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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

OCTOBER 1963

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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

OCTOBER 1963

This Status and Progress Report summarizes the unclassified portion of the Laboratory's work. Some of the topics are included every month, but the majority are reported on a bimonthly schedule.

REACTOR DEVELOPMENT

GAS-COOLED REACTOR PROGRAM

EGCR Startup Preparations (a Joint ORNL-TVA Effort). — Detailed calculations of the behavior to be expected of the EGCR during the initial buildup to a fully loaded core are continuing, and the quantitative results of the various measurements to be made are being predicted as a guide in planning the startup program. The critical loadings for several control-rod configurations and the loading stages leading to the full core are as shown below:

Number of Control Rods Inserted	Number of Loaded Fuel Channels Required for Criticality	Reactivity Effect of Adding One More Fuel Element
0	46	0.0029
1	71	0.0020
4	125	0.0013
5	141	0.0013
9	193	0.0014
13	>234	0.0011

The EGCR has a total of 21 control rods, and there are 234 channels available for fuel.

During loading, the core will be open to air. With this air poisoning, the full core will have a cold, clean multiplication factor of 1.142 without control rods and 0.888 with all 21 rods inserted. The calculations also show that at least six rods, in the most effective grouping, must be withdrawn to achieve criticality in the cold, clean condition with air present at atmospheric pressure.

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Since air pressurization of the EGCR is being considered as a means of uniform poisoning in control-rod calibration experiments, calculations were performed to estimate the rate of reactivity insertion that could result from a rupture of the pressure envelope and the ensuing loss of air and to ascertain the temperature rise of the fuel during the resulting supercritical excursion. While the cases examined thus far are not necessarily the worst, it appears that a reactor scram, initiated by a flux-level trip, would protect the reactor from the fastest attainable reactivity ramp.

A pulsed neutron source will be used in the EGCR for measurement of the subcritical multiplication factor. In order to permit selection of optimum pulser operation and detector locations, a digital-computer program was used to calculate decay time constants and flux shapes for the first ten flux modes for several subcritical EGCR configurations. The computer code was developed for predicting the transient behavior of a reflected reactor in response to a series of neutron bursts injected by a pulsed neutron source. The code permits description of the reactor in cylindrical, spherical, or unsymmetrical slab geometry.

Calculations of the prompt-neutron lifetime for various partially loaded cores with control rods inserted showed large changes with changes in core configuration, as indicated below:

Number of Fuel Channels Loaded	Control Rods Inserted	Reactivity Change	Prompt-Neutron Lifetime (msec)
44	0	−0.0097	0.925
44	1	−0.0814	1.285
44	4	−0.1677	1.352
124	4	+0.0048	0.688
124	5	−0.0174	0.783

It is to be noted that the lifetime increases upon insertion of the control rods, contrary to expectations. This results from a greater than expected effect of the reflector.

Fast Gas-Cooled Reactor Studies. — As part of a study of the characteristics of a fast gas-cooled reactor, the critical enrichment and breeding ratio were computed for a series of plutonium-fueled, helium-cooled reactors in which the fuel elements were UO_2-PuO_2 clad with stainless steel. The parameters varied were the coolant volume fraction (0.3, 0.5, 0.8), the fuel-to-cladding volume ratio (1, 2, 4), and the fuel specific power (50, 100, 200 w/g). The reactor thermal power was fixed at 2500 Mw. The plutonium composition was 61% Pu^{239}, 30% Pu^{240}, 6% Pu^{241}, and 3% Pu^{242}. The reactor blanket was a 2-ft-thick layer of $U^{238}O_2$ and steel surrounding the spherical core. Results of a few of the cases studied are:

Specific Power (w/g)	Fuel-to-Cladding Volume Ratio	Coolant Volume Fraction	Critical Enrichment (% Pu)	Breeding Ratio
100	2	0.5	9.52	1.57
100	2	0.3	8.64	1.54
100	2	0.8	14.17	1.66
100	1	0.5	11.06	1.39
100	4	0.5	8.65	1.69
50	2	0.5	8.62	1.53
200	2	0.5	10.88	1.61

How much the breeding ratio will be lowered by a more realistic core and blanket geometry will be determined by calculations now in progress.

A parametric study of the heat-transfer and pressure-drop characteristics of the fast reactor was also made for He, CO_2, and CF_4 as the coolants in a core containing fuel elements clad with iron- or nickel-base alloy and operating at surface temperatures up to 1500°F. Additional calculations are being made with helium as the coolant and with higher surface temperatures to determine whether it would be advantageous to use refractory-metal cladding materials for the fuel elements.

Calculations are presently being made to determine the performance of the fast reactor with SO_2 as the coolant. The calculated thermodynamic cycle efficiency for an SO_2-cooled direct-cycle system is summarized below:

Coolant Pressure (psia)	Net Cycle Efficiency (%) at Indicated Turbine Temperature				
	600°F	700°F	800°F	900°F	1000°F
1000	26	28	30	32	34
1200	26	28	31	33	35
1400	26	29	32	33	36
1650	26	29	32	34	36
1800	26	29	32	34	36
2000	25	29	32	34	37
2600	24	28	31	34	36

For the realistic design condition of a turbine operating at 1200 psia and 900°F, the 33% net cycle efficiency indicated above compares favorably with that of most water-cooled reactor systems. The SO_2-cooled system

has the additional advantages of lower system pressure and good breeding potential. In comparison with a gas-cooled reactor of the EGCR type, the primary advantages are a reduction in plant capital cost and a high heat capacity in the core. The latter consideration is an advantage from the safety standpoint.

Microradiography as a Means of Nondestructive Evaluation of Irradiated Fueled Particles. – Two pyrolytic-carbon-coated uranium carbide particles were successfully radiographed after irradiation. Useful microradiographs that showed the layers of the particles were obtained with 50-kvp x rays and a 1-min exposure. The radiation levels of the particles measured through a glass vial were 2 and 7 r/hr, and through 1 in. of lead, 30 mr/hr. Health physics monitoring throughout handling and radiography proved the procedures to be feasible from a hazards standpoint. With this technique, layer thickness and continuity of structure can be determined nondestructively.

Calibration of Apparatus for Thermal-Conductivity Measurements of Fuel Materials at 1000°C. – In preparation for measurements at 1000°C, the thermal conductivity of Armco iron, a calibration standard, was measured in the radial-heat-flow apparatus developed for thermal-conductivity measurements on fuel materials. These and previous measurements of Armco iron gave the following linear equations for the thermal conductivity, in w cm^{-1} $(°C)^{-1}$, between 100 and 1000°C:

$$k = 0.729 - (6.27 \times 10^{-3})t, \qquad 100 \leq t \leq 410°C,$$
$$k = 0.665 - (4.70 \times 10^{-3})t, \qquad 410 \leq t \leq 780°C,$$
$$k = 0.276 + (2.31 \times 10^{-5})t, \qquad 780 \leq t \leq 910°C,$$
$$k = 0.176 + (1.21 \times 10^{-4})t, \qquad 910 \leq t \leq 1000°C.$$

The apparatus gave results reproducible to 0.1% with an accuracy of ±1.5% at 1000°C.

Measurements of Instantaneous Fission-Gas Release from Coated Particles. – Uranium carbide particles (batch GA-309) and uranium-thorium carbide particles (batch GA-310) coated with three layers of carbon were irradiated in the apparatus designed for measurements of the instantaneous fission-gas release. The three coatings consisted of an inner layer of porous graphite, an intermediate layer of laminar pyrolytically deposited carbon, and an outer layer of columnar pyrolytically deposited carbon.

The triplex-coated uranium carbide particles reached 20 at. % burnup at 2500°F and were removed from the reactor. The fission-gas release was nearly constant for the first two weeks of irradiation and then increased slowly with burnup to about twice the starting release rates. The average fractional release rates (ratio of release rate to birth rate) remained low, however:

Kr^{85m}	3.6×10^{-5}
Kr^{87}	2.8×10^{-5}
Kr^{88}	3.3×10^{-5}
Xe^{133}	1.9×10^{-5}
Xe^{135}	0.7×10^{-5}

The triplex-coated uranium-thorium carbide particles have reached a burnup of about 9 at. % heavy metal at 2050°F. The fission-gas release has remained constant and is about one-tenth that given above for the particles irradiated at 2500°F.

ARMY PACKAGE POWER REACTOR PROGRAM

PM Irradiation Test Program. — A fuel holder containing two PM-1 reactor fuel tube sections and 24 fueled flat plate coupons were tested in the Oak Ridge Research Reactor pressurized-water loop in position A-1 for one reactor cycle (eight weeks), which ended October 25, 1963. The fuel specimens operated satisfactorily during this period. During the period between cycles the fuel holder was removed from the loop and examined visually in the ORR pool with an underwater periscope. The fuel specimens did not exhibit any significant change, and therefore the fuel test assembly was reinserted into the loop for another reactor cycle.

Water Chemistry. — The new sampling system, described in previous reports, has been connected to the pressurized-water loop of the ORR. Cooled water from the loop flows into the system at a measured rate and passes through a Millipore filter, a cation exchange column, and an anion exchange column before being returned to the loop. The system was used to sample the loop water during the first day of loop operation with the new experimental assembly in position. During this time, approximately 70 liters of water was handled, and 6.1 mg of solids was collected, making an average solids concentration of 90 ppb. Another test over the next three days involved 790 liters and gave an average solids concentration of 3.5 ppb. Spectrographic and radiochemical analyses of the samples collected on the filter and in the ion exchangers are being made.

APPLIED AND BASIC REACTOR PHYSICS

Measurements of α of U^{233}, U^{235}, and Pu^{239}. — For a reactor to achieve breeding — the production of more fuel than is burned — neutron losses must be kept to an absolute minimum. Since parasitic neutron capture varies strongly with neutron energy, neutron economy can be much enhanced by properly adjusting the mean neutron energy in the reactor. But parasitic capture in the fuel itself can be reduced only if the variation of the capture-to-fission ratio with energy is accurately known. Since this ratio, called α, is known to vary wildly with energy, we are measuring it over a wide range of energy (from thermal to 700 kev), with the best energy resolution that we can achieve, for the three principal reactor fuels, U^{233}, U^{235}, and Pu^{239}.

Our most recent measurements were done at the General Atomic LINAC Facility by a method in which a pulsed beam of neutrons struck the isotope sample positioned at the center of a large liquid scintillator. The scintillator detected gamma rays emitted either by the fission process or by the capture process, the two being distinguished by the difference between

their gamma-ray pulse-height distributions. The data for U^{235} in the energy range 10 to 20 ev reflect the same fluctuations in α that we observed in earlier measurements at Rensselaer Polytechnic Institute by using a fission chamber to distinguish between capture and fission events. The values measured at GA do appear to be consistently lower than those measured at RPI, but the reason for the difference is not completely understood. When we used the fission-chamber method at GA, the results were in complete agreement with those obtained by the pulse-height method. In all sets of data, including RPI measurements from 4 ev to 2 kev, the value of α for many energies was greater than 1.

We measured α of U^{235} for the energy range 12 to 690 kev at the ORNL 3-Mv Van de Graaff by a method similar to that described by Diven et al., except for using time-of-flight techniques to determine neutron energies below 100 kev. The resulting α values are in good agreement with the earlier measurements of Hopkins and Divins, ranging from 0.368 ± 0.040 at 12.3 ± 0.8 kev to 0.097 ± 0.018 at 690.0 ± 8.4 kev.

Comparison of S_n Transport Calculations with Experiments. — Of the various computer codes now available for reactor calculations, two that are being used with increasing frequency are the S_n neutron transport codes DSN and TDC developed by Carlson et al. of Los Alamos. Although these codes, together with the 6 and 16 energy-group cross sections of Hansen and Roach, have been shown to give results that agree with experiments for a number of hydrogen-moderated critical systems, their application to some important types of systems has largely remained untested, one reason being the unavailability of experimental data with which the calculations could be compared. Falling in this category were systems consisting of right-cylindrical annuli of fuel surrounding cylindrical cores of different materials. With the recent initiation of a series of experiments on cylindrical annuli of U(93.2) metal, some experimental data have now been obtained. Thus far the experiments have involved cylindrical annuli, with inside diameters of roughly 8 in. and thicknesses of 4 to 8 in., made critical with cores of air, carbon, beryllium, or potassium. In every case, calculations with the TDC code in the S_8 approximation yielded multiplication factors very close to 1 and thus demonstrated the adequacy of the method for these assemblies. In the one case in which a potassium core was used, however, the measured reactivity coefficient for potassium was $+4.3 \times 10^{-3}$ cent/g, whereas the calculated value was -13×10^{-3} cent/g, which indicates that the cross sections used for potassium in the calculation may be in error. The 16-group cross sections were used for this case and for all other cases except those in which the core was air.

As a further check of the S_n method for hydrogen-moderated systems, the DSN code and the 16-group cross sections were used to calculate multiplication factors for several water-reflected and unreflected aqueous solutions of uranyl nitrate and uranyl fluoride which had been made critical in spherical geometry. The spheres were all constructed of aluminum with radii of 11.5 to 27.9 cm for the reflected spheres and 16 to 61 cm for the unreflected spheres. As was expected, the calculated effective multiplication factors were very close to 1, ranging from 0.9870 to 1.0166.

One calculation was also performed for comparison with an experiment in which an aqueous solution of uranyl fluoride in slab geometry was made

critical with a hydrogen reflector on all sides. The experimental slab was 5.068 cm thick, 117 cm high, and 147 cm wide. In the calculations it was necessary to assume that the slab was infinite in two directions, with the thickness corresponding to that used in the experiment. When corrected for neutron leakage in the transverse directions, the calculated multiplication factor was 0.9955, which again verifies the validity of the calculational method.

Comparison of Monte Carlo Calculations with Fast Critical Assemblies. — The Monte Carlo calculational method has been widely applied to many neutron physics problems but, because of high computing costs and the lack of adequate cross-section data, has rarely been applied to criticality studies. However, now that more detailed microscopic data are becoming available and the cost per computing operation is decreasing, it seems reasonable that this method — the only general means for calculating complex-assembly properties — should be developed as a routine calculational tool to predict the properties of critical assemblies. As a first step in this direction, Monte Carlo calculations have been performed for several simple unmoderated, unreflected assemblies using 93.2% U^{235}-enriched uranium metal, each of which had been made critical experimentally.

The computer code used for the calculations was the 05R code, a general-purpose Monte Carlo neutron transport code. Where possible, cross-section values from BNL-325 were applied, and the angular distributions from BNL-400 were used to obtain Legendre expansion coefficients. The systems considered were a 17.42-cm-diam sphere corresponding to the Godiva I reactor, a 12.7- by 12.7- by 23.15-cm parallelepiped, and 17.77-, 27.93-, and 38.08-cm-OD cylinders whose respective heights were approximately 12.6, 8.7, and 7.6 cm. The calculations yielded multiplication constants which are in agreement with each other but are about 4% higher than the measured values. The calculations are continuing, with microscopic cross sections being varied within their experimental errors to determine the effect on the multiplication constant.

REACTOR FUELS AND MATERIALS

Behavior of High-Temperature Materials Under Irradiation. — Gas samples have been obtained from the capsules containing BeO in experiment 41-8 (650 and 1100°C, 8×10^{21} neutrons/cm^2), and the helium released during the irradiation period is to be determined.

The status of the current irradiations are summarized below. Variables associated with sample fabrication have included density, grain size, and specimen size.

