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# OAK RIDGE NATIONAL LABORATORY

# STATUS AND PROGRESS REPORT

APRIL 1965

**OAK RIDGE NATIONAL LABORATORY**

operated by

UNION CARBIDE CORPORATION

for the

U.S. ATOMIC ENERGY COMMISSION

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# OAK RIDGE NATIONAL LABORATORY

# STATUS AND PROGRESS REPORT

# APRIL 1965

DATE ISSUED

May 6, 1965

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

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# OAK RIDGE NATIONAL LABORATORY

## STATUS AND PROGRESS REPORT

## APRIL 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

GCR-ORR Loop No. 1. — Irradiation of experimental fuel assembly 14 continued. The fuel is a mixture of $(Th,U)O_2$ and $ThO_2$ sol-gel particles coated with pyrolytic carbon and is contained in two capsules, one of which is vented to the loop coolant. We are maintaining the central temperature of the fuel in the vented capsule at 2500°F by the addition of neon to the helium coolant (33% neon at present) to compensate for fuel burnup. We have observed no significant change in the release rate of the noble-gas fission products, which continue to show R/B (ratio of release rate to birth rate) values of approximately $1 \times 10^{-5}$ to $2 \times 10^{-5}$ for xenon and krypton isotopes. Fuel burnup is estimated at 2.7 at. % of the heavy metal (44% $^{235}U$ consumption).

LITR Capsule Irradiations. — Two capsules (L-CP9 and L-CP10), each containing pyrolytic-carbon-coated $UC_2$ particles in four separate compartments of ATJ graphite, continued to operate satisfactorily. The estimated burnup in capsule L-CP9 reached 16.0 at. % of the uranium, and the remaining operable thermocouple indicated a fuel central temperature of 1820°F. Capsule L-CP10 attained an estimated burnup of 4.6 at. % of the uranium, and indicated fuel central temperatures were 1580 to 1800°F.

ORR Poolside Irradiations. — Capsule 04-8, containing EGCR-vendor fuel material, reached an estimated burnup of 4340 Mwd per metric ton of $UO_2$ and continued to operate satisfactorily. The maximum indicated cladding temperature is 1542°F.

BeO-Graphite Compatibility Experiment. — In support of the TARGET reactor concept, an irradiation experiment is being conducted to investigate the compatibility of BeO in contact with graphite at a high temperature (1500°C).

The Mark II capsule continued to operate at the design graphite-BeO interface temperature of 1500°C. Temperature control by variation of the helium-argon concentration in the sweep gas has proved satisfactory. The

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helium concentration has varied from 25% at the beginning of an ORR fuel cycle to 10% at the end of the fuel cycle; this change indicates the variation in gamma heat with reactor fuel consumption.

A small leak developed in the capsule between the inner sweep gas system and the outer gas region. The outer-region gas system of the capsule was then isolated to prevent contamination of the highly purified purge gas of the inner region by the less pure outer-region gas. As a result the purge gas diffused into the outer gas region, increased the argon concentration there, and caused the primary containment wall temperature to climb to 1100 to 1200°F, from an initial 600 to 700°F. No detrimental effects are expected from the leak or from the increased containment temperature.

The Mark I capsule developed a similar leak during operation. We have outlined procedures for locating the leak in the Mark I capsule during postirradiation examination.

We evaluated several types of moisture monitors for the sweep gas system and selected an instrument that appears to be satisfactory. It will be installed between the sweep gas cleanup system and the capsule during the May ORR shutdown.

Thermal Conductivity of Fueled-Graphite Spheres. — In the recent series of tests of ORR poolside irradiation capsules designed to hold three 6-cm-diam fueled-graphite spheres, the temperature of the top sphere was monitored by a central thermocouple, as well as two thermocouples near its surface. Based on the temperature data obtained and the heat-generation rate derived from the neutron dose measured on flux-monitor wires, we calculated the thermal conductivity of the fueled spheres used in three experiments. The calculations were also based on the assumption of steady-state, spherically symmetrical heat flow in the sphere and its surroundings. Appropriate corrections were applied for self-shielding in the fueled region and variation of the heat-generation rate during the experiments.

The results show that the thermal conductivity of a fueled-graphite body is a significant function of temperature and that it may depend on materials and fabrication techniques. The conductivity k [in w $cm^{-1}$ $(°C)^{-1}$] of a sphere tested at an average fuel temperature T in capsule 08-7 was determined to fit the equation

$$k = 1/(0.00810\ T - 2.04)\ .$$

This sphere was prepared by hot pressing a premolded fuel insert inside a premolded unfueled shell at 750°C and baking the assembly at 1800°C. The conductivity of two other spheres, which were prepared by cementing warm-molded or hot-pressed fuel inserts into machined ATJ graphite shells, was found to be about 20 to 25% greater than that of the sphere tested in capsule 08-7. The results of the calculations further indicate that the thermal conductivity of fueled graphite is unaffected by thermal-neutron irradiation at temperatures above 800°C and to exposures of about $10^{20}$ neutrons/$cm^2$.

Phase Relations in the Pseudobinary System $UC_2$-$ThC_2$. — We measured the temperature coefficients of expansion of $ThC_2$ and of a series of

specimens of $(U,Th)C_2$ ranging in composition from 25.4 to 87.7 mole % $ThC_2$. These specimens were prepared either by electrical-arc-discharge machining of an arc-melted button or by cutting a cylindrical section from a drop casting. All specimens were radiographed for internal defects prior to measurement.

Dilation of each specimen was measured between 40 and 1000°C at least three times. The results, expressed as average coefficients over this temperature range, increased steadily from $8.42 \times 10^{-6}/°C$ for $ThC_2$ to $13.23 \times 10^{-6}/°C$ for a specimen containing 25.4 mole % $ThC_2$. The coefficient of a specimen containing 35.0 mole % $ThC_2$ was not significantly different from that of the specimen having 37.4 mole %, as would be expected, since both compositions are within the two-phase region of the binary system.

We also determined the lattice parameters of $ThC_2$ using an arc-cast specimen containing 0.06 wt % graphite. The specimen had been heat treated for 200 hr at 1350°C to minimize lattice strains associated with a high-temperature transformation. Analysis by computer program of the data from x-ray diffraction patterns yielded the following parameters for $ThC_2$: $a = 8.296$ A, $b = 4.232$ A, $c = 10.538$ A, and $\beta = 90.23°$. The monoclinic unit cell defined by these parameters represents a much smaller deviation from the orthorhombic cell than that reported by Gantzel and Baldwin (USAEC report GA-4839).

Fission Product Deposition. — We mounted small-diameter wires perpendicular to a turbulently flowing high-temperature helium stream containing trace amounts of iodine to determine whether the quantity of iodine deposited on the silver wires could be correlated with published experimental heat transfer data for systems having similar geometry and flow conditions. For the first array of three 0.010-in.-diam wires, we were able to predict the amount of iodine deposited to within 26% of the experimentally determined values. For three other arrays of wires farther downstream from the source of iodine, we were able to predict the amount of iodine deposited to within 45% of the experimentally determined value. We believe the poorer results downstream are due to uncertainties in determining the gas stream concentration of iodine.

Computer Programs for Shell Structure Analyses. — Structural analyses of pressure vessels are of primary concern in reactor containment design. In recognition of this fact, we have been working on the development of computer programs for the analyses. The two most useful codes thus far developed are the Unified Shell Analyzer (USA code) and the CERL-II.

The USA code will analyze a pressure vessel composed of several different types of elemental shell structures, for example, a cylindrical vessel with a spherical head and a toroidal transition. The edges of any elemental shell may be close or remote, and each shell can be joined to any logical combination of shells around it. Boundary conditions may be applied to any or all edges. The present code accounts for pressure, axial thrust, and thermal loadings. Complete stress and deformation data are included in the output. The final version of the code will include 25 different configurations. Eleven of the configurations are currently operational.

The CERL-II program is a code for calculating the stresses in a hemispherical shell with a radially attached cylindrical nozzle. The code will compute stresses in both the shell and the nozzle for any one of four loadings. Analyses based on axial symmetry are adequate for internal pressure and axial thrust on the nozzle; however, for transverse thrust on the nozzle and bending applied to the nozzle, asymmetrical analyses are required. The nozzle may extend into the vessel. The code is especially useful in large-parameter studies.

## REACTOR FUELS AND MATERIALS

Radiation Effects in Structural Materials: Stress Dependence of Dislocation Velocity in Iron. — Measurements have been made of the stress dependence of the dislocation velocity and the density of mobile dislocations in tensile samples of iron by applying a stress relaxation technique. The usual expression for the stress dependence of the velocity was found to be inconsistent with the observations, and it was necessary to introduce a threshold stress to the expression. When this was done, the observed stress relaxation data consisted of the motion of two species of dislocations corresponding to two distinct indices of the stress dependence m. The values of m determined this way remained invariant throughout plastic deformation. It was concluded that the two species represent the edge and screw dislocation segments.

Materials Compatibility Studies. — We are continuing to study the effect of oxygen on the compatibility of niobium with liquid potassium. Metallographic examination of niobium coupons containing at least 1000 ppm oxygen after exposure at 600 and 815°C to potassium containing 50 to 2600 ppm oxygen revealed two distinct features in addition to the surface corrosion-product layers previously reported. One phenomenon was the presence of numerous surface protuberances covered by the corrosion products. The protuberances were generally circular with a maximum diameter of 0.003 in. and a height of 0.0005 in. A study of cross sections of these projections showed that they resulted from the other phenomenon, a subsurface second phase that extended as deep as 0.007 in. below the surface of the niobium specimens. Small inclusions did not always cause protrusions. Apparently the surface bulged to relieve the growth of the subsurface products. Hence the second subsurface phase is probably less dense than the matrix. In confirmation, x-ray diffraction analysis with a microfocus camera identified the subsurface corrosion product as $NbO_2$.

Fuel Element Development. — Unaided sintering of uranium monocarbide to high densities is difficult even at excessively high temperatures. Our continuing work on sintering aided by $U_3Si_2$ reveals this to be a most promising method of fabricating carbide. Best results (>97% of theoretical density at < 1600°C) are obtained with the inexpensive UC powder derived from $UO_2$, and densification is not adversely affected by small variations in carbon content. The sintering mechanism for stoichiometric UC powder as charge appears to consist of sintering activated by silicon vapor, followed by liquid-phase sintering, with eventual dissolution of

the additive in the matrix. When the carbide powder contains much oxygen or excess carbon, the dicarbide secondary constituent present in the powder reacts with the $U_3Si_2$ additive to form grain-wetting liquid phases, ensuring good densification.

Behavior of High-Temperature Materials Under Irradiation: Magnesium Oxide Series of Irradiation Experiments. — Postirradiation examinations of MgO specimens irradiated in experiments 41-15 through 41-22 are nearing completion. X-ray diffraction studies of MgO samples irradiated at ~150°C revealed very little lattice parameter expansion. The fractional increase of the "a" parameter, $\Delta a/a_0$, was found to be about 0.0005 for samples irradiated to doses greater than $10^{21}$ neutrons/cm$^2$ (>1 Mev). The gross expansion of specimens irradiated under these conditions was about 1%, but the portion contributed by the expansion of the lattice parameter is only 0.15%. There is essentially no lattice parameter expansion of MgO irradiated at 800 and 1100°C.

Behavior of High-Temperature Materials Under Irradiation: Refractory Metal Carbides. — A low-temperature (130 to 420°C) assembly (experiment 41-23) of a series of experiments designed to determine the radiation stability of carbides of Ti, Zr, Nb, Ta, and W, translucent $Al_2O_3$ (Lucalox), and $Y_2O_3$ has been undergoing irradiation in the ETR since October 1964. The carbide samples in this assembly were fabricated by hot pressing. The assembly will be removed from the reactor flux after receiving a maximum fast-neutron dose of about $8 \times 10^{21}$ neutrons/cm$^2$.

A second low-temperature assembly (experiment 41-24) has recently been inserted in the ETR for an expected maximum dose of $4 \times 10^{21}$ neutrons/cm$^2$. This assembly contains both hot-pressed and slip-cast sintered specimens of each of the five carbides.

## FUNDAMENTAL HEAT TRANSFER AND FLUID DYNAMICS

Boiling in Multirod Geometries. — We completed a preliminary set of experiments designed to test the performance of a thin-walled ohmic-heated tube intended for use as the central element of a seven-rod cluster. This element, which was 1/2 in. in outside diameter and had a 10-in. heated length, was mounted vertically in a glass tube and exposed to an upward flow of saturated water. Wall temperatures were monitored by 20 thermocouples distributed over the outer surface and by a probe within the tube carrying two thermocouples. To examine the effect of a nonwetting surface, a 1-in.-wide band on the outer tube surface, 5 in. above the inlet end, was treated with a spray of emulsified Teflon particles; droplets of emulsion approximately 10 mils in diameter were deposited at a density of about 1000 per cm$^2$. These Teflon spots were cured in air at 800°F for 10 min. During this operation the tube metal oxidized appreciably, except near the ends, which were water cooled to prevent damage to the solder connections.

At power levels sufficient to induce surface boiling, we observed that the density of nucleation sites was moderately high on the unoxidized tube ends, very low on the oxidized portions of the surface, and very high in the Teflon-coated region, despite the oxidation. The inner

surface temperature, as mapped by the internal probe, showed no circumferential variation at any level and an axial decrease from inlet to outlet in accordance with the decreasing hydrostatic head. However, in the vicinity of the Teflon-treated band, the wall temperature remained essentially constant. This behavior was contrary to our expectation that the temperature would decrease at a faster rate in this region and perhaps exhibit a minimum. Axial wall conduction may account for the observed performance.

All but three of the thermocouples attached to the outer surface of the tube served as sites for bubble nucleation. At the nucleating position, the temperatures indicated appeared to depend upon the rate of nucleation and were observed to be as much as 11°F less than the corresponding inner surface temperatures. At the three nonnucleating locations, outer wall temperatures about 2°F below the inner surface temperatures were indicated by two of the thermocouples, and a temperature about 2°F above the inner temperature was indicated by the third. The latter result was obtained in a region where the surface had been oxidized by a near approach to burnout, and it is possible that the performance of the thermocouple had also been affected.

Vortex Dynamics: Hydromagnetic Stabilization of Vortex Flow. Experimental. — We operated the 4-in.-diam tangential-slit-fed vortex tube with an electrolyte (aqueous $NH_4Cl$ solution) at 200°F and found the peripheral tangential Reynolds modulus at the transition to instability on the cylindrical wall to be in the range from 1250 to 1500 with no magnetic field. With a peak axial field of 75 kilogauss, corresponding to a Hartmann modulus ($N_{Ha}$) of 172 at the mid-axial position (150 at the tube ends), the transition Reynolds modulus was increased to 10,350. This result is slightly below the value of 13,000 obtained by extrapolation of earlier data for 1.1-in.-diam tubes fed by two and four slits in which the maximum experimentally attainable Hartmann modulus was 49. These new data indicate that the transition Reynolds modulus varies approximately linearly with $N_{Ha}$ for $70 \leq N_{Ha} \leq 172$.

Preliminary observations of dye patterns on the end wall of the vortex tube opposite the discharge opening indicated a significant effect of the magnetic field on the undesirable, secondary, radial end-wall flow which bypasses fluid away from the interior of the tube. The suppression of this secondary flow is very strong at low values of the tangential Reynolds modulus (based on the primary flow), and the effect appears to decrease with increasing $N_{Re}$. However, uncertainties in the position of the dye-injection point relative to the wall make these conclusions uncertain. An adjustable end-wall dye-injection probe will be employed in future runs in an effort to eliminate this uncertainty.

Vortex Dynamics: Hydromagnetic Stabilization of Vortex Flow. Theoretical. — We extended the calculations for nonlinear hydromagnetic stability between insulating walls (ORNL-3788, p. 6) to a Hartmann modulus of 100, and we believe, on the basis of analogous linear calculations, that results for higher values of the Hartmann modulus can be obtained with sufficient accuracy by extrapolation. When the results are correlated by plotting the percentage increase in energy dissipation

produced by the nonlinear disturbance vs the percentage increase in Taylor modulus above the critical value, the resulting curves are changed by, at most, about 15% by increasing the Hartmann modulus from 0 to 100. Further increases in Hartmann modulus should have a negligible effect. A change in radius ratio from 0.25 to 0.8 changes the slope of the curves by about a factor of 3.

We completed the linear Couette flow stability calculations for oscillating axisymmetric disturbances at radius ratios of 0.25, 0.4, and 0.8 and at values of the Hartmann modulus up to about 300. Such disturbances begin to affect the stability when the Hartmann modulus exceeds about 20. At a Hartmann modulus of 300, the critical Taylor modulus is decreased by a factor of about 40 as a result of the oscillating disturbances.

