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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

APRIL 1963

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OAK RIDGE NATIONAL LABORATORY

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Oak Ridge, Tennessee
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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

APRIL 1963

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

REACTOR DEVELOPMENT PROGRAM

GAS-COOLED REACTOR PROGRAM

EGCR Startup Program. — The joint ORNL-TVA-ORO task force, set up to coordinate work on the physics aspects of the EGCR startup program, drew up an outline of tests to be performed and began writing detailed test procedures. An improved theoretical treatment was developed for describing the transient behavior of the EGCR during measurements of a subcritical multiplication factor with a neutron pulser. Group constants were computed for use in six-group calculations of the effective delayed-neutron fraction and the mean prompt-neutron lifetime required for interpretation of pulser measurements.

New calculations were made of the physics parameters for the lattice configurations used in the critical experiment performed in the Hanford Physical Constants Test Reactor for the EGCR program and in the HERO critical loadings at Windscale, England. New calculations were also made for the EGCR lattice, using dimensions and weights obtained from finished fuel elements, in contrast to estimates from drawings. Among other changes, the new calculations based on crystal-binding-model scattering kernels result in lowering the estimated infinite multiplication factor by about 0.01, which improves agreement with experimental data for 2.6%-enriched fuel but is a little less satisfactory for 1.8%-enriched fuel. This discrepancy is being investigated.

AVR (German Pebble-Bed Reactor) Fuel Studies. — A fueling scheme in which criticality is maintained by simultaneously removing original fueled balls and adding fresh fueled balls is being studied. In all cases considered, one-third of the original balls were fueled with 5 wt % fully enriched uranium plus thorium. The remaining balls contained graphite or borated graphite.

In one set of calculations a thorium-to-uranium ratio of 7.8 in the fueled balls was considered. In this scheme criticality is initially maintained by the removal and burnout of borated balls. When all the borated balls have been removed, criticality is maintained by the fuel-exchange method. In a second set of calculations a thorium-to-uranium ratio

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of 11.04 was used. In this series the fuel-exchange method is used throughout the reactor operating lifetime.

The fresh fueled balls were assumed in all cases to contain (1) the same materials in the same concentrations as the original fueled balls, (2) the same uranium concentration but only one-half the original thorium concentration, or (3) the same uranium concentration but no thorium. The only fueled balls removed from the core were those originally in the core. The total number of balls in the core was held constant. Any differences in the number of fresh balls added and original balls removed were made up with plain graphite balls.

The possibility of obtaining a flat temperature distribution for outlet gas by using unequal radial fuel loadings is being investigated. The core in these calculations is assumed to be divided into two equal-volume radial regions. The average fueled fraction of the core is fixed at one-third; however, each region has a different fueled fraction. Preliminary results for balls with a thorium-to-uranium ratio of 7.8 indicate that a loading in which 15% of the central-region balls are fueled and 48.3% of the outer-region balls are fueled will give very nearly an equal average outlet-gas temperature over the two regions. The temperature distributions for other thorium-to-uranium ratios and the expected fuel lifetimes are being investigated.

The control-rod worth in the AVR was calculated by four-group, two-dimensional diffusion theory, with the specification of effective boundary conditions at the rod surfaces. Full insertion of all four rods gives a δk of 8.8% in the case of a core uniformly fueled with balls with a thorium-to-uranium ratio of 7.8, and full insertion of three rods gives a δk of 6.2%. The temperature defect necessary to raise the core-moderator temperature from 300 to 900°K and the fuel from 300 to 1150°K appears to be about 6.3%, and the reactivity increase associated with the decay of equilibrium xenon is about 2.5%. It appears that a less heavily loaded core may give a higher worth for the control rods. The composition for the fuel originally planned for the AVR gives a control-rod worth of 12 to 13%.

Preliminary temperature-coefficient calculations at operating temperature gave the following results:

Core-moderator coefficient	$-6.1 \times 10^{-5}/°C$
Doppler coefficient	$-0.4 \times 10^{-5}/°C$
Reflector coefficient	$+0.5 \times 10^{-5}/°C$

These calculations were for a core containing equilibrium Xe^{135} and Sm^{149}, but no other fission products, and having a thorium-to-uranium ratio of 7.8.

Pyrolytic-Carbon Coating of Fuel Particles. — Two coating tests were made at 1400°C in a mullite tube furnace and two were made at 1800°C in the new high-temperature furnace. The base particles used were spheroidal uranium carbide particles of −60 +100 mesh (147 to 246 μ). The microstructures of the coatings applied at 1400°C were controlled by regulating the CH_4 partial pressures and flow rates, which, in effect, control the deposition rates. Limited data indicate that at this temperature a columnar structure is formed at deposition rates lower than 12 μ/hr and that

a laminar structure results at a rate of 25 μ/hr. It appears that deposition rates depend not only on the CH_4 pressure but also on reactant flow rate.

The tests at 1800°C were terminated after 2 hr because of carbon buildup at the entrance to the coating chamber. This buildup was probably a result of inadequate cooling of the gas supply. The supply-tube design is being changed to provide additional cooling of the entering gas. The coatings deposited at 1800°C were of a uniform but poorly developed columnar structure. Both the radiographs and polished sections revealed a layer, possibly a reaction zone, about 2 to 4 μ in thickness, between the particles and coatings. Methods being considered for identifying this layer are electron-probe microanalysis and x-ray diffraction analysis of a sample removed from the specimen by means of a microchiseling technique.

Fission Product Deposition. — An out-of-pile loop test was run in which irradiated uranium carbide was heated at 930°C to study the deposition of the released fission products on the loop walls as a function of wall temperature. This test differed from previous tests in that a section of the tubing in the normally cold zone, 11 in. above the sample, was heated to 300°C. In previous tests a large increase in iodine, which did not plate out on the wall but was caught in the end traps, occurred when a blower was provided to cool the walls to below 100°C. This indicated that at low temperatures the kinetics of the required chemical reaction might be too slow to cause the iodine to plate out and that if iodine plating were due to a chemical reaction, the current test should show a high I^{131} activity peak in the heated region and a decrease in trap activity. These predicted effects were observed; therefore it appears that iodine does plate out by a chemical reaction with the walls. Quantitative data on the I^{131} plated out are now being determined.

Instantaneous Fission-Gas Release from Coated Particles. — Capsule B9-10, which contained a third group of pyrolytic-carbon-coated uranium carbide particles from batch NCC-AD, was irradiated at 2100°F to 3.8 at. % uranium burnup. The average fractional fission-gas releases were between 1×10^{-6} and 7×10^{-6} for the five isotopes measured. Postirradiation visual inspection at 30× showed a few broken particles. The broken particles appeared to contain holes about one-third the particle diameter in size.

Capsule B9-11, which contained pyrolytic-carbon-coated uranium-thorium carbide particles from batch NCC-208-2, was irradiated at 2050°F to 8 at. % uranium burnup. The uranium-to-thorium ratio was about 6 to 4. The fractional fission-gas release rates were: Kr^{85m}, 1.1×10^{-6}; Kr^{87}, 0.6×10^{-6}; Kr^{88}, 0.8×10^{-6}; Xe^{133}, 2.4×10^{-6}; and Xe^{135}, 0.5×10^{-6}. These release rates are the lowest observed to date in this series of experiments. The release rates were very nearly constant over a 22-day irradiation period and did not increase with burnup. This experiment is continuing.

Coated-Particle Irradiations in ORR Capsules. — The helium-sweep capsule containing 1-1/2-in.-diam balls, manufactured by the National Carbon Company, of duplex pyrolytic-carbon-coated particles uniformly distributed in a graphite matrix, continued to operate without detectable release of fission gases. The capsule, which is being irradiated at a fuel surface

temperature of about 1500°F, has reached an estimated burnup of 3 at. % heavy metal. A second sweep capsule, containing pyrolytic-carbon-coated fuel particles in graphite inserts in machined graphite balls, continues to show a ratio of fission-gas release rate to birth rate of $\sim 4 \times 10^{-5}$ after 7 at. % heavy-metal burnup. Three additional capsules containing fueled-graphite balls manufactured by various vendors were assembled for irradiation.

High-Temperature UO_2 Irradiation in LITR Capsules. — The series of high-temperature UO_2 irradiation tests was completed. Each capsule was operated at a central temperature of ~2000°C. Irradiation periods for the four capsules were ~1 day, 1 week, 1 month, and 3 months. Temperature indications obtained with tungsten-rhenium thermocouples were unstable at high temperatures and drifted downward rapidly to lower levels of the order of 1800°C in a few hours or a few days. Relatively stable temperatures were observed for the remainder of the longer irradiation periods.

Irradiations of Advanced Clad Fuel Elements in GCR-ORR Loop No. 1. — A roughened-surface element having a heat rating estimated at 65,000 Btu hr^{-1} ft^{-1} operated satisfactorily for approximately two weeks and then failed during a calibration test. At the time of failure, the reactor power was 22 Mw, and the gas flow over the elements had been reduced to approximately one-half the normal flow rate. The test in progress was to establish different operating conditions from which heat-balance data would be used to establish limits of accuracy for power-generation measurements. Failure of the element resulted in radiation levels in the loop of 100 to 150 mr/hr, measured at contact with the primary piping. The activity dropped rapidly to near-normal levels when the element was withdrawn from the reactor.

GCR-ORR Loop No. 2. — Operation of GCR-ORR loop No. 2 continued without incident. A cell purge test was run to determine whether diffusion of Ar^{41} from the in-pile annulus could be controlled by a sweep flow of air. The test demonstrated that personnel could work in the cell during cold-finger insertion with the reactor at power. Samples of loop gas were analyzed daily for fission products. The total gaseous activity in the loop built up to approximately 800 μμc/ml with the cleanup system closed off and to approximately 140 μμc/ml with the cleanup system in operation. The fuel configuration of the reactor was changed during midcycle refueling to provide an additional fuel element adjacent to the loop No. 2 beam hole. Flux plotting of the beam hole is in progress to assess the effect of this change.

Operation of the set of two compressors installed in the loop continues to be satisfactory. This is the third reactor cycle of continuous operation.

The low-pressure graphite-oxidation and mass-transport experimental specimen operated for 450 hr at a specimen temperature of 1820 to 1830°F. Subsequent examination of the graphite specimen showed a weight loss of 0.87 g of graphite. The control of the CO and CO_2 levels during the test was erratic, however, because of poor flow control through the purification system. Consequently, the initial weight-loss measurement is not considered to be significant.

MARITIME REACTORS PROGRAM

NS "Savannah" Filter Testing. — In-place tests were conducted on the revised reactor compartment ventilation system of the NS "Savannah" while the ship was in the Todd Shipyards, Galveston, Texas. Participation by the Laboratory in this series of tests was less than in previous tests, as a result of efforts to assist the ship operator, States Marine Lines, to acquire the capability of performing these tests independently. The DOP aerosol tests of the particulate filters were performed by States Marine Lines personnel who had been trained at ORNL. A Laboratory observer was present, but assistance was not required. Both SML and ORNL personnel participated in tests of the iodine adsorption units. The iodine removal efficiency was measured by the radioiodine-tracer procedure and by normal-iodine activation analysis. It is anticipated that future DOP and nonradioactive-iodine tests will be carried out by the operator without assistance from ORNL.

Results of the DOP tests showed the efficiency of the particulate filters to be $> 99.98\%$. The I^{131} tests showed the iodine removal efficiency of the system to be $> 99.96\%$. Analyses of samples from the nonradioactive-iodine tests have not been completed.

