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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

JUNE 1965

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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

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Oak Ridge, Tennessee
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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

JUNE 1965

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

REACTOR DEVELOPMENT

GAS-COOLED REACTOR PROGRAM

EGCR Compressor Seal Design. — Tests of a modified atmospheric seal bushing for the EGCR vessel cooling compressors were successfully completed. The spillover of water into the bearing lubrication system was stopped by placing a slinger ring on the shaft immediately outboard of the atmospheric seal and by redesigning the labyrinth oil seal to incorporate a wind-back spiral. These changes permit the maximum pressure across the atmospheric bushing to be increased from 3-1/2 to 26 psi without spillover of water into the lubrication oil system. The normal operating pressure across the atmospheric bushing will be 10 psi.

GCR-ORR Loop No. 1. — Experimental fuel assembly 15 was completed and is ready for installation in loop 1 during the July 5 ORR intercycle. This assembly is similar to that irradiated in experiment 14; it consists of two fueled regions, one of which is vented to the loop coolant. The upper capsule contains a blend of three types of pyrolytic-carbon-coated particles — $(Th,U)C_2$, $(Th,U)O_2$, and ThO_2 — and will operate at a 2500°F central fuel temperature. The fuel region is vented to the coolant to permit measurement of the rate of release of noble gases during operation. The fuel particles are in two sizes to provide information on the segregation of mixed loose beds of particles during irradiation.

The lower capsule contains fuel in two small holes in a graphite cylinder. One hole contains a mixture of loose coated particles of two types; the other is filled with a bonded matrix of the same types of particles. This capsule is isolated from the loop coolant.

A bypass coolant cleanup system is being installed in loop 1. The principal components are: a copper oxide bed for oxidation of H_2 and CO, a molecular sieve for removal of H_2O and CO_2, a water-cooled heat exchanger, and a flowmeter. The addition of this cleanup system will permit the control of impurity levels for closer simulation of the coolant composition expected in high-temperature gas-cooled reactors.

LITR Capsule Irradiations. — Capsules L-CP9 and L-CP10, each of which contains pyrolytic-carbon-coated UC_2 particles in four separate compartments of ATJ graphite, continue to operate satisfactorily. The estimated

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burnup in capsule L-CP9 reached 19.5 at. % of the uranium, and the remaining operable fuel thermocouple indicates a central fuel temperature of 1810°F. Capsule L-CP10 attained an estimated burnup of 8 at. % of the uranium, and the indicated central fuel temperatures are 1490 and 1750°F.

ORR Poolside Irradiations. — Capsule 04-8 containing EGCR-vendor fuel material reached an estimated burnup of 5110 Mwd per metric ton of UO_2 and is continuing to operate satisfactorily. Maximum measured cladding temperature is 1548°F.

BeO-Graphite Compatibility Experiment. — Irradiation of the Mark II experimental assembly for studying BeO-graphite compatibility is continuing satisfactorily at 1500°C. However, the argon concentration required in the sweep gas to maintain this temperature has increased. This may be due to degradation of the heat-reflective baffles that would increase the radiant heat transfer within the capsule.

Bench tests were made on an infrared analyzer (Analytic Systems Company, model 709AX) for use in measuring the CO content of the sweep gas. The tests indicate that this analyzer should be satisfactory, and it is scheduled to be installed in the Mark II assembly during the July 5 ORR intercycle.

Fission-Gas Release from Pyrolytic-Carbon-Coated Fuel Particles. — In the ORR-B9-type sweep gas experiment the fission-gas release from coated particles is monitored continuously under controlled temperatures and burnups. In this way we can determine the conditions under which the coatings fail. Among the more recent experiments, several batches of particles have operated to burnups as high as 20% of the heavy metal and temperatures as high as 1400°C without any coating failures. From these experiments we have arrived at the following important conclusions regarding the fission-gas release:

1. The xenon diffuses through the coating at a slower rate than the krypton.
2. Essentially none of the fission gas generated in the fuel kernel escapes from the coated particle.
3. The fission gas observed comes from fuel contamination in the pyrolytic carbon coating. A correlation of fission-gas release rate (expressed as R/B, the ratio of release rate to birth rate) with coating contamination is shown below:

Capsule	Fuel	Coating Contamination Based on Alpha Count (μg of U)	10^6 R/B for ^{88}Kr at 1400°C, First Week of Irradiation
B9-22	OR-245	310	11
A9-2	OR-298	27	0.1
C1-16	OR-343	15	0.05
B9-26	OR-354	230	5
C1-15	GA-314	850	25
B9-25	GA-315	270	7

REACTOR FUELS AND MATERIALS

Materials Compatibility Studies. — The diffusivities of oxygen in polycrystalline niobium at 600 and 815°C were determined in connection with studies to determine the effect of oxygen on the corrosion of unalloyed niobium in liquid potassium. These corrosion studies had shown that oxygen diffuses from the niobium to form complex corrosion products at the niobium-potassium interface. The reaction kinetics were such that the surface oxygen concentration was reproducible and constant. By analyzing the oxygen depletion in the niobium as a function of time and applying the appropriate solution to Fick's law, we found the diffusion coefficients to be 7.9×10^{-10} and 6.4×10^{-9} cm^2/sec at 600 and 815°C respectively. These values are slightly smaller than, but in reasonable agreement with, recent literature values in which diffusivities were calculated from the rate at which oxygen gas at 760 torrs penetrated the niobium.

Fuel Element Development. — Previously, tungsten-rhenium alloy tubing deposited by hydrogen reduction of WF_6 and ReF_6 in the temperature range 450 to 700°C varied axially in composition and deposition rate due to a difference in the stabilities of the halides. A 1-in.-long moving hot zone provided by an induction coil has replaced the 12-in.-long static hot zone used in the initial studies. The composition of alloy deposited by this technique depends upon the composition of reacting gases, and the deposition rate depends upon temperature, mass feed rate, and rate of hot-zone movement. Initial moving-hot-zone experiments were conducted under the following conditions: deposition temperature, 700 to 750°C; gas preheat temperature, 300°C; hot-zone movement, 0.75 in./hr; pressure, 10 torrs; H_2 flow, 250 and 500 cm^3/min; WF_6 flow, 15 and 30 cm^3/min; ReF_6 flow, 5 and 10 cm^3/min. Under these conditions, deposition rates were about 5 mils/hr, and the efficiency of halide reduction was 100%. A typical deposit had an average rhenium content of 24% and a constant deposition rate of 4.3 mils/hr over the middle 4 in. of a 6-in.-long deposit. Thus, the technique shows promise for depositing alloys of uniform composition and thickness. Rhenium contents and deposition rates were lower on the ends of the deposit.

Behavior of High-Temperature Materials Under Irradiation. — Two low-temperature (130 to 420°C) assemblies (experiments 41-23 and 41-24) containing carbides of Ti, Zr, Nb, Ta, and W; translucent Al_2O_3 (Lucalox); and Y_2O_3 are now being irradiated in the ETR. Specimens of the same materials irradiated in the ORR are now undergoing postirradiation examinations. Assemblies designed for irradiation of the same types of specimens at high temperatures (1000 to 1400°C) are being prepared for insertion in the ETR early in FY 66.

FUNDAMENTAL HEAT TRANSFER AND FLUID DYNAMICS

Liquid Alkali Metal Boiling Studies: Superheat Phenomena. — A bubble-growth-rate calculation applicable to vapor bubbles rising in the Stokes-flow regime through a liquid pool at saturation temperature was completed.

The results were developed in nondimensional form and hence are applicable to pool boiling of any liquid metal; detailed attention is being given to the specific case of potassium pool boiling.

It is concluded that growth of the bubble is initiated by the Stokes' rise disturbing the equilibrium, under the assumptions that the bubble growth is limited solely by the mass inertia of the displaced liquid and that the liquid temperature remains constant. A single parameter is required to specify both the superheat conditions and the bubble rise velocity. Numerical evaluation of the general expression in dimensionless form shows that three stages of growth may be distinguished: (1) an initial dormant period in which the equilibrium situation is gradually being disturbed, (2) a period of rapid increase in growth rate, and (3) a final phase in which the increase in growth rate is slowed by the liquid mass inertia. The dormant time and the rate of increase of the growth rate both increase with diminishing initial bubble size.

Superheats required for pool boiling of potassium were obtained for a variety of initial bubble sizes, overpressures, and pool depths. The degree of superheat was found to increase with decreasing initial bubble size and decreasing overpressure; the pool depth had little influence. The potassium vapor bubble was found to leave the Stokes' rise regime within the first millisecond, after which a constant value of 25 cm/sec was assumed for the rise velocity. Except for the largest assumed initial bubble size, the growth rate, $\dot{R}_\infty$, reached a constant value that depended primarily on the initial bubble size, R_0, which is given by the following empirical formula:

$$\dot{R}_\infty = 0.879 R_0^{-0.721} .$$

General Boiling Studies: Light-Activated Switch Tests. — Several tests were conducted with the new G-E L9 light-activated switch (LAS) coupled to a radiant source through American Optical Company fiber optic light guides in order to evaluate the switch as a burnout protection device. The experimental arrangement consisted of a 1/8-in.-OD "A" nickel heater enclosed concentrically by a vertical Lavite shroud tube.

With the LAS positioned to view the internal heater through a hole in the shroud at a separation of 3/4 in., switch operation was effected at a temperature of 1150 ± 21°F. When a straight AO type IC-1-12 image conduit (1/8-in. hexagon, 12 in. long) was inserted between the LAS and the heat source so that one end was 1/8 in. from the heater surface and the other end was flush with the window of the L9 switch, the average switching temperature was 1242 ± 45°F. An identical test with a straight AO type CR-1-12 light-transmitting clad rod (1/2-in. round, 12 in. long) gave an average switching temperature of 1265 ± 11°F. Thus, with the light guides, it was possible to increase the separation between switch and heater by a factor of about 16 while taking a penalty of an increase in the heater temperature of only ~100°F before switch operation. A final test to study the effect on switch operation through a curved light path was made with an AO type LGM-3 fiber optic bundle (1/8-in. round, 12 in. long); switching in this case occurred at the appreciably higher temperature of 1435 ± 32°F.

HFIR Flow Reversal Studies. — A potentially critical condition exists for the HFIR during the period of core removal, after the completion

of an operating cycle, when the core coolant flow changes from downward forced convection to upward natural circulation. In this "turn-around" period, there is some concern that the fuel-element cladding temperatures may become excessive. A brief investigation of this situation was therefore initiated.

As a preliminary to such tests, natural-circulation heat transfer with an aluminum flat-plate test section used in earlier HFIR studies was reexamined. The test channel was 0.498 in. in internal width and had a 16.9-in. heated length when installed submerged in a drum containing water; the coolant gap was 0.050 in. Experiments conducted to date with the water pool at a temperature of 38 to 43°C have shown that incipient local boiling occurred at a heat flux of $\sim 10^4$ Btu hr^{-1} ft^{-2}, that the critical heat flux was 6.48×10^4 Btu hr^{-1} ft^{-2} (in comparison with values of 6.15×10^4 and 7.06×10^4 found in the original studies with this test section), and that the maximum wall temperature attained and the equilibrium wall-temperature distribution were the same for both ramp and step power changes at heat fluxes to at least 3.2×10^4 Btu hr^{-1} ft^{-2}.

Initial tests for studying the temperature history during flow reversal were performed by raising a hose above the submerged test section at various rates until flow reversal occurred. In no case was a temperature overshoot above the normal equilibrium level observed. Quantitative tests are planned for which a design will be used that will allow the downflow rate to be reduced according to a calculated schedule characterizing the situation in the HFIR. At all times, an open reverse-flow loop will be available to accommodate the change in flow direction following establishment of natural circulation.

POWER REACTOR FUEL PROCESSING

Close-Coupled Process Development: Evaluation of Simple Contactors for Solvent Extraction of Uranium and Thorium. — Simple, remotely operable methods are being investigated for removing at least 90% of the neutron poisons during the processing of irradiated reactor fuel. The decontamination step must also ensure that impurities which would adversely affect the refabrication procedures do not build up during multiple cycles. Simple operation and satisfactory decontamination have been obtained in laboratory experiments by extracting thorium and uranium from nitrate solutions with tributyl phosphate (TBP), using a differential contactor or unpacked spray column.

A comparison was made of the coextraction of uranium and thorium from a solution containing 375 g of thorium and 30 g of uranium per liter. The solution was 0.1 N acid deficient. The extractant was either 30 or 42% TBP in n-dodecane, and a differential contactor was used. With 30% TBP, more than 99.9% of the uranium and 48.5% of the thorium were extracted in 3.3 volumes of solvent. The factor for decontamination of the uranium from gross gamma activity was 1400. Continuing the extraction until 9.8 volumes of solvent were used resulted in the extraction of 99.2% of the thorium, with a factor for decontamination from gross gamma activity of 68. With 42% TBP, more than 99.9% of the uranium and 44% of the thorium were extracted in 2.2 volumes of solvent. The factor for

decontamination of the uranium from gross gamma activity was 600. Continuing the extraction until 7.6 volumes of solvent were used resulted in the extraction of 99.4% of the thorium, with a gross gamma decontamination factor of 58. Either concentration of solvent would, therefore, result in satisfactory recovery of the uranium and thorium. The use of 42% TBP resulted in a more rapid extraction but a slightly lower decontamination factor than with 30% TBP. A second organic phase was formed during the initial part of both experiments but did not interfere with satisfactory operation.

In unpacked spray columns, the solvent was passed up the column and overflowed at the top of the aqueous layer when 30% TBP in n-dodecane was used. With 30% TBP in CCl_4, the solvent was passed down through the aqueous phase and drained off at the bottom of the column. A second organic phase is not formed when CCl_4 is used as the diluent for the TBP, but CCl_4 would probably not be acceptable for fuel processing, because under irradiation it produces hydrochloric acid, which accelerates the corrosion of stainless steel equipment.