Boundary-Layer Transient Phenomena. — In the analytical treatment of the performance of the hot-film surface probes, we used a heat-generation function based on a linear relationship between the square of the voltage to the film and the mean film temperature. This assumption was experimentally tested, and a plot of the square of the voltage against the film resistance (proportional to the temperature) was found to exhibit the expected linear dependency. At the same time, these tests established that the present probes are not sufficiently stable (the $E^2$ vs R plots have shown changing slopes and a drift in the resistance). We believe that this difficulty may derive from dimensional changes in the epoxy backing material under the action of heat and long exposure to water. New probes are being fabricated using a number of different resins and curing procedures in an attempt to eliminate this problem.

We examined the possibility of using the hot-film probe to detect the laminar-to-turbulent transition, and the results were encouraging. A relatively crude experiment yielded a value for the transition Reynolds modulus of approximately 2370 in the 2-in.-diam pipe. This test also indicated that the surface contouring of the probes was sufficiently accurate to prevent the creation of parasitic turbulence.

## THORIUM UTILIZATION PROGRAM

Thorium-Uranium Recycle Facility (TURF). — The TURF is being built to demonstrate thorium-base fuel preparation and fabrication improvements growing out of the Kilorod Program and out of development work to be done in a facility now being equipped for preparing oxide and carbide microspheres. The TURF will permit process demonstration of the entire fuel cycle at high radiation levels. The four operating cells that will make up the facility are each 20 ft wide and have a combined length of 106 ft. The walls are to be made of normal concrete, 5-1/2 ft thick.

Bids for the construction were opened on March 25, 1965. Blount Construction Company, the low bidder, bid $3,240,000, which is about $200,000 in excess of the engineer's estimate. ORNL's participation in the project has been carefully reviewed and reestimated to ascertain the minimum expected cost. As a result of this careful study, it was possible

to reduce the estimated cost of certain items and to lessen the contingency allocated on various items. The total estimated ORNL outlay, exclusive of cost of making and installing the oxide fabrication equipment, is $2,269,200. With this amount, and with the anticipated expenditures and already incurred costs for other items, the funds remaining for the fixed-price contract are adequate for building the facility and providing a contingency sum of about 7%.

A deviation request was issued for deletion of the process-waste retention tanks. This was done in compliance with the wishes of the Plant Waste Disposal Department and will provide for common use of the TRU retention tanks by both the TRU and the TURF. Each facility will measure its quantity of waste and monitor it for radioactivity, and each system has an automatic device to switch the flow of liquid from the TRU retention basins to HFIR basin 2.

Procurement of various items to be supplied by ORNL is proceeding generally according to schedule. A minor change was made in the design of the frame for the alpha-seal unit of the viewing windows. The basis for the change was development work which showed that stronger pins were needed to support the units.

Work was continued on the preparation of in-cell equipment layouts. The items being given greatest consideration at present are those that pertain to the uranium-storage and sol-preparation equipment. Layout of the sol dryer and other solids treatment and handling equipment will be made when the exact configuration and size of the equipment is definitely determined. The equipment is being located such that its accessibility for remote operation and maintenance is optimized.

Sol-Gel Equipment. — A continuously fed vertical-tube calciner for converting thoria gel to dense oxide is being developed for use in the TURF. Emphasis has been on developing operating procedures acceptable for remote operation. A device for detection of the gel level at the top of the furnace was installed and tested. It gives a sensitive indication of level changes and may be adequate to detect gel bridges rapidly and to control a feed apparatus. In early tests it was found that bridges formed if the product-removal mechanism was turned off at 1150°C, but that they did not form when the static bed temperatures were kept below 950°C.

The solids sampler for the fired oxide stream from the vertical-tube furnace was tested and appears to give a representative sample. The initial model of the sol sampler, in which a ball valve is used, was not satisfactory because of a lack of complete closure between the flow-through and the sample-discharge positions.

Fueled-Graphite-Process Development. — The study of fluidization of iron shot in a 3-1/2-in. Plexiglas column has been continued in an effort to find ways to decrease the cycling time of particles. It is believed that in fluidized-bed coating furnaces the particles are coated only at the bottom part of the chamber, where the coating gas is introduced. The coating efficiency can be increased by bringing the particles back to the coating zone more rapidly.

With respect to rotating-drum-model experiments, the presence of doublets and of agglomerates in most batches of coated fuel particles

indicates insufficient agitation. We are attempting to increase this agitation by placing fins and baffles in the plastic models. A 9-in.-ID Plexiglas cylindrical model is used, in which holes have been drilled to facilitate installation and change of baffles. To measure the degree of mixing, a large number of samples were taken from various locations, and a statistical analysis of the results is being made.

Detailed design of the components of the prototype drum coater continues. This coater will be used for depositing pyrolytic carbon on sol-gel microspheres. Some component drawings were accepted, and a work order for fabrication was issued. Contamination control during maintenance work dictates that we must include a design for an alpha-control enclosure.

With regard to the TURF fueled-graphite fabrication process, the preliminary process flow diagram for such a fabrication line, which could be installed in the TURF, was extensively revised. The primary reason for the revision is the addition of process steps to bind coated microspheres into pellets. Also, a high-temperature gas-cooled-reactor fuel element, modified for remote fabrication, was conceived and drawings were prepared. Conceptual equipment layouts based on the process steps and fuel-element configuration have also been prepared.

An effort is being made to find ways of remotely controlling fluidized-bed particle-coating devices. The principal problems consist in (1) measuring and controlling the degree of fluidization in a coater and (2) determining when to terminate coating runs. In previous work, a resonating device that measures the degree of bed fluidity was developed. Such a device will be made for evaluation. In addition, other methods that might be used to measure and control bed fluidity, such as gamma scanning, sonic transmission devices, and pressure measurement, will be investigated. We will also investigate methods such as sampling and measuring coated microspheres by means of air gaging, electronic gaging, projection microradiography, and screening to determine when to terminate coating runs. The investigation will include methods for on-stream monitoring of the size of the coated particles.

Complete conversion of $ThO_2$ to $ThC_2$ occurred when a 20-g batch of 200-μ sol-gel $ThO_2$ microspheres coated with 19.7 wt % of porous pyrolytic carbon (36.5% open porosity) was heated (without the addition of carbon flour) at 2100°C for 6 hr while rotating in a graphite crucible at 44 rpm. Agglomeration (sintering) of the particles was severe, as it was in a previous experiment where the coated particles were heated at 2150°C for 6 hr. An appreciable quantity of coating remained on the particles after the conversion, and the fuel particles protruded from the excess coating, suggesting an exaggerated "amoeba" effect. This approach to conversion, however, has considerable merit, particularly for maximum yield of fully converted particles and a minimum quantity of carbon.

Complete conversion was obtained in 300- to 400-μ sol-gel $ThO_2$ particles after 6 hr at 2150°C during rotation at 44 rpm with 1:1 and 0.2:1 (approximately stoichiometric) carbon-to-thoria ratios. The microstructure of the latter was distinctly unlike that of previously converted particles. No oxide was observed metallographically in the particles, but not all the particles showed the typical twinned dicarbide structure normally seen; some did not react to polarized light and were stained by the etchant, unlike the monocarbide or dicarbide.

Sol-Gel Microsphere Preparation. — Process Development studies were continued for the preparation of thoria microspheres by dispersing sols into drops, extracting water (into an organic solvent) to gel the drops, and drying and calcining to give the oxide microspheres. A 100-hr run was completed, including drying, calcining, and sieving of the product. The sol feed rate was 280 cc/hr, or 230 g of $ThO_2$ per hr. A large number of solvent and gel microsphere samples were taken for analyses and for tests to determine the effects of long-term use of the solvent. A total of 14.9 kg of thoria microspheres, including 9.3 kg of 210- to 250-μ-diam product from the run, was made available for pyrolytic-carbon-coating studies.

A single charge of 2-ethylhexanol and 0.5 vol % Amine 0 solvent was used during the run, with distillation to remove water and addition of 0.1 vol % Span 80 every 24 hr as the only solvent treatments. This continued use of the solvent produced no obvious deleterious effects. During the first 24 hr, the amine concentration (from the Amine 0) decreased from 0.0100 M to 0.0080 M, while the nitrate concentration increased from 0 to 0.0080 M. During the final 72 hr, both the nitrate and the amine concentrations decreased approximately exponentially to 0.0012 M at the end of the run. These results indicate that the extraction of nitrate into the solvent is due to reaction with the amine. The amine nitrate is apparently lost by a first-order mechanism, which may be adsorption on the thoria droplets or a decomposition reaction.

Sintering and Grain Growth of Sol-Gel $ThO_2$ and $(Th,U)O_2$. — Thermogravimetric (TGA) and differential thermal (DTA) analyses in static air were made on crushed samples of thoria gel that had not been formed into microspheres and so contained no residual organic material. The crushed gel yielded well-defined rate-of-weight-loss peaks at 80°C (water loss) and at 430°C (decomposition of residual nitrate), with corresponding DTA endotherms. There was also a small rate-of-weight-loss peak at 560°C, with a corresponding DTA endotherm. This may be due to the removal of more strongly bound water.

Microspheres gave very complex DTA and TGA patterns. In addition to the water-loss and nitrate-loss peaks found with the crushed gel, there were at least three strongly exothermic rate-of-weight-loss peaks in the range 180 to 350°C. These are believed to be associated with various stages in the oxidation of residual organic material. This oxidation appears to be complete at about 400°C.

In addition to characterization of the materials, another reason for the DTA's was to see whether the decrease in crystallite surface area during heating of the gels would be sufficiently pronounced for the accompanying energy release to give an identifiable exothermic DTA peak. The surface area of 51.2 $m^2/g$ for the microspheres corresponds to a total surface energy of about 5 cal/g. This is comparable with the energy absorbed by the water-volatilization reaction (about 9 cal per gram of gel) or the nitrate-decomposition reaction (about 13 cal per gram of gel) in the crushed gel, and both of these reactions gave well-defined DTA endotherms. Thus, if most of the surface energy were released over a fairly narrow temperature range (for example, about 50°C), there would be an identifiable DTA peak. No such exothermic peak has yet been found. This

indicates either that the surface-energy release occurs over a wide temperature range or that any major release coincides with an endothermic reaction, such as the decomposition and release of residual nitrate.

Temperature calibration of the hot-stage microscope is nearly complete, and shrinkage runs on microsphere specimens should commence in the next month.

Irradiation of Thorium Fuels. — The microstructures of rods irradiated to the highest levels of burnup yet achieved with vibratorily packed fuels were examined and compared with those of similar capsules in the MTR-1 series. The diffusion-controlled changes in microstructure are quite apparent. The sol-gel fuel shows the evolution of a distinct equiaxed grain structure in the central region with increased irradiation time. From submicroscopic grains, the size has increased to 6 μ at 375 full-power days of exposure (burnup of 40,000 Mwd per metric ton of Th + U) to a maximum of 60 μ at 691 full-power days of exposure (81,000 Mwd per metric ton of Th + U). A more distinct boundary for equiaxed grain growth can now be defined with an

$$\int_{T_0}^{T_{eg}} k \, d\theta$$

of 31.5 w/cm. Since such phenomena depend on time as well as temperature, this somewhat lower value at longer times is the result of the improved structural definition and therefore easier detection.

Examination of the high-heat-rated rods (ETR-1) is continuing; fission-gas samples were obtained from the fourth, and last, rod in this group — a binary size mixture of air-fired fuel. The quantity of gas was high, like that for the ternary size mixture of the same fuel, indicating a possible influence of calcining atmospheres on the release rates. Exact percentages will depend on the gas analysis and burnup. The rod was sectioned and showed a central void the full length of the fuel.

A possible explanation for the failure to produce central melting in these high-heat-rated rods may have been found in a review of the plot of initial exposure cycle vs reactor power. During the first 9-1/2 hr of operation, the reactor power never exceeded 50%. The resultant flux level was sufficient to produce central temperatures in these rods high enough for columnar grain growth. Since the phenomenon is a rapid process and produces densification, a central void, and improved thermal conductivity, the central temperatures at full power could have been reduced accordingly.

Additional burnup samples were obtained and will be analyzed. The high-magnification microstructure examinations have not yet been made.

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

Fabrication of rods containing pure $ThO_2$ for testing in ETR group III was completed. Seven Zircaloy-2 tubes were each filled with about 310 g of sol-gel $ThO_2$. Bulk densities of about 89% of theoretical were obtained by vibratorily packing the tubes on a modified, spring-loaded NAVCO BH 1-5/8 pneumatic vibrator.

The crude material was jaw crushed, and a yield of 58 wt % was obtained for the −6 +16 mesh fraction. However, this fraction contained a large amount of flaky material, which is not desirable for vibratory packing. Therefore, the −6 +16 mesh fraction was ball-milled for 10 min, and an additional 12% of the fraction was lost. About a third of the remainder of the fraction was vibratorily packed, and additional breakup occurred.

Fabrication and preirradiation inspections are complete on these rods, which will be shipped to the reactor. Gamma scanning showed a variation in packed density along their length.

Clad-Oxide-Fabrication Development. — Experiments are in progress to investigate the role of radial acceleration of the tube wall in the compacting process. In view of difficulties in mounting accelerometers on tube walls, it now appears that we will have to be satisfied with relative rather than absolute measurements.

Apparently there is a correlation between bulk density and the rms value of the radial acceleration. There also appears to be a sharp threshold of rms values of the radial acceleration, above which high bulk densities are obtained.

As to the fabrication of sol-gel oxide bodies, $ThO_2$—8 wt % $UO_2$ pellets were sintered and evaluated. Similar pellets were divided into two sets and sintered to 1450°C for 2 hr, one set in air and the other in hydrogen. The results show that pellets of comparable isostatically pressed densities did not sinter as well in hydrogen as they did in air. Also, a lower density (8.8 g/cm$^3$) was obtained with this oxide than with $ThO_2$—3 wt % $UO_2$ (9.3 g/cm$^3$) under similar fabrication conditions.

Pellets from the $ThO_2$—8 wt % $UO_2$ are to be fabricated and sintered at higher temperatures (up to 1750°C) in an effort to reach densities of at least 9.0 g/cm$^3$.

Process Demonstration and $^{233}U$ Distribution. — Oak Ridge National Laboratory serves as a material storage and distribution point for $^{233}U$. A processing facility is being installed in the Pilot Plant (Building 3019) to permit cleanup and packaging of $^{233}U$ in various forms as requested by users.

About 17 kg of $^{233}U$, containing 250 ppm of $^{232}U$, was received and stored in the dry storage holes in the Pilot Plant penthouse. This completes the present series of shipments. Eight dissolvings of $^{233}U_3O_8$, comprising a total of 3.4 kg of $^{233}U$, were completed during the month. This completes feed preparation for an 18-kg processing commitment.

The new containment and shielding walls for the $^{233}U$ solution storage facility were constructed during the month. Installation of the tanks and piping will begin shortly.

Gas Evolution from Sol-Gel Thorium-Base Oxide Fuels. — A more precise knowledge of the moisture-adsorption characteristics of thoria and thoria-urania fuels would be useful in the specification of fuel-production standards. Moreover, the minimization of the moisture content would tend to reduce the aqueous portion of the gas pressure in the sealed fuel elements under reactor operating conditions. Finally, there is a suggestion that occluded water tends to react with some of the gases evolved from thoria, with the subsequent reduction of the available surface area.

In view of the foregoing factors it was considered advisable to begin, along with the gas-evolution studies, auxiliary isopiestic studies on the moisture-adsorbing characteristics of thorium sol-gel material. For this preliminary survey, three samples of sol-gel thoria (mesh size ≤270) were equilibrated with water at varying vapor pressures as well as at different equilibrium temperatures, the latter ranging from 100 to as high as 176°C.

Several trends are apparent: (1) accurate, reproducible results can be expected for equilibrium periods of three days or longer; (2) the amount of water retained by the sol-gel thoria represents a significant fraction of the total weight, ranging as high as 9%; (3) raising the equilibrium temperature does not materially alter the ability of the gels to bind water, thereby indicating a strong solid-liquid bond (in the present series, the samples tend to retain slightly more water at 176 than at 100°C).

Coated-Particle Development Laboratory. — Enclosures for equipment to be installed in the Coated-Particle Development Laboratory have been received and are being installed by the cost-plus-fixed-fee contractor. The contractor expects to complete his portion of the work shortly. Installations of the Astro coating furnace and associated equipment in the enclosure will begin at this time. Microsphere-forming equipment is now being installed.

## MOLTEN-SALT REACTOR PROGRAM

Molten-Salt Reactor Experiment. — The advanced training session was completed, and the operators and supervisors were given comprehensive written examinations.

During the month the reactor design and the plans for operation were reviewed by three committees: the ORNL Reactor Operations Review Committee, the AEC Advisory Committee on Reactor Safeguards, and a reactor safety committee from the AEC Oak Ridge Operations Office. The ACRS recommended addition of a period scram for added protection against unforeseeable incidents related to the fluid state of the fuel. This system is being designed. Other recommendations were confined to procedures.

