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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

FEBRUARY 1960

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

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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

February 1960

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

PROGRAM 2000 – SPECIAL NUCLEAR MATERIALS

Power Reactor Fuel Processing Pilot Plant. – On November 20 a chemical explosion in an evaporator in cell 6 of Building 3019 resulted in release of an estimated 615 mg of plutonium, 545 mg outside the building and 70 mg inside. The offices and control rooms were decontaminated to a transferable contamination level of < 3 d/min per 100 cm^2 and were reoccupied on January 14.

Other areas of the plant, including the attic, makeup area, sampling gallery, pipe house, and pipe tunnel, are being decontaminated sufficiently so that residual plutonium activity may be immobilized with three coats of "color coded" paint. Floors are being decontaminated by grinding with a terrazzo machine or, when necessary, by chipping of selected spots followed by pouring of new concrete. (AEC Activity 2461)

[redacted]

PROGRAM 4000 — REACTOR DEVELOPMENT

GAS-COOLED REACTOR PROGRAM

Physics. — A comparison has been made of the characteristics of BeO-moderated and graphite-moderated gas-cooled reactors. Approximately equal leakage is obtained if the graphite-moderated core is twice the diameter of the BeO-moderated core. The conversion ratios, which do not vary appreciably with the core diameter or void fraction, are uniformly lower for graphite-moderated cores than for BeO-moderated cores.

Recent measurements have indicated a resonance integral of 1200 barns for Pa^{233} (rather than the 700 barns previously reported). Thus the loss of neutrons due to capture by 27-day Pa^{233} will have a significant effect on the conversion ratio of a one-region reactor using thorium as the fertile fuel. The relative conversion ratios calculated for a number of large BeO-moderated systems, with 1200 barns used for the resonance integral and 70 barns for the thermal cross section, are given below for 1000, 2000, and 4000 atoms of moderator per atom of fuel:

Specific Power (kw/kg)	Relative Conversion Ratio		
	1000	2000	4000
1000	0.94	0.96	0.97
4000	0.80	0.85	0.89
8000	0.67	0.74	0.80

(AEC Activity 4141.1)

Thermodynamics. — Calculations have been made concerning the removal of the heat generated in the control rods of the Experimental Gas-Cooled Reactor (EGCR) by the absorption of neutrons and gamma rays. The maximum heat flux was determined to be 15,000 $Btu \cdot hr^{-1} \cdot ft^{-2}$ at the center of the core, and, since the central control rod is likely to operate fully inserted, it represents the extreme case for control rod cooling considerations. Positive cooling of the rod is difficult because the rod holes are located at graphite block junctions. Thus graphite shrinkage will increase the clearance between blocks and decrease the rod cooling. Approximate calculations indicate that about 70% of the control rod heat can be radiated to the graphite and thus be absorbed in the fuel element annulus coolant flow. ? This heat transfer is dependent, however, on achieving an emissivity of 0.6 for the stainless steel cladding and allowing the control rod surface temperature to reach 1600°F. The remaining 30% of the heat generated must be removed by coolant flowing between the graphite blocks and through the control rod holes (annuli). It

may be desirable to allow the coolant to flow through the center of the control rods. The coolant flow, in any case, will be greater than would be required if the control rod holes were located at the center of the blocks. (AEC Activity 4141.1)

Fuel Element Development. – A test stand for investigating the stability of the EGCR rod-cluster fuel element under full flow conditions was completed. In initial tests at air flow rates up to 3000 lb/hr, no visual or audible evidence of vibration was noted.

The pilot plant fabrication of 50 EGCR fuel elements has been undertaken in order to obtain information on the influence of manufacturing variables on the procedures. Based on experience gained in the manufacture of the first 20 elements, arrangements will be made for the fabrication of the remaining 30 elements under conditions that will permit a realistic determination of the problems and costs to be encountered by potential vendors. Four assemblies have been completed.

The gross thermal performance of annular fuel elements of one, two, and three rings has been compared with the performance of seven-rod EGCR fuel elements. The annular elements being studied have a central filler rod to support the rings and to provide low-temperature storage for the fission gas. The comparison was made to estimate the performance of these elements if they were placed in the EGCR core structure. The results for the EGCR design conditions of core inlet and exit temperature, core pressure level, and core pressure drop are presented in the following table:

Number of Rings	Channel Inside Diameter (in.)	Ratio of Heat Generated in Annular Element to Heat Generated in EGCR Element
1	4	1.16
1	3.75	1.06
1	3.5	0.96
1	3.25	0.86
1	3.0	0.76
2	4	2.08
2	3.75	1.87
3	4	2.67
3	3.75	2.29

The fabrication of annular fuel elements poses several problems that require further investigation. These include rolling and forming of stainless-steel-clad UO_2 plates, edge-closure welding of the cladding, and attachment of the fission gas carrier tubing. (AEC Activity 4141.1)

Irradiation Experiments. – A tabulation of typical temperature conditions and estimated burnup data for the group of fuel capsules presently being irradiated in the ETR is presented below. These capsules are full-diameter prototypes of the EGCR capsules.

