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OAK RIDGE NATIONAL LABORATORY

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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

December, 1956

This status and progress report summarizes somewhat less than one-half of the Laboratory's activities. A few of the activities are reported every month but most of them are reported on a bimonthly schedule. Program 4400 is reported separately.

PROGRAM 2000 - SPECIAL NUCLEAR MATERIALS

Thorex Process - The Pilot Plant equipment was decontaminated sufficiently to permit modifications by alternate contacts with 30% HNO_3 and 20% NaOH--2% tartaric acid. Eight contacts with each reagent were needed in the feed adjustment and rework systems and 3 to 5 in the rest of the equipment. External surfaces of equipment and cubicle walls were decontaminated by spraying with various solutions and by manual scrubbing with detergents.

Conversion to operation on the co-decontamination flowsheet, i.e., co-extraction and stripping in the first cycle and partitioning in the second cycle, was completed. This permits two solvent-extraction cycles for both products in five columns, instead of the seven required by the old flowsheet. In one run with thorium irradiated to 3500 g/ton and decayed 400 days, losses and gross decontamination factors were satisfactory.

In laboratory studies on process improvement, thorium decontamination from ruthenium is critically dependent on solution acidity, especially near the neutral point. The decontamination factor was increased 10-fold by decreasing the acidity from 0.1 N H^+ to 0.1 N acid deficiency and an additional 5-fold by decreasing to 0.5 N acid deficiency. Some but not all the decontamination losses resulting from extraction from neutral or acidic feed can be regained by using an acid-deficient scrub. The addition of acid or acidified tributyl phosphate to the last two stages of the extraction section decreased the loss of thorium resulting from highly acid-deficient feed and did not greatly affect the decontamination.

Additional tests confirmed the observation that ruthenium in the U^{233} product can be converted to a form that does not sorb on cation-exchange resin by treating with $NaNO_2$ at 70-80°C. With synthetic solutions containing 0.4 g U/liter, 0.05 M HNO_3, and Ru tracer, 2 g $NaNO_2$/liter was required. When only 1 g/liter was added, 50% of the ruthenium tracer was sorbed on the resin. (AEC Activity 2423)

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In-Line Instrumentation - An in-line colorimeter in an ORNL Metal Recovery Plant test operated satisfactorily. The colorimeter continuously records the uranium concentrations, measured by light absorption, in the uranium product stream. The development and construction of a prototype of a high-sensitivity in-line a-c polarograph is 75% complete. It will monitor uranium concentration in the range of 0-10 ppm in the presence of interfering ions, such as those found in the Purex process waste. An in-line instrument for continuous measurement of uranium and acid concentrations in Purex process feed solution is also 75% complete. It measures both the specific gravity and the conductivity of the feed solution.

A prototype of an in-line radiofrequency monitor for the continuous recording of HNO_3 concentration in enriched U^{235} process feed solution is in operation with a nonirradiated test stream. A prototype in-line gamma monitor is about 75% complete. It will be used to measure continuously the total gamma radiation from specific radionuclides in those streams. Other items being developed include a recorder-controller for tributyl phosphate fraction in process solvent, an interface level indicator, and radiochemical process samplers. (AEC Activity 2724)

PROGRAM 3000 - WEAPONS

Special Separations - The fabrication of plutonium fuel elements for irradiation in the MTR to provide the heavier isotopes of plutonium is proceeding satisfactorily. Two of the first seven fuel elements were completed at ORNL and shipped to the MTR. Components for the other five were received from Los Alamos; these elements should be completed in January. Seven additional elements are being scheduled. (AEC Activity 3610)

HOMOGENEOUS REACTOR EXPERIMENT

Homogeneous Reactor Test - At the completion of the 100-hr engineering test at 280°C and 1500 psig, numerous alterations and repairs were made in preparation for continuous operation with water and natural uranium. In the course of the repairs, it was observed that several of the flanged joints were badly pitted. Metallographic examination revealed typical stress-corrosion cracks in addition to the pits. Close examination revealed several leak-detector tubes which were leaking through cracks of a similar type. The cracks were judged to be the result of the chloride contamination originally present in these tubes, and which was the reason for extensive flushing and cleaning operations during September.

It is believed that the only certain remedy for these chloride-induced stress-cracking difficulties is to replace the contaminated leak-detector lines and ring-joint flanges. While material is being obtained to make these replacements, the series of engineering tests will be continued; starting in January, the reactor system will be test-operated continuously on oxygenated water as had been previously planned. (AEC Activity 4103.3)

HRT Chemical Pilot Plant - Shakedown operations of the high-pressure system of the chemical plant were concluded with the successful demonstration that gross quantities of solids could be removed from the hydroclone underflow receiver. In the final run, 70% of the solids charged (1468 g) was recovered in the drain and two rinses of the underflow receiver; there were no indications of plugging. In two runs prior to this, when 225 g and 665 g of solids were injected, recoveries were 68 and 75%, respectively. Solids' concentrations in the solids feed loop were decreased by the hydroclone with an average half period of about 1 hr. However, the volume of this loop is only 25 liters as compared with the reactor fuel volume of 500 liters.

The design of the modifications of the low-pressure system, for dissolution of solids, is complete. Shop fabrication of components for this system, including the tantalum-lined dissolver and two 100-gal decay storage tanks, is also complete except for another sampling facility. Field installation is 50% complete and is scheduled for completion January 31. (AEC Activity 4103.1)

HRT Fuel Processing - In the presence of various mixtures of hydrogen and oxygen over simulated homogeneous-reactor fuel solutions, the stability of iodate in the solution was sharply decreased by raising the temperature of the solution from 100° to 120°C. Increasing the mole fraction of hydrogen in the hydrogen-oxygen mixtures to greater than 0.66 increased the amount of free iodine present in such solutions at 100°C. In tests at Vitro Laboratories it was shown that the presence of ferrous or ferric ion had no effect on the rate of stripping of iodine from simulated fuel solutions at 225°C by a stream of oxygen. The iodine-absorption capacity of platinized alundum pellets, identical to those to be used in the HRT, was 6.5 mg/kg of pellets at 500°C and 30 mg/kg at 350°C in an oxygen atmosphere; in an atmosphere of 25% H_2 and 75% O_2 the capacity was 29 mg/kg at 500°C and 300 mg/kg at 350°C.

No evidence for the decomposition of silver iodide in silver-impregnated alundum pellets was found at 350°C, but definite decomposition at 400° and 550°C was observed.

Zirconium oxide formed in a high-pressure loop by hydrolysis of zirconium sulfate solution was precipitated as a dense, adherent scale but precipitated ZrO_2, injected into the loop, circulated freely and was removed almost quantitatively by the hydroclone. Synthetic HRT solids were more than 99% dissolved in two cycles with 10 ml of 10.8 M H_2SO_4 per gram of solids. A cycle consists of a 6-hr reflux at 10.8 M followed by dilution with H_2O to 4 M and a 6-hr reflux; the added H_2O is then distilled off and the process repeated.

