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STATUS AND PROGRESS REPORT

APRIL 1954

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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

April, 1954

F. T. Howard

and

W. H Sullivan

Date Issued: MAY 6 1954

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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

25 Processing - Facilities for recovering enriched uranium from the fuel rods of the Savannah River reactors are being designed for installation at ORNL. All criticality limitations of process equipment have been established except the diameter of the continuous dissolver. In general, no geometry control is required where U^{235} concentrations are kept below 12 g/liter. The recovery process to be used includes continuous acid dissolution of the alloy, two tributyl phosphate (TBP) solvent extraction cycles, product concentration by evaporation, and sorption of residual fission products on silica gel. In the tentative flowsheet, designed for an overall uranium loss of less than 0.04%, the first cycle extractant is 6% TBP and the feed is 0.1 M in nitric acid, 1.8 M in aluminum nitrate, and contains 1.92 g of uranium per liter. The uranium is stripped with 0.01 M nitric acid, evaporated, and converted to second cycle feed by addition of nitric acid. The second cycle extractant is 15% TBP; the feed is 3 M in nitric acid and the strip solution is 0.01 M nitric acid.

In preliminary laboratory-scale studies (1-in.-dia. column) on continuous dissolution of Savannah River type extruded slugs (5% uranium--95% aluminum), the dissolution rate, corresponding to 8 kg of uranium per day in a 5.5-in.-dia. column, was optimized at 6 M nitric acid, and a solution was obtained with aluminum, acid, and uranium concentrations easily adjustable to those required for the solvent extraction flowsheet. In studies on Hanford type cast slugs (7.5% uranium--92.5% aluminum) in 5.5-in.-dia. columns, a dissolution rate of 12 kg of uranium per day was observed. In this large-scale dissolver, off-gas removal from the middle of the column was shown to give a higher dissolution rate and more concentrated final product than removal from the bottom.

Tests of the first cycle of the solvent extraction flowsheet in glass 2-in.-dia. pulse columns indicated that with 7-in.-dia. columns a 16-ft extraction section and a 22-ft stripping column will be needed to keep the uranium loss at less than 0.01% in each column. (AEC Activity 2XXX)

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PROGRAM 3000 - WEAPONS

Special Electromagnetic Separations - In a second plutonium isotope separation run the two-arc calutron was operated for 24 hours at an average Pu^+ ion current of 30 milliamperes; this corresponds to a plutonium separation rate of 275 mg/hr. The feed ($PuCl_3$) assayed 93.9% Pu^{239}, 5.7% Pu^{240}, and 0.4% Pu^{241}. The three products obtained were: 6 gm 99.5% Pu^{239}, 0.4 gm 61% Pu^{240}, and 0.18 gm 16% Pu^{241}. (AEC Activity 3601)

PROGRAM 4000 - REACTOR DEVELOPMENT

HOMOGENEOUS REACTOR PROJECT

Homogeneous Reactor Experiment - Dismantling of the HRE continued at a faster pace during April and is now about 50% completed. All of the equipment was removed from the heat exchanger cell and most of the parts from the "soup" cell. Personnel exposures have been kept to reasonable levels, but this is becoming more and more difficult. An offer from the Naval Reactor group to supply enlisted personnel for the dismantling work has been accepted, and these men are already participating in the operation.

Partial decontamination of the HRE, following the procedure noted in the preceeding report (ORNL-1703), was completed and the decontamination solutions analyzed. The first decontamination solution, 200 liters of 8 M nitric acid, removed about 1900 curies of rare earths, 1000 curies of barium, 230 curies of zirconium, and much smaller amounts of other fission products. This nitric acid solution had a gamma activity of 6×10^9 c/min-ml, about 7.5 times higher than the HRE fuel solution with a comparable volume on the same date. Barium and the rare earths have never appeared as important contaminants before; it is postulated that they had precipitated in the system. The alkaline tartrate peroxide, which followed the first nitric acid treatment, removed some 200 curies of niobium, more of this element than was removed by any other reagent.

The most striking observation resulting from the decontamination is that the decontamination solutions have removed a total of over 40 times as much gamma activity as the fuel solution itself contained at the time, but only 1.4 times as much beta activity. This is evidence of the tendency of many of the fission products which are strong gamma emitters to adsorb on the equipment or to precipitate in the system. (AEC Activity 4103.1)

Homogeneous Reactor Test - It has been decided that the HRT would be operated with the core and blanket solutions at the same temperatures. In order to simplify such problems as pressure balancing, phase separation, and corrosion, it is now specified that the outlet temperatures of both fuel and blanket be 300°C when the blanket is either D_2O or 60 gm U/Kg D_2O. When the blanket is

operated at a concentration of 355 gm U/Kg D_2O, the outlet temperature for both systems will be reduced to 260-270°C. Plutonium production in the case of the 60 gm U/Kg D_2O blanket would be 1.25 gms/day which is expected to be sufficient for chemical separations development.

Overall design of the HRT is perhaps 50% complete. Development work for the core and pressure vessel is proceeding according to schedule, and experimental testing of the mock-up is expected to be completed by the end of July. Bids have been received for the manufacture of the main heat exchangers for both the core and blanket systems, but a fabricator has not been chosen. Orders have been placed for most of the stainless steel piping requirements. (AEC Activity 4103.1)

Engineering Investigation of Slurries - Circulating ThO_2 slurries do not appear to attack 347 stainless steel at velocities below 20 fps. At higher velocities attack becomes more severe, depending on the type of ThO_2 solids and the geometry. Stellite type 98M2, which approaches the hardness of the ThO_2 particles, and 400-series stainless steels are also attacked at high velocities. Titanium, zirconium, Zircalloy, gold, and platinum are not seriously attacked even at velocities approaching 100 fps. These facts not only point toward and explanation of the attack mechanism but also indicate a solution to the erosion problem with ThO_2 slurries. (AEC Activity 4109.31)

Chemical Investigation of Slurries - The least abrasive thorium oxide slurry that has been obtained at a thorium concentration of 800 to 1000 g/liter was prepared by precipitating thorium hydroxide from thorium formate solution with ammonium hydroxide, dehydrating it at 500°C, and autoclaving the resulting oxide in water at 250°C.

Studies of the properties of thorium formate show that it exists as the monohydrate at 100°C, becomes anhydrous at 125°to 175°C, and decomposes at 225°to 250°C. The thorium concentration of a saturated aqueous solution of the formate was 52 to 62 g/liter at room temperature and 24 g/liter at 97°C.

The addition of 10 to 40 mole % of ZrO_2 and Bi_2O_3 and 20 to 30 mole % of Al_2O_3 to ThO_2 by the coprecipitation of the hydroxides and subsequent calcination of the mixtures at 545°C did not appear to inhibit the crystallization of the ThO_2 during calcination. The addition of 10 mole % Al_2O_3 in a similar experiment partially inhibited the crystallization process.

Titanium metal exhibited approximately the same abrasive resistance as 300-series stainless steel when tested with slurries of SiC and high-burned ThO_2 in laboratory jet-impingement abrasion tests. The greater resistance of titanium to erosion as compared to stainless steel in the slurry pumping studies may result from the formation of a stronger oxide film in water at 250°C or its more rapid replacement when abraded. (AEC Activity 4109.31)

Radiation Chemistry - Studies of the effect of pile irradiation on the corrosion of type-347 stainless steel by uranyl sulfate solution have again suggested the existence of a radiation effect. Out-of-pile tests on mirror polished surfaces yield reproducible results, with an initial corrosion rate of about 20 mils/yr. After this short initial period the rate drops quickly, leveling off at less than 0.1 mil/yr. In the in-pile tests, although the initial corrosion rate is essentially the same, the curve does not level off as fast and after about three days actually shows a greater corrosion rate. At the end of five days of irradiation, when the experiments were terminated, the rate was about 6 or 7 mils/year. Work is being continued to ascertain the factors responsible for the observed effect.