Samples	Temperature (°C)	Dose (neutrons/cm^2)	Status
MgO, Al_2O_3, BeO, pyrolytic graphite	200	$\sim 1 \times 10^{21}$	Irradiation completed
MgO, Al_2O_3, BeO	200	$\sim 2 \times 10^{21}$	Irradiation in progress
MgO, Al_2O_3, BeO, and single crystals	800 and 1100	$\sim 6 \times 10^{21}$	Assembled and shipped to ETR
		$\sim 3 \times 10^{21}$	To be assembled

FUNDAMENTAL HEAT TRANSFER AND FLUID DYNAMICS

Reynolds Stress Measurements. — A series of Reynolds stress measurements was made in pipe flow of water at a Reynolds number of 175,000. The $\overline{v_x v_\theta}$ and $\overline{v_r v_\theta}$ components of the stress scattered about zero at all positions, as expected. The $\overline{v_x v_r}$ component was proportional to radial distance, again as expected; however, the value extrapolated to the wall value was only one-half that expected from the smooth-tube friction-factor correlation. It is believed that this low value was the result of using a wire that was too long (0.1 in.) in comparison with the pipe diameter (3 in.). The eddies the wire sensed partially averaged out along the length of the wire. The values of the three normal stresses ($\overline{v_r^2}$, $\overline{v_x^2}$, and $\overline{v_\theta^2}$) generally followed expected trends; again, however, they were approximately a factor of 2 lower in absolute magnitude than expected for smooth pipes.

Magnetohydrodynamic Studies of Vortex Flow. — Experiments were performed that demonstrated the stabilizing influence of a strong axial magnetic field on the tangential flow of an electrolytic conductor. The flow was generated by impingement of a pair of two-dimensional wall jets on the concave interior surface of a 1.1-in.-diam tube. With no applied magnetic field, this flow situation is characterized by an apparent oscillatory boundary-layer instability, which is observed to occur at very low values of tangential Reynolds modulus and which appears to initiate general turbulence throughout the flow field, thereby seriously limiting the potential of vortex-type flow for certain advanced energy-conversion applications of current interest.

Use of a concentrated aqueous solution of ammonium chloride at temperatures up to 210°F as the working fluid and a 74-kilogauss magnetic field (double-coil large-volume test magnet in the ORNL Magnet Laboratory) enabled attainment of Hartmann moduli of up to 49, sufficient to increase the peripheral tangential Reynolds modulus for transition to instability from approximately 400 with no field to approximately 1600 with the maximum field. The observed transition Reynolds modulus is found to correlate linearly with the square of the Hartmann modulus. As a consequence of the stabilizing effect of the magnetic field, the fractional recovery of injection velocity as tangential velocity near the periphery is significantly increased. The maximum effect of the field occurs when the magnetic interaction parameter is near unity. The field is also found to be effective in suppression of boundary-layer instability on the end wall opposite the discharge opening, thereby increasing the ratio of tangential-to-radial flow on the end wall.

Numerical Calculations of Hydromagnetic Stability. — A computer program was written to solve the equations of hydromagnetic stability for viscous flow between rotating concentric cylinders. The fluid was assumed to have finite electrical conductivity and viscosity and to be flowing in the presence of an axial magnetic field. In order to check the program, calculations were made for two types of flow for which results were

already available in the literature. These are the limiting case of flow in a very narrow gap in the presence of a magnetic field and flow in a large gap with no magnetic field. In both cases satisfactory agreement, within about 0.5%, was obtained. It is concluded that the method of finite differences presently being used for the calculations is sufficiently precise to give useful results and that the computer program is now running correctly.

POWER REACTOR FUEL PROCESSING

PuO_2-UO_2 Dissolution. — Dissolution studies with unirradiated PuO_2-UO_2 fuel pellets aimed toward producing solvent-extraction feed solutions 1 M in HNO_3, thereby eliminating the need for further feed adjustment, are continuing. When pellets containing 17.8 and 20% PuO_2 in UO_2 were dissolved in boiling 5.5 M HNO_3, 80% dissolution was achieved in 2 hr; subsequently, the rate slowed markedly, and 90% dissolution required 30 hr.

Protactinium Adsorption. — Laboratory studies have demonstrated that protactinium may be recovered from thorium—nitric acid solutions by preferentially sorbing the protactinium on unfired Vycor glass powder. These adsorption experiments have been extended to determine if other adsorbents would be equal or better. Linde molecular sieves 5X, 10X, and 13X (alkali-metal aluminosilicates) adsorbed 40 to 92% of the protactinium from aqueous solutions containing 4 g of thorium per liter and Pa^{233} equivalent to 9×10^5 counts min^{-1} ml^{-1}. The solutions were 1 to 10 M in HNO_3. The feed solutions were contacted for 24 hr with 5 g of 60- to 80-mesh adsorbent per liter. The maximum adsorption occurred in the more concentrated acid solutions. By comparison, unfired Vycor glass adsorbs 70 to 95% of the protactinium under similar conditions.

Chloride Volatility. — In the chloride volatility processes being developed in the laboratory, a satisfactory method of collecting volatilized chlorides such as $ZrCl_4$, $AlCl_3$, $NbCl_5$, and $ThCl_4$ is needed. One attractive method being investigated involves collecting these chlorides in the vapor phase above refluxing ethyl alcohol, where the chlorides are first wet by the vapor and then sorbed into the solution. The alcoholic solution can then be transferred to a waste storage vessel and the alcohol evaporated and recycled.

Initial studies in these systems involved determining the boiling points of the alcohol—metal chloride solutions during concentration, the maximum chloride solubilities, and the compositions of the solid residues. The boiling points of solutions of $ZrCl_4$, $NbCl_5$, or $AlCl_3$ in ethyl alcohol increased from about 80°C, at solute concentrations of 1 M, to 125, 146, and 154°C, respectively, at 6.6 M. At 6.6 M, precipitation and foaming began in all three solutions. At this concentration, 85% of the ethyl alcohol had been evaporated and recovered. No further recovery of alcohol was possible, except for the $ZrCl_4$ solution, from which 89% was recovered. After oven drying for 12 hr at 80°C, the zirconium and niobium residues contained 0.7 and 0.6 mole of chloride and 0.3 and 1.1 moles of carbon

per mole of zirconium and niobium respectively. The aluminum residue is now being anzlyzed. The studies with $ThCl_4$ have not been completed.

In future runs, drying at a higher temperature in air, to completely convert the residues to oxides, will be tried.

Corrosion. — Corrosion studies in support of the development of the chloride volatility process were continued. In addition, alloy DC-8 was evaluated for use in the Darex process.

"A" nickel corroded at maximum rates of 2.5 mils/month on exposure to 37 cycles of oxygen (800°C, 5 hr) followed by chlorine (500°C, 5 hr). When CCl_4 was substituted for chlorine, the rate increased from 2 or 3 mils/month for the first two or three cycles to more than 15 mils/month for the fourth and later cycles.

In a three-step cycle consisting of (1) exposure to oxygen at 800°C for 4 hr, (2) exposure to Cl_2-CCl_4 at 500°C for 5 hr, and (3) leaching with 6 M HCl for 15 min at 50°C, Inconel, Hastelloy N, Corronel 230, Hastelloy B, Nichrome V, "A" nickel, and Haynes alloy No. 25 showed rates of 19 to 51 mils/month for three cycles of exposure.

Alloy DC-8 (Duriron Company) corroded at rates of 3.5 and 1.1 mils/month for a 72- and a 48-hr exposure in initial and midpoint Darex dissolver solutions respectively.

Uranium Carbide Dissolution. — Recent hot-cell studies revealed that irradiation of either impure or high-purity uranium monocarbide to 5000 to 7500 Mwd/metric ton drastically affects the chemical reactivity of this fuel material. The irradiated samples were almost unreactive in water at 80 or 90°C, whereas unirradiated material of the same original composition is completely hydrolyzed by water to uranium dioxide in 4 to 6 hr under comparable conditions. Attempts to break the passivity with very dilute nitric acid, hydrochloric acid, or hydroquinone were unsuccessful. Thus it appears that the hydrolysis-dissolution head-end flowsheet may be useful only for processing unirradiated uranium monocarbide scrap material.

An alternate head-end process was developed in which unirradiated uranium monocarbide (4.8% C) is dissolved in excess 60% HNO_3 in 4 hr, producing a dark-brown, solids-free solution about 0.42 M in $UO_2(NO_3)_2$ and 11 M in HNO_3. Degradation of organic acid impurities to forms that do not interfere with solvent extraction is achieved by extended reflux and distillation of the excess acid. During adjustment of the uranyl nitrate concentrate to prepare Purex feed solution [1.36 M in $UO_2(NO_3)_2$ and 3 M in HNO_3], the remaining organic impurities are eliminated by 2 hr of reflux boiling with 0.1 M $KMnO_4$. The feed solution is then clarified in order to remove the manganese dioxide precipitate.

Hot-cell tests of the direct nitric acid dissolution process were conducted with hypostoichiometric uranium monocarbide irradiated to 6500 to 7500 Mwd/metric ton. No difficulties were encountered which could be attributed to radiation-induced effects. Feed solutions thus prepared were batch extracted, scrubbed, and stripped under simulated Purex conditions, with only a 0.02% loss of uranium and plutonium. Decontamination from fission products appeared normal for the Purex system. The loss of uranium and plutonium to the manganese dioxide precipitate was 0.001 and 0.01% respectively.

A third head-end process for uranium monocarbide is being developed. In this process, which uses high-temperature-steam combustion, unirradiated uranium monocarbide (4.8% C) reacted smoothly at a rate of 0.2 g/min with water vapor (mole ratio $H_2O/UC = 35$) at 800°C to produce high-purity uranium dioxide containing less than 0.05% carbon. Off-gas was evolved at 100 cc/min from a 4.5-g sample. Under similar conditions with some air present, U_3O_8 is produced. After 2 hr of dissolution and refluxing of the uranium dioxide in 8 M HNO_3, a feed solution is produced that performs adequately in batch extraction and stripping tests under simulated Purex conditions. Traces of carbon collect at the interface, but this imposes no difficulties. Uranium recovery is nearly quantitative.

Preliminary hot-cell evaluation of this process with hypostoichiometric uranium monocarbide irradiated to 7500 Mwd/metric ton indicates that 98% conversion to U_3O_8 may be obtained in 5 hr. The present apparatus does not permit the exclusion of air during reaction. The oxide product contains less than 1% of the original carbon. After dissolution in 8 M HNO_3, the resulting feed solution performed well under Purex batch extraction and stripping conditions. Uranium and plutonium recoveries were nearly quantitative. The results thus far suggest that the process is free of deleterious irradiation effects.

Shear-Leach Process. — Stainless-steel-clad UO_2 and ThO_2-UO_2 reactor fuels can be sheared to 1/2- to 2-in. lengths and put into a perforated basket, which is suspended in a batch dissolver containing recirculating acid for leaching the thorium and/or uranium from the acid-insoluble clad. Basket-design features (size and shape of the perforations, percentage of free area, and permeability to liquids) are being studied for their effect upon leaching rate and equipment reliability. Leaching experiments are also being extended to the refractory sol-gel ThO_2-UO_2 material. All studies are being done with unirradiated fuel.

Several basket designs were tested in order to evaluate ThO_2-UO_2 dissolution time, ease of fabrication, and independence from air-sparging. The use of a stainless-steel-sheet basket perforated with 0.062-in.-diam holes to give 2.1% of free area and a water permeability of only 20 gal min^{-1} ft^{-2} at a 0.1-psi pressure drop resulted in a 99% dissolution time of 8 to 9 hr for ThO_2-UO_2. A wire-cloth basket (30- by 250-mesh Dutch-twilled) gave poor results; only 80% of the charge dissolved in 14 hr. Although the measured water permeability of the clean cloth was good (70 gal min^{-1} ft^{-2} at 0.1 psi), permeability to the dissolvent was greatly reduced by the "blinding" effect of small particles of ThO_2-UO_2.

Batch leaching of sized ThO_2-UO_2 sol-gel material, free of stainless steel clad, in the 10-liter batch leacher showed an extrapolated 99.9% recovery time of 60 to 70 hr for 10- to 16-mesh material. The 12 to 14 hr required for typical ThO_2-UO_2 pellet fuel was obtained only on a −200-mesh fraction of sol-gel ThO_2-UO_2. The very low dissolution rate obtained for sol-gel material indicates that operation with a fuel heel may be required.

A prototype fuel bundle consisting of 77 loosely packed Zircaloy-2 tubes (0.420 in. in outer diameter, with a 30-mil wall), filled with UO_2 pellets, was sheared into 0.5-, 1.0-, 1.5-, and 2.0-in. lengths in the 250-ton horizontal shear. Some sparking was observed (shearing is done in air), but there seemed to be no real danger of igniting either the

sheared pieces or the parent assembly. A metallurgical examination of sheared Zircaloy-2 particles that ranged in diameter from 10,000 to less than 44 μ revealed that the temperature from shearing was not high enough to form a detectable oxide skin, to induce oxygen contamination, or to anneal the grain structure. Thus there appears to be no significant safety hazard associated with the shearing of Zircaloy-2 if done slowly. The Zircaloy-2 cladding fractured (circumferential cracks and longitudinal splitting) when sheared, which allowed about 55 to 83% more of the core to dislodge than in tests with the more ductile stainless-steel-clad fuel. A maximum shear force of 92 tons was required to shear the Zircaloy-2 bundle, compared with 63 tons for stainless-steel-clad UO_2 fuel assemblies.

Fluoride Volatility Pilot Plant. — In order to test the volatility process for high-enrichment uranium-zirconium alloy fuels, high-burnup short-cooled fuel was processed in the Volatility Pilot Plant. Cooling times for the fuel used in the runs ranged from 6 to 13 months. Operation during the latter portion of the series was smooth. All systems in the plant functioned as designed, and decontamination was better than had been thought possible; several decontamination factors (DF's) of more than 10^{10} and one of more than 5×10^{10} were achieved. The technical feasibility of recovering such fuel by the volatility process was concluded to have been satisfactorily demonstrated.

There were five runs in the series, and two fuel elements were processed in each. The first run used fuel cooled 13 months. The remaining four runs processed fuel from a single reactor core. At the time of the first of these runs, the fuel had cooled six months; by the time the last run was made the fuel had cooled ten months.

Dissolution times ranged from 9.5 to 12.7 hr for 90% completion, and from 12.2 to 16.5 hr for 100% completion. Fluorination was almost complete (and greater than economically necessary) in all runs; residual uranium in the waste salt ranged from 2 to 4 ppm.

Total cationic impurities in the UF_6 product should not exceed 300 ppm (based on the uranium content), according to AEC specifications for returned UF_6. Total cations in the products from these five runs ranged from less than 171 to less than 714 ppm. (Some degree of purification will be achieved in transferring these products to a shipping cylinder.) The principal contributors to the total cationic impurities were sodium (100 to 326 ppm) and molybdenum (as high as 241 ppm). The levels of Tc^{99} (less than 1 to 31 ppm) and Np^{237} (3 to 48 ppm) were satisfactory.

Decontamination factors for the series were excellent, as shown below:

Fission Product Description	Range of Decontamination Factors	
	Overall	Final Run
Volatile	$>10^6$ to $>10^9$	$>10^7$ (Sb^{125}) to $>10^9$ (Ru^{106})
Nonvolatile	$>10^9$ to $>5 \times 10^{10}$	$>10^9$ (Sr^{90}) to $>5 \times 10^{10}$ (Zr^{95}-Nb^{95})

THORIUM UTILIZATION PROGRAM

U^{233}-Thorium Rod Fabrication. — Operation of the Fuel Rod Facility (Kilorod Facility) for the engineering-scale demonstration of the fabrication of thoria-based fuel rods containing U^{233} continued. The facility is currently being used to prepare about 1000 fuel rods for a Brookhaven National Laboratory critical experiment. As of September 30, the 240th rod passed inspection, thus completing the requirements for the second shipment of rods to Brookhaven.

In the past two scheduled production weeks, 120 fuel rods were made, which is as many acceptable fuel rods as were produced in the operating time of about four months prior to this reporting period. Maintenance and repair operations were at a minimum and were minor. The results of a material balance show that only about 0.1% of the material processed is unaccounted for.

Thorium oxide production by the pyrolysis of thorium nitrate at 500°C continued at a rate of 13 kg of ThO_2 per operating day. To date, 957 kg, or about 60% of that required to fill the BNL commitment, has been prepared.

The inventory of purified U^{233} was adequate to meet the needs of the program, so none was purified in September. A waste evaporator (P-65) was placed in service to concentrate the aqueous raffinate from the extraction column. This evaporation is necessary to concentrate the thorium-bearing stream for reuse in the preparation of solvent-extraction feed solution. Also, limited aqueous storage volume makes the volume reduction necessary.