In equipment tests, flanges equipped with Belleville (washer type) springs to increase the flexibility of the bolting during thermal cycling did not leak during heating and cooling cycles. (AEC Activity 4103.1)

Plutonium Producer Blanket Processing - The behavior of plutonium when added gradually at 250°C to 1.4 m UO_2SO_4, contained in type-347 stainless steel, was the same whether the plutonium was added as Pu(IV) or Pu(VI) and whether the gas overpressure was 150 psi O_2 or 100 psi O_2 plus 200 psi H_2. All the added plutonium was found in solution as Pu(VI) until the concentration was ~ 35 mg/kg H_2O. The concentration then dropped to 5-10 mg/kg H_2O. Addition of more plutonium did not increase the amount remaining in solution. In these experiments the ratio of wall surface to solution volume was 1.75 cm^2 to 1 cm^3, and essentially all the plutonium lost from solution was uniformly adsorbed on the stainless steel wall. In one experiment plutonium addition was continued until 40 $\mu g/cm^2$ had been adsorbed. Chromium from stainless steel corrosion in these experiments was probably responsible for oxidation of Pu(IV) to Pu(VI), since it has been shown that addition of Cr(III) or Cr(VI) to 1.4 m UO_2SO_4 at 250°C under 100 psi O_2 and 200 psi H_2 acts as a catalyst for oxidizing Pu(IV) to Pu(VI).

In cooperation with the Analytical Chemistry Division, the near infrared spectra of various plutonium valence states were determined in 1.4 m UO_2SO_4. Simultaneous equations were derived which make it possible to determine quantitatively plutonium in any valence state at concentrations greater than 30 mg/kg H_2O. (AEC Activity 4103.1)

Thermal Breeder Blanket Processing - Exploratory tests indicated that ThO_2 particles in H_2O slurry have a large irreversible adsorption capacity for many ions. The anion adsorption followed the sequence $P_2O_7^{4-} > PO_4^{3-} > SO_4^{2-} > NO_3^- > ClO_3^-$; for $P_2O_7^{4-}$ the adsorption was 1600 ppm and for SO_4^{2-} it was ~ 1000 ppm. The adsorption of uranium on ThO_2 from a UO_2SO_4 solution was ~ 2000 ppm. The ThO_2 pretreated with 0.01 N H_2SO_4 sorbed less than 10% of this amount. ThO_2 pretreated with 1% Na_2SiO_3 solution adsorbed ~ 9000 ppm, but the uranium was not irreversibly held as it was on the untreated or SO_4^{2-}-treated ThO_2, being readily removed by 2 M H_2SO_4. Trace amounts of Zr^{95}, Y^{91}, Ru^{103}, and Nd^{147} behaved similarly.

Hectorite-W, a hydrated magnesium silicate, has a greater affinity for fission products than ThO_2. When equal weights of irradiated ThO_2 and Hextorite-W in water were slurried, 40% of the total fission-product activity was sorbed by the ion exchanger, together with 21% of the protactinium. However, separation of the ion exchanger and the ThO_2 was difficult. (AEC Activity 4103.1)

Homogeneous Reactor Thorium Blanket Studies--Chemical Development - Seven aqueous thorium--0.5% enriched uranium oxide slurries, containing 1500 ppm of PdO or 20,000 ppm of MoO_3 as radiolytic-gas recombination catalysts, were not noticeably affected by irradiation in the LITR at 300°C for 20, 90, 130, 192, 192, 251, and 288 hr. The first two were simple oxide mixtures; the others were prepared by thermal decomposition of the coprecipitated oxalates. Two ThO_2 slurries irradiated 130 and 355 hr were also unchanged. All the slurries contained 750 g Th/kg H_2O.

Stoichiometric H_2 and O_2 mixtures over an aqueous thorium--0.5% uranium oxide slurry (1000 g Th/kg H_2O) containing 0.05 m MoO_3 combined, in the presence of 2000 ppm sulfate (based on thorium), at a rate of 1.8 moles H_2/hr-liter of slurry at 277°C and a H_2 partial pressure of 500 psi. Activation of the slurry by heating with H_2 (1000 psi at 25°C) for 1.1 hr at 280°C increased the rate to 36.4 moles H_2/hr-liter of slurry at 170°C. At 4000 and at 6000 ppm sulfate the rate decreased to 29.4 moles H_2/hr-liter of slurry at 176°C.

In the presence of 1000 to 5000 ppm of silicate, the gross settling rates of the "standard" ThO_2 slurry were 1 to 3 cm/sec, about the same as in a sulfate system, but the settled beds were a little harder to disperse. Upon agitation at low temperatures and high silicate concentrations ($\geq$ 5000 ppm), heavy foams were formed; the foams were avoided when the mixture was preheated to 300°C. The settling rates of ThO_2 slurries containing 1000 and 10,000 ppm of aluminum, as Al_2O_3, were 0.17 and 0.13 cm/sec, respectively, after the slurries had been tumbled overnight at room temperature. The rates were decreased to 0.08 and 0.04 cm/sec, and the pH was decreased from 9.4 to 7.4 and from 9.2 to 6.8, respectively, by autoclaving the slurries overnight at 300°C. The settled concentrations after the slurries had stood for several days were 3200 and 2300 g Th/liter, respectively. Both slurries were completely dispersed by 4 hr of stirring at 98°C. As with pure slurries, the conductivities increased after stirring at 98°C.

In thoria sol development studies at Houdry Process Co., nitrate-stabilized sols became unstable at 300°C. Thoria gels containing 25-μ spheres were prepared. (AEC Activity 4103.1)

Slurry Component Development and Slurry Testing - A slurry containing 1000 g Th/kg H_2O, 2% U added as $UO_3.H_2O$, and 3000 ppm sulfate (based on thorium) has been circulating routinely in S-loop for 75 hr at 300°C and 1600 psig with oxygen overpressure.

A continuous-feed system, a flowmeter, and modified sample stations were added to the 200-A loop and the loop was put back in service for comparison tests of the influence of uranium added to the thorium oxide slurry. Loading to 1000 g Th/kg H_2O was started with a slurry containing 3000 ppm sulfate, since this is the sulfate concentration that was present during the successful long-term run A-5 (2000 hr). There is some evidence from the flowmeter and from gamma "scans" that the slurry is moving in laminar flow with slightly lower density in the top part of the horizontal piping. (AEC Activity 4103.1)

Special Slurry Problems - In studies of different methods of ThO_2 particle-size degradation (as substitutes for pumping in the 100- and 200-gpm loops), the Mikro pulverizer gave the greatest reduction in size. The mean particle diameter of a sample of oxide calcined at 800°C was reduced from 3 μ to 0.8 μ after four passes through the machine. (AEC Activity 4103.1)

Slurry Blanket System Development - In preparation for the first long-term run with slurry, the blanket test system was altered by the removal of the heat exchanger and the addition of a continuous-feed facility. Loading began with a slurry containing about 3000 ppm sulfate (based on thorium) and continued until about 500 kg of ThO_2 had been charged. This amount corresponds to about 350 g Th/kg H_2O. The system continues to operate smoothly and the slurry concentration is being raised. Evidence from sampling and gamma scans of the blanket shows that the slurry concentration in the lower hemisphere is greater than in the upper hemisphere. (AEC Activity 4103.1)

Blanket Engineering Research - Measurements, at room temperatures, of the rheological properties of the slurry, circulated for 2000 hr at 300°C in test 200-A-5, were completed. The coefficient of rigidity at any particular concentration was between the extreme values found for other similar slurries pumped for shorter times. However, the yield stress at any particular concentration was about 50% greater than the highest value found for similar slurries pumped for shorter times.

