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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

FEBRUARY 1964

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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

FEBRUARY 1964

DATE ISSUED

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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

FEBRUARY 1964

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

Instantaneous Fission-Gas Release Experiment. — Based on fission-gas release data from single-crystal UO_2 specimens, a tentative model for high-temperature fission-gas release was formulated. The essential features of the model include the following premises: (1) defects in the UO_2 structure will trap impurity atoms, (2) defects are formed by fission-fragment tracks, and (3) the fission-gas release is controlled by the probability of the gas being trapped rather than by a diffusion process. With this model we can explain qualitatively all the observed characteristics of the in-pile gas release.

The gas release from fine-grain UO_2 specimens of the same size and density as the single-crystal specimens, but having many more internal defects (since the grain boundaries are considered defects), was less than one-fifth the amount from the single-crystal UO_2 for the same irradiation conditions. A decrease in gas release is predicted by the model.

Postirradiation Examination of Fueled Graphite Spheres. — The first three irradiated AVR-size (6-cm-OD) fueled graphite spheres are being examined. These spheres were fueled with pyrolytic-carbon-coated particles of UC_2 or $(U,Th)C_2$ and had either molded or machined unfueled graphite shells.

No changes in the spheres were apparent by visual inspection. Measurements of spheres irradiated at 1300 to 1600°F showed no diameter or weight changes greater than 0.1%. However, spheres irradiated at 1700 to 2000°F had diameter changes of 0.1 to 0.8% and weight losses of 0.1 to 0.2%. Compression tests performed on selected spheres indicated that spheres with the unfueled shell molded directly to the fueled core were appreciably stronger than spheres in which the fueled core was cemented inside a machined unfueled shell. The average compression strengths of two spheres of each type were 2400 and 1400 lb respectively.

GCR-ORR Loop No. 1 Irradiations. — A new fuel element assembly was installed in loop No. 1 as a fission product source in an experiment for determining the behavior of radioactive fission products in a circulating helium stream. For convenience, we chose for the source UO_2 fuel contained

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in a stainless-steel-clad assembly ventilated with a small (0.013-in.-diam) hole. The fuel was manufactured by the Westinghouse Corporation and should be representative of the highly retentive fuel manufactured for the EGCR. This fuel is advantageous for the experiment because its fission-gas release rate is well known as a function of temperature and operating conditions. We are currently operating under EGCR conditions to provide information applicable to analysis of failed elements in that reactor. No measurable amount of fission gas was observed in the loop during the first week of operation.

LITR Capsule Irradiations. — Two capsules, L-CP7 and L-CP8, containing pyrolytic-carbon-coated particles in each of four compartments, continue to operate satisfactorily after 8 and 6 months of exposure, except that indicated fuel temperatures have decreased ~200°F for each capsule. We estimate that the burnup of the fuel in the various compartments varies between 5 and 12 at. % of heavy metal.

ORR Poolside Capsule Irradiations. — Irradiation of capsule 01-7 containing pyrolytic-carbon-coated particle fuel in spherical graphite-matrix elements was terminated after ~4 at. % heavy-metal burnup. At shutdown the element in the sweep section had an R/B (ratio of fission-gas release rate to birth rate for Kr^{88}) of $\sim 3 \times 10^{-4}$. This element has a molded unfueled shell. The capsule was replaced with a new assembly, 01A-7, containing two fuel elements having unfueled shells machined of ATJ graphite and a third element of molded graphite. We will operate the spheres in this capsule at central temperatures of 2500°F.

Eight-Ball Experiment. — The fourth eight-ball experimental assembly containing 1/2-in.-diam spheres fueled with pyrolytic-carbon-coated UC_2 fuel particles is operating at the design sphere surface temperature of 1700°F. We erroneously described this unit as the third assembly in the report last month.

Fueled-Graphite Fabrication. — Baking studies were completed on liquid phenolic-resin bodies compounded to have 7 and 10 wt % residual ungraphitized carbon after bakeout from each of two fine-grained synthetic graphite flours, A38 and A325. The A38 graphite is the coarser grained of the two. On a four-day baking schedule to 1000°C, we obtained higher cured densities from the coarser grained matrix than from the finer grained matrix. Also, the baking produced higher cured densities from the material with 7 wt % ungraphitized carbon than from that with 10 wt % ungraphitized carbon.

Crushing Loads of Fueled-Graphite Spheres. — Crushing loads of fueled-graphite spheres from three vendors were determined on approximately one hundred 1-1/2-in.-diam and 6-cm-diam spheres in the as-received condition and after irradiation and oxidation tests. The crushing loads after irradiation showed little or no change for all but one sphere, which had a crushing load 750 lb lower than the average value for six similar unirradiated spheres (2620 lb vs 3370 lb). The irradiated spheres had fracture modes which were the same as those for the unirradiated spheres.

Oxidation weight losses greater than 1% significantly reduced the crushing loads, for example, from an as-received average crushing load of 1440 lb to 960 lb. However, the oxidized spheres were generally softer, giving much larger "flattened zones." Thus the loads were not as concentrated on the oxidized spheres as on the as-received spheres.

Phase Relationships in the Th-U-C System. – Approximate melting temperatures (in vacuum) were determined for a range of arc-melted ThC_2-UC_2 alloys. The results ranged from 2640 ± 50°C for ThC_2 to 2475 ± 50°C for UC_2. Each alloy contained approximately 1 wt % free carbon, which may have lowered the melting temperature compared with that for stoichiometric alloys. However, the reduction should be less than the estimated experimental error. The temperatures reported are those at which a major portion of the thin sliver used became molten in a small carbon-tube furnace. We could not determine liquidus and solidus temperatures, but the figures do give an approximate melting temperature.

High-temperature x-ray diffraction studies have confirmed that at all compositions examined so far, the high-temperature phase is face-centered cubic. The transformation temperatures ranged from 1415 ± 10°C for ThC_2 to 1820 ± 20% for UC_2. At UC_2 contents of 33% and higher, the temperatures are only approximate, since we measured them by taking pyrometer readings of the surface of the x-ray diffraction specimen at the point where the high-temperature cubic phase disappeared on cooling. Refinement of the measurements on the high-temperature cubic thorium dicarbide (using ThO_2 lines as standards) gave a lattice parameter of 5.808 ± 0.003 A at 1500°C.

Steam-Graphite Reaction Rate Experiment. – A third graphite fuel channel sleeve of the type used in the EGCR was exposed to superheated steam at a constant pressure of 155 psig and a temperature varying from 1200 to 1580°F in order to study hazards problems associated with a steam-graphite reaction in the EGCR. The generation rate and composition of gas were measured as a function of temperature to determine the graphite reaction rate. The results indicated a reaction rate of 3.4 g hr^{-1} ft^{-2} at a steam temperature of 1580°F and 2.2×10^{-3} g hr^{-1} ft^{-2} at 1200°F. These values are substantially smaller than the estimated ratio used in the EGCR hazards report. The activation energy in the range 1300 to 1600°F was 43 kcal/mole, which is similar to that obtained in other laboratory experiments.

REACTOR FUELS AND MATERIALS

Radiation Effects in Structural Materials. – Slowly cooled samples of Cu-Ni alloys ranging in composition from 35 to 75 at. % Ni were irradiated to a dose of 10^{17} neutrons/cm^2 (E > 1 Mev) at a temperature of –180°C. Following the irradiation, the electrical resistivity was measured upon isochronal annealing. Similar measurements were made after deformation at temperatures below –100°C. There was very little resistivity change during the irradiation at –180°C, but a decrease of 1 to 3% was observed upon isochronal annealing between –100 and 300°C. The resistivity

increased to its original value between 320 and 420°C and continued to increase as the annealing was extended to higher temperatures. The low-temperature deformation introduced a 12% increase in resistivity, which was recovered upon isochronal annealing between 0 and 400°C. The decrease in resistivity of the irradiated samples is attributed to radiation-enhanced segregation of the copper and nickel into clusters, whereas the increase in resistivity upon deformation is believed to be associated with the breaking up of clusters in the partially segregated starting material.

Behavior of High-Temperature Materials Under Irradiation. — Gross-damage data on BeO specimens irradiated in experiment 41-8 (maximum dose, 8.2×10^{21} nvt) have been evaluated and compared with data of experiment 41-9 (maximum dose, 4.0×10^{21} nvt). As expected, there was greater gross damage in 41-8 than in 41-9 at both 650 and 1100°C, and the damage was much greater at 650 than at 1100°C. Specimens of low density (~2.7 g/cm^3) and small grain size (~20 μ) were damaged less than specimens of high density or large grain size at both temperatures. No definite flux intensity effect could be observed in comparisons of gross damage to specimens receiving the same dose at different fluxes in the two experiments.

The second or low-dose section of the current high-temperature materials experiment containing MgO, Al_2O_3, and BeO has been installed in the ETR. It is currently on test and will be irradiated to a scheduled dose of 3×10^{21} nvt. The high-dose section has already received an approximate dose of 2×10^{21} and is scheduled to operate until 6×10^{21} nvt has been accumulated.

FUNDAMENTAL HEAT TRANSFER AND FLUID DYNAMICS

Magnetohydrodynamic Studies of Vortex Flow. — The experimental investigation of the effects of an axial magnetic field on the stability of jet-driven confined-vortex flow of an electrolytic conductor was continued. Increasing the number of injection slits from two at 180° to eight at 45° circumferential spacing increased the tangential Reynolds modulus for transition to instability in the concave wall boundary layer (as detected by dye-trace injection) from ~400 to ~900 with no applied magnetic field and from ~1600 to ~4000 with an applied magnetic field of 74 kilogauss (double-coil large-volume test magnet located in the ORNL Magnet Laboratory). In contrast with the behavior of the two-slit model, for which instability in the wall boundary layer appeared to trigger general instability throughout the flow field, the eight-slit model gave evidence of initial instability in the interior of the flow field. Furthermore, the fractional recovery of injection velocity as tangential velocity near the periphery was lower for the case of the eight-slit tube, which may be related to the fact that the jets entered with a small inward radial velocity component, whereas in the two-slit model the jets entered tangentially.

Numerical Calculation of Hydromagnetic Stability. — Calculation of the effect of an axial magnetic field on the flow of a viscous, electrically conducting fluid between rotating concentric cylinders was carried out for two types of velocity profile. The first was nonrotational flow

in which the tangential velocity varied inversely with the distance from the axis. Diameter ratios from 0.1 to 0.8 and Hartmann moduli from 0.1 to 1000 were considered. For this type of flow it was found that small magnetic fields could produce instability. When the Hartmann modulus was greater than about 30, however, any further increase in the magnetic field had a stabilizing influence on the flow.

The other velocity profile considered was that obtained when the inner cylinder was rotated while the outer was held stationary. Again, diameter ratios from 0.1 to 0.8 and Hartmann moduli from 0.1 to 1000 were considered, and the magnetic Prandtl modulus was assumed to be small. In this case, an increase in the magnetic field always tended to stabilize the flow. The stabilizing effect of the magnetic field with the outer wall stationary was found to be greater than for similar calculations reported in the literature in which both walls were taken to be rotating, so that at a Hartmann modulus of 1000 the critical Taylor modulus was about 200 times what might have been expected.

Low-Pressure Two-Phase Swirl-Flow Boiling and Burnout Study. — Assembly of loop components continues. An entrainment separator utilizing both centrifugal and inertial impingement mechanisms of separation was designed, fabricated, and installed. A study was made of the critical flow limits of this loop, and it appears that at pressures of 30 to 50 psia and qualities of 50 to 90 wt %, the critical mass velocity will be ~200 lb sec^{-1} ft^{-2}, which corresponds to an inlet velocity of ~3.3 fps.

Tests were performed on a centrifugal flowmeter made by bending a tube into a 360° circle and measuring the pressure difference between the inside and outside of the curved flow path. Cold-water tests with tube and coil radii of 0.094 and 3.098 in., respectively, showed good reproducibility and yielded a head differential of 50 in. H_2O at a meter velocity of 34 fps. Meter Reynolds moduli were varied from 2180 (2 fps) to 39,000 (39 fps).

The onset of local wall overheating at the attainment of the critical heat flux will be detected by solid-state light-activated switches mounted inside a shroud tube, which will be concentric with the test section. A study was made of the sensitivity of these switches, which are of the subminiature silicon PNPN type and are characterized by an effective triggering irradiance of ~3 mw/cm^2. They are sensitive to visible and near-infrared wavelengths. When mounted with the axis perpendicular to the incident radiation, the switch latches into conduction at a source temperature of 1300°F at a distance of ~0.5 in., and at a source temperature of 1800°F at a distance of ~2.0 in. When the cathode is pointed toward the thermal source, the sensitivity is somewhat less, and the cut-on distance for a given source temperature is typically approximately half that described above. In the burnout detector application, the closing of the switch will activate a relay which will interrupt the heating current to the test section.

POWER REACTOR FUEL PROCESSING

Shear-Leach Process. — In the shear-leach process for multitubular stainless-steel-clad or Zircaloy-2-clad uranium and/or thorium oxide or

alloy fuels, the assemblies are first sheared to expose the fuel, which is then leached from the cladding with nitric acid. The leached cladding is then sent to waste.

In preparation for shearing tests with prototype Consolidated Edison core B fuel, new enlarged gags, a fixed-blade holder, fixed blades, and a feed envelope were made for the ORNL 250-ton prototype shear. Slight modifications to other shear components are being made in the field to ensure satisfactory operation.

For continuing batch leaching tests, a cylindrical batch dissolver (8.4 in. in inner diameter, 8 ft high) employing thermal circulation was built. It is made of Pyrex glass and stainless steel, and it uses a perforated basket 6 in. in diameter and 18 in. long. The dissolver was built for leaching studies with sheared, unirradiated, stainless-steel-clad UO_2 and UO_2-ThO_2 pellets. The off-gas produced during batch leaching of 1-1/2-in. lengths of sheared stainless-steel-clad UO_2 fuel in boiling 7 M HNO_3 gave a value of 0.64 to 0.69 g-mole of NO per g-mole of uranium dissolved in an oxygen-free system. This was in good agreement with the expected value of 2/3 g-mole of NO per g-mole of uranium. Composition of the off-gas was 97% NO; the remainder consisted of approximately equal parts of N_2, NO_2, N_2O, and CO_2.

Dissolution Studies on ThO_2-UO_2 Produced by the Sol-Gel Process. — Methods are being investigated for the dissolution of high-density ThO_2-UO_2 particles produced by the sol-gel process. Typical fuel, containing 4% UO_2, has a density nearly 100% of theoretical and has a particle size distribution as follows: 60%, 6 to 16 mesh; 15%, 70 to 140 mesh; and 25%, less than 200 mesh. The most practical dissolvent found for this material is boiling 13 M HNO_3–0.04 M HF–0.04 M $Al(NO_3)_3$. Batch dissolution of unirradiated fuel samples in this reagent required 75 hr for completion; the final solution was about 1 M in thorium. The amounts dissolved as a function of time were 70, 84, and 92% in 7, 24, and 36 hr respectively. Complete dissolution could be achieved in 35 hr by successive 7-hr digestions with fresh reagent; however, the thorium concentration after combining all the solutions was only about 0.2 M.

Dissolution Studies on Irradiated Uranium Carbide and Plutonium Carbide. — Tests with unirradiated samples of uranium and thorium carbides showed that they hydrolyzed readily in water. Subsequent tests with irradiated fuel samples indicated that irradiation renders the carbides nearly completely unreactive with water at 25 to 100°C. A sample of 100% PuC irradiated to 1000 Mwd/metric ton was also passive when refluxed in 13.2 M HNO_3 for over 2 hr. The carbide dissolved rapidly, however, upon the addition of 0.05 M fluoride ion (as KF) to a terminal concentration of about 125 g of plutonium per liter. Dissolution was 88% complete in 3 hr and complete in 6 hr. Excess nitric acid was removed by distillation to produce a stable Purex solvent extraction feed solution containing 2 M HNO_3 and 270 g of Pu per liter. Detection of colored organic products was not possible in the deep-green Pu(IV) solution.