Two hundred eighty kilograms of mixed ThO_2-$U^{233}O_2$ satisfactory for use in fabricating fuel rods was prepared. Chemical and mechanical performance of the process is excellent. This amount of 1150°C-fired mixed oxide, which contains 3.00 ± 0.05 wt % U^{233}, is about 28% of the amount to be prepared for the BNL experiments.

The equipment required to fabricate pellets for the 50 BNL fuel rods to be used in flux distribution measurements has been set up in glove boxes. The method is based on wet blending of ThO_2 and U_3O_8. Investigation of an alternative method is beginning in the Ceramics Laboratory to determine the feasibility of making pellets directly from sol-gel material instead of by the above-mentioned method of wet blending the powders. The outcome will determine which of the two methods will be used for pellet fabrication. No major equipment differences exist between the methods.

Thorium-Uranium Fuel Cycle Development Facility (TUFCDF). — Design of the TUFCDF, a facility to be used in the development of processes for remotely processing and fabricating Th-U^{233} fuel assemblies, has been in progress since January 3, 1963. The title I design effort has progressed to the point that almost all drawings and specifications will be ready for transmittal to the AEC within about a week. Drawings were given an unusually thorough review, compared with that for the ordinary preliminary proposal review. The specifications for shielded viewing windows for in-cell cranes and manipulators and for stainless steel materials and welding are adequate, with only one minor revision for final issuance. Planning, scheduling, and limited design effort are under way for the title II design of process equipment and to prove the feasibility of certain concepts and criteria set forth in title I process equipment design.

Sol-Gel Process Development. — These studies are directed toward simplifications in the sol-gel process flowsheets for the preparation of thorium and uranium oxides and carbides.

The sizing of dried gel before calcination is being tested on an engineering scale, based on the procedures developed in laboratory studies. The ThO_2 and $UO_2(NO_3)_2$ were prepared, and the vacuum drying oven was reassembled. Several 2-kg batches of sized product are being prepared.

Thoria-carbon sols prepared from sol-gel thoria and channel black mixed at 80 to 90°C were unstable and separated into a supernatant dispersion and a settled solid phase over a 72-hr period. With sols having initial mole ratios of C/Th = 4 or more, the settled phase had a constant mole ratio of 8.6. Agitation for 24 hr reduced the relative heel-to-supernatant volumes from 1/1 to about 1/8. Greater stability must be achieved to ensure good performance of the sol-gel process in the production of carbide fuels.

Development of a sol-gel process for the production of dense UO_2 fuels from uranyl nitrate solutions would significantly extend the usefulness of the sol-gel procedure. The ease of U(IV) oxidation to U(VI) leads to problems in the densification operation to form UO_2 as final product. Simultaneous reduction of uranium to U(IV) and removal of nitrate ion, which oxidizes the U(IV) in the drying and heating steps, was done electrolytically; ion exchange electrodialysis membranes and both platinum and mercury electrodes were used. Uranium oxide sols were prepared by selectively electrolyzing both nitrate ion from uranyl nitrate solutions and uranyl ion from uranyl nitrate solutions by the use of anion and cation membranes respectively. Although only 50 to 90% reduction was achieved, depending on conditions, and although the sols flocculated on drying, the product appeared to be similar to those that form high-density UO_2.

The study based on precipitation with ammonia followed by peptization by washing was continued as a means of forming U(IV) sols. Transitions from one form of U(IV) (e.g., flocculated) to another (e.g., colloidal) are very slow at pH's near 5.5, requiring up to eight days for completion.

Sol-Gel Microsphere Development. — A new medium consisting of 2-ethylhexanol-1 as the drying solvent and an ethoxydized soybean oil as the additive to prevent coalescence was developed for the formation of gel microspheres. This new system is an improvement over the previously used CCl_4—isopropyl alcohol—sorbital mono-oleate system in that it eliminates the possibility of chloride contamination of the product, the physiological hazards of CCl_4, and the problem introduced by the formation of a ternary azeotrope during distillation to remove water. The diameter of the sol droplet obtained by using a two-fluid atomizing nozzle can be varied from less than 10 μ to 1000 μ by varying the fluid flow rates. The conditions controlling the formation of gel spheres from sol dispersed in a drying medium have been delineated.

Sol-gel microspheres of thorium oxide were coated with pyrolytic carbon, and tests were conducted to determine the thermal stability of the coated product. Mixtures of fine particles of graphite and ThO_2 and graphite and UO_2 were heated to selected temperatures to establish the temperature thresholds for the reactions. Thermogravimetry was used to follow the reactions, which proceed with a weight loss due to the evolution of carbon monoxide. No weight changes were observed when the coated

oxides were heated to 1740°C, indicating no reaction. Mixtures containing uncoated oxides and graphite lost weight at temperatures under 1400°C. It appears that if reaction occurs in the coated particles the products are retained.

Dispersion Hardening of Thorium Alloys. — Hot pressings of thorium metal containing selected amounts of either thoria, indium, or zirconium were prepared in order to study the effect of these materials on thorium hardness. A problem with carbon contamination was traced to the use of CCl_4 as a surfactant in the ball-milling operation.

Irradiation Behavior of Ceramic Fuels. — The examination is continuing of thoria pellets containing 4.4% urania which were irradiated in the Materials Testing Reactor to nominal burnups of 40,000 and 68,000 Mwd per metric ton of thorium. Preliminary examination of the pellets, which were not prepared from sol-gel material, showed that none of them were intact. This fact and the general appearance of the pellets indicate relatively poor performance under the irradiation test.

The importance of dislocation movement in the release of fission gas from ceramic fuels is becoming more evident. The possibility that an increased surface area due to specimen cracking might contribute to the higher release rates observed has been eliminated. Techniques other than direct loading for inducing dislocation movement (e.g., quenching and sintering) have also produced a similar effect on the release of Xe^{133}.

MOLTEN-SALT REACTOR PROGRAM

Molten-Salt Reactor Experiment. — The design, procurement, and construction of the MSRE was about 85% complete at the end of October 1963.

The radiator was delivered to the reactor site and installed in the cooling-air duct. The coolant pump was installed, and the coolant piping was 75% complete.

The fuel pump was installed on the assembly jig. The welding was completed on the fuel and coolant piping. Preparations were made to remove components from the assembly jig and install them in the reactor cell.

The H. K. Ferguson Company continued to make progress in the installation of the auxiliary piping, instrument, and electrical services; the Ferguson portion of the work is ~35% complete.

Reactor Analysis. — Two-group two-dimensional diffusion calculations (Equipoise Burnout) were carried out to obtain flux distributions in the core and thermal shield of the MSRE with the core subcritical and an extraneous source in the thermal-shield source tube. The purpose was to determine the combination of source strength and detector sensitivity required for monitoring the reactivity during the initial loading of enriched uranium and in routine startups. Results show that as k_{eff} is increased from zero to 0.95 the thermal-neutron flux in the instrument shaft increases by a factor of 17; at the thimble located 150° from the source tube the factor is 12, but in the 120° thimble the flux increases

by only a factor of 4. These numbers reflect the contribution of neutrons reaching the detectors from the source by scattering around the gap between the core and the thermal shield. At $k_{eff} = 0.95$ the servo-driven fission chambers (6 in. long with a counting efficiency of 0.026 count neutron^{-1} cm^{-2}) in the instrument shaft give a count rate of 2 counts/sec with an external source of 7×10^6 neutrons/sec. Long BF_3 chambers in the instrument shaft would give a count rate approximately 200 times as high.

Development of Components and Systems. — Operation of the engineering test loop was continued for testing of the sampler-enricher mockup. Salt of MSRE fuel composition has now circulated through the graphite in the loop for 4000 hr at 1200°F. Fourteen samples were taken from the pump bowl under representative operating conditions, and for all but two of the samples the capsule was clean of exterior contamination by salt; for these two the capsule had only milligram quantities in scratches on the surface. Chemical analyses of the samples agreed with the analyses of samples taken with the method previously developed for use with nonradioactive systems. One capsule of enriching salt was added, also under simulated operating conditions. The 125 g of salt (76 g of uranium) melted out of the capsule in 5 min with only one small drop of less than 0.2 g remaining.

An analytical model was completed to describe a test of the stripping of CO_2 from water that contains no bubbles. The CO_2-water test was designed to provide experimental information about the efficiency of stripping of gas from liquid in the pump bowl for use in evaluating xenon migration in the MSRE. A stripping experiment was conducted at a reduced flow rate through the equipment to keep the liquid free of bubbles, and the correlation between the model and the experimental results was good. Future tests will be made to determine the effects of bubbles and variations in the liquid and purge-gas flow rates through the pump bowl.

The prototype control-rod drive, thimble, and rod assembly were operated continuously at a 1200 to 1300°F thimble temperature through 17,438 complete cycles (102 in. total travel up and down) and for more than 1000 scrams. All components operated satisfactorily except for the gear-reduction portion of the drive unit. The aluminum-bronze worm gear was worn completely out owing to lack of lubrication. This portion of the reduction unit is being redesigned.

Testing of the modified freeze valves was continued to establish operating conditions for use with the automatic control circuit. The automatic controls are such that the average melt time is approximately 10 min, and the freeze time is 15 to 20 min; these conditions are acceptable for use with the MSRE.

Several maintenance tools were received, and testing was started. Included were tools for handling the leak-detector disconnects, heaters and their electrical disconnects, and thermocouple disconnects; an offset pipe-aligning tool for freeze flange 100; and the revised lower end of the clamp-operator tool. All the major long-handled tools for the MSRE have now been delivered. Procedures for the preoperational testing of the maintenance capabilities are being prepared.

MSRE Pump Program. — Operation of the MSRE prototype fuel pump was stopped after 370 hr of circulating molten salt $LiF-BeF_2-ZrF_4-ThF_4-UF_4$ (67.9-24.9-5.3-1.0-0.9 mole %) when it became evident that the annulus between the shaft and the shield plug was plugged. Inspection revealed frozen salt in the annulus, the presence of which is believed to be due to misoperation of the purge-gas flow down the annulus. During the test the gas pressure in the pump tank was reduced from a high value by stopping the flow of purge gas to the annulus. This permitted gas and droplets and aerosols of salts to flow into the relatively cold annulus.

The prototype pump was cleaned and reinstalled and is being used to conduct proof tests of the lubrication stands and the supports for the fuel pump for MSRE.

A pneumostatic test indicated the presence of leaks in the cooling coil of the first vessel being fabricated for the drive motors of the MSRE pumps. The method of attachment of the cooling coil was changed from silver brazing to welding; another coil has been attached and the vessel is now undergoing stress-relief heat treatment. The three other vessels are ready for attachment of the cooling coils.

Instrumentation Development. — Performance of the ball-float level transmitters on separate installations in the level test and MSRE prototype pump test facilities continues to be satisfactory. Twenty months of operation have been accumulated on the installations in the level test facility. The device on the prototype pump has been operated for approximately three months. During this period the instrument underwent several shutdown and startup cycles.

Tests are being run to determine the best method of installing thermocouples for measurement of molten-salt temperatures in the MSRE drain tanks. An assembly was tested that simulated a design in which mineral-insulated sheathed thermocouples are attached to the inside of a 1-1/2-in.-OD by 0.065-in.-wall tube inserted in a 1-1/2-in. sched-40 bayonet tube well in the drain tank. The thermocouple was attached at the hot junction by means of a special fitting. Under steady-state conditions, there was no observable difference in the reading of a thermocouple located on the outside of the bayonet tube and the test thermocouple after corrections were made for thermocouple calibration errors.

A device has been developed, for use with Foxboro ECI pressure transmitters, that will provide the secondary containment barrier required when the transmitters are connected to the reactor primary system. The device, which consists of a floating-diaphragm assembly in a housing, is connected to the reference port of the pressure transmitter and is referenced to atmospheric pressure. During normal operation the device acts as a free-floating slack diaphragm and introduces less than 0.1% error in the transmitter signal. If the bellows assembly in the transmitter ruptures, the device will contain the released radioactive gases. Use of this device eliminates the need for several radiation monitors and safety block valves.

Work was started on the development of a system for obtaining a single-point indication of the position of the MSRE control rods. This index signal is required in order to correct the reading of the control-rod position indicator for errors introduced by creep in the flexible metal hose which supports the poison elements and is connected to the rod

drive. A system is being investigated which measures the pressure drop of control-rod cooling air across an orifice in the control-rod thimble. When the rod is near its lower limit, the area of the orifice decreases sharply and the pressure drop increases. A system of this type, installed in the MSRE rod-drive test facility, produced usable signals when operated with a single rod under out-of-pile conditions. The cooling-air piping is being revised to simulate three-rod operation. Further tests will be run to determine whether revisions in the reactor-system cooling-air piping will be required to obtain usable signal levels under reactor conditions.

A NaK-filled high-temperature differential-pressure transmitter was developed which meets the requirements for use in the MSRE system. Development of this transmitter was a cooperative effort between Taylor Instrument Company and ORNL. Three transmitters were fabricated and tested. Performance of the instruments is within the original ORNL specifications.

A marker system was developed for use with the MSRE temperature scanner system; the system will enable the operator to quickly identify any one of the 99 points in the temperature profile presentation on the oscilloscope.

NUCLEAR SAFETY

Melting Experiments in TREAT Facility. — Two experiments were performed in the TREAT reactor to study the release of fission products from fuel melted under transient heating conditions. The miniature fuel specimens employed in these experiments contained 30 g of 10%-enriched high-density UO_2 clad with 35-mil-thick type 347 stainless steel. The specimens were preheated electrically to 800°C in a purified argon atmosphere and exposed to different reactor transients. A 330-Mw-sec transient at a period of 108 msec was used in the first experiment and 338 Mw-sec at a period of 87 msec in the second experiment.

The stainless steel cladding melted away from both UO_2 specimens. No significant degree of fragmentation of UO_2 occurred in either experiment, in contrast to the shattered specimens produced in earlier ANL experiments of a similar nature. The UO_2 pellets in the first experiment increased 2% in diameter and exhibited considerable surface cracking, but there was no noticeable evidence of flowing of molten UO_2. The pellets in the second experiment increased 18% in diameter, and approximately 10% of the UO_2 flowed out of the pellets. Metallographic examination showed that the "melted" UO_2 was very porous (or "foamy"), but it did not have the large columnar grain structure typical of melted UO_2 fuel pellets. Determinations of the release of fission products and the transport of particles are in progress.

Development work has been completed for adapting the experimental system for the use of dry-air and steam-air atmospheres. Stainless-steel-clad UO_2 specimens identical with those used in the first two experiments will be employed in experiments 3 and 4, and the specimens will be heated electrically to 800°C in dry-air and steam-air atmospheres, respectively, before exposure to the reactor transients.

Innocuous Simulation of Accidents and Hazards. — A new program has been started to develop ways of simulating the radioactive materials which would be released during a reactor accident, making use of test materials which are not hazardous themselves. Such innocuous simulants would be of great value for investigating the transport of materials by gases in reactor installations. Tests could be carried out in the installation after construction, and possibly during operation, to determine the behavior of released fission products in the various stages of containment and in gas-cleaning equipment. Simulants must be found for the various forms of fission products, including molecular dispersions of the appropriate elements and their compounds and particles of sizes which are produced in reactor accidents.

A simulant for radioactive fission-product iodine has been developed. Natural iodine in the elemental form is released into the gas system being tested, and the amount of iodine at various points is determined by activation analysis. This method has been used for in-place testing of iodine adsorbers on the NS "Savannah." Additional and faster methods of analysis of natural iodine are being investigated. The search for methods of simulating other fission products and other forms of iodine will continue.

Release of Fission Products on Out-of-Pile Melting of Reactor Fuels. — Results of a previous test of the proposed ORR charcoal and absolute filter containment system on a laboratory scale were reported in the Nuclear Safety Progress Report for June 30, 1963 (ORNL-3483, p 35). Interest was centered on retention of iodine by two beds of activated charcoal in series and on possible indications of lower iodine pickup obtained in tests with melted uranium-aluminum alloy fuel as the source of radioiodine. This test showed highly satisfactory performance of the system, with an overall efficiency of 99.99%.