Studies of the yield of nitrogen gas from pile irradiated thorium nitrate solutions give a value of the order of 10^{-3} molecules per 100 ev. This is less than previously reported values for uranyl nitrate solutions by a factor of at least 100, but the conditions differ in that energy input from fission is almost absent in the thorium nitrate irradiations. (AEC Activity 4109.31)

Metallurgy - Corrosion resistance of a fully austenitic stainless steel weld metal (18% Cr, 13% Ni, 5% Mn, Type-"H" alloy developed in the program of the AEC Advisory Committee on Stainless Steels) was found superior to that of other welds containing from 8 to 11% ferrite, after 400 hours in 0.17 M uranyl sulfate at 250°C and 20 fps under highly turbulent conditions.

Moist-hydrogen treated, chromized, type-304L stainless steel specimens showed negligible weight loss in 0.06 M uranyl sulfate containing 0.016 M sulfuric acid, after 200 hours at 250°C and 68 fps. Film studies on these specimens showed presence of high-parameter (8.42 kx) spinel structure and chromic oxide. (AEC Activity 4109.4)

Fuel Reprocessing - Further solubility tests (ORNL-1695) of two mixtures of rare earth sulfates and $Ce_2(SO_4)_3$, $La_2(SO_4)_3$, and $Nd_2(SO_4)_3$ in simulated uranyl sulfate reactor fuel showed that the solubility of these substances is less than 0.2 g/liter at 250°C and even less at 300°C. At this concentration the rare-earth fission products contribute less than 2% to the reactor neutron poisoning. The Vitro Corporation has confirmed these results, by a filter-bomb technique.

Pilot-plant testing of the calcium fluoride process for removing rare earths was completed by Vitro. The cost for producing a fluoride-free product was estimated to be about 0.5 mill/kwh, while the cost of the CaF_2 process without a fluoride removal step was about 0.3 mill/kwh of electricity produced.

As part of the blanket-processing study for the K-49 plutonium producer, the solubility of neptunium (as NpO_2^+ or NpO_2^{++}) in 1.25 M UO_2SO_4 at 300°C in contact with H_2 and O_2 was shown to be greater than 0.07 g/liter. At the reactor equilibrium concentration, 0.033 g Np^{239}/liter, neutron capture by

Np^{239} followed by beta decay would produce significant amounts of Pu^{240} if the capture cross section of Np^{239} is of the same order of magnitude as that of Pu^{239}. In order that the Np^{239} capture cross section might be determined, a solvent extraction process, which is a modified single-cycle Purex process, was developed and designed to isolate milligram quantities of plutonium from LITR-irradiated depleted uranium. The plutonium in the solvent extraction product solution is concentrated to a small volume by a ferrous hydroxide precipitation, dissolved in hydrochloric acid, and further purified from iron and traces of uranium by a fluoride precipitation. (AEC Activity 4109.8)

AIRCRAFT NUCLEAR PROPULSION PROJECT

Aircraft Reactor Experiment (ARE) - Refabrication of the ARE heat exchangers has progressed rapidly, and it will be possible to start initial testing of the system sooner than expected. The fuel heat exchangers were reinstalled, and the sodium heat exchangers will be ready for reinstallation by mid-May. Fabrication of pump impellers in the shops is on schedule and is not expected to delay installation of the pumps for the fuel and sodium systems, which is scheduled to begin May 15. The monolithic concrete shielding plug for the reactor was set in place. The electrical work for the system is nearing completion. (AEC Activity 4401.1)

Experimental Engineering - The ARE fuel-system mockup was filled successfully in tests with 1-1, 2-2 tetrabromoethane. Complete filling and satisfactory degassing were obtained at a flow rate of 57 gpm and a gas pressure of 2 psig. Satisfactory recirculation for degassing inside the ARE pumps was demonstrated. Six rotating elements for ARE pumps were assembled, and four have satisfactorily passed hot testing. Two fabricated impellers were received. Tests were completed on three gas-sealed, vertical-shaft, sump-type centrifugal pumps for pumping fluoride mixtures in in-pile loops; two of the pumps were delivered for installation. Fabrication of a horizontal pump designed for in-pile use is about 50% complete.

An HNO_3-H_3BO_3-$Pb(NO_3)_2$ solution was found to be satisfactory for removing $NaZrF_5$ from Inconel surfaces. (AEC Activity 4401.1)

Critical Experiments - The results of preliminary assembly experiments on the AC-100, air-cooled, water-moderated reactor were transmitted to the General Electric Company for analysis. The recent experiments included the calibration of poison-type control rods, evaluation of reactivity coefficients of experimental materials and their arrangement within the assembly, and the measurement of fission-rate distributions. These experiments were done to provide the data needed for ascertaining the deficiencies in the calculations which caused the predicted critical parameters to be significantly different from those measured.

A reactor safety fuse designed by GE was tested. An air stream was used to release amorphous boron from a reservoir in the fuse through a rupture disk into a shell surrounding a fuel element; the reactivity decrease was about equivalent to one-half of that due to delayed neutrons. In an operating unit, the dispersal of the boron could be triggered by a temperature increase.

Materials are being fabricated for a critical assembly of a reflector-moderated reactor. Experiments have been planned that will provide more fundamental information than was obtained in the previous experiments which were made to establish design parameters. The fuel region of the new assembly will consist of alternate sheets of uranium metal, 0.004 in. thick, and a fluorocarbon plastic, Teflon. By varying the thickness of the Teflon, it will be possible to obtain uranium densities of up to 0.5 g/cm^3. Beryllium metal will be used as the reflector and the moderator. (AEC Activity 4401.4)

Radiation Damage - The LITR fluoride-fuel loop, consisting of a U-shaped length of Inconel tubing, a heat exchanger, a pump, and a venturi flowmeter, was assembled, with the exception of the pump. The pump is being positioned, with respect to the loop and its gamma shielding, prior to being welded to the loop. The attachment of heaters, thermocouples, and neutron shielding to the loop was completed, and connections to the control panel are being made. The shield into which the loop will be withdrawn after operation was finished, and handling facilities for disassembly of the loop in the hot cells are 80% complete. Bench testing of the loop remains to be done before the loop and its controls can be moved to the LITR. Installation of services and utilities for the loop has proceeded as far as possible until actual connections can be made to the loop and its control panel.

A detailed study of 13 Inconel capsules containing fluoride fuel that were irradiated in the MTR shows that the capsules can be divided into two groups: (1) those in which the Inconel was penetrated to a depth of 3 $\pm$ 1 mils and (2) those in which the penetration was greater than 10 mils and in which there were large grains. The four capsules with greater than 10-mil penetration showed no experimental trend other than temperature effects. At some time during irradiation, they were above 1900°F for a period of 1 to 100 hours. (AEC Activity 4401.4)

ANP Reactor Chemistry - The solubility of UF_3 in molten $NaF-ZrF_4$ mixtures at 600°C was found to increase from about 1.5 mole % to more than 3 mole % when the ZrF_4 concentration was increased from 47 to 57 mole %. The NaF-LiF-KF eutectic dissolved more than 3.5 mole % UF_3 at temperatures above 510°C. The NaF-LiF-RbF eutectic dissolved approximately 1.3 mole % UF_3 at 500°C and at least 2.9 mole % at 600°C. The solubility of beryllium metal in molten sodium, as determined by equilibration, filtration, and analysis of the filtrate, is less than 10 ppm at 800°C.