Close-Coupled Process Development: Use of Inorganic Ion Exchangers to Decontaminate Uranyl Nitrate and Thorium Nitrate Solutions. — Inorganic ion exchangers were tested for decontaminating uranium and thorium nitrate solutions. Work with nitric acid solutions spiked with mixed fission products showed that the highest fission product distribution coefficients were obtained in 0.1 M HNO_3 with zirconium phosphate, MnO_2, and Decalso. Distribution coefficients, based on gross gamma activity, were 53, 55, and 114, respectively, in 24-hr equilibrations at room temperature. Similar experiments with a 0.73 M $UO_2(NO_3)_2$–0.29 M $Th(NO_3)_4$–0.1 M HNO_3 solution containing mixed fission products gave fission product distribution coefficients of only 2, 12, and 2.5 respectively.

Fission product scavenging with MnO_2 was also studied briefly. Solutions of 0.73 M $UO_2(NO_3)_2$–0.29 M $Th(NO_3)_4$–0.1 M HNO_3–0.05 M $KMnO_4$, containing mixed fission products, were heated nearly to boiling, and MnO_2 was precipitated by adding a slight excess of 0.5 M $Cr(NO_3)_3$ solution. Fission product decontamination factors (based on gross gamma analyses) were only 2 to 5.

THORIUM UTILIZATION PROGRAM

Sol-Gel Microsphere Preparation. — We continued process development studies for preparing thoria microspheres by dispersion of a sol into 2-ethylhexanol, extraction of water to give gel microspheres, and drying and calcination to give high-density oxide microspheres. The present emphasis is on studies of large-capacity sol dispersers and solvent recycle to permit high throughput and long-term operation.

A "shear-dispersion nozzle," in which sol flowing through 0.010-in.-diam holes in a central tube was sheared off as drops by solvent flowing in an annulus, was made and tested. The advantages of this shear-dispersion nozzle are that it has fewer variables to control and therefore is simpler to operate than the rotary dispersers currently under study. Also, it has many orifices and thus a higher capacity than the two-fluid nozzle used in most of our development work. The initial test with this

new nozzle resulted in 52 wt % of the calcined microspheres being in the 45 to 50 mesh size (297 to 350 μ in diameter).

Process Demonstration and ^{233}U Distribution. — After the processing of an 18-kg shipment to ORNL last month, the process vessels were exhaustively flushed, and more than 98% of the ^{233}U was accounted for.

The solution carrier, containing 7700 g of uranium, was returned during the month. The isotopic composition of the uranium, as analyzed by the shipper, showed 91.21% ^{233}U and 5.68% ^{235}U, compared with 97.14% and 0.18% when originally shipped from ORNL. In addition to the above recycle material, 11 "bird cages" containing an estimated 5950 g of ^{233}U as slag ($MgO + U_3O_8$) and metal were received during the week.

One shipment of 25.2 g of U_3O_8 (20.05 g of ^{233}U) was repackaged from the existing stored oxide and shipped to Switzerland.

Installation of the solution storage system by the contractor is 95% complete. ORNL forces are scheduled to finish the tie-in piping by July 1.

Fabrication of the intermediate-scale solvent extraction facility (1 kg of ^{233}U per day) and equipment calibration were completed.

Sintering and Grain Growth of Sol-Gel Thoria and Thoria-Urania. — Differential thermal analysis (DTA) of crushed thoria gel yielded a preliminary value for the average surface energy of the thoria over the temperature range 700 to 1200°C. With the BET-determined surface area of the gel, and a cell calibration determined from the water-loss endotherm at 100°C, values of 840 and 890 ergs/cm^2 were calculated from two successive runs. These results must be revised to allow for edge and corner effects at the very small crystallite sizes involved, and to allow for variation in cell calibration with temperature. This latter effect is being investigated.

The DTA apparatus is yielding such well-defined and reproducible surface-energy release peaks that peak profile analysis seemed to be justified. A review of DTA theory indicated that, under the present conditions where the reaction rate is low, the temperature gradient across the sample is low, and the measured differential temperature is small, reaction rate as a function of temperature might be extracted from profile analysis of the DTA peak. This conclusion has to be tested by use of well-characterized reactions. However, the profiles of surface-energy release peaks obtained so far could be readily interpreted in terms of first-order reactions with measured activation energies, in four separate runs, of 22, 25, 23, and 27 kcal/mole.

Fueled-Graphite Development: Fluidizing Studies in Transparent Models at Room Temperature. — The gas flows required for fluidizing particles in coating processes are very sensitive to changes of particle density, particle diameter, and temperature. Since these parameters vary considerably during the coating operation, it is difficult to adjust the coating process for maximum efficiency and uniformity. On the other hand, our calculations indicate that the velocity required for entraining particles in a stream of gas should have much less dependence on particle diameter and density than on the fluidizing velocity, and should be practically independent of temperature. This could be the basis of spouted-bed, teeter-bed, or conveyor-bed processes that would be much more easily

controlled than the usual fluidized-bed process. These possibilities are being tested by calculations, literature search, and experiments in glass models.

The 5-in.-ID entrained-bed Plexiglas model was assembled, and several spot-check experiments were made. No major differences between entrainment characteristics were found for the 3-1/2- and 5-in.-ID models when nozzle diameter, distance between nozzle and insert, and shape of the insert were the same. The carryover capacity decreases only slightly with increasing insert height (from 18 cm to 31 cm). With increasing distance between nozzle and insert, the carryover capacity and carrying capacity per unit volumes of gas increase considerably. The pressure difference ΔP across the gap between insert and bottom cone was found to be a function of nozzle diameter, distance between nozzle and insert, shape of the insert, particle size, and gas flow. For gas flows only slightly larger than those of minimum entrainment, ΔP is positive, and some gas passes through the packed bed between the insert and the column wall. At high gas flows, ΔP is negative, and all the gas flows through the insert. Tapering both ends of the insert lowers slightly the carryover capacity for a given set of other variables, but $\Delta P = 0$ is reached at considerably lower gas flows. Up to 50% of the total gas may pass through the packed part of the bed (independently of the packed-bed height above the lower edge of the insert) at the maximum positive ΔP for a given set of conditions. This would be undesirable in a coating process, because agglomeration of particles can be expected. Therefore, this differential pressure is considered to be a useful indicator in the external control of the state of fluidization and entrainment in this type of coating device.

Fueled-Graphite Development: Rotating-Drum Model Studies. — Studies of the mixing of microspheres in a 9-in.-ID cylindrical Plexiglas model of a rotating drum have continued. Originally, ~100-g samples were drawn by suction from various locations in the charge. For example, up to 20% of the charge was removed for sampling in the lower drum-loading range (loading to less than 30% of the drum radius). To reduce the depletion in charge and hence the error in true local composition of the mix with rotating time, the influence of sample size (50, 20, and 10 g per sample) and location on sample composition was studied. Preliminary conclusions are that smaller particles (417- to 500-μ-diam iron shot) are more readily entrained by the sampling method than bigger ones (590 to 833 μ). This was shown by drawing consecutive 10- and 20-g samples from the same location. Therefore, the tip of the suction tube has been fitted with a cap with radial openings to entrain particles preferentially from a horizontal plane and reduce vertical lifting at the point of entrainment.

Conversion of ThO_2 to ThC_2. — Studies were continued to establish time-temperature relationships for converting 300- to 400-μ sol-gel ThO_2 microspheres to ThC_2 in the single-fin crucible. It was expected that the conversion would be complete after 6 hr at 2050°C, since conversion was almost complete in 5 hr in a previous run made under identical conditions except that different batches of oxide particles were used. Failure of the conversion to go to completion is attributed to intrinsic differences in the oxides used. The relation of oxide particle properties to conversion will be studied further.

Conversion of sol-gel microspheres of ThO_2–25% UO_2 to $(Th,U)C_2$ was assumed to be complete after 2 hr at 2150°C, but this has not yet been confirmed by x-ray analysis. The assumption that reaction was complete was made on the premise that the conversion of $(Th,U)O_2$ particles proceeds in the same manner as the conversion of ThO_2 particles, where the conversion proceeds toward the center of the particle and where any unconverted oxide is at or near the center. However, some of the converted $(Th,U)O_2$ particles contained numerous small spots that appeared to represent a different phase, voids, or both. This phenomenon has not been observed in studies of the conversion of ThO_2 particles. The $(Th,U)O_2$ sol-gel particles appeared to be very dense metallographically but showed considerable variation in structure from particle to particle. These variations may be related to the variations in appearance of the converted particles, but no direct correlation can be attempted until further conversion studies are made.

Irradiation of Thorium Fuels. — Metallographic examinations of the fourth rod from the high-heat-rated ETR-1 group have not yet been completed. Macroscopic examination of longitudinal sections from each end of the fuel column in this rod shows a distinctly different void termination. The lower end is in contact with the stainless steel end plug, and the void terminates by gradually reducing in diameter. The upper end had the void packed with Fiberfrax, an Al_2O_3-SiO_2-ZrO_2 fibrous insulating material. Here the void ends in a dome-shaped fuel layer, with evidence of incipient melting of the adjacent insulating material. The insulation has a reported melting point of about 1760°C; so, with the microstructural examinations, we may be able to estimate the maximum temperatures in this area. One unusual crystal, attached to the inside surface of the void near the top of the rod, but standing in relief, was a water-white, well-formed octahedron. Since such crystals can be grown by vapor deposition of ThO_2, an attempt will be made to recover this crystal along with similar, less well formed, ones for possible identification by electron microscopy or emission spectroscopy.

Chemical analysis of the second burnup samples from the ETR-1 group of rods is still not complete, because of an unidentified residue left after dissolving the sample. Since the dissolution involved protracted refluxing of the fluoride-containing solutions in glassware, the precipitate may be silica. The precipitate will be analyzed. Meanwhile, the original isotopic analyses for uranium have been used with a newly developed computer program to provide what are considered more reliable burnup figures, which agree to within 10% of the anticipated burnup of 22,000 Mwd per metric ton of U + Th, based upon the exposure history.

All capsules currently under irradiation are continuing to function without incident.

Thorium-Uranium Recycle Facility. — Construction of the building started during the past month. The fixed-price contractor has established most of his subcontracts. Work so far has concentrated on the installation of underground services to tie into existing facilities, and excavation for the below-grade work. Some of the underground ductwork, purchased by ORNL, has been turned over to the contractor, and most of the below-grade stainless steel embedments are on hand. At the request of the contractor, ORNL is expediting the delivery of quality-H stainless

steel fittings ahead of schedule. The date for the receipt of bids on the bulk shielding window has been extended at the request of the bidders.

As to TURF process equipment, the sub-base assembly for the vibratory compactor and the fuel blend-transfer device was completed and was installed in the mockup area. The mechanical portion of the vibratory compaction unit was completed and shipped by the outside fabricator; installation of the unit in the mockup area is now beginning. Completion of the mechanical portion of the blend-transfer device has been delayed because of late delivery of several key purchased components.

With respect to sol-gel process equipment, work was continued on the design of in-cell equipment layouts, equipment enclosure frames, interframe piping, and intraframe process piping for both the uranium storage-metering system and the sol blending-metering system. Work was started on the design of a circular sol dryer and a device for mockup testing of a ball-and-socket type of mounting and anchoring system for in-cell equipment frames.

MOLTEN-SALT REACTOR PROGRAM

Molten-Salt Reactor Experiment. — Criticality was attained for the first time on June 1, with a ^{235}U loading very close to the value predicted by multigroup diffusion theory calculations. About 99% of the ^{235}U was added in four major batches, 44.2, 14.9, 5.1, and 4.3 kg. These additions were made to the salt in a drain tank, and after each addition the core was filled and the neutron multiplication was determined. Finally, eight capsules of enriching salt (85 g of ^{235}U each) were added through the sampler-enricher, bringing the total in the salt to 69.4 kg of ^{235}U (61.4 kg in the circulating loop, the remainder in the drain tank). The reactor was critical with this loading at 1191°F with fuel circulation stopped, two rods fully withdrawn, and the third rod 95% withdrawn. The ^{235}U concentration was only 1.2% above the value predicted for these conditions by calculations using the MODRIC diffusion code with GAM-2 cross sections.

After initial criticality, more ^{235}U was added in 85-g increments through the sampler-enricher to produce the concentration required for operation at power with fission products in the salt and with control rods partly inserted. Control rod sensitivity over the entire range of one rod is being measured during these additions. Also measured are the ^{235}U concentration coefficient of reactivity, temperature coefficient, pressure coefficient, and reactivity effects of circulation. Seventy-three capsules of enriching salt have been added; about 17 more will be required. Preliminary results indicate that the rod worth, concentration coefficient, and temperature coefficient are close to the predicted values. The effect of circulation is 0.2 to 0.3% $\delta k/k$, somewhat less than expected. There was no measurable effect of pressure.

Aside from the planned experiments, the operation has been practically uneventful. A few minor difficulties were encountered in the operation of the sampler-enricher, and the manipulator boot was ruptured once by misoperation of valves. Steps have been taken to prevent recurrence of these troubles.

Acceptance of the on-line digital computer was delayed by difficulties with input amplifiers. The program runs without apparent problems.

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Development of Components and Systems. — The fuel sampler-enricher has been used to isolate 44 fuel salt samples and to make 73 uranium enrichments during the critical and zero-power experiments. The final testing of the equipment and procedures is also being conducted during this period. The shielding is about 75% complete and will be finished after the zero-power tests.

The control rods and drives were installed in the reactor and have been in operation since the start of the critical experiment. During drop tests shortly after the rods were installed, an unnecessarily tight mechanical fit between the rod-drive limit switch actuators and their guides caused the switches to malfunction after a scram. The clearances were adjusted, and there have been no further difficulties with the drives. Measurements made during three weeks of operation indicate that the rods have changed less than 1/16 in. in length, which is acceptable.

Routine operation of the freeze valves continues to be complicated by a poor temperature distribution across the valve and mechanical difficulties with the electrical leads. Modifications to the heater control circuits will be made so that a more uniform heat distribution can be obtained. The manufacturer has made modifications to the heater elements, and a prototype will be tested. A temperature controller was installed in the cooling air system for the reactor drain valve and appears to be operating quite successfully. This type of control will be installed on the other operational valves.

MSRE Pump Program. — Operation of the spare rotary element for the MSRE fuel pump with molten salt was continued to conduct back-diffusion tests with the radial clearance between the shaft and the shield block increased to 0.005 in. Preliminary results indicate that ^{85}Kr does not diffuse into the seal region even when the down-the-shaft purge rate is as low as 300 liters/day.