The mechanical work in the drain tank cell was completed, the tanks and piping were heated, and the charging of fuel carrier salt was started.

The construction of the chemical processing facility was completed, and, after calibrations and preliminary testing, the line from the reactor drain system was completed. The flush salt, which had been used in prenuclear testing, was transferred into the facility for $H_2$-HF treatment, the purpose being to remove oxides and to provide a final test of the facility.

Installation of nuclear and special instrumentation for the zero-power experiments continued.

The fuel sampler-enricher was completed and connected to the fuel pump.

The water tank in the vapor-condensing system was installed.

Development of Components and Systems. — Installation of the fuel pump sampler-enricher system was completed except for the radiation shielding. Equipment shakedown tests and the checkout of the sampling-enriching procedure are under way and will be completed in time for sampling during the critical experiments.

Alterations to several of the freeze valves in the fill and drain systems were completed. In the coolant salt system, tests had shown that the drain valve would not consistently thaw on a power failure unless emergency power was used, so it was replaced with a valve of the type used in the fuel salt system. The cooling air flow to all the freeze valves was increased from 15 scfm to greater than 25 scfm by enlarging the supply lines. The valves should now perform as originally planned.

The flowmeters in the sample lines of the two Meeco moisture analyzers used in the helium purification system were calibrated for helium flow. The results indicated that the flow settings during the previous tests on the model W-Vac have been higher by about 100% than the settings on the model W. This error has been attributed to the square-root type of pressure correction which was made but which does not apply in the region of flows as low as 100 cc/min. When the model W-Vac flow was corrected in accordance with the calibration, moisture readings on the two meters were found to agree within 0.5 ppm or less in the 0- to 10-ppm range.

The viewing equipment that will be used to inspect the inside of the reactor vessel through the 2-7/16-in.-diam graphite access plug was fabricated and tested. This equipment includes the lighting required and a sheath for containing the glass of a broken bulb as well as for preventing the contamination of the periscope. It is planned that this equipment will be used during the pre-power maintenance period following the critical experiments.

MSRE Pump Program. — Assistance with the operation of the MSRE pumps was continued. During the final cold shakedown of the spare rotary element for the MSRE coolant pump prior to delivery to the MSRE, indications of roughness were observed in the recorded trace of power. These indications are thought to originate in the motor-generator system that supplies the test drive motor, and the matter is being investigated.

The spare rotary element for the MSRE fuel pump is being operated in the prototype hot pump test facility. The radiation densitometer for determining the concentration of undissolved gas in the circulating molten salt was installed, and data were taken. Preliminary analysis indicates that the concentration of bubbles in the circulating stream is about 1-1/2% by volume with the normal level in the pump tank and that it increases to 5% as the level is lowered.

The Inconel centrifugal pump (PKP type), which utilizes a bellows-mounted shaft seal identical to that used in the MSRE pumps, was started up and is circulating molten salt at 1200°F.

The transparent mockup of the reservoir for the MSRE lubrication system was assembled and subjected to preliminary tests. It is now ready to evaluate the effectiveness of flow passage baffles which will be used to eliminate the entrainment of gas in the circulating lubricant.

Instrumentation Development. — Design, development, and fabrication of an ultrasonic level probe system for the MSRE fuel storage tank were completed. Installation of the probe system at the MSRE is nearing completion. Preliminary test results indicate that the system should work satisfactorily. Final testing of the system will be performed when salt is transferred to the fuel storage tanks.

The design of the MSRE system is essentially the same as that of a prototype that was successfully tested in the MSR Instrument Test Facility; however, the probe is of heavier construction to compensate for the high corrosion rate expected during operations of the fuel processing system. This imposed problems which required redesign of the probe and a change in operating frequency from 50 kc to 25 kc. Basic design work on this probe system was done by Aeroprojects, Inc., as an extension of similar work being done on an AEC contract. ORNL participation included establishment of criteria, review of the design to ensure conformity with MSRE design criteria, fabrication of components requiring special materials or techniques, and installation and testing of the completed system.

Developmental assistance to MSRE operations and in checkout and modification of MSRE instrumentation is continuing.

## NUCLEAR SAFETY

Release of Fission Products During Melting of Fuels Under Transient Reactor Conditions. — Radiochemical analysis was completed for the two experiments with transient-heated fuel, irradiated to a burnup of 10 Mwd/ton in 1000-psia steam. These experiments were designed to permit release of the high-pressure steam to a condenser immediately after the reactor transient. The condensate was collected in water traps, and noncondensable gases (hydrogen, xenon, etc.) passed through a series of membrane filters and charcoal-filled papers into a gas-collection autoclave. Twenty minutes later, argon gas was passed slowly through the fuel-containing autoclave to completely sweep out the remaining aerosol.

The first experiment of this type contained stainless-steel-clad $UO_2$, and the complete cycle of reactor transient, steam release, and argon purge was completed. The cladding melted, oxidized heavily, and adhered to the $UO_2$ pellets, which remained intact and unmelted. The distribution of fission products is shown below:

| Location | Percentage in Each Location[a] | | | | |
|---|---|---|---|---|---|
| | $^{131}I$ | $^{129}Te$ | $^{137}Cs$ | $^{89}Sr$, $^{140}Ba$ | $^{95}Zr$, $UO_2$, $^{144}Ce$, Ru |
| $UO_2$ fuel + SS cladding (SS cladding) | 99 (5.3) | 99.6 (1.1) | 99.9 (2.1) | 99.99 (0.12) | 99.998 (0.1) |
| Alumina heater (400°C) | 0.5 | 0.3 | 0.06 | 0.0014 | 0.0006 |
| Fuel autoclave (300°C) | 0.046 | 0.004 | 0.006 | 0.002 | 0.0004 |
| Steam released from fuel autoclave | 0.16 | 0.09 | 0.012 | 0.003 | 0.0001 |

[a]In each of the last two columns, the distributions of the species listed were determined individually, were found to be nearly identical, and were averaged for this table.

Of the $^{131}I$ carried out of the fuel autoclave in the steam release, 79% was found in the condensate, 18% in the walls of the condenser and water traps, 0.06% on the membrane filters, and 2.5% in the charcoal-filled papers. An identical filter pack was tested out-of-pile in order to determine the iodine adsorption characteristics under the TREAT experiment conditions; it was determined that the $^{131}I$ which reached the filter pack in the TREAT experiment was a "penetrating" form and was not elemental $I_2$. Metallographic examination showed no evidence of reaction of $UO_2$ with stainless steel or stainless steel oxide.

The second experiment with 1000-psia steam used Zircaloy-2 cladding. In this experiment the steam was not released, due to plugging of the 1/16-in.-ID condenser tubing with a material which is being investigated for identification. The cladding and the $UO_2$ were fragmented and dispersed within the $Al_2O_3$ crucible. The cause of the fragmentation is not known, but heavy oxidation greatly weakened the cladding. The maximum $UO_2$ temperature was approximately the same for both steam experiments. Loss of grain boundary integrity in the $UO_2$ occurred in the Zircaloy-2 experiment but not in the stainless steel experiment. Metallographic examination showed regions where Zircaloy-2, $ZrO_2$, and $UO_2$ had merged in a fluid state. The distribution of fission products in this experiment is shown below:

| Location | Percentage in Each Location[a] | | | | |
|---|---|---|---|---|---|
| | $^{131}I$ | $^{129}Te$ | $^{137}Cs$ | $^{103}Ru$, $^{106}Ru$, $^{89}Sr$, $^{140}Ba$ | $^{95}Zr$, $UO_2$, $^{144}Ce$ |
| $UO_2$ + Zr-2 cladding (Zr-2 cladding) | 90 (2.5) | 95 (11) | 77 (5) | 97.7 (10) | 98.9 (7) |
| Alumina heater (400°C) | 4.4 | 2.6 | 21 | 2.1 | 1.7 |
| Fuel autoclave (300°C) | 5.7 | 0.4 | 1.1 | 0.2 | 0.11 |

[a]In each of the last two columns, the distributions of the species listed were determined individually, were found to be nearly identical, and were averaged for this table.

Based on present knowledge, the greater release of fission products from fuel and cladding in the Zircaloy-2 experiment must be a result of the fragmentation.

A previous experiment used stainless-steel-clad $UO_2$ in 45-psia argon, and the $UO_2$ reached approximately the same maximum temperature (2600 ± 200°C) as in the 1000-psia steam experiments. Although the $UO_2$ had no previous irradiation, the release of mixed Br, Kr, I, and Xe averaged 23% even though the $UO_2$ remained intact. Thus at this time a very significant decrease in fission product release under transient heating conditions must be attributed to the high-pressure steam atmosphere.

Removal of Fission Products from Gases Under Reactor Accident Conditions. — The removal of methyl iodide (and other alkyl iodides) from

moist air systems is under study. Removal from dry air systems is not a problem. Possible methods for accomplishing adequate retention of $CH_3I$ by adsorbers in gas systems are: (1) control of the humidity of the gas system, thereby keeping the moisture content of the activated charcoal bed at a low level; (2) application of a material or combination of materials which specifically react with $CH_3I$ and render it immobile; (3) chemical modification or decomposition of the $CH_3I$ and adsorption of the resulting product(s). Humidity control as a solution to the problem has received some study. Tests with both radioactive and normal $CH_3I$ have been made using gamma spectrometry and gas chromatography for determining efficiencies. It was demonstrated that steam can be removed from a mixture of $CH_3I$, steam, and air by steam condensers and that the $CH_3I$ can be removed from the moderately dry air stream (relative humidity ~25%) by activated charcoal. A material which will remove $CH_3I$ (and higher alkyl iodides) from a gas stream, as activated charcoal does elemental iodine, would be preferable in reactor safety applications. Initial efforts are being made to survey possible materials and to develop a material, if necessary, which will react with or adsorb methyl iodide from moist air streams. Activated charcoals have been impregnated with or treated with various organic amines, oxidizing agents, and inorganic compounds and tested for methyl iodide retention. Only partial success can be reported at this time.

Behavior of Fission Products on In-Pile Destruction of Reactor Fuels. — Radiochemical analyses have now been completed for in-pile experiments 19, 20, and 21, which simulate reactor accidents of the loss-of-coolant type. The fission product release results for these experiments are shown in the table. The results for experiments 17 and 18 are shown for comparison.

Material Released from High-Temperature Zone of the Furnace in In-Pile $UO_2$ Melting Experiments (%)

| Experiment | Atmosphere | $^{131}I$ | $^{132}Te$ | $^{137}Cs$ | $^{106}Ru$ | $^{89}Sr$-$^{140}Ba$ | $^{95}Zr$-$^{144}Ce$ |
|---|---|---|---|---|---|---|---|
| 17 | Steam-He-$H_2$ | 73 | 87 | 30 | 1.2 | 3.6 | 0.47 |
| 19[a] | Steam-He-$H_2$ | 73 | b | 45 | 0.11 | 0.68 | 0.08 |
| 21[a,c] | Steam-He-$H_2$ | 51 | b | 67 | 0.01 | 0.12 | 0.01 |
| 18 | Moist air | 94 | 81 | 73 | 11. | 1.4 | 0.46 |
| 20 | Moist air | 97 | 39 | 59 | 10. | 0.90 | 0.10 |

[a]High-burnup fuel.

[b]Data incomplete.

[c]Intermediate-temperature experiment.

The conditions of experiment 19 were similar to those of experiment 17, except that the fuel in experiment 19 had been preirradiated to a burnup

level of 21,000 Mwd/ton. The conditions of experiment 21 were similar to those of experiment 19, except that the maximum temperature of the fuel in experiment 21 was only about 2000°C. Experiments 17, 19, and 21 are typical of all experiments in the absence of air, in that the releases of ruthenium and iodine are relatively low. Oxidation of these materials probably accounts for their high releases in air atmospheres, as shown by the data of experiments 18 and 20. The radiochemical data for the fibrous filter analyzer and composite diffusion tube from experiment 20 are being interpreted to obtain information about the forms of the fission products.

Experiment 23 has been performed to investigate the desorption of iodine from the apparatus in the hot cell after in-pile melting. Such desorption has been observed in recent experiments and may account for the low inventories of iodine which are generally observed. This phenomenon may also cause redistribution of iodine within the apparatus. To investigate this phenomenon the apparatus of experiment 23 was selectively cut in the hot cell after the $UO_2$ melting was performed, and a composite diffusion tube followed by a charcoal trap was attached to each major component. Helium was then passed through each system for 24 hr to cause desorption. Analysis of the diffusion tubes is in progress, but preliminary data indicate by the presence of iodine in the diffusion tubes that desorption of iodine did occur.

Characterization of Gas-Borne Fission Products. — Some aspects of the reactivity of the nonelemental vapor forms of radioiodine produced by the dichromate method have been investigated with composite diffusion tubes. The nonelemental forms include the species characterized by its tendency to deposit on rubber, designated the R species, and a more volatile substance, probably methyl iodide. Iodine was introduced into a stream of moist air at 25°C; it was then subjected to a temperature of 115°C in a quartz tube for about 1 min and, further downstream, to this same temperature in a silver-lined tube for about 0.1 min. Neither of the two nonelemental iodine species appeared to have been permanently altered during transit in the quartz tube. Indications were, however, that both species react appreciably either directly or indirectly (i.e., after dissociation or hydrolysis) with silver at 115°C. Estimated extents of reaction with hot silver under the conditions employed are ~50% for the R species and ~25% for the more volatile species.

Preparation of High-Specific-Activity Methyl Iodide for Use in Testing Charcoal Absorber Systems. — Methyl iodide has been found in aerosols produced during simulated reactor accident experiments and in radioactive iodine sources produced by a variety of methods. This compound is poorly retained by presently available trapping methods. Therefore, pure radioactive methyl iodide is needed to test the retention efficiency of trapping materials for this potentially important material. Production of $CH_3{}^{131}I$ with high specific activity by the isotopic exchange reaction between $Na^{131}I$ and $CH_3{}^{127}I$ in 95% ethyl alcohol has been studied. The procedure involved adding carrier-free $Na^{131}I$ to an alcoholic solution of microgram amounts of $CH_3I$ and extracting the product into n-decane after diluting the alcohol with an equal volume of water.

After equilibrating the mixture for 12 hr at room temperature with a concentration of 0.25 mg of $CH_3I$ per ml of ethyl alcohol, 50% of the iodine activity was found to be in the organic ($CH_3I$) phase. With 0.025 mg of $CH_3I$ per ml, the exchange process took about 13 days to attain 50% completion. Tests at both 90 and 150°C showed an enhancement in the exchange rate over the rate at room temperature, but also indicated that some thermal decomposition of the $CH_3I$ occurred. It was also shown that the $CH_3I$ could be separated from small quantities of n-decane by sparging the solvent with air and then trapping the decane with a dry ice—acetone bath (−80°C). The purity of the methyl iodide product was 99.9+%.

Simulation of Reactor Accident Aerosols. — Three meltdown tests have been completed in the Containment Mockup Facility (CMF) to characterize the behavior of iodine and other fission products in the presence of aerosols produced from a simulated $UO_2$—fission product mixture. The primary importance of such tests is to provide increased confidence in the use of simulants in larger test facilities, such as the Nuclear Safety Pilot Plant, the Containment Systems Experiment, and the Loss-of-Fluid Test. An appraisal of sampling devices was also an important part of the study.

One fuel specimen (run 12-9) was clad with stainless steel; two (runs 2-24 and 3-25) were clad with Zircaloy-2. Another important difference among the three runs was the atmosphere over the hot fuel sample. Air was used in runs 12-9 and 2-24, while in run 3-25 the fuel was heated in helium and steam; but the resulting aerosol was released into an air-steam mixture in all three runs. A comparison of the results with those from a previous run (10-28), which was performed with iodine tracer released by melting $UO_2$, is shown in the table below.

Distribution of Iodine in the CMF

| | Experiments with Simulated $UO_2$ Fission Product Mixture | | | Tracer Iodine with $UO_2$ Vapor, Run 10-28[a] |
|---|---|---|---|---|
| | Run 12-9 | Run 2-24 | Run 3-25 | |
| Cladding | SS | Zr-2 | Zr-2 | SS |
| Furnace atmosphere | Steam-air | Steam-air | Steam-He | Steam-air |
| Iodine, percent of inventory | | | | |
| Total remaining airborne | 7.9 | 6.3 | 9.7 | 32 |
| Retained on filters | 0.5 | 0.1 | 0.15 | 0.01 |
| Penetrating 0.75-in. charcoal bed | 0.02 | 0.02 | 0.02 | 0.5 |
| In penetrating form | 1.3 | 0.7 | 0.8 | 21 |

[a]The tank aerosol in run 10-28 contained 0.2 vol % mixed hydrocarbon gases. Aging time was 9.5 hr.