Apparatus for drawing liquid fuel samples from the ARE during its subcritical and low-power operation was tested and found to be satisfactory. (AEC Activity 4401.5)

Corrosion Research - Analyses of the fluoride mixture from a second Inconel loop operated at 1500°F for 500 hr with a zirconium-base fluoride mixture containing both UF_3 and UF_4 confirmed the lack of attack found metallographically. The analyses indicated, however, an apparent loss of uranium. The final uranium content of the fluoride mixture varied from 4.9 to 5.5%, while the original batch contained 6.0%. No systematic variation of the uranium content with position of the sample in the loop was found.

The results of additional experiments with Inconel loops in which zirconium-base fluoride mixtures were circulated confirmed that the mass transfer in these loops is linear with time, is not sensitive to the original purity of the fluoride mixture after the first few hundred hours, and is greater at higher operating temperatures.

Two stainless steel loops circulated lithium for 1000 hours. One loop operated with a temperature gradient of 150°C and a hot-leg temperature of 810°C. The other operated with a temperature gradient of 60°C and a hot-leg temperature of 785°C. Very little mass transfer occurred in either loop, only 0.1 g being mass transferred in the loop with a 150°C temperature gradient. In the previous tests with lithium, the austenitic stainless steel loops would only operate for a very short time prior to stoppage of flow because of mass transfer. Part of the increased life can probably be attributed to the higher purity of the lithium used in these tests.

Various borides, including TiB, ZrB_2 and Mo_2B, were tested in a fluoride mixture to determine their suitability as bearing materials. The titanium and zirconium borides showed fair resistance to attack, whereas Mo_2B was heavily attacked.

In the evaluation of high-temperature brazing alloys, oxidation tests of various alloys on Inconel joints were made at 1500 and 1700°F for 200 hours. It was found that Nicrobraz, low-melting-point Nicrobraz, Coast Metals alloy 52, General Electric alloy 81, and the ORNL research alloys 75% Ni-25% Ge, 65% Ni-25% Ge-10% Cr, 80% Au-20% Cu showed very good oxidation resistance. The following brazing alloys showed poor oxidation resistance: Ni-Mn, Ag-Pd-Mo, Ni-P, Ni-P-Cr, and Ni-Sn. (AEC Activity 4401.5)

Metallurgy - Fabrication - A sodium-to-air radiator with 3-in. high-conductivity fins was fabricated. Heliarc machine welding with the new rotating torch was used for making the 180 tube-to-header joints. The tube-to-fin matrix consisting of approximately 4500 joints was brazed with Coast Metals Alloy 52. It is encouraging to find that it is possible to build such a complicated assembly. Since the high-conductivity fin placed a maximum upper operating temperature of 1083°C, the melting point of copper, on the brazing operation and the brazing alloy had a melting point of 1020°C, the furnace was carefully controlled so that an overrun would not cause a failure.

In the past there was a need for very closely spaced tube-to-header joints, but with the cone-arc welding machine it was impossible to get a center-to-center distance of less than twice the tube diameter. By using the new automatic heliarc welding rotating torch, it is possible to make helium-leaktight joints with 0.225-in-OD, 0.025-in-wall tubing with a center-to-center spacing of only 0.250 inch.

The variables affecting tube-to-header welding with the automatic rotating torch were studied. Type-316 stainless steel test samples were constructed from 1/8-in. header plates and 0.185-in.-OD tubing with 0.025-in.-wall. The arc current was varied from 20 to 60 amperes, the welding time ranged from 4 to 13 sec, and the arc length was varied from 0.03 to 0.07 inch. It was found that (1) the weld penetration was directly proportional to arc current and greater at an arc distance of 0.05 in. than it was for arc distances of either 0.03 or 0.07 inch; (2) the hole closure was not excessive; and (3) weld penetration and weld width were inversely proportional to the welding speed. (AEC Activity 4401.5)

Heat Transfer Research - Thermal conductivity measurements were made on the fluoride mixture RbF-LiF (57-43 mole %). Preliminary data show

$$k_\ell = 1.25 \text{ Btu/hr-ft-}^\circ\text{F} \qquad 510^\circ < t < 728^\circ\text{C}.$$

On a plot of k_ℓ vs molecular weight, the value for the RbF-LiF mixture falls approximately on the middle of the curve obtained by plotting the values for the other fluoride mixtures studied to date. The solid and liquid thermal conductivity values of NaF-KF-LiF (11.5-42.0-46.5 mole %) and NaF-KF-UF_4 (46.5-26.0-27.5 mole %) are, in comparison,

	k_s	k_ℓ
NaF-KF-LiF	3.5	2.6
NaF-KF-UF_4	0.75	0.55

These materials give k_s/k_ℓ ratios of ~1.4, whereas metals, heat transfer salts, and water give

$$1.5 < \frac{k_s}{k} < 3.$$

(AEC Activity 4401.5)

Physical Properties Research - The viscosity of the RbF-LiF (57-43 mole %) mixture was determined and is compared below with the viscosities of other fluoride mixtures.

Temperature (°C)	Viscosity, (cp) RbF-LiF (M = 71)	NaF-KF-LiF (M = 40.4)	$NaF-ZrF_4-UF_4$ (M = 110)
530	11.0	10	22.0
700	3.9	3	9.5
800	2.5	2	7.0

The addition of small amounts of uranium to the RbF-LiF mixture would increase the viscosity slightly.

A report (ORNL-1702) was prepared on the densities of molten fluorides. The experimental data are summarized, and correlation expressions are given which permit predictions (accurate to within a few percent) of the liquid densities of fluoride mixtures over a wide range of temperatures if the compositions of the mixtures are known. (AEC Activity 4401.5)

Analytical Studies - Calculations of free-energy changes for the reactions between anhydrous HF and such metallic oxides as UO_2, ZrO_2, Fe_2O_3, and Cr_2O_3 were made. On the basis of these changes in free energy, a method was proposed for the determination of oxygen in oxides. The increase in conductivity of the HF, which results from the following reaction,

$$M_2O_3 + 6\ HF \longrightarrow 2MF_3 + 3H_2O,$$

is a function of the oxygen content of the oxide. A preliminary design was prepared of an apparatus for use in applying this method to the determination of oxygen in fluoride fuels.

In connection with the determination of U^{3+} in the presence of U^{4+}, dioxane was found to dissolve UCl_4 selectively from UCl_3. No solubility effect was noted on the fluoride salts. Further work with the acetyl-acetonates of U^{3+} and U^{4+} revealed that the complexes were too unstable for analytical application to the fluoride fuels.

Studies on the use of iodine in methanol as a one-step oxidant for U^{3+} indicated that, if U^{4+} were originally present and the conditions were similar to those for the quantitative oxidation of U^{3+} to U^{4+}, as much as 25% of the U^{4+} was oxidized U^{6+}. Further investigation of U^{6+} as an oxidant for U^{3+} revealed that these competitive reactions were involved:

$$2U^{3+} + UO_2^{++} + 4H^+ \longrightarrow 3U^{4+} + 2H_2O$$

$$2U^{3+} + H^+ \longrightarrow U^{4+} + 1/2\ H_2$$

Efforts to eliminate the oxidation by hydrogen ion were unsuccessful. (AEC Activity 4401.5)

Shielding Research - The reactor at the Tower Shielding Facility is now being operated on a three-day week basis. The flux distributions in both the reactor and the detector tanks are being mapped as a preliminary part of the differential shielding measurements.