A sample of 80% UC–20% PuC, irradiated to about 20,000 Mwd/metric ton by United Nuclear Corporation, was nearly passive in 80°C water. About 10 cc of off-gas per gram of carbide was evolved during the first 20 min of

reaction, and an additional 4 cc per gram was evolved at a decreasing rate over the following 6 hr. After the test a sample of the water contained trace quantities of uranium, plutonium, and fission product cesium. The irradiated carbide was then satisfactorily dissolved in 13 M HNO_3 in less than 3 hr. The resulting solution was refluxed to degrade soluble organic compounds and distilled to remove excess acid to produce a solution containing uranium (50 g/liter) and plutonium (10 g/liter) for subsequent solvent extraction tests. If reddish-brown organic products had formed, as in the test with pure UC, they could not have been detected because of the deep-green color of the Pu(IV) ion.

Corrosion. — Types 309SCb and 304L stainless steel are being evaluated for the process in which graphite-UC fuels are burned and the ashes are leached. Welded specimens of type 309SCb stainless steel were tested in eight consecutive cycles consisting of burning a fuel sample in O_2 at 800°C for 5 hr, followed by leaching in boiling 5 M HNO_3 for 2-1/2 hr. Corrosion rates were negligible. Another proposed cycle consists of disintegrating the graphite fuel in 90% HNO_3–10% H_2SO_4 at about 50°C, followed by leaching in boiling 5 M HNO_3 and then "burning" the leached graphite in 10% HNO_3–90% H_2SO_4 at 150°C or above. Type 304L stainless steel exposed for 24 hr in the third part of the cycle (10% HNO_3–90% H_2SO_4 at 150°C) corroded at rates of 0.2, 11.1, and 14.6 mils/month in the vapor, interface, and solution phases respectively. Other alloys will be tested in this environment.

Boiling 7 M HNO_3–0.05 M HF has been proposed as a dissolvent for PuO_2-UO_2 fuel. Type 309SCb stainless steel, Hastelloy F, Corronel 230, and Haynes alloy No. 25 tested with this dissolvent, in the absence of simulated fuel, corroded at maximum rates of 13.7, 28.2, 1.9, and 6.1 mils/month, respectively, for a 48-hr exposure to the leaching solution.

Two proposals for gas-phase head-end processing involve (1) the use of a 99/1 volume ratio of O_2 to HF at 600°C and (2) the use of superheated steam at 825°C. Welded specimens of "A" nickel exposed in O_2-HF for 264 hr showed small weight gains. After 840 hr of exposure to steam at 825°C (1 atm), both "A" nickel and type 304L stainless steel showed weight gains corresponding to a corrosion rate of less than 1 mil/month.

ARMY PACKAGE POWER REACTOR PROGRAM

PM Irradiation Test Program. — The PM fuel specimens operated satisfactorily in the ORR pressurized-water loop during cycle 51 (from December 20, 1963, to February 9, 1964). One problem was encountered in the operation of the loop. The equipment-room radiation monitor showed an increase in room activity from 120 mr/hr at the beginning of the cycle to approximately 400 mr/hr at the conclusion of the cycle. Water samples removed from the loop and analyzed during cycle 51 indicated that the major contributor to this increase in radioactivity was Co^{60} and not fission products.

The fuel was retained in the loop during the intercycle period. On startup of cycle 52 (February 15, 1964) the reactor was held at 26.74 Mw

so that a heat balance could be made on the loop. During the heat balance the inlet temperature of the in-pile section was 345°F. The heat balance results when extrapolated to ORR full-power conditions (30 Mw) indicated a heating rate of 214.3 kw. This would be nearly 13.5% over nominal design conditions for the fuel specimens. Therefore, in order to maintain the maximum center-line temperature of the fuel specimens near 900°F, the loop inlet temperature was reduced to 270°F. With this coolant inlet temperature the full-power output of the loop was determined to be 210.1 kw.

During the early stages of cycle 52, the radioactivity levels in the loop coolant water were studied carefully. Analysis of water samples taken at the inlet and outlet of the water purification system indicated that the ion exchangers were continuing to operate with a decontamination factor for Co^{60} of approximately 10. The Co^{60} level in the loop water continued to increase such that the activity level was now a factor of 10 greater than it was at the beginning of cycle 51. Because the Co^{60} activity level in the loop coolant was continuing to increase, it was decided to remove the fuel specimens from the loop at the next refueling period in order to inspect the fuel and the Nivco spacers in the fuel holders. Nivco is a cobalt-nickel alloy containing nominally 73.50% cobalt, 22.50% nickel, and small amounts of several other materials such as Ti, Zr, Al, Fe, etc. These spacers are used as flux suppressors and are plated with nickel. With the Co^{60} activity in the loop water rising quite rapidly, it was suspected that the Nivco spacers might be undergoing substantial corrosion. Therefore the A-1 and A-2 holders containing the fuel specimens were removed from the loop on February 22; they will be examined as soon as the fuel can be transferred to the ORR hot cell.

Water Chemistry. — The concentration of suspended solids (crud) in the water of the ORR pressurized-water loop has averaged 15 ppb over the first four months of the present test series, during which the continuous sampler has been in use. The crud level is very low, comparable with the best results reported for large reactor systems which have operated for extended periods of time. The low crud level makes the ORR facility especially suitable for the present test program in which prototype fuel elements are subjected to very high heat fluxes.

In a previously reported test, cooled water from the pressurized-water loop was filtered, heated to 260°C, and passed over a sample of Fe_3O_4 being tested as an adsorbent. The data have recently been evaluated and show that over 90% of the iron in solution was adsorbed on the Fe_3O_4, even though the final surface concentration was so large that the calculated average area available per adsorbed atom was only 2.5 A^2. This suggests that some diffusion into the oxide lattice had occurred, a process which could greatly enhance the effectiveness of Fe_3O_4 as a high-temperature adsorbent. Little dissolved cobalt escaped from the adsorption column, but only 20% was actually found on the Fe_3O_4; the remainder was presumably adsorbed on the stainless steel and nickel surfaces of the equipment holding the Fe_3O_4.

THORIUM UTILIZATION PROGRAM

Uranium-233 Purification and Sol-Gel Oxide Preparation. — The solvent extraction facility was not operated in January; however, a 4200-g piece of U^{233} metal from the Jezebel Reactor was dissolved in order to prepare feed for a solvent extraction run scheduled for February.

The new steam superheater for the thorium nitrate denitrator was received and will be installed to permit the resumption of ThO_2 preparation as needed.

During January, 140 kg of mixed thoria-urania was prepared, bringing the total to 900 kg. During the calcination of batch 85 the heating element in the calciner burned out (after 34 cycles). It was removed and replaced, and operations were resumed without major difficulty. Also during the month, trouble developed in the calciner program control. The proportioning control unit did not function, and the gas sequence switches were erratic and out of phase with the rest of the program. The cause is still undetermined.

Rod Fabrication. — The last of the 900 long rods were fabricated for the Kilorod Program during this reporting period. A scheduled cleanup, inventory, and criticality review is in progress. Some modifications to the process are being made so that the requested 200 shorter rods can be made. Equipment is being installed for the preparation of the pellet rods to be used for calibration. These rods constitute the last for the Brookhaven Kilorod Program.

Thorium-Uranium Fuel Cycle Development Facility. — 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 been completed. A detailed review of the preliminary proposal revealed that about $360,000 can be saved. This amount, when added to the contingency, raises it from 8.9% to 15.2%.

The design work on the oxide fabrication line is proceeding according to schedule. Work on solids preparation has centered around the drying and calcining equipment. In the rod-fabrication work, improvements are being sought in the ball milling and vibratory compaction equipment. Work on other steps, such as end capping and welding, fuel-rod cleaning and transfer, gamma-ray scanning, and fuel-rod inspection, is related to problems associated with wholly remote operation.

Particle Sizing of Sol-Gel Thoria and Thoria-Urania. — Several different fuel-particle sizes are necessary for successful vibratory packing into fuel tubes. A method for obtaining ThO_2 and ThO_2-UO_2 particles in a range of sizes at the gel stage, rather than by grinding the calcined oxide, appears promising, according to a method involving the introduction of the sol into a boiling organic drying solvent. Gel spheroids having 20 to 30 wt % in the −325-mesh (< 44-μ-diam) fraction have been obtained this way. Lowering the temperature of the heated solvent produced a product of generally larger particle size and increased porosity.

Sol-Gel Microsphere Preparation. — As to sol dispersion, by using a two-fluid nozzle and laminar flow it was possible to produce thoria gel spheres of which 75 to 80% were in the 150- to 250-μ-diam range. Under conditions of turbulent flow the same nozzle yielded 50 to 55% of the spheres in the same size range. The yield was increased to 88 to 89% by discharging the sol into a coaxially flowing stream of organic solvent.

With respect to column development, the tapered column containing a centrally located, tapered, metal rod to reduce uneven fluidization was used to prepare 530 g of 1150°C-calcined ThO_2 spheres in the size range 150 to 250 μ. It now appears that a straight column with a tapered center rod may have an even more stable flow pattern than the present tapered column.

Some sols were made by a special procedure. A 3.5-liter batch (3 M) of highly enriched 8 mole % UO_2–ThO_2 sol was made for the preparation of enriched sol-gel microspheres. This is sufficient sol to prepare about 2.7 kg of oxide. These microspheres will be used for accelerated irradiation tests by ORNL and General Atomic. The preparation procedure was the special one developed for 8% UO_2–ThO_2 sols reported last month, with the exception that a slightly different nitrate/metal mole ratio (0.064) was used, and the sol concentration was lowered from 4 to 3 M to decrease thixotropy.

Work in the microsphere-fabrication laboratory was based on optimizing the design of the equipment for the remote fabrication of fueled graphite elements, and an analysis of critical features in fluidized beds was begun. Of principal concern are the design of the gas entrance port and the variations in flow rate of the inert gas to maintain a stable action of the bed. Results suggest that some improvements can be made.

Irradiation of Thorium Oxide Fuels. — Metallography of vibratorily packed arc-fused and sol-gel oxide fuels from capsules irradiated to an estimated 35,000 Mwd/metric ton (Th + U) revealed voids and grain growth for the arc-fused materials, as contrasted to an absence of voids and much finer equiaxed grains in the sol-gel-prepared fuel. Preliminary results from the dissolution tests indicate that irradiated sol-gel-prepared fuel dissolved more quickly than unirradiated material.

Metallography of ThO_2–4.4% UO_2 pellets from two capsules subjected to metal burnups of 50,000 and 100,000 Mwd/metric ton was completed. The pellets from the capsule achieving the highest burnup showed a circumferential band of high porosity, less of a glassy material, and a relatively fine, uniform grain size, in contrast to the material from the other rod.

Vibratory Compaction. — Powder compaction experiments performed with a mechanically produced coarse fraction and a fine fraction produced by using heated organic solvent showed that the resultant mixture could not be packed to as high a density as a comparable mixture prepared from fractions both of which were mechanically produced. Densities in the latter case were 3 to 4% higher than in the former.

Design changes in a NAVCO BH-3 pneumatic device produced acceleration levels in excess of 30,000g. This high level is required to achieve uniform density in 10-ft-long fuel rods. Accompanying the large longitudinal impulse is a transverse wave that causes fatigue in the vibrator as well as the fuel tube.

Dispersion Hardening of Thorium Alloys. — Hot tensile-strength tests were run on thorium-base alloys containing indium, zirconium, and dispersed ThO_2. Indium is an extremely effective strengthening agent. The yield strength of alloys containing additions of 2 to 5 wt % In was increased more than twofold over the strength of the unalloyed metal. Dispersions of ThO_2 in thorium metal ranging from 4 to 13 vol % of oxide were nearly as effective as indium. Zirconium additions ranging from 2 to 5 wt % were relatively ineffective.

Permeability of Vibratorily Packed Fuels. — The effects of particle size on gas permeability and density of compacted thoria fuels are being studied. Permeability increases rapidly with increasing particle size, and packed fuel densities greater than 85% of the theoretical crystal density were obtained without using thoria smaller than 325 mesh (i.e., by using thoria particles larger than 44 μ). It is suggested that consideration be given to elimination of material smaller than 325 mesh in rod production. The increase in fuel permeability should substantially reduce the amount of entrapped water.

Sol-Gel Preparation and Properties of ThC_2 and UC_2-ThC_2. — The advantages of pyrolytic-graphite-coated microspheres of the carbides of nuclear fuels to be used in high-temperature reactors provide the incentive for seeking economical means to prepare them. Present studies are directed toward developing sol-gel methods for the preparation of the microspheres. There are three steps in the approach under study: preparation of carbon-containing sols, formation of gel spheres, and reaction at temperatures above 1450°C to produce carbides.

The best sol prepared so far was made with a channel black of relatively low surface area (460 m^2/g) dispersed in a 2 M ThO_2–3% UO_2 sol at a carbon/metal mole ratio of 4.4. Use of carbon having a larger specific surface area might lead to faster reaction rates in forming the carbides and to more complete reaction, but sols prepared with high-specific-surface-area (> 1000 m^2/g) channel blacks are highly thixotropic and will not feed well from an orifice. Thixotropy of the sols is increased by vigorous stirring and is decreased by additives such as Dow Methocel HG-400.

Sol prepared as suggested above was formed to microspheres by injection from a small orifice at right angles to a flowing stream of solvent in a drying column. Cracking was observed in spheres 500 to 1000 μ in diameter when solvents having a high capacity for water were used. The most satisfactory solvents tested were 2-ethylhexanol and 2-methylpentanol. Surfactants did not appear to be necessary to the formation of good spheres, but a small amount of carbon "shedding" from the surface of the gel spheres was suppressed by the addition of 1 to 2% of nonionic surfactants.

About 100 g of gel microspheres was prepared from the sol prepared by the optimum method mentioned above. These will be used in a firing-program study. A small sample has already been fired to produce good-appearing spheres having a density of about 6.9 g/cc.

Equilibrium Studies in the System ThO_2-UO_2-UO_3. — The equilibrium between orthorhombic U_3O_{8-y} and the cubic solid-solution ThO_2-UO_{2+x} has

been determined at the oxygen pressure of 1 atm over the temperature range from 700 to 1600°C. In addition, portions of the isobaric surface, P_{O_2} = 0.2 atm, within the one-phase, cubic, solid-solution region were determined. The boundary composition of the cubic phase at 700°C is very close to $X_{ThO_2} = 0.4$, $X_{UO_3} = 0.267$, $X_{UO_2} = 0.333$, and changes very little with temperature to above 1200°C. At higher temperatures the UO_3 mole fraction (excess oxygen content) increases smoothly as X_{ThO_2} decreases. A maximum oxygen content close to $X_{UO_3} = 0.4$ is reached at about 1525°C, the U_3O_{8-y}-to-UO_{2+x} transition temperature. The mole fraction of ThO_2 reaches zero at this temperature. Compositions of the U_3O_{8-y} phase are in good agreement with those given in the literature.

Fission-Gas Release from Thoria Fuels. — Calcium additions were made to samples containing ThO_2–3% UO_2 in a continuation of the study to determine the effects of stress on fission-gas release at elevated temperatures. It was expected that the increased plasticity resulting from the calcium addition would increase the magnitude of the fission gas released when stress was applied. Instead, the reverse was true, and efforts are now directed toward understanding the experimental observations.

Physical and Chemical Properties of Molten Chlorides. — In the study of the separation of protactinium, a series of neutron irradiations of $AlCl_3$-$ThCl_4$ mixtures was initiated in order to explore the possibility of removing the bred Pa^{233} by sublimation from the solid, along with the volatile $AlCl_3$. Protactinium, presumably present as the chloride in the $ThCl_4$, was not volatilized by heating for 21 hr at 150°C. Further studies at higher temperatures are planned.

The solubility of $ThCl_4$ in $AlCl_3$ at 210°C was indicated to be slightly less than 1.0 mole %.

MOLTEN-SALT REACTOR PROGRAM

Molten-Salt Reactor Experiment. — Progress was made in all activities concerned with the fabrication and installation of equipment for the MSRE.