Port Surveys. — Assistance was provided in surveying a number of U.S. and European ports which are to be visited by the "Savannah" in the next few months. The port survey teams were made up of a senior deck officer of the ship and a nuclear engineer from the Laboratory. The purpose of the advance visits was to select berth and remote anchorage locations, to inform the local authorities of safety procedures and requirements, and to make operational arrangements. During the current report period, surveys were made of the ports of Baltimore, Boston, New York, Bremerhaven, Hamburg, Oslo, Copenhagen, and Southampton. Discussions of port-survey procedures were held with Euratom officials in Brussels and in Rotterdam.

ARMY POWER REACTORS

The ORR pressurized-water loop is, as a result of a program change, to be used for irradiation testing of advanced cermet fuels. A mockup of the first series of specimens to be tested was irradiated in the loop to confirm the analytical methods being used to design the tests. Preliminary analysis of the mockup data indicates that the analytical techniques being used are valid.

An unfueled mockup of the proposed specimen assembly is in the loop, undergoing flow tests. It will be examined prior to insertion of the actual experimental assembly in the loop to determine whether any design modifications will be required in the fuel holders because of wear, corrosion, or any other problem.

The design of the experimental-fuel holders and fabrication of their parts are complete. Equipment for pre- and postirradiation measurement is being designed. Loop modifications and experimental-hazards analyses were started.

FUNDAMENTAL HEAT TRANSFER AND FLUID DYNAMICS

Transient Heat Transfer. — Work continued on the installation of apparatus for studying heat transfer. Tests for determining a procedure for brazing Chromalox heaters to the solid stainless steel test section demonstrated that the best results were obtained if the test section and heaters were nickel-plated before they were brazed with Nicrobraz-50 alloy. The plating is being done. The interior of the 200-gpm vacuum deaerator was treated with a rust-preventative paint (Apexture), and the Raschig-ring packing was installed. Checkout of the system is under way, and a number of minor modifications are being made to facilitate operation.

Boiling-Potassium Studies. — The new high-flux boiler was received from the vacuum-brazing operation in a damaged condition. The damage was apparently incurred in transit. The boiler will probably have to be disassembled, machined, reassembled, and rebrazed; this will delay operation by approximately three months.

Additional study of the superheat required to initiate vapor bubbles in liquid metals indicates that a typical surface which requires a 30°F superheat to initiate a vapor bubble with water at atmospheric pressure may require a superheat of approximately 100°F with potassium at atmospheric pressure and one of approximately 250°F with sodium at a comparable pressure. For very smooth surfaces, the superheat for bubble formation with potassium may be as high as 320°F, based on comparison with data for degassed, distilled water. Temperature oscillations indicating superheats of approximately 200°F have been observed in forced-circulation and natural-circulation boiling-potassium loops.

POWER REACTOR FUEL PROCESSING

Hydrolysis of Thorium Carbides. — The amount of uranium in uranium carbides is readily determined gravimetrically by burning the specimen in oxygen at 950°C, cooling, and weighing the residual oxide (U_3O_8) at room temperature. Unfortunately this procedure is not satisfactory for thorium carbides. The finely divided ThO_2, even after being heated at 1250°C, gains weight through surface sorption during cooling, resulting in a significant error (about 2%) in the thorium analysis. This difficulty has been overcome by burning weighed samples in a thermobalance and determining the weight of the ThO_2 at 950°C.

The ignition temperature in oxygen for thorium dicarbide is about 540°C, and for thorium monocarbide about 630°C. At 650°C, 95% of the thorium dicarbide reacted within 30 min, whereas only 85% of the monocarbide reacted after 3 hr. Both samples were −10 +50 mesh. The use of particles larger than 10 mesh is not recommended for this analysis, since the ThO_2 product tends to form a protective barrier that hinders further oxidation.

Extraction of Protactinium from Nitric Acid–Hydrofluoric Acid Solution. — Multiple-extraction and Martin-technique experiments using tracer

Pa^{233} have demonstrated that essentially all the protactinium in 5 M HNO_3 is extractable in 30% tributyl phosphate in n-dodecane. The protactinium distribution coefficient is about 4. When 0.02 M HF was added to the 5 M HNO_3, about 94% of the protactinium became unextractable, and the distribution coefficient of the extractable species was reduced to about 0.07, indicating that almost all of the protactinium was complexed by fluoride.

Protactinium Adsorption. — The adsorption coefficient of unfired Vycor for protactinium adsorption from 10 M HNO_3 was decreased by factors of 10, 100, and 1000, respectively, when 0.01, 0.02, and 0.03 M HF was present. About 7-1/2 moles of aluminum per mole of fluoride was required to completely complex the fluoride in solutions containing 0.04 M HF, so that the distribution coefficient was not affected. The distribution coefficient was approximately 1400 after a 24-hr batch equilibration between 10 M HNO_3, about 6 g of thorium per liter, 1.7×10^6 counts of Pa^{233} per minute per milliliter, the fluoride, and 20 g of 60 to 80 mesh unfired Vycor per liter. With feed solutions containing 112 g of thorium per liter, the coefficient decreased from 1200 with no fluoride present to 345 when the fluoride concentration was 0.04 M. The presence of 7-1/2 moles of aluminum per mole of fluoride increased the coefficient to about 800.

Chemical Analysis of Advanced Reactor Fuels. — The studies on the use of NaF in concentrations of ~0.75 M as an electrolyte in the coulometric titration of uranium were continued. A synthetic fuel typical of that used in the Experimental Boiling Water Reactor (EBWR) was prepared and analyzed for uranium by the controlled-potential coulometric method. The synthetic fuel was made to contain 0.07 M $UO_2(NO_3)_2$, 0.8 M $Al(NO_3)_3$, 0.8 M NaF, 0.04 M $Zr(NO_3)_4$, and 5 M HNO_3. Samples of 0.5 ml, each containing 9.07 mg of U(VI), were fumed with H_2SO_4, diluted to 10 ml with 0.75 M NaF, adjusted in pH to 7.0 with dilute NaOH, then titrated at a potential of −1.0 v vs the S.C.E. The results are shown below:

Sample	Titration Time (min)	Uranium Found[a] (mg)	Deviation (mg)
A	25	9.08	+0.01
B	21	9.08	+0.01
C	24	9.08	+0.01
D	26	9.03	−0.04
E	22	9.03	−0.04
F	28	9.02	−0.05
G	20	9.06	−0.01
H	20	8.99	−0.08
		Av 9.05	−0.02

[a]9.07 mg taken.

These results show that NaF is a satisfactory electrolyte for the determination of uranium in EBWR fuel solution. The method is essentially free of bias. The standard deviation is of the order of 0.5% in solutions containing 20 mg of U per ml.

THORIUM UTILIZATION PROGRAM

Thorium Materials Development. — Stress rupture tests at 800°C were made on specimens from an extruded thorium compact hardened with 10 vol % ThO_2 as a dispersion. The results of two tests indicate that this material has a minimum creep rate of about one-fifth that of the strongest thorium alloy evaluated by the Advanced Technology Laboratories.

Studies on fabrication techniques to produce dispersion strengthening of thorium and thorium-alloy powders are continuing. Stearic acid, used to aid in grinding to small powders, proved to be difficult to remove from the surface of the powders. Since stearic acid tends to embrittle thorium if allowed to react with it, all traces should be eliminated. Oleic acid, which is more volatile than stearic, is being tried as a substitute.

Hot-hardness tests on Th—5% Zr and Th—5% Zr—10% ThO_2 alloy pressings prepared from hydrides were made. The addition of the oxide increases the hot hardness at 800°C by about 50%.

Vibratory Compaction. — Fourteen irradiation capsules, 7/16-in. OD × 0.025-in. wall, with type 304 stainless steel cladding, were filled with thoria-urania oxides prepared by the sol-gel process. Bulk densities from 8.9 to 9.0 g/cm^3 were obtained by vibratory compaction. The experiments are designed to determine the effect, on irradiation performance, of the atmosphere used in the sintering step of the sol-gel process. Air, nitrogen, and a mixture of argon with 4% hydrogen were used.

Kilorod Program. — The installation and checking of the rod-fabrication part of the Kilorod Facility were completed; cold runs are in progress, using depleted uranium substituted for 3 wt % U^{233} in the ThO_2-UO_2 fuel. The startup period precipitated a number of minor changes to increase the efficiency of the process. Operators were trained during the startup period; therefore productive operation can begin immediately after completion of the cold runs.

Irradiation Results. — The postirradiation examination of a trefoil cluster irradiated in an ORR pressurized-water loop is in progress. The experiment (identified as L-1) had been prematurely discharged from the reactor because of a failure in one of the Zircaloy-2-clad fuel rods. Two of the vibratorily compacted ThO_2-UO_2 rods were clad with 0.460-in.-OD × 0.015-in.-wall Zircaloy-2 tubing, and the other was clad with type 304 stainless steel of like diameter and thickness. The rods were subjected to the following operating conditions: cladding temperature, 500°F; coolant pressure, 1750 psi; heat flux, 330,000 to 400,000 Btu hr^{-1} ft^{-2}; linear heat rating, 43,000 to 52,000 Btu hr^{-1} ft^{-1}; burnup, 2500 to 3000 Mwd per metric ton of oxide.

The failure, opposite the gas plenum above the fuel column, apparently resulted from a corrosion reaction; in addition, a crack propagated

around the periphery of the rod. The reaction is believed to be hydriding, initiated either internally by the release of absorbed gases or externally by the radiolytic decomposition of water. The degree of hydriding will be determined by metallographic and chemical analyses.

Macroscopic examination of the fuel from this rod clearly revealed sintering, but its extent remains to be determined. These rods were run with a higher heat rating and cladding temperature than any previous sol-gel experiments, so that the occurrence of sintering is not unexpected.

Fuel Cycle Development: Chemistry of Solid Fuel Element Materials. — Samples of urania-thoria solid solutions containing 70% or more urania displayed two-phase equilibrium when equilibrated in air at temperatures from 600 to 1500°C. In all cases x-ray diffraction patterns and metallographic examination identified the phases as U_3O_{8-y} and a cubic phase. Analytical data indicate that the phases are U_3O_{8-y} and the cubic solid solution containing 60 to 70 mole % urania.

In contrast, finely ground mixtures of UO_2, UO_3, and ThO_2 in similar proportions (70% or more uranium oxides) yielded the U_3O_{8-y} phase and essentially pure ThO_2 when equilibrated in air at 900°C. These results indicate that there is no interaction between U_3O_8 and ThO_2 and, therefore, that the appearance of U_3O_8 as a second phase from the solid solution is an irreversible process.

Fuel Cycle Development: Fuel Preparation. — In studies of the kinetics of the carburization of ThO_2 and of 5 mole % UO_2—ThO_2, where the reactant mixtures were prepared by the sol-gel process, reaction rate constants were increased tenfold by a 100°C temperature rise in the 1500 to 1700°C range. Increasing the carbon/metal mole ratio of the reactants from 4.3 to 5.1 doubled the reaction rates for corresponding temperatures, but a further increase in ratio produced no further effect. When uranium was also present, rates corresponding to those for ThO_2 alone were achieved at about 100°C lower. Carburizations were more than 99% complete in 4 hr of heating at 1500 to 1600°C when 5 mole % uranium was present.