An extension of this test has been conducted in the Hot Cell Containment Mockup Facility, which permits more realistic environmental release conditions, including selected aging periods for the released aerosol in a large stainless steel vessel. Provisions for additional studies are included in the Mockup Facility, such as fission product plate-out, steam condensate washout, and aerosol particle analysis. Only preliminary results are available from the first two tests, made with highly irradiated, long-cooled uranium-aluminum alloy heated to 750°C in air. The total gamma activity as a measure of fission product release reaching the containment vessel was less than 0.5% (excluding xenon and krypton), and this activity was essentially all Cs^{137}. Of this, almost two-thirds remained airborne for 2 to 3 hr and only one-third plated out on the inside of the holdup tank. An apparent efficiency of the three-part filter pack for the Cs^{137} aerosol was determined from the amount of activity on the prefilter and the activity on the two absolute filters in the first test. These values were 1.5×10^9, 3.9×10^6, and 1.8×10^3 dis/min, or almost a 10^6 to 1 ratio between the first and the third filter. No particle-size data were obtained in the first test, but particles collected during the second test by an electrostatic precipitator on a specially prepared screen were found to be spherical and to have diameters ranging from 0.03 to 1.5 μ. It was also found that the ratio of the amount of iodine on the first charcoal bed to that on the second had decreased to 20 to 1 in the first test (instead of 4000 or 6000 to 1 in the laboratory-scale test)

after 1 hr of operation with an air flow of 3 liters/min (velocity 50 fpm) through the 0.6-in.-ID beds. However, in the second hot-cell test, conducted under better experimental conditions, including a higher level of iodine activity, a 60 to 1 ratio was found between the first and the second charcoal bed, and the amount of iodine found in two backup charcoal beds indicated an overall efficiency of 99.86% for the first two filters and two charcoal beds.

Release of Fission Products on In-Pile Melting of Reactor Fuels. — Radiochemical analyses of the eleventh experiment on the in-pile melting of miniature UO_2 fuel elements have been completed. The percentage release values are as follows:

Origin	I^{131}	Te^{132}	Cs^{137}	Ru^{106}	Sr^{89}	Ba^{140}	Zr^{95}	Ce^{144}	U
Fuel[a]	97.7	99.4	96.3	48.4	18.3	26.3	12.3	7.58	5.08
High-temperature zone of furnace[b]	89.0	69.3	86.4	0.198	2.93	8.01	0.568	1.24	1.64

[a]Includes miniature fuel element and holder.

[b]Includes fuel and thermal insulation; minimum temperature, 1000°C.

The tenth and eleventh experiments were performed to study how the helium sweep-gas flow rate affects fission product release. The flow was increased from 400 cm^3/min, used in previous experiments (3–9), to 800 cm^3/min in experiment 10 and to 4000 cm^3/min in experiment 11. Other conditions remained the same as in previous experiments. The conclusions regarding the effects of flow rate are: (1) An increased helium flow rate lowered the maximum temperature of the fuel. On the basis of the extent of melting and the appearance of the fuel, the maximum temperature of experiment 11 was between those of experiments 7 and 8, which did not melt completely. (2) Release of fission products from the fuel itself did not appear to be affected by flow (except as flow affects temperature), since the release values for experiment 11 fell between those of experiments 7 and 8. (3) Release of the more volatile elements iodine, tellurium, and cesium from the high-temperature zone of the furnace did not appear to be affected by an increased flow rate, but the release of the less volatile elements increased fourfold with a tenfold increase in rate. (4) The above effects are apparently insignificant for moderate increases in flow rate, since they could hardly be detected in experiment 10.

Analysis of the distribution of released fission products and uranium in diffusion tubes yields information about gas-borne particles in the diameter range 10 to 100 A. Previous calculations of particle diameters and associated percentages of material (ORNL-3377, p 18) have been refined, and new calculations have been performed for all other experiments except experiment 11. The average diameter found by this technique was

25 A. The amount of fission products associated with these particles expressed as a fraction of the release from the furnace and averaged over all the experiments was 15 to 35% for iodine, tellurium, and cesium and 0 to 10% for the others. For most UO_2 melting experiments, no ruthenium, zirconium, or cerium was associated with millimicron particles. In the case of uranium carbide—graphite burning experiments, ruthenium, strontium, barium, and uranium were not detected by this technique.

Characterization and Control of Accident-Released Fission Products. — It has been previously mentioned that compounds of iodine are observed to occur in air along with the elemental form when air and what is presumed to be I_2 vapor are mixed. Since the behavior of these compounds could greatly affect the operating efficiency of iodine removal systems, considerable emphasis is being placed both here and elsewhere on the determination of the origin and identity of these compounds. As has been reported earlier, the diffusional deposition technique gave a diffusion coefficient (at 25°C and 1 atm) for one of the iodine compounds in air of about 0.05 cm^2/sec, which corresponds to a molecular weight in the vicinity of 300, and for I_2 about 0.08 cm^2/sec, which corresponds approximately to its known diffusion coefficient. The value 0.05 was obtained from the profile for the deposition of I^{131} activity in a rubber tube, and the 0.08 in a silver-lined tube. More recently, tubes lined with powdered activated carbon have been used to measure the diffusion coefficient of the compound or compounds not retained by a rubber or a silver surface. This species exhibited a diffusion coefficient of 0.10 cm^2/sec, indicative of an iodine compound having a relatively low molecular weight. An estimated value for HI in air is 0.115 cm^2/sec, so whether or not the value 0.10 reflects HI is questionable. In this connection, diffusion coefficients of known compounds will be determined by means of the diffusion tube method so as to provide more positive identification.

In a related study, equilibrium partial pressures of iodine and iodine compounds corresponding to various reactions and reactant concentrations are being calculated from thermodynamic data. The problem is not simple, inasmuch as some of the iodine concentrations of interest are extremely low and the chemical form and behavior of the iodine could vary markedly with the levels of the impurities found in air.

Nuclear Safety Pilot Plant. — All designs for the Nuclear Safety Pilot Plant have been completed, and construction is nearing completion. Installation of equipment and process piping is nearly complete; approximately one week's work remains. The electrical power system is approximately 60% complete; all conduit work is done, tie-ins to the building bus have been made, and wiring of the plasma torch power supply has been completed. Cable runs to other components in the cell remain to be done, plus installation of wiring in much of the conduit outside of the cell and tie-ins of all components other than the plasma torch.

Instrument installation, which was held up pending the installation of the facility proper, is now under way. The control panel was installed in the building on October 9. All internal piping and wiring were installed in the shop. Tubing runs and panel-to-instrument wiring remain and will be completed in November.

A hazards review of the Nuclear Safety Pilot Plant has been completed, and approval of the proposed operation has been given by the Laboratory Director's Review Committee.

A memorandum describing the experimental program is being prepared.

Reactor Containment Handbook. — A target date of December 1 has been set for the completion of the rough drafts of the three remaining chapters of the handbook — chapters 1, "Introduction"; 6, "Credible Accidents"; and 12, "Containment Research."

Chapters 2, "Codes and Criteria," and 4, "Energy Sources," have received external review and are being revised. Chapter 3, "Radioactivity Generation, Release, and Transport," chapter 5, "Analytical Techniques," and chapter 8, "Specific Containment Systems," are all completed and are in the process of being published for external review.

Chapters 7, "Containment Proof Tests"; 9, "Design Details"; 10, "Containment Accessories"; and 11, "Economics," are being furnished under subcontract by the Bechtel Corporation. Preliminary drafts of all four chapters have been prepared and have been reviewed internally. One copy of the final draft for external review of chapters 7 and 9 has been reviewed and returned with comments; similar drafts of the other three chapters are expected by November 1, and copies for external review of these four chapters will be available around the first of the year.

Nuclear Safety Information Center. — A considerable portion of the effort of the newly established Nuclear Safety Information Center continues to be directed toward the collection and storage of nuclear safety information that has been generated over the past few years. A full-time information specialist was added to the Center's staff in September 1963. This brings the staff to the equivalent of six full-time individuals, including four technical persons.

The Center has received and answered 19 technical inquiries since its inception. In addition, it has received 38 requests to be added to the distribution of reports prepared by the NSIC and 28 requests for additional information on the functions of the Center.

The Center's second report (ORNL-NSIC-2), a compilation of nuclear safety contracts, has been issued. It is intended to serve primarily as a guide for the Center's specialists and as an AEC management tool. Three other reports are well along and should be issued early next year. They include (1) radioiodine release and monitoring at four major AEC facilities, (2) safety problems in fast gas-cooled reactors, and (3) iodine release and transport phenomena within containment structures.

WASTE DISPOSAL

Clinch River Study. — Dye tracer tests were conducted to determine the effects of power releases from Melton Hill Reservoir on the dispersion of radioactivity released from White Oak Creek. In the most significant test the release of the dye at White Oak Dam continued for 168 hr. On the weekdays of the test a typical power release was made; there were no releases made on the weekend. The twice-daily power releases of 16,000 and 8000 cfs created flow conditions in the White Oak Creek embayment analogous to flow in a tidal estuary. As a result, discrete twice-daily pulses of dye were released to the river. After 58 hr of no flow over the weekend, the dye was observed to extend over a 2.5-mile reach of the

river. The peak concentration resulting from this buildup of dye in the river on the weekend was considerably higher than the twice-daily peaks observed on other days at the measuring station, 6.4 miles downstream from White Oak Creek.

Mineral Exchange Studies. — Cesium was desorbed from six representative clay minerals containing trace levels of Cs^{137}. The cesium on both kaolinite and montmorillonite was completely desorbed by 1 M sodium acetate after contact for 1 hr. When 1 M HNO_3 was used, the hydrobiotite and biotite released all of their cesium; both illite and muscovite retained 75 and 85% of the cesium even after 24 hr of contact with the nitric acid. Since acids are known to decompose the clay minerals, the difference in release by the minerals reflects the ease of their decomposition. The trioctahedral biotite and hydrobiotite are more susceptible to acid attack than the dioctahedral illite and muscovite. Similar desorption tests on sediments from the White Oak Lake bed and from the Clinch River suggest that the cesium associated with these sediments is bound in dioctahedral micaceous minerals.

Comparison between Dowex 50-X4 and alumina (F-20) shows that sorption of trace strontium is more efficient by alumina in simulated waste solutions whose sodium concentration is above 0.1 M and when the strontium (and calcium) concentration is less than 10^{-4} M.

Disposal in Deep Wells. — The complete drawings for the shale fracturing facility have been issued. The injection pump and bulk storage tanks have been received; construction of the facility has been started. For each injection, Halliburton Company, Duncan, Oklahoma, will furnish six operators, one mechanic, and two supervisors to handle the dry materials, operate the injection pump, and handle the standby pump. Halliburton will be responsible for maintenance and repair of the injection pump after ORNL forces have done the necessary decontamination.

A third water injection test was started; 50,000 gal of water with a fluid loss additive was injected into a fresh fracture about 20 ft above the previous fracture. The object of the test is to discover whether the water loss observed in the two previous injections was by a mechanism that would be affected by a fluid loss additive (through hairline cracks in the shale, for instance).

The 2-7/8-in. tubing string was placed in the well, and the previous fracture was plugged with cement. After the plug had set, the jet gun that will be used in subsequent injections was used to cut the slot in the casing. A sand-water slurry was pumped for 20 min, the well was backwashed, and the jet was recovered. Pressure (to 4500 psi) was applied to fracture the formation, but no fracture occurred. It was concluded that the casing had not been completely cut; so the slotting operation was repeated — this time for 1 hr. When pressure was applied again, the formation fractured at 2000 psi. Injection pressure was 1170 psi at an injection rate of 340 gpm.

Foam Separation Studies. — The scavenging-precipitation foam-separation process for decontaminating low-radioactivity-level process waste water includes the following steps: precipitation of hardness by making the water 0.005 M each in NaOH and Na_2CO_3 and adding 2 to 10 ppm Fe^{3+}, removal of the resulting solids in a gently stirred sludge column, and

foam separation in a countercurrent column with sodium dodecylbenzenesulfonate as surfactant. This process is very effective in removing strontium (Sr^{90}) and rare-earth ions; however, less than 50% of the Cs^{137} present in ORNL process waste water is removed. For this reason several alternative methods for removing cesium are now being studied. The first of these is the addition of illite clay (which strongly sorbs cesium) to the water before the caustic and carbonate are added. Besides the clay, two other materials were tested for their cesium-sorption capacities. Finally, tests have been initiated to determine whether surfactants that contain active phenolic groups will remove cesium in the foam column.

Magnesium ammonium phosphate and a refined asbestos, asbestos CMB (a product of Union Carbide Corporation), were compared with Grundite clay (an impure illite clay) in beaker tests of removal of Cs^{137} from caustic-carbonate water of the composition described above. Two samples of Grundite clay were used; the first had particle sizes larger than 50 mesh and the other smaller than 325 mesh. Each solid was tested at 0.5 and 5 lb per 1000 gal of water in 30-min contact periods. Neither the magnesium ammonium phosphate nor the asbestos compared favorably with the Grundite clay. At both concentrations, the phosphate removed only 6% of the cesium; the asbestos removed only 5 and 11% respectively. For comparison, Grundite of larger than 50 mesh removed 54 and 76% of the cesium, whereas clay smaller than 325 mesh removed 72 and 92% at the two solids concentrations.

The first surfactant containing an active phenolic group that was tested for cesium removal in the foam column was RWA-100, which is a monobutylphenylphenol sodium monosulfonate. Only 20 to 30% of the available cesium was removed in the foam column at a surfactant concentration of about 200 ppm. This poor performance may have been caused by the bad mechanical properties of the foam, which consisted of unusually small bubbles (only 0.1 to 0.2 mm in diameter) in very turbulent motion and which broke very extensively in the expanded head of the foam-drainage section. Although this particular surfactant does not show much promise for cesium removal, other surfactants with active phenolic groups will be tested, since these are known to react selectively with cesium at high pH (about 12) in ion exchange and solvent extraction systems containing sodium salts.

A feed distributor, designed to give 37 feed streams on an approximately 3-3/4-in. triangular spacing in a 24-in.-diam column, gave uniform flows and low discharge velocities and reduced the extent of channeling below that observed with previous distributors in this large-diameter foam column. The low feed and discharge rates necessary to reduce channeling were obtained by using perforated, parallel, horizontal 3/8-in.-OD tubes across the column as liquid distributors. The orifices (perforations), of 0.037-in. diameter, were baffled to aid in the reduction of foam breakage.

The effects of dodecylbenzenesulfonate and Na^+ concentrations on foam stability in the 24-in.-diam column with a 48-in. foam height were studied at various gas and liquid flow rates. At a concentration of 25 ppm of dodecylbenzenesulfonate in the liquid, a stable foam is obtained, whereas at 10 ppm, gross bubble growth over three-fourths of the initially generated surface was broken.

High-Activity-Waste Calcination. — Fixation of fission products in very insoluble ceramic materials is being studied as a means of preparing high-level waste residues which can be shipped with greater safety to permanent disposal sites. The successful development of a fixation process will largely depend on the ability to control corrosion rates, to remove mercury prior to the high-temperature processing step, and to achieve processing rates at least as great as those obtained in pot calcination (20 to 25 liters of feed per hour).

In the laboratory, continued investigation of ceramic solid compositions for the fixation of sulfate-containing Purex wastes resulted in the development of materials that melted below 800°C, retained more than 98% of the sulfate, and were noncorrosive to the fixation pot. Gas sparging, even with inert gas, increased sulfate loss to about 20% and caused catastrophic corrosion of the fixation container. Glasses softening at or below 600°C were developed for the fixation of nonsulfate Purex wastes.