Preliminary measurement of the rheological properties of an unpumped slurry in the high-temperature viscometer in the temperature range 100° to 250°C showed essentially no effect of temperature on the yield stress (to within ±15%), while the coefficient of rigidity was doubled. These trends are consistent with previous results obtained for the temperature range 30° to 145°C. (AEC Activity 4103.1)

Core Development - The corrosion-specimen holder for the HRT core vessel was inserted into the full-scale core mockup so that the flow behavior adjacent to the holder could be observed. The dye pattern disclosed downward flow past the samples in the lower hemisphere. At 8 in. above the equator the flow was upward at all points. It is expected that under conditions of heat generation, buoyant forces will counteract the downward currents observed in these tests. (AEC Activity 4103.1)

Fuel Circulating Equipment - The three HRT-prototype fuel circulating pumps (400A-1, 400A-2, and 300A) continued to circulate 0.04 m UO_2SO_4 solution at 250° to 300°C. The 400A-1 pump has operated for 5900 hr without trouble since the thermal barrier was seal-welded. The 400A-2 pump, fitted with a 1/16-in.-wide gold-plated stainless thermal-barrier gasket, operated for 650 hr without apparent thermal-barrier seal leakage. Following this run, the effectiveness of the HRT method of handling gas accumulation in the back of the pump was demonstrated. The 300A pump, after 4730 hr of trouble-free operation in the HRT mockup, was dismantled for modifications prior to use on a high-concentration UO_2SO_4 run. The thermal-barrier seal weld was found to have cracked, and considerable bearing wear was observed. The latter was believed to have been caused by solids injected into the system. The pump was equipped with some titanium parts and the bearings were replaced, prior to being returned to operation.

The 4000-gpm Byron Jackson pump, after being operated for several days on water, was found to have a ruptured rotor can. The Inconel can was replaced with one of type-347 stainless steel.

The 20-cfm canned-rotor blower was installed and was run for 13 hr, including 50 starts and stops. At this time, noise was noted and the blower was dismantled; excessive wear of the hydrostatic bearings was found.

Seven feed-pump disphragm heads and three purge-pump heads have been operated in several development loops. The diaphragms under investigation range from 0.019 to 0.031 in. in thickness, and their hardness also varies. They have been run from 106 to 5994 hr (2280-hr average). There were no diaphragm failures during the past two months except for one purge pump at the HRT site; it failed after only 319 hr of service. (AEC Activity 4103.1)

Miscellaneous Equipment - Tests were concluded on a 7-1/2-in.-long standard freeze jacket on 1/2-in. pipe at 300°C and 2000 psi. The maximum water leak that could be frozen at these conditions was 11 cc/min. At 24°C and 2000 psi, the maximum leak rate that could be frozen increased to 78 cc/min.

A series of tests on the HRT sampler mockup disclosed that 60 to 80 liters of distilled-water flushing was required to reduce salt concentration in successive samples by a factor of 5×10^3. (AEC Activity 4103.1)

Heat Removal Equipment - A small tube bundle, consisting of nickel-plated type-347 stainless steel tubes in a type-347-clad-carbon-steel tube sheet, was thermal cycled in a water-oxygen-chloride environment. There was no visible cracking in the bundle or in type-347 stressed specimens exposed simultaneously. (AEC Activity 4103.1)

Gas Handling Equipment - The high-pressure recombiner loop was operated for approximately 100 hr to complete a 180-hr run at 500 psi and a peak recombiner temperature of 400° to 425°C. Numerous stress-cracking failures occurred in the stainless steel loop during this period, but the desired exposure of Zircaloy-2 and titanium samples was completed. (AEC Activity 4103.1)

Solution Systems Development - A 1925-hr run with 0.04 m UO_2SO_4 solution at 280°C and 1400 psi was completed in the HRT mockup without any maintenance shutdowns. The following mockup components were inspected after the run: circulating pump, heat exchanger, pressurizer, letdown heat exchanger, letdown valve, ring-joint flanges, and feed-pump check valves. Except for considerable wear and corrosion in the circulating pump and the letdown heat exchanger, the system remained sound. The mockup was reassembled, and the circulation of UO_2SO_4 at a concentration of 300 g U/kg H_2O was begun.

Flange test loop No. 2, containing eight ring-joint flanges, has been thermal cycled 18 times between 212°F and 614° to 635°F. The ferruled bolts apparently are able to keep the flanges tight if leaks do not develop in the early cycles. (AEC Activity 4103.1)

PACKAGE REACTOR DEVELOPMENT

Army Package Power Reactor (APPR-1) - The Laboratory continued to furnish technical support to the Army Reactors Branch on the 10-Mw pressurized water reactor being constructed at Fort Belvoir, Virginia by Alco Products, Inc. Over-all construction is approximately 85% complete. All primary system components are now on hand and are being installed.

Fabrication of fuel assemblies by the Laboratory proceeded satisfactorily. It is now expected that sufficient fuel assemblies for the initial reactor loading will be completed by early January.

Experiments performed by Alco Products, Inc. in their zero power facility revealed pronounced peaking of the thermal neutron flux in the region between the absorber and the fuel sections of the control-rod assemblies. It is planned that the Laboratory will add absorber combs to the end of the control-rod fuel section to diminish the flux peak, and to assure that there will not be excessive heat generation in the end portion of the central plates.

Irradiation of APPR-1 fuel and control rod absorber samples in the MTR continued. A full-scale APPR-1 type control rod absorber section was substituted for the absorber section of a MTR fuel-shim assembly. Irradiation began November 23 and will continue for several MTR cycles to approximate the absorber burnup expected in APPR-1. (AEC Activity 4201.1)

Gas-Cooled Reactor Development - In the design of the proposed gas-cooled loop for use in the ORR, an alternate design of the in-pile section, in which structural gamma heat is transferred to the reactor water, appears sufficiently promising that it is being detailed in preference to the original scheme which utilized a secondary helium coolant system. Considerable simplification and increased safety should result.

In other areas of the design, problems were encountered. The proposed means for emergency cooling of the

loop in the event of electrical power failure appeared to be more costly than warranted, and other possibilities are being investigated. It became necessary to consider alternate lattice locations because of possible interference with other experiments. Provision of means for insertion and removal of samples has proven to be a very difficult design problem, as was expected; a proposed method is being detailed, but it is not clear that it will be completely satisfactory. There is some evidence that the graphite-lubricated compressor selected for circulating the primary coolant gas may wear excessively in the proposed dry nitrogen atmosphere.