Conductometric titration of the system thoria-carbon-water showed a critical nitrate demand similar to that for the system thoria-water for complete dispersion to stable sols. Gels obtained by evaporation of these sols were maximally reactive during carburization.

The size of spherical particles produced by spraying the sol into stirred CCl_4 can be controlled by varying the velocity of the tips of the agitator blade. A modified Reynolds number with respect to CCl_4 of 17,000 for the agitator tips gave particles having a geometric mean size of 194 μ, with a standard deviation of 1.5. The feasibility of redispersing off-size gel particles into a sol was established.

Addition by spray of 10 wt % water to a 3 mole % UO_2—ThO_2 gel that had been dried at 135°C shattered the dried gel and produced coarse- and medium-range particle sizes. Ball milling the same type of gel for 1 hr produced particles more than 95% of which were —200 mesh. Calcining a mixture of 75 wt % shattered and 25 wt % ball-milled gel by the sol-gel procedure produced an oxide mixture that was easily vibrated to 86% of theoretical density.

Twenty-seven stainless steel irradiation-test capsules containing sol-gel UO_2-ThO_2 or PuO_2-ThO_2 have been or are being irradiated at burnups of 5000 to 110,000 Mwd per metric ton of heavy metals. Other irradiation

conditions include linear heat ratings of 25,000 to 60,000 Btu hr^{-1} ft^{-1}, center-line temperatures up to 4000°F, and cladding temperatures to 1300°F. Seventeen specimens removed from the reactor were examined. Accumulated data indicate that the sol-gel oxides are potentially satisfactory fuels, with thermal conductivities of about that of UO_2 at corresponding temperatures.

MOLTEN-SALT REACTOR PROGRAM

Molten-Salt Reactor Experiment. — The second fuel drain tank and the radiator coil were completed in the Y-12 Shops. Installation of the radiator coil in the radiator enclosure was started. In the Paducah Shops, fabrication of the drain-tank furnaces and the freeze flanges was started.

Installation of support structures and auxiliary piping continued in the reactor cell, the drain tank cell, and the coolant cell. The off-gas holdup piping was installed.

The MSRE schedule was revised. The overall project was 69% complete on April 1; the new schedule shows an estimated 98% completion by June 30, 1964.

Reactor Analysis. — The nuclear characteristics of the MSRE were calculated for three different fuel compositions, using the most recent data and improved methods.

One fuel contained 1 mole % thorium and 0.29 mole % fully enriched uranium. Another contained no thorium and only 0.18 mole % fully enriched uranium. The third contained no thorium, but for reasons of fuel chemistry the uranium was raised to 0.8 mole % by reducing the U^{235} enrichment to 35%.

Critical concentrations were calculated by a 33-group diffusion code, using cross sections appropriate for the particular core composition. Flux and power distributions and reactivity coefficients were computed by a 20-region, two-group model with constants generated by the multigroup calculation.

Particular attention was given to improving the accuracy of the calculated temperature coefficients of reactivity. It was found that the dependence of the thermal-neutron spectrum on the fuel-salt temperature is not negligible, as had been previously assumed. Allowance for this effect reduced the graphite temperature coefficient but increased the fuel coefficient. It was also found necessary to raise the cutoff energy for the thermal group (to 1 ev at 1200°F) to eliminate a dependence of the calculated temperature coefficients on the cutoff energy.

Development of Components and Systems. — Fuel salt with a target composition of $LiF-BeF_2-ZrF_4-UF_4$ (66.9-29-4-0.1 mole %) was loaded into the fuel drain tank of the engineering test loop in March and treated with HF and H_2. A total of 113.7 g of H_2O, the equivalent of 625 ppm oxygen, was removed from 161.8 kg of salt during two weeks of treatment. After the loop and graphite had been flushed with flush salt, operation with the treated fuel salt at 1200°F was begun on April 4.

The installation of the fuel sampler-enricher prototype in the engineering test loop was completed, and testing was begun. All instrument

interlocks performed as designed, except for a rupture disk which had been specified at too low a rating. A disk with a slightly higher rupture point was installed, and testing was continued.

A prototype drain-tank heater was built, and operation at temperature was begun. The removable heater unit is designed for a total of 4.55 kw maximum heat output and contains eight ceramic heater elements enclosed in a type 347 stainless steel shell. The unit is operating satisfactorily.

The drain-tank cooler test was terminated because of cracks in two of the cooling tubes after 2600 thermal cycles simulating cooler startup conditions. Examination of the Inconel tubes is under way. The number of severe thermal cycles before failure in the test was greater by a factor of at least 10 than the number to which the drain tank coolers should be subjected in the MSRE.

Tests were conducted on a special version of the freeze-flange operating tool for the flange in the line between the reactor and the pump. Due to the crowded conditions in this area, some difficulty was encountered in engaging the jaws of the tool on the appropriate surfaces of the flange. However, once engaged, the tool operated satisfactorily. It was concluded that with some slight modifications to the support structure in the reactor cell and some alterations to the design, this tool will perform adequately in the MSRE.

MSRE Pump Program. — The MSRE prototype fuel pump has operated satisfactorily for 4100 hr, circulating molten $LiF-BeF_2-ZrF_4-ThF_4-UF_4$ (70-23-5-1-1 mole %) at 1200°F, 1200 rpm, and 1200 gpm. Additional measurements of the diffusion of radioactive gas up through the pump-shaft annulus against a purge flow of helium indicated no diffusion against purge flow rates greater than 50 cc/min. The concentration of undissolved blanket gas (helium) in the circulating molten salt was measured to be 1-1/2 to 2% by volume at the above-mentioned pump operating conditions. Additional measurements of gas concentration will be made at lower pump speeds and flows.

The endurance test of the Inconel centrifugal molten-salt pump (PKP type) was interrupted by a failure in the electrical insulation of the drive motor. The pump had been operated continuously for 9816 hr, circulating molten $LiF-BeF_2-ThF_4-UF_4$ (65-30-4-1 mole %) at 1225°F, 1950 rpm, and 510 gpm. While the drive motor is being repaired, the pump will be inspected and the lower shaft seal will be replaced with one containing a Graphitar nosepiece sealed to the mounting bellows assembly with a clear baking varnish.

Instrumentation Development. — Long-term tests of thermocouples, a ball-float level transmitter, a single-point level probe, and a temperature scanning system were continued with satisfactory results. The prototype temperature scanning system was moved from the level test facility to the MSRE engineering test loop in order to permit operating personnel to gain experience in operating this system and in order to evaluate the scanner performance under conditions which more nearly approximate those to be found in the MSRE.

Fabrication of components for a second ball-float level transmitter is complete. Installation of this device on the MSRE pump-test loop is being delayed pending completion of other tests on the loop.

A program to investigate the feasibility of using resistance elements for obtaining precise temperature measurements at MSRE temperatures was initiated. Procurement of four commercially available resistance elements, rated for short-term operation at 2500°F, was initiated, and preparations are being made to determine the maximum temperature at which stable, drift-free measurements can be made with these elements.

Evaluation of methods and devices for obtaining three-dimensional closed-circuit television viewing during remote-maintenance operations was continued. Existing TV equipment was reconditioned and set up in the MSRE remote-maintenance facility. Comparative evaluation of a two-channel and a single-channel stero TV system is in progress.

NUCLEAR SAFETY

Release of Fission Products on Out-of-Pile Melting of Reactor Fuels. — A few intermediate-level-activity experiments in hot-cell equipment were performed, in order to test ease of operation and safety of containment for higher-level hot-cell release experiments involving large quantities of fully irradiated UO_2. A low-melting fuel, an aluminum alloy irradiated to 23.6% burnup, was used in these tests.

No data on the burnup effect in aluminum-alloy fuel has been previously reported; consequently, a comparison was made with the tracer-level release data reported earlier (ORNL-2616). While comparisons are limited by the small number of results obtained in tracer-level experiments, it appears that all release values are increased by a factor of almost 10 by full irradiation. This corresponds closely with the effect of burnup on fission product release during the oxidation of UO_2 and the melting of metallic uranium observed previously. The efficient release of iodine from this alloy is being used to advantage in the testing of the filters and the charcoal iodine adsorption system proposed for the ORR and the HFIR.

Release of Fission Products on In-Pile Melting of Reactor Fuels. — The tenth experiment on in-pile melting of miniature UO_2 fuel elements has been performed. Radiochemical analysis is approximately 75% complete. This experiment was the first of a series to investigate the effects of carrier-gas flow on the release of fission products. The gas flow in the tenth experiment was twice that used in the first nine.

The F-3 facility, in which fuel for future meltdown experiments is preirradiated to obtain high burnup, is operating satisfactorily.

Melting Experiments in TREAT Facility. — Approval was obtained for ORNL fission product release studies in the TREAT facility at the NRTS following a review of the hazards analysis by TREAT management. The release of fission products during transient melting, in an argon atmosphere, of a 10%-enriched UO_2 compact clad with type 347 stainless steel will be investigated in experiments conducted in this reactor. The samples will be preheated to temperatures in the 800 to 1200°C range, and the released aerosol will be sampled by admission to an evacuated autoclave through a diffusion tube and filter.

Two experimental assemblies are being built for the first irradiations. These experiments will use 30 g of 10%-enriched UO_2 clad with type 347 stainless steel and preheated at 800°C in 1 atm of argon. The reactor energy release will be approximately 250 Mw-sec with a period of 100 msec. A preliminary low-energy transient will be performed during the first experiment, so that thermocouples on the sample cladding will show the exact relation between reactor energy release and sample heating rate.

Characterization and Control of Accident-Released Fission Products. — Studies pertaining to the removal of ultrafine radioiodine-bearing particles from air streams have been continued, using the diffusional deposition technique. In some of these tests, evidence that part of the airborne iodine was neither elemental nor particulate was observed. To eliminate the possibility that the spark gap used in generating particles was a factor in this effect, tests were performed in which intentional particle generation was omitted and, also, the air was rather elaborately filtered before it was mixed with the continuously injected iodine vapor. The resulting concentration of the latter, $I_2^{127} + I_2^{131}$, was about 10^3 $\mu g/m^3$. The air-iodine mixture flowed through a volume which was at 25°C and which permitted about 6 sec of reaction prior to side-stream sampling at the diffusion-tube port. The deposition data indicated the presence in appreciable quantities of two distinct species of iodine other than elemental. One species appeared to be retained almost quantitatively by the inner surface of rubber tubing and the other by activated carbon, in both cases at the low and not prolonged diffusion flow used. Neither was quantitatively retained by silver metal. British workers in this field have previously reported that their data indicated some formation of iodine compounds when elemental iodine is heated with atmospheric air. Experiments are being initiated to determine more accurately the quantities and, if possible, the identities of each of the two or more species observed in the diffusional deposition experiments. At the same time, the question of their universality will be investigated.

Nuclear Safety Pilot Plant. — The shielded cell in which the pilot plant is to be located was repainted, and the model containment vessel was set in place. All other components except the furnace are on hand. The furnace is being fabricated, with completion scheduled in June 1963. Shop fabrication of piping is under way, and field installation is scheduled to begin in May.

Mechanical design of the in-cell components is complete except for the recycle loop. Design of the out-of-cell equipment was started. Electrical drawings are 50% complete. Instrument procurement is substantially complete; detailed design is just starting.

Eight remote-handling tools were built. Several modifications to existing working shields were designed and are being fabricated. A periscope for observing fuel meltdown and manipulation of radioactive components was ordered.