A mockup of the oil reservoir used in the MSRE lubrication system was tested to investigate the entrainment of gas in the oil and the difficulty in priming the standby pump. The tests revealed that operation of a jet pump, which sparges oil from the bearing housing, contributed greatly to the entrainment of gas in the oil. The condition disappeared after the jet pump was removed from the system, and the standby pump primed satisfactorily. The piping is being modified to the configuration of the oil return line for the MSRE fuel pump, and the tests will be repeated to determine whether to remove the jet pump from the MSRE lubrication system.

The lower bearing in the drive motor used in the PKP molten-salt pump test failed. Insufficient lubrication is suspected. The bearing was replaced and the test will be resumed. The test pump having a molten-salt-lubricated journal bearing was assembled and was installed in the hot test facility. It will be placed in operation in the near future.

Instrumentation Development. — Installation of the ultrasonic level probe in the MSRE fuel storage tank was completed. Except for some problems associated with drift of oscillator frequency, the system performed well during initial operation of the MSRE chemical processing system and was a valuable aid to operations. The oscillator frequency drift is objectionable, since this frequency must be maintained at the resonant frequency of the probe system to obtain satisfactory level indication. An effort will be made to stabilize the oscillation and eliminate the drift.

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Miscellaneous activities related to providing assistance to MSRE operations, improving performance of MSRE instrumentation, and completing development work are continuing.

NUCLEAR SAFETY

Retention of Methyl Iodide by Charcoal Beds Loaded with Water. — On the basis of experiments recently completed, it was concluded that: (1) Coconut charcoal, either untreated or impregnated with 5% morpholine, extracted methyl iodide from water-saturated air at about 95°C better than did the coal-based charcoal (Pittsburgh SGL). (2) After methyl iodide is fixed on coconut charcoal, it is strongly held and cannot be readily removed by exposure to water-saturated air streams at temperatures up to 95°C. (3) Hydrolysis of methyl iodide does not occur to a significant extent in steam-air mixtures at 120°C. (4) The retention of methyl iodide by a deep bed of coconut charcoal, even when it is nearly saturated with water, may be good enough to prevent escape of dangerous amounts of iodine activity in a reactor accident.

Nuclear Safety Information Center. — The state-of-the-art study, "Secondary Shutdown Systems of Nuclear Power Plants," has been completed and was issued in draft form for external review. The report will receive extensive dissemination as ORNL-NSIC-7 upon completion of the review. The Center's second computer-sorted bibliography has been completed and submitted for publication. It will be distributed in July as ORNL-NSIC-9.

NSIC recently issued a report (ORNL-TM-1136) summarizing the first two years of the Center's operating experience. The report, which describes the services that the Center offers, the manner in which it operates, and the problems that it has encountered, was distributed to all participants at an AEC information and data-center meeting in Washington, D.C., on June 3 and 4, 1965, where directors of all AEC centers met to discuss their operating modes and problem areas with representatives from AEC program divisions, DTI, and DTIE.

Computer programming for NSIC's selective dissemination of information (SDI) program has been completed, and shakedown of the program is commencing. Routine dissemination of information to a selected group of management-level personnel according to their interest profiles will be initiated soon.

During May and June, the Center received and answered 24 requests for information. In the same period the staff conferred with 23 visitors.

Nuclear Safety Pilot Plant. — Analysis of data from run 8 continued during the report period. Further examination of the gas sample data indicates that the activity collected in the backup charcoal traps principally resulted from bypassing of the May packs rather than breakthrough.

Fair material balances were obtained. In the case of iodine about 9.9% was released from the fuel and only 2.4% entered the model containment vessel (MCV). The only other fission products released in significant quantity were tellurium (2.3%) and cesium (6.9%); only a small fraction of the released material (0.4 and 0.2% respectively) was transported to the MCV.

Of the iodine found in the MCV, 50% was recovered in condensate, 42% was found in decontaminating solutions, and 8% was vented during purging of the MCV.

Run 9 was made on May 20, 1965. This comprised melting irradiated stainless-steel-clad UO_2 in a steam-hydrogen atmosphere. With a slightly modified plasma torch and a lower speed of feeding the fuel under the torch, very thorough melting was achieved. The decay of activity by natural means was observed for 24 hr, after which the MCV water spray was operated for 1 hr and observation was continued for an additional 5 hr. Results of the spraying procedure have not been evaluated.

Preliminary study of the May pack results from run 9 shows that the iodine distribution is similar to that in run 8, with the iodine being principally collected on charcoal paper and charcoal beds. Only part of the analyses has been reported.

RADIOACTIVE WASTE DISPOSAL

Disposal in Deep Wells. — On May 19, injection No. 6 was started using a mix containing Kingston fly ash as a substitute for some of the Portland cement. To control the proportioning of low-solids mixes with more consistency than was possible with the Densometer, a flowmeter was installed in the mixer hopper. During the injection, difficulty was experienced in maintaining flow of solids from the storage bins to the mixing hopper; the cause was found to be plugging of the valves by large clumps of solids. Difficulties with the flowmeter were also experienced; in this case, a plastic cone, about 12 in. in diameter, came loose and plugged the hopper. Plugging of the hopper resulted in leakage of grout out into the mixer cell. The cell was decontaminated by flushing the grout into the concrete waste pit and pumping the contaminated wash water into the fracture.

After approximately 64,000 gal of waste had been injected, a leak developed in the high-pressure piping in the wellhead cell. After the leak was located at a connection between the plug container and the connecting piping, the well, filled with grout, was cleared by pumping water down the annulus, up the central tubing, and out through the leak in the wellhead. The wellhead cell was decontaminated with water, and contaminated grout was flushed down to the emergency pit. The injection well was left open in order to determine the bleed-back characteristics of the grout. Bleed-back is expected if phase separation occurred in the mix. The results thus far suggest that phase separation did occur, and studies are under way to formulate a mix which will not undergo phase separation.

High-Activity Waste Calcination. — Low-melting ($\leq 750°C$) phosphate-glass compositions were developed for the incorporation of both sulfate-containing and sulfate-free wastes. Resulting glasses contained up to 35% waste oxides with sulfate waste and up to 40% with sulfate-free waste. Rates of cesium leaching corresponded to gross solution rates of about 10^{-5} g cm^{-2} day^{-1}, about the same as for commercial glasses. Loss of SO_3 from sulfate melts was $\leq 5.5\%$, as determined by thermogravimetric analysis.

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A thermal conductivity probe designed to determine the levels of solution, calcine, and glass in the melt pot was tested. The probe consists of thermocouples which were periodically either heated above or cooled below the temperature of the surroundings. The rate of recovery is sufficiently different for air, solution, dry calcine, and melt to distinguish between these phases. This probe will be tested further.

The ORNL Rising-Level Potglass Process was successfully tested at the Pacific Northwest Laboratory (Hanford). The equipment and materials differed only in minor details from those used in similar tests at ORNL. The volume of simulated waste solution after addition of additives was equivalent to 80 gal per ton of uranium processed. The glass product was equivalent to 6.4 gal per ton of uranium. Postrun examination revealed no large void spaces in the product and no excessive corrosion of the pot.

Disposal in Natural Salt Formations. — All drilling in experimental rooms 1, 3, and 4 is complete. Installation of upper liner sections in room 1 is finished, and work is in progress in room 4.

Shipment of most of the experimental equipment to Lyons has been completed; the necessary thermocouples and connecting cables have been delivered.

Based on a critical-path schedule of the remaining installation and checkout operations, it appears that Project Salt Vault can be initiated around November 1, 1965. This date will coincide with a shutdown in the reactor operating cycle at the ETR, making fuel elements available for the project.

Engineering, Economic, and Safety Evaluation. — The costs of permanent storage of reactor fuel processing wastes in both acid and alkaline form have been estimated for a 22,400-Mw nuclear power economy. The wastes arise from chemical dissolution of the fuel cladding and solvent extraction purification of a combination of uranium and thorium converter fuels. The economics were examined for three representative types of financing: government ownership, private ownership, and a combination of government and private ownership. In each case it was assumed that during a 20-year period of waste accumulation, a permanent tax-free fund is established of such size that the annual tax-free interest will be sufficient to provide for periodic replacement of tanks and for the annual operating expenses of the facility. Capital costs were estimated for each case, then used in a computer code to estimate total costs for each method of financing as a function of tank size, tank life, and fission product concentration in the waste.

Total costs of raffinate waste storage, in terms of the cost of electricity generated, were least for storage of acid wastes in tanks of 75-year life with government ownership, and greatest for storage of alkaline wastes in tanks of 25-year life with private ownership. For acid wastes, the optimum tank capacity is about a million gallons, and minimum total costs range from 0.0165 to 0.0272 mill/kwhr. For alkaline wastes the optimum tank capacity is 2.5 million gallons, and minimum total costs range from 0.0177 to 0.0294 mill/kwhr. Doubling the concentration of fission products in the wastes, as stored, reduced the total costs between 15 and 30%.

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Total costs for storing wastes ranged from 0.0215 to 0.0398 mill/kwhr. These costs were greater than those for raffinate storage because of the greater volumes of cladding wastes.

Incorporation of Intermediate-Level Radioactive Waste in Asphalt: Flowsheet Demonstration with Radioactive Materials. — Thirty liters of ORNL tank-farm intermediate-level waste, containing about 95 mc/gal, was evaporated to a twentieth of the original volume, and the ionic concentrations were adjusted to the flowsheet conditions for fixing such wastes in asphalt. This feed, average specific gravity 1.28, was spiked with 3-year-old mixed fission products to the 5-curie/gal radioactivity level, and six fixation experiments were made incorporating 300 ml of "waste" into 158 g of RS-2 (rapid-setting) emulsified asphalt. Maximum temperature of the final asphalt-waste mass was 185°C in one test and 165°C in the others.

The radioactivity of the condensate from the fixation, containing from 28 to 56 curies/gal, was due to entrained $^{134-137}Cs$; almost no fission products were found on the Millipore off-gas filter or in the caustic scrubber solution that followed the condensate catch vessel.

Leaching rates for sodium and gross-gamma emitters ($^{134-137}Cs$) for the first three weeks of static leaching in demineralized water were comparable to rates for tracer-level and nonradioactive samples (about 10^{-3} g cm^{-2} day^{-1}). No pressure increase was noted for one batch of asphalt-fission product waste that was sealed into a system connected to a pressure recorder.

Incorporation of Intermediate-Level Radioactive Waste in Asphalt: Laboratory Studies. — Nine commercial emulsified asphalts were tested to determine their suitability for use in the process for incorporating intermediate-level radioactive waste in asphalt. These asphalts were made from four different crude asphalt bases and included rapid-, medium-, and slow-setting types. Eight of the emulsifiers were anionic (containing a sodium-soap emulsifying agent), and one was cationic (probably containing an amine emulsifying agent).

Eight of these asphalts yielded satisfactory products. They were mixed at 200 rpm with intermediate-level waste, the water was evaporated, and the product was drained at 165°C. One medium-setting type foamed excessively when heated. The most satisfactory product from the standpoint of homogeneity, viscosity, and radiation stability was obtained from the rapid-setting type which used the cationic emulsifier. This product increased only slightly in volume when irradiated to an absorbed dose of 10^8 rads. All products previously tested had swelled only slightly at 10^7 rads but had increased in volume about 30% at 10^8 rads.

Numerous safety tests indicate that there is no appreciable difference between the rates of burning of the final product and of the asphalt base itself. These tests are attempts to initiate rapid exothermic reactions by externally heating the specimen and, at the same time, suddenly overheating it locally by an electrical resistance wire embedded in the specimen.

PHYSICAL RESEARCH

PHYSICS AND MATHEMATICS

High-Voltage Experimental Program: Conversion Coefficients from Coulomb Excitation and Lifetime Measurements. — Several papers have pointed out that information on total internal-conversion coefficients for E2 transitions can be obtained from the comparison of B(E2)'s found from lifetime measurements with those obtained from Coulomb excitation results (either inelastic-scattering cross sections or gamma-ray yields). From such a comparison of ten rare-earth nuclei, Fossan and Herskind [Nucl. Phys. 40, 24 (1963)] concluded that the experimental alphas differed from the theoretical alphas by an average of 12 to 14%. Bernstein [Phys. Rev. Letters 8, 100 (1962)] has suggested that the deviations from theory exhibit an N/Z dependence.

In the course of compiling a list of B(E2) values for nuclei, the problem of total E2 conversion coefficients has once again been studied, since it is desirable to use the large amount of precise information available from lifetime measurements to supplement the Coulomb excitation results. Taking all the available information on 23 even-even rare-earth nuclei, we conclude that the average deviation of the experimental and theoretical alphas is only about 1%. Sliv's theoretical values are used for α_K and α_L, and we take the contribution from higher shells to be $0.33\alpha_L$. A statistical analysis suggests that the observed fluctuations in the individual values of $\alpha_{exp}/\alpha_{theor}$ are what one would expect from the errors assigned to the experimental measurements.

High-Voltage Experimental Program: Anisotropic Energy Losses in a Face-Centered Cubic Crystal for High-Energy ^{79}Br and ^{127}I Ions. — Channeling effects on the energy losses of fast heavy ions in solids were investigated. Multicomponent beams of Br and I ions with energies from 20 to 80 Mev, obtained from the Oak Ridge tandem accelerator, were passed through thin (0.5 μ) gold single crystals which could be rotated about an axis in the (100) surface. When low-index crystal directions ([001], [110], and [112]) were aligned with the beam, a single energy loss group was observed which was significantly less than the value obtained for polycrystalline gold of the same thickness. The fractional loss varied from 50 to 85% of the polycrystalline loss, depending on the energy, the species of the bombarding particle, and the particular crystal direction. When the crystals were oriented with low-index planes parallel to the beam direction, two energy loss groups appeared, one having the normal loss and the other having a lower loss which could be correlated with channeling in the interplanar space. Both the fraction of the ions which were channeled and the angular acceptance aperture into the channel were found to decrease with increasing ion energy, which is in accord with expectations based on an active channeling mechanism. The fact that two distinct groups were observed, rather than a continuum of energy losses, implies that most of the channeled particles were focused almost immediately upon entering the crystal and remained in these channels during the remainder of their trajectories through the solid.

Nuclear Data. — Nuclear Data Sheets, Volume 6, Set 5, is in press. Set 6, which is ready for press, is the last set to be published by the National Academy of Sciences. Work continues on the Nuclear Data Sheets for A = 182 through 189, which will be published by Academic Press.