Capsule No.	Temperature (°F)				Calculated Burnup (Mwd/metric ton)
	Design	Maximum	Minimum	Average	
E-1[a]	1300	1293	1250	1273	1005
E-1A[b]	1300	1330	1215	1273	664
E-2[b]	1300	1327	1078	1246	34
E-3[b]	1300[c]	1320	1092	1270	33
E-4[b]	1300	1353	1250	1304	664
E-5[b]	1300	1100	1065	1080	734
E-6[b]	1300	1160	1075	1125	765
E-7[b]	1300	1310	1195	1258	777
E-8[b]	1300[c]	1340	1141	1265	56
E-9[b]	1300	1292	1226	1267	961
E-10[b]	1300[c]	1332	1212	1275	57

[a]Temperature readings as of 10-13-59; reactor at 175 Mw; burnup is final calculated value; capsule withdrawn 10-19-59.

[b]Temperature readings as of 1-5-60; reactor at 175 Mw; burnup calculated as of 1-10-60.

[c]Design temperature to be raised to 1600°F in early February.

In the instantaneous fission gas release experiments being conducted in the ORR, it has been found that, at 1950°F, 0.06% of the Kr^{87} and Kr^{88} and 0.3% of the Xe^{133} escape on a steady-state basis from the 95%-dense UO_2 plates now being irradiated. The shorter half lives of the kryptons account for the lower release percentage. By using a slab model, a diffusion coefficient of 5.6×10^{-14} cm^2/sec was obtained for the kryptons and 3.8×10^{-14} cm^2/sec for Xe^{133}.

Fission-gas sampling and pressure measurements have been completed on the four hollow-pellet-containing capsules irradiated in the ORR. The results are presented below:

Capsule No.	Cladding Temperature (°F)	Internal Pressure at 86°F (mm Hg)	Fission Gas Release (%)*	Burnup (Mwd/metric ton)
01	1300	800	2.5	1680
02	1600	800	2.5	1700
03	1300	830	5	1900
05	1600	900	7	2100

*Percentage of fission gas generated.

(AEC Activity 4141.1)

Pressure Vessel Welding Studies. — In order to simulate heating and cooling in particular regions of the heat-affected zone of a pressure-vessel weld, full-size Charpy V-notch impact specimens have been cycled to 1400 and 2400°F at Rensselaer Polytechnic Institute. The specimens heated to 1400°F at 50 kilojoules/in. showed an improvement in notch toughness. This cycle caused refinement of the grain size and resulted in a soft, ductile structure. On the other hand, specimens heated to 2400°F with an energy input of 50 kilojoules/in. had a very coarse-grained mixed structure consisting largely of brittle martensite. Charpy V-notch specimens having this structure were brittle. When stress-relieved at 1150°F for 4 hr, however, the martensite was tempered, and the notch toughness of the resulting structure was comparable to that of the as-received plate. Cycling of specimens to 2400°F with an energy input of 100 kilojoules/in. produced a very coarse-grained structure, consisting essentially of ferrite and pearlite, which are characteristic of slow cooling. The ductile portion of the impact curve was improved, but the transition temperature was still too high. It appears that satisfactory structures can be produced in the heat-affected zone by carefully controlling the energy input during welding. (AEC Activity 4141.1)

Materials Compatibility Studies. — Three high-pressure thermal-convection loop tests have been completed with helium as the circulated gas. The test specimens consisted of AGOT-grade graphite, various iron-base alloys, and a dummy EGCR fuel element assembly. The tests were of 500, 1000, and 5000 hr duration. The weight gains at 500 hr were nearly as great as those at 1000 and 5000 hr, and thus it appears that most of the reaction takes place within the first 500 hr of operation. In all three tests the stainless steels gained less than 1.0 mg/cm^2, whereas the low- and medium-alloy steels gained in excess of 6 mg/cm^2. The gas impurity concentrations were reduced to relatively low levels in the first 150 hr of each test. No carbon mass transfer to the cold leg has been found. The test section temperature in these tests was 1225°F and the system pressure 325 psig.

The surfaces of T-1 and Croloy steel specimens exposed in natural-convection tests at 1100 to 1200°F to atmospheres produced by the out-gassing of graphite have characteristically exhibited relatively heavy, poorly adherent reaction films. The films are composed of a silvery outer layer and a black inner layer, both of which are highly magnetic. Analyses have indicated that the silvery layer is primarily FeC and that the black layer is largely Fe_3O_4. Thermodynamic calculations indicate that oxidation of the low-alloy T-1 material would have been possible only during initial stages of the test. Following this period, conditions would favor carburization and deoxidation of these specimens. It is believed therefore that the silvery component of the film is formed subsequent to the buildup of the black oxide material and results from a combination of reduction of the iron oxide and deposition of carbon at the gas-film interface. (AEC Activity 4141.1)

Graphite Permeability. — Room-temperature experiments have been conducted in which the diffusion of helium and argon through various graphites was measured. The absolute permeabilities and diffusion coefficients for each type of graphite show definite correspondence. For example, both the permeability and diffusion coefficients for AGOT graphite are roughly 2.5 times greater than the comparable values for Speer No. 1 graphite. Plots of the available

data as a function of reciprocal pressure strongly suggest that the diffusion coefficients for each gas (argon and helium) approach the same value at 400 psia. If this trend is real, it may be concluded that "classical" diffusion will be the controlling constant-pressure transport mechanism at projected reactor pressures. (AEC Activity 4141.1)

THERMAL BREEDER REACTOR PROGRAM

Homogeneous Reactor Program

Homogeneous Reactor Test. — In run 21, after the reactor had operated continuously for more than three months at power levels up to 5 Mw, the rate of mixing of fuel between the core and blanket regions suddenly doubled. An inspection of the interior of the core revealed a second hole in the Zircaloy vessel. It was located below the equator of the vessel and was estimated to measure about 1 by 1/2 in.