Reactor Containment Handbook. — A preliminary draft of Chapter VIII, "Specific Containment Systems," was completed and is being reviewed. Preliminary drafts of Chapter V, "Analytical Techniques," and Chapter III are 50% complete. Arrangements were made with Stanford Research Institute,

Armour Research Foundation, and Ballistics Research Laboratory for the preparation of material on their containment research for inclusion in the handbook.

Chapter II, "Codes, Criteria, and Regulations," and Chapter IV, "Energy Sources," were issued as ORNL CF memorandums.

The Bechtel Corporation completed preliminary drafts of four chapters under subcontract. Chapter VII, "Containment Proof Tests," was reviewed, and comments were submitted to Bechtel. Chapter IX, "Material Properties," Chapter X, "Design Details and Accessories," and Chapter XI, "Economics," were received for review.

Nuclear Safety Information Center. — Agreement was reached with the AEC with respect to the establishment of the Nuclear Safety Information Center at ORNL. The specific subjects included within the initial scope of the Center are containment, fission product release, meteorology, reactor instrumentation, safety and control systems, reactor kinetics and transients, and radioactive-effluent control and monitoring. The Center will serve as a focal point for the collection, storage, evaluation, and dissemination of information in these areas.

WASTE DISPOSAL DEVELOPMENT

Clinch River Study. — The possibility of release of low-level radioactive liquids to the Clinch River at several points other than the monitored release point at the White Oak Dam gate structure has been investigated. To assess the watertightness of White Oak Dam, the gates in the dam were closed and a fluorescent dye was mixed with White Oak Lake water along the upstream face of the dam. After 24 hr, water samples were collected for fluorescent analysis from very small seeps and small puddles on the creek overbank immediately downstream from the dam. No fluorescent dye was detected in the samples, but flow in the small seeps appeared to have increased slightly, and the creek overbank had become more wetted as a result of the higher lake level, thus suggesting a minor amount of seepage through the earth-fill dam structure. A visit to the Tower Shielding Facility and an examination of surface-water samples obtained from several small streams in the Oak Ridge Reservation failed to disclose any additional release points.

Recent examinations of aluminum plates which have remained submerged in the Clinch River at miles 19.1, 15.0, and 5.4 for 3 to 4 months have indicated that calcite had not precipitated on the plates during submersion. However, calcite had precipitated on the steel bolts and nuts holding the plates. The rough metal of the bolts and nuts may have provided a surface to which the precipitated calcite could adhere more readily than it could to the polished aluminum surfaces.

Man may be exposed to internal and external sources of ionizing radiation due to the discharge of radioactive fluids to the Clinch River. Preliminary calculations demonstrate that doses received from internal sources will be of greatest significance. Mathematical relationships were developed that equate dose received or body burden of radionuclides with the variable intake of such radionuclides. Factors are incorporated in

the equations to allow for differences in water or food intake and size of the critical organ as a function of age. The mathematical expressions have been coded for use in the CDC 1604.

Mineral Exchange Studies. — Sorption of trace quantities of cesium by vermiculite can be improved by pretreatment with potassium to increase the number of collapsed lattices available for edge fixation. Continued leaching of the treated vermiculite with 0.5 M $NaNO_3$, containing traces of cesium (1.7×10^{-7} M), releases the interlayer potassium and negates the pretreatment. Addition of potassium in the influent was found effective in maintaining potassium-treated vermiculite in a collapsed state. A concentration of 0.0005 M KNO_3 in 0.5 M $NaNO_3$ gave optimum cesium sorption (K_d = 366 ml/g) and a final exchange capacity of 46.8 meq/100 g, compared with the initial exchange capacity of the potassium-saturated vermiculite of 5.9 meq/100 g. Higher concentrations of potassium were more effective in maintaining lattice collapse, but reduced cesium sorption due to increased ion competition.

Interlayer fixation of trace quantities of strontium was not observed when the vermiculite lattice was collapsed by addition of 0.01 to 0.04 M KNO_3 to the 0.5 M $NaNO_3$ influent. The sorption of strontium was reduced by increases in the potassium concentration of the influent, and approximately 80% of the strontium was removed during the determination of the exchange capacity after column saturation.

Disposal in Deep Wells. — For disposal by hydraulic fracturing, a mix which exceeds original specifications for ORNL waste has been developed. The cement-base waste solution mix also contains bentonite, calcium lignosulfonate, silica flour, and carboxymethylhydroxyethyl cellulose. A formulation which did not show phase separation gave a thickening time of 50 hr and a fluid loss of 56 cc/30 min under 1000 psi. The slurry set in about 5 days; it had a compressive strength of 1050 psi after 7 days and 3440 psi after 28 days. When the concentration of dissolved salts in the simulated waste solution was increased ten times, the thickening time was decreased to 15 hr, but the compressive strength after 7 days of curing was 1225 psi.

Tests on the sorption of strontium in macro concentrations of calcium ions show that as the pH is lowered in the acidic range, the decrease in sorption of both ions is relatively constant. In basic solutions, however, the removal of strontium is higher than the removal of calcium, suggesting precipitation of a slightly soluble salt of a weak acid.

Disposal in Natural Salt Formations. — A cavity in salt, 8 ft by 10 ft by 2 ft high, has been heated to an air temperature of approximately 165°C for 35 days. Temperatures in the salt range from 140°C at the cavity surface to 40°C at 8 ft above the cavity roof. These are temperature rises of 120°C to 20°C. Cumulative movement between floor and ceiling on the center line of the room was 1.32 in. at 5 ft from the front and 1.35 in. at 8 ft 1 in. back (near the rear wall). At a point adjacent to the side wall and 5 ft from the front, movement was 1.01 in. The maximum recorded movement was 1.44 in. at 5 ft from the front and 2 ft from the center line toward the wall. The two side walls converged approximately 1 in., maximum travel on this gage.

In order to investigate the possible release of chlorine as a result of radiolytic decomposition of solid sodium chloride, samples of salt were irradiated in a Co^{60} gamma flux in sealed tubes at 35 to 50°C, to a dose of 3×10^8 rads. The air and salt in the tube were analyzed for the power of oxidizing a potassium iodide solution. No oxidizing material was found in the air, though when the salt dissolved in water it showed an oxidizing equivalent of 2×10^{-6} mole/g. When an irradiated sample was heated for 4 hr at 225°C prior to analysis, an oxidizing equivalent of 3×10^{-8} mole/g was found in the air and an equivalent of less than 10^{-9} mole/g was left in the salt.

Engineering, Economic, and Safety Evaluation. — The loss of activity from the high-level storage tanks at a hypothetical waste disposal facility at ORNL is being considered as a possible source of atmospheric contamination. The parameters describing the atmospheric conditions at the time of release were obtained from an earlier meteorological survey of the Oak Ridge area. The downwind concentrations are strongly dependent on these parameters, as shown by a variation of several orders of magnitude resulting from a change in only one parameter by less than a factor of 2. Assumed unit releases from the tanks have been treated, using Sutton's dispersion model to estimate the activity concentration at points downwind from the tank farm. Typical concentrations for a moderately stable atmosphere range from 5×10^{-4} unit/cm^3 at 1 mile downwind to 4×10^{-6} unit/cm^3 at 18 miles. The corresponding values for turbulent conditions are 3×10^{-5} and 2×10^{-7} respectively.

Foam Separation Studies. — The composite laboratory unit, consisting of a 3-in.-diam sludge column to precipitate most of the calcium and a 1.5-in.-diam foam column to remove strontium, was operated with tap water spiked with Sr^{85} tracer. The flow rates were 20 gal ft^{-2} hr^{-1} in the sludge column and 60 gal ft^{-2} hr^{-1} in the foam column. Calcium and magnesium precipitation were induced by making the water 0.005 M each in Na_2CO_3 and NaOH, while coagulation was aided by the addition of about 4 ppm Fe^{3+}. Gas-to-liquid flow ratios in the foam column were 10 to 15. Strontium decontamination factors were 10^3 to 10^4, without filtration of the decontaminated water.

Engineering studies were continued in 6- and 24-in.-diam foam separation columns, using dodecylbenzenesulfonate as the surfactant. The 24-in.-diam foam column with feed introduced through 19 tubes in a 5-in. triangular spacing was operated at varying flow rates, and the channeling below the feed tubes was observed visually. Channeling was visible 18 to 30 in. below the tube ends for 40 gal ft^{-2} hr^{-1} and 10 to 22 in. for 15 gal ft^{-2} hr^{-1} liquid feed rates. When 20-mesh screens were used across the column cross sections in an attempt to distribute the liquid uniformly and thus reduce channeling, liquid accumulated on the screens and "dumped" through at short intervals to give very severe channeling. The capacities of air-operated sonic whistles as foam breakers were 0.05 to 0.10 ft^3 of foam per standard cubic foot of operating air; these capacities were significantly reduced by thin (0.001 to 0.002 in.) rubber or aluminum membranes between the whistle and the foam. The concentrations for a continuous three-stage surfactant recovery system test agreed with calculated values for a surfactant distribution coefficient of 7×10^{-4} cm.

PHYSICAL RESEARCH PROGRAM

PHYSICS AND MATHEMATICS RESEARCH

High-Energy Physics. – An attempt was made to analyze K^- interactions with helium where two charged heavy particles, a lambda, and a π^- were produced. The events were run through the IBM 7090 program HEGUTS, assuming one heavy particle to be a deuteron and the other a proton, and again reversing the assignments. So far, 29 events were found for which only one assumption was compatible with kinematics, 7 events had both assumptions compatible, and 70 events would not fit either assumption. Missing mass calculations were done for all events that fit the assumption $K^- + He \rightarrow \Lambda + d + p + \pi^-$, in search of a resonance in the Λ-p as well as the Λ-π^- interactions. With the limited statistics, one hesitates to interpret the observed broad peaks as resonances.

High-Energy Electronuclear Machines: The Mc^2 Cyclotron. – An appraisal is being made of the residual radiation levels that will result from operation of the Mc^2 Cyclotron. The calculation of the residual gamma-ray dose rates was programmed for the IBM 7090 computer. The calculation, which takes into account contributions from all the spallation products, is made for varying bombardment and cooling times. Comparison of calculated residual radiation decay curves for C, Al, Fe, and Cu with experimental data obtained at the CERN Synchro-Cyclotron suggests that the calculation can be used with confidence to predict the residual radiation dose rates in the Mc^2 Cyclotron installation.

High-Energy Electronuclear Machines: A Superconducting Magnet. – A large nine-coil superconducting magnet is being wound with Nb–33% Zr (inner sections) and Nb–25% Zr (intermediate and outer sections). Assembled, this magnet will have a 2-in. ID, a 7-in. OD, and will be 5 in. high. In each section a secondary of copper wire is wound in a bifilar manner with the superconductor to dissipate the stored energy when a superconducting to normal transition occurs. All coils are wound on epoxy forms and potted in epoxy. Eight of the nine sections were completed; six of them (the inner and intermediate coils) were tested. The inner sections separately have carried as much as 35 amp; their capacity depends on the training and the external magnetic field. The intermediate coils connected in series carried 22 amp. These coils enhance the current-carrying capacity of the inner sections. During the last series of tests, fields as high as 25 kilogauss were measured.

