPMENT

HOMOGENEOUS REACTOR PROJECT

Homogeneous Reactor Experiment - The HRE experimental program was concluded in February with the completion of the internal recombination experiments. By using copper ion as an internal catalyst it was possible to operate at 1350 kw with no net gas production in the reactor core. A maximum power level of 1600 kw was attained.

The HRE equipment is now being dismantled to permit installation of equipment for the Homogeneous Reactor Test (HRT). Dismantling of the shield and reactor was postponed until the middle of March to allow short term reactor operation for the production of fission gases which are processed to give sufficient Xe^{135} for neutron absorption cross section measurements. Three 24-hour runs for this purpose were made during February. External components, not required for these operations, are already being dismantled. Excavation was completed for a temporary storage pool in which partially dismantled reactor components will be held prior to examination for evidence of corrosion.

The operating history of the HRE may be summarized as follows:

4,558 hours of fuel circulation under all conditions, critical and non-critical

2,010 hours of operation with the reactor critical

1,110 hours of operation at a power level above 100 kw

330 hours of operation at or above 1000 kw.

(AEC Activity 4103.1)

Homogeneous Reactor Test - Design and exploratory experimental work on the Homogeneous Reactor Test (HRT), which will replace the HRE, continued at a faster pace during February. (The conceptual design of the HRT was described in the January report, ORNL-1676.) A contract was let to Newport News Shipbuilding Company for development work leading to the construction of the 32-inch diameter zirconium core contained in a stainless steel-clad pressure vessel for operation at 2000 psi. Drawings for this assembly and the main heat exchangers for fuel and blanket systems are essentially complete. Orders were placed for the canned-rotor circulating pumps for both systems. Specifications were issued, or written, for the heat exchangers, circulating pumps, stainless steel piping and fittings, and stainless steel welding procedures.

The feasibility of fabricating the HRT high pressure blanket system from titanium was explored. The use of titanium is entirely feasible, but it was concluded the construction of the HRT would be delayed by several months.
(AEC Activity 4103.1)

Engineering Investigation of Slurries - Circulation studies of thorium oxide (Ames product) slurries at 250°C in 347 stainless steel loops were extended to tests in which titanium pump impellers and flow restrictors were included in the system. The titanium appeared highly resistant to attack by the slurry under the conditions of high turbulence that had been found to result in substantial attack on 347 stainless steel. (These conditions are more severe than are expected in any slurry reactor system.) In several tests, a shiny surface, having the appearance of chrome plate and apparently very resistant to abrasion, was found in the high velocity regions of the loop and pump. (AEC Activity 4109.31)

Chemical Investigation of Slurries - Thorium oxide, containing less than 10 parts per million (ppm) of nitrate, was prepared from thorium oxalate containing 0.5 to 1% nitrate by calcining the oxalate first at 370°C for 16 hr, then at 650°C for 16 hr. A product of the same purity was obtained also by heating thorium oxalate at 200° to 370°C, washing, and then calcining at 600°C. Fine-particle thorium oxide products containing less than 30 ppm of nitrate were prepared by pyrohydrolysis of the oxalates in water at 250°C in an aluminum-lined vessel. Calcination of the recovered solids at 250°, 500°, or 585°C removed organic decomposition products. The solids were then redispersed as aqueous slurries containing more than 1000 g of thorium per liter. Reautoclaving of the slurries at 250°C in stainless steel did not impair their properties.

Thorium oxycarbonate, prepared either by eluting thorium from an ion exchange column with ammonium carbonate or by metathesis of the hydroxide with carbonate, was decomposed at 500°C to yield oxide products containing less than 50 ppm of nitrate. (AEC Activity 4109.31)

Solution Chemistry - Further study of the UO_3-rich portions of the system UO_3-SO_3-H_2O indicated the existence of a basic salt, $4UO_3 \cdot SO_3 \cdot XH_2O$, at 25°C and the existence of anhydrous UO_3 at 250°C, both being formed only in limited concentration ranges. Studies in the UO_3-NiO-SO_3-H_2O system showed that, at 25°C, the addition of excess $Ni(OH)_2$ to a solution of uranyl sulfate results in quantitative precipitation of the uranium from the solution. The uranium precipitate appears to be $\alpha UO_3 \cdot H_2O$ and not nickel uranate or diuranate. The solid phase which appears at high temperatures in uranyl fluoride solutions of low concentration was identified as a solid solution, the components of which are $UO_2(OH)_2$ and $UO_2(OH)F \cdot \frac{1}{2}H_2O$. (AEC Activity 4109.31)

Metallurgy - Impact tests on titanium and Zircalloy-2 specimens indicated that exposures to low velocity, oxygen-pressurized uranyl sulfate solutions containing 5 g U/l and excess acid for periods of 529 to 693 hours at 300° to 320°C do not increase susceptibility to embrittlement or bring about other detrimental chemical or mechanical effects in these materials.

Metallographic examination of welded austenitic stainless steel pins, exposed in UO_2SO_4 solutions (300 g U/l) of high concentration at low temperatures (150°-175°C), revealed an intergranular corrosion type of penetration in the

sensitized portions of susceptible steels and preferential attack on the ferrite microconstituent in predominantly austenitic steels. "Loop" tests on electro-polished welded austenitic stainless steel pins, which were given a variety of heat treatments and tested in 40 g U/l UO_2SO_4 for 400 hours at 250°C and at flow rates of 23-26 feet per second (fps), indicated that, relative to the "as-welded" condition, only the 1000°F treatment (corresponding to commercial stress-relief annealing) does not adversely affect the dynamic corrosion resistance of the welds tested.

The combined pretreatment of chromizing (enrichment of surface in chromium) and selective oxidation in moist hydrogen enhanced the corrosion resistance of 18-8 type stainless steels in 15 g U/l UO_2SO_4 at 250°C and flow rate of 68 fps. (AEC Activity 4109.4)

Fuel Reprocessing - Preliminary solubility data for a mixture of rare-earth sulfates in 0.02 M UO_2SO_4--0.005 M H_2SO_4 indicate that the total solubility of rare earth sulfates at 300°C, the reactor operating temperature, is not greater than 0.1 g/liter. This concentration is low enough that the neutron poisoning of these elements may be kept at an acceptable level by filtering off the insoluble rare earth sulfates; however, the addition of $CaSO_4$ to effect additional removal may be desirable.

The precipitation of $CaSO_4$ at 270°C from a solution saturated with $CaSO_4$ at 40°C consistently removed 65 to 70% of the rare earths, with Eu^{155}-tracer, from dilute uranyl sulfate solution. In continuous column tests as much as 95% of the rare earths was removed by this method.