A program of maintenance and major repair operations was begun in order to accomplish removal of the core diffuser screens, reversal of the direction of flow through the core, repair of the core-vessel holes, and replacement of several pieces of equipment.

During the shutdown of the reactor following run 21, about 400 lb of D_2O condensate leaked into the shield sumps. Helium leak testing revealed that a purge line to the core circulating pump had cracked at a weld. The line will be removed with the pump, which is to be replaced. (AEC Activity 4151.3)

Thorium Oxide Slurry Development. — Photomicrographs of the product of flame denitration of thorium nitrate solutions containing aluminum nitrate or tetraethyl orthosilicate showed predominantly rounded particles of thoria containing 0.5 to 5.0 wt % of either SiO_2 or Al_2O_3. The thoria particles with these additives were spherical for up to 8 wt % uranium oxide. The same additives in flame denitration of uranium solutions gave much smaller particles of an uncertain shape.

Qualitative microscopic examination of solution drops from the flame denitration feed nozzles showed approximately 150-μ-dia drops for the usual feed conditions and down to 30-μ-dia drops for higher atomizing gas flows. Literature correlations indicate that the drops should be in this size range. Since 5- to 10-μ-dia drops of feed solution contain sufficient thorium nitrate to give the 1- to 2-μ-dia ThO_2 particles obtained, additional particle division must occur in the reflected flame. (AEC Activity 4151.1)

Heterogeneous Equilibria in Aqueous Systems (See AEC Activity 5330, ORNL-2906). — The temperatures at which second-liquid-phase formation occurred were determined after concentrating to progressively higher levels fuel solutions initially 0.025 M in UO_2SO_4 and having mole ratios, UO_2SO_4:$CuSO_4$:D_2SO_4, of 1:1:1, 1:1:1.5, and 1:1:2. These series showed minimum two-liquid-phase temperatures of 326, 340, and 350°C at six, four, and three times, respectively, their initial concentration. (AEC Activity 4151.1)

Reactions in Aqueous Solutions. — The decomposition of peroxide in a simulated reactor fuel solution based on uranyl nitrate was studied from 60

to 100°C. In all the experiments the disappearance of the peroxide was observed to be first order with respect to the peroxide. The addition of acid increased the observed rate. The effects of adding small amounts of iron, copper, chromium, and nickel to the solution in the form of dissolved salts were determined. The iron acted as a very effective homogeneous catalyst; the rate of decomposition increased a factor of 20 for each ppm of iron in solution. The catalytic effects of copper, nickel, and chromium were about one-thousandth that of iron. The addition of any of the above-mentioned ions did not materially influence the temperature dependence of the rate constant. The calculated energy of activation is 25 kcal/mole. The addition of copper to an iron-containing solution enhanced the observed catalysis. All these results are consistent with results previously obtained in the sulfate system. (AEC Activity 4151.1)

Analytical Chemistry. – In corrosion and stability tests of UO_2SO_4 solutions for use as reactor fuels which may also contain H_2SO_4, $CuSO_4$, other additives, and diverse corrosion products, the cation/SO_4^{--} ratio and changes thereof are factors of primary importance. A method was therefore devised for the determination of total SO_4^{--} which is simpler and more rapid than conventional titrimetric methods because it is unnecessary to remove UO_2^{++}, Cu^{++}, Fe^{+++}, Zr^{4+}, or, in general, ter- or quadrivalent cations. The SO_4^{--} is precipitated in an acetic acid medium by adding a known amount of barium acetate in excess, and then the excess barium acetate, which is basic in acetic acid, is titrated potentiometrically with $HClO_4$. Free HNO_3 and HCl and also sulfate salts of those uni- and bivalent elements which, like barium, form basic salts in acetic acid will interfere with this method, whereas H_3PO_4 and HF can be tolerated. In the range from 1 to 6 mg of SO_4^{--}, the coefficient of variation is better than 1%.

An evaluation was continued of flame-photometric methods for rare-earth elements in simulated UO_2SO_4 fuel solutions and ThO_2 slurries which are under test for use in homogeneous reactors. Specifically, it was demonstrated that 5 μg of europium per milliliter can be determined flame photometrically with a coefficient of variation of less than 5% without significant interferences from uranium, thorium, or other rare-earth elements (promethium was not tested) present in amounts up to ten times the concentration of the europium. After extraction of the uranium and thorium by thenoyltrifluoroacetone (TTA) in chloroform, the pH of the aqueous phase is changed from 2 to 5, following which the europium is extracted with TTA in hexone. The emissivity of the extract is then measured at 459.4 mμ. (AEC Activity 4151.1)

Slurry Circulation Experiments. – Following a run in the 200B loop in which thorium-uranium mixed oxide slurry had formed undesirable deposits, a similar run was started with a slurry of pure thoria which had been fired at 1600°C. No plugs or cakes were observed. Heat transfer measurements at 180 to 275°C, with a slurry concentration of 830 g of Th per liter, suggest that turbulent flow is not fully developed until the velocity is about 14 fps. The heat transfer coefficient at that velocity is close to that observed with water at the same conditions; however, the coefficient decreases with velocity much faster than the usual 0.8 power.