High-Energy Electronuclear Machines: Cyclotron Analogue II. – The Analogue, an eight-sector electron cyclotron, models the beam dynamics of the 810-Mev Mc^2 proton cyclotron now under study. A beam extraction efficiency consistently in excess of 90% is now obtained; the maximum efficiency reported previously was 85%. A resonance method is used to magnify the turn separation prior to extraction, and an electrostatic deflector separates the extracted and circulating beams. The improved performance is a result of careful optimization of the extractor location as suggested by computer studies. Measurements also show that the extraction efficiency is relatively insensitive to dee voltage and to the phase of the electrons prior to extraction.

Medium-Energy Nuclear Physics: 22-Mev Protons. – A computational technique for calculating spectroscopic factors for single- and multiple-particle pickup reactions was developed and programmed for machine calculation. The method can be used with or without isobaric spin formalism.

Self-supporting thin films of potassium metal were prepared and successfully transferred to the scattering chamber for use as a target in nuclear reaction studies. A deuteron energy spectrum was obtained from the reaction $K^{39}(p,d)K^{38}$ with such a target.

The 20,000-channel pulse-height analyzer and data-handling system was received and is undergoing acceptance tests.

Medium-Energy Nuclear Physics: Theory. – In the study of elastic scattering of deuterons with the optical model, it was found that many potentials give equivalent fits to the data. Some potentials which yield these equivalent fits are characterized by the same well-shape parameters and different well depths. This can be understood as arising from the fact that a discrete increment in the real well depth will admit one more half wavelength of the important partial waves without changing these waves asymptotically, provided that the imaginary well depth is adjusted to keep the ratio of attenuation length to mean free path constant. Subsequent fitting of He^3 and alpha-particle scattering data has shown this ambiguity to be important in these cases also. Theoretical studies of proton scattering from heavy nuclei suggest that the same ambiguity might appear in nucleon-scattering analyses.

Preliminary distorted-wave studies of the pickup of (1) alpha particles from nuclei in the (d,Li^6) reaction and (2) tritons in the (p,α) reaction indicate that the theory is capable of predicting magnitudes as well as shapes of angular distributions.

Medium-Energy Nuclear Physics: Heavy Particles. – The angular distributions for the reaction $Al^{27}(O^{16},N^{15})Si^{28}$ were measured at several energies on the Tandem Van de Graaff accelerator. At the lowest energy, 28.5 Mev (lab), the distribution is in agreement with the predictions of the Breit-Ebel semiclassical theory. The ratio of the proton widths for the ground state and the first excited state of Si^{28} can be determined with the use of this theory to be 1 to 4.3. Proton transfer reactions for various light elements leading to O^{15} were investigated and found to be in reasonable agreement with the predictions of the semiclassical theory. An eight-counter arrangement was built for the study of fluctuations in the cross sections for heavy-ion-induced reactions.

Cyclotron Operations. – The rf system of the ORIC was tested at various frequencies ranging from 7.48 to 19.06 Mc in an attempt to debug the system. Abnormal heating around the liner transition piece occurred. To alleviate this situation, the liner was welded onto the transition piece around its periphery to ensure a better rf contact. In subsequent tests the excessive heating was eliminated.

Installation of magnetic channels 1 and 2 and their associated power supplies, cooling-water systems, and controls is under way, and preliminary tests of some of the components have been performed.

Deflection of beams of deuterons, alpha particles, and low-energy protons is being studied experimentally.

Physics of Fission. – The fragment mass distributions for thermal- and resonance-neutron-induced fission of Pu^{239} were determined from correlated energy measurements of fragment pairs. Surface-barrier detectors were used in conjunction with low-noise amplifiers; events were serially recorded by a 128 × 128 correlation recorder. Specially designed fast-coincidence and inspection circuits were used to eliminate events in which alpha-particle pileup occurred. Fission events resulting primarily from the 0.3-ev resonance of Pu^{239} were obtained by filtering techniques. The data obtained thus far indicate that the mass distribution peak-to-valley ratio for the 0.3-ev resonance is 2.0 ± 0.4 times as large as the ratio for "thermal" fission. This result supports the suggestion by Wheeler that fission from different spin states of the compound nucleus might show different ratios of asymmetric-to-symmetric fission. When appropriate corrections are made for the contribution of the 0.3-ev resonance to the "thermal" cross section, this result is in agreement with previous radiochemical determinations. The experiment is being extended to the use of cold neutrons to increase the relative contribution from the bound state.

Scintillation Spectrometry and Instrument Development. – Measurements of the temperature dependence of the internal magnetic field in Fe-Rh reported previously were extended, with special attention given to the region near the ferromagnetic-antiferromagnetic transition. From ORNL data and those of Bertaut it is possible to give values for $\langle S_z \rangle_{Fe}$ and $\langle S_z \rangle_{Rh}$ in the antiferromagnetic phase. Bertaut gives 1.52 ± 0.07 and 0.31 ± 0.07 for the expectation value of the iron spin and the rhodium spin in the ferromagnetic phase. In the antiferromagnetic region he gives $\langle S_z \rangle^2_{Fe} + \langle S_z \rangle^2_{Rh} = 2.72$. The ratio of the iron hyperfine splitting just above the transition to that just below is equal to the ratio $\langle S_z \rangle_F / \langle S_z \rangle_{AF} = 1.074$. Thus $\langle S_z \rangle_{Fe} = 1.42 \pm 0.1$ and $\langle S_z \rangle_{Rh} = 0.85 \pm 0.1$ can be obtained for the expectation values of the spins in the antiferromagnetic region. The large increase in the rhodium spin is somewhat surprising in view of the ORNL measurement, which shows that the saturation iron spin is nearly the same in both the ferromagnetic and antiferromagnetic states.

High-Voltage Experimental Program: Coulomb Excitation of Cu^{63} and Cu^{65}. – Properties of levels Coulomb-excited in Cu^{63} and Cu^{65} with 4- to 8-Mev alpha particles have been investigated. The B(E2)'s for excitation, as determined from the yields of gamma rays observed in singles and coincidence spectra, for the 668-, 961-, 1323-, and 1412-kev levels in Cu^{63} are, respectively, (1.16 ± 0.12), (3.49 ± 0.34), (4.2 ± 0.5), and (0.9 ± 0.2) × 10^{-50} e^2 cm^4; for the 771-, 1115-, and 1474-kev levels in Cu^{65}, they are (1.02 ± 0.11), (3.47 ± 0.37), and (4.0 ± 0.7). From angular correlation studies, the 961- and 1115-kev levels are each assigned spin 5/2, and the 1323-kev level spin 7/2. Mixing ratios obtained from these measurements along with the B(E2)'s establish values of 0.09 and 0.4

$e\hbar/(2Mc)^2$ for the B(M1)'s of the 961- and 1115-kev transitions. The B(M1)'s found for the $7/2 \rightarrow 5/2$ cascade transitions are 0.2 and 0.3. Except for the 1412-kev transition, the ground-state transitions have B(E2)'s which are 13 to 16 times the single-particle B(E2). This similarity of enhancements is predicted by the single-particle—core coupling model. The small B(E2; $3/2 \rightarrow 3/2$) of the 1412-kev transition, which is $6 \times B(E2)_{sp}$, can be explained by mixing of the corresponding model states which have $J = 3/2$.

High-Voltage Experimental Program: Elastic Scattering of Neutrons from C^{12}. – Relative yields of 17- to 21-Mev neutrons elastically scattered from a carbon annulus having diameters of 6 and 3 in. and a thickness of 1.1 in. have been measured for laboratory angles of 51, 60, and 85°. The neutrons produced by the (d,t) reaction were detected with a 1 × 1 in. stilbene crystal placed 18 in. from the tritium target. Pulse shape discrimination techniques were employed. At each angle a broad resonance was observed for a laboratory neutron energy of 19.3 ± 0.25 Mev. Its width at half maximum was approximately 0.8 Mev; its peak height was between 10 and 20% of the value of the continuum. This resonance, which is believed to result from a state in the compound nucleus C^{13}, appears similar to the resonance reported previously [Phys. Rev. 99, 332 (1955); Bull. Am. Phys. Soc. 7, 285 (1962)] for elastic scattering of 22.5-Mev protons on C^{12}. The total cross section has also been measured for neutrons with energies between 13.5 and 20 Mev. Here the resonance effect was not evident.

Electronuclear Machines. – During continuing performance tests of the ORIC, deuterons and alphas were accelerated to energies up to the full design values of 40 and 90 Mev respectively. Two methods were used for determining acceptable values for the magnet trimming-coil currents: (1) empirical adjustment by observation of the probe current and (2) computation from measured field data. No deviation from the computed values was necessary to bring the beam to full radius.

A test model of the second section of the magnetic extraction channel was fabricated and installed. This "compensated iron" channel uses iron shielding in combination with currents to obtain a field reduction of up to 6 kilogauss for the extracted beam, while leaving the field for the circulating beam essentially undisturbed. Two separately controlled compensating circuits are used. The conductors for one are located outside the rectangular iron shielding pipe, and the conductors for the other are located inside the pipe. Proper balance between the two circuits permits operation over a wide range of cyclotron fields.

Neutralization of the rf power amplifier at each frequency setting has been found to be necessary to ensure stable operation of the system. Operation from 7 to 15 Mc is now without difficulty. The remaining range to 22 Mc has not yet been thoroughly explored at high power.

Stable Isotopes Development. – New shims for the 255° separator were installed in an XBX calutron tank, and measurements of the magnetic field were completed. The radial field achieved showed a deviation of 0.02% less than that theoretically sought. Computations made from these measurements fix the possible focal width of collected ion beams at 0.025 in.

The system is being prepared for testing with argon and then tantalum. Upon satisfactory completion of these tests, both XBX calutrons will be equipped with inhomogeneous shims; a regular large-scale potassium separation is scheduled for these units, with emphasis on the collection of K^{40} and K^{41}.

Studies are in progress to determine the effect of introducing a mirror magnetic field into the arc region of a calutron ion source. Preliminary results show that a slight gain in output may result from the use of such fields. The extent of ion-beam defocusing introduced by this field would be troublesome only in high-mass separations.

A second direct deposition of Th^{232} on graphite was completed. In this run an attempt was made to reach a saturation value and to establish a comparison between the penetration depth noted in this instance and that noted under conditions where saturation was not achieved.

Recent filament problems have been traced to the use of ultrapure tantalum in filament fabrication. This material was of such purity that prolonged heating resulted in the formation of large crystals which permitted the filament to distort and become useless. Future calutron filaments will be fabricated from tantalum rods containing specific levels of impurities which will retard crystal growth in the heated filament.

Limited success has been achieved in development of a sputtering-type ion source. This unit is being considered as a replacement for the electron-bombardment source now required for use in Pt-Pd separations. In initial efforts, failure of supporting insulators limited the run time to ~3 hr. Present runs are of 20 to 24 hr duration, but the ion output being achieved is much less than that arbitrarily chosen as an acceptable standard for this type of source.

Special Separations. — Estimated weight of separated plutonium isotopes and the range of enrichment achieved are as follows:

Isotope	Estimated Weight for Period (g)[a]	Accumulative Estimated Weight (g)[a]	Purity Range (%)
Pu^{239}	18.0241	34.4597	94.88–94.96
Pu^{240}	31.7867	59.1200	97.24–98.96
Pu^{241}	8.0163	15.0011	89.20–95.54
Pu^{242}	4.0970	7.4659	84.22–90.48
	61.9241	116.0467	

[a]Based on monitored ion currents.