Corrosion tests of candidate materials for the waste evaporator indicated rates less than 0.2 mil/month for type 304L stainless steel galvanically coupled to titanium 45A in a Purex waste solution to which H_3PO_4, $LiNO_3$, and $Al(NO_3)_3$ had been added to produce the desired solid product on fixation. Uncoupled type 304L stainless steel and Hastelloy F showed maximum rates of 0.1 mil/month, whereas titanium 45A showed slight weight gains.

About 99.8% of the mercury was removed from 100 column volumes of Purex waste solutions by passing it through a column packed with iron tacks. On the other hand, a column packed with stainless steel turnings was ineffective even when a cathodic current was imposed on the bed.

In the unit operations pilot plant, a continuous glass-melting pot has been operated for 30 hr at feed rates up to 20 liters/hr with TBP-25 glass mixture as feed. The glass-melting pot consists of an 8-in.-diam type 304L stainless steel pipe 40 in. long, operated horizontally. The liquid feed enters through four separate water-cooled lines spaced along the length of the pot. The melted glass accumulates to a depth of 3-1/2 in. and is withdrawn through an overflow pipe at one end. A skimmer baffle and freeze-trap valve are incorporated with the glass withdrawal piping. The off-gas is removed from the opposite end and is condensed. Continuous feed rates of 22 liters/hr were demonstrated.

A batch glass test was made with TBP-25 glass mixture as feed, employing the rising-level technique in a vertical pot, 8 in. in diameter and 90 in. high. An average feed rate of 7.3 liters/hr was obtained, and considerable trouble resulted from plugging of the off-gas line. Corrosion of the calciner pot and thermocouple wells was very severe. Complete penetration of the 70-mil-thick thermocouple wells and 150-mil penetration of the calciner pot occurred during the total operating time of 70 hr.

A continuous, horizontal glass-melting vessel incorporating a low-speed agitator has been designed and will be constructed of 8-in. type 304L stainless steel pipe, 36 in. long. Mild agitation of the glass-calcine mixture, with mechanical cleaning of the heat transfer surfaces, should give improved glass-melting capacities.

Low-Activity Waste. — The low-activity-level pilot plant was revised, and operation of a fixed-bed cation exchange process has been started.

The process includes anthracite-bed filtration of ORNL low-level waste to remove suspended solids, followed by ion exchange. A weak-acid resin (Amberlite IRC 50) is used to remove calcium and magnesium, and a strong-acid resin (Dowex 50) for sorption of cesium and strontium. Further study of the scavenging—ion exchange process will test a new 4-ft-diam high-throughput agitated clarifier and an alumina adsorption column for the removal of phosphates, which interfere with the scavenging portion of the process. Foam separation equipment for the pilot plant is being designed. It will include a countercurrent foam column for removal of radioactivity, three recovery columns for foam stripping and recycle of the foaming agent, and equipment for foam breaking, which will include orifices and a centrifuge.

Disposal in Natural Salt Formations. — Design of the demonstration experiment of the disposal of high-level solid radioactive wastes using canned fuel assemblies in the Lyons, Kansas, salt mine continues. A contract for the construction of a new shaft collar, headframe, and cages has been let. Prototype fuel-element canister parts are currently being fabricated, and the design of the canister and hole liner is essentially complete. Hole-liner design has now been modified so that part of the liner and the fuel canister can be removed from the mine floor into the underground transporter. A critical-path schedule of the overall experiment has been developed as an aid to procurement and fabrication sequences. About 30 additional creep-measuring gages have been installed in the area around the experiment site. Drilling of holes for internal strain measurement is under way.

PHYSICAL RESEARCH

PHYSICS AND MATHEMATICS

High-Voltage Experimental Program: Cross Sections for (α,n) Reactions for Medium-Weight Nuclei. — Thick-target (α,n) neutron yields were measured from threshold to about 11 Mev alpha-particle energy for 20 targets ranging in mass number from 27 to 120. The targets of nickel, copper, and zinc were isotopically enriched. The absolute neutron yields, which were measured at approximately 100-kev energy intervals, are accurate to within ±4%. The neutron detector was the 4π flat-energy-response graphite sphere detector developed at ORNL. Cross sections for the (α,n) reactions were obtained with an accuracy within ±15% by differentiation of the smooth thick-target yield curves. For those nuclei in which the (α,n) cross section is thought to account for a significant percentage of the total reaction cross section, the observed cross sections are considerably larger than those predicted by Shapiro for his larger radial parameter of $1.5A^{1/3}$ fermis. Both the shapes and the absolute values of the observed cross sections agree fairly well with the optical-model analysis of reaction cross sections for alpha particles by Igo.

High-Voltage Experimental Program: Resonance Self-Shielding in Neutron Capture Cross Section Measurements. — The resonance self-protection correction for neutron capture was developed for a thin circular disk. Formulas suitable for calculation with an electronic computer or desk calculator were derived. The results will be published in Nuclear Instruments and Methods.

High-Voltage Experimental Program: Gamma Rays from Neutron Resonance Capture in 2s-1d Shell Nuclei. — The capturing states formed by s-wave neutron capture in 2s-1d shell nuclei have even parity, and the ground states, as well as low-lying levels, have the same parity. In odd-Z nuclei (F, Na, Al, P, Cl) the spectra following thermal neutron capture are dominated by transitions to the ground state or to low-lying levels and are presumably of M1 type. On the other hand, for the even-Z nuclei in the same mass range (Mg, Si, S) the spectra are dominated by E1 transitions to levels at excitation energies of about 3 Mev or higher. In order to further investigate this behavior, an experiment on gamma-ray spectra emitted in resonance neutron capture by these nuclei was performed using a pulsed Van de Graaff and a 9 × 12 in. NaI spectrometer. For those resonances where spin and parity assignments are possible, the following phenomena were observed: (1) Odd-parity resonances in odd-Z nuclei decay in weak E1 transitions to low-lying levels. The same levels are fed by strong M1 transitions when the capturing state has even parity. (2) Odd-parity resonances in even-Z nuclei decay strongly via E1 transitions to low-lying levels. The same levels are reached by relatively weak M1 transitions from even-parity capturing states.

Stable Isotopes Development. — A separation of cadmium being used to test the recently completed 255° inhomogeneous-field separators has been completed. The two instruments are now being tested with mercury and lead.

The results achieved with cadmium show each 255° system to be superior to alpha-type calutrons, and separations of the mercury isotopes surpass those attained in the alpha calutron prior to the application of refrigeration to the liner system. The 255° separator having the corrected shims exhibits better focal patterns and produces samples of higher isotopic purity than the separator having the original set of magnetic shims. In general, a 30 to 60% reduction in contaminants is noted in samples separated in the corrected system. This improvement in the samples must be attributed to a successful reduction in the degree of radial aberration associated with the corrected magnetic lens.

An improved feed system for mercury has been designed and incorporated in one of the two 255° separator sources; liquid mercury is placed in a container near the arc chamber, and the feed rate is controlled manually by use of a valve located between the charge bottle and the arc region. The design places limitations on the quantity of vapor that can be conducted to the valve, and the use of a large-diameter, short-length tube connecting the valve and the arc chamber provides adequate conductance to prevent condensation of vapor at this point. The system is stable, and all possibility of flooding the vacuum chamber with mercury is avoided.

The computer program being used to evaluate magnetic fields suitable for use with sector-type separators establishes the feasibility of utilizing a 180° system having 0.5 inhomogeneity. Proper machine parameters can be chosen to compensate for fringing fields. A 24-in.-radius system producing a focal width of only 0.02 in. appears possible without resorting to complicated boundary shaping.

The half-scale model ion source has received some preliminary testing with argon and is currently charged with samarium. Ion outputs are greater than anticipated, but operating parameters have to be adjusted with precision to minimize oscillatory arc conditions. The miniature source which incorporates unitized construction with slip-fit joining of parts is assembled, but actual testing has not been attempted.

Spectrographic studies of the calutron ion beam continue to reveal singly charged ions having energies approaching twice the applied potential. The data are consistent, and credence must be given to the observation. These ions probably are formed by a charge-exchange process acting on ions which were originally doubly charged. Certain high-energy ions of this type contribute to isotopic contamination, and their existence must be minimized in the interest of process improvement.

Special Separations. — The plutonium separation series was completed on September 19. Calutron processing of plutonium has now been suspended in order to provide for a complete cleanup of equipment and for maximum recovery of feed material.

The estimated weights (based on monitored ion currents) of separated plutonium isotopes and the ranges of enrichment achieved are as follows:

Isotope	Estimated Weight for Period (g)	Total Estimated Weight for Series (g)	Purity Range (%)
Pu^{239}	9.5397	71.7178	94.88–94.96
Pu^{240}	17.0353	126.1935	97.63–98.96
Pu^{241}	4.3014	32.2836	89.20–95.88
Pu^{242}	2.1447	16.0052	84.22–93.76
Total	33.0211	246.2001	

Approximately 10 kg of depleted U^{238} has been purified to a low-fired oxide to be used as feed material for the first U^{238} separation series. Equipment to recycle this feed has been installed and is being tested.

Medium-Energy Nuclear Physics: The 86-Inch Cyclotron. – Isotopically enriched targets of Os^{189} were irradiated, and the iridium activity produced by the (p,2n) reaction was analyzed with multirange magnetic spectrographs. Over 100 separate transitions attributed to 41-hr Ir^{188} were resolved up to an energy of 2.5 Mev. Experimental data indicate that the gamma-vibrational ($K = 2^+$) structure may be extended to an $I = 4^+$ rotational state at 965 kev. A second $K = 2^+$ configuration is established at an energy of 1458 kev, with a rotational sequence at 1620 kev (3^+) and 1808 kev (4^+). Electron-capture decay of Ir^{188} directly excites additional 2^+ states in Os^{188} at 1729, 1843, 2099, and 2215 kev. The decay of Ir^{188} may weakly populate a number of odd-parity levels of spin 2 at 1305, 1461, and 2350 kev. Considerable experimental evidence now exists concerning the properties of the higher energy levels, such as rotational configurations and systematic decay branching ratios.

Medium-Energy Nuclear Physics: Heavy Particles. – The analysis of the excitation function for the reaction $C^{12}(O^{16},\alpha)Mg^{24}$ reveals that a significant part of the cross section is due to a direct-interaction process. This is revealed both by a statistical analysis of the deviation of the cross section from its mean and by the asymmetry of the angular distribution averaged over a 3-Mev interval.

Experiments have been initiated on the tandem accelerator to measure the angular distribution of N^{13} from $N^{14} + N^{14}$, below the Coulomb barrier. Excitation functions for nucleon transfer reactions are being measured from the nitrogen bombardment of manganese and scandium.

Cyclotron Operations: The Isochronous Cyclotron. – Stable internal beams of 40-Mev deuterons, 70-Mev protons, and 90-Mev alpha particles have now been achieved. Work directed toward completion of the beam extraction system is proceeding on a three-shift basis.

The iron-and-coil magnet channel, the last element in the extraction system, was installed and tested. Molecular hydrogen ions at 35 Mev were passed through the entire system. The beam could not be directed through the extraction port, however, because unexpected end effects of the extraction channel prevented adequate reduction of the magnetic field. A new channel of improved design was completed and is being readied for installation.

The dee voltage regulator was placed in operation and is providing regulation of about 1 part in 1000. The regulation of the dee voltage resulted in a marked improvement in beam stability and is greatly facilitating the development studies of ion dynamics.

The power amplifier and resonator system was operated stably over the 3-to-1 frequency range from 7.3 to 22.18 Mc/sec. The normal operating dee potential of 70 kv was maintained over the full frequency range.

Protons were accelerated to full hill radius of 32.5 in. at 22.18 Mc/sec. Energy measurements were not made, but the proton energy was calculated as 70.5 Mev.

A sine-cosine potentiometer system was installed to provide for separately controlling the azimuth and the magnitude of a first harmonic introduced by the harmonic coils. These controls greatly reduced the time required to optimize the beam for new operating conditions.

CHEMISTRY

Instrumental Methods for Continuous Chemical Analysis. — A new high-sensitivity direct-reading linear recording conductometric titrator, based on operational amplifier systems and having automatic temperature compensation or proportional temperature control, has been designed, constructed, and evaluated. The design criteria that were established for this instrument are the following: An electronically controlled 10-mv peak-to-peak, 1000-cps sinusoidal voltage is applied to the electrodes of a dip-type conductivity cell so that concentration polarization is minimized and also so that the resulting current which is amplified, rectified, and recorded is directly and linearly related to conductivity. The cells are designed so that the capacitive admittance is as high as possible in comparison with the conductance. The maximum error of the conductivity value from 1 mho to 1 micromho is ±1%. The linearity of recorded voltage vs conductance is within ±0.1%. The automatic temperature compensation is effective between 15 and 60°C except for work of the highest sensitivity, in which case proportional control is used to keep the cell temperature constant to within ±0.01°C. At high sensitivity, the initial conductance current is also compensated.

The proportional temperature controller that was designed provides close and essentially stepless control of titration cell temperature, so that low-noise titration curves are obtained at very high sensitivity. The short-term (15 min) regulation of the temperature of a 300-ml solution is within ±0.002°C. By use of the initial conductance current compensation circuit with a cell having an initial resistance of 1 ohm, a change of as little as 10 milliohms can cause a full-scale change on the recorder; this recorded information is also related linearly to conductance to within ±0.1%. The compensation circuit is used with cells having an initial conductance greater than 2 millimhos. The titrant is delivered at an appropriate rate by means of a motor-driven precision syringe; the change in the total volume of the solution is negligible.

Titrations have been performed with about 40 different systems, including acid-base, complexometric, precipitation, redox, and metal ion titrations. Some titrations, for example, acid-base and free-acid determinations, were performed by means of the initial conductance compensator

in the presence of very large excesses of foreign electrolyte. The instrument has also been used with this compensator to measure the specific conductances (approximately 1 mho/cm) of NH_4Cl samples at 5° intervals from 25 to 100°C. The performance of the conductometric titrator meets or exceeds the design criteria. In addition to the preceding tests, further investigations of certain conductivity titrations are now being made.

Analysis of Molten Salts by Electrochemical Methods. — Anodic stripping voltammetry was previously examined as a method for determining iron at a concentration of 265 ppm in a molten salt mixture of LiF, NaF, and KF at 500°C (ORNL-3508, p 23). The method has been extended to the determination of nickel (10 to 20 ppm) in similar mixtures of molten salts. In the first experiment, the molten mixture, which contained ~10 ppm nickel and 250 ppm iron, was subjected to controlled-potential electrolysis at −0.8 v vs a platinum quasi-reference electrode to remove these constituents. The current-voltage curves which were recorded following cleanup of the melt gave no evidence of the presence of nickel ($E_{1/2} \approx -0.25$ v) and only slight evidence of a wave for iron ($E_{1/2} \approx -0.6$ v). Pyrolytic graphite (1/8 in. in diameter) sheathed in boron nitride was used as the indicator electrode. To ascertain whether an anodic stripping curve for nickel could be obtained, the nickel was plated onto the pyrolytic graphite electrode for 4 min and then subsequently removed anodically. On measuring the areas under the stripping curves obtained before and after the addition of known amounts of nickel, it was estimated that the residual nickel in the melt following the cleanup step was less than 1 ppm. In further experimental work, a calibration curve was established for nickel up to 20 ppm, in which there is slight deviation from linearity; nevertheless, the method demonstrates analytical utility as long as a calibration curve is used. Beyond 20 ppm there is pronounced curvature in the graph. At this level, however, it should be possible to relate the limiting current (wave height) of the current-voltage curve to the concentration of nickel in the melt.

Protactinium Chemistry. — Arrangements were made to use a hot cell in Building 3019 for the purification of the 45 to 50 g of Pa^{231} that is to be returned to England as soon as possible. This protactinum was employed to prepare U^{232} for use in the measurements of nuclear properties; 1 to 2 g is present as impure oxide, a small amount is in various solutions, and about 46 g is in 10 liters of 8 M HCl–0.6 M HF. Noteworthy impurities are U^{232}, Th^{228}, a few grams of aluminum, and probably some natural niobium.