It is apparent that a much lower percentage of penetrating iodine was formed under conditions of the simulation runs than with tracer iodine and $UO_2$ vapor in the presence of organic gases. The most likely explanation for the high fraction of penetrating iodine in run 10-28 is the presence of hydrocarbons in the vessel atmosphere, but other possibilities cannot be excluded on the basis of the information presently available. The reproducibility of results in the simulation experiments was exceptionally good.

Coincident with these tests, a pressure-type gas sampler was tested and compared with a low-pressure sampler that uses a rubber diaphragm and a hypodermic needle. The low-pressure sampler had produced somewhat erratic results in earlier experiments. The airborne-iodine concentration data obtained with the pressure-type sampler were much more consistent than the data obtained with the diaphragm-needle sampler and could be fitted to a smooth deposition curve. Use of the low-pressure sampler will probably be discontinued.

Nuclear Safety Information Center. — In March the Nuclear Safety Information Center invited ACRS members, AIF safety committee members, and selected management-level personnel in the AEC and at ORNL to participate in an experimental program in the selective dissemination of nuclear safety information. The Center will use the replies of the participants detailing their specific interests in nuclear safety to establish profiles of key-word selectors for each individual. Computer programming is being developed so that the bibliographic items of interest along with the abstracts will automatically be sent to the participants after each updating (estimated once a week) of our IBM 7090 store of nuclear safety information.

The Center's first computer-sorted bibliography, Indexed Bibliography of Current Nuclear Safety Literature — 1, ORNL-NSIC-8, has been issued. Programming now under way will enable the staff to make retrospective searches of its computer files soon.

Copies of the Center's two latest publications, ORNL-NSIC-8 and Behavior of Iodine in Reactor Containment Systems, ORNL-NSIC-4, were distributed to all attendees at the International Symposium on Fission Product Release and Transport Under Accident Conditions, held in Oak Ridge in April.

In March and April the Center received and answered 18 requests for information. In the same period the staff conferred with 20 visitors.

Nuclear Safety Pilot Plant. — Preparations for run 8 involved adaptation of the sampling devices and the plasma-torch heat source for operation in the steam environment to be used in the next series of tests. The run was made April 1–3. Irradiated, stainless-steel-clad $UO_2$ was heated with the plasma torch for 8 min (2-1/2 passes over the fuel element). Although the torch operating conditions chosen were expected to produce melting, post-run examination has indicated that the fuel was heated only very close to the melting point; only a slight indication of flow of the $UO_2$ was observed. However, approximately half of each pellet was overheated sufficiently to cause gross alteration of the grain structure. The remainder (lower half) was not visibly affected.

Samples were collected with six May-pack samplers, six tape air samplers, deposition and fallout samplers, and condensate collectors. More than 1500 samples have been analyzed, principally by gamma spectrometry. The analytical work is only partially complete.

Preliminary results are as follows:

1. Gross release of volatile fission products occurred, even though the fuel was not actually melted.

2. The iodine distribution pattern on the May packs was radically different from that normally found in air atmospheres, in that most of the iodine was in the charcoal and less was on the silver screens.

3. A large amount of $I_2$ (the exact amount is being determined) passed through the May packs, and was collected on backup charcoal traps held at dry-ice temperature.

4. Several painted test coupons using coatings under consideration for the LOFT containment vessel were exposed during the run, counted, decontaminated in accordance with procedures recommended by Phillips engineers, and recounted. Decontamination factors of the order of 4 were measured.

## RADIOACTIVE WASTE DISPOSAL

Mineral Exchange Studies.— Ion exchange capacities of clay minerals are due to isomorphic substitution within the clay lattice and to ionization of hydrogen ions bound to hydroxyls on the clays. The latter type of exchange site would contribute to the capacity in solutions of higher pH and salt concentrations, particularly if they behave as weak acids. Ion exchange capacities of selected materials were determined at pH 6 and pH 9. The montmorillonite mineral, which exhibited ideal behavior as predicted by mass action laws, increased in capacity from 84 to 89 meq/100 g.

The samples of illite and pyrophyllite, which had shown nonideal behavior in sorbing radiostrontium, increased in capacities from 15 to 21 and from 2 to 4 meq/100 g respectively. The sample of pyrophyllite, which normally does not have isomorphic substitution, was tested further for "exchangeable hydrogen" by titrating the material with NaOH; a capacity of 1.5 meq/100 g was measured between pH 6.3 and pH 9. When the pyrophyllite was titrated in the presence of 1 M NaCl, the titratable acidity increased to 7 meq/100 g in the same pH interval. These initial studies suggest the need to define more accurately the capacity of mineral sorbents in different salt concentrations and at different pH's in order to define the mechanism of strontium sorption by soil minerals.

Disposal in Deep Wells. — Preparations were started for injection 6. Procurement orders were written for chemicals, cement, fly ash, attapulgite, and grundite. Arrangements were made for transporting and mixing the cement and the fly ash.

Modifications to the injection plant were started. These modifications included the installation of additional shielding to permit handling of more active waste solution, the addition of flowmeters to make possible better control of solids-to-liquid proportioning, and improvements to the sampling and off-gas systems.

The mass flowmeter proposed for use in injection 6 was tested at Duncan, Oklahoma, with dry solids. This was done in order to calibrate the meter, to determine whether various solids behaved similarly and dynamically in a mixing test, and to determine how well the meter would integrate with existing equipment. The usual difficulties were encountered, but in general it was determined that:

1. considerably better control of the solids-to-liquid ratio is possible with the mass flowmeter than with the present control system;
2. solids flow control is acceptable up to 2000 lb/min; at higher flows the calibration becomes unreliable;
3. changes in the proportions of the various solids tested had no apparent effect on flowmeter readings.

Low-Activity Wastes. — A new process, the water-recycle process, is being developed. It is based on the demineralization of all new water entering a nuclear installation and reuse of used waters which have been demineralized and decontaminated to 2% of the $MPC_W$ for the 168-hr week. Feasibility studies are being made in the laboratory with ORNL low-level waste to extrapolate to the recycle-water case. The system currently includes (1) batch clarification, using zeta-potential-controlled additions of coagulant (alum) and coagulant aid (activated silica); and (2) continuous demineralization of the supernatant by cation-anion exchange followed by sorption on granular activated carbon. Results of a run that included separate columns (beds) of cation and anion resins are given below and are compared with some results of the previously reported run, which included mixed-bed ion exchange.

Best clarification was obtained with the addition of 20 to 30 ppm alum [calculated as $Al_2(SO_4)_3$] and 0.5 ppm $SiO_2$, in that order. This corresponded to changing the zeta potential from its original range of −14 to −17 mv to +2.5 mv.

Demineralization capacity (0.2% specific-conductance breakthrough) for the separate-bed case was about 90% of that for the mixed-bed case in which 470 volumes of waste was processed per bed volume of cation resin before breakthrough. The projected demineralization capacity of 2500 bed volumes for water-recycle processing would probably diminish by a corresponding amount if split-bed ion exchange were used.

The overall decontamination factors at 0.2% breakthrough were 340 for $^{144}Ce$, 12,000 for $^{60}Co$, 2600 for $^{137}Cs$, 430 for $^{131}I$, 200 for $^{106}Ru$, 300 for $^{125}Sb$, 6300 for $^{90}Sr$, 160 for $^{95}Zr$-$^{95}Nb$, 11,000 for total rare earths, 130 for gross gamma, and 1200 for gross beta. The decontamination factors for $^{106}Ru$ and $^{60}Co$ are the highest ever attained in the ORNL development program. Breakthrough of the individual activities occurred either simultaneously with bulk ionic breakthrough or thereafter. Thus specific conductance could be used as a convenient and economical method to indicate exhaustion of a resin column.

Intermediate-Activity Wastes. — Nuclear installations generate large volumes of intermediate-level wastes, such as evaporator concentrates, second- and third-cycle solvent extraction raffinates, and slurry or

solid residues from low-level treatment processes. These wastes are characterized by their modest levels of radioactivity, so that heating and radiation dose levels are not serious problems, and by their high salt or solids contents, which prevent their efficient treatment by conventional ion exchange, precipitation, or foam separation methods. The ORNL intermediate-radioactivity-level waste-treatment program includes development of processes to remove the water from these wastes and to incorporate the dried solids in a cheap, inert, insoluble material prior to burial or contained storage. Incorporation of these wastes in asphalt or other plastic and elastic substances appears to be simple, inexpensive, and relatively insensitive to the type of material being incorporated, and, within limits, independent of local geologic and hydrologic conditions. Asphalt has good adhesive and coating properties and is insoluble in water, but its sensitivity toward radiation limits the radioactivity level of included material.

In order to evaluate the resistance of the asphalt product to irradiation, samples of the product containing 60 wt % solids from waste were irradiated by means of a $^{60}Co$ source to absorbed doses ranging from $10^6$ to $10^8$ rads. A dose of $10^6$ rads had negligible effect on the asphalt sample, but a dose of $10^7$ rads caused it to swell very slightly. A dose of $10^8$ rads caused the material to increase about 36% in volume. Leach tests on a sample irradiated to a dose of $10^8$ rads indicated that the irradiation has no effect on the leach rate.

Design of a pilot plant to incorporate 100 gal of ORNL waste-evaporator bottoms in emulsified asphalt per 8-hr shift by the above batch process has been completed, and installation has begun. "Warm-cell" and hot-cell studies to determine the long-term effect of incorporated radioactive elements on the physical (in particular, leach rate) and chemical (in particular, radiolytic gases formed) properties have been initiated.

Disposal in Natural Salt Formations. — Mining of the experimental area for Project Salt Vault has been completed. Construction of the topside waste-charging shaft facility is well under way. Drilling of gage holes and installation of plastic flow gages are nearing completion. Thermocouple holes, array holes, and pillar heater holes in rooms 2, 3, and 4 remain to be completed.

A preliminary analysis has been made of plastic flow gage data up to March 8, 1965. As expected, floor-to-ceiling convergence gages were found to be good indicators of increasing stress in pillars. It was also found that increased stresses in the existing mine pillars (due to mining of the experimental area) could be detected at distances up to about 200 ft away from the experimental area. The behavior of the roof in the experimental area appears to be similar to that in a normal salt mine operation.

The third phase of the prototype test in the existing mine floor was halted after about 11 weeks of heated operation. This phase was with no air flow in the annulus between the salt and the hole liner; therefore the moisture which comes from the shale in the floor of the existing mine was not removed. As suspected, the presence of the water

caused severe corrosion of the lower portion of the liner, resulting in what appears to be stress corrosion cracking. This type of problem is not anticipated in the operation of the demonstration or of an actual disposal facility in the purer salt.

Engineering, Economic, and Safety Evaluation: Waste Storage. — A detailed estimate of the costs of "perpetual" tank storage of wastes from the processing of power reactor fuels is being undertaken to establish a basis for comparison with costs of alternative waste-management schemes. In this study, storage of Purex and Thorex solvent extraction raffinates as both acid and alkaline solutions is considered, and storage of Zirflex and Sulfex decladding wastes in alkaline form is postulated. Tanks are enclosed in steel-lined concrete vaults for secondary containment and are similar to those at Savannah River. Decay heat is removed by circulating water through banks of coils in the waste.

The estimated capital costs of tank farms for storage of acid raffinate waste range from $21 million to $47 million, and farms for storage of alkaline raffinate cost from $27 million to $50 million. In terms of the electricity originally produced from the fuel, the costs are 0.007 to 0.015 mill/kwhr for acid waste farms and 0.009 to 0.016 mill/kwhr for alkaline waste farms. The capital cost for cladding wastes was $53 million, corresponding to 0.017 mill/kwhr.

A detailed breakdown of these costs is being incorporated in a computer code, where they will be used with allowances for operation, maintenance, amortization, and tank replacement to calculate total present-worth costs for perpetual storage.

Engineering, Economic, and Safety Evaluation: Clinch River Study. — A second interpretation can be made of the internal-dose calculations completed during the Clinch River safety analyses. As a consequence of radionuclides with long effective half-lives, and in particular $^{90}Sr$, the dose to the critical organ will continue for many years after the exposure period. Therefore, there is a dose commitment for the future, at least in part, rather than just a dose actually received during the period the individual was present in the area. Dose commitments for future years associated with the consumption of Clinch River and Tennessee River water are calculated by use of mathematical models coded for the CDC 1604-A. These are the cumulative doses that persons of various ages may receive, beginning in 1964 and extending to age 65 for each individual; they result from retention of radionuclides in critical organs due to ingestion of contaminated water during the period 1944 through 1963. The dose commitment to the skeleton of the nine-year-old who begins to drink Clinch River water in 1944 is largest, about 13 rems; for reasons previously enumerated, standard man has a smaller dose commitment, 7.6 rems. In all cases the dose commitments are well below prescribed limits. Information on dose commitment may be necessitated by a change in population exposure limits that are considered acceptable, by the desire of a new installation to utilize the diluent capacity of a surface water, or by the requirement for corrective action in the event of an accidental release of radioactive material.

# PHYSICAL RESEARCH

## PHYSICS AND MATHEMATICS

Physics of Fission. — Our earlier fission-fragment energy correlation experiment for $^{239}Pu$ thermal-neutron-induced fission has been analyzed in terms of the recently reported absolute energy calibration method. It is found that, as in the cases of $^{235}U$ and $^{252}Cf$ (ORNL-3788, p. 23), the fragment kinetic energies are significantly higher than those of the double time-of-flight experiments of Milton and Fraser. For example, the average total fragment kinetic energy from the present experiment is 177.6 ± 1.6 Mev, and that from the double time-of-flight experiment is 175.0 ± 1.7 Mev. The disagreement for masses near symmetry is ~16 Mev. Further analysis of the effects of fragment scattering from the walls of the drift tubes in the time-of-flight experiments indicates that this, in fact, is the source of the large discrepancy in all cases studied to date and reported in the literature.

Analysis of the division of energy in fission, based on the present new kinetic energies, permits comparison of empirically determined Q values for fission with those calculated from currently used semiempirical mass formulas.

Low-Temperature, Nuclear and Solid-State Physics. — We have observed the recoilless emission of gamma rays from $^{61}Ni$ in nickel metal after the 67.4-kev energy level of this nucleus was populated by Coulomb excitation. The projectiles used for the excitation were 25-Mev oxygen ions from the ORNL Tandem Van de Graaff. The Mössbauer effect observed by this method for $^{61}Ni$ was compared with previous measurements using the radioactive source $^{61}Co \xrightarrow[99\ min]{\beta^-} {}^{61}Ni^*$. The total Mössbauer absorption measured in the present experiment is equal to that observed previously; but the spectrum is wider, and the absorption at zero velocity is (74 ± 6)% of the expected value (4.05 ± 0.15)%. Measurements of the absorption at zero velocity m(v = 0) were made vs irradiation time of the target. A least-squares fit to these data indicates that there is no observable change in m(v = 0) with irradiation time. Any radiation damage in the target seems to reach saturation rapidly and to remain constant. This experiment establishes the feasibility of Mössbauer measurements using gamma rays from Coulomb excitation. Since this method is applicable to all stable nuclei with low-lying energy levels, it can be an important tool both in nuclear and solid-state physics.

High-Voltage Experimental Program: Total Neutron Cross Sections of Hydrogen and Carbon in the 20- to 30-Mev Region. — Precise n-p scattering and polarization measurements in the region 0 to 42 Mev are needed as an aid to theoretical interpretation of the nucleon-nucleon interaction, since theoretical fits to the present data are not sufficient to describe the interaction unambiguously. Measurements of the n-p differential scattering cross section in the region 20 to 30 Mev are now in progress at the Laboratory. The total n-p cross section has been measured and will be used to normalize the n-p angular distributions.

The transmissions of polyethylene and carbon were measured to determine their respective total cross sections, and a difference technique was used to extract the hydrogen cross section. Neutrons of the desired energy were produced by bombarding a T-Zr target with deuterons from the ORNL 5.5-Mv Van de Graaff and Tandem accelerators. The deuteron energy calibration was obtained relative to the $^{7}Li(p,n)^{7}Be$ threshold on the 5.5-Mv accelerator and relative to the $^{27}Al(p,n)^{27}Si$ threshold on the Tandem accelerator.

The polyethylene sample was chemically analyzed at ORNL and was found to have a hydrogen-to-carbon ratio of (2.038 ± 0.020):1 with no significant impurities. The carbon sample was fabricated from CGB grade graphite. This is an experimental grade graphite and is specified to be 99.98% pure. The scatterers were checked by x-ray photography to ensure the absence of voids and other inhomogeneities. These samples were machined in the shape of right cylinders with a diameter of 1 in. Sample length and density were measured to within 0.1%.

Stilbene scintillators served for both the neutron monitor and the detector. The detector was located at 0° and 25 in. from the target. Backgrounds measured with a zirconium blank were always less than 0.5%, while the background from room scattering was less than 0.6% for all runs. The geometry of this experiment was chosen so that sample alignment was not critical. In all cases corrections for in-scattering were less than 2%.