Approximately 560 mg of 93.27% Pu^{241} from recent calutron separations was converted to PuF_3 by a wet chemical method and will be used to prepare targets by a vacuum evaporation technique.

Approximately 68 mg of 99.9963% U^{233} was recovered from salvage solutions and made available for research purposes. Development is proceeding on a wet chemical procedure for preparing UF_4 to be used in making targets by vacuum evaporation.

CHEMISTRY RESEARCH

Thorium Oxide Studies. – X-ray and metallographic examination of thoria preparations (P-82 pellets, arc-fused pellets, and sol-gel fragments) irradiated wet (250 to 300°C) and dry (estimated maximum temperature less than 300°C) in the LITR to exposures of 2.5×10^{18} fissions/g were completed (ORNL TM-409 and -444). Relatively little change occurred as a result of the irradiation, indicative of the marked resistance of cubic structures to radiation damage. The material irradiated under D_2O showed a slight lattice expansion (less than 0.5%), while that irradiated dry appeared relatively unaffected. Apparent crystallite sizes for the irradiated P-82 and sol-gel materials ranged from 950 to 2000 A, compared with original crystallite sizes uniformly greater than 2000 A. Hence there may have been some slight crystallite damage. Diffraction patterns of the irradiated arc-fused pellets, which were originally monocrystalline, indicated that no extensive formation of large angle boundaries occurred as a result of the irradiation.

Metallographs of the irradiated samples resembled those of the original materials. The polished and etched sections of the P-82 pellets showed grain sizes similar to those of the unirradiated pellets. Neither the irradiated nor the unirradiated arc-fused and sol-gel-prepared materials showed a grain structure after polishing and etching.

Equipment Decontamination. – Satisfactory decontaminants were developed for stainless steels that had been contaminated in a gas phase at 650°C with mixed fission products, and for stainless steels contaminated by aqueous solutions and then baked in helium at about 500°C.

The most effective solutions were mixtures of 0.5 M oxalic acid with 0.1 M fluoride, made either slightly corrosive (0.02 mil/hr) with 0.015 M hydrogen peroxide or nearly noncorrosive (0.003 to 0.005 mil/hr) with 0.1 to 1 M peroxide, and used at 95 ± 3°C. The slightly corrosive solution dissolved thin black oxide coatings formed on stainless steels heated to 500 to 650°C in the presence of a trace of air. For this reason it is more effective as a decontaminant for steel than is the noncorrosive solution. For example, a piece of type 304 stainless steel tubing contaminated at 650°C in a helium gas loop at Battelle Memorial Institute was decontaminated from mixed fission products by a factor of 41 (to only twice background level) by immersion in the slightly corrosive solution at 95°C for 1 hr. The decontamination factor in noncorrosive oxalate-citrate-peroxide solution at pH 4 and 95°C (a solution used for carbon steel decontamination) was only 2. Other contaminated samples from the same loop, but from cooler areas (340 and 540°C), were decontaminated in the pH 4 solution by factors of 125 and 64, however.

Decontamination factors for aqueous-contaminated and helium-baked (500°C) stainless steel coupons, when treated with the fluoride-containing solutions for 2 to 4 hr at 95°C, were 100 to 200 for Zr-Nb^{95}, 200 to 300 for Ce-Pr^{144}, 70 for Ru-Rh^{106}, and 70 for Ba-La^{140}.

Stainless steel pipe contaminated by exposure to helium flowing over heated neutron-irradiated UC_2 was easily decontaminated by suitable oxalate-peroxide reagents. Iodine and tellurium deposits collected below about 300°C responded well to the pH 4 solution, and those collected above this temperature were dissolved by the oxalic acid-peroxide-fluoride solutions.

Surface Chemistry. – In the preparation of ThO_2 for heat-of-wetting measurements, certain samples of low specific surface area acquired a light-gray color when outgassed at 400 to 500°C. Coloration of ThO_2, as well as some other oxides, upon outgassing has been variously attributed either to the presence of impurities (possibly carbon) or to the production of anion-deficiency color centers. In order to ascertain whether or not the appearance of color had any effect on the heat of wetting, several color-producing samples were treated with purified oxygen after the usual outgassing procedure. No measurable difference in the heat of wetting was found.

Pyrolysis and Radiolysis of Organic Liquids. – Studies of the liquid-phase pyrolysis of biphenyl at 425°C were completed in which the vapor-to-liquid volume ratio was varied from 1:2 to < 1:13. The yields of benzene indicated that the vapor-phase reaction was relatively unimportant. Studies of the effect of surface on pyrolysis showed no appreciable difference between the results obtained with the use of quartz wool and those obtained with borosilicate glass wool. In both instances a marked decrease in the yield of benzene accompanied the increase in surface area (ORNL-3426, p 23). A similar marked decrease in the yield of benzene was associated with the presence of stainless steel wool; however, the stainless steel surface may have catalyzed further degradation, since there was an approximately eightfold increase in the (very low) yield of methane.

Radiation Chemistry of Organic Materials: High Polymers. – The volatile decomposition products from polystyrene exposed to gamma radiation were analyzed. Product yields were determined after separating volatile products into two fractions, one condensable at 196°C and the other noncondensable at this temperature. Gas chromatographic analysis revealed that the noncondensable fraction consisted of hydrogen and trace amounts of methane, while the condensable fraction was composed of benzene. Hydrogen yields were quantitatively determined by measuring the volume of the noncondensable fraction in a Toepler pump. Benzene yields were determined from the peak height of the chromatograph tracing. The initial yield of hydrogen was 0.016 molecule per 100 ev of energy absorbed. The initial yield of benzene was 0.008 molecule per 100 ev. This contrasts with the radiolysis of most hydrocarbons, which generally give low-molecular-weight hydrocarbons in addition to the high-molecular-weight products.

Analysis of Molten Salts by Electrochemical Methods. – In a continuing study on the voltammetry of reducible species in molten fluoride salt systems, a cathodic wave for the reduction of $Zr^{4+} \rightarrow Zr^{\circ}$ and an anodic wave which represents the oxidation of U^{4+} in a molten mixture of LiF-NaF-KF at approximately 500°C were observed. Zirconium and uranium were added to the salt mixture as ZrF_4 and UF_4 prior to melting the salts. The apparatus was described previously (ORNL-3377, p 27).

In general, the correlation between the limiting current (wave heights) and the concentration of the electroactive species was poor. This may be due in part to the surface condition of the electrode as well as to traces of oxide impurities in the melt; the impurities could lead to partial precipitation of UO_2 and ZrO_2.

The reduction wave for zirconium at both platinum and tungsten microelectrodes exhibits a half-wave potential of approximately –1.4 v vs the platinum quasi-reference electrode. Generally, the waves were better defined at a tungsten indicator electrode. That the zirconium undergoes a four-electron change was indicated from reverse scans in which reasonably well-defined stripping curves were obtained in all cases. On reverse scans, the current passed through zero at approximately +1.25 v; however, the stripping curve exhibited no hesitancy in passing through zero current, a fact which suggests that zirconium is deposited at essentially unit activity and that the electrode process is reversible.

For the uranium wave which exhibits a half-wave potential of approximately +0.5 v vs the platinum quasi-reference electrode, the slope of a logarithmic plot of log $(i_1 - i)/i$ vs E is in excellent agreement with theory (Kolthoff and Lingane, Polarography, vol 1, pp 205–9, Interscience, New York, 1952) for a one-electron change. This suggests the oxidation of U(IV) to U(V) under the prevailing conditions.

Comparative Inorganic Chemistry. – As part of an extensive study of the adsorptive properties of sulfides, it was found that a number of these react with selected metal ions sufficiently rapidly for chromatographic applications. In many cases this reaction involves essentially a metathesis, the original sulfide being converted to another one by reaction with the metal ions of the solution. The relative solubility of the sulfides seems to determine the possible reactions. Particularly interesting in this context was cadmium sulfide, which was found to react very rapidly with ions such as Cu^{2+}, Hg^{2+}, and Ag^{+} and permit their removal from flowing streams at high flow velocities with small sulfide beds. The reaction was found to be essentially quantitative; for example, almost 2 moles of Ag^{+} could be "adsorbed" on 1 mole of CdS.

Radiation Chemistry. – In the radiolysis of gases, there has long been evidence for the importance of ions to stages of the process later than the initial act, and there is currently great interest in establishing the extent and manner of the participation of ions. The old observation of increased radiolytic yields of some reactions in the presence of the rare gases (Lind, ca. 1920) was neatly explained by the concept of charge transfer from primary ions He^{+}, Ne^{+}, or Ar^{+} to such molecules as H_2 and O_2, both of which have lower ionization potentials than any of these rare gases.

An obvious further test of this concept would be to use an inert gas of ionization potential between those of two reactants, and this has now been done with the reaction

$$2H_2 + O_2 \rightarrow 2H_2O ,$$

with CO_2 as the component with intermediate ionization potential. It was demonstrated to be inert by complete recovery of the amount added.

Like the rare gases, CO_2 proved to increase the radiolytic yield, and the results could be quantitatively explained by the assumption that CO_2 was about 60% efficient in transferring charge to O_2, the component with the lower ionization potential.

Corrosion Tests for the TRU Facility. – A specimen of titanium-75A remained chemically inert for the first 800 hr in the vapor above non-aerated 6 M HCl solution at room temperature. Increased exposure, however, produced a sudden loss of passivity. The incremental rate for an additional 300 hr was 2.0 mils/month; attack was uniform. A type 304L stainless steel specimen exposed simultaneously in the vapor with the titanium specimen corroded at a nearly constant rate of 4.0 mils/month during the 1100-hr test.

Specimens of cast Homalite plastic exhibited excellent behavior during a 1000-hr exposure period at 80°C in 1 M mono-2-ethylhexylphenylphosphonic acid and in 30% Alamine 336 chloride in diethylbenzene. Other than a darkening of the original color, very minor dimensional and weight changes were observed.

Two specimens of tantalum plate on Hastelloy C and stainless steel base metals have successfully withstood attack by boiling 6 M HCl solution for 500 hr. Previous specimens tested in a similar manner failed during the initial 100 hr.

Target Preparation. – Radioisotope targets are being fabricated in an evaporator system constructed from a 4-in. pyrex glass pipe "T"; the evaporator is enclosed in a Plexiglas glove box within a sealed hood to provide double containment. Targets produced by this technique include $Pu^{239}F_3$, $Pu^{241}F_3$, $Th^{232}F_4$, $U^{235}F_4$, and $Ra^{226}Br_2$ films of 10 to 100 $\mu g/cm^2$ thickness.

Targets of UFe_2 alloy, a refractory material incapable of being rolled, were fabricated in a combined effort with the Y-12 Metallurgical Development Department. The method used consisted in arc melting the material, casting it into specified shapes, and machine grinding it to the desired dimensions in an inert atmosphere.

Further development of electron bombardment techniques for the evaporation of high-melting elements has resulted in electrical circuit changes designed to improve flexibility so that both conducting and non-conducting elements can be evaporated. Evaporation of bulk silicon and manganese metals with subsequent condensation into self-supporting thin film has been studied, using this heating technique. Most of these films were found to be of poor quality because of numerous pinholes in the films.

Preparation of single-crystal silver films is being studied. Such films were produced by using cleaved, single-crystal NaCl (1:0:0 face) as the substrate and were verified by electron diffraction and microscopy.

By maintaining several substrates at different temperatures during film condensation, a substrate temperature of 350°C was found to be most conducive to crystal formation.