Certain changes in the proposed procedure for purifying this material (ORNL-3508) are being studied. The U^{232} will be removed by passing the solution through an anion exchange column as it is pumped into the hot cell. This simple step quantitatively removes the uranium. Also, initial tests indicate that the Pa^{231} loading on the anion exchange column is comparable when either boric acid or aluminum chloride is used as the fluoride ion complexer, so boric acid will probably be used. (The boric acid is more easily separated at a later stage in the process.) For example, when boric acid was used, the accumulated effluent contained about 2% of the inflowing Pa^{231}, whereas the eluant contained 3% of the thorium, 20% of the niobium, and the balance of the Pa^{231}. The prevention of the

2% "loss" of Pa^{231} is being studied, and it is thought that it may be due to hydrolysis, polymerization, or both.

In one test with Dowex 50 cation exchange resin and appropriately adjusted feed solution, the prior removal of the thorium was complete, while the niobium and Pa^{231} passed through the column.

Studies of the fundamental chemistry of protactinium in sulfate media are continuing; solvent extraction is being used to study the apparent polymerization. The effect of the concentration of protactinium on the distribution between 4.2 M H_2SO_4 and 0.03 M trilaurylamine in diethylbenzene is somewhat similar to that reported previously for 2.5 M H_2SO_4. At low protactinium concentrations the extraction coefficient is nearly constant at 4.2 to 4.5, as compared with about 1 for the 2.5 M acid. At a concentration of about 2×10^5 counts min^{-1} ml^{-1}, there is a sharp increase to a coefficient of about 50. At higher protactinium concentrations, the coefficient decreases slowly (with about the 0.33 power) with increasing protactinium concentrations in the aqueous phase. This decrease at high concentrations had not been observed previously, when lower acid concentrations were used.

Qualitative observations during some protactinium-recovery studies indicated that hydrofluoric acid has less effect on the distribution of protactinium between organic solvents and aqueous sulfuric acid than it does with hydrochloric acid solutions. For example, fluoride at a concentration of about 0.003 M causes a sharp decrease in the extractability from hydrochloric acid solutions [J. Inorg. and Nucl. Chem. 10, 289–305 (1959); Proc. U.N. Intern. Conf. Peaceful Uses At. Energy, 2nd, Geneva, 1958, 17, p 216]. But, with sulfuric acid, it appears that a fluoride concentration of about 0.01 M is needed before the extractability decreases significantly.

Pyrolysis and Radiolysis of Organic Liquids. – Measurements of the kinetics of the initial stages of the liquid-phase pyrolysis of pure biphenyl in evacuated borosilicate glass ampoules at 422, 438, and 446°C have been essentially completed. At each temperature the rates of formation of hydrogen, benzene, and polyphenyl (sum of terphenyl and quaterphenyl) were constant during biphenyl decomposition up to 1.1%. The initial rates of formation of these products per mole of biphenyl at 422, 438, and 446°C, respectively, were: hydrogen – 0.59, 1.9, and 2.8×10^{-5} mole/hr; benzene – 1.1, 3.1, and 4.8×10^{-4} mole/hr; polyphenyl – 1.1, 2.8, and 4.0×10^{-4} mole/hr. These data result in calculated activation energies of 55 to 65 kcal/mole for hydrogen, benzene, and polyphenyl formation. Satisfactory material balances for these pyrolyses indicated that the data were reliable.

The presence of 0.07 mole % oxygen in biphenyl increased the rates of product formation at 420°C by a factor of 3. Large amounts of CO were found in the gaseous products from such experiments.

Properties of Aqueous Solutions: EMF and Solubility Studies. – In any desalination program involving distillation, it is desirable to know the vapor pressure of the solution as a function of temperature and the concentration of the sea salt mixture. Vapor pressures were calculated and compared with experimental values from 25 to 175°C and from zero to 28 wt % total solids (over 6 m in electrolytes) by using the extended

Debye-Hückel equation and osmotic coefficient data for $NaCl-H_2O$ solutions over the range 0 to 4 m from 25 to 100°C and from 1 to 3 m from 100 to 270°C. Even at the extrapolated concentrations the calculated vapor pressures agreed with the observed values within the apparent accuracy of the latter (i.e., within about 2% in most cases).

Properties of Aqueous Solutions: Species and Thermodynamic Properties. — Activity coefficients in multicomponent electrolyte solutions are of great interest, since assumptions involving them underlie such diverse endeavors as studies of complexing reactions, estimation of molecular weights by ultracentrifugation and light scattering, and interpretation of solvent extraction and ion exchange in concentrated solutions. Relative to the importance of the subject, little work has been carried out in the past; much of what has been done has involved systems for which reversible electrode systems are available. In perchlorate media, for example, which are widely used in complexing studies, and for which there are no electrodes suitable for activity coefficient measurements, virtually no work appears to have been done.

The possible application of equilibrium ultracentrifugation to this problem is complicated by the fact that in ultracentrifugation the concentration distribution is measured by the refractive index of the solution, which is nonspecific with respect to solute. In principle, analysis of results is nevertheless possible, since by the Bjerrum cross-differentiation restriction, the activity coefficients of the two solutes are not independent; this relation requires that, if an empirical equation is written for log $\gamma_{\pm}$ of one solute involving parameters for the limiting two-component solution and cross terms involving the concentration of the second solute, the same coefficients of cross terms will be found in the corresponding equation for the second solute.

In analyzing results, one selects the values of these coefficients which give agreement between computed and experimental refractive index patterns. For a test case, the system $HCl-BaCl_2-H_2O$, which has been investigated by the electromotive force method by Harned and Gary, was selected; HCl in these solutions obeys Harned's rule, and the results can therefore be summarized by the coefficient α_{12} which represents the linear variation of log $\gamma_{\pm HCl}$ with the ionic strength of $BaCl_2$ at constant total ionic strength:

Ionic Strength	α_{12}	
	Ultracentrifugation	EMF
2	0.0653	0.0653
3	0.0678	0.0672
4	0.0730	Not measured

The agreement, though partly fortuitous in view of known uncertainties of the ultracentrifugation method, is gratifying.

The improvement of the values over those obtained in earlier stages of this study results from several refinements in the analysis. Two of

particular importance were correction for the effects of pressure on refractive index by the Gladstone-Dale or the Lorentz-Lorenz equation, and inclusion in the least-squares fit of another adjustable parameter which allowed for variation of α_{12} with ionic strength.

Radiation Chemistry. — Measurements were made of the radiolytic decomposition of BrO_3^- ion and of the formation of oxidizing fragments in Li^6-containing preparations of crystalline Li^7BrO_3 which had been irradiated in the ORNL Graphite Reactor. The radiation doses imparted by Li^6 fission recoil particles were estimated from the atom percent of Li^6 in the crystals, the thermal neutron flux, the effective neutron cross section for the reaction $Li^6(n,\alpha)H^3$, the energy release per fission, and the irradiation time, assuming that the recoils were stopped completely within the solid.

The initial 100-ev radiolytic yield for decomposition of bromate, $G_0(-BrO_3^-) = 1.4$, was nearly five times the yield for Co^{60} gamma rays, indicating a strong dependence of the extent of radiolysis on the linear energy transfer (LET). No dependence on dose rate was observed, however, even when the latter varied by tenfold. The large yield of intermediate bromate decomposition products observed (i.e., oxidizing fragments) and the ease with which these products were decomposed by mild thermal annealing appeared to be inconsistent with a "thermal spike" hypothesis for the mechanism of radiolysis. The difference in $G_0(-BrO_3^-)$ for gamma rays and Li^6 fission recoil particles may, however, arise because of the difference in the rate of energy transfer in the crystal following ionization and excitation by these types of radiation.

Physical Chemistry of Gaseous Thermodynamic Working Fluids. — Studies of the thermal transpiration phenomenon are currently being conducted in an effort to further explore the possible relationship between this phenomenon and the translational component of the thermal conductivity. In addition to the experimental work in progress, all of the available thermal transpiration data are presently being compiled and analyzed to determine whether they conform to present theory.

Since the rate of interdiffusion of gases is dependent upon the intermolecular forces involved and, to a high degree of approximation, on only the unlike-molecule interactions, the diffusion process affords an invaluable tool for the elucidation of these interactions. A prototype apparatus has been constructed for studies of binary mutual gaseous diffusion. The gas pair helium-argon is currently being investigated, primarily to compare the results obtained by the present method with those previously obtained by other workers.

Target Preparation. — An electron-bombardment heater system has been developed which can reach temperatures in excess of 3000°C — sufficient to vaporize tungsten metal with special tubular tungsten or tantalum crucibles. Collimation of vapors generated by this technique has been achieved; a hundredfold improvement in vapor condensation has resulted. Self-supported thin films having a thickness greater than 300 $\mu g/cm^2$ have been prepared by using only 20 mg of silicon.

Self-supporting films of germanium, antimony, and beryllium have been prepared in the thickness range 100 to 1000 $\mu g/cm^2$. Thin self-supporting films of more than 30 elements can now be prepared for use in physical

research requiring the complete absence of reactions between bombarding particles and a supporting material.

TRANSURANIUM-ELEMENT PRODUCTION

Chemical Process Development: Acid Control in Tramex Feed. — All aspects of the control of acid in Tramex feed by the addition of methanol are encouraging. Not only has the methanol been effective in counteracting acid loss by radiolysis (the solution was stable at 0.03 to 0.06 M HCl for 25 days at 18 w/liter), but several other advantages were also noted. Extraction of acid, which causes lower actinide extraction, is depressed by the addition of methanol, and as a result more acid can be allowed in the Tramex feed. Phase separation is much more rapid with methanol present; however, there is the possibility that the extraction is somewhat slower. Preliminary tests indicate that there is no explosion or fire hazard with as much as 10% alcohol in Tramex feed (10 M LiCl); no fire or explosion occurred even with an electrical spark in the vapor phase during a slow warming and eventual evaporation to dryness.

Chemical Process Development: Tramex Safety. — Small-scale tests are in progress to determine the possibility of fires and explosions from Alamine 336·HNO_3 in diethylbenzene. In all tests to date, extreme conditions were required in order to cause a fire or a mild explosion. For example, a mild explosion occurred when 4% Alamine 336 in diethylbenzene was in contact with concentrated nitric acid, but only by using an electric spark in the vapor phase after the solution was evaporated almost to dryness (pot temperature, 147°C). The addition of 11 M LiCl (16% of total volume) to the above mixture greatly reduced the fire hazard. With air bubbling through the solution and an electric spark in the vapor phase, a flash occurred only after the pot temperature was increased to 265°C. These conditions are much more extreme than any planned for the Curium Processing Facility.

Carboxylic Acid—Versene—Organophosphorus Extraction System for Separation of Transplutonium Elements from Lanthanides. — In the new method being developed for trivalent actinide—lanthanide separation by phosphorus reagent extraction from aqueous complexes (TALSPEAK process), it has been found that the recovery of americium and curium from carboxylic acid—aminopolyacetic acid solutions used in actinide-lanthanide separations is too slow. This difficulty can be overcome, however, by increasing the carboxylic acid concentration. For example, after the lanthanides have been extracted with a hydrocarbon solution of di(2-ethylhexyl)phosphoric acid from 1 M lactic acid containing diethylenetriaminepentaacetic acid, americium and curium can be extracted readily with a more concentrated solution of the same extractant if the lactic acid concentration is increased to 3 M.

Americium-Curium Separation at High Activity Levels. — Oxidation of Am^{3+} to Am^{5+} in 3 M K_2CO_3 with 0.6 M NaOCl was successful at a Cm^{242} alpha activity level of 80 w/liter. The insoluble $KAmO_2CO_3$ formed readily; 97%

of the americium was precipitated, and only about 3% of the curium was carried. The surprisingly good separation at such a high activity level suggests this as a suitable method for removing americium from Cm^{242}.

Process Equipment Development: Cell Mockup. – Extensive adjustments and alterations of the equipment-transfer case were necessary. A leak-tight seal to the cubicle liner (against a water pressure difference of 1.5 in.) and successful transfer of 400-lb equipment racks were demonstrated after the revisions were made. Additional tests and improvements will be made on the transfer door and its locking mechanism.

Process Equipment Development: Components. – The seating of glass balls in check-valve seats was greatly improved by pressing a hardened steel ball a distance of 0.015 in. into the valve seat. Three sets of check valves were operated dry with a diaphragm pump, and gave a vacuum of 19-1/2 to 20 in. Hg. The feasibility of the service-line disconnect concept was demonstrated by use of an alignment jig. Disconnects aligned in the jig registered perfectly with the corresponding equipment rack.

Process Equipment Development: Pulsed Column. – Testing of the phosphonate process for the separation of transplutonium elements into an americium-curium fraction and a transcurium fraction is under way in the 3/4-in.-ID glass pulsed-column complex. Europium tracer, a stand-in for californium, was extracted from 1.3 M HCl by 1 M 2-ethylhexylphenylphosphonic acid in diethylbenzene and stripped into 3 M HCl. All tests were made at a pulse amplitude of 0.2 in., a column temperature of 25°C, and flows of 10-10-5 ml/min (feed-organic-strip).

The extraction stage heights decreased from 19 to 10 in. as the pulse frequency was increased from 25 to 60 pulses/min, while the stripping stage heights remained nearly constant (17 to 14 in.) over the range 25 to 70 pulses/min.

The effect of radiolytic gas release on the operation of the column was simulated by introducing air into the bottom of the column. The maximum flow of air without flooding was 4 ml/min at 40 pulses/min and 9 ml/min at 60 pulses/min, compared with a calculated gas release rate of 0.5 ml/min from Cm^{242} at a concentration equivalent to 10 w/liter.

HFIR Target Development. – The six prototype target rods for the irradiation experiment were completed and shipped to the Engineering Test Reactor. They are scheduled to be inserted during the October 28 shutdown. Radiographs of the welds and pellets were made. Dimensional measurements and weights were recorded for each component and the final assembly.

Bench testing is in progress for most of the equipment items in cubicle 3 of the Transuranium Facility. The only unsatisfactory component has been the cap-powder dispenser. In cubicle 2, the pellet inspection and loading device is undergoing testing and modification. The design effort is being accelerated to ensure that the present schedule can be met.

Transuranium Facility. – Construction of the Transuranium Processing Plant and the design, procurement, and fabrication of equipment for the facility continue on schedule. Work at the site includes the construction of column footings, grade beams, and retaining walls; installation of underground piping; and the qualification of welders. Test sections

of the conveyor housing were welded by the vendor to ascertain that the fabrication procedures are satisfactory, and bids for the conveyor were issued by the Purchasing Department. Fabrication of the engineering model of a cubicle floor pan has been started at the Nooter Corporation in St. Louis.

Fabrication at ORNL of the first-cycle solvent extraction rack structure was completed, and welders were qualified for Zircaloy-2 fabrication of the pulsed columns and surge tanks that mount on the rack. Purchase orders were placed for all Zircaloy-2 tubing, plate, and bar required, except for material for product storage tanks. These orders totaled $90,900, about 22% below estimates. Design of the process equipment is now 40% complete.

Construction of in-cell equipment in the development facility in cell IV, Building 4507, was completed. Installation of maintenance equipment on top of the cell bank will be completed within a month.

METALLURGY AND MATERIALS

Preparation of Pure Materials. — A two-year fellowship of the International Atomic Energy Agency has now been completed in which the subjects of study were the purification of beryllium by solvent extraction with acetylacetone and the preparation of substantial quantities of high-purity beryllium hydroxide. The work represented an extension of earlier studies (ORNL-3323). All-plastic equipment was assembled in which 20 moles of beryllium could be treated at once, with a total processing time of 36 hr, through the successive steps of solvent extraction, precipitation, filtration, and drying, to give a beryllium hydroxide product suitable for calcination to BeO. Major attention was focused on the chemistry of the solvent extraction steps, in which beryllium ion is chelated by acetylacetone (HX) and is dissolved in carbon tetrachloride as BeX_2, while most metallic impurities are retained in an aqueous phase in the form of strong chelate complexes with ethylenediaminetetraacetic acid (EDTA). Spectrophotometric analytical procedures were developed, based on the absorbancies of the various acetylacetonate species, and Be^7 tracer was used in many of the studies. The variation of the following system parameters with aqueous ionic strength and organic solution composition at 25°C was studied: formation quotient for BeX^+, formation quotient for BeX_2, distribution quotient for BeX_2, distribution coefficient for HX, solubility of BeX_2, and solubility of HX. Decontamination factors, over a pH range from 2 to 7, for some 15 cations, including important contaminants of beryllium, were estimated from the known or measured complex formation of these ions with EDTA. The kinetics of the formation and hydrolysis of BeX_2 were studied, and it appeared that the rate-determining step is $BeX^+ + H^+ \rightarrow Be^{2+} + HX$.