A series of runs was completed in the 200A loop, with a thoria slurry at concentrations of 0 to 1000 g of Th per liter, temperatures of 240 to 280°C,

and velocities of 2.7 to 12.5 fps. A gamma-ray densitometer was used to scan the slurry concentration at various elevations in a horizontal pipe. At the highest temperature, evidence of nonuniform concentrations was obtained at high velocities. The experimental data, which provide a complete description of the slurry distribution over the range investigated, are being analyzed.

Construction and shakedown of the 50A loop (a 50-gpm system) were completed, and a slurry of 1600°C-fired thoria, containing the relatively large average particle size of 3.65 μ, was circulated in the initial run. The attack on loop components was comparatively mild. (AEC Activity 4151.1)

Slurry System Development. — A number of improvements and modifications to the 300-SM system were completed, and some preliminary tests with water indicated that the revisions were mechanically sound. The major changes were the installation of a full-flow heat exchanger in the high-pressure loop, improvement of the pressurizer flow configuration, improvement of valve trim materials, elimination as far as possible of mechanical tubing connections in contact with slurry, and simplification of the feed-pump—slurry-dump-tank configuration. (AEC Activity 4151.1)

Core and Blanket Vessel Development. — A study was made of two-region breeders consisting of a blanket containing thoria pellets around a cylindrical core vessel containing uranyl sulfate solution. Assuming that satisfactory pellets can be fabricated, it was found that a 400-Mw reactor with a pellet blanket is economically more attractive than the corresponding reactor with a slurry blanket. A practical 40-Mw model of the larger reactor was also devised. The pellets were assumed to reside as fixed granular beds cooled by the flow of heavy water. Occasional transport of the pellets, to regulate their irradiation, would be accomplished with water jets.

A small blanket test model containing lead and steel shot was operated successfully at room temperature. A mockup of the blanket of the 40-Mw reactor was designed for use in more sophisticated tests.

Fabrication was completed of a cylindrical 2 by 6 ft flow model of the core of the 40-Mw reactor; this vessel will also serve as the scale model of the core of the 400-Mw reactor. Testing of the model will provide information necessary for calculation of the temperature distribution which could be expected in a reactor vessel of this type. (AEC Activity 4151.1)

Component Development. — Testing of titanium impeller wear rings under rubbing conditions in an oxygen atmosphere created a fire which burned through the stainless steel discharge pipe of the test pump. As a result of the tests of Zircaloy and titanium impellers under adverse conditions, it was recommended that wear-ring clearances should be greater than normally specified and that these materials should be operated in water with cutoff protection in the event of gas accumulation.

A slurry low-pressure system was operated in which it was demonstrated that a feed pump could be coupled to a slurry tank with a simple, sloping pipe. The system was adapted for use in the 300-SM loop. (AEC Activity 4151.1)

Development of Remote Maintenance. — Optical equipment was provided for locating a second hole in the core tank of the HRT. Existing tool designs are being modified in order to obtain an impression of the new hole and to

patch it. Tools are also being developed for obtaining a sample of Zircaloy from the vicinity of the hole and for cleaning scale off the core.

Tools for removing the screens from the HRT core were prepared for use. (AEC Activity 4151.1)

Molten-Salt Reactor Program

Chemistry. — In the system $NaF-ThF_4-UF_4$, the compound $4NaF \cdot ThF_4$ dissolves UF_4 extensively, and ThF_4 also has a significant solubility in $3NaF \cdot UF_4$; the phase fields for these solid solutions will be difficult to establish because they appear to grade continuously. Since more than 18 mole % UF_4 can be substituted for ThF_4 in $4NaF \cdot ThF_4$, it now appears that $4NaF \cdot UF_4$, not heretofore observed, can occur metastably in the binary system $NaF-UF_4$.

A representative one-region breeder fuel, $LiF-BeF_2-UF_4-ThF_4$ (67-18.5-0.5-14 mole %), begins to precipitate a solid solution based on $3LiF \cdot ThF_4$ at 499 ± 4°C. This solid solution continues to precipitate as the primary phase even below 438°C, whereas approximately the same melt with no UF_4 present yields $2LiF \cdot BeF_2$ as a secondary phase at 448°C.

Further explorations of uranium oxide precipitation have been carried out. For example, the addition of 18 g of BeO per kilogram of $LiF-BeF_2-ThF_4$ (67-18-15 mole %), containing 1875 ppm uranium as UF_4, decreases the uranium concentration to 40 ppm within 30 min. No further changes in uranium concentration had occurred when the experiment was terminated after 7 hr.