Transuranium Element Processing

Effect of Solvent Diluent in Phosphonate Process. — Solvent diluent has an important effect on the phosphonate process for splitting transplutonium elements into an americium-curium and a transcurium fraction. A correlation has been noted between the dielectric constant of the solvent used to dilute 2-ethylhexylphenylphosphonic acid [2-EH(ΦP)A] and distribution coefficients of americium and californium between 1 M 2-EH(ΦP)A and 1 M HCl. The californium distribution coefficient decreased from 45 for heptane diluent, with a dielectric constant of 1.9, to 3 for toluene diluent, with a dielectric constant of 2.4. Americium distribution coefficients with these two diluents were 0.37 and 0.028 respectively.

Separation of Actinides and Lanthanides in Carbonate Solutions. — Methods of separating actinides and lanthanides in carbonate solutions are being investigated. If satisfactory methods can be found to replace methods using chloride systems, corrosion problems will be greatly reduced. Scouting tests indicate that it may be possible to separate americium and curium from most lanthanides by either extracting the lanthanides into quaternary amines or loading them on strong-base anion exchange resin from dilute carbonate solutions. The group separation of actinides and lanthanides does not appear possible because the heavier actinides have greater distribution coefficients than the lighter lanthanides. Distribution coefficients between 30% Aliquat 336 in diethylbenzene and 0.5 M $NaHCO_3$ were as follows: Am, 0.52; Cm, 0.65; Cf, 2.03; Es, 2.51; Ce, 1.0; and Eu, 4.8. The practical application of this method is limited by the solubilities of actinides and lanthanides in the dilute carbonate solutions. Solubilities have not been determined, but they are probably very low.

Preparation of PuO_2 for HFIR Targets. — A method was investigated for preparing dense, coarse particles of PuO_2 for use in HFIR target prototypes. A solution of 0.5 M $Pu(NO_3)_4$ in 4 M HNO_3 was rapidly mixed with an equal volume of 8 M NH_4OH to precipitate $Pu(OH)_4$. The precipitate was washed thoroughly with 2 M NH_4OH and dried at 150°C. The dried solid was fired in air at 1200°C to produce a glassy solid with a density of 10.99 (96% of theoretical). Since most of the particles were from 1 to 3 mm in diameter, this method, followed by careful grinding and screening, appears to be satisfactory. When PuO_2 particles less than 10 μ in diameter are mixed with −325 mesh aluminum powder and pressed into cermets, the oxide phase is continuous, and the thermal conductivity of the pellet is low. With PuO_2 particles ranging from 20 to 200 μ, the aluminum phase will be continuous, and conductivity will be satisfactory for irradiation in high neutron fluxes.

Determination of Californium in TRU Pu-Al Rods. – A nitric acid solution of eight plutonium-aluminum alloy rods, irradiated for the transuranium element program, was analyzed for californium; two anion exchange cycles from chloride solution were used to isolate the transplutonium elements, and chromatographic elution from cation exchange resin with α-hydroxyisobutyrate solution was used to separate the californium. The californium was analyzed by both alpha pulse and fission counting. The fractions of transplutonium alpha counts due to Cf^{252} were 3×10^{-7} and 6×10^{-7} for duplicate determinations. The difference in these two values probably indicates the accuracy of the determinations. Based on these numbers, the eight Pu-Al alloy rods contained between 0.45 and 0.90 μg of californium. The balance of the TRU rods (which have been irradiated longer) should contain 100 to 150 μg of californium.

Preparation of Pure Materials. – The fourth LiF (99.99 at. % Li^7) crystal has been analyzed by emission spectroscopy. The top (~3-mm layer) was found to contain 1, 30, 5, 5, 75, 1, and 50 ppm, respectively, of Al, Cu, Fe, K, Mn, Na, and Ni. The surface contacting the container half way up the crystal was found to contain 1, 1, 5, and 5 ppm, respectively, of B, Fe, K, and Mn. The bulk of the crystal contained only 0.5 to 1.0 ppm Mn; no other element was detected.

A fifth crystal of LiF, weighing 290 g and containing 96.50 at. % Li^7, was produced in March 1963 and loaned to the Materials Science Center at Cornell University. No strain was detected in this crystal by observation with polarized light.

Approximately 75 g of purified and dehydrated LiF (4.38 at. % Li^7) was produced and was supplied to the Analytical Chemistry Division for use in developing a thermal-neutron activation analysis for oxygen in the ppm range.

Crystal Physics. – Optically clear ThO_2 crystals up to 3 mm on an edge have been grown in unseeded melts. Also, seeds suspended in stirred melts have produced medium-quality crystals approaching 1 cm on an edge. These crystals were grown at 1220°C from $Li_2O \cdot 2WO_3$ melts having a temperature gradient of 11°C/cm between the nutrient and the seed. The only impurity detectable by spectroscopic analysis in these crystals was 100 ppm Ca. In support of this work, a portion of the ternary system Li_2O-WO_3-ThO_2 has been investigated. At least two incongruently melting compounds have been detected below 1200°C. One of these compounds has been isolated and is being chemically and structurally analyzed.

Crystals of BeO greater than 2 mm on an edge have been grown from the solvent, molten V_2O_5. Impurities detected by emission spectroscopy in these BeO crystals were 100 ppm Si and 200 ppm V. Hence these well-formed crystals are better than an order of magnitude more pure than those grown from alkali oxide polytungstate or polymolybdate.

The intermetallic compounds UAl_3 and UAl_4 have been grown as single crystals, with their longest dimension up to 5 mm. Melts of the desired U-Al composition were cooled at a rate of 2°C/hr in a temperature gradient of 10°C/cm over a range where the specific compound was stable and then quickly cooled to near room temperature.

CONTROLLED THERMONUCLEAR RESEARCH

Plasma Physics and Arc Research. – The study of the plasma formed between magnetic mirrors by the pressure-gradient arc has continued. A condensable vapor of evaporated gold is being used to determine plasma dimensions and density. Neutral atoms ionized by the hot plasma electrons are trapped in the magnetic field and diverted from their original path. The evaporated gold is collected on a glass plate, and the resulting shadow of the plasma is examined by measuring transmission of light or by directly analyzing the gold by an x-ray fluorescence method. The ratio of gold deposited inside the shadow to that deposited outside the shadow is the fraction of atoms ionized and a measure of the electron density. The measured density, 10^{11} electrons/cm^3, is in agreement with other measurements of this type of plasma.

DCX-1 Facility. – For some time there has been evidence that the Z scattering of the plasma to larger volumes is associated with the ion clumping instability. Some investigations have been made in an effort to determine if this scattering might be peculiarly sensitive to the shape of the magnetic field. These investigations were studies of the effects on proton containment of a magnetic-field perturbation produced by rapidly moving a soft iron slug along a radius in the median plane from large radii, where there was no measurable ($\leq$1 or 2 gauss) perturbation of the containment field along the proton stable-orbit radius, to R = 9 in., where the perturbation along the stable orbit ranged from about 35 to 5 gauss. No effects on containment were noted for trapped-proton containment times less than about 0.1 sec. At longer containment times, insertion of the slug produced a decrease both in the steady-state charge-exchange reaction rate and in the containment time. Both effects became more noticeable as the pressure was reduced. At the lowest pressures the containment time without the perturbation was about 10 sec; the perturbation reduced the steady-state charge-exchange reaction rate by about 1/2; and it reduced the containment time for the remainder of the plasma by about 1/4. The change in the steady-state charge-exchange reaction rate upon inserting the slug occurred with a time constant of about 1 sec, which was several times longer than the amount of time required to insert the slug and which appears to be a measure of a plasma phenomenon rather than a ferromagnetic effect. The mirror loss of fast particles did not increase upon inserting the slug. It is believed that the increased proton loss rates associated with the perturbation are the result of enhanced radial diffusion, though there has not yet been a successful measurement of the current to the walls. It does not appear that the slight field perturbations (again $\leq$1 or 2 gauss) normally present over the stable orbit give rise to any significant scattering.

Lorentz trapping experiments in the DCX-1 were extended to include the injection of 600-kev H_2^+ formed from the collisions of 60-kev H_3^+ (from a von Ardenne source) in a water-vapor cell positioned between the ion source and the accelerator tube. The results were most encouraging. Three determinations of the Lorentz trapping fraction yielded values of 5×10^{-5}, 7.5×10^{-5}, and 10^{-4}. The input H_2^+ beam in these experiments was only about 100 μa, and the pressures were not particularly good – the

longest containment times were 5 sec. Even under these conditions, at base pressure the plasma density obtained with Lorentz dissociation of the 100-μa beam was equivalent to that obtained with gas dissociation of the full 10-ma H_2^+ beam in normal operation. Since the Lorentz trapping fraction is independent of the background gas density, the density of a plasma established via Lorentz dissociation will scale upward with increases in containment time. The H_2^+ current yield from the water-vapor cell was only 3% of the input 3-ma H_3^+ current and was limited by the maximum conversion cell pressure, which in turn was fixed by the maximum pressure at which it was deemed safe to operate the accelerator tube. Current efforts are devoted to a cell redesign that should permit operation with a conversion efficiency more nearly approaching the 13% maximum, and on studies of the ion source and beam-focus equipment to improve the yield and focus of the H_3^+ component.

DCX-2 Facility. – Lithium arcs have been run in the DCX-2 with simultaneous injection of a molecular ion beam. The first experiments were with the "backstream" of a short lithium arc in one end section. Later, an arc was run axially through the plasma region with anode and cathode in opposite end sections. It was hoped that these arcs would suppress the instability causing the trapped hot plasma to be lost in bursts radially outward. Both arcs suppressed the bursts to a considerable degree, and under some conditions eliminated them. The plasma decay appeared to be the result of charge exchange. The density has been limited to about 4×10^8 cm^{-3}, probably by energy degradation. Energy-spectrum measurements showed a spread to greater than 600 kev of the trapped protons when the direct arc was used.

Vacuum. – Modifications of the instrumentation in the getter test facility were made which allowed pressure measurements to be obtained down to about 2×10^{-10} torr, with as yet an uncertain accuracy, but one which is probably within a factor of 5.

Several molybdenum-wound filaments have been designed and tested, with the goal of increasing the life span of such metal-vapor sources. At low evaporation rates these filaments have provided the satisfactory quantity of about 0.1 mole of molybdenum before failure. Evaporations at suitably high rates to date have led to early filament failure.

The pressure behavior in the test facility has been studied in the range 2×10^{-10} to 2×10^{-8} torr as a function of temperature in the range 80 to 120°K. Desorption behavior from this 15-ft^3 system has indicated the existence of at least two desorption peaks, one with an enthalpy of desorption of the order of 4.5 kcal/mole and the other of about 7 kcal/mole. Desorption enthalpies have been determined for argon, methane, and carbon monoxide in the same system and have been found to be about 3, 4.3, and 3.5 kcal/mole respectively. The base pressure of this system thus appears to be limited by the desorption rate of physically adsorbed species.

Carbon monoxide was found to adsorb on the cold surface with a sticking factor of as much as 0.5 mole adsorbed per second for each mole striking the surface.

The bakeable-flange test facility was used in this report period to make a study of leak rate as a function of gasket deformation of a 0.086-in.-diam aluminum 1100-0 alloy wire gasket. To effect a minimum leak

rate, a 0.026-in. total deformation was needed with good reproducibility. A leak rate of less than 3×10^{-10} atm cc of He per sec for this 32-in.-long gasket is obtained at this deformation, using eight 1/2-in. bolts loaded to about 60 ft-lb of torque.