The possibility of conducting mock-up experiments in the BSR to determine nuclear characteristics of the proposed loop is being investigated. If such experiments prove feasible, the reactivity worth of the loop in various situations and the effect of the loop on thermal neutron flux in regions of interest will be evaluated. (AEC Activity 4201.1)

GENERAL REACTOR RESEARCH

Basic Reactor Research - The group-constant preparation routine (GCPR) and the reactor-constant preparation routine (RCPR) of Corn Pone, a reactor code for the Oracle, were essentially debugged. The GCPR takes a specified lethargy group structure and averages the cross sections of specified elements from the master cross section tape. It also computes slowing-down parameters for the specified elements. The RCPR uses the results of the GCPR to make up input by group and region for the integration routine. Reactor characteristics and composition are supplied to the RCPR by the user. Five of the nine codes making up the integration routine were written and typed.

A multigroup reactor calculation was coded for the Oracle specifically to make material parameter studies for the resonance-energy critical assembly. It was found that for an assembly of enriched uranium, 37.5% U^{235}, in the form of UF_4 homogenized with Teflon (CF_2) the inventory of uranium required to obtain mean fission energies in the range 1 to 100 kev would be prohibitively large. An assembly of highly enriched uranium metal plates suspended in liquid C_8F_{16} is more attractive from the viewpoint of versatility and minimum inventory. A specific calculation was made for a mean fission energy of 11 kev; an expected critical mass of 394.4 kg of uranium would be required in a cylindrical assembly 50.5 cm in radius and 93.5 cm high.

A series of experiments, designed primarily to set critical parameters of UF_6 containing enriched uranium, is in progress at the Critical Experiments Facility. The material is a homogeneous mixture of Teflon and UF_4 pressed into blocks of composition UCF_6 and a uranium density of 3.1 g/cc. The uranium is enriched to 37.5% U^{235}. Assemblies of this material without reflector or additional moderator show the critical quantity of U^{235} to be 467 kg in a 33-in.-dia by 28 in. high cylinder and 491 kg in a 29.2-in. cube. Measurements of fission rates were made at the center of the array with chambers containing U^{235} and U^{238}; the ratio of these counting rates was 25.8. Surrounding the U^{235} chamber with cadmium reduced this ratio to 25.5. These results were substantiated by observations of fission-product activities collected on

aluminum catcher foils adjacent to irradiated U^{235} and U^{238} foils. The addition of layers of graphite blocks, 2.88 in. thick, between 2-in. layers of UCF_6 reduces the critical mass of the cube to 427 ± 3 kg U^{235} at a $C:U^{235}$ atomic ratio of 42.2. Results from these experiments are being applied to the specification of materials for the core of the 10-kev (mean fission energy) critical assembly to be used in the study of nuclear properties in the resonance energy range.

Several critical assemblies were built with a hydrogenous material latticed with the UCF_6, and, as expected, the critical mass is observed to be quite sensitive to the presence of the hydrogen. The heterogeneity of these hydrogen-moderated arrays has made interpretation of the data difficult.

The photoneutron flux in the water surrounding the Bulk Shielding Reactor was calculated, and the results were compared with the results of an experiment performed earlier (ORNL-1714). There is only a 4% discrepancy between the calculated and the measured neutron flux; this is well within the calculated error.

An experiment to determine the voltage-current saturation for 50-cc and 4-cc ionization chambers was performed at the Bulk Shielding Facility. A 50-cc chamber failed to become saturated for currents greater than 10^{-6} amp, which represented an $\sim 10^4$ r/hr radiation field. Saturation for a 4-cc chamber was reached in a 10^7 r/hr field. (AEC Activity 4520)

Power Reactor Waste Disposal Engineering Studies - A series of laboratory studies of liquid-liquid extraction, using a number of compounds for the removal of fission products from an acid aluminum nitrate waste was completed. Tributyl phosphate, di-2-ethylhexyl phosphoric acid, tridecyl phosphine oxide, tri-n-octyl amine and thenoyl trifluoroacetone were found to be effective in extracting one or more of the cerium, yttrium, zirconium, niobium, and ruthenium tracers used in this study. After converting ruthenium and niobium to an extractable form, more than 99% of the tracers was extracted into undiluted tributyl phosphate. Cesium was not extracted, and strontium was slightly extracted. The extracted radionuclides were stripped from tributyl phosphate with water, dilute hydrofluoric acid and nitric acid, permitting reuse of the organic phase.

In studies of the self-sintering method for high-level waste disposal, the amount of heat required to produce one gallon of sintered cake at 900°C was calculated. One gallon of final cake would require initially 1.7 gallons Hope solution, 5.7 lb Conasauga shale, 1.7 lb limestone, and 1.7 lb soda ash. The heat required, assuming no heat loss, can be broken down into four steps; 1) heat required to raise the temperature of the mixture from 100°F to 212°F, 2) heat required to evaporate the liquid, 3) heat required to dissociate the $Al(NO_3)_3$ in the Hope waste, and 4) the heat required to raise the temperature of the cake from 212°F to 1652°F (900°C). The total heat required was calculated to be 42,150 Btu/gal of cake, of which 27,000 Btu/gal represented the dissociation of $Al(NO_3)_3$. (AEC Activity 4630)

ISOTOPE PRODUCTION

Isotope Separation - The processing of palladium with the high-temperature, electron-bombardment ion source was completed during the month of November. An estimated 12.4 grams of isotopes (masses 102, 104, 105, 106, 108, and 110) was collected. The collection of special high-purity Li^6 was completed; from meter readings the amount of the isotope was calculated to be 153 grams. A short run was made to collect high-purity Li^7. A calculated 15.9 grams was separated, of which 11 grams is expected to approach 99.995+% purity in Li^7. Collection of tin isotopes has been underway in XAX for several weeks; the estimated collection of separated isotopes totals 135 g to date. Tin has masses of 112, 114, 115, 116, 117, 118, 119, 120, 122, and 124. Collections of platinum are currently being made in XBX with approximately 38 grams of isotopes masses 190, 192, 194, 195, 196, and 198, separated so far. (AEC Activity 5121).

Isotope Chemistry - New lots of enriched B^{10}, B^{11}, Si^{29}, Si^{30}, Dy^{162}, Pb^{204}, Pb^{206}, Pb^{207}, and Pb^{208} were chemically refined and added to the inventory. Returned samples of Fe^{58}, Zn^{64}, and Zn^{66} were chemically reprocessed and made available for distribution.

Targets of $Rb^{87}F$, $K^{39}F$, and $K^{41}F$ were encapsulated in silver containers for cross section measurements by the ORNL group using the Van de Graaff time-of-flight method. Five isotopes of tin, as well as Ge^{70}, Pb^{206}, and Pb^{207}, were reduced to elemental form for various ORNL personnel.

Two thin foils of Zn^{64} and Zn^{66} were prepared electrolytically for the University of California Medical College. (AEC Activity 5121)

PHYSICS

High Voltage Program - In a study of the spectrum of protons from the $B^{11}(He^3, p)C^{13}$ reaction, energy levels of C^{13} were observed from 0- to 13-Mev excitation. Proton groups, corresponding to most of the known levels in this energy range, were seen in the spectrum. In addition, two groups corresponding to new levels appeared weakly but consistently. The new levels are at 5.49 ± 0.08 Mev and 6.09 ± 0.08 Mev.