Medium-Energy Nuclear Physics: Experimental. — The analogs of the ground states of ^{88}Sr and ^{89}Y were observed in the reactions $^{89}Y(p,d)^{88}Y$ and $^{90}Zr(p,d)^{89}Zr$ respectively, as produced with 36-Mev protons from the ORIC. Although excitation of these states was predicted, they had not been observed in our earlier measurements because of insufficient energy in the 20-Mev runs and inadequate resolution in the earlier 36-Mev runs. Even in our latest runs the resolution is no better than 150 kev full width at half maximum, and the analog-state deuteron groups become lost in the background for angles larger than about 25°.

The polarization of 40-Mev protons elastically scattered from ^{59}Co, ^{120}Sn, and ^{68}Zn was measured. This brings to eight the total number of elements for which we have the differential cross section and the polarization. The differential scattering cross section and the polarization of protons from calcium were measured over the range 10 to 170° and at 35, 40, and 45 Mev in an attempt to understand the details of the behavior of the scattering amplitudes. The analysis of these results is not yet complete.

The elastic and the inelastic scattering of 65-Mev alpha particles from ^{90}Zr, ^{91}Zr, ^{92}Zr, ^{94}Zr, and ^{96}Zr were measured at angles from about 55 to 76°. A shift of the diffraction structure was evident for ^{90}Zr relative to the other isotopes.

Total cross section, range, and angular distribution studies were completed for the $^{10}B(^{14}N,^{13}N)^{11}B$ reaction for E_{lab} = 9 to 16 Mev. Below about E_{lab} = 12 Mev the data are well described by the tunneling theory of neutron transfer. Above this energy, nuclear absorption effects occur. Data from the $^{40}Ca(^{14}N,^{13}N)^{41}Ca$ reaction were obtained for E_{lab} = 19 to 27 Mev. The total cross section data do not follow the energy systematics obtained from other targets. Further studies are necessary to determine if other reaction mechanisms are involved or if target impurities may produce the new effects.

Preliminary data were taken for the $^{48}Ca(d,^{3}He)^{47}K$ reaction from 13 to 37°. The 150-$\mu g/cm^2$ ^{48}Ca target was obtained from Argonne National Laboratory. These data will be used to extract the mass of ^{47}K, to study the proton configurations present in the ground state of ^{48}Ca, and to locate low-lying excited states of ^{47}K. Since the ^{48}Ca target was partially oxidized, data from the $^{16}O(d,^{3}He)^{15}N$ reaction were also obtained. The positive parity doublet ($1/2^+$, $5/2^+$) at 5.3 Mev excitation in ^{15}N is observed in this reaction, indicating the probable presence of $(1s)^4$ $(1p)^{10}$ $(2s, 1d)^2$ configurations in the ^{16}O ground state.

Cyclotron Operations: ORIC. — During May and June the ORIC was scheduled for 1227 hr of operation, distributed about as follows: 78% to nuclear research bombardments, 11% to cyclotron beam and machine development, 7% to installation and modification of associated equipment, and 4% to scheduled shutdowns for routine maintenance. The beam was "on target" 61% of the 1227 hr, or about 13.4 hr/day. No major component failures were encountered. Extracted beams of alpha particles at 68 and

80 Mev, deuterons at 34 Mev, and protons at 36, 44, 48, and 60 Mev were routinely delivered to the experimenters. In a first attempt in a program to provide a usable ^{3}He beam, the ^{3}He ions were accelerated and deflected at about 51 Mev.

Electronuclear Systems: Advanced Acceleratory Studies. — A high-intensity proton accelerator, such as the separated-orbit cyclotron, is potentially capable of producing a neutron flux of great intensity. The Chalk River Laboratory (Canada) and the Rutherford Laboratory (England), as well as ORNL, are evaluating this possibility.

In collaboration with the Chalk River Laboratory, measurements were made at the Brookhaven Cosmotron to determine the yield of low-energy neutrons resulting from high-energy proton bombardment of thick targets. Six targets were studied: 4-in.-diam by 24-in.-long targets of Sn, Pb, and depleted U; an 8-in.-diam by 24-in.-long Pb target; a 24-in.-long truncated-cone Pb target 4 in. in diameter at one end and 8 in. in diameter at the other; and a rectangular Be target 4 × 4 × 24 in. All targets were bombarded with protons at energies of 0.5, 0.75, 1.0, and 1.5 Bev; additional measurements were made with the 4-in.-diam lead target at 2.0 Bev. Neutron yields were determined by measuring the spatial distribution of neutrons in a large water moderator surrounding the targets. Neutron flux measurements were made with copper foils, gold foils (both cadmium covered and uncovered), and with a vanadium β-current detector.

General features of the measurements are: a linear increase in neutron yield with increasing proton energy for all targets, and increasing neutron yield with increasing atomic weight of the target. A pronounced increase in yield in going from the lead target to the uranium target indicated the important contribution from fission.

Specific results for the 4-in.-diam lead target are:

1. The neutron yield per incident proton increases linearly with energy from 8 neutrons/proton at 0.5 Bev to 40 neutrons/proton at 2.0 Bev.

2. Above a proton energy of 1.0 Bev the beam energy required to release a low-energy neutron is 50 Mev.

3. At 1 Bev the 2200-m/sec neutron flux at the surface of the lead target is 1.6×10^{11} neutrons cm^{-2} sec^{-1} for a proton beam current of 1 μa.

Medium-Energy Electronuclear Machines: Electronuclear System Development. — In continuation of the beam development program for the ORIC, the lower limit of the energy range was extended downward from 36 Mev to 25 Mev for protons and from 68 Mev to 55 Mev for alpha particles. These lower limits will remain until new power supplies for the extraction system, now on hand, are installed and put in service. These extensions of energy ranges were achieved by using the valley coil system to correct the average field gradient near the extraction radius. Initial development on obtaining an ^{3}He beam resulted in accelerating ~51-Mev ^{3}He ions to the extraction radius and deflecting them to the extraction magnet. Further development will be scheduled before the extracted beam of ^{3}He will become available for experimental work.

The broad-range spectrometer was tested and accepted at the vendor's plant. Field uniformity of 1 part in 2000 was achieved over the usable portion of the magnet for most excitation levels. This system will accept

particles with radii of curvature between 30 and 63 in. and with rigidities up to that of 70-Mev tritons. The system was received and is being assembled for testing in the large experiment room. It is hoped that this system can be put in use in September.

CHEMISTRY

Instrumental Methods for Chemical Analysis: Potentiostatic DC Polarography Directly in Solvent Extracts. — The feasibility and utility of highly efficient and selective solvent extraction separations followed by a potentiostatic polarographic determination of the concentration of the extracted species directly in the organic extract have been demonstrated for several cations and extractants. A method involving a 1:1 volume extraction (no concentration relative to the aqueous sample) was developed for the determination of uranium; uranium is extracted from a 0.5 M HNO_3 solution with 0.1 M tri-n-octylphosphine oxide (TOPO) in cyclohexane, and an equal volume of 0.2 M $LiClO_4$ in ethanol is added to the extract to provide a supporting electrolyte (ORNL-3561, pp. 32–33).

Very reproducible first-derivative dc polarograms of 5×10^{-6} M uranium were obtained, so another advantage has subsequently been investigated: an increased concentration in the extract relative to that in the aqueous solution. With a 5-ml cell, a net 25-fold concentration advantage has been achieved by a single, quantitative 50:1 aqueous to organic volume ratio extraction step. For example, this permits 250 ml of aqueous samples containing as little as 0.01 μg of U(VI) per ml [$\sim 5 \times 10^{-8}$ M U(VI)] to be analyzed by this polarographic procedure. To obtain a larger concentration advantage, a 250-ml sample solution in 0.5 M HNO_3 that has been purified by reaction with sulfamic acid is extracted with 5 ml of 0.025 M TOPO in cyclohexane, 4 ml of the extract is evaporated in a graduated cone to 0.4 ml, 0.4 ml of ethanol 0.2 M in $LiClO_4$ is added, and 0.5 ml of this solution is transferred to a 0.5-ml cell. The net concentration advantage is 250-fold. Aqueous samples that are as low as $\sim 5 \times 10^{-9}$ M in U(VI) can be analyzed.

The precision of analysis of 0.6 μg (0.002 microequivalent) of U(VI) in the 0.5-ml cell is ~4%. A cursory determination of uranium in a seawater sample (~3 μg/liter) indicated that the method could be applied to this analysis.

Preparation and Properties of Actinide-Element Oxides. — Repeated electrophoresis and free diffusion of colloidal thoria in a Tiselius-type apparatus confirmed that a large difference exists between receding- and advancing-front velocities of a thoria sol 1 M in 140-A crystallites (measured by x-ray line broadening). The sol had electrophoretic mobilities of about 1.2×10^{-4} cm^2 v^{-1} sec^{-1} at the receding boundary, and 9.9×10^{-4} at the advancing boundary. The difference in mobilities is strongly concentration dependent and does not appear to extrapolate to zero at zero concentration. The boundaries, after the colloid had undergone electrophoresis, had apparent velocities of diffusion differing by a factor of 8, with the boundary which was faster in electrophoresis being also faster in diffusion. The sphere apparent particle sizes, calculated according to the Einstein diffusion formula, were 170 and 1400 A at the fast and slow boundaries respectively.

Studies of mixed colloids were started by preparing mixed nitrate-stabilized thoria—iron oxide colloids in ratios ranging from thoria rich to iron rich, without evidence of gross segregation or mutual flocculation of the sols. These sols were dried to gels that maintained a vitreous character.

Protactinium Chemistry. — Further work on the solubility of protactinium oxide in hydrochloric acid solutions did not confirm the results previously reported. The previous results indicated an increasing solubility from about 8 mg/liter in 7 N HCl to about 90 mg/liter in 11.5 N HCl. A somewhat different procedure was carried out on duplicate samples. An excess of Pa_2O_5 powder was mixed with 10 N HCl and agitated without heating for several days. The sample was then centrifuged and analyzed. One sample indicated a solubility of 40 mg/liter, and the other 35 mg/liter, significantly different from the value of 80 mg/liter obtained in the previous work. A third experiment, in which 1.5 liters of 10 N HCl was contacted with excess solid and then passed through an anion exchange resin column, showed that only 27 mg of protactinium had dissolved, or 18 mg/liter.

Attempts were made to obtain ultraviolet spectra of these solutions in 10 N HCl, with confusing results. Different dilutions of the same solutions yielded quite different spectra, and none agree with those reported in the literature. All previous workers report a very strong band at about 213 mμ and a much weaker one at 260 mμ. In this work a band at 270 mμ was consistently obtained, which could be correlated with the protactinium concentration. Some solutions showed no peak at all at lower wavelengths, and some showed a peak at 208.5 mμ. The heights of these peaks could not be correlated with the protactinium concentration.

Three different absorption spectra have been observed for different protactinium solutions in concentrated sulfuric acid (18 M), with absorption peaks at 2500, at about 2250, and near 2000 A. Upon dilution with water the 2000-A peak maintained almost the same extinction coefficient (in contrast to the 2500-A peak reported previously, which disappeared), and a shoulder appeared at about 2800 A for acid concentrations below 13 M. Dilution of the 18 M acid solution to 2.4 M yielded a solution that gave two well-defined peaks, at about 2000 and 2800 A. Although shoulders at about 2800 A have been reported previously, this is the first time a peak has been observed.

The solubility of protactinium in concentrated sulfuric acid is low, about 0.01 mg/ml, but the first dissolution of a small amount of protactinium oxide (leaching with 18 M H_2SO_4) yielded an apparent solubility substantially higher than that for subsequent dissolutions. Similarly, the ultraviolet spectrum was also higher, but it may not have been in direct proportion to the solubility. The first two dissolutions made on a few milligrams of the oxide obtained initially from Harwell, with concentrated sulfuric acid, gave apparent solubilities of 25 and 6 mg/liter, respectively, and molar extinction coefficients of 2.4×10^4 and 1.7×10^4 for a peak at 2160 A. With subsequent dissolutions the peak shifted to shorter wavelengths, near 2000 A, but the solubility remained about 6 mg/liter.

Irradiation Effects on Heterogeneous Systems. — Studies of the radiolysis of water adsorbed on two silica gels and on porous Vycor glass

were continued. Earlier studies showed that these systems exhibited energy transfer of absorbed nuclear radiation from the solid to the sorbed water but that the initially high yields of hydrogen decreased with irradiation time. Current experiments are devoted to attempts to find conditions under which the high yields can be maintained.

No success in maintaining high yields was obtained with silica gels having surface areas of 682 and 227 m^2/g. A G_{H_2} value of 0.4 to 0.6 was maintained for 60 hr of irradiation by keeping the porous Vycor glass (155 m^2/g) under water-saturated argon. However, these values are not much higher than for pure water ($G_{H_2} = 0.45$), and the technique did not work for the higher-surface-area gels, for which the energy transfer is more efficient.

Analysis of Molten Salts by Electrochemical Methods: Voltammetric Measurements in Molten $LiF-BeF_2$. — It is now believed that the reduction waves observed at both pyrolytic graphite and platinum indicator electrodes at approximately −1.2 v vs the platinum quasi-reference electrode are due in part to the reduction of hydroxide ion impurity in the melt. As previously pointed out (ORNL-3814, p. 31), however, the overall electrode reactions appear to be very complex.

After making a standard addition of LiOH to the melt, it was observed that the overall limiting current at both indicator electrodes increased and then slowly decreased over a period of several days to a reasonably constant value. This behavior raised the question whether the OH^- remained in the melt as such or slowly reacted with the fluoride ($OH^- + F^- \rightarrow HF + O^{2-}$) to form oxide and HF. To check this possibility further, a cold trap that contained a known amount of standard sodium hydroxide was placed in the vacuum line leading to the cell, and then a second addition of 220 mg of LiOH was made to the melt. The cell was evacuated, and the off-gas was pumped into the cold trap and analyzed at 24-hr intervals. These tests revealed that approximately 50% of the LiOH added appeared to react with the melt to evolve HF over a period of approximately six days, after which only traces of HF (or other acidic substances with the capacity for neutralizing sodium hydroxide) were found in the cold trap.