The CaF_2 process (see ORNL-1643) for removing rare-earth fission products from the uranyl sulfate fuel solution of a homogeneous reactor, having a zirconium core tank, does not look attractive at present. Fluoride contamination of CaF_2-treated fuel solution has not been decreased below 55 ppm by other than an expensive process involving evaporation to dryness and roasting of the solid UO_2SO_4 at 300°C. Preliminary data on the corrosion of zirconium by dilute uranyl sulfate solution containing fluoride indicate that 50 ppm of fluoride probably accelerates corrosion. (AEC Activity 4109.8)

AIRCRAFT NUCLEAR PROPULSION PROJECT

Aircraft Reactor Experiment - All main system piping was finished, and the remaining welding involves only miscellaneous jobs on widely different phases of the work. The heaters on the reactor and all heaters on the main circuit valves were installed. The pump coolant system was simplified by substituting direct water cooling for the rather complex dibutyl carbitol system. The driving motors in the sodium-circuit helium loop were changed from 3 to 5 horsepower to provide a margin of safety. One pump was hot-tested and is ready for installation. (AEC Activity 4401.1)

Experimental Engineering - Three out-of-pile sump pumps for in-pile loops were subjected to "cold" shake-down tests, and two were tested at high temperature. Gassing in these pumps because of improper liquid entry was corrected. An experimental pump for in-pile installation performed well with water. This pump has a gas-sealed horizontal shaft, and centrifugal action prevents leakage.

All ARE sump pumps have undergone mechanical shake-down, and each pump is being tested with hot sodium, with at least one thermal cycle. Careful inspection of every seal part and selective assembly of parts has reduced oil leakages to tolerable (very low) rates.

In bearing tests, an extreme amount of mass transfer has occurred even at iso-thermal conditions because of, in part, the variety of metals present. However, in a system comprising primarily 300 series stainless steel, chromium carbide was run successfully against titanium carbide in fluoride fuel at 1200°F.

A solution of boric and nitric acids proved useful as a solvent for cleaning fluorides from used systems. (AEC Activity 4401.1)

Critical Experiments - A critical experiment was assembled to test the designed core dimensions and core composition of the supercritical-water reactor. A 38-in., equilateral, cylindrical, aluminum tank was used for this experiment. An organic liquid ($C_5H_4O_2$), which has a hydrogen density similar to that of water in the supercritical state, served as the neutron reflector and as part of the moderator. The fissionable material, enriched uranium in an aqueous solution of UO_2F_2, was contained in 1-in.-dia stainless steel tubes. The effective loading for initial criticality was about 5 kg of U^{235} without stainless steel inserts in the core. Stainless steel was inserted in the core by loading 3/16-in.-OD tubes into the UO_2F_2 solution.

In the current experiments, the designed dimensions and composition of the core are being approached by varying the height of the organic liquid reflector and moderator (assumed to be the effective core height) as a function of the number of fuel tubes loaded at increasing steel-to-uranium ratios.

A preliminary assembly of the AC-100, air-cooled, water-moderated reactor of the General Electric Aircraft Propulsion Project was made. The fuel comprised enriched uranium metal disks placed between steel disks. The disks were mounted inside aluminum tubes 4 in. in diameter. The fuel section was 30 in. high. Thirty-seven aluminum tubes, in a pattern designed to give uniform radial power distribution, constituted the core. The core was immersed in water, which served as the neutron moderator and as an effectively infinite reflector. In order to make the system initially critical, it was necessary to deviate significantly from the prescribed loading by increasing the uranium from 26.6 to 45.6 kg and by decreasing the steel content by about one-half. A series of measurements is being made to ascertain the cause of the discrepancy between the reactivity of the experiment as designed and as actually assembled. (AEC Activity 4401.4)

Radiation Damage - The presence of large grains and intergranular cracking in a few of the Inconel capsules, containing fuel mixtures that were irradiated under static conditions in the MTR, suggested the need for careful investigation of temperature control. The temperature of the capsule wall was measured and controlled by means of thermocouples placed on the surface of the capsule. The experimental results showed that, for the worse thermocouple, a temperature error of 250°F would be obtained for the hottest portion of the inside surface of the capsule, i.e., an actual temperature of 1750°F for a thermocouple reading of 1500°F. (AEC Activity 4401.4)

ANP Reactor Chemistry - The first scheduled shipment of 120 lbs of purified fluoride salt mixtures, comprising "$NaZrF_5$" and "$NaZrF_5$" + UF_4, was sent to Pratt and Whitney. The next shipment, totaling 900 lb, should be ready by April 1.

Construction was started on the new, small-scale fluoride-mixture production facility in Building 9201-3. The remodeling of the facility in Building 9928 to permis safe operation with BeF_2 was started. These facilities are expected to be completed in April.

Evidence has accumulated that the crystalline compound to which the formula $NaZrF_5$ was previously ascribed actually has a composition very close to 47 mole % ZrF_4 and 53 mole % NaF with the probable formula being $Na_9Zr_8F_{41}$. The evidence for this is the following: (1) Homogeneous crystalline material with uniform optical and x-ray diffraction properties separated from a large, slowly cooled mass gives, by analysis, the approximate composition 47% ZrF_4. (2) Slowly cooled samples with compositions between 47 and 57% ZrF_4 consist of two phases. One of these is the 47 mole % ZrF_4 compound; the other is a compound previously recognized and believed to contain 57 mole % ZrF_4. (3) Slowly cooled materials with compositions between 40% and 47% ZrF_4 consist of a single crystalline phase with optical properties and x-ray diffraction patterns varying linearly between those of the end-member phases at 40% and at 47%.

Fuels, containing UF_3 with essentially no UF_4, were prepared for corrosion testing by each of two similar reactions:

$$(1)\quad 3ZrF_4 + 4U^o \rightleftharpoons 3Zr + 4UF_3$$

and

$$(2)\quad 3UF_4 + U^o \rightleftharpoons 4UF_3$$

The reaction, in each case, was conducted at about 800°C in a fused mixture of NaF and ZrF_4. Preliminary tests of corrosion by such UF_3-bearing mixtures in thermal convection loops (see Corrosion Studies) were most encouraging. It should be noted, however, that only 3 wt % of trivalent uranium was soluble in this mixture at 600°C.

Experimental study of the reaction of Ni^o with FeF_3, using molten $NaZrF_5$ as the solvent, showed the reaction to be

$$Ni^{o} + 2FeF_3 \rightleftharpoons 2FeF_2 + NiF_2 .$$

The reaction proceeded so nearly to completion that the equilibrium constant could not be determined reliably from analytical data.

Additional studies of the reduction of NiF_2 and FeF_2 in molten $NaZrF_5$ by hydrogen were conducted over the temperature interval 600 to 800°C. The reaction of NiF_2 with hydrogen was strongly catalyzed by Ni^{o}; the apparent first-order reaction rate which prevailed during a large portion of the process probably resulted from the fact that the rate-controlling step was the stripping of HF from the melt. In similar reductions of FeF_2 by hydrogen, Ni^{o} again acted as a catalyst but was quickly poisoned by the Fe^{o} produced; stripping of HF was more rapid than the chemical reaction, and an apparent one-half order process resulted. (AEC Activity 4401.5)

Corrosion Studies - An Inconel loop was operated with a zirconium-base fuel in which UF_3 was used instead of UF_4. No subsurface voids of inter-granular penetration were found after 500 hr at 1500°F. A nonmetallic deposit was found that was thought to have been formed during cooling.