The welding of brackets in place of the support ring that had to be removed from the reactor vessel was completed. The vessel was stress relieved, and the welds on the brackets were radiographed. One weld contained a crack, and it was repaired. The vessel was placed in a vertical lathe for final machining in the region of the brackets.

A stainless steel shell with many windows was made for use in testing the heat exchanger. Flow tests can now be run with the tube bundle in this shell for evaluation, visually and with instruments, of the effects of changes to the tube bundle and the shell on the vibration of the tubes.

All drain tanks were placed in their furnaces in the drain-tank cell. Much of the piping in the cell was completed. The freeze valves are being altered to accommodate the heating units and the jackets for directing the cooling air; this work was nearly completed.

Installation of the coolant-salt equipment and piping and the thermocouples and instrumentation directly connected to the vessels and piping was completed. Preparations were made for leak testing the entire coolant system with helium. Installation of heaters and insulation was started on some of the equipment.

A review of the status of the construction and the cost of the MSRE was finished. The cost of the Experiment is now estimated to be $10.2 million; the previous estimate was $9.4 million. The total of expenditures plus the value of materials and equipment completed but not yet billed through the end of January was $8.9 million. Based on expenditures and costs, the project was 87% complete on February 1; the revised critical-path schedule showed the project to be 90% complete. This latter value does not properly account for contingencies that were included in the cost estimates.

Reactor Analysis. — The reactor normally will be kept well above the freezing point (840°F) when it contains fuel salt; so it was not required that the control rods provide nuclear shutdown at lower temperatures. If the drain line were to become plugged and if it were necessary to allow the reactor to cool to ambient temperature without draining, criticality could be prevented by the addition of a neutron poison, such as Li^6F, to the fuel salt before it froze. Multigroup neutron diffusion calculations indicate that less than 0.26 kg of Li^6, dispersed in the 70 ft^3 of fuel salt, would ensure that the core would be subcritical at 70°F.

Nuclear calculations for the final design of the MSRE used multigroup cross sections generated by the GAM-I program. Since GAM-I does not include Li^6, Li^7, or F^{19} cross sections, their effect was simulated in the GAM-I spectrum calculations, and group cross sections for these nuclides were compiled separately from basic data. The GAM-II program, which recently became available, includes the lithium isotopes and fluorine. A trial calculation was made of the MSRE critical concentration, using the MODRIC program with GAM-II cross sections, for comparison with earlier calculations by MODRIC and GAM-I. The critical concentration of U^{235} predicted by the new calculation was 0.88 of the earlier prediction. The difference appears to be due to inelastic scattering by fluorine, included in GAM-II but not earlier, which reduces the age and the fast-neutron leakage probability.

Development of Components and Systems. — Testing of the prototype control rod and drive was continued. The front bearing of the servo motor in the drive had to be replaced again because it overheated and the lubricant failed. The air flow through the drive is being changed to improve the cooling, and we are looking for a better high-temperature lubricant. The flexible control rod has operated through 32,158 full-stroke cycles without difficulty.

A production model of the removable heater and insulation boxes for the 5-in. pipe was received from the Mirror Manufacturing Company, and it is being tested. The heat loss from this unit is about 12% less than the loss from the original test unit when both are operated at the same temperature.

Operation of the equipment for removing oxygen from helium was continued at 1160°F with a helium flow of 10 liters/min in order to determine the capacity of the absorber bed. When the inlet concentration of oxygen in helium was 220 ppm, the exit concentration did not rise above 1 ppm until 58% of the titanium had been consumed. The quantity of oxygen removed was estimated to be equivalent to a year or more of operation of the reactor gas system. These results were considered satisfactory, and no further testing is contemplated.

The engineering test loop has operated continuously for 4400 hr. During this time 35 samples of salt were removed and 11 capsules containing a total of 1528 g of enriching salt (930 g of uranium) were added without difficulty by means of the sampler-enricher mockup. The additions were intended to raise the concentration of uranium from 0.1 mole % to 0.2 mole %; however, the analysis of samples from the loop gave a concentration approximately 10% below the calculated value. Investigation of the discrepancy is continuing.

The enriching capsule chosen for use in the reactor is 3/4 in. in outside diameter by 6-3/8 in. long and contains 150 g of enriching salt (92 g of uranium). The salt melted and drained completely from the capsule in less than 5 min in the pump bowl.

MSRE Pump Program. — The pump tanks for the fuel and the coolant pumps for the MSRE were delivered to the reactor site several months ago. We have now completed the testing and modification of the lubricating-oil systems for both pumps and have delivered them to the site.

The supports for the fuel pump, which permit the pump to move to compensate for thermal expansion of the piping, received their final tests during 168 hr of operation of the prototype pump. The tests indicated that the supports will permit the required horizontal and vertical movement and that the vibration dampeners are unnecessary. The force required to counterbalance the weight of the pump drive motor is about 1650 lb greater than the value used in the design, but this can be supplied without overstressing the piping or components.

The first pump motor passed the electrical tests at the Westinghouse plant, and it will be shipped soon. The rotary element — shaft, impeller, shield block, bearings, seals, and housings — for the pump was assembled, and testing of the assembly was begun. The rotary element for the coolant pump was partly assembled.

Instrumentation Development. — Instrumentation development was devoted to continuation of testing and development previously reported and to assisting in the design of the MSRE instrumentation system.

A method was proposed for obtaining a precise single-point indication of the position of each control rod in the MSRE. An orifice would be installed in each control-rod thimble near the lower end, and an abrupt increase in pressure to maintain a flow of cooling air through the rod and thimble would provide the indication as the end of the rod entered the orifice. A mockup of the system was built, and experiments were performed to determine whether a usable signal could be obtained under reactor conditions. Results of these experiments indicate that usable signals could be obtained if minor modifications were made in the control-rod cooling-air piping.

NUCLEAR SAFETY

Decontamination Factors for Particle Deposition in Leaks as Applied to Leak Testing of Containment Vessels. — Further study has been made of the process whereby a varying degree of decontamination of escaping gas results from diffusional deposition of radioactive particles in containment-vessel leaks. Such particles would be expected to originate during a nuclear reactor accident and would present a serious hazard. The earlier results showed that the calculated decontamination factor is a strong function of leak size and of particle size and that it increases as either decreases. The more recent study of this attenuation effect has been primarily concerned with application of the results to containment-vessel leak testing, since potentially large credit factors may be claimed through the use of leak testing methods that are both sensitive and reliable. For example, calculated decontamination factors of 50 and higher are obtained for particles 2 μ and less in diameter if leaks corresponding to $>10^{-6}$ cm^3/sec, as measured under the accident conditions, are eliminated. Prior to applying these results in practice, additional experimental investigation may be required of the assumption, used in making the decontamination factor calculations, that the leak wall is a perfect sink for the particles. Such an investigation would need to include determination of the resistance to movement, during subsequent air flow, of a deposit of particles. Achieving the required detection sensitivity and eliminating the leaks would be very costly, but the hazard reduction could conceivably more than offset the cost. Theoretical analysis indicated that conventional leak testing data could be utilized readily in estimating pessimistic decontamination factors for particles even though detailed information on leak size and structure was not available.

Melting Experiments in TREAT Facility. — Two experimental assemblies are being constructed to investigate the release of fission products from fuel melted under transient heating conditions. These assemblies are similar in design to those previously described (ORNL-3483, -3508, -3532) and which were used in the first two experiments. Previously, unirradiated stainless-steel-clad UO_2 fuel specimens were heated in an argon atmosphere in both experiments, which simulated a transient accident with a clean core. In order to investigate a transient accident with a reactor which has been operating for some time, the next two experiments will employ fuel specimens which have received previous irradiation (10^{17} nvt) equivalent to 100 times that to be received during transient heating in TREAT. The atmosphere will be of moist air in both experiments, but the reactor transient characteristics will be varied to investigate the effect of different degrees of melting and specimen damage on the release and transport of fission products.

Innocuous Simulation of Accidents and Hazards. — Study is continuing to develop ways of simulating the radioactive materials which would be released during a reactor accident, but making use of test materials which

are less hazardous themselves. The behavior of radioiodine and its volatile compounds is being given first consideration, since radioisotopes of iodine are generally considered to be the most significant of the volatile fission products from a hazards standpoint. Previously, it had been shown that supposedly pure iodine sources contain small amounts of CH_3I, HI, ICl, and higher-molecular-weight compounds, in addition to molecular I_2. The behavior of these compounds in small-scale activated-charcoal systems is being studied under the varying parameters of humidity, temperature, and type of charcoal. Initial results indicate that the presence of moisture (relative humidity ~60%) in the system adversely affects the adsorption behavior of the volatile fraction to a much larger extent than it does the less volatile fraction. Elevated temperature (~100°C) and the presence of metallic impregnants in Whetlerite charcoal (Ag, Cu, Cr) together seem to enhance the adsorption or retention of the volatile iodine fraction by charcoal. These studies are being continued, and it is expected that with the knowledge obtained, sampling techniques can be developed for studying the behavior of these volatile iodine compounds in large-scale reactor systems.

Properties of Aerosols Produced from Molten Reactor Fuels. — Two experiments on melting trace-level-irradiated stainless-steel-clad UO_2 in the confinement filter test facility have been completed. The UO_2 was melted over a period of several minutes while a stream of air flowing at 3 liters/min was passed over the crucible to transport the smoke and volatile fission products into a 9-ft^3 stainless steel confinement tank. The two experiments differed only in that the first one had a portion of unfiltered cell air, approximately 25% of the tank volume, accidentally added to the tank after melting of the fuel was completed. This difference in the conditions of the two experiments had a significant effect on the chemical form of the released radioiodine. The proportion of penetrating forms (possibly organic iodine compounds) was increased by a factor of approximately 10. This was the ratio between the amounts of iodine found to penetrate a 1-1/2-in. charcoal bed (ORR specifications) in these tests. Summarized below are data on the iodine behavior, total fission product release from the UO_2, and the amount of each fission product deposited during a 3-hr aging period in the confinement tank.

It is interesting that the rate of removal of radioactivity from the aerosol was low in these experiments, suggesting that the particles remained too small to settle quickly even after agglomeration. An attenuation of airborne iodine by only a factor of 2 was observed during the 3-hr aging period. Analyses of sections of the diffusion tube and of the absolute filter and charcoal bed showed that approximately equal parts of particulate, molecular, and penetrating gaseous iodine were present in the confinement tank at the end of the 3-hr period.

UO_2 Aerosol–Fission Product Distribution in Filtered and Unfiltered Air[a]

	Fission Product Distribution (% of inventory)					
	Iodine		Tellurium		Cesium	
	A	B	A	B	A	B
Total release	83.8	63.8	11.7	11.8	29.2	37.2
Hot-zone deposition[b]	6.3	10.6	3.3	1.9	3.2	16.3
Total in aerosol tank	46.5	23.8	5.1	3.4	20.3	9.3
Activity remaining airborne after 3 hr	31	29.6	3.3	6.5	5.7	12.6
Amount of iodine in penetrating form	3.7	0.4				
Iodine loss through 1.5 in. of charcoal	1.3	0.14				

[a]Run A was performed in filtered air but was diluted by ~25% with unfiltered air. Run B was performed entirely in air filtered by absolute filters and a charcoal bed.

[b]Temperature in the "hot zone" varied from an undetermined value close to that of the fuel to approximately room temperature.

Release of Fission Products on In-Pile Melting of Reactor Fuels. – Two additional experiments have been conducted to study the behavior and characteristics of fission products released when UO_2 is melted in-pile under conditions simulating a reactor accident. The atmospheres in these experiments consisted of moist helium and moist air, respectively, each having a dew point of 23°C. Other conditions were similar to those in experiments previously reported (ORNL-3286, p. 10). Radiochemical analysis of one experiment is approximately 50% completed; the other is being disassembled in the hot cell.

In all these experiments, fission products appear in varying proportions in various locations, and sample compositions are, in general, different from those predicted for normal fission yields. This fractionation occurs because differences in physical and chemical characteristics of individual fission products result in differences in their behavior in release, transport, or deposition processes. Fission product fractionation is expected to occur similarly in reactor accidents. A vapor transpiration model which predicts fission product fractionation has been derived and has given satisfactory agreement with the data from the in-pile fission product release experiments. This model deals with the fractional vaporization of fission products from the heated fuel at various temperatures and their removal by the carrier gas to regions of lower temperature, where they are rapidly cooled and deposited without further change in

composition. The partial pressures of fission products in fuel material are assumed to be represented by Henry's law. A complete mathematical derivation of this model was reported (ORNL-3547). The model predicts that the fractions of any three suitable fission products, f_1, f_2, and f_3, found in the various samples deposited at various locations, are related so as to generate a straight line on log-log coordinates if the ratio f_2/f_1 is plotted vs the ratio f_3/f_1. The slope of the line is given by the ratio of the differences of the heats of vaporization of the specimens that vaporize during the temperature excursion. The intercept is a function of the entropies and heats of vaporization, as well as of the integrals of the vapor pressures of the pure fission products over the temperature limits of the excursion and of the total quantities of fission products vaporized. It must be noted that all these quantities can be calculated a priori with one exception, the quantities of fission products vaporized.

The success of the vapor transpiration model in predicting the fractionation of fission products during in-pile melting of UO_2 validates the assumptions of the model and identifies the mechanism of fractionation. The chemical forms of the fission products during vaporization are also identified in the same operation. In most of the experiments with stainless-steel-clad UO_2, the strontium, barium, and zirconium were vaporized in the metallic form; however, in experiment 5 they were in the oxide form. This type of investigation is being continued, specifically being applied to the other fission products for which data are available: iodine, tellurium, cesium, ruthenium, and cerium.

Knowledge of these fission product release mechanisms should make possible the prediction of the hazards of fission product release associated with reactor accidents, and will help in the recognition of conditions that are important in their effects on fission product release, transport, and deposition.

Characterization and Control of Gas-Borne Fission Products. — The behavior of iodine released in reactor accidents will depend upon its chemical form. Iodine compound formation in gases is being investigated with elemental iodine prepared by two rather different methods — the palladium iodide and the dichromate. It has been observed that when air is passed over an iodine source produced by the palladium iodide method, while it is thermostated to a temperature around −70°C, there is a rapid initial transport of I^{131} activity from the source, followed by a slower, reasonably constant rate of transport. This and other observations indicate that these iodine sources contain, but not necessarily at all times, two or more components, one component more volatile than elemental iodine and the other component with a volatility equal to that of elemental iodine.

Mass spectrometry indicated that the iodine source contains masses corresponding to those of HI, ICl, CH_3I, and I_2. Diffusion-tube analysis showed that the more volatile fraction deposited on a charcoal-lined diffusion tube with a diffusion coefficient of approximately 0.10 cm^2/sec, but not on silver or rubber surfaces. The less volatile fraction deposited primarily on silver with a diffusion coefficient of 0.08 cm^2/sec, characteristic of molecular iodine. Previously it was reported that iodine

compounds with very similar diffusion coefficients were observed in iodine sources produced by the dichromate method. Further work on iodine compounds is planned so that their occurrence and behavior can be recognized in nuclear safety experiments and predicted in reactor accidents.

Nuclear Safety Pilot Plant. – Installation of equipment and piping was completed, and calibration of instruments and debugging of subsystems are under way. Concrete shield blocks required for later high-level runs and containing access openings through which remote handling operations will be performed were designed. A gas sampler capable of taking 12 independent selective samples, in which each sample allows differentiation of molecular, particulate, and penetrating forms of iodine, has been designed; and procurement of a prototype has been initiated. Planning of the experimental program is continuing.

Reactor Containment Handbook. – The 12 chapters of the handbook are in various stages of review as follows: two (Chapters 2 and 4) are being prepared in final form, seven (Chapters 3, 7, 8, 9, 10, 11, and 12) have been distributed for external review with comments requested on or before April 1, and the remaining three (Chapters 1, 5, and 6) will be distributed for external review by the middle of March. It is planned that necessary revisions on all chapters will be made so that the handbook can be submitted for printing by the end of June 1964.

Nuclear Safety Information Center. – The collection, evaluation, and storage of the backlog of pertinent information continue to receive major emphasis at the Nuclear Safety Information Center. To date, over 1300 sources of information have been extracted into the Center's storage system.