The Bulk Shielding Reactor was operated at a power level of one megawatt for a few minutes to determine the operating characteristics of this type of reactor at high power with unimpeded convective cooling. (In similar experiments with the LITR, convection was limited because the return path included the reflector which had relatively small cooling passages.) The reactor operated very satisfactorily at the high level, except that the radioactive N^{16} produced in the water was carried by the convection to the pool surface, where it raised the radiation level to well above tolerance. Other experiments are planned in which the convective flow will be broken up by, for example, a cross-jet of water above the reactor. The short half-live N^{16} will thus decay throughout the whole pool rather than primarily near the surface. (AEC Activity 4401.7)

GENERAL REACTOR RESEARCH

Package Reactor Project - Approximately one-half of the private concerns contacted in regard to a lump-sum contract for the design and construction of the package reactor replied, with interest ranging from mild to very strong. It is expected that invitations to bid will be issued in early June. A preliminary review of the reactor design was held by the Reactor Safeguards Committee on April 21 at Cincinnati, Ohio. The Committee was very much interested in the possible use of the proposed stainless steel-clad fuel plates in other reactors, especially in research reactors. The development of fabrication techniques for the fuel plates is progressing satisfactorily. Preliminary testing of the control rod drive mechanism, by the American Machine and Foundry Company, was started. (AEC Activity 4510)

General Reactor Radiation Damage - A "rocking-curve" apparatus, incorporating an x-ray source and a GM tube, was devised which permits the measurement of lattice parameters in large single crystals with an accuracy of $\pm$ 0.005%. With additional refinements it is expected that this precision can be increased to $\pm$ 0.002%. Preliminary measurements on copper single crystals irradiated with 3.0×10^{19} neutrons/cm^2 reveal no changes in lattice parameter within the limits of measurement.

A study was made of radiation effects on polyvinyl chloride plastics which contained various proportions of plasticizer. Formulations (furnished by the B. F. Goodrich Research Center) were plasticized with 52, 25, and 0 parts of dioctyl phthalate per 100 parts of polymer. In addition, two unplasticized formulations were loaded with, respectively 3 parts TiO_2 plus 17 parts $CaCO_3$, and 10 parts of TiO_2. The dynamic elastic modulus and the internal friction were determined for the irradiated materials. After

exposure for one week in the ORNL Graphite Reactor the unplasticized formulations were slightly softened; the formulation with the smallest proportion of plasticizer was little changed; the formulation with the greatest proportion of plasticizer was hardened. This behavior suggests opposite radiation effects for polymer and plasticizer. The predominant radiation-induced changes are chain cleavage for the polymer and crosslinking for the plasticizer. This explanation fits the behavior of certain previously tested polyvinyl chloride formulations containing less plasticizer that were observed to soften for short irradiation periods, but to harden for long irradiation periods. (AEC Activity 4540)

Heterogeneous Reactor Fuel Reprocessing - Results of preliminary experiments on processing of the stainless steel-clad uranium dioxide fuel plates for the proposed 10-Mw package reactor indicate that the chemical passivity of the cladding can be destroyed by introducing carbon into the stainless steel at an elevated temperature and precipitating chromium carbide. A section of an element was heated for about 1 hr at about 1000°C in a stream of nitrogen and methane. The iron, nickel, and some of the chromium then dissolved readily in 2 M copper sulfate containing 0.05 M sulfuric acid. The solution was discarded, with a uranium loss of 0.03%, and the element, now coated with copper, was dissolved by boiling with 12 M nitric acid for 2 hr. The insoluble residue from this step contained 0.72% of the uranium. The uranium loss increased in the stainless steel dissolution step when the sulfuric acid concentration was increased, and in the uranium dioxide dissolution step when the stainless steel was not completely dissolved. The copper sulfate reagent is much less corrosive than 6 M H_2SO_4, which is commonly used for dissolving uncarburized stainless steel. (AEC Activity 4581)

Dissolution Studies - A rate equation was derived which fits the data for caustic dissolution of uranium-aluminum slugs under all conditions studied. The specific rate constant was 2.3×10^{-4} liter/min-cm^2 in 6 M sodium hydroxide. This rate is slightly higher than the theoretical maximum rate for the HNO_3-Hg^{++} system with 4.5 M nitric acid and zero aluminum concentration. Unlike the acid dissolution rate, which is optimized at 4.5 M concentration and decreases with increasing aluminum concentration, the caustic rate is directly proportional to the sodium hydroxide concentration, at least over the range 3 to 7.65 M sodium hydroxide. No significant difference was noted between rate of dissolution of cast and extruded slugs.

The average uranium loss on filtration of the uranium-caustic slurry through a G-porosity stainless steel filter was less than 0.05%. Filtration rates were reasonable, i.e., greater than 0.6 ml/min-cm^2, at initial sodium hydroxide concentrations of 6 M and increased by a factor of 10 as the caustic concentration decreased by a factor of 2. (AEC Activity 4581)

ISOTOPE PRODUCTION

Stable Isotope Production - Another separation of zirconium isotopes (masses 90, 91, 92, 94, 96) was completed in March; an estimated 35.1 grams of the separated isotopes was obtained. Gram amounts of enriched Cr^{54} are being collected from which targets will be fabricated for the Cr^{54} (p,n) Mn^{54} reaction in the 86-inch cyclotron. Mn^{54} has applications as a radioactive isotope and the calutron-cyclotron production of Mn^{54} is economically and technically feasible.

A simplified process was developed for the preparation of pure solutions of rare earths from gadolinite ore. The metallic elements are dissolved from the milled ore with the concentrated nitric acid. After dilution to 6-10 N acid, the rare earth elements are separated from iron, beryllium, and other similar impurities by countercurrent extraction with tributyl phosphate. A preliminary extraction for removal of the less desirable cerium earths is particularly advantageous. The rare earths are then extracted from the tributyl phosphate with 3 N nitric acid. This procedure eliminates all precipitations and ignitions.

Enriched Gd^{157} was added to the inventory of isotopes for the first time. New lots of Fe^{54}, Fe^{57}, Ti^{48}, Sm^{147}, Sm^{148}, Sm^{149}, Sm^{150}, Sm^{152}, Hf^{176} and Hf^{178} were also added as replenishments.

Forty-one shipments were made, consisting of 33 different isotopes of 23 elements. (AEC Activity 5121)

PHYSICS

High Voltage Program - The absolute neutron spectrometer was modified to utilize two proportional counters and a scintillation counter. The neutron spectrum from $T(d,n)He^4$ was measured with 7% energy resolution at an angle of 118° with respect to the incident deuteron beam. The neutron spectrum from $Li^7(d,n)Be^8$ was measured with 6% to 10% statistics. Only one excited state (at 3 Mev) was found in Be^8, up to an excitation of 9 Mev.

The Li^6 total neutron cross section, previously reported in ORNL-CF-52-8-165, was rerun with new samples of higher purity. The new results confirm the earlier data.

The angular distribution of neutrons elastically scattered from Li^6 was measured at neutron energies of 220, 260 and 300 kev. Scattering is asymmetrical about 90° in the center-of-mass system, being forward below resonance and backward at and above resonance. (AEC Activity 5211)

Scintillation Spectrometry and Instrument Development - Investigation of the radiation from Sb^{124} with a 3" x 3" NaI scintillation spectrometer showed the presence of two new gamma rays with energies of 0.99 Mev and 1.38 Mev, and with respective intensities which are 5.4% and 6.2% of the intensity of the 0.603-Mev transition. Because of poor resolution measurements previously obtained these gamma rays had not been separated from pair peaks and Compton edges. The presence of these gamma rays, together with the intensities of the radiation, allowsa coherent decay scheme to be arranged which agrees in energies and intensities with earlier beta-ray measurements.