A quartz thermal-convection loop, with inserts of beryllium in the hot- and cold-leg sections, was operated with a hot-leg temperature of 815°C and a cold-leg temperature of 550°C to determine the mass-transfer characteristics of beryllium in molten lead. Operation was terminated after 456 hr because of plug formation. X-ray examination of the mass-transferred material showed the presence of beryllium oxide and free silicon. It is therefore concluded that the beryllium reduced the quartz.

Samples of Chromalloyed type 304 stainless steel and Chromalloyed type 1035 plain carbon steel were tested in sodium and in alloys of lead, cadmium, and tin. Examination showed the "as-received" sample of type 304 stainless steel to have a 0.0001- to 0.0002-in. layer of chromium, with no diffusion layer beneath it, and the chromium layer was cracked in many places. The as-received type 1035 plain carbon steel was found to have a 0.0005- to 0.001-in. chromium layer, with a 0.001-in. diffusion zone beneath it. The chromium layer was brittle and cracked in many places. After testing at 850°C for 100 hr, both materials showed intergranular attack to an appreciable depth. Since these samples were attacked only in the regions where there were flaws in the chromium layer, it is thought that a chromium layer would afford good corrosion protection if it were free of cracks.

Tests of systems containing more than one metal at 815°C for 100 hr in the seesaw furnace revealed that when Inconel and type 316 stainless steel are exposed together in the fluoride mixture $NaF-ZrF_4-UF_4$, the steel is attacked more heavily than the Inconel even when the steel is in the cold zone. The amount of material mass-transferred from one alloy to the other was quite small.

Special high-density graphite crucibles were tested with the fluoride mixture $NaF-ZrF_4-UF_4$ and with sodium at 815°C for 100 hours. The sodium penetrated

the walls of the crucibles and caused them to crack and crumble. The crucible containing $NaF-ZrF_4-UF_4$ sustained a weight loss of 0.012%, and the surfaces in contact with the fluoride had an etched appearance. (AEC Activity 4401.5)

Metallurgy--Welding - Cone-arc welding experiments demonstrated the feasibility of joining 187-mil-OD, 25-mil-wall Inconel tubing to 125-mil Inconel headers. Several headers containing 16 tubes each were welded, and the conditions necessary for obtaining consistent results were established.

The cone-arc welding machine was made more versatile by the installation of an attachment on the nozzle to facilitate arc movement around the tube periphery. This attachment will be useful for the welding of large tubes, i.e., 3/8 to 1/2 in. ID. Experiments indicate that it will also minimize some of the difficulties encountered with cone-arc welding of smaller diameter tubing; it will reduce the tendency of the arc to concentrate at one point on the tube periphery and, consequently, to overhead the tube wall in that vicinity. (AEC Activity 4401.5)

Metallurgy--Mechanical Properties - The effect of the annealing temperatures on creep behavior of Inconel specimens was determined. Examination of the creep curves for fine-grained Inconel showed a slower creep rate for the first 200 hr than that for coarse-grained Inconel. After 200 hr, a transition occurs, and a new and faster creep rate was established and maintained until fracture. Thus, up to 700 hr of test life and up to approximately 4.5% elongation, the fine-grained material had superior creep properties, but after 700 hr, the coarse-grained material had more resistance to plastic flow. (AEC Activity 4401.5)

Heat Transfer Research - The enthalpy and heat capacity of $NaF-ZrF_4$ (50-50 mole %) were determined in the liquid and solid states. The results are represented as follows:

Solid (216 to 490°C)

$$H_T(\text{solid}) - H_{0^\circ C}(\text{solid}) = 0.19\ T$$

$$c_p = 0.19 \pm 0.01$$

Liquid (552 to 875°C)

$$H_T(\text{liquid}) - H_{0^\circ C}(\text{solid}) = -13 + 0.33\ T$$

$$c_p = 0.33 \pm 0.02$$

where H is the enthalpy in cal/g, T is the temperature in °C, and c_p is the heat capacity in cal/g °C. The heat of fusion was about 59 cal/g. The thermal conductivity of NaF-KF-LiF (11.5-42.0-46.5 mole %) in the solid state was found to be about 2.8 Btu/hr-ft-°F.

Additional forced-convection heat transfer experiments were conducted in pipe systems which carry liquids containing volume heat sources; the heat was generated electrically. The experimental system was modified to facilitate making laminar flow experiments. The laminar flow data fell about 30% below the values predicted by previously developed theory; this deviation may exist because of free convection or entrance phenomena. The experimental turbulent flow data reported previously fell within ± 30 percent of the predicted values. (AEC Activity 4401.5)

Physical Properties Research - The density of $NaF-ZrF_4-UF_4$ (62.5-12.5-25 mole %), a mixture to be used in an in-pile system, was found to be,

$$\rho(g/cm^3) = 4.836 - 0.00111T \quad (650^\circ C < T < 900^\circ C).$$

The viscosity for this material varied from about 17.1 centipoises at 670°C to about 8.3 centipoises at 900°C. (AEC Activity 4401.5)

Analytical Studies - Hexavalent uranium was tested as an oxidant for trivalent uranium in connection with studies on the determination of UF_3 in the presence of UF_4. Various acidic media for the reaction $2U^{3+} + UO_2^{++} + 4H^+ \longrightarrow 3U^{4+} + 2H_2O$ were used. In all cases, oxidation was attributed partly to the hydrogen ion in addition to the uranyl ion. For example, in 1 N oxalic acid, only 50% of the UF_3 was oxidized by hexavalent uranium.

Iodine in methanol-water solutions was also tried for this one-step oxidation. By increasing the concentration of methanol, the oxidation potential of iodine was lowered to the extent that the following reaction took place:

$$U^{3+} + 1/2\ I_2 \longrightarrow U^{4+} + I^-$$

Based on this reaction, quantitative results were obtained in an 80% methanol medium.

Uranium trifluoride and uranium tetrafluoride were converted to UCl_3 and UCl_4, respectivly, by fusion with $NaAlCl_4$ to take advantage of the greater solubility of the chlorides. The chlorides were extracted with a mixture of acetone and acetylacetone, which formed the corresponding acetylacetonates. By titrating the hydrogen ion liberated upon the reaction of U^{3+} or U^{4+} with acetylacetone, it was shown that 3 moles of acetylacetone combined with one mole of trivalent uranium and that 4 moles combined with one mole of tetravalent uranium. These chelate compounds are not particularly stable, however, as shown by studies of their ultraviolet spectra. The molecular extinction coefficients at maximum absorbancy (272 mμ) were: $U(acetylacetone)_3$, 515,000; and $U(acetylacetone)_4$, 107,000. (AEC Activity 4401.5)

Shielding Research - Acceptance tests were run on the hoists for the Tower Shielding Facility with a maximum load of 85 tons in the central position. With the exception of minor modifications for no-load conditions, the hoists were approved by the Laboratory. It is expected that the facility will begin operation during the month of March.

Measurements were made on the two types of recoil-proton fast-neutron dosimeters for use at the Bulk Shielding Facility to obtain intercalibration factors. One dosimeter has two chambers filled with 2 atm of methane, and the other has three chambers filled with 1 atm of ethylene. The measurements of neutron flux showed a constant relationship between the two dosimeters. Measurements on a third type, a three-chamber dosimeter with 2 atm of methane agreed with those on the first two types.