Arrangements have been made to supply a miniature information center on the subject of nuclear facility containment for the Third International Conference on the Peaceful Uses of Atomic Energy at Geneva, Switzerland, August 31 to September 9, 1964. About 1000 to 1500 pertinent documents on containment will be extracted and indexed in the miniature center. Each document will be available on microfiche from which a copy will be made, of a particular document, for those desiring it. So far, 400 sources of information have been selected for use in the Geneva exhibit.

One additional study is being undertaken by NSIC. At the request of the AEC-ORO, the Center will investigate the use of and experience with reactor backup shutdown systems.

In January and February the Center received and answered 17 specific requests for information. Four of the requests were received by telephone. Four additional names have been added to the distribution list for reports prepared by the Center.

WASTE DISPOSAL

Clinch River Study. – Differences in the longitudinal pattern of radioactivity in the Clinch River for winter and summer flow conditions have

been examined for seasonal changes. In the first few miles downstream from the mouth of White Oak Creek – that is, Clinch River Mile (CRM) 20.8 to 12.5 – the increased velocities of flow in the winter period appear to cause scour of sediment deposits. During the summer period, thermal stratification, the beginning of which centers at about CRM 9.5, does not seem to affect the distribution of radioactive sediments in the lower reaches of the Clinch River, CRM 9.5 to 1.3.

A survey of radioactivity in bottom sediments in sloughs along the Clinch River was conducted with the Flounder in order to determine whether significant levels of radioactivity existed in areas outside the normal river channel. The radioactivity in the sloughs surveyed was within the range of the radioactivity in the main river channel, but maximum activities in the sloughs were not as high as maximum activities in the main channel. Bottom sediment radioactivity varies directly with the depth of water in several of the deeper sloughs. This condition was probably due to (1) the exposure of the portions of the sloughs less than 6 ft deep to subaerial erosion during winter low water in Watts Bar Lake and (2) the washing of sediment down the sides and into the deeper portion of the sloughs during periods of high rainfall and high surface runoff.

A study of interrelationships between stable-chemical constituents in Clinch and Tennessee River water has been made by processing the results of weekly or monthly chemical analyses through a digital computer program. The results of the analysis indicate that although water in both rivers is of the calcium bicarbonate type, Tennessee River water contains somewhat less calcium, magnesium, bicarbonate, and suspended and dissolved solids, and somewhat more sodium and chloride, than does Clinch River water. Correlations of constituent pairs show that iron and manganese, and at some stations silica and potassium, vary in the same manner as the suspended material in the water, suggesting that those constituents are present in the water primarily as solid particles. The suspended sediment content, turbidity, and color of Tennessee River water vary directly with discharge, as one might expect, but this pattern is not apparent in Clinch River water.

Mineral Exchange Studies. – Mass-action equations predict that in the sorption of tracer concentrations of a cation in a media containing a high concentration of a competing cation, the distribution coefficient (K_d) of the tracer ion is inversely proportional to the concentration of the competing ion. In ideal situations the log-log plot of K_d vs the competing cation concentration yields a straight line whose slope is dependent on the ratio of the charge of the cations; and for exchange of tracer levels of strontium in sodium nitrate solutions, the line should have a slope of −2.

Plots obtained with trace-level strontium in sodium nitrate solutions at pH 7 show near-ideal response with sodium-saturated forms of Wyoming montmorillonite, South Carolina hydrobiotite, and Wisconsin illite. Sodium-saturated Georgia kaolinite and Illinois illite (the latter is generally regarded as a reference illite sample) showed lines whose slope was less than −2. Activated alumina and corundum did not show appreciable decrease in sorption with increasing sodium nitrate concentrations, which suggests that these materials sorb strontium by a different mechanism.

In dilute sodium nitrate concentrations the highest K_d was found for the hydrobiotite sample; in the highest concentration (1.0 M $NaNO_3$), activated alumina gave the highest K_d. Of the clay minerals investigated, Illinois illite had the highest K_d in the concentrated solution but had only a moderate K_d in dilute solutions.

Disposal in Deep Wells: Permeable Formations. – The relative behavior of tritiated water and chloride ions as water tracers has been investigated experimentally with hydrobiotite columns. Repulsion of the chloride ion by the negatively charged surfaces of the hydrobiotite (negative sorption or anion exclusion) can be described on the basis of electric double-layer theory. In the case of the vermiculite-type layers of hydrobiotite, the successive layers are close enough (5.55 A) so that Donnan membrane theory can be used to estimate interlamellar anion exclusion. The response of anion exclusion to the expansion and collapse of hydrobiotite makes this technique useful for obtaining data on the distribution of surface charge density. Surface charge density is seemingly quite important in determining the ease of lattice collapse of the clay minerals.

Disposal in Deep Wells: Hydraulic Fracturing. – Construction of the injection plant was completed in February, and operation of the water system, the low-pressure waste system, the solids handling system, and the high-pressure slurry system was checked. A 20-min run was made to check the calibration of the densometer system.

The existing fracture at 966 ft in the injection well was plugged, and the well was slotted at a depth of 944 ft. The proportioning hopper was placed in the mixer. An injection of 35,000 gal of dyed synthetic waste solution mixed with 23,400 lb of attapulgite was made on February 13. The injection required 2 hr 20 min.

The proportioning hopper was removed, and the waste and solids storage tanks were refilled. A dye and 30 curies of Au^{198} were added to the solution in the waste tanks. The injection well was slotted at a depth of 924 ft. On February 21 an injection of 27,300 gal of synthetic waste solution mixed with about 6 lb of a cement-base mix per gallon was completed. The injection required 2 hr. The grout sheet was detected in the observation well (up-dip at 150 ft north of the injection well) at a depth of 904 ft.

High-Activity-Waste Calcination. – Methods for fixation of fission products in thermally stable solids of very low solubility are being investigated as an alternative to evaporation of the wastes to dryness and calcination of the residues. A systematic survey of the system containing lithium and aluminum phosphates and Purex waste was initiated to determine the ranges of composition in which glassy solids of low solubility can be obtained without volatilization of sulfate from the system. Effects of simulated fission product mixtures at concentrations expected in wastes from the processing of power reactor fuels of high burnup are also being evaluated.

A fixation engineering experiment was performed in the close-coupled stainless steel evaporator-pot calciner equipment, using lithium phosphate and Purex (FTW-65) waste for feed. (FTW-65 waste is formaldehyde-treated waste that will be produced in 1965). Calcium nitrate solution was added

to the pot with the waste to reduce sulfate volatility. Only minor upsets were caused by dust deposition in the off-gas line. At an average feed rate of 8.2 liters of concentrated waste per hour, this test yielded glassy solids having a bulk density of 1.9 g/cc and a macroscopic density of 2.8 g/cc. It contained 130 ppm of nitrate.

In corrosion tests, titanium 45A showed negligible corrosion when exposed in boiling Darex waste solution (the nitrates of the components of stainless steel) for 336 hr, whereas type 304L stainless steel corroded at a rate of 14.7 mils/month over the same exposure period in this solution. Coupling 304L to titanium lowered the corrosion rate of the 304L to 7.6 mils/month with no increase in the corrosion of the titanium.

Intermediate-Activity-Waste Treatment. — Laboratory studies are under way to evaluate methods for converting intermediate-activity wastes to disposable solids. Simulated ORNL waste-evaporator bottoms (6 M $NaNO_3$–NaOH solution) were incorporated in asphalt by mixing 2 volumes of waste with 1 volume of emulsified asphalt and holding the resulting mixture at 130°C until nearly all the water was distilled off. The final product had a volume about half that of the original waste and contained about 60% by weight of solids from the waste solution. After 10 weeks of leaching with distilled water, cesium was leached from the asphaltic product at a rate of 2×10^{-4} g cm^{-2} day^{-1}, and sodium was leached at a rate of 3×10^{-4} g cm^{-2} day^{-1}.

Incorporation of the waste-evaporator bottoms in cement gave a product from which cesium and sodium leached at rates about 200 times greater than those for the asphaltic product. The volume of the cement product was about 70% greater than that of the original waste solution.

Low-Activity Waste: Pilot-Plant Work. — An additional sludge dam was installed in the agitated clarifier, and some minor equipment and piping changes were made in the scavenging-precipitation head-end portion of the pilot plant. A series of tests with tap-water feed is in progress for studying the effects on clarifier-effluent total hardness of (1) feed rate, (2) copperas concentration, (3) clarifier-agitator design and rotational speed, (4) combined-vs-separate addition of copperas and caustic when phosphate ion is present in the feed, and (5) withdrawal of sludge from the top of the sludge blanket instead of from the bottom.

Low-Activity Waste: Experimental Work. — Two runs (micro pilot-plant scale) of a modified scavenging-precipitation ion exchange process demonstrated that the addition of grundite clay to the sludge eliminated the need for evaporation of eluant from the cation exchange column and improved cobalt decontamination. In the precipitation step, the cesium decontamination factor (DF) was 11, and the DF for cobalt was increased to greater than 7 from about 2.

The runs also showed that the strong-base anion exchanger, Illco TAD-1, is not effective in removing ruthenium and cobalt — two radioactive elements that remain after precipitation (without grundite added) and cation exchange. Although DF values of 2 to 5 were obtained for ruthenium,

probability plots of the small amount of breakthrough data available suggested that some ruthenium leaked from the beginning of each run. No selectivity for cobalt was indicated.

Two methods are being considered for clarification of raw waste prior to decontamination by continuous ion exchange. Jar tests on waste that approximates the analysis of tap water (pH of 7 to 8, initial total hardness of 125 ppm, and a specific conductivity of 250 micromhos/cm) show that the addition of 5 ppm of copperas followed by 70 ppm of lime will produce a dense, rapid-settling floc and crystal-clear water. The same can be accomplished by the addition of 10 to 20 ppm of alum. The former method elevates the pH to about 10, removes about 40% of the total hardness, and consequently reduces the amount of resin needed for decontamination and demineralization. The latter method lowers the pH less than 0.5 of a unit, has no effect on total hardness, results in stoichiometric precipitation of the aluminum ion added, and produces less floc than the former method.

Disposal in Natural Salt Formations. — Erection of the new headframe at the Lyons mine is essentially completed. The main structural steel has been erected, and the sheave wheels are ready for installation. Temporary hoisting arrangements will allow access to the mine by the first week in March 1964, pending delivery of the new man cage and counterweight crossarm. The mining machinery has been delivered at the site and is currently being disassembled for lowering into the mine. Contracts have been let for the approach slabs to the headframe and the electrical and structural renovations of the hoist house.

A contract for the underground transporter shield assembly has been let to the Athey Products Company, and design is proceeding. A prototype hole-liner assembly is being fabricated and should be delivered during March 1964.

Engineering, Economic, and Safety Evaluation. — Fish which feed in the Clinch River and Tennessee River downstream of White Oak Creek assimilate some of the radionuclides that are released to the river system. A preliminary analysis of the effect of contaminated fish on the intake of radionuclides by man considered all data collected during the period 1960 to 1962 as a single sample, distinguishing only between bottom feeders and sight feeders. All calculations assume fish were consumed at the rate of 37 lb/year, a value believed applicable to commercial fishermen, an admittedly high exposure group. Estimates for the Clinch River indicate that the bone of the postulated exposure group may have received the largest dose; on the average, the fraction of maximum permissible intake (MPI) attained by consuming the flesh and the total fish (bottom feeders) was 0.06 and 0.28 respectively. The flesh of sight feeders would have contributed only 0.03 of the MPI. Fish consumption by the general population is likely to be influenced by all fish harvested in East Tennessee; consequently, the estimated fraction of MPI attained would be reduced by factors ranging from 2 to 4.

PHYSICAL RESEARCH

PHYSICS AND MATHEMATICS

Medium-Energy Nuclear Physics: Heavy Particles. — The existence of a new alpha emitter in the rare-earth region was established. Whereas only the electron capture decay of Eu^{148} (54-day) was previously known, it was found that Eu^{148} also decays by the emission of 2.61-Mev alpha particles. The ratio of alpha emission to electron capture was determined to be $\sim 8.5 \times 10^{-9}$, which corresponds to a partial half-life for alpha emission of 1.7×10^{7} years. From the simple barrier-penetration formula and the measured decay energy, the theoretical partial half-life for alpha emission was calculated to be 1.3×10^{7} years. The agreement of the partial half-life calculated from theory with the partial half-life calculated from the emission rate, within experimental error, indicates that the alpha decay of this odd-odd nucleus, Eu^{148}, is unhindered.

Transfer reactions induced by N^{14} ions were investigated in the energy region 30 to 42 Mev, and excitation functions were obtained for the following reactions: $O^{16}(N^{14},N^{13})O^{17}$; $O^{16}(N^{14},N^{15})O^{15}$; $Sc^{45}(N^{14},N^{15})Sc^{44}$; $Sc^{45}(N^{14},N^{15})Sc^{44m}$; $V^{51}(N^{14},N^{13})V^{52}$; and $Mn^{55}(N^{14},N^{13})Mn^{56}$.

Medium-Energy Nuclear Physics: The 86-Inch Cyclotron. — Isomeric decay of the 160-day metastable state in Re^{184} may populate three states at excitation energies of 188 kev (8^+), 104 kev (4^-), and ground (3^-). The 188-kev isomeric state de-excites mainly (70%) by M4 radiation of 83 kev energy to the 104-kev level. The 104-kev transition ($4^- \rightarrow 3^-$) is an M1/E2 = 15 admixture, and is interpreted as a rotational excitation of the K = 3 ground-state configuration. The 83-kev M4 transition is slower than the single-particle estimate by a factor of 100. Hindrance factors for M4 radiation usually range from 1 to 10. The observed retardation may be attributed to violation of the K selection rule; the M4 radiation is one degree K-forbidden.

Cyclotron Operations: The Isochronous Cyclotron. — The operation schedule for ORIC now allows about 3 days per week for installation of components of the external ion optics system and experimental apparatus. The remaining time is used for testing and evaluation of various features of the cyclotron. Beam time for testing averages 40 to 60% of available time.

The beam positioning and switching magnets and the beam piping were installed and preliminarily aligned for conducting the extracted beam to all four target stations in the high-intensity experiment room; test beams were measured at each station. As soon as the quadrupole magnets for focusing the beam are in operation, the beam pipes will be finally aligned and the block section of the shield wall will be reassembled around the lines.

Beam quality measurements, still in progress, give an energy dispersion of between 70 and 100 kev for an extracted beam of 18-Mev protons, accelerated as 36-Mev molecular hydrogen. Beam measurements indicate radial and axial emittances of 55 and 14 mm-milliradians respectively.

In a test of the variable-energy feature of the cyclotron, the energy of an extracted beam of molecular hydrogen was varied from 27 to 40 Mev. To test the operation of the extraction system for variable beam intensities, beams in excess of 30 μa of molecular hydrogen, deuterons, and alpha particles were all extracted with uniformly good efficiency.

Two systems for measuring the characteristics of the external beam were built and tested. One system determines the profile of the beam in the beam pipe by means of a rotating wire, with oscilloscope readout. Several of these devices are now being fabricated. The other system uses a transmission line beam catcher for measuring the shape of the beam pulse; it indicates a pulse width of 5×10^{-9} sec for 36-Mev deuterons.

Physics of Fission. — A system for the measurement of correlated energies and times of flight of fission fragments has been developed. This system includes a fast-response transformer coupling scheme in which the linear signal, originating in a surface-barrier detector, passes through the transformer primary to a low-noise, charge-sensitive amplifier. The fast timing signal is obtained from the transformer secondary, and the noise added to the linear signal is negligible. When fission fragments from spontaneous fission of Cf^{252} were incident on back-to-back, closely spaced detectors, the measured time resolution was ~1.8 nsec, full width at half maximum. When a narrow band of pulse amplitudes from each detector was selected, the measured time resolution was <0.4 nsec, full width at half maximum. In fission-fragment and heavy-ion measurements, the system is used in conjunction with a zero-time detector consisting of an electrostatic accelerating and focusing lens, plastic scintillator, and photomultiplier tube for electron detection. The overall time resolution for this system was ~2 nsec, full width at half maximum.