Investigations were continued with the purpose of developing a scintillation spectrometer that gives only one peak for each gamma ray detected. While this could be accomplished with a very large NaI crystal (about 10" in diameter), such crystals are not available. A very large tank (28" in diameter) was constructed to contain solution phosphor around a 3"-diameter NaI crystal so that any radiation partially detected in the NaI crystal will probably also yield a pulse in the solution phosphor. With anticoincidence circuits these events are not counted so that a spectrum is obtained which includes principally the full energy peaks. This arrangement has given a response to the 1.114-Mev gamma ray of Zn^{65} which has no point in the Compton distribution with an intensity higher than 1/25 of the intensity in the peak. Changes in the electronic circuits will probably improve these results so that this arrangement will make a very good spectrometer for continuous gamma-ray distributions below energies of 3 Mev. (AEC Activity 5211)

Nuclear Physics, 86-Inch Cyclotron - Cross sections and excitation functions for several (p,n), (p,2n), (p,pn), (p,f), (p,α), and (p,He^3) reactions were measured. The principle conclusions are: (1) The nuclear radius for uranium and thorium is approximately 1.6 $A^{1/3} \times 10^{-13}$ cm; (2) the sticking probability for 22-Mev protons incident on elements as heavy as iron and nickel is considerably less than unity; (3) the ratio of (p,pn) to (p,2n) cross sections is, in general, far larger than can be explained theoretically (previous explanations can be shown to be invalid); and (4) the cross section for the (p,He^3) reaction on Ga^{71}, the only (p,He^3) reaction reported, is about equal to the theoretical estimates. (AEC Activity 5211)

The 86-Inch Cyclotron - Tests were continued for determining the yields for various cyclotron-produced isotopes. A 24-hr bombardment of a chromium target at an average proton current of 1020 μa produced 12.1 mc of Mn^{54} and 2900 mc of Mn^{52}. Yield data for the production of Mn^{54} by (p,pn) on Mn^{55} indicate that it is produced at the rate of 24 mc/mah; 170 mc was produced in 7 hours. A sodium target was bombarded at 400 μa for 28.5 hr to produce non-carrier-free Na^{22} by the (p,pn) and (p,2n) reactions; 62.5 mc of Na^{22} was produced with a specific activity of 6 mc/g Na. In a 36-hr bombardment of nickel at an average current of 1015 μa an estimated 1.8 curies of carrier-free Co^{57} was produced; this material will be used in preparing a point source for determining the suitability of the isotope for radiographic applications. (AEC Activity 5261)

Electronuclear Machines - In connection with the study of a multiple-particle heavy-ion cyclotron an investigation of a low-pass filter coupling to cyclotron dee stems was completed. Data concerning the operation of the filter as a function of frequency indicates that a low-pass filter could be used to couple the oscillator to the dee stems of a variable-frequency system, provided a variable phase-shift circuit were included in the grid circuit of the oscillator.

Also in connection with the study of the multiple-particle cyclotron, an automatic tuning system was developed which should prove applicable to such a machine. The tuning system operates very reliably in tuning a small oscillator to the resonant frequency of a loosely-coupled secondary circuit; the error of the control in tuning is of the order of 0.1%. This is thought to be within the accuracy required of the multiple-particle cyclotron. (AEC Activity 5261)

Isotope Analysis Methods Laboratory - The first reported Zeeman spectra of a transuranic element were obtained for plutonium in a magnetic field of 25000 oersteds by means of an Echelle spectrograph. These spectra show that the singly-ionized plutonium atom contains six electrons in the 5f shell.

Continuing studies on the quadrupole perturbed hyperfine patterns of U^{235} resulted in the detection of three seven-component patterns. Precise intensity measurements on one of these indicates it to be an unresolved eight-component pattern. These data revise the nuclear spin of U^{235} from 5/2 to 7/2.

Strong Fermi resonance between the CH bending and the CF stretch in the infrared vibrational spectra of DCOF have been confirmed by precise integrated absorption measurements of the spectra. (AEC Activity 5261)

Mass Spectrometer Laboratory - Data for the ratio of isotope masses 58 and 60 can be obtained for as long as 10 to 15 minutes on one microgram of nickel deposited as the sulfate on the iridium ribbon heater in the ion source of the mass spectrometer. This is adequate data for the quantitative measure of nickel in the HRE fuel stream.

Isotopic abundance measurements on ruthenium and iridium yield new values as follows:

Ruthenium

Mass	Abundance (%)
96	5.47 ± 0.03
98	1.84 ± 0.02
99	12.77 ± 0.05
100	12.56 ± 0.05
101	17.10 ± 0.06
102	31.70 ± 0.06
104	18.56 ± 0.05

Iridium

Mass	Abundance (%)
191	37.3 ± 0.1
193	62.7 ± 0.1

Application of the isotope dilution method of analysis to ARE fuels was extended to include measurement of uranium losses due to corrosion, whereas previously only uranium losses due to burnup were measured.

Mass spectrometer analysis revealed that about 2% methanol was an unsuspected impurity in krypton which gave rise to desirable operating characteristics in a group of krypton-filled counters.

A compact, self-contained bias supply was designed and built to replace the many batteries used in conventional mass spectrometer circuits. The supply converts 115 volt ac to any multiple of 55 volts dc, with less than 0.01% ripple. (AEC Activity 5261)

The 63-Inch Cyclotron - During a shutdown period, adjustments and re-alignments were made which resulted in much improved reliability and stability of operation of the cyclotron. The reactions produced by nitrogen on carbon were compared with the reactions produced by protons on Mg^{25}; both bombardments lead to the same nucleus, Al^{26*}, if the reactions proceed by way of a compound nucleus. The nitrogen-induced reactions seem to favor the emission of alpha particles. The coulomb excitation of the 137-kev gamma ray in Ta^{181} was observed. Progress was made in the study of the excitation functions for the reactions $N^{14}(N^{14})N^{13}N^{15}$ and $N^{14}(N^{14})O^{15}C^{13}$. (AEC Activity 5261)

Research and Development, Stable Isotopes - The collection of calutron-separated isotopes of inert gases on a collector consisting of a rotating drum dipping in oil was satisfactory, although decomposition of the oil presents some uncertainty as to actual gas composition. The use of gallium and some of its low-melting alloys for increasing gas retention is being investigated.

Twelve grams of very high purity Li^6 metal was cast into three separate containers for Van de Graaff experiments. The technique for casting large quantities of lithium metal was applied to the preparation of a special 25-lb shield for neutron spectrometer work.

In cooperation with the 86-inch cyclotron group, cyclotron targets of enriched isotopes are being investigated for the production of useful radioactive isotopes. Enriched isotopes of lithium, germanium, nickel, hafnium, molybdenum, chromium, platinum, and tellurium are of interest as targets. (AEC Activity 5261)

CHEMISTRY

Inorganic Solution Chemistry - The anion exchange behavior of the alkaline earths, Mg, Ca, Sr, Ba, and Ra, in citrate solutions was studied with the strong-base anion-exchange resin, Dowex-1. Increasing adsorption was found

with decreasing atomic weight, as well as a very large difference in adsorbability of Ca and Mg. The separation of the elements, Mg, Ca, Sr, and Ba was demonstrated with a 20-cm column of the citrate form of the resin. Under the same conditions there was only a partial separation of radium and barium.

Since it had been found that osmium could be strongly adsorbed from chloride solutions by Dowex-1 and since iridium under the same conditions showed very little adsorption, attempts were made to isolate the Ir^{191} isomer, which was suspected to be formed in the decay of Os^{191}. By using an anion exchange technique and very short columns, it was possible to isolate the Ir^{191} isomer and to determine its half life to be 4.9 sec.

In continuation of the systematic survey of the anion exchange behavior of metals in hydrochloric acid solutions, the adsorbabilities of Ag(I), W(VI), and Mo(VI) were studied. Both Mo(VI) and W(VI) showed strong adsorption in 6 to 12 M HCl (distribution coefficient D = $\sim$100). At lower HCl concentration (1 M HCl), the D for Mo(VI) was $\sim$1. Due to the precipitation of tungsten, its distribution coefficients were not determined below 6 M HCl. Ag(I) showed only slight adsorbability in strong HCl solutions; increasing adsorption occurred with decreasing HCl concentration. The distribution coefficient for Ag(I) in 2 M HCl was larger than 100.