Testing of the front section of the mockup of the shield which had been designed for the G-E R-1 reactor was completed at the Bulk Shielding Facility. As was reported for the rear section of the shield, the peaks of radiation occurred beyond the voids. This mockup will now be moved to the Tower Shielding Facility for complete testing, including air scattering and crew-box attenuation.

The Fermi age of a neutron from a point source in $NaZrF_5$ ($\rho = 4$ g/cm^3) was calculated to be 433 cm^2. The calculation was based on the assumption that the Fermi age equation holds for the material. A 25% error was estimated.

A Lid Tank measurement of the neutron-removal cross section of B_2O_3 gave a value of 4.4 ± 0.14 barns/molecule. This value is in fair agreement with other measurements of oxygen and boron. (AEC Activity 4401.7)

GENERAL REACTOR RESEARCH

Package Reactor - The package reactor specifications are being prepared for submission to private contractors for a lumped-sum bid. Several prospective bidders are now reviewing the preliminary specifications. A full scale mock-up of the control rod drive is being assembled and will be ready for testing shortly by the American Machine and Foundry Co. It will be operated at elevated temperatures and pressures to simulate actual reactor conditions. The reactor calculations are being reviewed and refined. Methods of achieving a greater degree of moderation in the reactor are being considered. (AEC Activity 4510)

General Reactor Radiation Damage - The electrical resistivity of Karma alloy (approx. 73% Ni, 20% Cr, plus Al and Fe; manufactured by Driver-Harris Co.) was shown to increase up to 2% during 600 hr of irradiation in a beam hole of the LITR at a temperature of 230°F. The wire was tested because it has been considered for use in bonded-wire strain gages for inpile measurements. The large change in resistivity is attributed to the radiation-induced ordering of the metal lattice. (AEC Activity 4540)

PROGRAM 5000 - PHYSICAL RESEARCH

ISOTOPE PRODUCTION

Stable Isotope Production - The processing of hafnium was completed in February with the production of 26.8 g of separated hafnium isotopes (masses 174, 176, 177, 178, 179, 180). The processing of hafnium from $HfCl_4$ charge material was characterized by low ion output, and further complicated by the deposition of hafnium metal inside the arc chamber. The cause and prevention of this phenomenon have not been established.

Separation of the isotopes of zirconium (masses 90, 91, 92, 94, 96) is now in progress. Calutron operation with $ZrCl_4$ charge is strikingly similar to operation with $HfCl_4$.

The separation of potassium, using charge material that was enriched from 0.01% to 0.02% K^{40} by pile irradiation, was completed near the end of February. An estimated 20 mg of the K^{40} isotope was obtained, and results from a single mass analysis indicate that much of this product will have an isotopic purity of about 40%.

Enriched Ru^{96} (250 mg), Gd^{158} (6.35 g), Gd^{160} (7.625 g), Ir^{191} (975 mg), and Ir^{193} (1.15 g) were added to the inventory of isotopes for the first time. New lots of Ti^{46}, Ti^{47}, Ti^{49}, Ti^{50}, La^{138}, La^{139}, Sm^{154}, and Hf^{180} were also placed in the inventory as replenishments. Continued progress was made in the purification of the rare earth elements by extraction with tributyl phosphate, emphasis at the present being on the separation of europium and dysprosium from the other rare earths.

Forty-one shipments involving 34 different isotopes of 21 elements were made. Present shipments are 25% higher than for the same months last year.

During the years 1946 to 1953, inclusive, approximately two thousand shipments of enriched isotopes were made, at an average rate of five shipments per week. These isotopes have resulted in over four hundred articles, letters-to-the-editor, and abstracts of papers published in Physical Review during this period. (AEC Activity 5121)

PHYSICS

High Voltage Physics - Study of the Li^6 (d,t) reaction was extended to the 4-Mev energy range by using the 5-Mev ORNL Van de Graaff accelerator. The cross section shows a further rise to about 0.29 barn (see ORNL-1676). The Li^7(d,t) yield studies were also extended to 4 Mev, but are discrepant with the earlier data near 2.5 Mev. The need for further experimental clarification of the discrepancy is being considered. A paper on the Li^7(n,t) reaction has been accepted for publication in Science.

Yields and spectra of gamma rays produced by 2.8 to 4.0 Mev protons on B^{11} were measured with a 3" x 3" NaI crystal. The capture radiation to the ground

state in C^{12} is resonant at 3.55 Mev ($\Gamma \approx 200$ kev), whereas the cascade transition to the 4.43-Mev level in C^{12} is resonant at 3.17 Mev ($\Gamma \approx 80$ kev) and 3.55 Mev. A 2-Mev gamma ray from inelastic scattering exhibits a known resonance at 2.8 Mev and a new level at 3.55-Mev bombarding energy.

A new electrostatic lens was installed in the 2.5-Mev Van de Graaff. Ions from the rf ion source are focussed by a saddle-shaped field into a field region 7 cm below the ion source exit canal. The ions are then accelerated into a long field-free region and thence into the accelerator tube. The focusing characteristics are improved over those of the old lens; however, a large fraction of the beam continues to be unsatisfactorily focused.

The differential cross section for elastic scattering of neutrons from nitrogen was measured at 0.80, 1.28, and 1.54 Mev. Neutrons produced by the $T(p,n)He^3$ reaction were elastically scattered from a cylindrical sample of LiN_3 and detected by a propane recoil counter. Differential cross sections were measured over the region of cos φ (center-of-mass angle) from + 0.9 to -0.7. For cos $\varphi < 0.5$ the distributions are essentially isotropic, in agreement with results previously found by observing nitrogen recoil energies in a counter. At 1.28 and 1.54 Mev the differential neutron measurements show an increased cross section for cos $\varphi \geqslant 0.5$.

A careful study of Be^8 and Be^9 by means of the $Li^6(He^3,p)Be^8$ and $Li^7(He^3,p)Be^9$ reactions was made possible by use of the 20-channel pulse height analyzer. The Be^8 state at 3 Mev showed, with 2% error in counting statistics, no evidence of structure. The $Li^7(He^3,p)Be^9$ reaction gave evidence in Be^9 of three rather sharp levels at 2.04, 2.53, and 3.18-Mev excitation and a broad level at 4.86-Mev excitation. (AEC Activity 5211)

Scintillation Spectrometry and Instrument Development - Investigations of gamma-ray attenuation in thick absorbers were made in order to obtain the degraded spectrum through the absorber and the broad beam attenuation coefficients. A new method was developed for analysis of the smearing of a continuous distribution that ends abruptly. With this method of analysis, values of the broad beam attenuation coefficients were obtained which are within 0.25% of the values given, for an attenuation of 5×10^6 or greater, by the National Bureau of Standards for lead and aluminum. Moderately smooth photon distributions for the degraded spectra in these materials were also obtained. Attempts to use the new method of analysis for previous absorption data on NaI still yielded values for the attenuation coefficients which were slightly lower than those listed by the National Bureau of Standards.