Following this experiment, the cell was pressurized with helium, and the electrodes were inserted into the melt. Subsequent current-voltage curves at both platinum and pyrolytic graphite indicator electrodes again revealed an increase in the limiting current at approximately −1.2 v. The increase in current is of qualitative significance, because the melt did not completely clear up after the second addition of LiOH. The magnitude of the limiting current as seen at both indicator electrodes has remained relatively constant.

Previous experience with the electrolysis cell has revealed that it is difficult to maintain a good vacuum ($<30\ \mu$) within the cell with the electrodes in place; hence most of the work has been done with a helium overpressure within the cell. Recently, however, by utilizing vacuum grease and Glyptal paint around the electrodes and Swagelok fittings, it was possible to evacuate the cell to approximately 3 μ. Current-voltage curves recorded with the melt under vacuum were not significantly different from those recorded with the melt in a helium atmosphere; however,

some interesting observations were made. When the melt was electrolyzed at +1.2 v under vacuum, gas bubbles could be seen escaping at the anode. An unknown anodic wave has consistently been observed at approximately +1.1 to +1.2 v in these fluoride melts; this is the first time that a gas has been observed as a consequence of this electrode reaction. A few gas bubbles could be observed escaping from the melt on electrolysis at −1.2 v (potential of OH^- reduction). The evolution of gas was not, however, as pronounced as in the case of the anodic experiments. Further experiments along these lines are planned, and an effort will be made to identify the evolved gases.

Spectrophotometry of Solutions over Wide Ranges of Temperature and Pressure. – We reported (ORNL-3788, p. 28) studying the spectra of pure H_2O and stock D_2O (0.29 at. % H) in the condensed states between 0.7 and 2.0 μ from −20 to 250°C. Recently, we have measured the spectrum of D_2O as a function of both temperature and added H_2O, and deduced therefrom the spectrum of pure D_2O. The light-water contaminant in stock D_2O exists essentially in the form of the mixed molecule, HDO, which, like H_2O, absorbs strongly above 1.3 μ, and adds apparent breadth and complexity to the D_2O bands.

The vibrational bands of HDO responsible for the interference in the spectrum of D_2O are probably the $(2\nu_3)$ band at 1.4 μ and the $(2\nu_2 + \nu_3)$ and $(\nu_1 + \nu_3)$ bands in combination between 1.6 and 1.7 μ.

The spectra of pure H_2O and D_2O are qualitatively similar; however, the D_2O bands occur at longer wavelengths by a factor of 1.35, are narrower, less intense, and less temperature sensitive.

Radiation Chemistry. – It has been recently established that the e^-_{aq}, instead of the H atom, is the principal primary reducing species in the radiolysis of water by ^{60}Co gamma radiation. Understandably, the Lea-Platzman model for the primary physical processes of energy absorption gained currency over the Samuel-Magee model. Their disagreement concerned the fate of the electron from primary ionization of water: the electron escapes from the parent positive ion in the Lea-Platzman model, while Samuel and Magee have deduced that the electron does not escape. In the original Samuel-Magee model, charge neutralization was assumed to yield an H atom and an OH radical. However, Magee has recently proposed that charge neutralization yields H_3O and an OH radical and that H_3O is the precursor of the e^-_{aq}.

A critical survey of the literature indicates that the electron is not the precursor of intraspur H_2. Furthermore, homogeneous kinetics was used instead of diffusion kinetics to express the dependence of intraspur G_{H_2} on solute concentration. The rate-determining step for H_2 formation is not the combination of reducing species but first-order disappearance of "excited water." Two physical models of "excited water" are considered. In one model the H_3O + OH radical pair is assumed to undergo geminate recombination in a first-order process, with H_3O combination to form H_2 as a concomitant process. In this model, solute decreases G_{H_2} by reaction with H_3O. In the other model, "excited water" yields freely diffusing H_3O + OH radicals in a first-order process, and solute decreases G_{H_2} by reaction with "excited water." The dependence of intraspur G_{H_2} on solute concentration indicates $\tau_{H_2O^*} = 10^{-9}$ to 10^{-10} sec.

TRANSURANIUM-ELEMENT PROCESSING

Process Development: Purification of ^{242}Cm by Anion Exchange. — About 0.5 g of ^{242}Cm was purified by LiCl anion exchange and transferred to the Source Fabrication Facility of the Isotopes Division. This material was recovered from recycle solutions during solvent extraction processing of $^{241}AmO_2$ targets earlier this year.

During this purification, various ion exchange columns were operated at radioactivity levels substantially higher than those previously encountered. The LiCl anion exchange column operated satisfactorily at a resin loading equivalent to about 600 w/liter; however, completely satisfactory results were not obtained with an α-hydroxyisobutyrate cation exchange column, operated in an attempt to separate ^{242}Cm from ^{241}Am by chromatographic elution. A resin loading equivalent to 5000 w/liter was achieved in this experiment, and gassing of the solution was effectively suppressed by the use of methanol; however, resin degradation was severe, and about one-third of the ^{242}Cm remained irreversibly embedded in the resin during elution. This curium was recovered by leaching the charred resin in 6 M HNO_3 for 24 hr.

In future experiments, efforts will be made to alter loading characteristics and elution rates in order to prevent severe resin degradation.

Process Development: Preparation of Dense PuO_2 for HFIR Targets. — About 290 g of PuO_2 containing 255 g of ^{242}Pu has been prepared for incorporation into HFIR targets. These targets will constitute the first loading of the HFIR.

The oxide was prepared by the hydroxide precipitation method (see ORNL-3452, pp. 114–15) and was ground to the desired particle-size range with a mortar and pestle. The following sizes were obtained:

Screen Size	Grams of PuO_2	Percent
−70 +100	135.30	46.7
−100 +200	66.78	23.1
−200 +325	38.40	10.5
−325	57.14	19.7

Previously, the −325-mesh fines, generated during grinding, were recycled; however, thermal conductivity experiments are now being conducted to determine the feasibility of incorporating this material into HFIR targets.

In additional processing, about 23.5 g of PuO_2 was recovered from metallurgical waste (rejected aluminum pellets). This oxide, as recovered, should be suitable for reincorporation into HFIR targets. All additional ^{242}Pu on hand must be recovered from process waste prior to conversion to dense oxide.

Process Development: Dissolution of PuO_2. — In-cell dissolution experiments with segments of irradiated prototype HFIR targets are in progress. Experiments will be conducted with the rods designated TRU-1 (about

40 at. % burnup) and TRU-2 (about 80 at. % burnup). Preliminary results indicate significant dissolution of the PuO_2 content of TRU-2 in HCl, although this reagent is not effective in dissolving unirradiated PuO_2.

Process Development: Preparation of Actinide and Lanthanide Oxide Microspheres. — Laboratory studies of praseodymium and ^{241}Am sol-gel preparations have continued. Previously reported variations in praseodymium sol preparations appear to be due primarily to impurities present in the tetramethylammonium hydroxide used to precipitate praseodymium from nitrate solution. This reagent is frequently contaminated with CO_2 and is reported to contain amine impurities, which may be detrimental. To date, efforts to purify this reagent have only been partially successful, and a good method for removing CO_2 has not been found. Despite this, the procedure was used successfully to prepare a second ^{241}Am sol. Microspheres made from this sol calcined to oxide spheres at 1150°C. Equipment necessary to determine the density and crushing strength of the spheres is not yet available.

To simplify the sol-gel process, nitrate extraction with neutral amine is being investigated. In this procedure, microspheres are produced by coextraction of nitrate and water from droplets of concentrated nitrate solution which are slowly agitated in the amine solvent phase. To date, it has been possible to produce small transparent spheres which calcine to oxide spheres. As yet, sufficient quantities have not been prepared to evaluate density and crushing strength.

A variety of sols, gels, and microspheres have been examined by ultraviolet, visible, and infrared spectroscopy. Organic matter and carbonate have been detected in some preparations, but good correlation of detected impurities with product appearance has not yet been attained. The ratio of nitrate to praseodymium in sol products can be rapidly determined by spectroscopy.

Curium Recovery Facility: Solvent Extraction. — Preparations for a second ^{242}Cm production run have continued. Efforts have been directed toward repair, maintenance, and modification of equipment and toward the replacement of the house vacuum pump with a steam-jet system. Decontamination of the area in the vicinity of the house vacuum pump was completed, and the pump-system shield was then enclosed in a special containment enclosure. The shield and pump were dismantled and removed. Preparations are in progress to pour a section of new floor in that area. Design of the steam-jet vacuum system that will replace the pump is complete, and the equipment is on order. Fabrication of the tantalum mixer-settler is on schedule in the shop, and all the associated equipment and material are on hand or on order. Other modifications scheduled at lower priority include design and fabrication of a vacuum-energized solution-transfer system (replacing the feed-transfer pump) and a remote adjustment for feed flow control. These items are now in the conceptual design phase. Fabrication of the tantalum liner for the new curium product storage tank is complete; the liner has been set in the tank, and fabrication of the head is in progress.

The occurrence of significant amounts of corrosion products in the entrained aqueous solutions collected with the waste organic solution

led us to suspect a breach in the glass liner of the tank. Exposure-test data indicate that no perforation exists up to the 10-liter level.

Experimental Engineering Studies: Cell Mockup. — The proposed leak-detection system for the master-slave manipulator double booting was tested in the mockup. A boot tear equivalent to a 1/9-in.-diam hole is easily detected in either the outside or inside boot. The leak-detection system consists of a controlled air supply of 700 cc/min through an orifice to the cubicle interior, with the space between the boots interconnected to the air flow system. A change in pressure inside the air system indicates which boot is leaking. The pressure between the boots was set intermediate between atmospheric and cubicle pressure. The inside boot registered a pressure change of 0.6 in. H_2O gage and the outside boot a pressure change of 0.3 in. for the 1/8-in. hole. Holes less than 1/16 in. in diameter can be indicated by careful testing. The system, of course, is not operable while the manipulators are in use, but a test can be made in less than a minute after the manipulator is stopped.

The transfer-area port closure and carrier-charging system were operated successfully in the mockup after minor adjustments. Final development work is being done on the canister-sealing device, and the entire system should soon be available for installation.

HFIR Target Fabrication Development: Plutonium Target Fabrication. — Fabrication of the HFIR "critical-experiment" target elements was begun, and the fabrication of pellets for 14 elements has been completed. About 40 simulated target elements will be fabricated to supply 31 elements for a target assembly, plus spares. Fabrication of the pellets that contain uranium oxide and tantalum is proceeding very smoothly. By use of a batch-blending vibratory-feeder dispensing method, we are maintaining the pellet lengths within approximately ±0.006 in. of the nominal for the pellets. Their density is 90% of theoretical. Fabrication of the 40 target elements should be completed within the next month.

Before beginning fabrication of the simulated target pellets, tests were performed to find a better way to blend and dispense a homogeneous mixture of the core powders, using tungsten carbide (density of 14.3 g/cc) to simulate PuO_2 (density of 11.5 g/cc). It developed that batch blending and vibratory feeder dispensing was the best system investigated, and was definitely better than the TRU blender-dispenser setup used in the fabrication of previous $^{242}PuO_2$-containing pellets.

HFIR Target Fabrication Development: HFIR Target Irradiations. — The third prototype target rod, TRU-3, removed from the reactor last month, has been received from Idaho, and the hot-cell examinations were started during the middle of June.

The examination of TRU-2 is continuing. This prototype rod operated at high surface heat flux and was damaged after discharge from the reactor. The burnup sample has been dissolved with some difficulty, but the analyses have not yet been made. Five 1/2-in.-long sections, roughly one pellet each, are being used for dissolution studies. Similar sections from TRU-1 will also be dissolved.

As reported in April (ORNL-3814, p. 35), six "rings" nearly free of oxide were observed on the outside of the target tube. At that time we suggested that the flexing action during postirradiation handling had

caused the heavier scale over the fueled regions to crack and flake off. Examination of normal metallographic samples showed very little oxide scale on the outer surface of the target tube. To investigate the premise that the scale was lost during sample cutting prior to mounting, a 1-in. length of rod was first mounted and then sectioned longitudinally by carefully grinding. This technique resulted in the retention of the scale by the mounting material. With this section we were able to measure the film thickness, the average value being 0.0016 in. over the fueled region and 0.0013 in. over the unfueled region. These films were made up of two layers, with the layer adjacent to the metal being 0.0011 in. over the fuel and 0.0009 in. over the unfueled pellet end caps. The film in the region of the "rings" was darker and had a maximum thickness of 0.00015 in. The layering of the film in the undisturbed areas and the difference in appearance and thickness of the film in the "ring" areas suggest the following: first, during irradiation the variable-thickness layer of film adjacent to the metal was formed; second, during storage in the reactor canal an additional layer of film was accumulated, probably as bayerite ($Al_2O_3 \cdot 3H_2O$); third, both layers flaked off at the rings during preparation for shipping, and the film found in these areas was formed through exposure to air during shipment and hot-cell examination. The separate layers in the retained-scale regions may have resulted from the air drying and conversion of the trihydrate bayerite to the more common monohydrate boehmite. Unlike the films formed on alloy X-8001 out of pile, even the base layer contained very few metallic inclusions. This same premounting technique will be used, and the film structures will be examined, to evaluate the effects of the different heat ratings.

The fourth prototype target rod is still being irradiated, at an unperturbed neutron flux of 3.4×10^{14}; one more step in flux is planned.

Transuranium Processing Plant Design and Construction. — Beneficial occupancy of the Transuranium Processing Plant, Building 7920, was accepted on May 14, 1965, subject to a 30-page list of exceptions that the contractor agreed to finish as soon as practical. The H. K. Ferguson Company began the installation of process equipment under a cost-plus-fixed-fee contract on May 17, 1965. All design has been completed for that portion of the work. The intercell-conveyor port closures and the cell-cubicle top doors at the Process Equipment Company are proceeding approximately on schedule. A July 1 delivery is anticipated. Work orders were written for the fabrication of the equipment transfer case shield at Paducah and the decontamination glove-box facility at ORNL. A purchase order was written for a monorail hoist to be installed in the mechanical equipment room.