BIOLOGY AND MEDICINE PROGRAM

TERRESTRIAL AND FRESH-WATER ECOLOGY

Evaluation of Fission Product Distribution and Movement in White Oak Drainage System. — A preliminary investigation of the bed of White Oak Creek and its flood plain has revealed that bottom sediments at mile 2.3 contain Sr^{90}, Cs^{137}, Co^{60}, and total rare earths (TRE), exclusive of Y^{90}, in concentrations, down to a depth of 18 in., which range from 100 to 3450, 4000 to 76,900, 290 to 2200, and 0 to 14,000 μμc/g respectively. In general, for Sr^{90} and TRE, the highest concentrations occur in the uppermost few inches of the sediment; whereas for Cs^{137} and Co^{60} the higher concentrations were found at a depth of 6 to 12 in. The concentrations of radionuclides found in the flood plain along the bank of the creek are roughly the same as those observed in the creek bed.

A flood time-of-travel study was made in White Oak Creek on March 12, 1963, while discharge at the gaging station "White Oak Creek below ORNL" was about 200 cfs, using Rhodamine B as the tracer. Minimum time of travel for the 5900-ft reach between the gaging stations "White Oak Creek at ORNL" and "White Oak Creek below ORNL" was 37 min; peak concentration time of travel was 42 min. In contrast, a study made in October 1962 while discharge was 4.5 cfs showed a minimum time of travel of 3 hr 23 min for this reach and a peak concentration time of travel of 3 hr 50 min. No dye was detected at White Oak Dam during the flood of March 12, possibly due to dilution.

During the week of March 18–22, water levels were measured in 247 wells in White Oak Creek Basin for the purpose of constructing a water-table contour map during the period of water-table high for the 1963 water year.

RADIOLOGICAL PHYSICS, HEALTH PHYSICS, AND INSTRUMENTATION

Internal Dose Estimation. — A study of I^{131} uptake and metabolism was made. Five male volunteers ingested organically bound I^{131} in milk, and their thyroid-gland uptakes were measured with a gamma-ray scintillation counter. Three subjects ingested 150 μμc/day for 11 consecutive days; two of these three subjects also ingested a single dose of 92,000 μμc. The third subject then ingested 1840 μμc/day for 4 days along with two other subjects. Of these other two subjects, one took in 1840 μμc/day for 8 consecutive days; the other took in the same level for 63 consecutive days. The latter subject submitted 24-hr urine samples during the

entire period of intake and for two weeks after the intake stopped. The biological half-lives of I^{131} for these individuals ranged from 37 to 153 days, and f_W, the fraction going from the gut to the thyroid gland, ranged from 0.08 to 0.28. Some of these values differ significantly from the ICRP-NCRP values, and others are approximately the same; but there is a large variance. However, it is found that the ICRP-NCRP predicted thyroid burdens are generally higher – by about a factor of 4 for the subject ingesting 1840 μμc/day for 63 days – than the individually extrapolated burdens, and thus they can be considered to be "conservative" predictions for adults. As nearly as could be judged from this study, the organically bound I^{131} seemed to be as available for uptake as an ionic iodine.

Aerosol Studies. – Dispersion of particles and vapors in semi-isolated environments is being studied, particularly regarding deposition and retention on environmental surfaces. Laboratory-scale studies have been made on the redispersion of 1-μ and of 5-μ ThO_2 particles from various surfaces by air impingement. In general, the smaller particles are more difficult to redisperse by air blowing onto a surface than are the larger particles. Air impinging on a stainless steel surface at a speed of 72.5 m/sec removes 90% of the 5-μ particles, whereas only 50% of the 1-μ particles are removed. For mixtures of the two particle sizes on the same surface, the redispersion rate is approximately the same as that for the larger particles. Thus the presence of large-particle nuisance dust can enhance the redispersion of surface contaminants which may be of a much smaller particle size.

NUCLEAR ENERGY CIVIL EFFECTS

Dosimetry Applications. – The major portion of the data from Operation BREN was analyzed, and results were correlated with weapons test results. Spatial dose distribution measurements were made for 4500 ft along a line which extended horizontally from the base of a 1524-ft tower.

Data reduced to 1.293 g/liter show that the relaxation lengths in air for neutrons and gamma rays from a point fission source 1125 ft above the air-ground interface are 198 and 258 yd respectively. Similarly, the relaxation length in air for gamma rays from a point isotropic Co^{60} source at 300 ft above the air-ground interface is 182 yd.

The fast-neutron spectrum was measured with a thick-radiator proton recoil spectrometer placed in a collimator at 750 or 1000 yd from the base of the tower. Neutron spectra were measured with the collimator aimed directly at the reactor and at a polar angle of 90° from that direction; the neutron spectrum does not change appreciably with distance or angle. The gamma spectrum produced by the reactor was measured with a magnetically analyzing Compton recoil spectrometer located 500 yd from the base of the tower and permanently aimed at the 1125-ft elevation on the tower. An NaI crystal was utilized for additional gamma measurements following reactor pulse operation, with a 1200-curie Co^{60} source in place of the reactor.

Auxiliary experiments have been conducted at a Van de Graaff generator to refine the curves of response vs energy for the recently developed or modified dosimeters and detectors. Emphasis at present is on the quantitative evaluation of the gamma radiation produced by inelastic interactions of neutrons in air and building materials. An experimental evaluation of low-atomic-number metaphosphate glasses as long-term integrating dosimeters was completed; the detectors are accurate to within ±10% for periods greater than one year.

TRAINING, EDUCATION, AND INFORMATION PROGRAM

Professional Health Physics Training. — A course in applied radiation physics is being conducted at Vanderbilt University. The course began February 4 and ends May 24. Thirteen AEC Fellowship students, one Air Force officer, one citizen of Pakistan, and four Vanderbilt students are enrolled. Other items include the following:

1. A series of lectures on radiation units was given to personnel of Applied Health Physics.
2. A. Gopez from the Philippines Atomic Energy Commission arrived on April 4 for three months of field training.
3. The Applied Health Physics and Waste Disposal groups participated in a one-week training program for a U.S. Public Health Service group from Cincinnati, Ohio.

ISOTOPE DEVELOPMENT PROGRAM

Recovery of Fission Products. — In extractions of strontium from 3 M $NaNO_3$ solution with 0.3 M Sunaptic Acid A (a commercial naphthenic acid) in Amsco 125-82, the extraction coefficient increased to about 70 with increase in pH to 7.5 and then decreased rapidly with further increase in pH. Maximum coefficients were much lower (2 to 6) in extractions from citrate-complexed simulated formaldehyde-treated Purex waste (FTW). In the latter tests, strontium coefficients were about threefold higher when pH adjustment was made during, rather than before, extraction. The strontium/sodium separation factor in the tests with FTW solution ranged from 60 to 150.

In a batch countercurrent test using two extraction stages and one water-scrub stage, 94% of the iron was extracted from simulated FTW with 0.24 M Primene JM in Amsco 125-82, while only about 5% of the cerium was extracted. Greater than 98% of the cerium was then extracted by contacting the raffinate with six successive volumes of the same solvent at an aqueous/organic ratio of 10/1.

Curium-242 Preparation. — Details of the Tramex process are being studied to ensure successful operation for Cm^{242} isolation from irradiated AmO_2-aluminum cermets. The principal effect of alpha radiation in

Tramex feed is a loss of acid. Acid-depletion G values (G_{-H^+}) were determined for synthetic feeds of 10 M LiCl containing 2.5 and 15 w of Cm^{242} per liter. Previously reported G_{-H^+} values of 4.0 and 5.6 were averages for a wide range of acid concentration. Recent data show that G_{-H^+} values are dependent upon acid concentration. Over the range 0.4 to 1.0 M HCl, G_{-H^+} values of 10 to 20 were measured. At lower acidities, G_{-H^+} values were 2.2 at 2.5 w/liter and 1.0 at 15 w/liter. If G_{-H^+} is slightly greater than 1 at 10 w/liter, which is the proposed operating level, acid will be lost at the rate of 0.1 mole per liter per day. Since acid concentration must be held between 0.01 and 0.1 M in order to obtain both a stable feed and sufficient extractability of the transplutonium elements, acid loss by radiolysis means that adjusted feeds may be stable for less than a day unless acid is replenished. Acid can be replenished by HCl gas bubbled through the feed; however, the equilibrium acidities of 1.35 M at 60°C and 0.4 M at 120°C for HCl gas at 1 atm are too high for satisfactory extraction.

Careful investigation of the effect of $AlCl_3$, LiCl, and HCl concentrations in Tramex feed on americium distribution coefficients is in progress. Concentrations must be carefully controlled for the Tramex process to work satisfactorily. For example, the americium distribution coefficient between 0.6 M Alamine 336·HCl–diethylbenzene and 10 M LiCl–0.1 M $AlCl_3$–0.02 M HCl is 4.0, but is decreased to 1.0 by either increasing the acid to 0.14 M or decreasing the $AlCl_3$ plus LiCl salt concentration to 9.5 N.

Radioisotope Research and Development. – The production of Ir^{192} from neutron irradiation of natural iridium in the ORR was investigated to determine the rate of production, specific activity attainable, and flux depression effects. A study of the flux depression produced by an iridium rod 4 in. long by 1/16 in. in diameter showed that the effective neutron flux was reduced by a factor of 2 in the immediate vicinity of the iridium. Flux depression within the iridium further lowered the effective flux so that the overall flux depression factor was 7.5 for the 1/16-in.-diam rod. With an assumed allowable reactivity change of –0.15% in the ORR, ~85,000 curies of Ir^{192} could be produced per year at a specific activity of 325 curies/g, based on the irradiation of 44 g of iridium for each 49-day cycle in a flux of 1.8×10^{14} neutrons cm^{-2} sec^{-1}.

The electrolytic (ac) dissolution of metals has been investigated for Pd, Pt, Rh, Ir, and Al. The dissolution of the noble metals in hydrochloric acid and of aluminum in nitric acid was successfully demonstrated. The rate of dissolution was determined as a function of acid concentration and amperage. The ac dissolution method will be applied to the processing of reactor-irradiated metals to reduce the time required in preparing product solutions for distribution.

The preparation and properties of compounds suitable for the fixation of radioisotopes in radiation sources are being investigated. The primary emphasis is on compounds of Ce, Sr, and Pm, which will be available in large quantities from fission product recovery operations within the next

few years. Experimental quantities of CeB_6, $CeSi_2$, CeBe, SrB_6, and SrF_2 have been prepared for evaluation as suitable source compounds.

Radioisotope and Stable Isotope Production. — Ion currents equivalent to 2380 g of separated isotopes were monitored during March and April. Isotopes of Ca, Ta, Si, and Pb were separated and collected, using 28 of the calutrons associated with the program.

Hundred-gram batches of silicon, silicon carbide, silicon sulfide, a mixture of aluminum sulfide and silicon dioxide, and barium fluosilicate were supplied for experimental runs in silicon isotope separations. Smaller amounts of lead oxide, chromium oxide, titanium oxide plus carbon, and cerous oxide plus carbon were supplied for evaluation of internal chlorination techniques.

Recovery of 11,802 g of Yb_2O_3 from an original supply of 13,383 g of the oxide gives an 88% recovery factor for the recent ytterbium series.

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