A study was made of the gamma-radiation in K^{42} by means of an anti-coincidence absorption technique which eliminated the inner and external Bremsstrahlung radiation associated with the decay. A previously unreported gamma ray with an energy of 0.309 Mev was found.

Investigations of the conduction current in various types of semi-conductor diodes were continued in a search for the diodes best suited for high speed

coincidence circuits, pulse lengtheners, and other special electronic circuits. The current-voltage characteristics of the diodes were investigated during the intervals following the sudden application of a moderate current (10 ma) and after the sudden removal of such a current. Heavy conduction currents that appeared to be caused by a high concentration of holes were observed, and a search was made for diodes with a minimum of such conduction. The most satisfactory diodes found up to the present are the 1N26 mixer crystals (Sylvania); other mixer crystals give a fairly good performance. Most of the dc restorer type of diodes (especially those of recent construction) are very poor.

The performance of the A-7 linear amplifier proved to be very satisfactory in actual use, and the final engineering design was started. This is an anti-overload linear amplifier for use in spectrometry. It has extremely good differential linearity as well as exceptional baseline stability when presented with violently fluctuating counting rates. (AEC Activity 5211)

Nuclear Physics, 86-Inch Cyclotron - Measurements of angular distributions of fragments from 22-Mev proton-induced fission in U^{238} were completed and similar measurements in U^{235} were initiated. Cross sections and excitation functions for several (p,n), (p,2n), (p,pn), (p,2p), (p,He^3), and (p,f) reactions were measured. Two new isotopes were identified. A 1.7-m activity was produced by the (n,p) reaction on enriched Fe^{57}. It was identified as Mn^{57} by chemical separation and by both cross section and gamma spectrum measurements. The Mn^{57} decays principally by the emission of a 2.5-Mev negatron; a weak 690-kv gamma ray was also measured. A previous assignment of a 7.2-day activity to Mn^{57} is very probably in error. (AEC Activity 5211)

The 86-Inch Cyclotron - Carrier-free Fe^{55} was produced at an estimated rate of 10 mC/mah by the (p,n) reaction on manganese. The target was prepared by electroplating manganese directly on the aluminum target base. During a seven hour period the proton current on the 6" x 10" target averaged 1170 micro-amperes.

A 6" x 10" magnesium target, water-cooled at 100 gpm (50 fps), was bombarded with 1200μa of protons (25 kw) for a period of over 10 hours. Carrier-free sodium-22 was produced by both Mg^{25} (p,α) and Mg^{26}(p,αn) reactions. Since these two isotopes together have an abundance of only about 20% the yield of Na^{22} was estimated at only 0.4 mC/mah. (AEC Activity 5261)

Isotope Analysis Methods Laboratory - New nuclear magnetic resonance frequency ratios were obtained as follows: Li^6/D = 0.958638, Li^7/Na^{23} = 1.469225, Na^{23}/D = 1.723167, Al^{27}/Na^{23} = 0.985055, P^{31}/Li^7 = 1.041611, P^{31}/Na^{23} = 1.530366, Ga^{69}/Na^{23} = 0.907349, Br^{81}/Na^{23} = 1.020965, Rb^{85}/D_2 = 0.628985, and Tl^{205}/Tl^{203} = 1.009816. Average precision is about four parts per million.

A device was developed for measuring optical densities, d, from 0 to 4, for red light and 0 to 6 for green light. (Optical density, $d = \log_{10} 1/T$, where 0 = opacity and T = transmission).

Spectrochemical tests on the Echelle-Littrow spectrograph showed that its sensitivity, for Be, Ca, Cu, Mg, and Ni in uranium samples burned directly in the arc, is 1 ppm or better.

In the x-ray fluorescence analysis for high concentrations of constituent rare earths, a precision of about $\pm$ 5 percent was obtained. (AEC Activity 5261)

Mass Spectrometer Laboratory - Iridium was found to be very useful as a hot cathode for vaporizing solid materials in the ion source of a mass spectrometer. Strips of iridium foil 0.003" x 0.060" in cross section were spot welded to tantalum support legs and heated with currents up to 12 amperes. An outstanding feature of the iridium filaments is that they can be heated to over 2000°C in air without oxidizing appreciably. This enables one to sinter a few tenths of a milligram of metal or oxide powder on the filament while observing it under a low power microscope. The filament is then transferred to the ion source of the mass spectrometer.

With the iridium cathode it is possible to analyze the more refractor materials and to analyze samples directly, without preliminary chemical processing. The following materials were analyzed isotopically in this manner: U_3O_8, UO_2, Pd, Pt, Cr_2O_3, HfO, Rh, and NiO. Data, never before satisfactorily available, were obtained on Pt, Rh, and NiO.

It was also demonstrated that two or three different samples can be loaded on the iridium filament in descending order of their fusing temperatures, then analyzed in the mass spectrometer in reverse order as the temperature of the filament is slowly increased. This technique greatly reduces the instrument time required per sample. (AEC Activity 5261)

The 63-Inch Cyclotron - The range-energy relations for nitrogen ions in nickel and aluminum were measured. Improved measurements on the charge of nitrogen ions as a function of their velocity confirm the previously reported (ORNL-1665) equilibrium charge of 6.2 for 28-Mev nitrogen ions emerging from a very thin nickel foil ($\sim$0.5 mg/cm^2). (AEC Activity 5261)

Research and Development, Stable Isotopes - Preliminary experiments were made in a new approach to the problem of collecting isotopes of the inert gases in a calutron. The proposed ion collector consists of a rotating drum dipping in oil. Since the solubility of inert gases in certain liquids is appreciable, this type of collector offers the possibility of operating on an almost continuous basis with relatively high retention of the gas. Oils have been found which appear to have suitable characteristics.

Highly enriched Li^6 metal ($\sim$93%) was prepared for use as target material in high voltage accelerators.

A satisfactory technique was developed for casting large quantities of lithium metal in various forms needed for shielding experiments. Oxidation is pre-

vented by flooding the surface of the molten metal with mineral oil. Up to fifty pounds can be cast. (AEC Activity 5261)

CHEMISTRY

Inorganic Solution Chemistry - An anion exchange method for the separation of lithium from other alkali metals was developed. It was found that lithium could be adsorbed from basic solutions (pH 10) of ethylenediaminetetracetic acid (Versene) by the strong-base anion-exchange resin Dowex-1. The adsorption is large enough to permit separation of Li from Na, K, and Cs. The lithium can be removed from the columns at a lower pH.

Ultracentrifugation of Bi(III) in perchlorate solutions was carried out under conditions where earlier investigators postulated relatively high molecular weight hydrolytic polymers. The ultracentrifugation results indicated that, in the initial steps of the hydrolysis of bismuth, relatively low molecular weight polymers are formed (degree of polymerization between 4 and 6) and that in this respect Bi(III) is thus similar to, though slightly more polymerized than, Zr(IV) and Hf(IV). (AEC Activity 5311)

Chemical Physics - The presence of atomic hydrogen in H_2SO_4, H_3PO_4, and $HClO_4$, irradiated at liquid nitrogen temperature with cobalt gamma rays, was established by paramagnetic resonance measurements.