At the conclusion of the program, the total quantity of blended dried beryllium hydroxide, carefully analyzed by a number of methods and available as reference material for research and analytical studies, was equivalent to 2100 g of BeO of 99.998% purity.

Reactions at Metal Surfaces. — The usefulness of metals is often limited by their reaction with the surrounding atmosphere. Despite considerable information on the oxidation of metals, the fundamental mechanisms are inadequately understood. In a continuing investigation of the oxidation characteristics of the refractory metals niobium and tantalum, we are emphasizing the effects of oxygen dissolving in the metal during oxidation.

We have studied the effect of anodically deposited oxide films on the reaction of niobium and tantalum with gaseous oxygen at 500°C. The anodic film decreased the initial rate of oxygen uptake only slightly, particularly in the case of tantalum. For both metals, the thickness of the surface oxide remained virtually unchanged during reaction with oxygen, indicating that the oxygen consumed dissolved directly in the metal. This discovery has enabled a more detailed study of the process by which oxide platelets precipitate in these metals. In both metals the oxide platelets formed were considerably larger and better defined than those obtained during oxidation without a prior anodic oxide film. Platelet formation could be induced in niobium under conditions of temperature and oxygen pressure at which platelets do not usually form.

The anodic films themselves appeared to be particularly inert at temperatures from 400 to 500°C. They were amorphous and could be neither crystallized nor dissolved in the metal by annealing in a vacuum for several hundred hours.

Deformation of Crystalline Solids. — To better understand the deformation of metals and subsequent recovery and recrystallization, we must understand the absorption of deformation energy by the metals. In particular, knowledge of the energy stored during deformation of single crystals is lacking. The methods that are used for polycrystalline materials are too insensitive to measure the smaller energies involved in single crystals. However, Farren and Taylor [Proc. Roy. Soc. A107, 422 (1925)] used a method that depends on the rapid adiabatic deformation of a tensile sample in which the stored energy is the difference between the work used to deform the sample and the thermal energy released in the sample. Using modern instrumentation, we have applied this method with appreciably higher sensitivity to single crystals of copper. A major gain in sensitivity of thermal-energy measurement was achieved by bucking a thermocouple on the sample with a potentiometer, amplifying the differential emf, and recording the signal with a digital voltmeter. While the accuracy is short of that achieved with polycrystalline specimens, which store larger amounts of energy, energies less than 0.01 cal/mole could be measured — one-tenth to one-hundredth the minimum by other methods. The fractional energy storage was about the same as for polycrystalline copper, and as in polycrystalline materials the stored energy was proportional to the square of the flow stress. However, a different dependence on strain was found for the single crystals. We conclude tentatively that dislocations, and only dislocations, are important in both the deformation process and the energy-storage process.

Spectroscopy of Ionic Media. — In nuclear energy and other modern technological fields, increasing use is being made of high-temperature liquids, such as molten salts. However, there is very little funda-

mental knowledge concerning the molecular entities that exist in these media. A continuing program is using spectroscopic techniques to gain better understanding of high-temperature ionic liquids.

We have identified the tetrahedral tetrachloro copper(II) complex in molten salt solutions by means of its spectrum in the near infrared. The tetrahedral complexes of copper(II) are rare; only $CuBr_4^{2-}$ has been observed previously in a liquid medium. The present measurements used molten cesium chloride and tributyl-2,4-dichlorobenzylphosphonium chloride as solvents. These results provide new evidence that anionic complexes having low coordination numbers have an enhanced stability in molten halides with large cations.

By extensive computer calculations we have fitted our spectroscopic data on tetrahalogeno nickel(II) complexes in crystals and solutions to the ligand-field theory, which has been developed in recent years to explain the structure and properties of inorganic complexes. The results demonstrate that the magnetic interaction between spin and orbital motions of the electrons cannot be neglected, as is often done in applying the theory. In the light of these results we are reevaluating certain previous measurements to derive new values of important parameters of the theory.

Theory of Alloying. — Systematic development of new alloys with improved properties requires better understanding of the fundamental physics of the metal crystal and how it is influenced by alloying and temperature. In a continuing program we are studying several fundamental properties, emphasizing the early transition elements Zr, Nb, Mo, Hf, Ta, and W, which have found use in the technologies of nuclear energy, space, high temperatures, and superconductivity.

We have measured the electronic and lattice specific heats of titanium-zirconium and hafnium-zirconium alloys from 1.1 to 4.2°K over the entire range of compositions in these continuous-solid-solution systems. The electronic specific heat and the Debye temperature both vary nearly linearly with the hafnium-to-zirconium ratio. In the titanium-zirconium system a pronounced maximum occurs in the electronic specific heat near 50% Ti. By a current theory of superconductivity [Bardeen et al., Phys. Rev. 108, 1175 (1957)], this is in agreement with a maximum in superconducting transition temperature at this composition. The average interaction parameter in this theory, determined from these measurements, remains constant from titanium to 60% Zr and then rises to the value for zirconium. Although the Debye temperatures rise monotonically from zirconium to titanium, they deviate negatively from a linear dependence on composition. The effects in the titanium-zirconium system apparently correspond to significant relative energy shifts in the multiband electronic structure of these alloys.

A galvanomagnetic study of nine high-purity tungsten crystals shows, in agreement with experiments of Fawcett [Phys. Rev. 128, 154 (1962)], that the Fermi surface of tungsten is closed and compensated, that is, contains equal numbers of holes and electrons. In this property of compensation, tungsten corresponds to other even-atomic-number elements, and its experimentally small electronic specific heat is related to its closed surface. We have observed magnetoresistance (the fractional increase in electric resistance due to a magnetic field) values for tungsten as large as 100,000, which is larger than those previously measured for this element.

Fundamental Research in X-Ray Diffraction. – Investigations of mechanisms of radiation damage in crystalline lattices are important both to the development of practical reactor materials and to the understanding of the fundamental physical processes involved. Beryllium oxide is a particularly suitable material for radiation-damage experiments because of its favorable nuclear properties and because of the variety of damage effects that it exhibits. We are investigating the damage mechanisms in BeO by x-ray diffraction techniques.

Single crystals of beryllium oxide, grown from high-temperature oxide fluxes, have been irradiated at 100, 650, and 1050°C to integrated fast-neutron doses of over 10^{21} nvt. At 100°C the radiation debris appears to be distributed in small agglomerates (about 10 to 100 defects per agglomerate) which do not affect the coherent scattering of x rays from the crystal lattice, as previously reported for a crystal irradiated at 54°C. At 650°C there appears to be macroscopic (>10,000 A in diameter) agglomeration of the debris, and the crystal is severely fractured on basal planes. There is some reason to regard the agglomerates as amorphous BeO and to attribute their growth to the high rate of diffusion at 650°C. Irradiation at 1050°C produces less drastic changes in the optical, physical, and x-ray scattering properties of BeO single crystals. We no longer observe basal-plane fracture or unambiguous regions with macroscopic debris agglomerates. We conclude that defect agglomerates are unstable at this high temperature and that in-pile annealing occurs.

Further experiments are in progress in which the effects of lower integrated dose, varying dose rate, and different methods of crystal growth on the irradiation damage to BeO single crystals will be studied.

Crystal Physics. – Fundamental understanding of the properties of many materials is aided by experiments with single crystals. Suitable single crystals are frequently difficult to obtain, particularly for refractory substances. Therefore, we are conducting a continuing program on crystal growth, both to provide crystals needed in research and to devise and improve methods for growing crystals.

Paramagnetic impurities have been sought and not found by electron spin resonance (ESR) in the previously reported high-quality ThO_2 crystals. This suggests that the concentrations of Mn, V, Cr, and some of the rare earths are less than 10 ppm. We are now growing ThO_2 crystals intentionally doped with specific rare-earth ions for ESR and optical studies.

We have grown BeO crystals at a rate of 0.6 mm/day in our first test of the recently described [J. Appl. Phys. 34, 2885 (1963)] traveling solvent method. This is considerably faster than the rates previously obtained by slow cooling or material transport using the same solvent.

Well-formed crystals greater than 1 mm on an edge of several tri- and tetravalent metal silicates have been grown from $Li_2O \cdot 2MoO_3$ solutions, using mass transfer at temperatures in the range 1050 to 1250°C with thermal gradients of 10 to 20°C/cm. By x-ray diffraction we have identified one product as identical with the mineral thortveitite, monoclinic $Sc_2O_3 \cdot 2SiO_2$. Both forms of $ThO_2 \cdot SiO_2$ were prepared, tetragonal (thorite) and monoclinic (huttonite). Four other compounds formed have been tentatively assigned the following compositions and crystal systems: $Y_2O_3 \cdot 2SiO_2$ (monoclinic), $La_2O_3 \cdot 2SiO_2$ (monoclinic or tetragonal because of possible phase transfor-

mation), $Er_2O_3 \cdot 2SiO_2$ (monoclinic), and $HfO_2 \cdot SiO_2$ (tetragonal). The more exciting aspect is that the erbium silicate, which is light-rose colored, is strongly magnetic at room temperature. Quartz (SiO_2) crystals, about 1 cm on an edge, have been grown hydrothermally from 0.5 N KOH at 15,000 psi and 410°C in a graphite capsule within a stainless steel autoclave. This quartz is being used in ESR studies of the paramagnetic centers formed upon irradiation. Generally, synthetic quartz is grown from NaOH or Na_2CO_3 solutions in steel autoclaves and contains sodium impurity, which complicates the interpretation of the ESR data.

Fundamental Investigation of Radiation Damage in Solids: Effect of Plastic Deformation on the Production of F Centers, M Centers, and V Centers in KCl. — The effect of plastic deformation and heat treatment on the room-temperature production of F centers, M centers, and V centers by Co^{60} gamma rays was investigated. Deformation of an irradiated sample causes a reappearance of the "first stage" coloration for F centers when the sample is irradiated further. This deformation-induced enhancement of the colorability can be annealed out by heat-treating the samples in air at 450°C for 20 min. The coloration curves for the V_2 band are very similar to those for the F band, and they are affected by deformation, heat treatment, and optical bleaching in the same way.

Fundamental Investigation of Radiation Damage in Solids: Investigation of Transmutation Effects in Crystalline Solids. — X-ray diffraction, electron diffraction and microscopy, metallography, and thermal methods were utilized to study transmutation effects in a group of crystalline solids. Nearly homogeneous distributions of transmuted species were introduced into small single crystals of Au^{197}, Ag^{107}, and In^{115} and into powder samples of Ir^{193}, $In_2^{115}O_3$, and $Lu_2^{176}O_3$; the crystals were irradiated in a thermal neutron flux of 4.0×10^{14} neutrons cm^{-2} sec^{-1} at a temperature of ~85°C for periods of a few days to several months, so that considerable fractions of transmuted species were formed.

Small metal single-crystal spheres were converted to alloy single crystals in their solid solution ranges at temperatures as much as several hundred degrees below those required by normal laboratory methods in $Au_{0.82}Hg_{0.18}$, $Ag_{0.78}Cd_{0.22}$, and $In_{0.95}Sn_{0.05}$. A powder sample of Ir^{193} was transformed into the solid solution alloy $Ir_{0.68}Pt_{0.32}$, a high-temperature alloy, which would require a temperature in excess of 2000°C for its formation by laboratory methods.

The production of 22.5 at. % Hg^{198} and Hg^{199} in Au^{197} through nuclear transmutation revealed the nucleation of Au_3Hg particles with the hcp structure, whereas the introduction of 29 at. % Hg caused a recrystallization into two unidentified phases. Long-range order in single crystals of In^{115} was completely destroyed when 14 at. % Sn^{116} was introduced through nuclear transmutation.

Powder specimens of In_2O_3 enriched in In^{115} were irradiated to produce Sn^{116} concentrations as high as 38% of the metal atoms, with the result that a new phase with a smaller fluorite structure was formed which contains oxygen vacancies. The original In_2O_3 structure was largely recovered upon annealing at 1200°C, and SnO_2 particles were precipitated. Powder samples of Lu_2O_3 containing Lu^{176} were irradiated to produce 74 (metal) at. % Hf^{177}, and the resulting material was cubic (fluorite structure) with a lattice parameter approximately half that of the

original structure. This oxygen-deficient lattice was stable at temperatures up to 1200°C.

Fundamental Investigation of Radiation Damage in Solids: Electron Microscope Studies of Fission Fragment Damage in Molybdenum Trioxide. — High-resolution electron microscope images of suitably oriented thin crystals of molybdenum trioxide (MoO_3) show fringe patterns of 6.97 A spacing which relate directly to the (020) planes in the crystal. Crystals irradiated with fission fragments show track patterns which correlate with the passage of the fission fragments through the material. The disruption of the fringe patterns in the tracks indicates disturbance of the crystal structure, and displacement of the fringe patterns in the vicinity of the tracks indicates that internal stresses are present.

Fundamental Investigation of Radiation Damage in Solids: Phonon Scattering from F Centers. — It has been assumed in the past that the interaction between the F center and the lattice vibrations is limited to a possible change in force constant, or to strain field effects coupled with scattering from a quasi-localized mode at the F center. The interaction between the electron and the lattice vibrations has been ignored because of the large energy gap between the ground state and the excited states. The phonon scattering due to this mechanism has been calculated, and it was found that the scattering is an extremely sensitive function of phonon frequency, ω, increasing initially as ω^8. Therefore this mechanism leads to an appreciable scattering of high-frequency phonons. This effect may be seen as a depression in the thermal conductivity of the crystal at temperatures just above the low-temperature maximum.

Fundamental Investigation of Radiation Damage in Solids: Rates of Introduction of Lattice Defects into Semiconducting Materials by Fission Neutrons. — The apparent rate of introduction of lattice defects in semiconducting materials was determined by electrical property measurements subsequent to irradiation at ambient temperature with fission-spectrum neutrons. The biological thermal-neutron facility of the ORNL Graphite Reactor was used in order to minimize any background effects of fast (damaging) neutrons from the normal reactor-neutron spectrum, and all irradiations were conducted between two fuel-element-type fission plates. A reasonable agreement was obtained between the calculated fission-spectrum neutron flux and the experimental results for a wide variety of threshold detectors. The average number of conduction electrons removed per incident fission-spectrum neutron was then determined as 5 to 7 for n-type germanium, 3 to 5 for n-type silicon, and 4 for n-type gallium arsenide.

Fundamental Investigation of Radiation Damage in Solids: Rates of Production of Self-Trapped Holes in Potassium Chloride Doped with Lead or Thallium. — Preliminary investigation of the optical absorption near 350 mμ induced in potassium chloride by 40-kvp x rays at liquid-nitrogen temperature gave the following results. (1) Prior doping with lead or thallium ion enhances the absorption in this range (V_1, V_k region). Moreover, previous studies showed that heating of a lead-doped sample to 625°C in air causes an even greater enhancement. (2) The thermal stability of the absorption was found to be greater in the doped and heated

samples. Warming to 140°K caused a relatively small decrease in absorption. Since it is known that the V_1 band is unstable at this temperature, the implication is that it is the V_k band whose production is enhanced. (3) The initial rate of increase in the height of the V_k band with x irradiation in a heavily doped sample (1 mole % $PbCl_2$ in the melt) which had been heat-treated was more than five times that of the F band. There are indications that the growth rate of the V_k band decreases at the higher x-ray doses. (4) A sharp absorption band at 253 mμ was observed in the heavily lead-doped sample.