Preliminary results for the total neutron cross sections of hydrogen and carbon are presented below:

| E (Mev) | Cross Sections (barns) | |
|---|---|---|
| | Hydrogen | Carbon |
| 19.58 | 0.498 ± 0.012 | 1.51 ± 0.04 |
| 20.81 | 0.467 ± 0.012 | 1.43 ± 0.04 |
| 21.93 | 0.432 ± 0.011 | 1.34 ± 0.03 |
| 26.32 | 0.329 ± 0.012 | 1.32 ± 0.05 |
| 30.46 | 0.287 ± 0.011 | 1.32 ± 0.05 |

Corrections have been made for background and in-scattering effects. Neutron energies are known to better than 0.2%. The probable errors have been increased from the statistical error to include estimated maximum values of systematic errors. More exact calculations of these errors and corrections are in progress.

High-Voltage Experimental Program: kev-Range Total Cross Section Resonances in ($^{204}Pb + n$). – The location and total widths of about 20 resonances were determined in the energy range $6 < E_n$ (kev) $< 50$. The

transmission measurements were made with the ORNL 3-Mv pulsed-bunched Van de Graaff and a $^{10}$B-NaI neutron detector. In later runs we replaced the NaI with a gamma-ray detector composed of a lead glass—liquid scintillator [unpublished Harwell report AERE-R 3744 (1961)]. This technique resulted in reducing the overall time-of-flight resolution from about 4 nsec/m to about 2 nsec/m without loss in efficiency. The average s-wave spacing for ($^{204}$Pb + n) is $\langle D \rangle = 2.7 \pm 0.5$ kev, and the s-wave strength function is $S^0 = (0.55 \pm 0.2) \times 10^{-4}$. An appreciable number of small resonances are also present, probably due to p-wave interactions.

Nuclear Data. — Nuclear Data Sheets, Volume 6, Set 4 has been published. Set 5, which is ready for press, contains a 148-page collection of nuclear magnetic moments. A-chains 172 and 173 are finished. Work continues on A-chains 174 through 183. A computer program has been written to assist in selecting best spin values and multipole mixing ratios from experimental gamma-gamma angular correlation coefficients. A contract has been signed with Academic Press to create a journal devoted to compilations in nuclear physics.

Medium-Energy Nuclear Physics: Experimental. — Isotopically enriched $^{151}$Eu was irradiated, and the gadolinium activity produced by the (p,3n) reaction was analyzed with multirange magnetic spectrographs. Over 40 new transitions attributed to $^{149}$Gd were resolved up to an energy of 1 Mev. The multipolarity determinations suggest even-parity states at 534.3, 666.6, 813.2, and 939.3 kev (9/2), as well as odd-parity states at 749.0 kev (7/2) and 795.1 kev (9/2). A number of weakly excited states are designated at energies of 404.0, 672.5, 877.0, 956.9, 979.7, 1013.2, and 1097.8 kev.

The $^{65}$Cu(d,$^3$He)$^{64}$Ni reaction study was essentially completed. A comparison with the $^{63}$Cu(d,$^3$He)$^{62}$Ni data taken previously indicates that the $^{64}$Ni core contains fewer particle-hole proton configurations than $^{62}$Ni. This conclusion is based on the f:p-wave ratios required to satisfy the angular distributions for the reactions proceeding to the first excited states. For $^{64}$Ni this ratio is approximately 1:1, whereas for $^{62}$Ni it is 3:1.

Preliminary data were taken with the 34.2-Mev deuteron beam incident on a $^{51}$V target. Elastic-scattering data were collected from 10 to 60° to obtain the optical-model parameters for the incident channel of the $^{51}$V(d,$^3$He)$^{50}$Ti reaction. Data were taken for this reaction proceeding to the lowest $0^+$, $2^+$, $4^+$, and $6^+$ states in $^{50}$Ti. The $0^+$ and $6^+$ angular distributions show an L = 3 pickup, whereas the $2^+$ and $4^+$ angular distributions indicate the possible admixture of $(p_{3/2})^2$ proton configuration in these states.

Angular-distribution studies of $^{18}$F particles from the reaction $^{16}$O($^{14}$N,$^{18}$F)$^{12}$C were extended to 27- and 33-Mev incident $^{14}$N energies. The total cross sections obtained by integrating over the angular distribution are 3.6 and 11 mb at 27 and 33 Mev respectively. These values for the cross section agree well with results previously obtained from thick-target yields.

The angular distributions cover angles from ~10 to ~160° (c.m.). They are forward peaked, and each has a relative minimum and a secondary peak at angles less than 90°. At 33 Mev incident $^{14}$N energy, the differential cross section is 4 mb/steradian at 15° c.m., falls to 0.8 mb/steradian at 65°, and has a secondary peak of 1.0 mb/steradian at 80°. At

27 Mev the differential cross section is 0.5 mb/steradian at 15°, falls to 0.33 mb/steradian at 44°, and has a broad secondary peak at approximately 70° of 0.4 mb/steradian. The differential cross section being much larger for angles less than 90° c.m. indicates that the reaction proceeds predominantly by $^{14}N$ picking up an alpha particle from $^{16}O$ rather than $^{16}O$ stripping a deuteron from $^{14}N$.

The differential cross section for elastic scattering of 40-Mev protons was measured for $^{59}Co$, $^{120}Sn$, and $^{68}Zn$. The angular range from 10 to 170° was covered in 2.5° steps. Elastic scattering from $^{40}Ca$ at 45 Mev was also measured over the same range of angles.

Elastic and inelastic scattering of 67-Mev alphas from $^{90}Zr$ and $^{91}Zr$ were studied from about 11 to 70°. Excellent fits for the elastic data were found with the optical-model search program. At the same time, data were obtained on the $(\alpha,{}^3He)$ reactions for the same nuclei.

Cyclotron Operations: ORIC. — During the period of this report the Oak Ridge Isochronous Cyclotron was scheduled for 1200 hr of operation, distributed about as follows: (1) 79% to nuclear research bombardments, (2) 13% to cyclotron beam and machine development, (3) 5% to installation of additional beam optics (read-out circuitry), and (4) 3% to scheduled shutdown for routine maintenance.

The beam was "on target" 69% of the time scheduled for the nuclear research bombardments, noted in (1) above; the 31% "off target" time was used in setting up experiments and in adjusting and focusing the beam on target and by routine startup and machine shutdown, changing particles and energies, and some minor rf problems. No major component failures were experienced.

The cyclotron beam was actually "on target" for nuclear research bombardments for an average of ~11 hr/day. Extracted beams of 68- and 80-Mev alpha particles, 34-Mev deuterons, and 36-, 44-, and 48.5-Mev protons were routinely and reliably obtained.

Electronuclear Systems: Separated-Orbit Cyclotron. — Recent studies of the SOC have been directed at developing parameters for a 10- to 50-Mev multiparticle, variable-energy, high-current machine. The present concept is an SOC capable of accelerating 75 ma of protons to 50 Mev. The maximum energy of alpha particles will also be 50 Mev, and that of deuterons 25 Mev. For a machine design with a harmonic number of 24, the maximum beam radius will be 24 ft, the minimum radius 11 ft. The machine will have 12 sector magnets in a circular arrangement alternating with 11 accelerating cavities and a gap which will be used for beam extraction. A movable magnet in this gap provides for varying the energy in 2- to 4-Mev steps by intercepting the beam at the proper orbit. For this machine the cost is relatively independent of orbit separation over the range from 5 to 9.5 in.; the cost of the magnet poles and pole-tip coils goes down at about the same rate that the rf system costs go up as the voltage is increased to give larger orbit separation.

For a multiparticle machine using coaxial cavities, it is possible to choose the cavity dimension so that the required rf power is the same for protons, deuterons, and alphas. One arrangement that meets this condition is a gap center-line spacing of 0.415 $\beta\lambda$ for protons (0.830 $\beta\lambda$ for deuterons) and a gap width of 0.1 $\beta\lambda$ (protons).

Electronuclear Machines. — A study to examine the problems associated with the acceleration of heavy ions in the Oak Ridge Isochronous Cyclotron has resulted in an extracted beam of 43-Mev $^{12}C^{3+}$ ions. This beam was measured and verified by magnetic analysis and by means of a scattering experiment and de/dx measurements. The beam current obtained is still too low for practical use, <5 nanoamperes. Since it is limited mainly by ion source output, an investigation of ion source configurations for higher output of heavy ions is under way.

In an effort to extend the available energy range of extracted protons, the valley-coil magnet system was energized to investigate the effect on extraction efficiency. It was found that the position of the lower magnetic channel was not the optimum for extraction of 36-Mev protons when the valley coils are energized. Moving the lower magnetic channel to a more favorable position has resulted in extraction of 40% of a circulating beam of 36-Mev protons, and as an added benefit has increased the reproducibility of good extraction conditions for 68- and 80-Mev alpha particles. Continuing effort is being given to the problem, and the extraction of protons at energies below 36 Mev is being improved.

## CHEMISTRY

Preparation and Properties of Actinide-Element Oxides. — Aging effects in 116-A crystallite thoria sols stored in tightly stoppered Pyrex containers at room temperature have been investigated by viscometry and light scattering. The interparticle fluid of the sol was adjusted to pH 3.6 with nitric acid. Over a period of a year the crystallite size was unchanged, and the observed complexities of flow behavior are attributed to partially reversible aggregation of the crystallites into flocs that are highly anisotropic in shape and possess at least one dimension greater than 1000 A.

Intrinsic viscosities were large, ranging from 30 to 50 millipoises, but were insensitive to temperature and to age in sols containing more than 20 g of $ThO_2$ per liter. Sols less concentrated than this showed greater effects of temperature and age, generally decreasing as either of these increased. Light extinction increased fourfold with one year of aging, despite the decrease or near-constancy in intrinsic viscosity. The extinction obeyed Beer's law in concentration, but not Rayleigh scattering in wavelengths up to 9750 A.

Protactinium Chemistry. — The solubility of $Pa_2O_5$ in hydrochloric acid solutions has been determined by heating excess $Pa_2O_5$ in hydrochloric acid of various concentrations. The resultant solutions are cooled to room temperature and filtered through Whatman 42 filter paper, and the protactinium concentration is determined by alpha-ray analysis. The results indicate that at HCl concentrations below about 6.5 N, the oxide solubility is too low (less than 5 mg/liter) to be determined by the method employed. Above this concentration the solubility of protactinium increases approximately linearly with HCl concentration, reaching a value of nearly 100 mg/liter in concentrated HCl (12 N). On a log-log plot, this indicates approximately a 7th-power dependence on the HCl concentration.

This work will be continued, both to confirm these results and to use a different technique to determine the solubility values.

As to spectrophotometric studies of sulfuric acid solutions of Pa(V), additional spectra of protactinium in $H_2SO_4$ solutions were measured to study the effect of the source of protactinium on the spectra. A sample of oxide obtained from Harwell was mixed with concentrated $H_2SO_4$ for several days, and the resulting solution gave a slightly different spectrum than had others; the peak was near 2100 A.

The sharp peak observed near 2000 A in protactinium solutions heated to fuming has been studied further. Generally the solutions became discolored somewhat, turning a light brown, when heated to fuming. It is known that various colored decomposition products are formed if any organic matter is present, and it was thought that this may have been the source of the discoloration. To test this idea, several solutions were heated to fuming and even evaporated to dryness after addition of organic material (usually Kleenex or paper towel). These solutions did give several small peaks in the ultraviolet spectrum, but nothing even remotely like the 2000-A peak observed with the protactinium solutions. From this result we conclude that the change in protactinium spectrum on heating is probably a real effect, and further investigation is planned.

Irradiation Effects on Heterogeneous Systems. — Considerable evidence has been amassed at various laboratories to support the belief that nuclear radiation absorbed in the body of a solid may be transferred with significant efficiency to the surface of the solid. At the surface the transferred energy may bring about chemical changes in adsorbed materials. We have been studying energy transfer in silica-gel—water systems by measuring the amount of hydrogen gas produced by water decomposition.

The G value for hydrogen was measured as a function of water content on silica gel (surface area 65 $m^2/g$) and on microporous Vycor glass (155 $m^2/g$). Comparison of the results with those obtained previously for silica gels with surface areas of 227 and 682 $m^2/g$ indicates that energy transfer is more efficient when the material has a high surface area.

Transport Properties of Gases: Thermal Transpiration. — The thermal transpiration experiments, designed to test the feasibility of using this phenomenon for a study of inelastic collision processes, have been completed for the gases argon, xenon, carbon dioxide, and nitrogen. In no case was it possible to fit the data to an empirical formulation proposed by Miller without further adjustment of the several parameters employed.

The translation Eucken factors calculated from the transpiration data were 2.28 ± 0.05 for nitrogen and 2.25 ± 0.08 for carbon dioxide. These results are in good agreement with the values of 2.26 and 2.24, respectively, which have been previously reported in the literature.

Construction of a more versatile thermal transpiration apparatus is about two-thirds complete.

Transport Properties of Gases: Gaseous Diffusion. — The gaseous diffusion studies utilize the temperature dependence of the coefficient characteristic of binary diffusion to determine the intermolecular forces between the gas molecules involved. To this end, diffusion experiments are currently being conducted over the temperature range 0 to 120°C with the gas pairs He-Kr, Ar-Kr, and Xe-Kr.

Analysis of Molten Salts by Electrochemical Methods: Voltammetric Measurements in Molten Fluoride Salt Systems. — The derivative of an unidentified wave (from a modified ORNL-Q 1988 polarograph) in a molten fluoride salt system at approximately −1.2 v at both pyrolytic graphite and platinum indicator electrodes revealed three peaks, indicating that the overall wave is actually composed of three waves and that the electrode reaction is certainly not a simple one.

To ascertain if any of the waves could be due in part to the reduction of $OH^-$ impurity in the melt, a weighed portion (560 mg) of anhydrous LiOH was added to the mixture, following which current-voltage curves were recorded after the melt had been allowed to equilibrate overnight. At the pyrolytic graphite electrode the peak heights of the first and second waves did not change significantly. The peak height of the third wave increased approximately sixfold. For the waves at the platinum indicator electrode, the first wave increased approximately twofold, whereas no appreciable change was observed for the second and third waves. From these observations it is tentatively concluded that the reduction of a trace of $OH^-$ in the melt is responsible in part for the waves that are observed at approximately −1.2 v; however, the overall electrode reactions appear to be very complex. Further measurements of these waves will be made to observe if they change with time. It is planned to make further additions of LiOH to the melt and also to look for evolved HF. This process should be of some help in ascertaining whether the LiOH is remaining as such in the melt or is reacting with fluoride ($OH^- + F^- \rightarrow HF + O^{2-}$) to form oxide and HF.

Nuclear Chemistry: Spectroscopy. — Measurements of the half-life of $^{137}Pr$ showed that it is only about three-fourths as long as reported in the literature. The $^{137}Pr$ was produced both by a (p,2n) reaction on cerium enriched in $^{138}Ce$ and by a (p,$\gamma$) reaction on cerium enriched in $^{136}Ce$. Although no gamma-ray emission was reported in previous work on $^{137}Pr$, measurements with NaI and germanium detectors indicate a very complex decay scheme with at least 15 gamma rays. A quantitative relationship was established between the disintegration rates of $^{137}Pr$ and the 9-hr $^{137}Ce$ daughter. Less than 0.1% of the $^{137}Pr$ decays to the 34-hr isomeric level in $^{137}Ce$.

Radiation Chemistry: Adsorption by Amorphous Ice. — Water vapor condensed on a surface cooled by liquid nitrogen has the ability to adsorb large quantities of oxygen and other gases, indicating high surface area. This ability to adsorb gases disappears when the amorphous ice is warmed to −130°C. Amorphous ice prepared and maintained at −196°C, when subjected to gamma radiation yields hydrogen at a rate of 0.3 molecule/100 ev. If the ice is annealed by warming to −130° before irradiation, the hydrogen yield drops to about 0.15, and if the ice is allowed to crystallize by warming above −120°, the yield drops further to about 0.1. Almost all of the hydrogen produced in the unannealed amorphous ice is found in the gas phase, while hydrogen produced by irradiation of ice previously warmed to −10° remains in the solid when the solid is irradiated at −196°.

The ability of amorphous ice to adsorb and occlude oxygen might be considered for possible applications in space technology.