Redesign of the first-cycle solvent extraction rack was completed, and fabrication will start in the coming month. Modifications to the original rack are being completed, which will convert this rack for service in the second cycle. Progress on all shop fabrication continues on schedule. Design of several in-line radioactivity monitors for process control was started. An alpha detector and a neutron detector for monitoring the product stream from the Cf-Es-Fm ion exchange separation column will prove of great value, indicating when product cuts should be made. Monitors will also be installed on two waste streams.

METALLURGY AND MATERIALS

Preparation of Pure Materials. — The compound Li_3AlF_6 is a complex fluoride, formed from metals of low atomic weight and low neutron absorption cross section, that is of current significance in chemical reprocessing. Pure crystals of this compound have been prepared for use in obtaining fundamental thermodynamic and crystallographic information. The crystals were prepared by vapor deposition on a nickel substrate at 450 ± 5°C; the vapor was produced by distilling the molten mixture of the stoichiometric composition under reduced pressure. The process gave biaxial single crystals with edges longer than 0.5 mm. Their optical properties and x-ray diffraction patterns conformed with those previously reported for the compound (ORNL-3761). In unpublished work, Li_3AlF_6 had been observed to undergo five equilibrium transitions according to the following sequence:

$$\alpha \underset{}{\overset{225 \pm 50^\circ C}{\rightleftharpoons}} \beta \overset{475 \pm 10^\circ C}{\rightleftharpoons} \gamma \overset{575 \pm 10^\circ C}{\rightleftharpoons} \delta \overset{705 \pm 10^\circ C}{\rightleftharpoons} \epsilon \overset{771^\circ C}{\rightleftharpoons} \text{liquid}$$

The last four transitions are considered to be readily reversible. X-ray diffraction studies of pure crystals obtained from the nickel substrate suggest that they are orthorhombic, and we infer they correspond to the beta form in the sequence above.

Deformation in Crystalline Solids. — Face-centered cubic solid solutions have been used in most of the previous investigations of solid-solution strengthening. According to these investigations, in general, a metal becomes stronger due to an alloying addition. This is not true of tantalum and tantalum-base alloys. The thermal component of the flow stress changes only little due to alloying; for example, the flow stress of a Ta-9.2 at. % Nb alloy is slightly less than that of tantalum. This effect cannot be explained in terms of existing solid-solution strengthening theories, according to which the addition of any solute element increases the flow stress. The reduction in flow stress can readily be explained in terms of the double kink (Peierls) mechanism. If the alloying addition reduces the shear modulus of the solid solution below that of the base metal, it correspondingly reduces the activation energy of plastic deformation, which in turn reduces the stress necessary to maintain a given strain rate.

Three stages of work hardening have been observed in tantalum and in tantalum-base alloys. However, stage II exists only over a small temperature range (300 to 550°K). The Ta-9.2 at. % Nb alloy shows a similarly short temperature range for the existence of stage II, but its rate of work hardening is less. The activation volume is slightly greater in stage II than in stage I, whereas in face-centered cubic metals and solid solutions the activation volume is less in stage II than in stage I.

A tentative explanation of the temperature dependence of work hardening (i.e., the existence of stage II) is based on the conclusion that screw dislocations are much less mobile than edge dislocations at low temperatures (large effective stresses). At higher temperatures (small effective stresses) the effective difference in mobility of dislocation components is less because the athermal component contributes a larger

fraction of the flow stress. Stage II exists because the edge component of the dislocation interacts with other components to form a cell structure. Electron transmission micrographs show no cell structure in samples deformed at low temperatures, but cell structure is observed in samples deformed at higher temperatures.

Fundamental Research in X-Ray Diffraction. – A knowledge of the detailed arrangement of the atoms in a disordered alloy may be obtained only through measurements of diffuse x-ray scattering. Hence, interest in experiments to determine the distribution of the diffuse intensity is high. One of the factors tending to compromise the quality of such measurements is that contributions due to other sources are superimposed on the short-range-order diffuse scattering. These contributions are primarily the result of small displacements of atoms from their sites. The displacements may be either dynamic (due to thermal motion) or static (due to a disorderly distribution of atoms of different sizes on the sites of the crystal).

We have found and are testing a data-reduction method that promises to allow the separation of the various components of the diffuse scattering. No models for either the static or dynamic displacements are required, and measurements at one temperature suffice.

Reactions at Metal Surfaces. – Our study of the effect of existing anodic oxide films on the subsequent thermal oxidation characteristics of the refractory metals tantalum and niobium has been continued. Data recently obtained for the kinetics of oxygen uptake by anodized specimens of tantalum have permitted oxygen permeability values to be computed for the amorphous anodic films. From the temperature dependence of such data, the activation energy for migration of oxygen in the films was measured as 45 kcal/mole. This value appears to be low compared with that estimated for high-density crystalline material, although other factors may be involved which would limit the significance of such a comparison.

Theory of Alloying. – The sharp rise in the temperature of the superconducting transition of zirconium-rich alloys with niobium content has been studied by further measurements of low-temperature specific heats. Quenched alloys containing up to 5 at. % Nb have a hexagonal structure. As the niobium content was increased, we observed a sharp increase in the density of electronic states $N(\epsilon)$ and a rapid decrease in Debye temperature θ; these changes satisfactorily account for the transition temperature in terms of a slowly decreasing superconductivity interaction constant. Heat treatment of the quenched alloys sharply changes the transition temperature, $N(\epsilon)$, θ, and the resistivity of these alloys. These changes are due to precipitation of phases with high critical transition temperature and critical field. Specific heats were measured for dilute zirconium-molybdenum and zirconium-ruthenium alloys in the body-centered cubic structure retained after quenching. The $N(\epsilon)$ and θ values for these alloys and the superconducting transition temperatures were closely similar to those of the hexagonal zirconium-niobium alloys. Extrapolation to pure cubic zirconium gives values close to those for hexagonal zirconium. On this basis the allotropic transition in zirconium arises from changes in the specific heats at higher temperatures, in agreement with measurements of the vapor pressure of zirconium-cadmium alloys.

Fundamental Investigation of Radiation Damage in Solids: Thermally Excited Reorientation and Migration of V_K Centers in Potassium Iodide. — The thermally excited reorientation of V_K centers has been studied in KI in a series of isothermal annealing experiments near 80°K. Doped crystals of KI were irradiated at 78°K, and prominent V_K absorption bands were observed at 404 and 800 mμ. Bleaching with linearly polarized light at 404 mμ induced anisotropic V_K absorption. The crystal was then quickly warmed to a fixed temperature, and the anisotropy was measured as a function of time. In all cases it was found that the anisotropy A(t) was accurately described by $A(t) = A(0) \exp(-Kt)$, where K is a rate constant characteristic of the temperature. Two series of experiments were performed. In series I the electric vector of the orienting light was parallel to a <110> direction, and in series II the electric vector was parallel to a <100> direction. In both series it was found that K was described by $K = s \exp(-E/kT)$. Denoting series I and II experiments by the subscripts 110 and 100, we find that $E_{110} = E_{100} = 0.273$ ev, but at any temperature $K_{110} = 2/3\ K_{100}$. The difference between K_{110} and K_{100} is attributed to a preferential thermal reorientation of V_K centers. Analysis of the reorientation kinetics shows that $K_{110}/K_{100} = 2/3 + 1/3\ P_{90}/P_{60}$, where P_{90} is the probability per second that a V_K center reorients through 90°, and P_{60} is defined correspondingly. Comparison with experiment shows that $P_{90} \ll P_{60}$, from which we conclude that reorientation occurs almost entirely through one-step jumps in which the V_K axis changes direction by 60°.

The migration of V_K centers through the lattice is being studied at higher temperature. The absorption due to V_K centers decays according to second-order kinetics at small t. Analysis of the data will permit a determination of the activation energy for migration, and this will provide an estimate of P_0 compared with P_{60}.

Fundamental Investigation of Radiation Damage in Solids: Some Effects of Small Displacements from Equilibrium on the Structure and Properties of Single-Crystal Copper Electrodes. — Small displacements from equilibrium conditions were generated in systems of variously oriented copper single crystals exposed to acidic copper sulfate solutions by permitting a slow leak of air into the system and/or by application of an external emf. The effects of dissolution conditions on the orientation and shapes of structures formed on the surface were determined. An anomalous effect of a simultaneous corrosion reaction on electrolytic current-potential characteristics also was observed. Explanations for the phenomena in terms of kinetic processes occurring on the surface can be offered.

Fundamental Investigation of Radiation Damage in Solids: Anisotropic Energy Losses in Face-Centered Cubic Crystals for High-Energy ^{79}Br and ^{127}I Ions. — Channeling effects on the energy losses of fast heavy ions in solids were investigated. Multicomponent beams of bromine and iodine ions with energies of from 20 to 80 Mev, obtained from the Oak Ridge tandem accelerator, were passed through thin (0.4 μ) gold single crystals which could be rotated about an axis in the (100) surface. When low-index crystal directions ([001], [110], and [112]) were aligned with the beam, a single energy-loss group was observed, for which the loss was significantly

less than the value obtained for polycrystalline gold of the same thickness. The fractional loss varied from 50 to 85% of the polycrystalline loss, depending on the energy, the species of the bombarding particle, and the particular crystal direction. When the crystals were oriented with low-index planes parallel to the beam direction, two energy-loss groups appeared, one having the normal loss and the other having a lower loss which could be correlated with channeling in the interplanar space. Both the fraction of the ions which were channeled and the angular acceptance aperture into the channel were found to decrease with increasing ion energy, which is in accord with expectations based on an active focusing mechanism. The fact that two distinct groups were observed, rather than a continuum of energy losses, implies that most of the channeled particles were focused almost immediately upon entering the crystal and remained in these channels during the remainder of their trajectories through the solid.

Fundamental Investigation of Radiation Damage in Solids: Measurement of the Harmonic Generation of Ultrasonic Waves in Copper. — Large-amplitude ultrasonic waves are distorted in a nonlinear medium. This distortion is manifested in the generation of higher harmonics of an initially sinusoidal wave propagating in a solid. The amplitude of the second harmonic is proportional to the square of the fundamental amplitude and, for single crystals, to a combination of second- and third-order elastic constants, the exact form of which depends on the crystallographic direction in which the sound wave propagates. By using a capacitive detector, we have measured the absolute amplitudes of ultrasonic pulses at both the fundamental of the driving frequency and its second harmonic in neutron-irradiated copper single crystals.

Fundamental Investigation of Radiation Damage in Solids: Magnetic Structures of Metallic Erbium. — Single-crystal neutron diffraction data were obtained for metallic erbium at sample temperatures from 4.2 to 298°K. Three distinct regions of long-range magnetic order were observed. In the high-temperature region (52 to 80°K) the ordered moments are confined to the c axis direction and are sinusoidally modulated with a modulation wavelength of $3.5C_0$. The amplitude of this modulation saturates at 52°K with a maximum value of 7.6 Bohr magnetons per erbium atom. Below 52°K the basal plane component begins to order, and a third-order modulation of the c axis is observed. These effects are interpreted on the basis of a squared elliptical spiral with its major axis tilted relative to the crystal c axis. However, the structural details in this region are obscured by a temperature variation of the modulation wavelength and by the incommensurability of the modulation and the lattice. In the low-temperature region (4.2 to 20°K) a ferromagnetic spiral structure is indicated. At saturation the ferromagnetic c axis component is 7.9 Bohr magnetons per erbium atom and the spiral basal plant component is 4.3. The total ordered moment is then the free-ion value of 9.0.

Fundamental Investigation of Radiation Damage in Solids: Role of Crystalline Orientation in the Behavior of Copper as Electrode in Chloride Solutions. — The aim of this work has been to provide a better insight into the causes of the different kinetic behavior of differently oriented surfaces of copper single crystals in electrochemical systems. As one

of the aspects of the problem, it has been thought to be relevant to study such behavior in chloride solutions, where copper ions are overwhelmingly in the monovalent state and therefore the electrode processes are limited to the exchange of only one electron. Moreover, it is known that in solutions of similar composition the defect structure of copper single crystals can play a detectable role in determining the etching pattern.

In this research, copper single crystals having (100), (110), (111), and (321) orientations, as well as polycrystalline specimens, have been used as electrodes in copper-containing chloride solutions previously equilibrated with copper metal under a hydrogen atmosphere. Anodic and cathodic overvoltage values as a function of current density have been recorded in the current density range from 10^{-7} to 10^{-3} amp/cm^2. In addition to these electrochemical measurements, the kinds of faceting pattern formed by electrolysis at various current densities, as well as those obtained by corrosion in nonequilibrated solutions, have been examined, using both the optical microscope and the double-circle reflection goniometer. At the same time, information as to whether differently oriented electrodes possess different equilibrium potentials has been collected. It has been confirmed 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 charge-transfer reaction, and it was concluded that the properties of the surface structure were decisive kinetically.

In all instances it has been found that the behavior of (100) oriented surfaces was remarkably different from that of all other orientations investigated. The overvoltages recorded on (100) electrodes were considerably higher and exhibited an extremely high initial slope, the values of $(d\eta/di)_{i=0}$ being of the order of 10^5 ohms cm^2. Lower overvoltages were found on the other close-packed orientations, differences between them being only minor, and the lowest values were exhibited by (321) oriented electrodes. The difference in behavior between (100) and all other orientations was also found in the range of facets developed by anodic dissolution: whereas (100) developed only facets vicinal to (100), the other surfaces formed a completely different set of facets.

By correlating the overvoltage data and the faceting structure, some suggestions can be made concerning the relative influence of elementary processes, such as nucleation of new steps and then motion across the surfaces, on the kinetic behavior. The most likely conclusion at this stage seems to be that the rate-determining step is linked to the removal of atoms from or addition of atoms to the surface steps.