Cross sections, measured with the BF_3 counter assembly calibrated through a Mn bath with an Sb-Be source, were completed for $Nb^{93}(p,n)Mo^{93}$ and $Co^{59}(p,n)Ni^{59}$. The cross section for Nb^{93} increases from 2×10^{-4} mb at 1.3 Mev to 1.0 mb at 2.1 Mev. Published data on the neutron spectrum show no excited states in Mo^{93} below 1.4 Mev so that this yield results from ground-state neutrons. The Co^{59} cross section (threshold 1.887 Mev) is 0.3, 1.6, and 5.0 mb at proton energies of 2.0, 2.2, and 2.4 Mev. The excited state at 0.33 Mev in Ni^{59} makes the cross section uncertain by ± 8% at proton energies above 2.22 Mev. (AEC Activity 5220)

86-In. Cyclotron, Nuclear Physics - Studies of inelastic scattering of 23-Mev protons were continued. The proton spectra from Bi^{209}, Pb^{208}, Pb^{207}, and Pb^{206} are nearly identical, as are the deuteron spectra from (p,d) reactions. The spectra from Au are markedly similar to these. The pulse height in a CsI crystal vs energy was measured for both heavy and light fission fragments. The results for the two are essentially identical, and the relationship is nearly linear. The equilibrium charge of both heavy and light fission fragments was measured as a function of energy and pressure in hydrogen, helium, air, and argon. Pressure effects become important above 10 mm Hg, and introduce a limitation into the resolution obtainable from a fission-fragment mass separator. (AEC Activity 5220)

86-Inch Cyclotron, Applied Physics - The isotopes Al^{26} and As^{74} were produced in service irradiations for outside customers and F^{18}, $Tc^{95,96}$, and V^{48} were produced for use within the Laboratory. (AEC Activity 5220)

86-Inch Cyclotron, Operation - In a test of the deflector system, eighty microamperes, or 17% of the circulating beam, was focused on a 0.6-in.^{2} area 28 in. beyond the quadrupole focusing magnets. The deflector septum is made from 0.028-in.-wall, 0.250-in.-diameter aluminum tubes flattened to 0.090 in.; it has been in operation since June 1956 without a tube failure and has operated in circulating beam currents up to 900 microamperes. (AEC Activity 5220)

63-Inch Cyclotron, Physics - When potassium is bombarded with nitrogen ions, the compound nucleus Fe^{53} is formed. The following radioactive nuclides produced in this reaction were chemically separated and identified: Mn^{52}, Mn^{51}, Cr^{49}, Cr^{48}, and Ti^{45}. The cross sections for the formation of these and other as yet unobserved reaction products will be used to check the evaporation model for deexcitation of compound nuclei. Light particles from the nitrogen bombardment of aluminum are also being investigated.

An analyzing magnet was installed on the 63-inch cyclotron; it will be used to identify heavy reaction products from targets bombarded with 27-Mev nitrogen ions. The resolving power of the magnet, as checked with Po^{210} α-particles, was found to be 0.1%. (AEC Activity 5220)

Spectroscopy Research Laboratory - The infrared spectra of the molecules NT_3, NT_2H, and NTH_2 were observed in the region 5000 to 450 cm^{-1}. Frequencies were measured with a Perkin-Elmer single-beam double-pass spectrometer. Improved relative intensities were obtained with a Perkin-Elmer Model 21 double-beam spectrometer. Approximately 0.002 mole (150 curies) of NT_3 was synthesized, allowing a maximum pressure of 10 cm Hg in a 10 cm absorption cell. The NT_3 was prepared by the reaction of T_2O and Mg_3N_2. The tentative frequency assignments for NT_3 are given in the following table:

Molecule	ν_1	ν_2	ν_3	ν_4
NT_3	2190.5	654.0	-	997.0
NT_2H	-	748.0	-	1401.0 a* 1077.5 s*
	-	743.5	-	1075.0 s*
NTH_2	-	867.0	-	-
	-	851.0	-	-

* a and s refer to the vibrations asymmetric and symmetric with respect to a plane of symmetry passing through the nitrogen and hydrogen atoms.

A feasibility study for x-ray fluorescence analysis of yttrium in solids and solutions was completed. A precision of 1% of the amount present was obtained for concentrations greater than 5% in solids and 3 g/liter in solutions. Impurity concentration ten times as great as Y concentration has little, if any, effect on accuracy.

A compilation of about 3,000 classified lines of Th II was completed. An examination and compilation of the Zeeman patterns of 200 unclassified Th II lines was based on Zeeman data from MIT.

The Zeeman spectrum of Te II in the region 3500 to 2800 Å was analyzed. The results of previous measurements in the region 3500 to 6600 Å were compiled and evaluated. (AEC Activity 5230)

Electronuclear Machines - All parts of the main vacuum system of Cyclotron Analogue I are installed and leak testing of the completely assembled system is in progress. The winding of the coils is now approximately 50% completed; techniques were developed for winding precise coils of various unusual shapes. Tests on completed coils are being prepared to ascertain the precision to which the coils produce the fields as calculated on the Oracle. Power supplies, r-f oscillator, and miscellaneous electronics equipment are on hand and are being wired into the over-all machine control circuitry. The preparation of codes for the design of Cyclotron Analogue II is nearing completion. As soon as code preparation is complete, design calculations on CA II will be initiated. (AEC Activity 5240)

Mass Spectrometry Research and Applied Mass Spectrometry - Operational tests for circuit stability, systematic errors, reproducibility of data, and linearity of ion current measurements are now complete on the newly completed 6-in.-radius research mass spectrometer, RS-1. The resolution is excellent as demonstrated by the ability to separate completely the following ion pairs: (1) C_2H_4 and CO where the mass difference is 0.0365 mass units and (2) CO_2 and C_3H_8 where the difference is 0.0728. These complete separations yield a value for the resolution ($M/\Delta M$) of over 600).

Analyses of NO, N_2, CO_2, and BF_3 samples were made for the Chemical Isotope Separations group. The N^{15} production reached a purity level of 99% for several days. The BF_3 data make it possible to plot the B^{10}/B^{11} separation factor against such things as flow rate and temperature.

A series of runs was made on an equal-isotope mixture of separated rare-earths to determine their relative ionization efficiencies at various temperatures. Two major groupings of ions were found, with some elements in one group having ion intensities one-hundredth as much as the other group.

It was discovered that ZnF^+ ions can be obtained by surface ionization. (AEC Activity 5250)

Research and Development, Stable Isotopes - The eight hot-central-wire thermal diffusion columns were divided into parallel cascades of four columns each, operating from a common reservoir of about 10 liters capacity. End volumes of 0.5 liter were attached at the light product ends. The d-c power supply is functioning in a satisfactory fashion with the diode-connected Thyratrons. In five weeks of operation there has been only one tube failure. One cascade developed a small leak but this was located and plugged. The two cascades have produced to date about one liter of 20% A^{36} at a pressure of 0.7 atmospheres. The 10-liter end volume is refilled daily to provide an effectively infinite reservoir at the feed point.