Radiation Chemistry: Mechanism of Radiolysis of Water. — The predominant reducing radical produced in the radiolysis of water has been identified as the hydrated electron. There are also about 1/5 as many "H" atoms produced as there are electrons. The yields of these species were not accurately known, but on analyzing the existing information, Allen concluded that the reducing species $e^-_{aq}$, "H," and $H_2$ were in excess of the oxidizing species OH and $H_2O_2$, and suggested an additional oxidizing radical, possibly the O atom. We reevaluated the total yields of oxidizing and reducing radicals in the gamma radiolysis of water using carbon monoxide and oxygen as scavengers. Carbon monoxide is an efficient scavenger for OH radicals and electrons, and oxygen reacts rapidly with electrons and with the intermediates produced by the reactions of carbon monoxide. All of the radicals are thereby converted to $H_2O_2$ (via $HO_2$ or $O_2^-$). The measured initial yield was $G(H_2O_2) = 3.62 \pm 0.07$, and according to the proposed mechanism this is equal to $G_{H_2O_2} + \frac{1}{2}(G_{red.} + G_{ox.})$, which is also equivalent to $G_{H_2} + G_{red.}$. The initial yield of peroxide in oxygen-swept solution was remeasured as $1.03 \pm 0.02$ and is equivalent to $G_{H_2O_2} + \frac{1}{2}(G_{red.} - G_{ox.})$. The above measured values, when corrected to zero scavenger concentration to compensate for track (spur) scavenging, are 3.58 and 0.99. The difference in these two values, 2.59, is equal to $G_{ox.} = G_{OH}$. Taking the known value of $G_{H_2} = 0.45$, the total yield of reducing radicals is 3.13. Assuming the yield of H atoms to be 0.55 as reported, this gives 2.58 for the yield of electrons. The known yield of $H_2O_2$ is 0.72. These values give good material balance and eliminate the need for postulating an additional oxidizing radical. The equivalence of $G_{"H"}$ and the excess of molecular peroxide over molecular hydrogen suggests the possibility that "H" and part of the molecular peroxide are produced in the same reaction.

Radiation Chemistry: Radiation Stability of Sulfur Dioxide. — Sulfur dioxide is of interest as a potential fast-reactor coolant. However, the radiation stability of gaseous $SO_2$ had not been investigated. Therefore, room-temperature irradiations of $SO_2$ were made with alpha particles from radon.

Initially, visual evidence, such as a Tyndall beam, fog in the reaction vessel, and a slight deposit, indicated radiation damage. After the decay of the radon, pressure measurements showed no pressure change within experimental error.

Mass spectrometric analyses of the initial and the irradiated gases indicated little net change had occurred. Gas chromatographic analyses showed no detectable $O_2$ and only a few hundredths of a percent of $SO_3$.

These results, combined with the disappearance of the initial visual evidence of radiation damage, suggest that under the conditions of the experiment, back reaction of the initially formed products had occurred.

High-Temperature and Structural Chemistry. — In pursuance of our study of mixtures of metals with salts in the liquid state at elevated temperatures, we have begun an investigation of the state of dissociation of salts when dissolved in a liquid metal. [Only one previous such study seems to have been undertaken, that of $BiCl_3$ in liquid bismuth metal, in

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which the salt addition was found to produce three ("new") particles, namely, $3Cl^-$, besides $Bi^{3+}$, or $3(BiCl)$, or $3(BiCl)^{2+}$.] We chose to measure the depression of the freezing point of calcium metal by calcium fluoride and found it to be even smaller than that corresponding to the introduction of only one new particle. This obvious discrepancy was resolved by measurements of the unexpectedly high solubility of the salt (1 mole %) in solid calcium metal. The difference (0.5 mole % $CaF_2$) in the composition of the solid metal and the liquid eutectic phase in equilibrium therewith (1.5 mole % $CaF_2$) establishes the fact that $CaF_2$ is dissociated in the liquid metal either according to $CaF_2 \rightarrow Ca^{2+} + 2F^-$ or, less likely, according to $CaF_2 + Ca^{2+} \rightarrow 2(CaF)^+$ or to $CaF_2 + Ca \rightarrow 2CaF$. The latter mode of reaction might be excluded by measurements of magnetic susceptibility.

Chemical Physics: Molecular-Beam Studies with Cesium Ions. — The angular distribution of $Cs^+$ ions elastically scattered from Ar, Kr, and $N_2$ was measured in the energy range 2.3 to 11.6 ev. A measurement of the angular distribution of elastic scattering is expected to yield information about the interatomic potential between the interacting particles.

Rainbow scattering was observed for the three systems studied. This is manifested by a rise in cross section at some angle. The position of this peak is related to the parameters of the interatomic potential, particularly the potential-well depth.

The position of the observed peaks indicates potential-well depths three to four times greater than those expected from an interatomic potential whose attractive part is due only to polarization forces.

It is proposed that the additional binding energy may come from an attractive exchange force. There is some evidence that partial electron exchange, in the sense of Mulliken's charge transfer theory, can account for the stabilization energy of rare-gas compounds such as $XeF_2$ and $XeF_4$. Because the electron affinity of the $Cs^+$ ion is larger than that of a fluorine atom, it seems reasonable to expect that a partial electron exchange may contribute appreciably to the interatomic potential. Calculations are being done to estimate the size of this effect.

Chemical Physics: Molecular-Beam Studies of Hydrogen in Helium. — Measurements of the total scattering cross section of $H_2$-He and $D_2$-He by molecular-beam methods were undertaken in an attempt to resolve discrepancies in the relative scattering cross sections of $H_2$ and $D_2$ as determined from various transport property measurements. Experimentally unique was the use of a mixed $H_2$-$D_2$ modulated molecular beam and the arrangement of the detecting system so that mass 2 and mass 4 signals could be measured simultaneously and their ratios compared with and without scattering gas (He) in the scattering chamber. During these experiments a coherent He signal was observed which was due to small impact parameter ("knock-on") collisions. This phenomenon was then further studied, since such "knock-on" collision cross sections represent a direct measurement of large-angle scattering cross sections which are important because they are directly relevant to macroscopic gaseous transport properties (e.g., diffusion). However, their determination by molecular-beam techniques is uncommon. Furthermore, "knock-on" cross sections are important for measurements of total collision cross sections in which beam and scattering particles are alike.

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The 300°K $H_2$–77°K He total scattering cross section was determined to be 51.5 ± 0.8 $A^2$, and the corresponding "knock-on" cross section was determined to be about 7 $A^2$, a yield so large as to increase the value of the He-He total scattering cross section by ~20%.

TRANSURANIUM-ELEMENT PROCESSING

Curium Recovery Facility. – Recovery of the $^{242}Cm$ from the irradiated $^{241}Am$ targets was completed. A total of 4.0 g of $^{242}Cm$ was separated and recovered as product suitable for conversion to oxide and encapsulation. A second TRAMEX cycle was necessary to obtain $^{242}Cm$ of acceptable purity. In addition to the highly purified material, about 1 g of partially purified $^{242}Cm$ was transferred to cell 3 for flowsheet experimentation.

Solvent extraction losses were decreased from 2.6 to 1.3% in the second TRAMEX cycle by using a higher ratio of solvent to feed. In both runs, losses were split about equally between the waste organic and the raffinate.

A modified feed-adjustment procedure developed to obtain maximum cerium decontamination resulted also in increased ruthenium removal by volatilization. The procedure consisted in close control of feed acid throughout the run, coupled with periodic removal of oxidizing agents by volatilization (presumably hypochlorous acid and dissolved chlorine). An acidified reductant solution was added to hold cerium in the trivalent state. The effects of radiolysis were controlled by limiting the size of the feed batch and the curium concentration to give a maximum exposure to ionizing radiation of 600 whr per equivalent of reductant. The maximum curium power density that could be tolerated on this basis was 6 w/liter. Ruthenium and zirconium decontamination factors across the CLANEX-TRAMEX first cycle were 300 and 9000 respectively; those for $^{103}Ru$, $^{95}Nb$, and $^{144}Ce$ were 84, 500, and 160, respectively, in the second TRAMEX cycle.

A simultaneous failure of the feed-transfer pump and a manipulator resulted in a two-day shutdown of the facility; about 75% of the feed had been processed at the time. After the delay, several difficulties were observed while the remaining feed was processed; the effects of radiolysis products were intensified, and difficulties were experienced with interfacial solids.

After the second TRAMEX cycle, the product was converted from the chloride to the nitrate form in preparation for shipments to the Source Fabrication Facility.

Plutonium Target Fabrication. – Fabrication of the dummy HFIR target assembly (31 elements) for flow testing by the Reactor Division has been completed. Fabrication of a second flow-test assembly is being held and will be completed only if difficulties develop in the flow testing of the first assembly.

Preparations are being made to make a simulated plutonium target assembly and the first $^{242}Pu$-containing target assembly for the HFIR. The simulated target assembly, which will be capable of producing the maximum heat generation rates calculated for the presently proposed plutonium targets, will contain enriched uranium and tantalum. The $^{242}Pu$-containing assembly will contain 8 g of $^{242}Pu$ per element (31 elements

per assembly). We have been requested to have both assemblies made before August 15, 1965.

HFIR Target Irradiations. — The two prototype target rods still being irradiated in the ETR are continuing to function without incident.

Examination of the second rod removed from the reactor, which had been exposed to an average heat flux of 1 million Btu $hr^{-1}$ $ft^{-2}$, is in progress. As reported previously, an unsuccessful attempt was made in the reactor pool to remove the flow holder, which was stuck in the X-basket. During the hot-cell examinations, the problems created by doing this in the reactor pool became evident, and the reactor people have been advised not to do this in future experiments. The damage to the fuel rod is apparently superficial and not enough to harm the postirradiation evaluation, but it did prove the remarkable structural stability of the fuel-rod assembly. Apparently, a small-diameter rod was dropped or forced down through the X-basket with the intent of forcing out the holder. Sufficient force was exerted to break the stainless steel retaining bolt on that end and force the assembly through the holder until the opposite-end aluminum tip was stopped by the other retaining bolt. The bolt that did not fracture was embedded in the aluminum, leaving distinct thread marks. The other end of the target rod was curled up like a pig tail and forced against the side of the flow holder. A section was gouged out of the closure weld on this end, although apparently not deep enough to penetrate to the gas-plenum area. It was necessary to cut the ring off the flow holder at the pig-tail end before the fuel-rod assembly could be removed.

When the hex can was removed, the points of connection between the fins and the can were examined. There was no evidence of damage. The force on the pig-tail end had caused a bowing of the rod between the fins on that end and the central fins. This flexing action had broken the heavier oxide scale off the exterior of the target tube, leaving a series of six relatively scale-free rings next to the end fin.

The gross gamma scans showed no anomalies. Fission-gas samples are being analyzed. The diametral measurements, while showing the sinusoidal form of the unirradiated rods, are within the preirradiation maximum and minimum, indicating no significant swelling. Sectioning for metallography is under way.

As reported previously, a heat-treatment program on unirradiated, rejected pellets is being conducted to examine $PuO_2$-Al reactions and to investigate the temperature at which the aluminum substructures change. This information may make it possible to estimate irradiation temperatures.

Six pellets were heat treated for 24 hr at 50°C temperature intervals from 350 to 600°C. The density in all cases increased from 3 to 4%. There was no evidence of a $PuO_2$-Al reaction at any temperature. A one-week exposure to 600°C was completed, but the pellet has not yet been examined. The aluminum substructure was gone after 24 hr at 550°C and somewhat altered at 500°C. A one-week treatment at 475 and 500°C will be given two separate pellets in order to better define the limiting temperature and the effects of longer exposure. A third study at 450 or 525°C is planned, pending the results of these longer exposures.

Transuranium Processing Plant Design and Construction. — The application of floor and wall coatings, general cleanup, and the testing of ventilation systems are the principal tasks remaining to be performed under the lump-sum contract for the construction of the Transuranium Processing Plant. The work is expected to be completed late in May.

The bulk-shielding windows that had been leaking oil were inspected by representatives from Corning Glass Works, and all repairs and deficiencies were corrected, except for one window, which was returned to the factory for repair. The cell-cubicle cooling systems and the postirradiation carrier fabrications at Paducah are nearly 30 and 50% complete respectively. The delivery of the Hastelloy vessels from Nooter Corporation has been delayed slightly.

Preparation of the equipment-installation packages that define work to be done by the H. K. Ferguson Company on a cost-plus-fixed-fee contract is about complete. The drawings and specifications for package II (waste-pit piping) were issued "Certified for Construction." Piping drawings for all out-of-cell work were issued "Title II for Approval," and the associated instrument and electrical drawings were completed and issued as "Preliminary."

Fabrication of processing equipment is continuing on schedule in the ORNL shops. Installation of nozzles in the 36 process tanks, one of the major jobs remaining, was just started. Nearly all equipment will be available by September 1.

## METALLURGY AND MATERIALS

Preparation of Pure Materials. — A 325-g crystal of LiF, designated ORNL-10 and containing 99.99 at. % $^{7}$LiF, was produced and made available for use at the Materials Science Center at Cornell University as part of the AEC Pure Materials Program. In addition, 100 g of high-purity dehydrated LiF, used as the charge material for growth of ORNL-10, was furnished to the Cornell group for growth of LiF single crystals by the Czochralski method. As in previous ORNL preparations, the crystal grown here was prepared by the modified Stockbarger method, vacuum annealed, and subsequently handled only in vacuum or inert atmospheres. The thermal conductivity of ORNL-10 was determined to be 52 w $cm^{-2}$ $cm^{-1}$ $(°C)^{-1}$ at Cornell, approximately half that expected from current theory, whereas the thermal conductivity of crystals grown from the same starting material by the Czochralski method was in the theoretically expected range. Heavy-metal impurity content of ORNL-10 was determined by activation analysis to be about 1 ppm manganese, some three times that obtained in our best preparations (ORNL-3341). The irreproducibility of Stockbarger crystals with purity exceeding 1 ppm indicates that it will be necessary in the future to employ growth capsules which are free of manganese.

Deformation in Crystalline Solids. — Plastic deformation of metals causes the grains to take up preferred orientations. A knowledge of this preferred orientation is important for understanding the nature of deformation and because preferred orientation affects the physical properties. One aspect of such a study is the representation of the orientation data

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in the simplest and most direct fashion; for this purpose one can use an inverse pole figure in which the orientations of the physical directions are represented in crystallographic coordinates. There are no simple direct methods for deducing inverse pole figures from conventional data, although trial and error methods have been in use for some time. Since these methods are time consuming and may not give the best inversion, computer methods have been under investigation recently.

In our current method a set of peaks distributed uniformly over the crystallographic triangle represents the desired function. One can write a set of linear equations relating the experimentally measured intensity to the strength of the peaks. Since generally more equations are used than there are peaks to be evaluated, the computer performs the inversion in the least-squares sense. However, the resulting matrix, which now gives the value of each peak as a linear function of intensities, gives neighboring peaks that differ excessively in strength. When we wrote additional linear equations to limit the variation in strength of neighboring peaks and incorporated them into the inversion, the resulting matrix gave an inversion that was comparable to or better than that possible by trial and error methods. The increase in the error in the calculated intensities was unimportant.

Fundamental Research in X-Ray Diffraction. — A careful survey of reflection intensities that could be significantly changed if guest atoms or molecules were introduced into the holes of the calcium beryllate structure has been completed with an "empty" crystal of $Ca_{12}Be_{17}O_{29}$. This crystal will be used in experiments to determine if argon or helium can be introduced into the structure through treatment at high pressure and temperature.

Reflection profile analyses of BeO crystals irradiated at 0.4, 3.7, and $7.1 \times 10^{20}$ neutrons/cm$^2$ (fast) at <150°C have shown that sharp peaks disappear and diffuse maximums appear within this dose range for reflections with large $[\ell^2/(h^2 + k^2 + hk)]$ values. The shift of sharp components to higher Bragg spacings [e.g., $d_{002}$ (sharp) = 2.28 A for $3.7 \times 10^{20}$ neutrons/cm$^2$] has been confirmed.

By studying multiple diffraction from unirradiated and irradiated BeO crystals in goniostat geometry, we have developed a useful technique for the measurement of lattice parameters of large nonisotropic crystals whose shape precludes lining up more than one crystal direction in the diffraction plane. The usual method for obtaining a complete set of lattice parameters for such crystals involves tilting the crystal and making tedious and questionable corrections for the angle. Our method avoids this. It involves the recording of azimuthal rotation angles about a space-group-forbidden reflection at which multiple diffractions from other reflections produce maximums.

Reactions at Metal Surfaces. — We have continued our studies of the mechanisms of alloy corrosion in high-temperature water. Alloys tested included Fe–15% Cr–4% Al–1% Y, Al–20% Zn, and Al–5% Fe. All were tested at 200°C and about 250 psi in distilled water, and the iron alloy was also tested at 300°C.

Both the Al–20% Zn and the Al–5% Fe alloys exhibited small weight gains in tests lasting up to 24 hr. The corrosion resistance of both alloys was considerably superior to that of pure aluminum, and only slight

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grain-boundary penetration was observed in either material. Only boehmite, AlO(OH), was detected on either alloy after testing. Phenomenologically, the corrosion process resembled that previously reported for Al-Y alloys.

The Fe–15% Cr–4% Al–1% Y alloy formed only a thin interference film in 24 hr at 200 or 300°C. No weight change was detected, and the surface oxide, as determined by reflection electron diffraction, was either $Fe_2O_3$ or $Cr_2O_3$.