High-Voltage Experimental Program: Search for a Spin-Spin Effect in the Interaction of Polarized Neutrons with a Polarized Ho^{165} Target. — The need for including a spin-spin term in the optical-model potential can be tested by studying the interaction of polarized neutrons with polarized nuclei. A neutron energy of 350 kev was chosen, since high polarizations can be obtained from the $Li^7(p,n)$ reaction [Nucl. Phys. 31, 78 (1962)]. The pulsed and bunched proton beam from the ORNL 3-Mv Van de Graaff accelerator was used in conjunction with a time-of-flight spectrometer. The detector consisted of two 58-AVP phototubes optically coupled to a plastic scintillator. A fast coincidence of the anode signals was required to reduce the effect of photocathode noise. This system achieved a resolution of 5 nsec with a bias level of approximately 100 kev neutron energy. In order to understand the nature of the interaction between 350-kev neutrons and the highly deformed Ho^{165} nucleus, we first measured, using an unpolarized beam and unpolarized target, the angular distribution for elastic scattering at this energy and the total cross section between 300 and 400 kev.

High-Voltage Experimental Program: Coulomb Excitation of Vibrational Triplet States and Octupole States in the Even Cadmium Nuclei. — Oxygen-16 ions with energies of 42 to 49 Mev have been used for the Coulomb excitation of higher states in the even cadmium isotopes. Double E2 excitation of the 0^+ state at 1133 kev in Cd^{114} has been observed. The E2 transition

between the excited 0^+ state and the first 2^+ state is strongly enhanced: $B(E2, 0' \rightarrow 2)/B(E2, 2 \rightarrow 0) = 0.85 \pm 0.17$. The second 2^+ state and the 4^+ state in Cd^{116} are degenerate to within 5 kev at 1222 kev excitation. The values obtained for the ratio $B(E2, 4 \rightarrow 2)/B(E2, 2 \rightarrow 0)$ are 1.42 ± 0.19, 1.84 ± 0.23, and 1.83 ± 0.23 for Cd^{110}, Cd^{112}, and Cd^{114}. States at 2051 ± 16, 1968 ± 16, 1945 ± 16, and 1900 ± 16 kev are excited in Cd^{110}, Cd^{112}, Cd^{114}, and Cd^{116} respectively. If these states are interpreted to be the result of direct E3 Coulomb excitation, the values of $B(E3, 0 \rightarrow 3)$ are 11.3, 10.6, 9.0, and 7.5×10^{-74} e^2cm^6, accurate to within ±20%, provided the branching ratio of cascades to crossovers from the 3^- state is large compared with unity. These B(E3) values represent enhancements of 13 to 22. They are from one-third to one-sixth of the values derived from yield measurements with 14- to 15-Mev alpha particles at Copenhagen [Nucl. Phys. 42, 197 (1963)].

High-Voltage Experimental Program: Collective Excitations in the Even-Even Samarium Isotopes. — The transitional behavior of the low-lying collective energy levels of the even-even samarium isotopes Sm^{148}, Sm^{150}, and Sm^{154} was investigated with reference to its dependence on neutron number. Multiple Coulomb excitation was obtained with 49-Mev O^{16} ions from the ORNL-EN Tandem Van de Graaff accelerator. In Sm^{154} and Sm^{152}, transitions from 6^+ to 4^+, from 4^+ to 2^+, and from 2^+ to 0^+ levels were studied in the ground-state rotational bands. Additional data on branching ratios in de-excitation of the β and γ vibrational bands in Sm^{152} corroborate previous results [Phys. Rev. Letters 2, 211 (1963)] on the mixing of state functions of members of the ground-state and β vibrational bands. The more highly deformed Sm^{154} nucleus exhibits this mixing to a lesser degree, as evidenced by the purer rotational spectrum. In the vibrational nuclei Sm^{148} and Sm^{150}, the one-photon 2^+ level and members of the two-phonon triplet were observed.

Stable Isotopes Development. — Tungsten isotopes separated in the 255° separators are of the highest purity yet obtained:

Isotope	Purity Achieved in 255° Separator (%)	Purity Achieved in Calutron (%)
W^{186}	98.9	97.9[a]
W^{184}	98.2	95.7[a]
W^{183}	92.2	86.2[a]
W^{182}	97.5	94.4[b]
W^{180}	14.2	11.35[b]

[a]Alpha (48-in. radius).

[b]Beta (24-in. radius).

These runs included evaluation of various tungsten charge materials and use of restricted vapor flow to control arc chamber pressure. This mode of operation with the WCl_6 charge produced the highest ion output attained from any of the tungsten charges. The vapor control technique is being applied to operations where internal chlorination of oxide charges is performed. It is expected that in this application arc pressure can be rendered reasonably independent of reaction chamber pressures, and that an increase in chlorination with greater process efficiency will result.

Thorough understanding of the characteristics of the magnetic field produced by the 255° shims and subsequent programming of ion trajectories have made it possible to produce well-defined straight-line images of focused ions at the 255° collector. Achievement of a straight-line image instead of the conventional curve greatly simplifies receiver pocket fabrication and component assembly; the time for these operations is reduced by more than 50%. Straightening of the focal line at the collector was made possible by retaining the z-wise point-to-point correspondence between ions originating at the source and those arriving at the collector, but at the same time introducing an aberration in the radial curvature of the ion exit slit and accelerating electrodes. Since the electrodes have a relatively long useful lifetime as compared with the receiver pockets, this change represents a substantial gain in equipment design and program technique.

Special Separations. — An 11.7-g sample of 85.85% Pu^{242} from the recently completed plutonium series "a" was used as feed for a second-pass separation to obtain a sample containing about 99.9% Pu^{242}. Approximately 1.5 g of Pu^{242} was monitored at the collector, but assays are not yet available. The material in the receiver will be divided into four samples taken from different sections of the pocket to determine variations in purity within the receiver and to provide a small amount of material of very high purity.

Two hundred twenty grams of 99.64% U^{233} was electromagnetically processed to reduce the 800 ppb U^{232} content and thereby permit measurement of fission to capture cross-section ratios. Preliminary assays on the 30 g of collected material indicated an activity level equivalent to 8 ppb U^{232}. The Th^{228} daughter is being removed from the samples by ion exchange separation in order to evaluate the Tl^{208} hard gamma content. With this evaluation completed, the U^{232} content can be more accurately determined.

More than 600 g of high-purity U^{238} was collected in the current separation series, using 99.983% U^{238} feed material. Preliminary assays indicate that contamination from other isotopes was reduced to 2–6 ppm. A cocurrent solvent extractor using 30% tributyl phosphate in Amsco as the extractant is in operation to purify recycle material.

Development is continuing on techniques for electrodeposition of plutonium on nickel backing. Deposits up to 0.1 mg/cm^2 thick with good adherence have been obtained, but plating efficiency is only 50%. Efforts will be continued to achieve greater thicknesses at higher efficiency.

CHEMISTRY

Analysis of Molten Salts by Electrochemical Methods. — The data obtained to date on the approximate decomposition potentials of various metals in fluoride melts at 500°C are summarized in the following table.

Metal Electrode	Decomposition Potential (v vs platinum quasi-reference electrode)		
	LiF-NaF-KF	$LiF-BeF_2$	$LiF-BeF_2-ZrF_4$
Zirconium	−1.3	−1.5	−1.1
Iron	−0.6	−0.5	−0.1
Chromium	−0.55	−0.6	−0.5
Nickel	−0.2	−0.2	+0.4
Molybdenum	+0.3		
Tungsten	+0.5		
Silver	+0.8		
Platinum	+1.5	+1.6	

The values were determined by noting the potential which must be applied to a particular metal electrode to oxidize the metal anodically. From such values and assuming a reasonable degree of reversibility of the electrode reaction, $M - ne \rightarrow M^{n+}$, the potentials of the various oxidation and reduction waves of the metals in the fluoride melts can be approximated. For example, the decomposition potentials of iron and nickel in molten LiF-NaF-KF are approximately the same as the half-wave potentials for the reduction of these metals at a pyrolytic graphite indicator electrode.

The decomposition potentials of the various metals are about the same in the three different fluoride solvents, except for iron and nickel, which exhibit pronounced shifts in molten $LiF-BeF_2-ZrF_4$. The reason for this apparent shift is not known. If, however, the same pattern holds true for the reduction wave of iron, the possibility of separating the reduction waves for iron and chromium in $LiF-BeF_2-ZrF_4$ is better than in the LiF-NaF-KF or the $LiF-BeF_2$ solvents.

Where a negative decomposition potential is observed, this is interpreted to mean spontaneous dissolution of the metal when exposed to the molten fluoride.

Protactinium Chemistry. — All effort recently has been directed toward purification and recovery of the protactinium borrowed from England, so that it may be returned. Most of the material has been processed in an analytical chemistry hot cell in Building 3019, and the products from two runs, which together contain 35.9 g of protactinium (by alpha-count analysis), have been obtained in the form of the fired oxide. In addition,

a little over 4 g of protactinium from several sources is still being purified. About 80 mg of U^{232} has also been recovered during this work.

The protactinium in the product from the first run was obtained by elution of a resin column on which protactinium loading was poor because of incomplete removal of fluoride by complexing with borate. As a result, only about half the protactinium fed to the column was loaded. In contrast, the material in the product from the second run was obtained by elution of a column on which the remaining protactinium was loaded quantitatively. Analysis of the products indicates that the first is purer; in particular, it contains much less niobium (less than 0.1%, compared with 2.5% in the second product). Both products contain some calcium, aluminum, and silicon, probably introduced with the ammonium hydroxide used for precipitating the protactinium. The alpha radiation represents nearly pure Pa^{231} in both products, and the only significant gamma-radiating impurity is a small amount of Nb^{95} in the second product.

The appearances of the two products are different. The second (less pure) is uniformly white, very much like the protactinium oxide we have been previously producing. The first, which is purer, is very light brown and much less uniform. It should be noted that this first product is the only one we have seen which has not contained appreciable amounts of niobium contaminant.

Properties of Aqueous Solutions: Species and Thermodynamic Properties. — Characterization of the so-called isopolytungstates, formed on acidification of aqueous WO_4^{2-} solutions, has been the objective of a great deal of work, without attainment of generally accepted conclusions. We have carried out a study of this system in the region of p from 0 to 1.5, where p is the average number of protons bound per atom of tungsten(VI); over most of the acidity range of these measurements, p is essentially equal to the stoichiometric number of moles of acid per mole of tungstate. As in a similar study of molybdenum(VI) recently reported, the techniques employed were equilibrium ultracentrifugation, acidity measurements, and Raman spectroscopy. Since rates of reaction on addition of acid are slow at 25°, most of the measurements were carried out at 50°C, at which temperature equilibrium seemed to be attained in relatively short times.

At a p value of 7/6, a strong dependence of molecular weight (from ultracentrifugation) on the concentration of tungsten(VI) indicated an aggregation independent of acidity, that is, that the solutions contain two species having the same number of bound protons per atom of tungsten(VI). The degree of polymerization was between 6 and 12. At p = 1.5, the degree of polymerization appeared to be about 12.

Acidity measurements on 0.005 to 0.2 M tungsten(VI) solutions were carried out in approximately 3 M LiCl. A scheme involving the ions $H_7(WO_4)_6^{5-}$ (or equivalently $HW_6O_{21}^{5-}$), $H_{14}(WO_4)_{12}^{10-}$ (or $W_{12}O_{41}^{10-}$), and $H_{18}(WO_4)_{12}^{6-}$ (or $W_{12}O_{39}^{6-}$) gave satisfactory agreement with the data, and was consistent with the ultracentrifugation results. The species are written here without indication of bound alkali ions; there is some indication, more so with sodium than with lithium, that some binding of counterions occurs.

Confirmation of one aspect of the scheme was obtained by comparing the Raman spectrum of $Na_{10}W_{12}O_{41}\cdot 28H_2O$ with that of a concentrated solution of lithium tungstate brought to the same value of p (7/6). Close

correspondence between the two indicated that the $W_{12}O_{41}^{10-}$ species exists in solution.

Spectrophotometry of Solutions at High Temperature. — The effect of temperature upon the broad vibration-rotation bands of liquid NH_3, H_2O, and D_2O in the region of the third harmonic of the N-H, O-H, and O-D stretching modes has been determined, and the marked differences between NH_3 on one hand and H_2O and D_2O on the other hand are interpreted in terms of much weaker hydrogen bonding in liquid NH_3.

For liquid NH_3 the shift in the position of the band maximum (9565 cm^{-1} at the freezing point, −78°) is small and linear (0.27 $cm^{-1}/°C$) up to 90°, the band maximum finally reaching 9645 cm^{-1} at the critical point (132°C). For liquid H_2O and D_2O the shift in the position of the respective maxima (10,160 and 7510 cm^{-1} at the freezing point) is initially rapid (3 to 4 $cm^{-1}/°C$), leveling off at 120° to a linear shift (0.27 $cm^{-1}/°C$) equal to that of the NH_3 band. The large shifts observed for H_2O and D_2O near the freezing point are consistent with the presence of hydrogen bonds and their breaking with increasing temperature. Above 120° these liquids approach the behavior of NH_3 near its freezing point.

The comparatively narrow NH_3 band (half-width = 210 cm^{-1} at −78°C) shows a temperature broadening (+5% at 70°; +24% at 132°) due to kinetic effects, characteristic of weak intermolecular association. However, the broadness of the H_2O and D_2O bands near the freezing point (half-widths of 985 and 760 cm^{-1}) and the sharp decrease in width observed with increasing temperature (54 and 53% at 250°) are indicative of strong intermolecular hydrogen bonding and concomitant bond breaking with increasing temperature.

Although the maximum band heights for all three liquids increased with temperature in the range studied, those for H_2O and D_2O showed a rate of increase (in percent per degree) 3.6 times that of NH_3. This is also consistent with the presence of and breaking of many more hydrogen bonds in the water systems than in the NH_3 system.

Physical Chemistry of Gaseous Thermodynamic Working Fluids. — The thermal transpiration effect exhibited by gases appears to provide a convenient method for the investigation of inelastic gas-phase collision processes. Recent publications on the phenomenon indicate serious discrepancies between theory and experiment in the low-pressure (Knudsen) region. An attempt has been made to account for the deviations in terms of energy accommodation of the gas molecules. On the basis of the dusty-gas model, this type of gas-wall interaction describes the anomalies quite well, even though it can be shown to be physically implausible.

Thermal transpiration data have been obtained for argon and nitrogen and are currently being analyzed.

The program for the determination of intermolecular potential energy parameters by means of gaseous diffusion is proceeding satisfactorily. Calibration of the diffusion apparatus over the temperature range 0 to 120°C with the system helium-argon is almost complete. Investigation of the temperature dependence of the diffusion of the gas pair helium-xenon will be initiated shortly.

Target Preparation. — Development work on sorbed-tritium targets indicates that neutron output and target life can be enhanced. The most

positive improvement has resulted from new or modified fabrication techniques with evaporated titanium as the matrix for tritium sorption. With 150-kev deuterons, emission levels as high as 7.0×10^7 neutrons sec^{-1} μa^{-1} were achieved. Further studies with targets containing lithium tritide or tritium sorbed in evaporated thorium should result in even greater neutron production and longer target life because of high thermal stability and greater tritium concentration.

Using electron bombardment or resistance heating or both, enriched isotopes of magnesium metal were prepared in gram quantities from the oxide with no carbon impurity or aluminum contamination greater than 10^{-3}%. Vapor-phase condensation of magnesium metal is being studied as a technique for growing highly pure single crystals of this metal and as a method for obtaining similar crystals of other metals.

Thin adherent films of plutonium oxide were prepared by electron bombardment at >2000°C. A film thickness of 1 mg/cm^2 can be readily produced by using a few milligrams of PuO_2.