Pure molten $FeCl_3$ causes graphite tubes to crumble to an intercalated powder within 2.5 hr at 300°C; however, no readily apparent damage occurred in 7.5 hr at 200°C when the $FeCl_3$ was diluted with 46 mole % NaCl, or within 15 hr at 700°C when diluted with 70 mole % NaCl. These experiments, initially intended to demonstrate the effect of temperature, show that dilution also plays a relatively important role in determining whether or not intercalation occurs. (AEC Activity 4152.1)

Metallurgy. — Techniques have been developed to utilize the highly localized heat of an electron-beam welding machine for sealing fill tubes on fuel-graphite capsules used for in-pile testing. The ends of the tubes were welded shut after the capsule was completely filled with salt. The equipment was also used to braze metal sleeves on sections of these tubes for repair purposes, utilizing the localized heating effect to keep the salt from melting and rupturing the capsule.

Specimens of three additional grades of low-permeability graphite have been exposed at 1300°F to BULT 14-0.5 U salt for 100 hr under a pressure of 150 psig in order to determine the degree of salt permeation. These graphites compared favorably with the most impervious grades previously tested. In no case was more than 1.0% of the bulk volume of the specimen filled with salt. (AEC Activity 4152.1)

Corrosion Tests. — Eleven corrosion pump loops of INOR-8 and two of Inconel were operated during this month. Eight of these loops have been in operation in excess of one year. The 13 loops operate at high wall temperatures of 1200 to 1500°F, with various fluoride salts.

Thin metallic films with thicknessess of less than 1/2 mil have been observed on surfaces of INOR-8 loops which circulated fluoride salt mixtures

for periods of over 5000 hr. These films occurred in both hot and cold sections of the loops and have been identified by spectrographic analysis as containing nickel and molybdenum in a weight ratio of 2 to 1, indicating a loss of nickel in these areas. This ratio approximates that of the beta phase in the Ni-Mo system. (AEC Activity 4152.1)

Graphite Systems. — A second frozen-salt graphite-to-metal joint was established in a loop circulating molten salt at 1150°F. The seal formed in an annulus between the graphite and metal surfaces did not leak when a pressure differential of 18 psi existed for 24 hr. However, salt leakage did occur through the wall of the type S-4 graphite container. (AEC Activity 4152.1)

In-Pile Tests. — Operation of the first MSR in-pile graphite-fuel capsule experiment, inserted in the MTR the week of January 25, has been satisfactory. Sodium temperature in the vicinity of the capsules is 1220°F, which should give a fuel temperature of about 1300°F. The fuel pressurization system is operating at about 105 psia, or 5 psi above the specified operating conditions.

The second experiment, which will be shipped to the MTR the week of February 21, contains four INOR-8 capsules containing varying grades of graphite submerged in molten-salt fuel. The operating conditions will be similar to those of the first experiment: 1300°F fuel temperature, 200 w/cm^3 fuel-power density, and 100 psia applied to the fuel. (AEC Activity 4152.1)

Pump Development. — The molten-salt pump containing a salt-lubricated journal bearing operated for 1000 hr at 1200°F, 1400 rpm, and 100 gpm. The test was suspended for examination of the salt bearing, the face-type shaft seal, and the oil-lubricated ball bearing. This examination showed the salt bearing to be in good condition, with only a slight sign of rubbing between the bearing surfaces. The bearing sleeve and the shaft seal are suitable for operation in the future. The total oil leakage from the shaft seal during the 1000-hr test was approximately 30 cc. Assembly of the pump for start-stop and speed-variation tests is under way.

The journal-bearing tester was placed in operation with a carburized INOR-8 journal and a helically grooved INOR-8 bearing in salt 130 at 1200°F, 1200 rpm, and a load of 200 lb. After 80 hr of apparently smooth operation, the drive motor experienced a sudden power surge. The test was suspended for examination of the bearing and journal.

The MF-F endurance test has operated continuously for more than 23,160 hr, circulating salt 30 at 1200°F. The PKP pump rotary element was assembled and is ready for installation in the hot test stand for cold shakedown prior to renewing high-temperature endurance testing with NaK. (AEC Activity 4152.1)

Remote Maintenance Demonstration. — Dress-rehearsal types of operations are in progress on the remote removal and installation of the heat exchanger, molten-salt circulating pump, pump motor, and reactor. Closed-circuit television is used as the only viewing medium for all the remote mechanical maintenance operations. Design of the last remotely operated tool has been completed. (AEC Activity 4152.1)

Heat Transfer and Physical Properties. — The enthalpy equations for the BULT-14 salt mixture ($LiF-BeF_2-UF_4-ThF_4$, 67-18.5-0.5-14 mole %) as reported in ORNL-2895 contained several errors. Thus, for the solid (100–400°C), the equation should read,

$$H_t - H_{30}\ (\text{cal/g}) = 5.0 + 0.185t + (7.8 \times 10^{-5})\ t^2 \quad ;$$

and for the liquid (500–800°C),

$$H_t - H_{30}\ (\text{cal/g}) = 61.5 + 0.493t - (11.71 \times 10^{-5})\ t^2 \quad .$$

Accumulation of data on the heat transfer coefficients of BULT-14 salt flowing in electrically heated Inconel and INOR-8 tubes has continued. The system has been in continuous operation for nearly 3000 hr with only minor maintenance shutdowns; data obtained at approximately biweekly intervals show no significant variation of the heat transfer coefficient with time.