Fundamental Investigation of Radiation Damage in Solids: Simulated Etching of Close-Packed and Vicinal Surfaces. — On close-packed and vicinal surfaces dissolution usually occurs by the removal of atoms at steps, causing the steps to move, and the dissolution process can be described in terms of the motion of trains of these steps. An example is the etching of (111) surfaces of copper in HCl-HBr mixtures. Under certain conditions for this and other reactions of its type, steps aggregate to form multiatom ledges. In an attempt to explain this type of behavior for copper surfaces, a recent theory of step motion by Mullins and Hirth has been employed in which the rate of atom removal at a step, which is a

measure of the velocity of the step, is expressed as a function of the distance from its nearest neighboring steps. The dissolution of the crystals as a whole is represented by an infinite set of step velocity equations. Whereas in closed analytical form the theory can only be applied to very simple step configurations, a numerical approach, by high-speed machine calculation, allows one to treat large and complicated step configurations that should approximate actual surfaces. Calculation times are sufficiently short to make it practical to simulate dissolution experiments on the computer. The Mullins-Hirth theory has been tested by comparing computer experiments with laboratory experiments and was found to give a satisfactory explanation of ledge formation. In the laboratory, step velocities were measured under conditions for which ledges were both stable and unstable, and step interaction functions were estimated from these measurements. When a step interaction function that corresponded to stable ledge conditions in the laboratory was used in a computer experiment, it caused ledges to be generated; conversely, interaction functions corresponding to unstable conditions in the laboratory caused the computer ledges either not to be generated or to be unstable.

CONTROLLED THERMONUCLEAR RESEARCH

The DCX-2 Injection-Accumulation Experiment. — Continued development has resulted in a hydrogen arc that runs on a gas feed of ~0.3 cm^3/sec at a current of 75 to 80 amp and at 200 to 250 v. Earlier gas arc experiments in the DCX-2 required a gas flow of 10 cm^3/sec. Fluting of the arc column has prevented a reduction of the pressure in the plasma region below $\sim 2.5 \times 10^{-6}$ mm Hg. The trapped plasma density increases with increased gas feed to the arc, and the maximum density achieved thus far is 1 to 2×10^9 ions/cm^3 at a gage pressure of 5×10^{-6} mm Hg. Since both the percentage dissociation of the molecular hydrogen ion beam and the partial pressure of hydrogen in the plasma region increase with increased gas feed, it is uncertain which process is responsible for the higher density. This density is somewhat lower than that obtained with the lithium arc.

Experiments with hydrogen gas dissociation indicate that the density of the plasma produced increases with increased gas flow, reaching a maximum value of 5×10^8 ions/cm^3 at a gage pressure of 1.8×10^{-5} mm Hg. The energy of the trapped plasma also spreads, and the mean energy rises. The pitch angle distribution does not show a large group of particles at small pitch angles at the highest density, although such a group has always been associated with a large energy spread in the past. By contrast, nitrogen gas dissociation yields a lower plasma density of 1×10^8 ions/cm^3 which is approximately independent of pressure. Previous experiments with hydrogen gas dissociation had not revealed this behavior, and the reason may have been due to a high impurity level in the gas used. Even in this experiment, only 85% of the gas was hydrogen, and a much higher purity of hydrogen is needed if the plasma behavior is to be dominated by the hydrogen background. A serious magnetic-field-dependent vacuum leak and electrical shorts in the west mirror coil have necessitated the replacement of this coil.

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Active and Passive Stabilization of Plasmas and Ion Beams. — During the study of various boundary impedance networks as applied to the calutron beam, it was found that certain frequencies in the system shift as the background pressure is varied. These frequencies have been correlated with the oscillatory frequencies of electrons which are oscillating along the magnetic field through the positive potential well of the beam. From the measured frequency and energy the amplitude of the electron excursion can be calculated. This amplitude varies with operating conditions but at maximum is slightly more than the width of the ion beam.

Properties of Vacuum Arcs. — Magnetically confined (8300 gauss), high-current (120 to 300 amp), low-pressure ($\sim 10^{-6}$ torr), long (20 ft), carbon arcs have been studied under various conditions. The maximum ion temperature, as determined by Doppler broadening, now appears to be leveling off in these 20-ft-long arcs at about $T_{\perp} \sim 5 \times 10^6$ °K, suggesting a saturation of the ion heating mechanism. If our proposed model for heating ions by multiple inelastic collisions (see ORNL-3652, pp. 74–77) is valid, such a saturation might well be expected for the ion-heating collision cross section. A quite crude check on the appropriate "ln λ" value to be used in the expression for the rate of ion cooling on cold electrons was made in terms of the electrical resistivity of the carbon arc. The resulting "ln λ" was 1.0, as compared with 1.4 previously deduced from our revised cooling rate equation for which

$$\ln \lambda \rightarrow \ln (1 + x)^2 - \frac{2x}{1 + x} ,$$

where $x^2 = \lambda_d m v_+^2 / 2Ze^2$. The conventional ln λ is about 9 for the carbon arc.

Experimental Plasma Physics. — We have observed quiescent plasmas formed in special discharge tubes in which the bulk of the electrons are trapped electrostatically by the positive ions. These plasmas are so noise free that thermalization of the electrons apparently takes place only by the slow process of multiple scattering. Extremely small Langmuir probes must be used to study these plasmas or else electrons are removed so rapidly that the equilibrium electron energy distribution is destroyed. In these plasmas, collisionless ionic sound waves have been propagated, and the adiabatic compression coefficient of the electron gas, γ, is found to be unity. This is apparently the first clear-cut experiment which discriminates between the theoretically possible values of γ of 1, 5/3, and 3.

Vacuum Studies for Fusion. — Mass spectrometric analysis of the low mass peaks in the background gas of the DCX-2 experimental apparatus has been continued with a total of almost 1200 spectral scans having been obtained and tabulated to date. Correlation of residual gas composition with operating parameters of the machine, vacuum condition diagnosis during operation, and gas analysis are the three principal objectives of the analytical assistance. Gas analyses have been made of the argon, nitrogen, and hydrogen supplied to the experiment. Vacuum condition diagnosis may be called for during periods of erratic ion gage behavior to distinguish air and water vapor or to account for apparently slow pump-down cycles. The correlation of gas composition with operating parameters of the machine reflects a combination of many effects, but some conclusions appear to be relatively well founded.

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Operation of the titanium evaporators was associated with a decrease of water vapor and other inorganic gases (CO, CO_2, N_2, and O_2), while hydrogen tended to increase (especially at water pressures greater than 10^{-7} torr) along with methane. Magnetic-field effects appear to be largely instrumental. Lithium-arc operation differed markedly from hydrogen-arc operation in the chemical changes observed. While lithium-arc operation tended to provide a considerable pumping action (especially for hydrogen), the hydrogen arc was normally associated with a large (0.5 to 1.5×10^{-7} torr) increase in the partial pressure of methane. Operation of the hydrogen arc with a partial feed of deuterium gas led to a rapid increase in masses 19 and 20 (from deuterium oxide) and a slower increase of partially deuterated species such as lead to masses 58 and 59 and 45, 46, and 47. It was found that after only 2 hr of arc operation with hydrogen gas feed on the following day the deuterium-containing masses were at normal levels. This result was gratifying, since mass spectral confusion due to isotopic interchange in a device such as the DCX-2 experiment would virtually preclude the possibility of the development of a suitable analytical capability. The injection of the 600-kv hydrogen molecular-ion beam appears to be associated with a synthesis of acetylene and methane primarily (ΔP = 1 to 2×10^{-7} torr). This increase is accompanied by increases of approximately the same magnitude of carbon dioxide, carbon monoxide, and other organic compounds. The origin of these species is uncertain.

The apparatus used to study the hydrogen occlusion of molybdenum has been modified to allow operation of a glow discharge with hydrogen gas of higher purity than the 99.5% purity level which characterized the initial experiments.

BIOLOGY AND MEDICINE

MOLECULAR- AND CELLULAR-LEVEL STUDIES

Physics of Tissue Damage: Optical Properties of Polystyrene. — The optical properties of polystyrene are being studied in the wavelength region below 3000 A. Samples $1 \times 1\frac{1}{2} \times \frac{1}{4}$ in. are machined from a commercially obtainable polystyrene rod 1 ft long and 3 in. in diameter. Each sample is polished, annealed, and then lapped to 0.000033 in. of optical flatness. The polystyrene block is attached to a holder in the reflectivity box, which is attached to the monochromator, for the reflectivity measurements. Reflectance data for 20 and 70° incidence are obtained by using a McPherson model 235 vacuum ultraviolet scanning monochromator. From the reflectance curves the real (n) and imaginary (k) parts of the complex index of refraction are obtained. The n and k values are then used to obtain the real (ϵ_1) and imaginary (ϵ_2) parts of the complex dielectric constant ϵ (where $\epsilon = \epsilon_1 + i\epsilon_2$). The complex dielectric constant is related to the complex index of refraction by the relation $\epsilon = (n + ik)^2$. The energy loss function, $\mathrm{Im}(1/\epsilon)$, is calculated from the n and k values, and this curve indicates the energies at which losses occur.

Preliminary data obtained in the 3000- to 1500-A region show the presence of a major energy loss at 6.5 ev and two minor losses at 5.2

and 4.6 ev. The 6.5-ev loss agrees precisely with that observed from ultraviolet absorption data on thin films of polystyrene. This 6.5-ev loss is believed to be associated with the collective electron oscillations of the π electrons in the benzene ring. Data are now being obtained that extend these measurements to include the 21-ev loss which is believed to be associated with the σ electrons in the system.

TERRESTRIAL AND FRESH-WATER ECOLOGY

Aquatic Ecology: Deposition of Strontium and Calcium in Snail Shells. — A previous study of strontium and calcium deposition in snail shells showed that strontium uptake was dependent primarily upon the environmental concentration of strontium and that there was a negative interaction from environmental calcium concentrations. The response surface indicated an interesting area of minimal strontium uptake, dependent upon both calcium and strontium concentrations. An additional experiment was performed to further define the region of minimal strontium uptake. Environmental calcium concentrations ranged from 0.10 to 180 ppm, and strontium concentrations ranged from 0.02 to 8.0 ppm.

Both calcium and strontium uptake response surfaces, calculated from least-squares fits of the uptake data, were expressed by quadratic equations (without the Sr^2 term): Sr uptake (atoms $\times 10^{15}$) = $4.558 \times 10^{19} + 2.660 \times 10^{18}\ Sr_{input} - 5.775 \times 10^{17}\ Ca_{input} + 1.611 \times 10^{15}\ Ca^2_{input} - 6.571 \times 10^{15}\ Sr_{input}\ Ca_{input}$; Ca_{uptake} (atoms $\times 10^{16}$) = $-3.061 \times 10^{21} + 1.848 \times 10^{20}\ Sr_{input} + 8.205 \times 10^{19}\ Ca_{input} - 8.421 \times 10^{16}\ Ca^2_{input} - 1.014 \times 10^{18}\ Sr_{input}\ Ca_{input}$. Previously, a planar response surface was obtained for calcium uptake. The difference in calcium uptake between the two experiments is thought to be the result of an alteration of the magnesium concentration in the basic medium. The minimal strontium uptake was observed within the range of 100 to 150 ppm Ca and 0.02 to 4.5 ppm Sr. This study shows that both calcium and strontium amendments to an environment should be controlled carefully in order to effect a reduction in radiostrontium uptake. Other biological systems should be tested to see if they respond similarly to different calcium and strontium concentrations.

Aquatic Ecology: Feeding Rate of Carp in White Oak Lake. — The feeding rate of carp (*Cyprinus carpio*) and the importance of detritus in the diet of the carp in White Oak Lake are being estimated using a radioisotope method. An equation used to calculate the feeding rate is based on the amount of ^{137}Cs that must be ingested to maintain an equilibrium body burden when ^{137}Cs has a known assimilation and turnover by the organism. The equation is

$$q = (p + b)Q/a ,$$

where q is the rate of ingestion of ^{137}Cs, p is the rate of physical decay, b is the rate of biological elimination of ^{137}Cs, Q is the body burden, and a is the fraction of the ingested ^{137}Cs assimilated by the organism. The assumption that cesium enters fresh-water fish only through

the food chain has been verified in the laboratory. Autoradiograms of scales from carp taken from White Oak Lake demonstrate a uniform distribution of radioactivity in the scales, thus indicating that the carp have been in the lake since they were spawned and are in equilibrium with the lake environment.

Estimates of the parameters of the equation have been made. The rate of cesium turnover (p + b), determined in the laboratory, is 0.01/day at 16.5°C. The average body burden (Q) of ^{137}Cs in the carp from White Oak Lake is 24.2 μμc per g of live weight. The diet, determined by analysis of gut contents, was simplified to 40% algae and 60% detritus. Laboratory studies revealed that carp assimilated (a) about 7% of the ^{137}Cs from ingested detritus and 80% from ingested algae. The average amount of ^{137}Cs associated with algae and detritus samples from White Oak Lake is 18 and 9 μμc per g of wet weight respectively. Based on these parameter values, the calculated amount of ^{137}Cs ingested by a yearling carp (146 g) was 81.2 μμc/day. To obtain the ^{137}Cs, the carp would have to ingest, daily, 3.9 g of detritus and 2.56 g of algae. Since growth is not included, this food intake is for maintenance alone. Based on the measured calorie content of the algae and detritus, the intake for maintenance by yearling carp was about 6000 cal/day. A mammal of comparable size requires about 21,000 cal/day for maintenance.

Radiation Effects on Insect Populations: Radiosensitivity of Bagworm Eggs. — Radiation sensitivity of bagworm eggs was measured as an initial phase of a combined laboratory-field study of radiation effects on populations of herbivorous insects. Overwintering egg clutches of the evergreen bagworm, *Thyridopteryx ephemeraeformis*, were brought out of dormancy in the laboratory and irradiated in replicate sets of 25 in a ^{60}Co gamma source. At an exposure dose rate of 14.4 r/sec, eggs were irradiated at dose levels of $100(2^n)$, where n = 1, 2, ..., 9, and at increasing time intervals after breaking of diapause.

Hatching at each dose level was expressed as percent of controls. Mean hatching time for all controls was 14.1 ± 0.07 days, and mean percentage hatching was 82.0 ± 3.9%. Radiosensitivity decreased with increasing age of embryonic development. Two-day eggs showed no hatching at doses at or above 1600 rads, while a minimum of 25,600 rads was necessary to prevent hatching of 8- to 12-day eggs. Eggs during early embryonic development were about 16 times more radiosensitive than those at late stages immediately preceding hatching. One-day-old larvae were fourfold more radioresistant than prehatch eggs, as interpreted from lethality at 20 days postirradiation (time equivalent to longest development for eggs). One-day eggs were somewhat more resistant than two-day eggs, implying that eggs in diapause may not be as radiosensitive as those in early stages of development. In certain instances it appears that low doses may have an enhancing effect upon hatching; but verification of this phenomenon will require additional data. Besides the obvious effects of these acute irradiations upon egg hatching and subsequent larval survival, alterations in normal behavioral patterns (case building, locomotor activity, and feeding) were also detectable in the larval stage.