TRANSURANIUM-ELEMENT PRODUCTION

Plutonium Behavior in the Tramex Process. — Since the initial HFIR targets irradiated for only a few months will still contain significant amounts of PuO_2, the dissolution of PuO_2 in HCl and the behavior of plutonium in the Tramex process were investigated. Plutonium dioxide that had been calcined at 800°C was successfully dissolved in 6 M HCl that was 0.02 M in CuF_2. Dissolution of a 1-g sample in 50 ml of dissolver solution was complete within an hour. Both copper and fluoride ions are required, since either ion in the absence of the other was not effective in catalyzing the dissolution.

Plutonium that had been dissolved in 6 M HCl—0.02 M CuF_2 was adjusted to Tramex feed conditions and then extracted with 0.6 M Alamine 336·HCl in diethylbenzene. Plutonium valences in the adjusted feed were 86% Pu^{3+} and 14% Pu^{4+}. Plutonium extraction was 99.95% complete; however, only 3% of the extracted plutonium was stripped with 8 M HCl—0.1 M NH_2OH·HCl. When 1 M HCl was used as the strip solution, 95% of the plutonium was recovered.

HFIR Target Development: Preparation of Oxides. — About 90.4 g of dense high-fired oxides containing 79.8 g of Pu^{242} was prepared for incorporation into seven target elements to be fabricated. These prototype targets will be irradiated in one of the reactors at the Savannah River Laboratory. This oxide was prepared by the hydroxide precipitation method and then calcined at 1150°C. The calcined oxide was ground with a mortar and pestle so it would pass through a 70-mesh screen. Fines generated during grinding will be recycled. The following particle-size distribution was obtained:

Screen Size	wt %
−70 +100	38.3
−100 +200	30.3
−200 +325	14.0
−325	17.4

HFIR Target Development: Design and Fabrication. — Detailed design of the Pu^{242} prototype target rods that will be irradiated in one of the Savannah River Laboratory (SRL) production reactors, starting about April 1, was completed and approved. Hardware for fabricating the seven Pu^{242} rods plus two dummy rods was ordered. It appears that the delivery dates will permit us to meet the scheduled fabrication completion date of March 21. The plutonium-target fabrication equipment and glove boxes were prepared for operation, and this equipment is being transferred to the alpha laboratories in Building 3019. Fabrication of the Pu^{242} target rods is scheduled to begin by March 2. Any delay will make it virtually impossible to have the fabricated rods at SRL by the March 27 deadline.

As to the irradiation of the plutonium target, the initial four HFIR prototype target rods, containing Pu^{239}, are performing satisfactorily in the Engineering Test Reactor. It is planned to remove one of the capsules in April and examine it during May and June. It is estimated that this capsule will have achieved a burnup of about 40% of the original plutonium atoms.

Determination of Stability Constants for Am, Cm, Bk, and Cf Complexes. — The determination of stability constants, which evaluate the affinity of ligands for various metal ions, affords a method for evaluating ligands that are of potential interest in the TALSPEAK process and for the separation of transcalifornium elements. Americium, curium, berkelium, and californium stability constants (K_s) were determined for 1,2-diaminocyclohexyltetraacetic acid (DCTA) and for diethylenetriaminepentaacetic acid (DTPA). In the DTPA system, the K_s for americium is greater than for any of the rare earths; however, stability constants gradually decrease in the following manner: Am > Cm > Bk > Cf. The K_s for californium is about the same as that for europium. Values for actinide-lanthanide stability constants with DTPA correlate with extraction positions that have been obtained for the organophosphate extractants. Stability constants indicate that DCTA may be useful in obtaining transcalifornium-element separations.

Development of the TALSPEAK Process. — The TALSPEAK process for separating trivalent actinides from lanthanides by partitioning between hydrocarbon solutions of di(2-ethylhexyl)phosphoric acid (HDEHP) and aqueous aminopolyacetic acid solutions was tested with solutions containing up to 30 g of lanthanum per liter. The feed solutions, made by dissolving $La(OH)_3$ in 1 M lactic acid, were spiked with Am^{241}, La^{140}, and Eu^{152} tracers, and the behavior of each element in two different potential flowsheet arrangements was studied by batch countercurrent tests. In the first experimental flowsheet the lanthanides are separated by extracting them away from the actinides, which are recovered in a subsequent cycle. According to the second flowsheet, the lanthanides and actinides are coextracted and then separated by partitioning in a subsequent cycle.

For the first flowsheet the lanthanide separation cycle was tested by using only seven batch countercurrent stages. The feed, containing 20 g of lanthanum per liter in 1 M lactic acid, entered stage 4. The scrub was 1 M in lactic acid and 0.1 M in sodium diethylenetriaminepentaacetate (Na_5DTPA) at pH 3.0. The extractant was a 1 M solution of HDEHP in diethylbenzene. Volume ratios were: feed/scrub/extractant = 1/10/5.

At equilibrium the lanthanum extraction was better than 99.99%, and europium extraction was better than 99.98%, while more than 99.9% of the americium was left in the raffinate. Future tests will determine the extent by which the scrub/feed ratio can be reduced.

In the first cycle of the second flowsheet, both the lanthanide and actinide groups are completely extracted from lactic acid with HDEHP, giving at least partial decontamination from other elements. The extract then becomes the feed to a partitioning cycle. In the present batch countercurrent test the first-cycle feed contained 30 g of lanthanum per liter in 1 M lactic acid. The scrub was 1 M lactic acid, and the extractant was a 1 M solution of HDEHP in diisopropylbenzene (DIPB). Relative volumes were: feed/scrub/extractant = 5/2/10. The feed was introduced in the third of five stages. The extraction of americium, lanthanum, and europium was complete.

The extract was introduced into the third of a six-stage partitioning cycle. The aqueous phase was 1 M in lactic acid and 0.05 M in Na_5DTPA. The organic scrub was a 1 M solution of HDEHP in DIPB. Relative volumes were: feed/aqueous/organic scrub = 10/7.5/2. At equilibrium the extract contained 99.9% of the europium and 99.99% of the lanthanum, while about 99.95% of the americium was removed in the aqueous phase. This flowsheet does not require the stripping cycle necessary in the first flowsheet.

Curium Recovery Facility. — Process equipment for the recovery and purification of Cm^{242} and Cm^{244} is being installed and tested in cells 3 and 4, Building 4507. Nearly all equipment except the dissolver and the product removal station has been installed. Flowsheet and equipment tests were completed on simulated target dissolution, feed adjustment, continuous product boildown, and conversion of product from hydrochloric acid to nitric acid solution. Necessary modifications to eliminate air locks in the gravity flow lines on the mixer-settler rack were made. Other equipment modifications are continuing during cold testing.

Process Equipment Development. — With respect to testing the components, annealed test specimens taken from the last order of Zircaloy-2 diaphragm stock were readily sealed in the sampler pump. A test apparatus is being set up to test the life of pump diaphragms having different annealed states to determine the best heat treatment conditions. An additional seal configuration in the form of a tongue (0.003 in. high and 0.015 in. wide) was tested in an attempt to lower the manufacturing cost. The seal was leak-tight but did not indent the diaphragm as expected.

A disconnect (1/4-in. size, with 3/8-in. tubing connection) was pressure tested for possible use in the hydraulic forming of the fuel-rod sheaths. The disconnect held against 30,000 psi at a torque of 20 lb-ft on the TRU disconnect clamp nut.

Transuranium Facility. — Progress in the construction of the Transuranium Processing Plant is generally good. All concrete roof planking is in place, part of the second-floor concrete slab has been poured, and the major portion of the exterior walls on the south side of the building has been erected. ORNL-procured material has been supplied on schedule. The first five cell-viewing-window sleeves, the cell-pit-floor sumps, and the prefabricated 2-in. Hastelloy pipe assemblies are on hand.

Final design of the cooling system for the cubicle is complete, and conceptual designs for a solid-waste disposal system, a maintenance glove box for the intercell conveyor dolly, and a decontamination facility were issued for comment. The decontamination facility consists of a large unshielded glove box into which any piece of contaminated, but not highly radioactive, equipment from the cell bank can be placed for semidirect maintenance. Transfers from the cell bank to this facility are made in the completely contained equipment-transfer case.

Design of two chemical-process equipment racks and the plugs through which the intercell piping is routed was completed. Shop fabrication of the pulse columns and other components for the first-cycle solvent extraction rack was completed, and the cell 7 sample station was started.

The design of the dissolver and all accessory equipment to be installed in cell 3, Building 4507, was completed, and shop fabrication was started. This equipment will be used in conjunction with the Curium Recovery Facility for the production of gram quantities of Cm^{242}.

Corrosion. — One-thousand-hour tests in boiling and oxygenated 6 M HCl solution were completed in order to determine the effect of ferric ion on the corrosion of welded Zircaloy-2. Duplicate specimens of the alloy were exposed in solutions containing 5-, 10-, 15-, 25-, and 100-ppm concentrations of ferric ions, as $FeCl_3 \cdot 6H_2O$. A mild acceleration of attack rates was realized in the presence of added iron. In the absence of iron the average rate was 0.2 mil/month. With ferric ions in the solution the average rates varied from 0.4 to 0.6 mil/month. All specimens picked up hydrogen during the course of the 1000-hr tests. Final hydrogen values in specimens exposed to the iron-containing solutions ranged from 63 to 150 ppm. The specimen exposed in the iron-free 6 M HCl solution showed a final hydrogen content of 89 ppm. The unexposed Zircaloy-2 contained between 38 and 53 ppm of hydrogen.

Aeration with oxygen exerted a mild beneficial effect on the corrosion of welded Zircaloy-2 in boiling 6 M HCl solution. In a nonaerated solution after 1000 hr, the observed rate was 0.6 mil/month; with oxygen aeration the rate was reduced to 0.2 mil/month during a similar period. Corrosion rates for specimens exposed to the vapors were little influenced by the presence or absence of oxygen. In both cases the rates were less than 0.1 mil/month.

METALLURGY AND MATERIALS

Preparation of Pure Materials. — A standard method for preparing single-crystal lithium fluoride of <1 ppm impurity level has now been developed. Details of the preparation procedures are discussed in an ORNL topical report [ORNL-3341 (in press)]. Design and construction details of the apparatus and equipment are available in a set of ORNL blueprints [ORNL Engineering Drawings D-56550, -56551, -56552, -56553, -56554, -56555, -56556, -56557 (1963)]. The most efficacious innovation for improving the purity of the product was the use of a "clean" box supplied with filtered air to prevent exposure of reagents to particulate contamination from the laboratory atmosphere at any step of the low-temperature portion of the procedure.

A 350-g crystal of LiF, designated ORNL-7, containing 69.24 metal atom % Li^7 was produced and made available for use at the Materials Science Center of Cornell University as a part of the AEC Pure Materials Program. At Cornell, the effect of isotopic composition on phonon scattering is inferred from the results of low-temperature thermal conductivity measurements. Crystals ORNL-4 and -5 were found to be sufficiently pure that the proportionality constants of the Rayleigh scattering term could be shown to be dependent on the isotopic composition. Analysis of the ORNL-5 crystal by neutron activation methods indicated that the metallic impurity concentration did not exceed 30 ppb.

Reactions at Metal Surfaces. — Our investigation of the factors that control the degree of protectiveness of oxide films on the refractory metals tantalum and niobium was continued with a study of the rate of diffusion of oxygen through anodic oxide films on these metals. In both cases the oxide films were shown to be highly permeable to oxygen, the oxygen easily passing through the films and dissolving in the underlying metal. As a result, large stresses are generated in the surface layers of the metals. Information gained in this research provided the basis for the development of a metal-sectioning technique (ORNL-3543, p. 4). The sensitivity of the sectioning technique has permitted measurement of diffusion coefficients of Nb^{95} in tantalum over a temperature range of 900 to 1600°C. The smallest diffusion coefficient measured was 2×10^{-19} cm^2/sec, which is smaller by several orders of magnitude than any diffusion coefficient previously measured by a mechanical-sectioning technique.

Deformation of Crystalline Solids. — The plastic deformation that can occur when a metal is stressed enables fabrication of the metal and permits it to maintain its integrity during use. One result of deformation that is important, both in understanding deformation and in the ultimate use of the metal, is texture formation. This process is a reorientation of the crystallites that make up the metal. The two aspects of texture formation are the identities of the orientations developed and the rate at which the reorientation takes place. Although the first of these aspects has been studied extensively, the second has been studied very little.

We are concerned with the rate of texture formation in copper as a result of tension and compression. We have obtained x-ray diffraction data on several specimens, and the data are being reduced. With this information it should be possible for the first time to critically check the theoretical calculations reported by G. I. Taylor in 1938 on the rate and identity of reorientation. Using the computer, we have corrected and completed his calculations.

Spectroscopy of Ionic Media. — As part of our continuing study of the nature of the molecular entities that exist in high-temperature liquid media, we have been investigating the structure of metal complexes spectroscopically. The optical spectra of transition-metal ions in crystals and liquid solutions consist of weak bands, usually in the infrared and visible regions, and much stronger bands, usually in the ultraviolet region. The weak bands give information about magnetic electrons and are being intensively investigated here and at many other places. The strong bands are much less well understood, but it is known that they are caused

in part by electrons that contribute strongly to chemical bonding between the metal ions and surrounding anions in the medium. We are investigating these bands to obtain information on this chemical bonding. We have recently measured a number of the strong bands in the spectra of tetrahedral $NiCl_4^{2-}$, $NiBr_4^{2-}$, and NiI_4^{2-} complexes and have used these data to deduce the optical electronegativity of an electron energy level associated primarily with the halide ions. The results fit in quite well with the theory that is now being developed (at various places) to relate these strong bands to chemical bonding.

Theory of Alloying. — We have experimentally investigated the nature of the electron energy bands in beryllium. Several of the galvanomagnetic properties of a high-purity crystal have been measured at 4.2°K in magnetic fields up to 30 kilogauss. This has enabled new information to be obtained on the Fermi surface (a geometrical description of the most energetic electrons in the metal). Some rather exceptional effects are observed when these properties are measured with the magnetic field simultaneously perpendicular to both the current and the c axis of the crystal. These measurements are directed toward a theoretical understanding of the mechanical and thermodynamic properties of beryllium and the effects of alloying additives on these properties.

Fundamental Research in X-Ray Diffraction. — Investigations of mechanisms of radiation damage in crystalline structures are important both to the development of practical reactor materials and to the understanding of the fundamental physical properties 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. Our x-ray diffraction investigation of the structure of radiation-damaged BeO has continued with recovery of single crystals, grown from V_2O_5, Li_2WO_4, and Cs_2MoO_4 melts, after irradiation in the ORR at 147 and 670°C. Crystals irradiated to 7×10^{20} neutrons/cm^2 (neutron energy above 1 Mev) produce diffraction effects similar to those observed from other BeO crystals irradiated to equivalent and higher fast-neutron doses at comparable temperatures. Crystals irradiated to a lower dose (probably about 1×10^{20} neutrons/cm^2) produce new diffuse scattering effects not observed in diffraction patterns of crystals irradiated to high doses. We will attempt to interpret these new effects through a model of the damaged BeO structure that will be consistent with the model of the highly damaged structure previously proposed.

To understand the oxidation behavior of metal crystals, a detailed knowledge of the structure of the surface films is necessary. Some of this knowledge can be obtained by x-ray diffraction techniques. Continued research on very thin Cu_2O films formed on copper single crystals has revealed twin relationships of the oxide on {111} crystallographic surfaces of the metal. The predominant orientation of the oxide on this surface is related to that of the copper by a 60° rotation about the <111> axis normal to the surface. From crystal to crystal, the prevalence of this twin orientation varies from very slight to three times the prevalence of the oxide that has its cube axes parallel to those of the copper substrate.

Crystal Physics. — To use currently available materials more skillfully and to aid in designing special properties into new materials, basic information is seriously needed. In order to characterize many physical phenomena unambiguously, the use of single crystals is required. Suitable single crystals, particularly of refractory substances, are frequently difficult to obtain. Thus, we are conducting a continuing program on crystal growth to provide crystals needed in research, to create new materials with specific enhanced properties, and to devise and improve methods for growing crystals.