A separation procedure, devised for the separation of W(VI) and U(VI), is based on the fact that U(VI) in 6 M HCl-1 M HF is strongly adsorbed, while under the same conditions W(VI) is not adsorbed. Passing a mixture of the two elements into a small column of Dowex-1 yields W(VI) in the effluent, almost immediately, while U(VI) is retained by the column. Uranium can then be removed from the column with 0.1 M HCl. (AEC Activity 5311)

Chemical Physics - New paramagnetic resonance equipment was put in operation, and the atomic hydrogen hyperfine separation was measured in H_2SO_4, H_3PO_4, and $HClO_4$ irradiated at liquid nitrogen temperature with gamma rays from Co^{60}. In all cases the separation was within 1% of the atomic beam value. Atomic deuterium was also observed and measured in H_2SO_4 plus D_2O and the results also agreed with the atomic beam value.

Zeeman studies on the pure quadrupole spectra of Cl in barium chlorate were completed. No asymmetry in the field-gradient system was found, and the z component of the field gradient was located relative to the crystal axes.

The measurement of KBr, resulting from the reaction between molecular beams of K and of HBr at $\sim$500°K, was repeated. The new measurements confirm the first observation that a small but detectable number of collisions result in reaction. The shape of the product distribution curve is being more surely characterized by successive improvements in technique. Another run with K at about 500° and HBr at about 150°K also confirmed earlier indications that no presently detectable reaction occurs at these temperatures (AEC Activity 5311)

Chemistry of the Solid State - The solubilities of solid cesium chloride and iodide in liquid cesium metal at 550°C were found to be 16 and 19 mole %, respectively, i.e. somewhat lower than that of cesium fluoride, 26 mole %. However, because of their much greater temperature dependence the solubilities of the former two salts are, at lower temperatures, very much smaller than that of the fluoride. In contrast, the solubility of potassium chloride in potassium metal was found to be only slightly lower than that of potassium fluoride, reported previously, and its temperature dependence only very slightly larger. Earlier, the solubility of sodium chloride in sodium metal had been observed to be larger than that of the fluoride and to exhibit a smaller temperature dependence. A comparison of these six systems, considered in three pairs as above, shows that the partial heats of solution of the solid salts in the saturated solutions ($\Delta H_{soln.}$), as derived from the temperature dependences of the solubilities, correlate rather well with the heats of vaporization (ΔH_v) of the salts, which represent a qualitative measure of the cohesive energies. Note the parallelism in $\Delta H_{soln.}$ and ΔH_v, as the differences between the fluoride and chloride become more negative in both, when going from the sodium to the cesium system:

$$\Delta H^{NaF}_{soln.} - \Delta H^{NaCl}_{soln.} = +5.0 \; ; \; \Delta H^{NaF}_{v} - \Delta H^{NaCl}_{v} = +12.5 \text{ kcal}$$

$$\Delta H^{KF}_{soln.} - \Delta H^{KCl}_{soln.} = -1.5 \; ; \; \Delta H^{KF}_{v} - \Delta H^{KCl}_{v} = +2.5 \text{ kcal}$$

$$\Delta H^{CsF}_{soln.} - \Delta H^{CsCl}_{soln.} = -7.0 \; ; \; \Delta H^{CsF}_{v} - \Delta H^{CsCl}_{v} = -1.4 \text{ kcal}$$

This behavior is attributable to the larger polarizability of the cesium (and potassium) cation, which favors both the liquid and the gaseous states in the fluorides. (AEC Activity 5311)

Radio-Organic Chemistry - Carbon-14 isotope effects were shown to be present in the Schmidt reaction of acetone-1-C^{14} and in the auto-oxidation of benzaldehyde-α-C^{14}. The absence of an isotope effect has been positively demonstrated for the iodoform reaction of acetone-1-C^{14}.

In the distillation of labeled methanol, those molecules containing C^{14} and, to a lesser extent, those molecules containing C^{13} were shown to distill faster than the unlabeled molecules.

The carbon-14 solution scintillation counter was tested by comparing the calculated ratios of two standard benzoic-α-C^{14} acid solutions with the measured ratios agreed within one percent. (AEC Activity 5311)

Radiation Chemistry - In the radiation chemistry of water and aqueous solutions, most observations can be interpreted on the basis of the intermediate formation of H, OH, H_2 and H_2O_2 by decomposition of the water. To a first approximately, the yields of these intermediates are nearly the same in most solutions. However, it was definitely shown that these yields are somewhat sensitive to the reactivity and concentration of dissolved solute, and also

to the temperature. In 10^{-2} N H_2SO_4, the primary yield of H_2O_2 decreased from 0.75 molecules per 100 ev with no added halide ion to 0.56 with 10^{-2} M KBr and to 0.70 with 10^{-2} M KCl. The effect of Cl^- was greater in 0.8 N than in 10^{-2} N H_2SO_4 solutions. These observations were interpreted to indicate that halide ions can react with OH radicals in the particle tracks, and thereby reduce H_2O_2 formation. Likewise, some oxidants lowered the primary yield of H_2 in KBr solutions, presumably by reaction with H atoms in the particle tracks. The H_2 yield was lowered from 0.46 to 0.41 by addition of 10^{-3} M oxygen and to 0.17 by the addition of 1.5 M H_2O_2. Increased temperature lowered the primary yields of H_2 and H_2O_2. Increased temperature lowered the primary yields of H_2 and H_2O_2 and raised the yields of H and OH.

In the studies of reactions in the gas phase, a third experiment on the effect of CO_2 on the recombination of H_2 and O_2 in the presence of radon corroborates previously reported findings that CO_2 does not enter into the chemical reaction and only slightly accelerates the reaction rate. A new experiment shows that steam and CO_2 do not interact under alpha-particle irradiation. (AEC Activity 5311)

Chemistry of Fused Salts - The molar volume and equivalent conductance of the fused salt system potassium chloride-sodium chloride were determined. Although some calculations are incomplete, it is clear that the behavior of this system is very similar to that of molten potassium chloride-lithium chloride. This latter system does not, however, show strict additivity of molar volumes; there are very small positive deviations of the order of 0.7%. The equivalent conductance of this system exhibits a very shallow minimum at about 85 mole % potassium chloride, as in the potassium chloride-lithium chloride system at about 80 mole %. The minimum is interpreted as resulting from a contraction of the chloride semi-lattice of the melt on substitution of sodium ion for potassium ion, thereby hindering migration of the larger potassium ion. As in previous work, a sharp decrease in conductance was found as the temperature was lowered to a few degrees above the melting point. This effect presumably indicates structural ordering.

A radio-tracer technique was developed for determining the self-diffusion of ions in molten salts by making use of diffusion from capillary tubes. Preliminary measurements of self-diffusion of sodium ion in molten sodium nitrate at 310° and 326°C indicate that the self-diffusion coefficient under these conditions is in the range of 1 or 2 x 10^{-5} cm^2/sec. (AEC Activity 5311)

Math and Computation for Chemistry - An approximation to the energy equation for the "Cyclone Separator" was obtained which gives the kinetic energy, E(r), of a particle as a function of the distance, r, from the axis of rotation. The differential equation is

$$E'' + \left(2K + \frac{1}{r}\right) E' + \frac{2K}{r} E = -f(E', r)$$

where $f = Kr(E')^2/2E$. With the approximation $f = 0$ the solution is

$E(r) = a(1 + Kr) + be^{Kr}$ with a and b arbitrary constants. After the approximate solution has been evaluated, the function f will be expressed in terms of it and a second approximation obtained from the non-homogeneous equation resulting. (AEC Activity 5311)

Uranium Chemistry of Raw Materials - The extraction of uranium from acidic sulfate liquors with long chain tri-n-alkylphosphine oxides is increased by the addition of nitrate ion and, to a lesser degree, by the addition of chloride ion. In a 0.5 M sulfate liquor containing 1.3 g U/l, 5.8 g Fe/l and 3.5 g Al/l, the addition of 2% (w/v) NO_3^- increased the coefficient for extraction of uranium into 0.1 M tri-n-decylphosphine oxide in kerosene from 0.07 to ~200, while the same amount of Cl^- increased the coefficient to only 13. In solutions containing chloride as the only anion, very efficient and selective extraction of uranium is possible; the coefficients with 0.2 M trioctylphosphine oxide in 0.5 M chloride (pH = 1) are 260 for uranium(VI), 0.4 for iron(III) and 8×10^{-4} for aluminum, and from 4 M hydrochloric acid they are 10^4, 10^{-2} and 10^{-4} in the same order.