Zeeman studies on the pure quadrupole spectra of barium chlorate are near completion. No asymmetry in the field gradient tensor was found. (AEC Activity 5311)

Chemistry of the Solid State - In the first few experiments on the system CsF-Cs the solubility of solid cesium fluoride in liquid cesium metal was found to increase rapidly from about 11 mole % at 450°C to 23 mole % at 525°C and 36 mole % at 600°C. This result means that the solubility of cesium fluoride in cesium metal is more than ten times that of potassium fluoride in potassium metal and is of the order of one hundred times that of sodium fluoride in sodium metal. Such a result is strongly indicative of complete miscibility of the salt and the metal in the liquid state at temperatures not far above the melting point of the salt, 680°C. Predictions based on the combined favorable influence of low lattice energy of the salt and a large metal-atom:halide-ion size ratio were thus confirmed. (AEC Activity 5311)

Radiation Chemistry - In attempting to establish a general theory of the radiolysis of water and aqueous systems, yields of the earliest chemically detectable products (H, OH, H_2, H_2O_2), formed in the primary reactions, are important. Yields of hydrogen gas were found to be the same for many solutions in which the solute prevented secondary reactions of the hydrogen. The important secondary reaction is $H_2 + OH \longrightarrow H_2O + H$.

The yield of H_2 per 100 ev was 0.37 in 0.8N H_2SO_4 and in 0.8N H_2SO_4 solutions containing oxygen, 10^{-3} M H_2O_2 + O_2, 2 x 10^{-3} M $FeSO_4$ + O_2, and 10^{-3} M KBr or 4 x 10^{-4} M $Ce(SO_4)_2$ both in the presence and absence of oxygen.

At pH values 2 to 7, the yield of H_2 per 100 ev was 0.40 for solutions containing 10^{-6} M to 10^{-1} M KBr both in the presence and absence of dissolved oxygen. In oxygen-saturated pure water and in oxygen-saturated H_2SO_4 solutions at pH2, the H_2 yields were 0.21 and 0.23; these yields along with H_2O_2 measurements indicated that some of the H_2 reacted with O_2 to form H_2O_2.

The yields of hydrogen peroxide in oxygen-saturated KBr solutions, in 10^{-2} N and in 0.8 N H_2SO_4, decreased with increasing KBr concentration, and at concentrations above 10^{-1} M, free Br_2 appeared as part of the oxidant. Extrapolation of the peroxide yield to that at zero KBr concentration allowed evaluation of the yield of H_2O_2 formed in the primary decomposition of water. The extrapolated yields of H_2O_2 per 100 ev were 0.78 in 0.8N H_2SO_4 and 0.75 in 10^{-2} N H_2SO_4 solution.

These results were interpreted to indicate that Br^-, even at low concentration, can react with OH in regions of high energy density along the particle track, thereby preventing some H_2O_2 formation. (AEC Activity 5311)

Uranium Chemistry of Raw Materials - Extractions of uranium (VI) from acidic sulfate solutions with long chain amines in a hydrocarbon diluent have been described previously. These studies have now been extended to nitrate, chloride, and phosphate solutions. In the chloride and nitrate systems, extraction coefficients were quite low until the total anion concentration was raised above 5 M. In the phosphate systems, the extraction coefficients were fairly high at low levels of phosphate but fell off rapidly when the phosphate concentration exceeded 2 M. These results are qualitatively similar to those obtained in the sorption of uranium with anion exchange resins.

The extraction of uranium from acidified, dried Western ores in a non-aqueous system was achieved by using long chain amines in hydrocarbon diluent. For the best results the amine was converted to the amine salt, prior to extraction, by contact with dilute sulfuric acid solution. Uranium recoveries of 90-96% were obtained from the Marysvale and Uravan ores by this technique. No measurements of amine and diluent loss have been made yet to determine the practicability of the process.

From an ashed sample of Wyoming sub-bituminous coal, about 50% of the uranium present was extracted by treatment of an aqueous slurry with a mixture of sulfur dioxide, carbon dioxide, and air. Essentially all of the sulfur dioxide was absorbed and oxidized to sulfuric acid. The total amount of sulfur dioxide, and its concentration in the gas stream, corresponded to the quantity expected in the flue gas from a furnace burning the uraniferous coal. If similar results are attainable with actual flue gas, an important saving can be made in reagent costs for uranium recovery. (AEC Activity 5361)

Volatility Studies - A cost study was made of a hypothetical chlorine trifluoride processing plant for recovering uranium and plutonium from 3 metric tons per day of uranium containing 550 g of plutonium per ton. The recovery cost, assigning all charges to plutonium, was $14.50 per gram. With a direct-maintenance Purex plant of similar capacity, the plutonium recovery cost was estimated to be $20.00 per gram (including the cost of converting uranyl nitrate to UF_6). This small differential in favor of the CTF process is not believed sufficient to justify an intensive development program of volatility processes for recovering plutonium from irradiated uranium.

Scouting runs were made to determine the feasibility of recovering uranium from irradiated zirconium- or aluminum-uranium alloys by direct hydrofluorination of the metal in molten fluoride, with subsequent fluorination to UF_6 and volatilization of the UF_6. The dissolution rate of zirconium, 7 mils/hr, in the molten $NaF-ZrF_4$ (50-50 mole %), through which HF was bubbled at 575°C, was rapid enough to justify further study. In the absence of HF, the rate was 3 mils/hr, showing that part of the reaction was between the metal and the salt. A black precipitate that formed during the process dissolved in excess HF. Faster penetration rates were obtained with excess molten KHF_2 but the ZrF_4 formed was insoluble in the melt, and uranium could not be removed easily from the thick paste that resulted. (AEC Activity 5361)

Thorex Process Development - In further batch studies on protactinium recovery, 90 to 95% of the protactinium was removed from a concentrated extraction column aqueous effluent by adsorption on a chromate precipitate. The aqueous solution was concentrated by evaporation by a factor of 3.3, adjusted to 0.1 to 0.4 M chromate ion, and digested at about 100°C for 2 hr; the precipitate was separated by centrifugation or filtration. Since the separated precipitate is about 5% of the concentrated extraction column effluent volume, a concentration factor of 50 to 60 is realized for the protactinium. The precipitate can be dissolved or slurried with acid (nitric or hydrochloric). The protactinium can be purified by a diisobutyl carbinol extraction process (ORNL-1518, pp. 55-57), or, after a decay period, isotopically pure U^{233} can be obtained by a process similar to the "Interim-23" process.

"Acid-deficient" feed to the extraction column has, in laboratory-scale equipment, consistently permitted greater variations in solution composition and given greater decontamination factors than acid feed. (AEC Activity 5361)

Equipment Decontamination - Gamma-ray scintillation spectrometer analyses of type 347 stainless steel in various stages of decontamination established the fact that niobium is the principal gamma-emitting contaminant of stainless steel that has been in contact with a nitrate solution of mixed fission products. Niobium contamination penetrates the metal surface less than 0.1 mil. Stainless steel that is corroded during contamination is difficult to decontaminate. The most effective decontaminating reagent for either corroded or noncorroded stainless steel was sodium hydroxide--sodium tartrate--hydrogen peroxide solution, used after a preliminary wash with nitric acid. This

reagent removes 75 to 80% as much contamination in 5 min as it will remove in several days.