Fundamental Investigation of Radiation Damage in Solids: Growth of Copper Crystals of Low Dislocation Density. — A study was made of the perfection and the factors controlling perfection of copper crystals grown by a Bridgman technique. Crystals were grown from 99.999%+ and from oxygen-free high-conductivity (OFHC) copper; they were acid-sawed and polished, and dislocation densities were determined with etch-pit techniques. The orientation of the growth direction of unseeded crystals of both purities was random. There was a correlation between perfection and growth direction for the 99.999%+ copper crystals in that the more perfect crystals had growth directions far from the [111] zone. Dislocation densities as low as 1000 per square centimeter, with no subboundaries, were obtained in the purer as-grown crystals. The OFHC crystals were much less perfect and contained many subboundaries. After the purer crystals were annealed at 1075°C, slices with dislocation densities as low as 100 per square centimeter were obtained. The vacancy condensation mechanism for generation of dislocations apparently was not significant in accounting for the residual dislocation density in these crystals.

Fundamental Investigation of Radiation Damage in Solids: Cesium-137 Gamma Source. — Recently, a high-intensity Cs^{137} irradiation facility has been constructed for use in studying the effects of radiation on solids. The source contains approximately 9000 curies of $Cs^{137}Cl$, and the radiation intensity in the central specimen chamber is 1.5×10^6 r/hr. This permits investigations of the effect of high intensities of lower gamma-ray energy (0.67-Mev photons) than the 1.3-Mev Co^{60} photons, which were the only gamma rays available in high intensity previously. The intensity of the source corresponds to a flux of 1.2×10^{12} photons cm^{-2} sec^{-1}.

CONTROLLED THERMONUCLEAR RESEARCH

Plasma Physics. — In previous experimental work concerning the propagation of ionic sound waves in magnetically supported, collision-dominated plasma columns, the velocities of wave propagation were found to be higher than the theoretical values. Recent work suggests that the theory is in need of correction. Essentially, a strong longitudinal magnetic field inhibits energy transfer from the longitudinal to the transverse direction of the motion of the electron and the ion. Thus, at high magnetic fields the motion of the particles becomes, in effect, one dimensional; the value of the adiabatic compression coefficient of

the electron gas increases from 5/3 to 3; and the velocity of the wave is higher by a factor of 1.3 than the value predicted on the basis of a three-dimensional theoretical equation.

In previous studies of the plasma containment time and the plasma density in a magnetic-mirror apparatus, some inconsistencies were obtained (ORNL-3392, sec 3.5, p 38). These inconsistencies are now apparently understood. In this work, plasma density was measured in two ways: first, by measuring the frequency shift of a cavity resonator surrounding the plasma volume, and second, by collecting and measuring the ion current escaping through the magnetic mirrors. The discrepancy observed was that the plasma density as measured by the cavity resonator was always lower by a factor of between 30 and 400 than that measured by the charge-collection technique. However, the decay time constants measured by the two methods were identical.

The explanation of the discrepancy is quite simple — the plasma in the apparatus is now considered to be a hot-electron rather than a cold-ion plasma. Such hot-electron plasmas having electron temperatures of many electron kilovolts were indeed later observed in the same apparatus. This hot-electron plasma gives a decay time constant of the correct order of magnitude. In addition, each hot electron produces many secondary ions and electrons by ionizing the background gas in the apparatus. These secondary ions and electrons are scattered very rapidly out of the magnetic mirrors.

Thus, the cavity resonator apparently measured the actual density of plasma contained in the apparatus, while the ion-current collection technique apparently measured the current of secondary ions produced. The theoretical studies show that, as was observed, the secondary ions should be about 100 times as abundant as the primary electrons, and that the decay time constants for the two measuring processes should be identical.

DCX-2 Facility. — During this report period, hydrogen and lithium arcs have been operated in the DCX-2 during beam injection. The density produced with a hydrogen arc is about 5×10^8 for molecular ion beams of about 20 ma injected into the machine. Between 30 and 50% of the beam is dissociated. It appears now from a study of the energy spectrum of the charge-exchange neutral particles that the loss of energetic particles from the plasma can be accounted for by charge exchange within a factor of 2. With the hydrogen arc the trapped particles are observed to have a widely spread energy distribution, which results in lifetimes from 1/3 to 1/10 of those expected if the particle energy were unchanged from the injected energy. The angular distribution of the emitted particles has been measured. It shows that the trapped protons have essentially the same angular distribution as the injected molecular ions, implying a negligible energy scattering in the axial direction. Technical and mechanical difficulties with the lithium arc have prevented the accumulation of many data with it.

Vacuum. — The large-scale ultrahigh-vacuum test facility, which uses liquid-nitrogen cooling to effect adsorption of residual gases, has been assembled and operated after the detailed redesign of most elements of the apparatus. The newly constructed auxiliary controlled low leak rate apparatus has been operated and performs according to the

design specifications. Leak rates with good stability are maintainable in the range from 10^{-5} torr liters per second up to 10^{-2} torr liters per second in the present configuration of the apparatus. This device in conjunction with the large test facility (30,000 cm^2 surface area) allows the introduction of gas at a rate of about 10^{10} molecules cm^{-2} sec^{-1}, or less than 0.1 monolayer/hr. Continued attention to the achieving of lower temperatures using subcooled liquid nitrogen has resulted in a temperature of a copper baffle plate (15,000 cm^2 area) of near 70°K, as compared with the previous operating temperature of 80°K.

Two additional bakeable flange gasket assemblies have been tested and found to be inadequate for controlled thermonuclear research purposes.

Mass spectra of vacuum pump foreline containments showed a need for control of forepump oil quality. The use of diffusion pump oils (hydrocarbon type) has greatly decreased the background of volatile compounds in these systems. However, time-dependent contamination of forelines by aromatic compounds (e.g., toluene) has been observed even when diffusion pump oils are used.

BIOLOGY AND MEDICINE

TERRESTRIAL AND FRESH-WATER ECOLOGY

Evaluation of Fission Product Distribution and Movement in White Oak Drainage System. — A series of preliminary core samples has been taken in and around two of the earlier "open" type intermediate-level waste seepage pits that were recently abandoned and filled. Radiochemical analyses of segmented sections of these cores indicate that most of the activity is associated with the sludges and precipitates that have accumulated in the pits and on the first few inches of weathered shale that comprises the side walls and bottoms of the pits. Concentrations of Cs^{137}, Ru^{106}, and Co^{60} were observed to be as high as 85, 78, and 4 μc per gram of dry weight respectively. The sludge was also observed to be quite variable in thickness and distribution within the pits.

RADIOLOGICAL PHYSICS, HEALTH PHYSICS, AND RADIATION INSTRUMENTATION

Radiation Physics and Dosimetry: Magnetodynamic Instabilities in a Plasma-Beam System. — It is well known that Alfvén waves may be excited in a plasma-beam system. It has been found that such excitation is different for an ordinary (Alfvén-O) wave and for an extraordinary (Alfvén-X) wave. An Alfvén-O wave is characterized by a nonisotropic velocity distribution. However, the instability behavior for such a wave was found to be isotropic; that is, the excited frequency and the rate of growth are independent of the direction of propagation of the

wave. On the other hand, an Alfvén-X wave has an isotropic velocity distribution, but its instability behavior was found to be nonistropic. Expressions have been derived for the excited frequency and the rate of growth corresponding to any direction of an Alfvén-O or Alfvén-X wave. An analysis was made of an excited Alfvén wave aligned along the direction of the magnetic field and a relationship given between the instabilities of linearly and circularly polarized waves. A comparison has been made of instabilities produced by an electron and by an ion beam.

Radiation Physics and Dosimetry: Constitutive Equations for a Plasma-Like Medium. — A general formulation has been obtained representing the constitutive equations for a plasma-like medium. These equations are of the type $\mathbf{P} = \alpha_{ee}\mathbf{E} + \alpha_{em}\mathbf{B}$, $\mathbf{M} = \alpha_{me}\mathbf{E} + \alpha_{mm}\mathbf{B}$, where $\mathbf{E}$ is the electric intensity, $\mathbf{B}$ is the magnetic induction, $\mathbf{P}$ and $\mathbf{M}$ represent the electric and magnetic polarizations, respectively, and α_{ee}, α_{em}, α_{me}, and α_{mm} are appropriate macroscopic parameters characterizing the medium. These equations differ from the constitutive equations for a molecular medium, since in the latter case one has $\mathbf{P} = \chi_e\mathbf{E}$ and $\mathbf{M} = \xi_m\mathbf{B}$, where χ_e is the electric susceptibility and ξ_m is the "proper magnetic susceptibility." A formulation has been obtained for the constitutive parameters for two particular cases: (1) when the plasma-like medium is represented by a charge-equilibrated uniform electron beam of infinite width, and (2) when the plasma-like medium is represented by a charge-equilibrated electron gas characterized by an isotropic velocity distribution f(v).

Internal Dose Estimation. — Maximum permissible body burdens and concentrations in air, $(MPC)_a$, and in water, $(MPC)_w$, have been computed for 20 radionuclides of importance in the Transuranium Program. These values are for occupational exposure and are intended to supplement Table I appearing in the NCRP Internal Dose Handbook, NBS Handbook 69, and in the ICRP Internal Dose Handbook, ICRP Publication 6. Thus the same formulas as well as the same basic biological and physical assumptions of these publications were adopted for the computations of the new values.

The specific radionuclides for which the computations were made are: U^{240}, Pu^{243}, Pu^{244}, Am^{242m}, Am^{242}, Am^{244}, Cm^{247}, Cm^{248}, Cm^{249}, Bk^{250}, Cf^{251}, Cf^{253}, Cf^{254}, Es^{253}, Es^{254m}, Es^{254}, Es^{255}, Fm^{254}, Fm^{255}, and Fm^{256}. In each case, values were tabulated for (1) insoluble materials, using both the lungs and the gastrointestinal tract as organs of reference, and for (2) soluble materials, using total body, bone, gastrointestinal tract, and other organs wherever indicated as organs of reference. In cases where the radionuclides decayed to a stable state through a long chain of radioactive daughters, the computations became quite involved, and these determinations were programmed for the IBM 7090 computer.

One problem arose with the new group of radionuclides that was not encountered in estimating MPC's for the other radionuclides given in Table I. This was the matter of radionuclides in which the prominent mode of decay is spontaneous fission. Since the quantity and distribution of energy released in spontaneous fission are not precisely known

(for many of the radionuclides involved), it was decided, after consultation with the NCRP and the ICRP committees on internal dose, to use the same energy distribution for all isotopes of an element; and in cases where data were not available from the literature for an element, to extrapolate these values from neighboring elements.

These new values have been adopted by the ICRP Committee 2 after certain revisions resulting from an interchange of ideas as to basic philosophy, format, and parameters used. They will appear in the forthcoming ICRP Publication 6, Recommendations of the International Commission on Radiological Protection, as Amended 1959 and Revised 1962. NCRP Subcommittee 2 has the values under consideration.

ISOTOPE DEVELOPMENT

Nuclear Analysis Research and Development: Activation Analysis in Forensic Science. — The characteristic differences in the concentrations of trace elements in hair are being studied as a possible means of identification. The results reveal that the method used in washing the hair is an important step, since some methods do not remove all of the surface contamination. In the past, some workers have primarily used organic solvents for this purpose, but detergents and shampoos remove much more material from the hair than the solvents do. Many of the elements which were previously believed to be in hair appear to exist primarily as surface contaminants.

Nuclear Analysis Research and Development: Total Neutron Absorption Cross-Section Measurement. — As a consequence of recent interest in the total neutron absorption cross sections of reactor construction materials, the sensitivity of neutron absorption measurements (ORNL-3335) has been reevaluated. By using large samples (~6 liters) and two 2-curie Am^{241}-Be neutron sources, differences equivalent to 2 μg of boron per cubic centimeter have been detected in the total neutron absorption cross sections of such materials. This method appears practical for measuring neutron-absorbing impurities in aluminum, graphite, beryllium, and water. It is nondestructive and requires only about 10 min.

Nuclear Analysis Research and Development: Use of a 14-Mev Neutron Generator. — Elements with atomic number less than 30 have been irradiated at a flux of 5×10^8 neutrons cm^{-2} sec^{-1}. The reactions observed were cataloged, cross sections were calculated, and the gamma-ray spectra were recorded under standard conditions. These spectra may be utilized in accurately predicting the presence of interferences in and the ultimate sensitivity of direct gamma-ray-spectral analyses of irradiated materials for various times of irradiation and decay.

Recovery of Fission Products. — In testing the chemical and radiation stability of 4-sec-butyl-2-(α-methylbenzyl)phenol (BAMBP), which is an effective extractant for cesium, 1 M solutions of BAMBP in Amsco 125-82 were contacted for 51 hr with tartrate-complexed simulated Purex 1WW waste solution at pH 12 and with 0.05 M HNO_3 in the Co^{60} gamma-ray source for exposures up to 100 whr/liter. Parallel tests were also run

outside the radiation field. The extractant solution subjected to the acid treatment under irradiation and those subjected to the alkaline treatment, with or without irradiation, showed a two- or threefold increase in extraction power for cesium and sodium as compared with that for untreated extractant solution, whereas the extractant solution subjected to the acid treatment without irradiation showed no change. For all samples, phase-separation characteristics and selectivity with respect to extraction of other fission products (Zr-Nb, Ru, Eu) were not significantly changed.

Isopiestic measurements and infrared spectra indicate that BAMBP is dimeric to trimeric at 0.8 to 1.25 M concentration in octane diluent. The degree of polymerization increased within this range with increase in BAMBP concentration and was somewhat higher with water present than when water was excluded from the system.

Radioisotope Research and Development. — The production of Mn^{54} by the $Fe^{54}(n,p)Mn^{54}$ reaction was examined in different positions in both the LITR and the ORR. The most favorable position for maximum yield of Mn^{54} is the ORR-NF stringer, which has a fast neutron flux between 1 and 2×10^{14} neutrons cm^{-2} sec^{-1}. In this position, 10 g of natural iron can be irradiated in a single capsule at a fast flux of at least 1×10^{14} neutrons cm^{-2} sec^{-1} to produce a minimum of 54 mc of Mn^{54} in a 28-day irradiation period. Carrier-free Mn^{54} was recovered by an anion exchange separation in the hydrochloric acid system. No radiochemical contaminants were detectable in the Mn^{54} product solution.

Applications of P^{32} in certain biomedical studies require that the activity be administered as PO_4^{3-}. Small amounts of polymeric forms of phosphorus in the solution are undesirable. The ORNL P^{32} product was examined by solvent extraction and paper chromatographic techniques, which confirm that less than 1% of the P^{32} exists in a polymeric state. Further studies are in progress to determine whether commonly employed methods for the preparation of isotonic solutions will result in the formation of the undesirable polymeric species.

Recently, the first large Cs^{137} teletherapy source prepared for ORINS was returned for examination of the effect of 9.5 years of aging on the source capsule and the $Cs^{137}Cl$ source compound. The capsule showed no evidence of corrosive attack. Chemical processing of the $Cs^{137}Cl$ resulted in the recovery of about 1.5 g of stable barium resulting from the radioactive decay process. An isotopic analysis of the barium showed it to be 99.2% Ba^{137}, 0.2% Ba^{134} (from Cs^{134}), and 0.6% natural barium.

An enriched target of $Zn^{68}O$ (99.3% Zn^{68}) was irradiated with 23-Mev protons in the ORNL 86-in. cyclotron. Chemical separations and gamma-ray spectroscopy identified two radioactive isotopes produced in the target by the reactions $Zn^{68}(p,2p)Cu^{67}$ and $Zn^{68}(p,2n)Ga^{67}$. The production rates were 5 μc/ma-hr for Cu^{67} and 30 mc/ma-hr for Ga^{67}. Gallium-67 has an attractively high production rate and can be readily recovered as a pure, carrier-free radioisotope product.

Radioisotope and Stable Isotope Production. — Ion currents equivalent to 2327 g of separated isotopes were monitored during the past two months. Isotopes of Ca, Si, Cr, S, Rb, Cd, and Pb were separated and collected using 28 of the calutrons associated with the program.

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