Radionuclide Cycling in Terrestrial Ecosystems: Radiocesium Turnover in Forest Cryptozoa. — Radiocesium dispersion in a forest-floor food web is being studied in the *Liriodendron* (tulip poplar) stand tagged with

^{137}Cs. The cryptozoan animals are those living on the forest floor in the zone of leaf litter, rotten wood, mosses, ferns, stones, and upper organic layers of the soil. Pitfall trap collections made at weekly intervals form the basis for estimates of ^{137}Cs equilibrium concentrations in cryptozoan animals. Radiocesium distribution among the food materials (leaf litter, wood, roots, and mosses) and biological half-lives for radiocesium in cryptozoan arthropods have been reported previously. Current findings disclosed activity densities for ^{137}Cs along the generalized food chain to be about 25, 5.0, and 2.7 pc per mg of dry weight in leaf litter, saprovores, and predators respectively. These data yield consumer-to-food ratios of 0.8 to 1.0 for food chains based on green foliage. The reduced transfers among the cryptozoa probably were due to less efficient absorption of cesium from ingested detritus than from living material, as well as dilution of the biomass with untagged animals migrating from surrounding areas.

Radiocesium turnover rates were estimated for eight arthropod species whose body burdens of ^{137}Cs, elimination rates, and food material were known. The balance of input and elimination at equilibrium may be expressed by $I = kQM$, where I is the input (pc/day), k is the elimination constant (day^{-1}), Q is ^{137}Cs activity density (pc/mg), and M is the mass of the arthropod (mg of dry weight). For the species studied (three orthopteran, five milliped species), turnover of radiocesium ranged from about 9 to about 70 pc/day per individual. Turnover of ^{137}Cs per unit body weight was more similar for these species, however, and ranged from about 1 to about 3% of the body-weight activity density per day. These field turnover rates are much lower than those reported for herbivorous insects, for which turnover approaches 100% per day. Lower intake rates and lower turnover rates may be the rule for detritus feeders, however.

Radionuclide Cycling in Terrestrial Ecosystems: Transfer of ^{137}Cs from Trees to Forest Undercover Vegetation. – Although the root contributions from tagged trees are greater than contributions of rainfall and leaf fall to the surrounding environment, the latter are more promptly transferred to other living components of the forest system. Comparing the understory plant concentration and soil concentration of ^{137}Cs for the first two growing seasons, a reverse relationship exists between concentration of ^{137}Cs in plants and soil (sampled in August). Thus plant concentrations in the forest tagged in the overstory decreased from 0.056–0.116 μc/g in 1962 to 0.009–0.026 μc/g in 1963, while mean soil concentration increased from 0.0008 to 0.0064 μc/g, thereby decreasing the plant/soil ratios from 76–158 to 0.5–4.1. By contrast, a nearby plot in which ^{137}Cs was sprayed directly on litter showed only 0.0033 to 0.0076 μc/g on most plants (except for 0.078 for certain species that may have been directly affected by spraying). Since soil activity here was 0.162 μc/g, the soil ratios were 0.02 to 0.05 for most species (0.48 for the exceptional ones).

These latter ratios tend to confirm that herbaceous uptake of ^{137}Cs from soil is rather limited, though perhaps not so severely limited in intact, active forest floor conditions as in mineral soils with well-incorporated cesium. The ratios are so much smaller than those in either the first- or second-year results of the forest tagged via the overstory that results strongly confirm the earlier hypothesis that undercover vegetation in the tagged forest was contaminated almost completely by rainout

from the overstory, and by recycling of ^{137}Cs released during prompt decay of litter.

Evaluation of Fission Product Distribution and Movement in White Oak Drainage System. — The ^{137}Cs loss from the White Oak drainage system, following six months of operation and 27.92 in. of rain, amounted to 17.9% from plot 1 (bare), 14.1% from plot 2 (clipped meadow), and 6.0% from plot 3 (tall meadow). The relationship between ^{137}Cs loss and soil loss was found to follow the equation

$$y = \exp(m \ln x - b); \quad 0 < y < 100, \quad x > 16 \text{ g/m}^2,$$

where y is cesium loss (%), x is soil loss (g/m^2), m is 0.6758, 0.6257, and 0.6382, and b is 1.3345, 0.4505, and 0.9029 for plots 1, 2, and 3 respectively. Letting ln y = z, then for plot 1, z = ±0.0539; for plot 2, z = ±0.0468; and for plot 3, z = ±0.0077.

After 19.7 in. of rain, the ^{137}Cs on the vegetation was measured and compared with the earlier sampling following 7.60 in. of rain. In the tall meadow plot, the nuclide on the vegetation decreased from 39% to 26%. In the plant material of the clipped meadow plot, the percentage decreased from 31% to 23%; whereas in the litter lying on the ground surface, the percentage change was practically nil: 26% to 28%. It is possible that the apparent lack of decrease in the litter is due to transfer, by rain, of ^{137}Cs from the plant to the litter lying on the ground surface.

NUCLEAR ENERGY CIVIL EFFECTS

Ichiban Dosimetry Studies: Operation HENRE. — Operation HENRE (High Energy Neutron Reactions Experiment) is a large field experiment designed to characterize the radiation fields from a monoenergetic (14-Mev) source of neutrons supported at various heights above the ground on a 1500-ft tower. The experiment is to be conducted at the Nevada Test Site later this year. The neutron source is a positive ion accelerator utilizing a duoplasmatron ion source, the $^{3}H(d,n)^{4}He$ nuclear reaction, and an accelerating potential of 150 kv.

After evaluating the sensitivity of various dosimetry and spectrometry instruments to be used in the field experiment, it was determined that the source should have an average output of at least 10^{13} neutrons/sec for individual operating periods of 4 hr. In addition, the total weight of all system components, including the hoisting device containing the neutron source, must be less than 16,000 lb, and quick-release mechanisms must be incorporated for removing highly radioactive components.

Because the neutron source operates on a tower, it must be controlled remotely from an underground station located about 200 ft from the tower base. Hence control cables vary in length from 200 to 1700 ft. The accelerator target, ion source, diffusion pump casing, and other components must be cooled by a recirculating water system. A total of 90 kw must be removed by this system.

All components have been designed and constructed. Final testing is under way at Thermonuclear Division facilities. Results of tests to date have not revealed any major difficulties in system design. It is estimated that the field experiment will begin in early August.

Ichiban Dosimetry Studies: Revised Dose (Exposure) Curves for Hiroshima and Nagasaki. — Revision of the weapons yield figures and further data from weapons tests and the BREN experiment have permitted the calculation of revised "air dose" vs distance curves for Hiroshima and Nagasaki.

Dosimetry and Spectrometry Research. — During November and December of 1964 an experiment was conducted to measure the radiation leakage of the Ichiban critical assembly, which was designed by personnel of the Los Alamos Scientific Laboratory to simulate the Hiroshima weapon. Neutron dose measurements utilized Hurst proportional counters and threshold foil detectors. The neutron energy spectrum was determined from the threshold detector data and, in more detail, with an ^{6}Li semiconductor neutron spectrometer. Neutron-insensitive counters (miniature Geiger counters in lead-plastic shields) and metaphosphate glass rods were used for gamma dose measurements. Neutron output was normalized by using a ^{238}U fission counter, a BF_3 proportional counter, and sulfur pellets. LASL personnel determined the fission yield by performing fission product analyses on small fuel samples irradiated during high-power runs. Dose leakage at a distance of 2 m was corrected for room and air scattering and was normalized to dose per fission in the core. Angular and spatial distribution measurements were made to assure that the data taken at 2 m could be interpreted correctly. Neutron fluence leakage was determined directly with threshold foils and a moderated BF_3 counter and indirectly by dividing the dose per fission by the mean dose per neutron as calculated from the neutron energy spectrum.

As was expected, the massive casing caused the Ichiban neutron energy spectrum to be softened, so that the mean dose per neutron was 1.2×10^{-9} rad neutron^{-1} cm^{2}, and the neutron fluence leakage was reduced by a factor of about 2, compared with Godiva-type reactors, to 0.76 neutron/fission.

ISOTOPE DEVELOPMENT

Nuclear Analysis Research and Development: Activation Analysis with ^{3}He Ions. — Calculations of experimental cross sections of nuclear reactions induced by ^{3}He ions in light elements have been completed. The computer codes NET AREA and CLSQ were used for this purpose. Range-energy curves for ^{3}He particles, based on the tables by Williamson and Boujot (CEA-2189), were constructed for beryllium, Teflon, Mylar, and nylon. The energy losses for these targets were obtained from these curves and used to plot excitation functions. Cross-section curves for reactions of ^{3}He with beryllium, carbon, oxygen, and fluorine were obtained. Interpretation of these data from the viewpoint of nuclear-reaction mechanisms and for application in activation analysis is being carried out.

Stable Isotopes Development. — The feasibility of obtaining ^{196}Hg at 40 to 50% isotopic purity (natural abundance, 0.14%) without using

refrigeration has been demonstrated. The highest purity previously obtained was 8.5%. Enrichment of this isotope has previously been limited by the high vapor pressure of the feed materials, which required extensive use of dry-ice refrigeration. The advancements in purity are attributable to the development of improved receivers and the use of calcium vapor as a pumping and beam-stabilizing medium. Present samples have an ^{196}Hg to ^{202}Hg ratio between 5 and 6, whereas in prior series this ratio was between 0.1 and 0.5. This composition of 40 to 50% ^{196}Hg and the ratio of ^{196}Hg to ^{202}Hg makes samples useful in the formation of radioactive ^{197}Hg.

Small-scale calutron components have been designed for ion implantations, and recent modifications of equipment have enabled ^{32}P to be implanted as a radioactive tracer. This equipment appears capable of being used for other isotope tracer implantations which could be useful in surface studies.

Special Separations. — An isotope separation series to provide >6 g of >99% ^{242}Pu is in progress. One run yielded ~5 g of 99.82% ^{242}Pu. Isotopic recovery and feed recycle from 37 g of recycled feed from the second run are still in progress.

The program to provide 10 g of ^{233}U containing <0.5 ppb of ^{232}U continues. Approximately 4 g of ^{233}U has been collected from 30 g of feed containing 6 ppb of ^{232}U. Other runs are being performed simultaneously in which ^{233}U containing 400 ppb of ^{232}U is being processed to acquire additional feed to supplement the material being used in this high-purity separation.

Target Preparation. — The tritium sorption system for Operation HENRE targets has been completed and tested. Vacuum levels of $\sim 6 \times 10^{-9}$ torr have been routinely achieved using a 200-liter/sec ion pump. The electron bombardment evaporation system for erbium deposition is being modified. A prototype target using hydrogen rather than tritium is being prepared to study accelerator operation.

Calcium-48 metal was obtained by reduction-distillation of the carbonate; ~62% yield was obtained from a 50-mg sample. This metal was difficult to roll into 1-mg/cm^2 foil because of its extreme softness. Since the metal was doubly distilled, the extremely low impurity content probably accounts for the softness. A 2.5-g rectangular piece of ^{48}Ca was prepared to be used in an electron accelerator.

Construction of the heavy-element target laboratory is nearly complete. The facility should be ready for operation by July 1, 1965, and will allow fabrication of rolled foils, evaporated films, and other forms using microgram to gram quantities of alpha-emitting isotopes through ^{252}Cf.

Radioisotope Research and Development. — The equipment for isotopically enriching ^{85}Kr by thermal diffusion has been installed, and test runs with nitrogen and nonradioactive krypton are in progress. This equipment will be used to increase the isotopic concentration of ^{85}Kr in fission krypton from the present 5% to ~40%. Krypton-85 is useful for commercial applications such as tracers for wear studies, luminous signs, and beta gages.

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A 500-w ^{90}Sr heat source has been designed which will be used on a trial basis in a U.S. Navy underwater laboratory (Sea Lab II). The source will heat a container of water in an underwater way station outside the main underwater laboratory. Divers working under water will periodically immerse themselves in the warm water of the way station to restore body heat, which will enable them to work for longer periods before returning to the main laboratory. The doubly encapsulated source will be sealed in a finned container with an inner uranium shield and an outer steel shield which will provide the necessary heat transfer surface, shielding, and underwater integrity. Similar sources in the range between 1 and 5 kw are being designed for use in Antarctica to prevent fuel-oil tanks from freezing.

The application of cadmium neutron filtration to radioisotope production in order to reduce the production of unwanted radioisotopes was demonstrated. Tantalum-182 (half-life 115 days) is formed by thermal-neutron capture from ^{181}Ta with a 21-barn cross section. Subsequent neutron capture by ^{182}Ta (17,000 barns) produces five-day ^{183}Ta, which must be eliminated or reduced to negligible levels by decay before the ^{182}Ta is useful as a radioisotope tracer for medical purposes. Samples of Ta_2O_5 were irradiated with and without 40-mil cadmium covers at a thermal flux of 2×10^{14} neutrons cm^{-2} sec^{-1} for 7.5 days. The cadmium filter reduced the ratio of ^{183}Ta:^{182}Ta from 3.7 (without cadmium filter) to <0.03.

The separation and recovery of ^{22}Na from a magnesium disk target by vacuum distillation were attempted with negative results. The disk was irradiated with 10 μa-hr of 23-Mev protons to produce 5.1 μc of ^{22}Na. The disk was heated in a vacuum (3×10^{-5} mm Hg). No evidence of ^{22}Na distillation was detected until the temperature reached 680 to 700°C, when the magnesium metal also vaporized.

The feasibility of producing millicurie quantities of ^{33}P by the $^{36}Cl(n,\alpha)^{33}P$ reaction was demonstrated. A sample of 0.55 g of KCl (37% ^{36}Cl, 38% ^{35}Cl, and 25% ^{37}Cl) was irradiated for 4.6 months in a neutron flux of 2×10^{14} neutrons cm^{-2} sec^{-1} and allowed to decay for 3.4 months to reduce the ^{32}P content formed by the $^{35}Cl(n,\alpha)^{32}P$ reaction. From this target, 7.5 mc of ^{33}P was recovered containing 0.34 mc (4.5%) of ^{32}P.

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