The data have been recalculated based on heat capacities for BULT-14 evaluated from the enthalpy equations listed above; a constant value of 0.29 $\text{Btu}\cdot\text{lb}^{-1}\cdot(°\text{F})^{-1}$ was used in the initial data reduction. While little effect on the heat transfer coefficients was noted, heat balances (ratio of the fluid heat gain to the net energy input) were somewhat improved. (AEC Activity 4152.1)

Fuel Processing. — The solubilities of the components of a potential fuel salt for a one-region breeder reactor (LiF-BeF_2-ThF_4-UF_4, 67-18.5-14-0.5 mole %) were measured in aqueous 50% NH_4F solution at 100°C. After 1 hr all the BeF_2 and UF_4 had dissolved, the respective concentrations being 13 and 2.15 mg/ml, but only 10 to 15% of the LiF and ThF_4 had dissolved, their solubilities being 2.2 and 9.3 mg/ml, respectively. Thorium fluoride by itself is soluble to at least 45 mg/ml. After another day, during which there was a noticeable loss of ammonia gas, all solubilities were lower except that of BeF_2.

Detailed study of the relation between poisoning and chemical processing rates and methods showed that neutron loss to protactinium in the blanket is serious unless protactinium can be removed rapidly. In preliminary tests, neither fluoride volatility nor oxide precipitation appeared promising. From LiF-BeF_2-ThF_4-UF_4 (67-18.5-14-0.5 mole %) containing 0.012 wt % Pa^{231}, protactinium was not significantly removed by a 2-hr fluorination with elemental fluorine at 650°C or by addition of 1% CaO. (AEC Activity 4152.1)

Thermal-Breeder Evaluation

Thermal-Breeder Evaluation Studies. — A study has been made of the relative ease of keeping the Xe^{135} poison at a low level in three fluid-fuel breeder reactors. In the aqueous system the xenon poisoning in the core solution can be reduced to about 1% by iodine removal and to still lower levels by gas purging of the xenon. In the molten-salt and bismuth reactors, gas purging of the xenon should be effective in keeping the xenon poisoning in the fuel stream to less than 1%, provided the iodine precursor does not diffuse into the graphite. In all the reactors, the cost of keeping the Xe^{135} poisoning down to 1% is small enough to be justified on an economic basis. (AEC Activity 4160)

MARITIME SHIP REACTOR PROGRAM

Charpy V-notch specimens of A212B steel were prepared from excess of material used to fabricate the NS "Savannah" reactor closure head and were irradiated in the ORR pressurized-water loop for one ORR cycle. The temperature of the specimens during irradiation was approximately 500°F, and it is estimated that the maximum fast-neutron dosage to the specimens exceeded that to be expected during 20 years of operation of the NS "Savannah" reactor. Examinations of threshold detectors irradiated with the specimens will permit a determination of the fast-neutron dosage. Impact properties of the irradiated specimens are to be determined for comparison with previously measured values of the same material in the unirradiated condition. (AEC Activity 4510)

GENERAL REACTOR RESEARCH

Radiation Damage: Advanced Engineering and Development. — Diffusion-controlled atomic rearrangements in alloys are accelerated upon irradiation. Optimum properties are produced in many alloys by allowing a diffusion-controlled reaction to proceed to a critical degree at an elevated temperature. When the material is returned to room temperature, it is in a metastable state; but the metastability persists because of the low atomic mobilities. However, when the material is used in a radiation environment, the atomic mobilities are enhanced and the alloy may achieve the equilibrium atomic arrangement, with an attendant deterioration of properties. The phenomenon of radiation enhancement of atomic rearrangements in alloys is being studied on Cu-Al alloys. Irradiation at 100°C by reactor neutrons, Co^{60} gamma rays, or electrons from a Van de Graaff accelerator causes a decrease in resistivity to take place. The magnitude of the decrease in resistivity increases with the aluminum content of the alloy. For the 15 at. % Al alloy, the decrease amounts to about 0.2 μohm-cm, or about 2%. The neutron irradiation has also been carried out at -120°C, in which case no decrease in resistivity was observed during the irradiation, although a decrease in resistivity was observed when the temperature was subsequently raised above -50°C. The implication from this experiment is that the rearrangement does not take place until the radiation-produced defects can migrate through the crystal. With neutron irradiation at a series of temperatures between 45 and 250°C, the resistivity decreased to a lesser extent as the temperature of irradiation was raised, which indicates that the initial atomic arrangement corresponds more closely to the equilibrium state at the higher temperatures. (AEC Activity 4410)

Basic Reactor Research: Reactor Theory. — An Oracle code has been written to compute the Fourier transform of the P_1 approximation to the slowing-down density of neutrons from a plane source in an infinite homogeneous medium. Results are in good agreement with results obtained by Monte Carlo methods. A total time of 5 min is required to compute and record (on punched tape) 31 group results for a given value of the buckling. (AEC Activity 4410)

Basic Reactor Research: Pulsed-Neutron Work. — The ratio of the prompt-neutron generation time to the effective delayed-neutron fraction, L/β_{eff}, has

been measured in critical cores of the BSR-I, the Puerto Rican reactor, and a BeO-reflected reactor. The familiar pulsed-neutron technique was employed, with high-energy neutrons being obtained from the D-T reaction utilizing the 300-kv BSF particle accelerator. The preliminary values of L/β_{eff} were as follows: for the BSR-I, 8.54 msec; for the PRR, 7.9 msec; and for the BeO-reflected reactor, 11 msec. No reliable error analysis of these numbers has yet been made. Control-rod worths were also measured for the BSR-I and PRR cores. It is intended to compare these experimental results with the results of various machine code calculations when they are completed. (AEC Activity 4410)