The previously-mentioned use of concentrated HNO_3 in the refinement of certain isotopes was extended to the recovery of Li^7 in the $LiCl$-$BaCl_2$ residue remaining after the thermochemical preparation of Li^7 metal. Barium nitrate is precipitated quantitatively from 76% HNO_3, with the lithium remaining in solution.

Operation of extraction columns and mixer-settlers continued to segregate ytterbium and lutetium from a rare-earth mixture. Additional data were obtained on the distribution of samarium, gadolinium, dysprosium, and yttrium, between tributyl phosphate and nitric acid. (AEC Activity 5250)

Math and Computations, Physics - The Breit-Wigner constants were determined from xenon cross section measurements. The general curve fitting code completed in March, 1956 was used in the computation. The constants determined by this calculation and by an earlier experiment were used in calculating the cross section as a function of reactor temperature.

CHEMISTRY

Raw Materials Chemistry - As previously shown (ORNL-1903), extraction of uranium(VI) by dialkylphosphoric acid is greater from hydrochloric than from sulfuric acid solution. Extraction of uranium(IV) from hydrochloric acid solution is still greater, and is decreased to a lesser extent by an increase in aqueous acid concentration, as shown by the following extraction coefficients $E_a^o(U)$ with 0.1 M di(2-ethylhexyl) phosphoric acid/kerosene.

Aq. Acid:	0.5 M	1.5 M	6 M
U(VI), H_2SO_4	17	3	<0.1
U(VI), HCl	140	13	0.2
U(IV), HCl	700	100	8

In continued basic study of design and scale-up of mixer-settler equipment for the Dapex process, the effects of operating variables on phase separation were examined in a standardized unit with 12 ft^2 of settling area. In all cases the dispersion formed a uniform band across the entire area of the settler, the thickness of the band increasing exponentially with increased flow rate (e.g., from 1 in. at 2 gpm to 25 in. at 7 gpm total flow), and decreasing slightly with increased aqueous:organic phase ratio. It was independent of degree of dispersion in the mixer (0.005 to 0.2 horsepower input per gallon), and nearly independent of entrance flow distributor configuration. The nominal total flow capacity/ft^2 of settling area was established at 3 gpm for aqueous-continuous and 2 gpm for organic-continuous operation. (AEC Activity 5310)

Equipment Decontamination - Inconel, monel, and nickel samples exposed to waste $NaF-ZrF_4$ salt at 650°C for 72 hr were readily decontaminated by removing the waste salt with molten KHF_2 or KBF_4. The ZrF_4 forms the complex K_2ZrF_6, which, being water soluble, is easily washed out of a system. Plating of fission products on the metal samples was negligible compared with the activity in the $NaF-ZrF_4$ salt clinging to the metal coupons. Analysis of the $NaF-ZrF_4$ salt showed that $Zr^{95}-Nb^{95}$ and Ce^{144} were the only major gamma activities present.

Molten KHF_2 at 275°C is better suited for decontamination than KBF_4 at 550°C, mainly because of the excessive corrosion caused by the KBF_4 salt. A 15-min contact with the KHF_2 completely removed the waste $NaF-ZrF_4$ salt. Removal of the plated activity with KHF_2 is a rate-dependent process. (AEC Activity 5310)

Chemical Physics - The vapor pressure of NaCl was measured by an effusion technique in the region of the melting point (1086°K) from 1.19×10^{-2} to 6.52 mm Hg (940° to 1228°K). The heat of sublimation obtained from a least squares calculation of the temperature dependence is 53.26 ± 0.25 kcal/mole, which is in good agreement with previous measurements which extended from 760°K to 897°K and gave 53.0 ± 0.5 kcal/mole. The heat of vaporation is determined from the data to be 46.62 ± 0.25 kcal/mole, which by difference gives 6.64 kcal as the heat of fusion; this is in good agreement with the literature value of 6.8. The heats of vaporization measured by manometric methods disagree with this measurement and it is felt that the difference occurs because of association in the vapor phase. Calculations are in progress to demonstrate that this difference can be resolved by making corrections for association. (AEC Activity 5330)

High Temperature and Structural Chemistry - An interpolation of the data obtained for the miscibility of metal with salt, in the liquid state, for the four potassium-potassium halide and corresponding cesium systems, suggested that peculiarly in the intermediate series of the rubidium systems a miscibility gap might alternately occur and be absent. This prediction was borne out thus far in the present study of the RbI-Rb and RbBr-Rb systems. A miscibility gap of more than 50 mole % was observed in the RbI-Rb system at a monotectic temperature of 615 $\pm$ 1°C, with a consolute temperature of 634 $\pm$ 1°C, approximately 14° below the melting point of the pure salt. Complete miscibility in the liquid state was found, as expected, in the RbBr-Rb system. The study of the remaining two systems is underway; a miscibility gap is expected in the case of the chloride but probably not for the fluoride, because of the relatively low cohesive energy of RbF as a result of the polarization of the Rb^+ ion by the F^- ion.

The study of the electrical conductivity of the potassium metal solutions in molten potassium chloride was followed by measurements in the sodium-sodium chloride system. The specific conductance at 880°C was found to increase from 3.83 ohm^{-1} cm^{-1} for the pure salt to 6.90 ohm^{-1} cm^{-1} for a solution of approximately 4 mole % of sodium metal. This behavior is similar to that observed in the KCl-K system. The apparent impossibility of preparing, for these measurements, solutions with metal concentrations higher than 4 mole % seems to be in disagreement with the original phase diagram according to which 9 mole % should be soluble at 880°C. Other data have suggested a lower solubility, approximately 4 mole %, in line with this latest experience. A reinvestigation of the NaCl-Na phase equilibria was started. (AEC Activity 5330)

Radio-Organic Chemistry - A new problem was started, the preparation and study of organic compounds of uranium and thorium suitable for reactor fuels and blankets. The properties being studied are solubility in molten diphenyl, thermal stability, and radiation stability. Addition compounds of some phosphone oxides with some uranyl salts possess the required solubility. As expected, thorium acetylacetonate has a high solubility in diphenyl. The rate of thermal decomposition in molten diphenyl is now being evaluated. (AEC Activity 5330)

Molten Salt Thermodynamics - As part of a continuing program on activities in molten salts, vapor pressures were determined for molten KF-ZrF_4 solutions to provide an example of pronounced negative deviations from Raoult's law associated with the interaction of univalent and tetravalent cations with a common anion. Vapor pressure was measured as a function of temperature for nine KF-ZrF_4 compositions from 35 to 100 mole % ZrF_4. In this composition range ZrF_4 was the only volatile constituent; the method, a quasi-static technique devised by Rodebush and Dixon, was most convenient for pressures from 10 to 250 mm of mercury (710°-1288°C). At 912°C, the melting point of ZrF_4, the activity coefficient of ZrF_4 is given by the empirical relation $\log \gamma_{ZrF_4} = -N^2_{KF} (0.76 + 7.74 N_{KF})$ where N is the mole fraction; from the Gibbs-Duhem equation, the activity coefficient for KF is given by $\log \gamma_{KF} = -N^2_{ZrF_4} (12.37 - 7.74 N_{ZrF_4})$. The negative deviations are associated with a

large positive excess partial molal entropy of solution of ZrF_4, and the entropy effect can be qualitatively explained as the result of substitution of non-bridging fluorides for the fluoride bridges between zirconium ions. (AEC Activity 5330)