Development was previously reported of a process for recovery of uranium from Florida Leached Zone ore, using sulfuric acid digestion of an aqueous slurry to dissolve the uranium. Slurry-liquid extraction of uranium from the digest offers the important advantage of avoiding a difficult filtration, but it also has the disadvantage of increased opportunity for solvent loss. When the solvent was a long-chain amine (0.1 M) in a hydrocarbon diluent, used at phase volume ratio of 1:5 organic:aqueous, more than 15% of the diluent and 30% of the amine was lost to the slurry in a single contact.

The results of the extraction of uranyl sulfate from 0.01 M H_2SO_4 by didecylamine sulfate in benzene indicate that nearly complete "loading" is reached at a mole ratio of six amines to one uranium (0.1 M amine concentration). The results for uranyl sulfate extraction from 0.5 M and 1 M sulfate solutions (pH = 1) contacted with a secondary or tertiary amine in Amsco D-95 conform to approximately the same loading ratio. (AEC Activity 5361)

Volatility Studies - Recovery of uranium from reduction bomb slag by fluorination to form volatile uranium hexafluoride was shown to be feasible. Uranium losses of only 0.017% were demonstrated at atmospheric pressure and 500°C for -100 mesh slag with an initial uranium content of 1 to 3%. Fluorine consumption was limited to that used in fluorinating the uranium since the bulk of the material is unreactive magnesium fluoride.

Preliminary studies indicate that a number of possible components of fuel elements dissolve in a molten fluoride salt bath rapidly enough that an economical method may be developed for recovering uranium from low-uranium alloys. In a ZrF_4-KF-NaF (44.5-48.5-7 mole %) mixture at 675°C, through which HF was passed at a rate of 250 cc/min, penetration rates of 2 to 50 mils/hr were obtained with silicon carbide, types 304 and 347Nb stainless

steel, niobium, tantalum, manganese, mild steel, thorium, uranium, zirconium, chromium, titanium, Zircalloy-2, and 95% uranium-zirconium alloy. Substances that dissolved at too slow a rate to be practical were vanadium, silicon, monel, molybdenum, and tungsten. Tin and zinc melted and reacted instantaneously. The dissolution rate varied with the composition and/or the temperature of the bath, and suitable combinations were found to give practical dissolution rates for all substances in the zirconium-clad uranium-zirconium alloy fuel assemblies. (AEC Activity 5361)

Thorex Process Development - Decontamination factors for iodine in the extraction column were increased from 10 to 200 by pretreating the Amsco 125-90W solvent diluent with 96% sulfuric acid. This removes unstable and unsaturated materials so that iodine extraction is limited to that dissolved by the tributyl phosphate solvent. (AEC Activity 5361)

Analytical Chemistry - Investigations at present emphasize methods applicable to homogeneous reactor samples. The introduction of copper in the homogeneous reactor system required methods for copper and for uranium in the presence of copper. Interference by copper can be eliminated by its displacement with metallic cadmium, and the copper can be independently estimated by a polarographic technique with a standard deviation of 0.65%. A re-evaluation of the precision of all analytical methods used in analyzing homogeneous reactor samples is being made and will be extended to include studies on the effects of radioactivity on the precision.

A radiofrequency oscillator for measuring concentrations of highly conducting solutions, such as alkali hydroxides, without the use of internal electrodes has been developed and found sufficiently stable for process applications.

Studies are being made of the variation of temperature in the discharge in the porous-cup electrode system for spectrochemical analysis. The results indicate that the temperature near the counter electrode is nearly independent of the addition of various salts to solutions in the porous cup. This information may be useful in increasing the precision of porous-cup analyses and in minimizing foreign element effects.

METALLURGY

Ceramics Research - Petrographic data is being collected on fuel systems containing UF_3. It was found that a composition containing 50 mole % RbF and 50 mole % LiF, both of which are cubic, forms a compound that is anisotropic.

A mineralogical study on selected specimens of rocks associated with the Chattanooga shale, carried out in cooperation with the University of Tennessee, revealed possible methods for field correlation of various strata in the Chattanooga shale.

Density determinations were made on a series of specimens of thorium oxide containing 0.5%, by weight, CaO fired to 1700°C for various periods. These data show that maximum density is approached approximately exponentially and that 99% of the density obtainable is attained within two hours at 1700°C.

Initial tests on comparative fission-product loss were made on SiC-Si-UO_2 fuel sandwiches. Fission-product (xenon only) leakage was found to be reduced by a factor of 10^4 over the leakage from a specimen of powdered UO_2 or USi_3. (AEC Activity 5411)

Physical Metallurgy of Reactor Materials - The work on the carburizing of stainless steel fuel elements to improve chemical dissolution was continued. It appears desirable to use an atmosphere of methane and hydrogen to avoid the soot formation which occurred with methane and nitrogen. Carbon contents, after carburization, of up to 1.5% are desirable for complete reaction of the fuel element with the initial dissolving solution of slightly acidic $CuSO_4$. One sample of fuel plate material (0.020" core of 35 wt % UO_2 with 0.005" stainless steel cladding) was carburized and treated with $CuSO_4$ solution, HNO_3, and then with aqua regia. The uranium analyses of the various solutions were as follows:

	% Total U
$CuSO_4$ solution	0.03
HNO_3 solution	99.25
Aqua regia treatment of insoluble residue from HNO_3 solution	0.72

These results, while not typical of optimum procedures, indicate that very high uranium recoveries can be attained.

Transformation studies were continued on a 3.5 wt % Ag-Zr alloy. The phases present in this hypo-eutectoid alloy, after various isothermal transformations, were similar to those obtained in a hypo-eutectoid steel, i.e., a pearlitic structure of zirconium solid solution and $AgZr_2$, and a martensite of the same lattice structure as alpha zirconium but of larger lattice parameter. (AEC Activity 5411)

Fundamental Physico-Metallurgical Research - Studies in the alloying of Group-IVa elements were extended to a thermodynamic calculation of the idealized α/β-phase boundaries of titanium and zirconium. In this calculation the variation of internal energy of each phase with composition is assumed to be directly proportional to the valence difference of the solute and the Group-IVa element, and inversely proportional to the electron density/atom-ev at the Fermi surface of electronic distribution. The known phase boundaries were fitted to the expressions for the idealized boundaries, and the difference in the electron density of the two phases

was deduced. From the electronic contribution to the low temperature specific heat, the electron density in α-zirconium, for example, was taken to be 1.22 electrons/atom-ev, whereas the calculated value for the electron density of states of the β-zirconium is 1.18 electrons/atom-ev---showing that only slight differences in the density of electrons for the two phases are required to give a significant dependence of the phase boundaries upon solute valency.