Basic Shielding Research: Total Absorption Gamma-Ray Spectroscopy. — An investigation of a 5-in.-dia, 3-in.-long CsI(Tl) scintillation crystal has been completed. A resolution of 9.8% for the 662-kev gamma ray from Cs^{137}, together with a peak-to-total ratio of 74%, was measured. The expected improvement in peak-to-total ratio over a comparable NaI(Tl) crystal was observed to continue through 2.76 Mev. A comparison of these results with values predicted by an existing machine code is being made. (AEC Activity 4410)

Basic Shielding Research: Spectrum of Prompt Gamma Rays Associated with Thermal-Neutron Fission of U^{235}. — Analysis of experimental data is continuing. A new gamma-ray standard, to be used for pulse-height calibrations required in the conversion of the present pulse-height spectrum to an absolute gamma-ray spectrum, is being investigated. The standard utilizes the $B^{11}(p,\gamma)C^{12*}$ reaction, employing a thick boron target in the BSF 300-kv particle accelerator, with a proton energy of ~ 175 kev. Decay gammas from the excited state of C^{12*} are expected at 4.43 and 11.7 Mev, with a few at 16.1 Mev from the direct transition to the ground state. During the investigation of this standard an attempt is being made to measure the absolute source strength, using the standard Huber coincidence method. (AEC Activity 4410)

Power Reactor Fuel Processing: Zirflex Process. — In a series of cyclic Zirflex decladding-dissolution runs on unirradiated prototype PWR fuel pins (Zircaloy-2-clad UO_2), the amount of solids found in filtered decladding waste and product solutions increased with the number of cycles. The $ZrO_2 \cdot 2H_2O$ in the decladding solutions probably was formed when ammonium hydroxide was returned to the dissolver from the updraft condenser. Stannic oxide was the chief component of the solids in the core solutions. A gradual increase in the pore size of the glass filter used in these tests may be responsible for the appearance of increasing amounts of solids as the number of cycles was increased.

In Zirflex corrosion tests Haynes experimental alloy EB4359 was corroded at a rate of 6.8 mils/month, as compared with 8.0 mils/month for vacuum-melted Hastelloy F. The 24-hr corrosion rates of Ni-o-nel and type 309SCb stainless steel were decreased from 27 and 23 mils/month to 13.3 and 10.9 mils/month, respectively, by adding 0.1 M $Na_2B_4O_7$. However, the presence of 0.1 M dissolved Zircaloy counteracted the inhibiting effect of the borate. (AEC Activity 4441)

Power Reactor Fuel Processing: Sulfex Process. — In depassivation tests in which stainless steel dissolution was started in 8 and 10 M H_2SO_4 solutions

which were diluted to 4 M H_2SO_4 as soon as depassivation occurred, Ni-o-nel showed maximum corrosion rates of 3.9 and 9.5 mils/month, respectively. The presence of 35 g/liter of stainless steel dissolved in 4 M and 6 M H_2SO_4 decreased the corrosion rate of Ni-o-nel from 5.3 to 1.2 and from 11.5 to 1.6 mils/month, respectively.

In semicontinuous Sulfex decladding tests on prototype Consolidated Edison power reactor fuel (type 304 stainless-steel-clad ThO_2-UO_2), the reaction rate for the cladding in 4 M H_2SO_4 was approximately half the rate found for type 304 stainless steel plate. The reaction rate for tubular cladding was 1.0 to 1.5 $mg \cdot min^{-1} \cdot cm^{-2}$. The product metal loading was 25 to 30 g of stainless steel per liter.

The initial rate of dissolution of 95.8% ThO_2–4.2% $UO_{2.36}$ pellets (94% of theoretical density) in 200% excess of boiling 13 M HNO_3 increased from 0 to a maximum of about 13 $mg \cdot min^{-1} \cdot cm^{-2}$ as the fluoride concentration in the acid was increased from 0 to about 0.07 M. The rates were lower when the dissolvent contained aluminum.

A 2-hr test to determine whether ThO_2 particles of 63 μ mean diameter could be settled preferentially from a gelatin-precipitated siliceous slurry at 85°C showed no significant separation of the two solids. A steam jet–hydroclone unit was tested to determine its applicability as a critically safe solids separation device for Sulfex decladding and wash solutions. Crushed porcelain of < 74 μ particle size was used to simulate UO_2. It was estimated that 5% of the solids passed through the unit to the overflow. (AEC Activity 4441)

Power Reactor Fuel Processing: Solvent Extraction Studies. – Synthetic Consolidated Edison fuel processed by an Acid Thorex flowsheet with an acid feed was decontaminated from ruthenium, zirconium-niobium, protactinium, and rare earths by factors of 20, 2000, 200, and 1000, respectively. The use of this flowsheet for a second cycle following an Acid Thorex first-cycle flowsheet with acid-deficient feed should give thorium and uranium products which meet storage specifications.