Our preparation of erbium and other germanomolybdates (ORNL-3561, p. 39) has now been extended to additional rare-earth elements. We have grown millimeter-sized crystals of eight compounds of the formula R_2GeMoO_8 (R = Pr, Nd, Gd, Tb, Dy, Ho, Er, Yb). The erbium compound shows unusual magnetic properties. Although the magnetic anisotropy and the dependence of the magnetic susceptibility on field strength had implied ferromagnetic behavior, we now find that the temperature dependence of the susceptibility from 60 to 300°K and neutron diffraction data from 77 to 300°K indicate that this material is paramagnetic and not ferromagnetic.

We had previously developed a method of growing ThO_2 crystals up to 3 mm on an edge by mass transfer in the solvent $Li_2O \cdot 2WO_3$ at temperatures above 1200°C. More recently we have found that the addition of approximately 1 wt % B_2O_3 to the solvent increases the growth rate tenfold while still maintaining good crystal quality; the present growth rate is 0.5 mm in 24 hr. Well-developed, optically clear octahedral ThO_2 crystals up to 2 mm on an edge have been grown doped with the lanthanide ions. These crystals are being used in studies of the electron spin resonance and optical absorption of these rare-earth ions in the thoria host lattice.

Fundamental Investigation of Radiation Damage in Solids: Theory of Radiation-Enhanced Diffusion. — An analysis has been made of the interstitial and vacancy concentrations produced during irradiation in the presence of pair recombination and/or diffusion to fixed sinks as annihilation mechanisms. Previous treatments of the problem were extended. Assuming that the interstitials were the more rapidly moving defects and that they were in a quasi-steady state, various approximate solutions to the problem were developed. Consideration was given to the ordering that may be produced in alloys as a result of radiation-enhanced diffusion. It was concluded that many experiments are performed under conditions such that it is not safe to assume that the defect concentrations are at their steady-state values during the experiment. The dependence of enhanced diffusion on the defect production rate, temperature, defect motion energies, and sink density has been obtained for the various approximate solutions. The theory has been compared with recent experiments on radiation-enhanced ordering in Cu–15 at. % Al.

Fundamental Investigation of Radiation Damage in Solids: Paramagnetism of Germanium Glass. — Paramagnetic centers produced in germanium glass by irradiations at ~30°C in a cesium gamma source have been observed by electron spin resonance techniques at 78°K. The most intense line was asymmetric and was typical of an axially symmetric center in a glass. The line could be fitted with $g_\perp = 1.995 \pm 0.001$ and $g_\parallel = 2.002 \pm 0.001$. For

a dose of ~2×10^8 r an estimate of the spin concentration (8×10^{17} spins/cm^3) was an order of magnitude higher than the estimated impurity concentration. It is therefore assumed that the defect is intrinsic to the GeO_2. There was a decrease in the rate of increase of the spin concentration with increasing dose, although saturation was not complete. Another line was observed at a lower field whose intensity was ~0.1 that of the line discussed above.

Fundamental Investigation of Radiation Damage in Solids: Free Hydrogen in Synthetic Quartz Crystals. — The electron spin resonance of free hydrogen in irradiated silica glass at ~78°K has been reported. A resonance spectrum has been observed in a single crystal of synthetic quartz which has been attributed to free hydrogen. The specimen was irradiated with 1.7-Mev electrons at ~300°K and then with Co^{60} gamma rays at ~80°K, and measurements were made without warming. The lines, which are attributed to free hydrogen, have a separation of 520.6 oersteds when the applied field is parallel to the c axis and were easily saturated. The g tensor was isotropic within the accuracy of the measurement and had a value of 2.0020 ± 0.0003. The width of the lines was ~0.1 oersted and was attributed mainly to field inhomogeneity. An anisotropy of the hyperfine interaction was observed with a magnitude of 0.7 oersted. Additional interactions of the free-hydrogen paramagnetic electron with Si^{29} were tentatively identified on the basis of intensity ratios. An interstitial site can be found which has the required symmetry. The signals disappeared after the specimen was warmed to 300°K and then recooled to 78°K within a few minutes.

Fundamental Investigation of Radiation Damage in Solids: Test of the Kinematic Theory of Crystal Dissolution. — From the assumption that the dissolution rate at any point on the crystal surface is a function only of the crystallographic orientation at that point, the kinematic theory predicts the dissolution shapes of a crystal as a function of time. An important prediction of the theory is as follows: If one plots the family of curves which are the dissolution profiles of the crystal at successive times, points on successive profiles which have the same crystallographic orientation have abscissas which are proportional to time. This prediction has been tested for the anodic dissolution of copper crystals in mixtures of HCl and HBr solutions. Profiles of dislocation etch pits formed on (111) surfaces in these solutions have been determined from interference photomicrographs taken as dissolution progressed. The pit profiles developed as predicted by the kinematic theory, thereby substantiating the theoretical assumption for the conditions studied.

Fundamental Investigation of Radiation Damage in Solids: Role of Crystalline Orientation in the Behavior of Copper Electrodes in Chloride Solutions. — The study of the electrode behavior of copper single crystals has been carried out in the low-current-density range, where crystallization phenomena are most likely to be rate determining, and a halide solution has been chosen as the electrolyte, in view of the already established fact that dislocation etch pits tend to be formed by anodic attack in such solutions. Single-crystal copper electrodes having (100), (110), (111), and (321) orientations, as well as polycrystalline specimens, were

used as electrodes in copper-containing chloride solutions, previously equilibrated with copper metal. It was found that, in the range of current densities investigated, the rate-determining step for the electrode reaction could be attributed neither to the solution phase nor to the exchange reaction between metal and solution, and it was concluded that surface properties were decisive kinetically. Overvoltage values were found to differ considerably for different orientations; in particular, (100) oriented electrodes showed considerably higher overvoltages, having an extremely high $(d\eta/di)_{i=o}$ of the order of 10^5 ohm-cm. Differences between other close-packed orientations were not as great, and the lowest values were exhibited by (321) oriented electrodes. The facets developed by anodic dissolution also were different: (100) surfaces behaved in a sharply different way compared with all others; only orientations vicinal to (100) were formed, whereas other orientations developed a completely different set of facets, including {111} and {110} close-packed surfaces.

Fundamental Investigation of Radiation Damage in Solids: Mass Spectrometric Studies of Sputtered Particles from Copper Monocrystals. — Copper single crystals were bombarded with Ar^+ ions of 3 kev energy, and the positive ions ejected at 90° from the bombarding beam were mass analyzed. The mass spectrometer is comprised of a 6-in.-radius, 60° sector system capable of operating at a vacuum of $\sim 10^{-9}$ torr. Analyses were made of the positive ejected ions as a function of baking out the system. A clean surface as prepared for sputtering experiments was used as the basic surface; then several high-vacuum techniques were employed, and the degree of surface cleanliness was evaluated by comparing the spectra of the extraneous ejected positive ions. The peak heights and half-widths for mass 63 and mass 65 as a function of angle of incidence were investigated. It was found that the ratio of sputtered peak heights for $^{63}Cu^+$ to $^{65}Cu^+$ compared favorably with the isotopic ratio only for an angle of incidence between 72 and 80°.

Fundamental Investigation of Radiation Damage in Solids: Monte Carlo Range Calculations for a Thomas-Fermi Potential. — Earlier Monte Carlo calculations of the ranges of atoms having energies from 1 to 100 kev, slowing down in a random solid through binary elastic collisions, have been extended by using a Thomas-Fermi potential to represent the interaction between the moving atom and a lattice atom. The screening radius of the potential is that derived by Firsov. The calculations have been made for a wide variety of target-to-projectile mass ratios. Except for the highest energies, the calculated ranges are considerably shorter than those found previously with the exponentially screened Coulomb potential. Most of the experimental range data lie between the range curves calculated for these two potentials, although the Thomas-Fermi potential gives somewhat better overall agreement. The shapes of the calculated range distributions give close agreement with those found by experiment in amorphous solids. Average ranges calculated by integrating the reciprocal of the stopping power agree fairly well with these Monte Carlo calculations, especially when the mass of the slowing-down atom is large compared with that of a lattice atom.

CONTROLLED THERMONUCLEAR RESEARCH

Plasma Physics. — Rather extensive image-converter studies have been made of the velocities of moving striations in the inert gases, as a function of background gas pressure. The results have been interpreted in terms of ionic sound waves. The theory of ionic sound waves has been modified to include the loss of wave energy to the background gas through collisions between ions and gas atoms. The modified theory predicts that the waves should be slowed down by the collisions. Quantitative agreement between the ionic sound wave velocities predicted by the modified theory and the experimentally observed striation velocities is found. The theory also predicts a strong damping of the waves, which is not observed experimentally. However, the input power is much greater than the power loss by ion-atom collisions, so that energy is possibly available to overcome the damping.

The transition between a normal, magnetically supported vacuum arc and the Neidigh mode II pressure-gradient arc has been experimentally studied in more detail. The new experimental evidence strongly supports the previously discussed theoretical model (ORNL-3561) of the phenomena. The essential feature of this model is that the transition is due to a sudden, complete ionization or "burnout" of the neutral gas in the ion-emitting plasma.

DCX-2 Facility. — Continuation of injection experiments with the lithium arc have reached considerably higher ion densities since it was discovered that the plasma density was extremely sensitive to adjustment of the magnetic field. An arrangement for integrating the foil neutral detector currents following beam turnoff (measuring total stored plasma decaying by charge exchange) showed a strong maximum of stored plasma with a very small adjustment (~5%) of the small magnetic dip into which the beam is injected. With optimum adjustment, about one-half of the plasma decays in 10 to 20 msec, the remainder decaying with a time constant of 0.1 to 0.2 sec. Scans with the neutral energy spectrometer showed a strongly peaked emission of the charge-exchange neutrals perpendicular to the magnetic field at its center (~1 to 2° wide), as well as the two broader and weaker peaks showing the pitch angles at which the ions were trapped. This strong peak seems to be an effect of the plasma. The ion cyclotron radiation consists mostly of the third harmonic of the proton cyclotron frequency with the beam on; after shutoff the very weak fundamental becomes very much sharper in frequency and shows a small increase in frequency with time, disappearing at the proper frequency for Larmor motion of protons in the magnetic field. This shift, taking some tenths of a second, is believed to be due to diamagnetic change of the magnetic field. The shift of ~50 kc (out of 18.2 Mc) corresponds to a plasma density of $\sim 6 \times 10^{10}$ ions/cm^3 if the mean energy is 300 kev. This is a ratio of particle pressure to magnetic field pressure (β) of 0.005! A practical thermonuclear reaction might be imagined with a deuterium-tritium plasma at a β above 0.01 to 0.02. The plasma ion energy is spread wider than before, with appreciable numbers of ions up to 2 Mev; the mean ion energy is ~600 kev.

Vacuum. — The ultrahigh-vacuum test facility was operated for the purpose of making two studies, one a determination of the compression ratio of a high-performance diffusion pump for hydrogen, and the other a mass spectrometric observation of the gases evolved from a copper surface as the temperature was raised from 80°K to near room temperature. The compression ratio for hydrogen was found to be at least 10^4, some tens of times as large as a value reported in recent literature. The source of this discrepancy is not yet clear. The occurrence of eight or more desorption pressure peaks upon warming over the 200°K temperature interval appears to be associated primarily with desorption of specific compounds characteristic of the vacuum system and having heats of desorption from 4 to 15 kcal/mole. The tentatively identified order of desorption for the first few of the species is: CH_4, CO_2, C_2H_6(?), C_3H_8(?), H_2O, and possibly C_4H_{10}. These species desorbed at partial pressures of about 10^{-6} torr over the temperature interval 80 to 160°K. The mass spectrometer was relocated in the expectation of increasing the sensitivity for this type of experiment.

A retarding potential difference circuit was designed for the time-of-flight mass spectrometer to study the appearance potentials for fragments from organic compounds in greater detail as a potential diagnostic aid. The feasibility of adapting the time-of-flight spectrometer to the study of ions emitted from filaments and synthetic β-eucryptite, $LiAlSiO_4$, was established. From tungsten and tantalum filaments, the lower-molecular-weight potassium and sodium ions Na^+, Na_2^+, and K^+ were observed. From platinum, very little emission of these ion species occurs. Upon loading the filament with the silicate, the expected lithium ion was observed along with an apparent increase in masses 23 and 30. The nature of these peaks has not been determined (possibilities are Na^+, $NaLi^+$, LiO^+, Li_2O^+).

Performance tests of commercially available vacuum block valves representing five manufacturers were completed, and the results are being correlated in order to provide better specifications for the purchase and acceptance of such valves.

Previous tests of the effects of magnetic fields upon the sensitivity of ion gages (used in the measurement of low pressures) were conducted with the use of magnetic field strengths above 30 gauss; a new series of tests was begun in which field strengths down to as low as 1 gauss will be used.

BIOLOGY AND MEDICINE

TERRESTRIAL AND FRESH-WATER ECOLOGY

Floor Income and Distribution of Cs^{137} in Tagged Forest. — Previous progress reports have described several steps of the transfer of Cs^{137} in the tagged tulip poplar (*Liriodendron tulipifera*) forest. The following is a summary inventory of the amounts of cesium at the end of the first growing season and an interpretation of the above-ground and below-ground movement of cesium into the soil. Sampling of the soil, including

the organic forest floor layer and the mineral soil to a depth of 4 in., indicates the presence of 293 $\mu c/m^2$ (31% of the initial 936 $\mu c/m^2$) of Cs^{137} within this shallow layer of the forest floor, only 5 months after inoculation. The distribution of Cs^{137} among the components of this forest soil system (in $\mu c/m^2$) is as follows: litter, 15; rock (>2 mm), 9; roots (>1 mm), 57; roots (<1 mm), 25; fine organic, 84; soil (< 2 mm), 103. Surface income of Cs^{137} from rain leaching and from litter fall of canopy components accounts for a total of 63 $\mu c/m^2$, 21% of the activity found within this 4-in. surface layer. The remaining 79% (230 $\mu c/m^2$) is income via the root systems of the tagged trees. While 28% (82 $\mu c/m^2$) remains in inseparable roots, the larger fraction (51%) is associated with the finer fraction of the soil. This includes 29% (84 $\mu c/m^2$) still associated with fine organic tissue (including root hairs) which is digested by 1 N nitric acid and 22% (74 $\mu c/m^2$) which may already have reached mineral soil by root exudation, root leaching, or death of root materials. The above suggests that mineral turnover (at least for some elements) may be as great below ground surface as above ground surface in this forest ecosystem.

Translocation of Cs^{137} in Tulip Poplar Seedlings. — Seeds were collected from trees in the Cs^{137}-tagged tulip poplar forest. They were stratified, germinated, and grown under controlled environmental conditions. Harvests were made to determine the Cs^{137} content of developing seedling components. At 1 week of age the cotyledons contained 40.8% of the total plant Cs^{137}; roots, 26.3%; and stem, 12.8%. The remaining 20.1% was in the unattached woody carpel and wing. By 3 weeks the first pair of leaves were developed, and the distribution was as follows: cotyledons, 20.1%; roots, 38.1%; stem, 12.8%; leaves, 11.1%; and the remainder in the old carpel. By the seventh week, two pairs of leaves were developed, and the distribution was: cotyledons, 13.7%; roots, 38.3%; stem, 11.7%; leaves, 17.8%; and carpel, 18.5%. The total dry weight of the plant increased from 18.8 mg at 1 week to 71.3 mg at 7 weeks, the weight of the cotyledon increasing slightly from 2.3 to 4.3 mg. From the first to the third week of seedling growth, the cotyledons lost half of their Cs^{137} to other rapidly growing tissue. During the next 4 weeks, additional radiocesium was translocated from cotyledons to new leaf growth, while the levels in stem and root tended to remain stable. Further translocation is being followed as the seedlings mature, and attempts will be made to estimate the losses of Cs^{137} from these plants by leaching and by death of roots.