In the study of texture development as a function of amount of deformation, the fiber axis distributions have been determined for spherical diffraction specimens machined from the center of the front and middle sections of an extruded aluminum rod, and diffraction data were obtained by the usual method. Axis distribution charts showed both specimens to have [111] and [100] as the preferred directions of the fiber axis; however, the sharpness of the texture and the volume fraction of material associated with each component were widely different. The sharpness of the texture increased markedly from the front to middle of the rod. In the specimen cut from the front of the extruded rod, the ratio [111]:[100] was 2:1, corresponding to a [111] fiber axis in 67% of the volume and a [100] fiber axis in 33% of the volume. The ratio [111]:[100] was 9:10 for the specimen cut from the middle section of the extrusion. (AEC Activity 5411)

Fundamental Investigation of Radiation Damage in Solids - The residual electrical resistivity of copper single crystals deformed at 4.2°K indicates that severe tensile deformation increases the resistivity by about 3×10^{-8} ohm-cm. Data obtained from annealing experiments seem to show that at least two types of defects are responsible for this change in resistivity. One of the defects, assumed to be a point defect (interstitial atom or lattice vacancy), has a high mobility in the temperature interval from 78° to 300°K, while the other defect, assumed to be a line defect (dislocation), has no appreciable mobility below 600°K. It would seem that the creation of point defects may be a fundamental part of the deformation process since about 10^7 of these defects are formed per centimeter of dislocation line throughout the deformation process. The temperature dependence of the residual resistivity was measured in the region from 1.6° to 4.2°K. It was found that the temperature dependence of the resistivity resulting from the dislocations introduced by the deformation was normal; that is, the resistivity was temperature independent. After these defects had been rearranged by annealing at relatively high temperatures, the resistivity became temperature dependent, with an increase of about 3% being observed when the temperature was decreased from 4.2 to 1.6°K. Apparently the arrangement of the dislocations into grain boundaries and sub-boundaries resulted in the negative temperature coefficient. (AEC Activity 5411)

BIOPHYSICS

Waste Disposal Research - A series of eight test holes were drilled in the Melton Valley area (near the X-10 site) in order to define specifically the geologic structure, to locate the shale formations most favorable for waste pits, and to determine the hydrologic conditions that would effect underground travel of radioactive liquid wastes in this area. On the basis of these data, the location for an underground tank and several tentative sites for waste pits were selected.

In a laboratory study of the feasibility of storing high-level radioactive wastes, of "HOPE" composition, in earth pits, results so far indicate that the radioisotopes are not removed from acid solutions by either filtration or adsorption on clay, but that either process removes most of the radioisotopes from solutions made basic with NaOH. The rate of movement of both acid and basic solutions through clay may be decreased by treating the clay with Na_2SiO_3. (AEC Activity 6510)

Physics of Nuclear Radiations - The drift velocity of electrons in argon, methane, and in mixtures of the two gases was measured and are reported below as a function of E/P (volts per cm per mm Hg). The method follows that of Stevenson (A. Stevenson, Rev. Sci. Instr. 23, 93 (1952) in which the time of transport of electrons between two parallel plates, between which a uniform electric field is established is measured by means of proportional counters.

TABLE OF DRIFT RATES IN CM/PER MICROSECOND

E/P	Pure Argon	99% Argon 1% Methane	75% Argon 25% Methane	50% Argon 50% Methane	25% Argon 75% Methane	Pure Methane
1.2		0.97	6.25	10.00	10.90	11.50
1.0	0.389	0.97	6.81	10.00	10.90	11.50
0.80	0.359	0.97	7.31	10.00	10.90	11.50
0.66	0.350	0.98	7.64	10.00	10.90	10.90
0.53	0.327	1.00	8.00	10.00	10.00	9.20
0.40	0.319	1.06	8.00	8.80	8.57	6.80
0.266	0.303	1.22	8.00	6.81	5.71	4.20
0.20	0.298	1.36	7.00	4.91	4.00	2.80
0.133	0.273	1.68	4.68	3.00	2.40	1.70
0.100	0.256	1.90	3.63	2.10	1.71	1.20
0.066	0.222	2.11	2.33	1.31	1.33	0.75
0.04		2.22	1.11	0.69	0.60	0.41
0.033		2.06	0.88	0.56	0.46	0.34
0.0266	0.130	1.84	0.31	0.44	0.36	

The value of W, the average energy loss for each ion pair produced, was measured for purified methane by using the recoil atoms resulting from the alpha decay of Thorium C and Thorium C'. These measurements were made in a proportional counter at pressures between one and two centimeters of mercury. The individual pulses were identified as being produced from recoils by means of their coincidence with alpha particles leaving the source in the opposite direction.

The recoil particle from the disintegration of a Thorium C' atom is a Pb^{208} atom having a kinetic energy of 169,000 ev which gives it a velocity a little less than that of an electron having a kinetic energy of 0.5 ev. The value of W found for this particle was 129 ev per ion pair which means that each particle produces about 1300 ion pairs.

The recoil particle from the Thorium-C disintegration is Tl^{208} and has a kinetic energy of 117,000 ev. Since the value of W found for this particle is 139 ev per ion pair it produces only about 840 ion pairs. The absolute values of W obtained should not be considered to have an accuracy of better than $\pm$ 10% but the relative values are probably much better than this. (AEC Activity 6510)

Physics of Radiation Dosimetry - Nuclear emulsion films having low photon sensitivity are at present included in all film badges for use in case of a disaster. Tracks are counted on some films as a routine monitoring of personnel for neutron exposure. The use of the nuclear emulsion film for also monitoring high gamma exposure is feasible, provided that cognizance is taken of latent image instability in such an emulsion. The image fading was measured by exposing films to 100 r of Ra-γ; this gave an original density of 1.52. The same density was obtained after a delay of about 5 hr before development, but 20% of the original density was lost in a 36-hr delay, 50% in 160 hr, and 67% in 350 hr. Thus, an accidental high exposure can be evaluated sensibly only when the elapsed time between exposure and development is known. (AEC Activity 6510)

Professional Health Physics Training - In April four medical officers from the Armed Forces Special Weapons program completed a six weeks training period in Health Physics at the Laboratory.

During the second half of the academic year, members of the Health Physics Division are conducting a graduate course in Radiological Physics at Vanderbilt University as part of the course work for the AEC Fellows in Radiological Physics. (AEC Activity 7620)

RADIOISOTOPE SALES AND COSTS

Type of Transaction	March	FY to Date
Domestic Sales	$ 94,102	$ 682,562
Foreign Sales	2,778	42,543
Project-Cash Sales	19,804	102,126
Project-Transfer Credits	105	4,100
Technical Cooperation Program Credits	916	7,694
Plant Credits	3,383	82,325
Civilian Defense Credits	---	390
Cancer Program Credits	30,842	178,324
*Miscellaneous Income	-17	317
Total Income	$ 151,913	$1,100,381
**Income - No production costs incurred Subtract from Total Income	1,665	23,342
Net Radioisotope Income	$150,248	$1,077,039
Radioisotope Costs	$100,857	$ 842,009
Radioisotope Shipments	1,170	9,246

*Miscellaneous Income - D_2O, Freight, Missing parts, TCP.

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

GROSS OPERATING COSTS

	Cost for March	FY 1954 Cost to Date
Programmatic Operating Cost - Net	$ 2,385,806	$ 21,318,669
Plant and Equipment Cost	239,858	2,518,351
Construction Program "H"	112,659	433,395
Work for Other Parties - Transfers	54,166	401,177
Inventory Changes	93,074*	302,554*
Reimbursable Work for Other Parties	193,049	1,242,048
Deferred Charges	453	6,167
Total Laboratory Cost - Net	$ 2,892,917	$ 25,617,253
Estimated Cost for Next Month - Net	$ 2,950,000	$ 28,567,253

*Credit

[redacted]

PERSONNEL SUMMARY

	Number of Employees April, 1954	New Hires April	Terminations April
Administration	64	1	0
Operations*	117	2	0
Engineering, Shops and Mechanical	800	23	5
Laboratory and Research	1715	32	9
Protection	150	1	0
Service	381	9	2
Total	3227	68	16

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

A total of 675 Laboratory personnel are located in the Y-12 Area.

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Previous reports in this series:

April 1953	ORNL-1529
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
February 1954	ORNL-1695
March 1954	ORNL-1703

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