Analytical Chemistry - Investigations on the use of di-2-ethylhexylphosphoric acid (D2EHPA) as a reagent for the separation of ions by solvent extraction revealed the feasibility of D2EHPA for the quantitative separation of the rare earths. Yttrium is apparently not extracted from a chloride solution at pH 0.5 to 1.0. This same behavior is observed for the rare earths from gadolinium through ytterbium, atomic numbers 64 to 71. Lanthanum, however, is quantitatively extracted under similar conditions, as are cerium (trivalent), praseodymium, neodymium, and samarium.

The extraction coefficients (E^o) for hexavalent chromium in acidic systems by trioctylphosphine oxide (TOPO) were found to be: $E^o \rightarrow 100$ for 1 to 4 M HCl; 29 for 1 to 3 M H_2SO_4; and 6 for 1 M HNO_3, for 1 to 8 M H_3PO_4 and for 1 to 4 M $HClO_4$. Under optimum conditions as much as 10 mg of chromium is extracted quantitatively in a single extraction with 0.5 millimole of TOPO. The formula of the extracted species was shown to be $H_2Cr_2O_7 \cdot 2TOPO$. (AEC Activity 5330)

METALLURGY

Ceramics Research - The investigation of the variables in the production of large grained UO_2 continues. It was established that the peroxide cake (one of the starting materials) varies considerably in both composition and crystallite size and habit. Two crystal forms of the peroxide cake were demonstrated.

Electrical resistivity measurements of Pt wire indicate that anomalies noted in the specific heat values of the quenched Pt, used for container material in enthalpy determinations of other materials, are real.

In relation to waste disposal studies, several new mixes are being tested for fixing both Cs-Ba^{137} and mixed fission products. A reduction in weight of the solids needed to fix the isotopes was achieved. Many new clay materials are being tested with both Purex and Hope waste solutions. (AEC Activity 5420)

Fundamental Physico-Metallurgical Research - Continued investigation of the Cd-Zr phase diagram has shown that at least 5.5 at % Cd dissolves in beta zirconium at 1100°C. The first zirconium-rich intermediate phase was found to contain 37 ± 1 at % Cd and to be of face-centered cubic structure at 581° and 900°C. X-ray examination of this phase at 865° and 870°C revealed evidence of a tetragonally deformed modification of this crystal structure which may contain stacking faults and which may indicate that cooling rate has a significant effect on the observed structure.

A new investigation of the $\beta/(\alpha + \beta)$ phase boundary of the Zr-Ag system with purer alloys has shown that this boundary is of the form $T = 873 - 13.7 \pm 0.2\ (x)$ where x is at % silver. Additional data was obtained for the lattice spacings of hexagonal Zr-Ag alloys in the range 0-1 at % Ag. These spacings can be given to within 1 part in 10,000 by the linear relations

$$a = 3.2325 - 0.0036x$$
$$c = 5.1464 - 0.0033x.$$

A transformation from the normal face-centered cubic structure of cerium to a hexagonal close-packed structure was found to start at approximately 240°K. Upon heating, the hexagonal close-packed phase started to transform back to face-centered cubic at 375°K, and the transformation was complete at 425°K. The hexagonal close-packed structure differs from that previously reported for cerium in that the c-axis is approximately doubled. The lattice constants are $a_o = 3.688$Å, $c_o = 11.92$Å, and $c/a = 3.232$. (AEC Activity 5420)

Fundamental Investigations of Radiation Damage in Solids - It has previously been demonstrated that the gamma rays from radioactive Co^{60} produce Compton and photoelectrons of sufficient energy to produce radiation damage in semiconductors. Previous measurements with Van de Graaff electrons indicates a threshold energy for the formation of an interstitial Ge atom of about 31 ev, which is equivalent to a Compton or photoelectron energy of about 0.65 Mev. Radioactive Cs^{137} emits gamma rays of 0.66 Mev; preliminary experiments indicate radiation damage is produced in high purity n-type Ge by such gammas. By means of electrical measurements the removal rate of conduction electrons was found to be only 10^{-3} of that for Co^{60} gammas, which is in turn only 10^{-3} that for fission spectrum neutrons.

Measurements of minority-carrier lifetime in high purity (7 - 15 ohm cm) n-type Germanium were made before and after irradiations in the ORNL Graphite Reactor and with a Co^{60} gamma source. Reactor irradiation to a total dose of $\sim 10^{12}$ neutrons/cm^2 caused a change in the lifetime by a factor of 50 while the carrier concentration changed by one percent. For gamma irradiation, the change was only a factor of two for the same change in carrier concentration. The change in reciprocal lifetime (recombination probability) was found to be proportional to the seven-tenths power of the integrated flux for both fast neutrons and gamma-rays.

One of the main obstacles in the path of interpreting the low temperature relaxation of irradiation effects in metals is the lack of knowledge of the order of the process. It has been assumed by numerous workers that the process is of an order greater than unity. An experiment was performed in Hole 12 cryostat of the Graphite Reactor in which specimens of copper and aluminum were irradiated at 18°K to 1×10^{16} nvt_f and 5×10^{17} nvt_f. On subsequent isothermal annealing at 55°K it was found that for both copper and aluminum the fractional recovery of electrical resistivity as a function of time was the same, within experimental error, after the two exposures, even though the integrated fast neutron flux differed by a factor of fifty. This result is indicative of a first order annealing mechanism. (AEC Activity 5430)

OPEN LITERATURE PUBLICATIONS

Billington, D. S., "Irradiation Effects in Reactor Materials", Nucl. Metallurgy 3, 31 (AIMMPE, 1956)

Binford, F. T., "A Series for the F-Functions of Shielding Computations", Nucleonics 14, 62 (1956)

Blake, C. A., C. F. Coleman, K. B. Brown, D. G. Hill, R. S. Lowrie, and J. M. Schmitt, "Studies in the Carbonate-Uranium System", J. Am. Chem. Soc. 78, 5978 (1956)

Blanco, R. E., "Processing of Power Reactor Fuels", Nucl. Sci. and Eng. 1, 409 (1956)

Bonner, W. A., and C. J. Collins, "Molecular Rearrangements. X. Rearrangement During the Deamination of 1,2,2-triphenylethylamine with Nitrous Acid", J. Am. Chem. Soc. 78, 5587 (1956)

Bresee, J. C., et al., "Damaging Effects of Radiation on Chemical Materials", Nucleonics 14, 75 (1956)