A method was developed for possible use in decontaminating the homogeneous reactor. Surfaces are descaled with a single application of 1.2 M HCl--1.8 M H_2SO_4 containing 0.16 vol % alkyl pyridines HF, a corrosion inhibitor. In tests with contaminated plaques, the decontamination factor was 10^3 which was the maximum obtainable with the activity levels used. The addition of 0.2 M H_2O_2 to the decontaminating solution increased the rate of scale removal but also increased the corrosion rate from 30 to 600 mils/year, an excessive amount. The solution containing peroxide was used successfully to decontaminate a homogeneous reactor test loop. (AEC Activity 5361)

METALLURGY

Ceramics Research - Samarium oxide was found to produce a well sintered, strong ceramic body when it was first fired to 1500°C, pressed into shape, and refired at 1500°C. Densities thus obtained were 99% of theoretical and the body was nearly impervious (0.5% porosity). The thermal expansion coefficient was found to be 9.3×10^{-6} in/in-°C. There was no change in structure up to 1130°C, as shown by thermal expansion or differential thermal analyses. A reported phase change above 1150°C was tentatively verified by x-ray. Samarium oxide disintegrates in boiling water and reacts with BeO at ~1520°C and with Al_2O_3 at ~1600°C.

Gadolinium oxide (mol wt 351.8) has a slightly lower melting point (1950°C ± 20°C) than Sm_2O_3 (mol wt 348.8 and m p 2300° ± 20°C). It fires to a well-sintered ceramic if it is precalcined at 1350 to 1500°C. The density of the fused material was measured as 7.51, which is higher than the reported theoretical density (7.41) and due probably to the presence of 2% terbium. The thermal expansion coefficient of Gd_2O_3 ceramics was observed to be 10.3×10^{-6} in/in-°C. The cubic form was found, by thermal expansion behavior and differential thermal analysis, to be stable to 1000°C, as reported in the literature. Gadolinium oxide ceramics were found to be unaffected by boiling water. Their machining properties are good. (AEC Activity 5411)

Physical Metallurgy of Reactor Materials - Experiments aimed toward improving aqueous chemical dissolution of spent stainless steel-uranium oxide fuel elements by prior metallurgical treatment are continuing. A fuel recovery process involving carburization was shown by laboratory tests to be feasible. The technique of carburizing such elements followed by sensitizing heat treatment allows chemical dissolution to be accomplished with slightly acidic copper sulfate solutions. The sulfate solution produces disintegration of the fuel element without significant dissolution of the uranium oxide. The sulfate solution may then be discarded, and the stainless steel and uranium can be taken into solution with nitric acid and fed into standard uranium extraction columns. A study of proposed flow sheets indicates that this technique offers real economies in that the volume of solution and nitric

acid required are very much smaller than those contamplated in present processes, and the corrosion problem in the dissolvers is minimized.

Further testing of the UO_2-aluminum system, for enriched-uranium fuel elements, of interest to Savannah River and Hanford showed that reduction of UO_2 by aluminum occurs in a matter of hours at 600°C and seconds at 1000°C. (AEC Activity 5411)

Fundamental Physico-Metallurgical Research - Further studies of the phase diagrams of the Group IVa elements gave the following new results:

(1) In the Zr-Ag system, the composition of the eutectoid reaction $\beta \longrightarrow \alpha + \gamma(Zr_2Ag)$ appears to be closer to 3.6 atom percent silver rather than 2.8 atom percent as previously reported.

(2) In the Zr-In system, refinement of the $\alpha/\alpha+\gamma$ boundary demonstrated that the solubility of indium in α zirconium follows the usual relationship $\log S = -K/T$. Microstructures in the material, as cast, indicate that the composition of the eutectic relation $L \longrightarrow \beta + \gamma$ (Zr_3In) is 18-19 atom percent indium. A eutectic temperature of 1350-1450°C is suggested by the shape of the phase boundaries at lower temperatures.

(3) In the Zr-Pb system, the general form of the phase diagram near the $\alpha \rightleftharpoons \beta$ transformation in pure zirconium was established. The α phase is formed by the peritectoid reaction β(4.5 atomic percent Pb) $+ \gamma \longrightarrow \alpha$(5.9 atom percent Pb) at 900°C. Lead is more soluble than tin in both α and β zirconium. This result might be expected from the atomic size differences, for the lead and zirconium atoms are more nearly the same size than are tin and zirconium, if allowance is made for incomplete ionization of the lead.

The proposed relationship between the fiber axis distribution and pole distribution for fiber textures has now been confirmed experimentally. In order to obtain a specimen having a homogeneous and ideal fiber texture, a 0.19 in. diameter spherical specimen was machined from the center of a 0.89 in. diameter extruded aluminum rod. An analysis of the x-ray diffraction data obtained in the usual manner indicated that the preferred directions of the fiber axes were $[111]$ and $[100]$, that the axid distribution fell off rapidly from these positions but never reached zero along the $[110]$ zone, and that the ratio $[111]:[100]$ was 9:10. This fiber axis distribution was shown to reproduce the observed pole distribution for all diffracting planes to within a few percent.

In order to obtain quantitative preferred orientation data for rolled sheets and plates, it has been proposed to study the axis distribution by using the rolling, transverse, and normal directions as "fiber axes". Axis distribution data were obtained for a spherical diffraction specimen of laminated

brass sheet. Preliminary analyses of these data indicate that the proposed method gives quantitatively the information given only qualitatively by the pole figure method. (AEC Activity 5411)

Fundamental Investigation of Radiation Damage in Solids - Hall coefficient and electrical conductivity studies were made on both p- and n-type InSb. It was found that the p-type InSb is converted to n-type by irradiation. This is in contrast to the behavior of the semiconductor Ge in which n-type is converted to p-type by irradiation. It is also in contrast to the result of irradiating the semi-conductor Si in which there is an approach to the intrinsic condition (electrons = holes); both p- and n-type Si increase in resistance.

The magnetic susceptibility of a flawless, No. 1 color (colorless), emerald-cut diamond (on loan from R. G. Chapman of Kimball's, Knoxville, Tennessee) that weighed 640 mg (3.2 carats) was measured in the 88° to 294°K temperature range. This susceptibility was found to be 0.460 ± 0.003 cgs units at room temperature and was independent of temperature, within ± 0.1%, in the range studied. This work is part of a study of the effect of neutron bombardment on elemental semiconductors whose crystalline habit is the diamond lattice. (AEC Activity 5411)

PROGRAM 6000 - BIOLOGY AND MEDICINE

BIOPHYSICS

Waste Disposal Research - Studies on the use of open pits for storing concentrated radioactive wastes were extended. One type of pit under consideration has clay and asphalt walls and is filled with gravel and sand (see report ORNL-1638). Laboratory tests of the radiation, heat, and chemical resistance properties of pit materials are being made. Several small experimental pits have been constructed and are being evaluated.