Nuclear Volume Measurements. — A study is currently in progress which seeks to establish an index of radiosensitivity for economically important endemic woody species by determining the volume of interphase nuclei in stem apexes. Results of radiation experiments with northern species at BNL suggest a direct relationship between nuclear volume and relative radiosensitivity. Techniques for measuring the nuclear volumes of endemic woody species were tested during the past report period, and the following method proved most satisfactory. A tertiary butyl alcohol series is used for dehydration and infiltration. Paraffin sections are cut at 8 to 10 μ thickness, stained, and made into permanent slides. From the various

slides, numerous micrometric measurements are made of the interphase nuclei in the tunica-corpus tiers of the apexes. Two diameter measurements of each nucleus are made at right angles. The number of apexes utilized for each species varies from 1 to 10. The total number of measurements varies from 40 to 150 per species. From the mean interphase diameter the nuclear volume is calculated. Fourteen species were measured during January.

The average interphase nuclear volume ranged from 44.60 to 198.59 μ^3 for deciduous trees, and from 356.75 to 637.83 μ^3 for coniferous trees. The local coniferous trees have nuclear volumes similar to those of northern species and therefore should exhibit the same degree of sensitivity.

Selection of Radioactive Muds by Mud-Dauber Wasps. — Study is continuing on the transport of mud from waste disposal areas at ORNL by mud-dauber wasps. Mud nests of *Sceliphron* wasps were often built with radioactive mud found nearby, but nests of *Trypoxylon* wasps in the same places contained no radioactivity. Nests collected last autumn from various sites are being studied to discover what factors may be responsible for this difference in mud utilization by the wasps. Of tests performed so far, a consistent difference has been disclosed only by kiln firing of the nests. Forty mud nests from the same site, 20 of *Trypoxylon politum* and 20 of *Sceliphron caementarium*, were fired in a kiln at 2200°F. The firing disclosed variations in clay and mud usage which were not visible before firing. Additions of new cells to old nests and extensive patching of old nests were among the construction details made obvious by kiln firing. The only consistent difference between nests of the two wasp species was the presence of a thin outer layer of dirty gray manganese compound on the *Trypoxylon* nest tubes, which was lacking in the *Sceliphron* materials. Differences in mud utilization by wasp species will be studied further in field experimentation.

Strontium and Calcium Uptake by Snails. — With the availability of strontium-free calcium carbonate we were able to demonstrate precisely the relative effects of calcium and strontium in the environment on the deposition of calcium and strontium in the shell of the snail *Physa heterostropha*. Snails were grown in laboratory environments with contrasting calcium and strontium concentrations within the range of 0 to 10 ppm Sr and 0 to 200 ppm Ca; Sr^{85} and Ca^{45} were used as tracers to determine uptake. Response surfaces, representing uptake, were calculated from a least-squares fit of the uptake data. Calcium uptake, represented by a plane response surface, increased slightly with increasing calcium concentrations; the strontium concentration had a slight negative effect. Strontium uptake was described best by a quadratic expression and a corresponding curved response surface. At less than 100 ppm Ca, the strontium uptake was almost directly proportional to the environmental strontium concentration for a given calcium concentration. However, there was a slight negative effect of calcium on strontium uptake. At the higher calcium concentrations the opposite behavior was noted: increased environmental strontium depressed strontium uptake, and increased environmental calcium enhanced the uptake. This reversal of the relation of strontium deposition to different environmental concentrations of calcium and strontium results in an area of minimum uptake. If this minimum uptake should prove to exist for other species, it would have significant implications for the

reduction of Sr^{90} uptake in the event of high levels of environmental contamination. The study also showed that there was a poor correlation between radiostrontium uptake and the calcium concentration in the environment. The concentration of stable strontium in the environment was a much better indicator of the behavior of radiostrontium, and the environmental concentrations of both strontium and calcium are even more useful for predicting radiostrontium uptake.

Chromosomal Aberrations in Chironomus tentans. — Previously, it was reported that chronic low-level irradiation from radioactive waste in White Oak Creek increased the kinds of chromosomal aberrations found in the larvae of a natural population of Chironomus tentans. The number of larvae analyzed for chromosomal aberrations has been increased to 578 in the contaminated area and 568 in the uncontaminated areas. Two new inversions, each involving several bands, were observed one time in the additional 213 larvae from the contaminated area. No new inversions were observed in the 212 additional larvae collected from the uncontaminated area; however, one inversion that occurs in the contaminated area was found for the first time in the uncontaminated area. Now, 17 different chromosomal aberrations have been found in 578 larvae in the White Oak Creek population, which is receiving approximately 230 rads/year, whereas 6 different aberrations have been found in 568 larvae from the uncontaminated area. The observation of these two new inversions strengthens the previous conclusion that more chromosomal aberrations are occurring and being eliminated from the natural population exposed to chronic low-level irradiation.

Analog Computer Simulation of a Five-Compartment Ecosystem. — Movements of energy or materials such as radionuclides in ecological systems can be simulated with an analog computer. Study of the following five-compartment system is in progress:

$$\dot{X}_1 = \tau_{01}X_0 - \tau_{12}X_1 - \tau_{10}X_1 - \tau_{14}X_1 ,$$
$$\dot{X}_2 = \tau_{12}X_1 + \tau_{42}X_4 - \tau_{23}X_2 - \tau_{20}X_2 - \tau_{24}X_2 ,$$
$$\dot{X}_3 = \tau_{23}X_2 - \tau_{30}X_3 - \tau_{34}X_3 ,$$
$$\dot{X}_4 = \tau_{14}X_1 + \tau_{24}X_2 + \tau_{34}X_3 - \tau_{42}X_4 - \tau_{40}X_4 ,$$

where X_i is the quantity of material in compartment i (i = 0, 1, ... , 4, with zero denoting the environment, 1 the primary producers, 2 the herbivores, 3 the carnivores, and 4 the decomposers), and τ_{ij} is the instantaneous rate of transfer from compartment i to compartment j.

The response of the system to four input voltage signals through τ_{01} has been investigated: (1) $X_0 = 5$ v, (2) $X_0 = 5\ [1 - \sin\ (t/10)]$ v, (3) $X_0 = 2.5\ [2 - \sin\ (t/10)]$ v, and (4) $X_0 = 5\ [1 - \sin\ (t/20)]$ v. With parameter values of $\tau_{01} = \tau_{12} = \tau_{23} = \tau_{i4} = 0.05$, $\tau_{42} = 0$, $\tau_{10} = 0.1$, $\tau_{20} = 0.2$, $\tau_{30} = 0.3$, and $\tau_{40} = 0.4$, each compartment came to equilibrium within 40 sec (real time). Equilibrium values were $X_1 = 1.3$, $X_2 = 0.22$, $X_3 = 0.031$, and $X_4 = 0.19$ v. The sinusoidal inputs produced oscillatory steady states around these values, the amplitude of oscillation being inversely proportional to the input frequency. The low-frequency input (4) resulted in a widely fluctuating steady state.

This result has an interesting application to a classical ecological problem: population instability at high latitudes. Both marine and terrestrial communities of arctic and subarctic regions are subject to wide variations in time. This is usually attributed to the simplicity of the food webs in such regions. However, food webs also tend to be simple in arid and semiarid regions of temperate and tropical zones, but these environments are not noted for pronounced population cycles. Since the seasonal input of solar radiation (corresponding to X_0 in the above system) is increasingly of longer period toward the poles, the computer simulation suggests that this factor may be a primary determinant of population instabilities in these regions, with the simple food webs a secondary effect.

Evaluation of Fission Product Distribution and Movement in White Oak Drainage System. — The slug of tritiated water that was recently injected into the groundwater within the waste pit area has, after 2 months, spread southwestward and broadened into a tract of contamination that is about 8 to 10 ft wide and between 10 and 20 ft long. Variations were observed in the time of initial arrival and in the peak concentrations of activity at different wells located within the zone at the same distance from the injection points. These results show that the tritium is moving at many different rates in slightly different directions within the contaminated zone.

By x-ray diffraction techniques it has been established that the clay fraction of the sludge, which contains a large part of the activity in waste pits 2 and 3, is predominantly illite.

RADIOLOGICAL PHYSICS, HEALTH PHYSICS, AND RADIATION INSTRUMENTATION

Radiation Physics: Photoelectric Effect in Metals. — Photoemission from a bounded free-electron metal has been examined on the basis of semiclassical models of the surface and volume photoprocesses. We propose that the surface photoeffect may be considered to be the inverse of the transition-radiation process. In the present approximation it is possible to exhibit the connection between these two phenomena in detail. The vectorial effect in surface photoemission is related to the complete polarization of transition radiation in the plane of emission. The probability distributions for the two processes exhibit striking similarities. The effect of collective oscillations in a thin metal film may lead, under certain conditions, to a maximum in photoelectric yield, analogous to maximums in the yield of transition radiation observed at certain film thicknesses. The volume photoeffect is described on a phenomenological basis; transport of nascent photoelectrons to the outside of a metal may be calculated by using cross sections and transport theory employed previously in another connection [Health Phys. 9, 883 (1963)]. The effect of excitation of surface oscillations upon the transmission probability of a metal surface for photoelectrons has been examined.

Radiation Physics: New Technique for Measuring Range of Low-Energy Charged Particles in Solids. — Recent experiments in electron tunneling have suggested that thin film sandwiches might be used to measure the

ranges of low-energy particles in films too thin to be self-supporting. The technique consists in vacuum evaporation of a thin layer of silicon monoxide 10 to 100 A thick onto a conducting solid. A thin conducting film of the material to be studied is then evaporated onto the silicon monoxide. The latter provides electrical insulation between the solid and the film, so that the final stopping place of an incident charged particle may be determined by measuring the currents to the solid and the film. Any charged particle which may be generated in an ion source may be used, and the films may be as thin as 50 atomic diameters. Information may be obtained also on the charge state of the ion as it leaves the thin film, that is, on whether or not it has captured an electron. The thickness of the insulating layer is measured by determining its radioactivity after neutron irradiation in a reactor. For the thin film, the thickness is determined either by interferometry or by neutron activation.

Radiation Physics: Interaction of Low-Energy Electrons with Organic Molecules. — The interaction of electrons having energies from thermal energy to approximately 10 ev with organic molecules was studied by using the swarm method, which allows control of the distribution of electron energies.

Two essentially different processes, a nondissociative electron capture and a dissociative one, have been observed. In the case of C_6H_5Cl and C_6H_5Br, free electrons are captured by the nondissociative process at thermal energies, whereas the dissociative process peaks at an electron energy of ~0.76 ev for C_6H_5Cl and ~0.7 ev for C_6H_5Br. Energy-integrated cross sections for dissociation have also been obtained. Because of the fact that electron transport is considered to be of major significance in the biological function of organic molecules, an effort was made to discover the behavior of representative organic molecules of biological interest under our experimental conditions.

Radiation Physics: Effect of Surface Losses in Stopping-Power Theory. — The Bethe-Bloch stopping-power formula was derived by assuming that an energetic charged particle interacts with atoms randomly distributed in an infinite medium. The existence of charged-particle energy losses of a special type associated with the presence of boundaries has been postulated [Phys. Rev. 106, 847 (1957)] and has been confirmed experimentally [Phys. Rev. 110, 657 (1958); 115, 869 (1959); 116, 81 (1959); 118, 640 (1960)]. Such surface losses result in a larger total energy loss in a foil of given thickness than one would predict with conventional stopping-power theory. These losses affect the total energy loss appreciably in the case of thin metal foils in which conduction electrons account for an appreciable fraction of the total oscillator strength. An approximate correction formula, which may be used with the Bethe-Bloch formula to obtain $-dE/dx$ in such cases, has been written.

Radiation Physics: Temperature and Thickness Dependence of Transition Radiation from Thin Silver Foils. — The radiation emitted by vacuum-evaporated silver foils bombarded by 25-, 40-, and 60-kev electrons was investigated as a function of foil thickness (300 to 2300 A) and temperature (90 to 390°K). The intensity at the wavelength of maximum emission was found to oscillate with foil thickness as predicted theoretically [Phys. Rev. 126, 1935 (1962)]. The peak in the spectrum was observed to

increase in intensity and also to be shifted to shorter wavelengths when the temperature of the foil was decreased. The observed shift in wavelength was much greater than the value predicted for a pure plasma oscillation on the basis of a change in the electronic density alone. The additional shift may be due to the change in the Fermi distribution of the electrons that participate in the interband transition. The emission from silver thus appears to arise from a hybrid type of resonance, with the pure plasma resonance being shifted in energy by the interband transition [Phys. Rev. 128, 1622 (1962)].

Radiation Physics: Detection of Plasma Radiation from Electron-Bombarded Aluminum Foils. — The detection of monochromatic photons from the decay of plasma oscillations in electron-bombarded metal foils has been the subject of intense investigation for several years [Phys. Rev. 111, 1214 (1958); 126, 1947 (1962)]. A peak in the spectrum from silver at 3300 A has been observed previously, but the interpretation is complicated by interband effects. The band structure of aluminum has been shown to be nearly free-electron-like [Phys. Rev. 124, 1797 (1961)] and should thus provide a better test of the plasma oscillation theory. Self-supported aluminum foils 600 to 800 A thick were bombarded with 60- to 100-kev electrons. The light emitted at 30° from the foil normal was analyzed with a Seya-Namioka vacuum ultraviolet spectrometer. A sharp peak at 810 ± 20 A was found in the spectrum from aluminum, in good agreement with the results of characteristic energy loss experiments. The line shape and absolute yield agree well with the predictions of transition-radiation theory.

ISOTOPE DEVELOPMENT

Recovery of Fission Products. — Preliminary results were favorable in using ozone for oxidizing cerium before its extraction with di(2-ethylhexyl)phosphoric acid to obtain separation from trivalent rare earths. Nearly complete oxidation of 0.05 M $Ce(NO_3)_3$ in 6 M HNO_3 was obtained in 30 min at 60°C.

Radioisotope Research and Development. — The CDC 1604A computer was used to find the optimum conditions for producing P^{33} by the (n,p) reaction on sulfur enriched in S^{33}. On the basis of a flux of 2×10^{14} neutrons cm^{-2} sec^{-1} and an irradiation time of 1 year, the conditions necessary for producing 100 μc of P^{33} containing less than 5% P^{32} are as follows:

Target	Weight of Target (mg)	Time of Aging After Irradiation (days)
Natural sulfur	33×10^3	350
7% S^{33}	221	250
22% S^{33}	17.7	196
70% S^{33}	0.35	98
90% S^{33}	0.035	28

Radioactive glass beads about 0.1 mm in diameter were prepared by adsorbing various cations on 210 to 230 mesh Decalso zeolite, heating to 900°C for 2 hr to form glass beads, and irradiating these beads in a flux of about 1.6×10^{14} neutrons cm^{-2} sec^{-1} for 1 month. Activities of 0.1 to 5 curies/g were obtained from glasses doped with Sc, Eu, Zn, Ag, Sb, Cs, and Co. Leach tests were performed with water at room temperature, 1 M NaOH at 70°C, and 0.5 M NaOH at room temperature; europium and scandium were most resistant to leaching (1×10^{-5}% for water).

A Y^{87}-Sr^{87m} generator has been developed which yields Sr^{87m} activity containing no detectable Y^{87} activity (the sensitivity of the detection of Y^{87} is 1 part in 2×10^{8}). The Y^{87} activity was produced by the $Sr^{87}(p,n)Y^{87}$ reaction. The irradiated target and nickel foil wrap were dissolved in hydrochloric acid, and the solution was then adjusted to a pH of about 9. The Y^{87} radiocolloid was separated by filtration onto a medium-porosity frit, removed from the frit with hydrochloric acid, evaporated to dryness, taken up with 0.01 M ammonium carbonate solution, and transferred to a small BioRad AG-1 (Dowex 1) column. The first washing with 0.01 M ammonium carbonate showed traces of Y^{87}. Further washing over several half-lives of 80-hr Y^{87} removed only Sr^{87m} with no detectable Y^{87} contamination. The Sr^{87m} product was allowed to decay through about 20 half-lives; the sample demonstrated no other detectable activities.

Radioisotope and Stable Isotope Production. — Ion currents equivalent to 3916 g of separated isotopes were monitored during January and February. Isotopes of Ca, Si, S, Ni, Ba, Rb, and Pb were separated and collected by using 28 of the calutrons associated with the program.

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