Bruce, F. R., "The Processing of Two-Region Thermal Breeder Reactors", Chem. Eng. Progress 52, 347 (1956)

Campbell, E. C. and F. Nelson, "Rapid Ion Exchange Techniques for Radiochemical Separations", J. Nucl. and Inorg. Chem. 3, 233 (1956)

Cartledge, G. H., "The Passivation of Iron by Osmium(VIII) Oxide and the Origin of the Flade Potential", J. Phys. Chem. 60, 1571 (1956)

Cohen, B. L., A. F. Cohen, and C. D. Foley, "Energy Distribution of Mass-97 Fission Fragments from Thermal Neutron Fission of U^{235}", Phys. Rev. 104, 1046 (1956)

Davis, R. J., V. L. Sheldon and S. I. Auerbach, "Lethal Effects of Gamma Radiation Upon Segments of a Natural Microbial Population", J. Bacteriology 72, 505 (1956)

Epler, E. P., "The Materials Testing Reactor", Instruments and Automation 29, 2408 (1956)

Halperin, J., and R. W. Stoughton, "Batch Versus Continuous Processing in Fluidized Nuclear Reactors", Ind. Eng. Chem. 48, 2115 (1956)

Higgins, C. E. and W. H. Baldwin, "Ester Interchange for the Preparation of P^{32} Labeled Butyl Phosphates", J. Org. Chem. 21, 1156 (1956)

Holmberg, R. W., K. A. Kraus, and J. S. Johnson, "Hydrolytic Behavior of Metal Ions. VIII. Ultracentrifugation of Bismuth(III) in Perchlorate Solution", J. Am. Chem. Soc. 78, 5506 (1956)

Householder, A. S., "On the Convergence of Matrix Iterations", J. Assoc. for Computing Mach. 3, No. 4 (1956)

Kimball, A. W., "Approximate Confidence Intervals for Specific Locus Mutation Rates", Amer. Naturalist 90, 369 (1956)

Kneip, G. D., Jr. and J. O. Betterton, Jr., "Floating Zone Purification of Zirconium", J. Electrochem. Soc. 103, 684 (1956)

Levy, H. A., "Symmetry Relations Among Coefficients of the Anisotropic Temperature Factor", Acta Crystallographica 9, 679 (1956)

Lowe, H. N., W. J. Lacy, B. F. Surkiewicz, and R. F. Jaeger, "Destruction of Microorganism in Water, Sewage and Sewage Sludge by Ionizing Radiation", J. Am. Water Works Assoc. 48, 1363 (1956)

Lyon, W. S. and S. A. Reynolds, "Alpha Counting Efficiency of a Flow Type Proportional Counter", Nucleonics 14, 44 (1956)

Ritchie, R. H. and A. Y. Sakukura, "Asymptotic Expansion of Solutions of the Heat Conduction Equation in Internally Bounded Cylindrical Geometry", J. Appl. Phys. 27, 1453 (1956)

Ropp, G. A., and E. M. Hodnett, "Isotope Fractionation Studies of Reactions with Cleavage of a Carbon-14 to Hydrogen Bond in the Rate-Determining Step", J. Chem. Phys. 25, 587 (1956)

Sworski, T. J., "Reactions of OH Radical in 0.4 M Sulfuric Acid", Rad. Res. 5, 598 (1956)

Tobias, M., and H. C. Claiborne, "The Effect of Nuclear Parameters Upon Fuel Costs in Aqueous Homogeneous Reactor Systems", Nucleonics 14, 52 (1956)

Upton, A. C., K. W. Christenberry, G. S. Melville, J. Furth and G. S. Hurst, "The Relative Biological Effectiveness of Neutrons, X-Rays, and Gamma-Rays for the Production of Lens Opacities: Observations on Mice, Rats, Guinea-Pigs, and Rabbits", Radiology 67, 686 (1956)

Weeks, R. A., "Paramagnetic Resonance of Lattice Defects in Irradiated Quartz", J. App. Phys. 27, 1376 (1956)

STATUS OF CONSTRUCTION

Construction to expand the radioactive waste gas disposal facilities was started. The work will be performed by the H. K. Ferguson Company under a cost-plus-fixed-fee contract. This project consists of alterations and additions in the stack area, in the cell ventilation system, and the off-gas system. It includes the procurement and installation of a new 60,000 cfm fan and accessories, the installation of a new motor and drive for the existing 30,900 cfm fan to increase its capacity to 35,000 cfm, modification of the duct system in the vicinity of the existing exhaust stack 3039, alterations and additions to the off-gas system, including the procurement and installation of a new 2,000 cfm high-suction fan and changes in the off-gas piping system in the vicinity of the stack, and the installation of a new 750-kva substation.

The Laboratory prepared the plans and specifications for the facilities, will procure and furnish the fans, motor drives, and prefabricated sections of the ducts, and will perform that portion of the work which requires precise scheduling to minimize interruption of research and production programs. In advance of the completion of the preliminary proposal for the project the ORNL Health Physics Division made a survey in the stack area to establish the extent of radiation hazards which would be encountered during the installation of these facilities. This survey revealed that, on occasion, several areas have a radiation background sufficiently high to limit the working time of personnel. In view of such potential hazards, it was determined prudent to select a cost-plus-fixed-fee type of contract.

VISITORS

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Smith, Cyril Stanley

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Schmid, E., University of Vienna, Austria
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RADIOISOTOPE SALES AND COSTS

Type of Transaction	November 1956	FY 1957 to Date
Domestic Sales	$194,921	$647,332
Foreign Sales	2,081	50,822
Project-Transfer	712	5,595
Project-Cash Sales	4,105	22,889
Technical Co-operation Program Credits	0	3,380
Plant Credits	9,659	24,289
AEC Credits	18,816	84,914
Total Radioisotope Income	$230,294	$839,221
Total Radioisotope Costs	$ 86,477	$575,472*
Total Radioisotope Shipments	1,141	5,551
Helium		
Income	$1,621	$9,284
Costs	$ 465	$3,148
Shipments	2	11

*$42,606 added to FY 1957 to date costs due to adjustment in October reported costs on accounting report.

GROSS OPERATING COSTS

	Cost for November	FY 1957 Cost to Date
Programmatic Operating Cost - Net	$4,475,746	$20,867,135
Equipment	208,588	1,162,062
Construction	129,751	795,682
Work for Others - Transfers	188,278	426,021
Inventory Changes	8,810	(35,153)
Reimbursable Work for Others	70,790	359,967
Deferred Charges	114,173	(260,992)
Total Laboratory Cost - Net	$5,196,136	$23,314,722
Estimated Cost for Next Month	$5,000,000	$28,314,722

() Credit

PERSONNEL SUMMARY

	Number of Employees December, 1956	New Hires December	Terminations December
Administration	72	0	0
Operations*	128	1	1
Engineering, Shops, and Mechanical	848	7	4
Laboratory and Research	2171	22	23
Protection	127	1	0
Service	390	6	0
	3736	37	28

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

929 Laboratory personnel are located in the Y-12 Area.

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November 1956	ORNL-2232

Edited by F. T. Howard

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