Laboratory-scale experiments for testing the adaptability of ion exchange methods for the removal of radioactive contamination in military water supplies were completed. In these tests different ion exchange resins were tested with various radioisotopes and fission-product mixtures. The results indicated that the optimum pH of the feed solution depends on the type of radioactive contaminant, cycle of the resin used, chemical form of the contaminant, and other factors. A cubic foot of mixed-bed ion exchange resin can be expected to remove about 10 kilograins of hardness and over 99% of the radioactive materials from the contaminated water. (AEC Activity 6510)

Radio-Chemical Analysis - A new technique for the separation of tracer amounts of radioactive strontium from bulky quantities of calcium was developed. The separation is made possible by the ability of Versene to form stable complexes with calcium, when the pH is controlled within a limited range, whereas complexes with strontium do not exhibit such stability. The separation is made with a cation exchange resin, such as Dowex-50. It is possible to recover more than 99% of the Sr^{89} tracer from as much as 600 mg of calcium, with a separation factor of 10^3. This procedure can be used for isolating radioactive strontium isotopes from body fluids, bone, and bulky water samples. (AEC Activity 6510)

Experimental Radiation Measurements - A battery-operated scintillation detector weighing approximately 31 lbs, without the recorder, was designed and built for use in a car or light aircraft. This 6-12 volt unit contains a built-in calibration system which standardizes the amplifier gain, calibrates the rate meter, and checks the stability of the high voltage.

The scattering of gamma radiation from a concrete slab is under study. A Co^{60} gamma-ray source is used and the reflected rays are detected with a sodium iodide crystal scintillation spectrometer. The source and detector are suspended from a horizontal iron bar whose height above the concrete slab can be varied. For a source to detector spacing of 20 feet, it was observed that the measured intensity reaches a maximum when the height above the surface of the concrete is approximately 6 feet. Further measurements will be made at other spacings and with gamma-rays of different energies. (AEC Activity 6520)

Radioactive Particulate Problem - A histogram was plotted showing the average number of particles per 1000 cu ft of air collected weekly during the period of March 1949 through December 1953 by ten continuous air monitors stationed throughout the laboratory area. Every one of the twenty-four periods of RaLa operation coincides with a major peak of activity on the histogram.

Since May 1953, two complete cycles of the RaLa process were monitored for radioparticulate contamination of the air. Evaluation of the monitoring data seems to show that the major portion of the activity discharged through the stack is gaseous radioiodine. The gaseous activity masked the particulate activity and made it difficult to identify the nature of the collected particulates by radiochemical analysis or autoradiographic techniques. Samples containing particulates showed iodine and trace amounts of Pu, Ba, and rare earths. Samples containing no particulates showed iodine only.

Gaseous and particulate samples were collected simultaneously in the 3026-D Building and the 3039 Stack for a period of thirty hours. The concentration of particulates per unit volume of air inside the building was 3.2 times higher than in the stack effluent. The concentration in the building ranged from zero to 2600 particles/1000 cu ft of air. The average for one entire cycle amounted to 177 particles/1000 cu ft. Decay and absorption studies of the air samples collected in the process building indicated that a mixture of isotopes was present.

An underground tank (W-9) used for storing wastes from the RaLa process was also monitored during the second cycle studied. This tank proved to be a profuse source of airborne contamination. Peaks of airborne activity occurred whenever wastes were jetted from the process building to the tank. Air samples were taken at a position 18 inches from the vent in the top of the tank. The level of activity two days prior to the first jetting amounted to 10^{-9} μc/cc of air, while the average activity for the period covering the entire cycle of operation averaged 2×10^{-6} μc/cc. The highest activity observed for a 24-hour period was 2×10^{-5} μc/cc.

Peaks of activity at the tank vent were always accompanied by peaks of activity and particle counts at the continuous air monitors located about the laboratory area. The composition of the activity which was vented from the tank varied with the different stages of the process cycle. Radiochemical analyses showed that considerable quantities of rare earths, Sr, Ba, Te, and lesser amounts of I, Zr, Ru, and Nb were present. Individual particulates could not be resolved for measurement of size by the autoradiogram method. (AEC Activity 6520)

Consultation and Special Problems - The composition of the air contamination originating at the tank farm during the RaLa operations was studied to determine the maximum permissible concentration for such mixtures. Although the values obtained were significantly greater than the 10^{-9} μc/cc limit used when the composition is unknown, there was considerable variation in both concentration and composition during the run. (AEC Activity 6530)

RADIOISOTOPE SALES AND COSTS

Type of Transaction	January	FY to Date
Domestic Sales	$ 73,930	$ 520,933
Foreign Sales	8,550	33,140
Project-Cash Sales	14,715	67,690
Project-Transfer Credits	3,420	3,995
Technical Cooperation Program Credits	598	5,832
Plant Credits	7,634	77,526
Civilian Defense Credits	---	390
Cancer Program Credits	18,838	127,566
*Miscellaneous Income	33	252
Total Income	$ 127,718	$ 837,324
**Income - No production costs incurred, subtract from Total Income	1,557	18,525
Net Radioisotope Income	$ 126,161	$ 818,799
Radioisotope Costs	$ 95,152	$ 647,694
Radioisotope shipments	1,026	7,031

*Miscellaneous income - D_2O, freight, missing parts, TCP, etc.

**Income from H^3, He^3, B^{10}, and miscellaneous items in excess of handling cost.

GROSS OPERATING COSTS

	Cost for January	FY 1954 Cost to Date
Programmatic Operating Cost - Net	$ 2,380,037	$16,671,755
Plant and Equipment Cost	321,778	2,217,781
Construction Program "H"	1,979	24,208
Work for Other Parties - Transfers	40,016	302,642
Inventory Changes	47,021*	189,977* 1/
Reimbursable Work for Other Parties	138,014	906,342
Deferred Charges	4,980*	609
Total Laboratory Cost - Net	$ 2,829,823	$19,933,360
Estimated Cost for Next Month - Net	$ 2,850,000	$22,783,360

1/ Inventory budget adjusted to reflect transfer of the Returnable Containers 7/1/53 balance of $60,361 to Inventories.

*Credit

[redacted]

PERSONNEL SUMMARY

	Number of Employees February, 1954	New Hires February	Terminations February
Administration	64	0	0
Operations*	115	0	0
Engineering, Shops and Mechanical	757	11	2
Laboratory and Research	1673	11	7
Protection	149	0	9
Service	376	2	2
Total	3134	24	20

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

A total of 639 Laboratory personnel are located in the Y-12 Area.

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Previous reports in this series:

January 1953	ORNL-1486
February 1953	ORNL-1516
March 1953	ORNL-1529
April 1953	ORNL-1545
May 1953	ORNL-1562
June 1953	ORNL-1576
July 1953	ORNL-1600
August 1953	ORNL-1612
September 1953	ORNL-1622
October 1953	ORNL-1643
November 1953	ORNL-1651
December 1953	ORNL-1665
January 1954	ORNL-1676

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