Spectroscopy of Ionic Media. – As part of a continuing program to study the geometric and electronic structures of ionic liquids, we have completed an investigation of the optical absorption spectra of nickel(II) centers in melts of CsCl, RbCl, KCl, NaCl, LiCl, $MgCl_2$, CsBr, KBr, and LiBr. The measurements on CsCl, LiCl, $MgCl_2$, and LiBr extended from 4 to 40 kK (1 kK = $10^3$ $cm^{-1}$), and covered both d → d and charge-transfer absorption bands. The remaining systems were studied in the range 4 kK to between 22 and 26 kK, which included the d → d absorptions. The results were analyzed in terms of ligand-field theory to give information on the coordination geometry of halide ions about $Ni^{2+}$ ions and electronic structures in the various melts. In CsCl, CsBr, RbCl, KCl, or KBr, nickel(II) ions were surrounded by approximate tetrahedra of halide ions. In NaCl, LiCl, LiBr, and $MgCl_2$, two kinds of nickel centers were present, one of which was four-coordinate and the other had an unidentified geometry. The four-coordinate center predominated in NaCl and at relatively high temperatures in LiCl.

Fundamental Investigation of Radiation Damage in Solids: A Camera for Borrmann Stereo X-Ray Topographs. – In order to take Borrmann topographs for the study of dislocations in nearly perfect copper crystals, a relatively inexpensive camera for use with standard x-ray diffraction equipment has been constructed. The horizontal resolution is determined by the width of reflection of the nearly perfect crystals employed, and only a simple slit is needed to separate the $K_\alpha$ doublet. A vertical resolution of about 4 μ is obtained by utilizing a spot focus, a distance of 100 cm from the focal spot to the sample, and a distance of 0.5 cm from the sample to the Ilford L-4 plates used for recording the topographs. The camera has been used with both a G-E CA-7 and a CA-7H x-ray tube, and typical exposure times [for (111) reflections and $\mu t = 10$] were 16 hr for crystals 1 cm square in cross section. Topographs have been taken of copper crystals up to 0.2 cm thick with both silver and molybdenum characteristic radiation, and {111} topographs have been used to characterize dislocations in crystals for $\mu t < 20$. This camera has been used to take stereo pairs of Borrmann topographs by rotation around an axis normal to the diffracting planes, in contrast to Lang's method which involves topographs of hkl and $\bar{h}\bar{k}\bar{l}$ reflections. For the crystals studied thus far, a stereo angle of about 10° was found to be most desirable.

Fundamental Investigation of Radiation Damage in Solids: Orientation Dependence of the Emission of Alpha Particles from a Monocrystalline Source. – The recent experimental discovery by Domeij and Björkqvist of the anisotropic emission of alpha particles from a monocrystalline source can be discussed in terms of a model which considers the emission from alpha-active nuclei substitutionally located at lattice sites. An alpha particle starting at an angle near a crystallographic direction undergoes

a large scattering by the neighboring lattice nucleus. The angle between the crystallographic direction and the direction of the alpha particle after this scattering event is a minimum (Rutherford scattering) for an impact parameter of $(Z_1Z_2e^2d/E)^{1/2}$, where d is the lattice spacing. For angles greater than this cutoff angle, $\theta_c = 2(Z_1Z_2e^2/dE)^{1/2}$, the alpha emission per solid angle is $\sim(1 - \theta^2/\theta_c^2)^{-1/2} + (1 - \theta^2/\theta_c^2)^{1/2}$, where the mean free path, $\lambda(\theta > \theta_c)$, is greater than the source depth. Including thermal vibrations into the model brings the calculations into quite good agreement with experiment. Although the numerical work has been done with a particular experiment in mind, the model can be tested by similar experiments such as emission of fission fragments from a monocrystalline source.

## CONTROLLED THERMONUCLEAR RESEARCH

The DCX-2 Injection-Accumulation Experiment. — An attempt to increase the production rate of particles with small axial momentum by preventing precession of the initially trapped protons was described in the last report. The arc was moved 3 in. from the axis so that the arc-dissociated proton orbits were concentric with the axis. At the time of the report it had not been determined that the molecular ion helix was in the proper place. Later measurement showed that in fact it was, leading to the conclusion that increasing the density of countercurrent protons did not enhance the production of the class of particles having small axial momentum. Another possible reason for the existence of these particles is that they are produced by ion cyclotron heating of the cold plasma. The beam-injected particles may transer energy to cold hydrogen in this way if they are bunched. Heavier ions might also be heated if the rf disturbances are sharply localized, since their gyration frequency is at a subharmonic of the proton frequency. No additional neutrons were produced, however, when deuterium gas was injected into the plasma region. In other experiments the lithium arc was moved on-axis, and the beam injection was moved correspondingly. The performance of the machine was only slightly altered. With gas breakup, however, the indicated density increased with background pressure to a maximum of about $3 \times 10^8$ at a pressure of $3 \times 10^{-6}$ mm Hg. Most recently a hydrogen arc has been installed in the DCX-2. The anode and cathode configuration is being adjusted to minimize the pressure in the plasma chamber.

Active and Passive Stabilization of Plasmas and Ion Beams. — Both the experiments dealing with sources as well as those studying the response of the calutron system to a variety of boundary impedance networks continue. The reproducibility of the oscillatory condition in the ion source proper is a very delicate matter, and we are trying to better control this parameter. We are also trying to improve the usefulness of our Faraday cup for observing radio-frequency signals being propagated in the cold plasma.

Properties of Vacuum Arcs. — The unusually bright character of the blue $\lambda$ 4647-A spectral triplet line group of $C^{2+}$ in our magnetically confined arcs and the recent observation of laser action of this group [Appl. Phys. Letters 5, 91 (1964)] prompted a review of mechanisms for enhancing

the triplet. The singlet line λ 5696 A, which requires about the same excitation potential, is only about 1% as bright in our arcs. We conclude that the probable reason for the seeming anomaly in intensity is related to the lack of competing downward transitions from the upper levels of λ 4647-51 A ($2s3p\ ^3P^0_{0,1,2}$), whereas λ 5696 A can decay by either λ 5696 A, $2s3d\ ^1D_2 \rightarrow 2s3p\ ^1P^0_1$ ($A = 5 \times 10^7\ sec^{-1}$) or via λ 574 A, $2s3d\ ^1D_2 \rightarrow 2s2p\ ^1P^0_1$ ($A = 6 \times 10^9\ sec^{-1}$). Thus, assuming the same rate of excitation of the 2s3d and 2s3p levels, λ 5696 A should be about 1% as bright as λ 4647 A, in agreement with our observations. Cascade transitions into $2s3p\ ^3P^0$ are permitted to decay further only by λ 4647-51 A transitions ($A = 8 \times 10^7\ sec^{-1}$) to the $2s3s\ ^3S_1$ level, which then decays at a rate of $2 \times 10^9\ sec^{-1}$. The lasing property of the triplet group may be related to the very short life of the $2s3s\ ^3S$ state and the relatively long life of the $2s3p\ ^3P^0$ state, which permit level inversion of the $^3P^0$ state in optically thin plasmas.

Experimental Plasma Physics. — We have attempted to observe the reflection of ionic sound waves from glass and metal surfaces. No reflection was observed, which in our apparatus corresponds to less than $10^{-3}$ of the wave energy being reflected. Although this experiment appears to conflict with others in which standing waves were observed, it differs from the others in that this plasma is quiescent and formed by diffusion. The other experiments used noisy, active plasmas in which counterstreaming electron and ion beams were present.

Magnetics and Superconductivity. — For several experimental facilities new field calculations and magnet coil designs were made. The technological tests of the new construction elements of the 7-Mw, 13-in.-high homogeneity magnet coil were successfully completed, and the manufacturing of this solenoid was started. Field calculations and engineering designs were made for providing an additional magnetic shear field for the DCX-1.

Continued experiments with indium arsenide films demonstrated the advantages of using this material for cryogenic micro Hall probes. The developmental work on the new type of magnetometer progresses satisfactorily.

Flux jump observations on cylindrical Nb-Zr samples with small demagnetization factors in longitudinal external fields are being continued. Oscillograms show the propagation modes of the flux jumps from points located at the sample surface. The longitudinal propagation velocity of the superconducting normal boundary has been measured. Experiments with almost adiabatic field sweeps yielded results which are in agreement with a newly developed theoretical approach based on the H. London—C. P. Bean model. Samples in transverse fields show similar behavior; however, a quantitative analysis is not possible yet.

Vacuum Studies for Fusion. — Estimates of possibly large tritium inventories in conceptual thermonuclear reactors were made, based on assumptions regarding the concentration of hydrogen isotopes in metals when subjected to a flux of ionic or atomic hydrogen. An apparatus was designed and constructed in which hydrogen glow discharges could be operated in order to test the validity of some of the assumptions needed in the inventory estimation. Initial results indicate that molybdenum metal in

a glow discharge can occlude at least 0.2 atm cc of $H_2$ per cc of molybdenum in a glow discharge at 500 μ pressure, 2 ma/cm$^2$ current density, and 700 v applied potential. This value is approximately one thousand times the equilibrium concentration of hydrogen in molybdenum at this pressure calculated from literature data, thus indicating that chemical processing of tritium must include this factor.

Several hundred mass spectral scans were obtained on the DCX-2 experimental device, and analysis of this data has yielded some generalizations regarding the origin of certain species in the DCX-2 vacuum system.

## BIOLOGY AND MEDICINE

### TERRESTRIAL AND FRESH-WATER ECOLOGY

Environmental Radiation Botany: Prediction of Tree and Plant Community Radiosensitivity. — Based on the direct relationship between average nuclear volumes or estimated chromosome volumes, predictions of tree radiosensitivity and possibly plant community radiosensitivity can be made under field conditions. Interseasonal differences in the size of interphase nuclei of somatic cells of shoot meristems have been determined and cataloged for 14 local tree species which are representative constituents of various plant communities over a period of one year. Results indicate that 11 of 14 are most radiosensitive between April and September. The ratios of the changes in average nuclear volume, actively growing to dormant plants, varied from 1.2 to 2.0 between species, indicating a wide range in interseasonal radiosensitivity. These results also suggest that one might be able to predict intraseasonal variations in radiosensitivity for some species.

Environmental Radiation Botany: Effects of Fast Neutron Irradiation on Soluble RNA Characteristics in Mimosa. — Nine-day mimosa seedlings (*Albizzia julibrissin* Durazzini) were given a total dose of 1350 rads (tissue dose in air) of fast neutrons delivered over a period of 47 min. Epicotyls of irradiated and controlled seedlings were excised at three-day intervals following irradiation. Soluble and total RNA were quantitatively extracted and base compositions determined. Early results suggest that levels of sRNA (soluble RNA) decrease rapidly in irradiated seedlings, and more slowly in controls as a result of aging. Base compositions varied, with guanine and cytosine showing an immediate rise in concentration, while uracil and adenine correspondingly decreased. Previous correlations of sRNA base compositions with growth rates suggest that these postirradiation trends indicate an immediate flush of growth, followed by retardation. Such variations suggest shifts in mechanisms of sRNA synthesis, and possible protein synthesis, following irradiation.

Environmental Radiation Botany: Response of Vegetative Growth and Flowering in Xanthium to Day-Night Gamma Irradiation. — Based on previous observations that flowering in *Xanthium* is inhibited when the plants are irradiated simultaneously with photoinduction (short-day treatment to induce flowering), an experiment was designed to investigate differential

radiosensitivity during either the dark or light period of photoinduction. Both the dark- and light-irradiated plants showed an initial flush of vegetative growth over the nonirradiated, but photoinduced, controls. Early results indicate that light-irradiated plants show an inhibition in flowering as compared to the controls, while dark-irradiated plants show an initial stimulation. Future results should elucidate differential radiosensitivity of flowering in Xanthium, depending on the timing of irradiation with respect to dark or light periods.

Environmental Radiation Botany: Differential Radiosensitivity of Cottonwood Clones. — Dormant stem cuttings from ten clones of Populus deltoides Marsh. (Eastern cottonwood) representing ecotypes from Alabama, Mississippi, Kentucky, and Illinois were given an acute $^{60}Co$ gamma radiation dose of 7600 rads. These irradiated cuttings, along with controls from each clone, were placed in aerated nutrient solutions and maintained under constant environmental conditions in a plant growth chamber. Clones were scored for time of leaf emergence, callus formation, and root emergence. Shoot height was measured during the initial rapid growth phase (20 days), and shoot and root biomass was measured on harvested plants. Statistically significant growth differences were found between control clones, with five clones exhibiting a growth rate 1.5 to 2 times greater than other clones. Biomass differences of the same order were found. Statistically significant differences also were found between irradiated clones. Average retardation of shoot height ranged from 60 to 85% of control clones; shoot biomass reduction, 64 to 95%; inhibition of callus formation, 0 to 45%; and root biomass reduction, 56 to 100%. Formation of callus tissue was least affected, but root production was most affected. A linear relationship was found between clonal growth performance and radiosensitivity. Clones which exhibited the best growth were more radiosensitive when irradiated in the dormant condition. Histological examination of apical meristematic tissue showed no nuclear basis for differential radiosensitivity of clones.

Radionuclide Cycling in Terrestrial Ecosystems: Biological Half-Life of $^{134}Cs$ in Forest Arthropods. — The biological half-lives of $^{134}Cs$ in arthropods of detritus-based food webs were determined for 15 species as a continuation of the study of the biological behavior of radionuclides in forest ecosystems. The species utilized belonged to the natural fauna of the $^{137}Cs$-tagged Liriodendron forest and were selected from a variety of orders to serve as analogs for future field studies. These were fed on contaminated food in the laboratory, and, subsequently, eliminations were measured at 20°C. Disregarding the rapid initial loss rates due to gut voidance, the elimination curves were best characterized by a single exponential for lepidopteran larvae and some Isopoda, Diplopoda, Isoptera, Orthoptera, and Coleoptera. Multiple exponentials better described radiocesium elimination in other Isopoda, Orthoptera, and Coleoptera, with the longer rate functions responsible for loss of more than 50% of body burden. Loss rates approximate power functions for the millipedes Ptyoiulus impressus and Cambala annulata. Biological half-lives ($T_b$) ranged from 1.5 days for the wood roach Parcoblatta to 11.8 days for the isopod Armadillidium vulgare. Current studies are attempting to correlate $T_b$ with metabolic rates and species biomasses. Physiological stresses such as laboratory confinement can be important variables affecting elimination rates.

Molting is also significant, resulting in cessation of elimination preceding molt followed by a loss of less than 30% with the cast skin. Temperature appears to be the most influential abiotic variable affecting elimination, following a $Q_{10} \sim 2$ for those species examined (isopods Cylisticus convexus and A. vulgare, geometrid larvae, and the millipede Dixidesmus erasus).

Radionuclide Cycling in Terrestrial Ecosystems: Cesium Leaching from Terrestrial Microcosms. – Preliminary work on an analog computer model of $^{134}Cs$ leaching from terrestrial microcosms of varying complexity indicates that these systems may be simulated by a simple diffusion model.

In a litter-leachate system (cesium-labeled leaf litter) the rate of leaching was 7.3% per day. Addition of soil did not influence this rate, but only 0.2% per day of the removed activity appeared in the leachate, the remainder being transferred to the soil where it was held. In a litter-microflora-leachate system, 3.8% per day was transferred to the leachate and 3.3% per day to the microflora, so that total loss from the tagged litter was substantially as in the two previous systems. When soil and microflora were added together, however, the rate of activity loss from the litter was almost doubled, 14.1% per day. Of this, 8.3% went to soil, 3.3% to microflora, and 2.5% to leachates.

Work with a fifth compartment, soil arthropods, is continuing, and particular attention will be given to reconciling all of the results obtained with the experimental and computational methods used to obtain the data.

Radionuclide Cycling in Terrestrial Ecosystems: Radiocesium Inputs for Forest Floor Food Chains. – Cesium-137 concentrations in materials which form the bases for food chains were measured in the forest floor of the tagged Liriodendron stand during fall-winter 1964–1965. Weekly samples estimated radiocesium content of leaf litter, moss, roots, and woody materials as well as the soil. Leaf litter (including minute fragments) averaged about 55 dpm (disintegrations/min) per mg of dry weight during the six-month period; averages for moss (81 dpm/mg) and roots (102 dpm/mg) were not significantly different from leaf litter. Wood (13 dpm/mg) and soil (20 dpm/mg) had lower $^{137}Cs$ concentrations than did leaf litter. Radiocesium concentrations in leaf litter decreased from September to mid-October but thereafter showed a slight but statistically significant ($P = 0.01$) increase on both per unit weight and per unit area bases. Previous experimentation with litter bags and in the laboratory had indicated a continued decrease in radioactivity during the winter months. Possible explanations for the increase include a rearrangement of $^{137}Cs$ within the organic layer itself from larger fragments to organic particles, movement of $^{137}Cs$ from mineral soil into organic materials, movement via roots or microbial growths from soil into organic layer, or a progressive bias in the sampling procedures. A consequence of such a longer retention of $^{137}Cs$ in leaf litter is a greater $^{137}Cs$ availability to litter-feeding arthropods. No consistent change in $^{137}Cs$ content was detected in moss, fallen wood, or roots during the six-month period. In soil, $^{137}Cs$ concentrations increased from about 8 dpm/mg in September to about 20 dpm/mg in March, with the increase probably due to radiocesium accumulation from roots of the tagged Liriodendron trees.

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Systems Ecology: Probabilistic Estimates of Forage Intake. — Daily forage intake (F) may be calculated by the equation

$$F = 100 \cdot E \cdot E_c / (100 \cdot F_c - F_c \cdot M_c) \ , \qquad (1)$$

where E is the quantity of feces produced per day, $E_c$ is the percent cellulose content of the feces, $F_c$ is the percent cellulose content of the diet, and $M_c$ is the percent microdigestion of dietary cellulose. Generally, E, $E_c$, and $M_c$ are measured with one group of animals, and $F_c$ is measured with a second group of animals grazing with the first group. Estimates are obtained by sampling of the means and standard deviations of these variates and of correlations among E, $E_c$, and $M_c$, and it is assumed that these variates follow a normal distribution. No measure of precision of F is obtained by conventional procedures. An estimate of the precision of F may be obtained by generating a large number of such values and examining the distribution. For this purpose it is assumed that E, $E_c$, and $M_c$ are to be sampled from a trivariate normal population. This is accomplished by first generating three mutually independent standard normal deviates designated as u, v, and w. Values for x, y, and z are selected by letting x = u and then calculating y and z by Eqs. (2) and (3):

$$y = r_1 \cdot u + a \cdot v \ , \qquad (2)$$

where $r_1$ is the correlation between E and $E_c$, and a is calculated from the variance of y: $a = \sqrt{1 - r_1^2}$;

$$z = a \cdot u + b \cdot v + c \cdot w \ , \qquad (3)$$

where a is given as above, and b and c are calculated by (4) and (5);

$$b = \frac{r_3 - r_1 \cdot r_2}{a} \ , \qquad (4)$$

where $r_2$ is the correlation between E and $M_c$, and $r_3$ is the correlation between $E_c$ and $M_c$;

$$c = \sqrt{\frac{1 - r_1^2 - r_2^2 - r_3^2 + 2r_1 \cdot r_2 \cdot r_3}{a}} \ . \qquad (5)$$

The deviates x, y, and z (from a standard trivariate normal population) are "scaled" by Eqs. (6):

$$\begin{aligned} E &= x \cdot s_E + \overline{E} \ , \\ E_c &= y \cdot s_{E_c} + \overline{E}_c \ , \\ M_c &= z \cdot s_{M_c} + \overline{M}_c \ , \end{aligned} \qquad (6)$$

[redacted]

where $s_E$, $s_{E_c}$, and $s_{M_c}$ are the measured standard deviations, and $\overline{E}$, $\overline{E}_c$, and $\overline{M}_c$ are the means. Utilizing these procedures with data from experiments with grazing cattle and sheep, from 1000 to 10,000 values of F were generated and their distributions examined. For cattle this distribution of F was approximately normal, whereas for sheep it was skewed to the right. Coefficients of variation in percent for cattle in three experiments with decreasing herbage availability were 19, 24, and 20, whereas for sheep under identical conditions they were 31, 28, and 36. The practical implications of these studies are (1) more precise estimates of forage intake are obtained with cattle than with sheep, and (2) availability of herbage does not influence the precision of the estimate.

Systems Ecology: Prediction of Chemical Composition of Forage Components by Nonlinear Programming and Least Squares. — The relation between the composition of a given chemical in the ith mixed sample ($C_i$), the chemical composition of the jth species, plant part, or plant group in the sample ($P_j$), and the weight percentage of the jth species, plant part, or plant group in the jth sample ($W_{ij}$) is:

$$C_i = \sum_{j}^{m} (P_j W_{ij}) \ , \qquad (1)$$

where j = 1, 2, ..., m species, parts, or groups in the sample. Given data for $C_i$ and $W_{ij}$ we desire to estimate $\hat{P}_j$. This is accomplished by using least-squares and nonlinear programming routines to solve for the $P_j$ by minimizing a function Q as follows:

$$Q = \sum_{i}^{n} \left[ C_i - \sum_{j}^{m} (P_j W_{ij}) \right]^2 \ , \qquad (2)$$

subject to $0 \leqq P_j \leqq 100$ and i = 1, 2, ..., n samples. Known $P_j$ and $W_{ij}$ were used to test these techniques. Known variability of $P_j$ and $W_{ij}$ was obtained using random number generators, and known bias was included in some $W_{ij}$ values in 30 computer experiments. $\hat{P}_j$, predicted by the above methods from the generated $C_i$ and $W_{ij}$, were compared to $P_j$, the true values used in generating the data. Both least-squares and nonlinear programming gave acceptable $\hat{P}_j$ in most instances. Poor estimates of $\hat{P}_j$ were found only if $P_j$ were small and if the corresponding $W_{ij}$ were small and of low variability. These techniques were applied successfully to data from experiments with cattle and sheep to predict the crude protein content of five dietary species groups from the botanical composition and the crude protein in the mixed sample.

Evaluation of Fission Product Distribution and Movement in White Oak Drainage System. — The $^{137}Cs$ loss following 11.69 in. of rain amounted to 11.9% from plot 1 (bare), 5.1% from plot 2 (clipped meadow), and 2.6% from plot 3 (tall meadow). A linear relationship on a full log scale was observed between $^{137}Cs$ loss and soil loss. Although the total rainfall during this period was below normal, the intensity and the energy of the storms were higher than average. Of ten separate rainfalls, four rains,

in particular, were responsible for heavy soil and $^{137}Cs$ losses. The maximum intensity recorded was 3.60 in./hr for a total of 0.59 in. The four rains accounted for 91, 84, and 98% of the total soil loss and 81, 75, and 99.9% of the total $^{137}Cs$ loss for plots 1, 2, and 3, respectively, during the period.

Following 7.60 in. of rain, analysis was made of the $^{137}Cs$ content on the vegetation of plots 2 and 3. On plot 2 (clipped meadow), 12.5% of the cesium originally on the plant material was still remaining and 29% was still remaining on the clippings lying on the surface. On plot 3 (tall meadow), 39% of the cesium still remained on the vegetation.

## RADIOLOGICAL PHYSICS, HEALTH PHYSICS, AND RADIATION INSTRUMENTATION

Radiation Physics: X-Ray Investigation of the Pyrene and Naphthalene Excimers. — Pyrene oxygen-free solutions in cyclohexane and benzene have been excited by x rays, and the monomer-excimer spectra were studied as a function of x-ray intensity, x-ray energy, and solute concentration. A comparison was made of the shape and the relative excimer-monomer emission intensities as obtained by x-ray and ultraviolet excitation.

Data were obtained for the liquid emission spectra of 1,6-dimethyl naphthalene, 2-ethyl naphthalene, and 1,2-dimethyl naphthalene at various temperatures. The data were analyzed to determine the excimer binding energy.

Radiation Physics: Time-of-Flight Studies of Electron Transport in Gases. — Electron swarm experiments may be arranged to satisfy the boundary conditions applicable to a one-dimensional time-dependent transport equation. Under such conditions it may be shown that the distribution of the time-of-arrival of individual electrons at a point on a plane which is L centimeters from another plane releasing electrons at time $t = 0$ may be interpreted in terms of the diffusion coefficient D and the drift velocity w. The integrals of the distribution functions, that is, the total numbers of electrons arriving, before and after the addition of an attaching gas, are directly related to the attachment coefficient $\beta$. Since, in such experiments, time is a more accessible independent variable than is length, it is not surprising that much more information on the basic character of electron transport in gases is obtained than can be obtained from a Townsend experiment.

The original apparatus [J. Chem. Phys. 39, 1341 (1963)] has been improved by providing a direct experimental means for measuring the instrument fluctuation arising from such sources as variation in time of electron release from the photoelectric surface and the variation in electron detection time. Correcting for these fluctuations leads to improved accuracy for the measurement of D. New results are (1) better values of D for $C_2H_4$ as a function of E/P, (2) values for D in mixtures of $C_2H_4$ and $H_2O$, and (3) the ratio of the momentum transfer cross section in $H_2O$ to the momentum transfer cross section in $C_2H_4$.

The first of these methods used a gas-discharge single-electron detector and hence is limited in application to counting gases. A new method has been developed which employs a surface-type electron multiplier and is separated from the reaction chamber by differential pumps; hence the restriction to counting gases is removed.

Radiation Physics: Magnetodynamic Instability in a Plasma-Beam System. — Two types of magnetodynamic (hydromagnetic) instabilities have been analyzed. One type is produced in an ambient plasma by a stream of protons moving in the direction of an applied magnetic field $B_0$ (linear beam), and the other is produced by a stream of protons gyrating around the lines of the magnetic field (helical beam). In the case of a linear beam, there is a P instability; that is, the magnetodynamic waves excited by the beam have frequencies and phase velocities which differ very little from the magnetodynamic waves which propagate in the undisturbed plasma. In a helical beam the presence of a transverse velocity component of gyrating protons causes an additional B instability. The magnetodynamic B waves ("beam waves") occupy a relatively wide frequency range in the neighborhood of a P wave ("plasma wave") and represent a continuation of a P instability along the frequency axis. It is found in some instances that the qualitative behavior of instabilities is significantly different when the beam intensities increase from relatively small to relatively large values. When the beam is very tenuous, the magnetodynamic B instability is "weak"; that is, the rate of growth is smaller than that of the corresponding "strong" P instability. The contrast between the weak B instability and the strong P instability decreases for increasing beam intensity, and for less tenuous beams the situation is reversed; that is, the P instability is weak, whereas the B instability is strong.

## ISOTOPE DEVELOPMENT

Stable Isotopes Development. — Both ion retention and isotopic purity of $^{112}Sn$, which is used as target material for the preparation of $^{113}Sn$ and $^{113m}In$, have increased through the use of a different charge material, SnS plus $H_2S$. Ion retention (percent of ions monitored into the receivers and recovered) increased from 25.8 to >64%. Preliminary assays showed an increase from 73.6 to 83.3% in isotopic purity of $^{112}Sn$ (natural abundance, 0.95%). Elemental chlorine from the decomposition and ionization of $SnCl_4$, which was used as feed material, had chlorinated some of the isotopes and prevented the attainment of higher isotopic purity and ion retention.

The titanium series terminated with the purest isotopic species yet collected. The chemical pump concept of vaporizing calcium metal into the tank concurrently with the isotope collection was used to absorb neutral contaminants. The isotopic purity of $^{46}Ti$ increased from 86.4 to 90.1%, and that of $^{50}Ti$ from 84.1 to 87.8%.

Specific atoms can be implanted in selected crystals by ion bombardment, to evaluate the feasibility of preparing semiconductors by this method. During the past month, ~112 silicon and glass substrates were bombarded with aluminum, chromium, and phosphorus. The substrate materials were subjected to ionic dosages of $10^{17}$, $5 \times 10^{14}$, and $2 \times 10^{10}$ atoms/cm$^2$ at energy levels of 40, 30, 20, and 14 kev.

Special Separations. — Runs are being made to collect gram quantities of $^{230}Th$, $^{242}Pu$, and $^{236}U$. To date, ~1.2 g of thorium containing ~90% $^{230}Th$, 5 g of 99% $^{242}Pu$, and ~20 g of 94% $^{236}U$ have been separated. Two other uranium isotopes, $^{234}U$ and $^{235}U$, are being collected simultaneously

in the $^{236}U$ series. When sufficient quantities of enriched material are available, second-pass separations will be made to obtain ultrahigh-purity samples of uranium isotopes.

Target Preparation. — Approximately 10-g quantities of $^{40}Ca$ and $^{44}Ca$ were prepared by simultaneous oxide reduction and metal distillation. Calcium was condensed into quartz cylindrical receivers in the form of compact metal pellets which were then pressed into rectangular plates containing 2.5 g of the desired isotope. Use of these targets in electron scattering experiments has proven their integrity and low impurity content. Spectroscopic analysis showed that <30 ppm of lanthanum reductant was contained in the product calcium.

Strontium, europium, and erbium metals have been produced in small quantities suitable for rolling. The condensed strontium and erbium metals were highly reactive toward atmospheric gases and had to be kept in an inert atmosphere; however, both metals were successfully rolled into thin foils and could be used as accelerator targets. Erbium metal foils of various isotopes have been prepared by deposition on a nickel substrate followed by mechanical stripping of the film.

The evaporator system for fabrication of 14-in.-diam targets for project HENRE is complete. Vacuum levels of $10^{-7}$ torr have been achieved in the 32-in.-diam vacuum chamber without system bakeout; during erbium evaporation the chamber pressure decreased even further. The tritium sorption system and the glove box in which the system will be installed are nearly finished.

More uniform sorption and higher saturation of the titanium or erbium-coated copper disks with tritium have been achieved by using the ion-pumped tritium sorption system with an ultrahigh vacuum level instead of the oil diffusion systems previously used. Higher neutron yields for targets with metal coatings of 1 to 2 mg/cm$^2$ and a longer target half-life under deuteron bombardment have resulted. Oil contamination of the target sorption surfaces and oxidation of sorption sites have been reduced.

Radioisotope Research and Development. — A heat transfer study was conducted on a 5600-lb "Tucson" shipping container, 48 in. in diameter by 67 in. high with 9 in. of lead shielding, to determine if excessive heat would be given off during the shipment of 192,000 curies of $^{137}CsCl$ to France. A special block to hold the capsules in the carrier was built and was tested with four 250-w heaters to simulate the heat output of the shipment. The maximum temperature to be expected at the center of the $^{137}CsCl$ capsules will be ~400°C at equilibrium, while the temperature at the surface of the cask will be 90°C, and the hottest part on the fire shield will be 52°C. These temperatures are within the limits allowed by current shipping regulations.

Another test was conducted to determine the effect of $^{137}CsCl$ on the stainless steel source capsules during the ~30 days required in transit. Two loaded capsules were placed in the carrier and held at a temperature of 450°C for ten days. No evidence of interaction between the capsules and the source material was detected.

High-specific-activity $^{198}Au$ ($T_{1/2}$ = 2.7 days) was produced by epithermal neutron capture [$^{197}Au(n,\gamma)^{198}Au$], while the thermal neutron capture reaction [$^{198}Au(n,\gamma)^{199}Au$ ($T_{1/2}$ = 3.14 days)] was greatly reduced by

a cadmium shield. Samples of 1-mil gold and 0.13% gold-aluminum alloy were irradiated with and without cadmium shields for 5.5 days in the ORR at a monitored thermal flux of $1.3 \times 10^{14}$ neutrons $cm^{-2}$ $sec^{-1}$ and an epithermal flux of $1.1 \times 10^{13}$ neutrons $cm^{-2}$ $sec^{-1}$. Data given below indicate that the production of $^{199}Au$ can be greatly reduced by the cadmium shield, which removes the thermal component of the reactor neutron flux without greatly reducing the specific activity of $^{198}Au$. The production rate and the specific activity of $^{198}Au$ can be optimized by irradiating targets in the highest possible epithermal flux, which would be obtained in the reactor core near a fuel element.

Comparison of Specific Activity and $^{199}Au$ Content of $^{198}Au$ Prepared With and Without Cadmium Shields

| Sample | Without Cadmium Shield | | With Cadmium Shield | |
|---|---|---|---|---|
| | Specific Activity (curies/g) | $^{199}Au$ (%) | Specific Activity (curies/g) | $^{199}Au$ (%) |
| 0.13% Au–Al alloy | 530 | 77 | 370 | <1 |
| 1-mil gold foil | 495 | 65 | 135 | <1 |

To determine optimum conditions for cyclotron production of $^{197}Hg$ ($T_{1/2}$ = 65 hr), a stack of 28 1-mil gold foils was irradiated. Mercury-195 ($T_{1/2}$ = 9.5 hr), produced by a (p,3n) reaction, predominated in the initial foils in the stack; $^{197m}Hg$ ($T_{1/2}$ = 24 hr) and $^{197}Hg$ were produced in the deeper layers of the stack. Mercury-197m,197 produced by the (p,n) reaction from gold contains a much greater proportion of the 24-hr isomer than does $^{197m,197}Hg$ produced by the (n,$\gamma$) reaction on $^{196}Hg$.

Preliminary spectral and decay data indicate that the half-life of $^{197}Hg$ is 63.7 hr instead of 65 hr and that the internal conversion of both gamma rays in the isomeric transition of $^{197m}Hg$ produces no K x rays. A sum-coincidence method of assay shows promise as a suitable absolute assay method for both reactor- and cyclotron-produced $^{197}Hg$ preparations.

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