Tributyl phosphate may be removed from aqueous product solutions by a two-stage countercurrent contacting with 1/50 volume of Amsco. About 95% of the used Amsco may be recovered by steam stripping. (AEC Activity 4441)

Power Reactor Fuel Processing: Processes for Graphite Fuels. – Uranium and thorium recoveries were > 99% when -10 mesh samples of prototype ungraphitized Pebble Bed Reactor fuel were leached with boiling 13 M HNO_3–0.04 M NaF–0.1 M $Al(NO_3)_3$, as compared with 7% thorium recovery with 15.8 M HNO_3. (AEC Activity 4441)

Power Reactor Fuel Processing: Mechanical Processing. – The results of exploratory tests on the reaction that occurs when NaK is contacted with water under a mineral oil blanket indicated that a violent hydrogen explosion would not occur and that continuous reaction cannot be sustained unless steam, or some external heat, is used to agitate the solution and increase its temperature to above 100°C. (AEC Activity 4441)

Power Reactor Fuel Processing: Darex Process. – The PuO_2 produced in stainless-steel-clad UO_2 fuel sample pins irradiated to 16,100 Mwd/ton was found to be completely soluble in the Darex dissolvent, 5 M HNO_3–2 M HCl.

In Thorex dissolver solutions containing 0.1 M $Na_2B_4O_7$, titanium was corroded at maximum rates of 1.0 and 0.3 mils/month in beginning and final solutions, respectively. There appeared to be less pitting on these specimens than on those exposed to solutions containing Al^{+++}. After 41 Darex-Thorex (integral dissolution) cycles, the maximum corrosion rate of welded titanium specimens was 0.06 mil/month. Under total reflux conditions of Thorex feed adjustment solutions, welded titanium at the bottom of the condenser showed localized weld attack; over-all rates were as high as 1.9 mils/month. During Thorex feed adjustment by steam distillation, the maximum fluoride concentration in the condensate was 2.4×10^{-3} M; no appreciable corrosion of titanium was noted. After 840 hr of exposure to beginning Thorex dissolver solutions, tantalum showed a maximum rate of 0.30 mil/month.

Type 304L stainless steel exposed to Darex-Thorex waste solutions continued to be corroded at essentially constant maximum rates of 0.006, 0.030, and 0.065 mils/month, at 50, 65, and 80°C, respectively. (AEC Activity 4441)

Power Reactor Fuel Processing: Zircex Process. — In a variation of the Zircex flowsheet, uranium-zirconium fuels are first hydrochlorinated with HCl at ~ 500°C, to remove most of the zirconium as volatile $ZrCl_4$. The residual ZrO_2-UCl_3 is then contacted with a gaseous CCl_4-O_2 mixture to produce volatile $ZrCl_4$-UCl_4.

Corrosion rates of INOR-8, Nichrome V, and Illium R after two Zircex cycles were 1.9, 1.4 to 6.9, and 7.3 to 11.0 mils/month, respectively. (AEC Activity 4441)

Power Reactor Fuel Processing: Radiation Damage to Coatings. — In irradiation tests to evaluate protective coatings for use in the Power Reactor Fuel Processing Pilot Plant deionized water canal at 135°F and at a gamma-ray exposure of 3×10^9 r, Phenoline-300 experimental cover coats failed at 1×10^9 r, whereas Amercoat No. 74 surfacer, Amercoat Nos. 66 and 1680, and Dimetcote No. 3 were not damaged at this radiation level. (AEC Activity 4441)

Phase Studies for Fuel Reprocessing. — Studies of the phase equilibria in four fluoride fused-salt systems whose use will broaden the applicability of the Volatility Process for recovery of uranium from spent reactor fuels have been made. These are LiF-NaF-ZrF_4, NaF-BeF_2-ZrF_4, LiF-BeF_2-ZrF_4, and NaF-FeF_2-ZrF_4. Compositions of the lowest liquidus surfaces of each system have been established. Phase diagrams for the system LiF-NaF-ZrF_4 and its limiting binary systems have been constructed. (AEC Activity 4442)

Waste Disposal Research and Engineering: Geochemical Studies. — Data on increasing the selectivity for cesium by reducing the c-axis spacing of clay minerals to 10 Å have been previously reported. Experiments conducted recently have been designed to test whether cesium selectivity might be used to differentiate interstratified systems of clay minerals. An important application of this technique would be in establishing the purity of minerals. Thus illite is sometimes regarded as an interstratified mineral of two components, though only a 10-Å c spacing is observable by x-ray diffraction. If, as is suspected, the impurity is montmorillonite, heating to 500 to 600°C should collapse the montmorillonite layers, and the selectivity for cesium should be increased. Tests on Fithian illite (Ward's standard

specimen No. 35) show that hydrated montmorillonite-type layers do exist according to the cesium selectivity criterion. This technique when perfected should be a valuable additional tool for solving the difficult problem of quantitative clay mineral analysis of sediments. (AEC Activity 4452)

Waste Disposal Research and Engineering: Disposal in Natural Salt Formations. — On January 6, power was applied to the 3160 gal of simulated neutralized Purex waste in the field-scale experiment. Startup of the experiment with simulated acid waste was on January 18. The power to both cavities, which was initially 5.8 kw, is being decreased in accordance with the decay of two-year-cooled wastes (800 gal/ton, 10,000 Mwd/ton, 33 Mw specific power).

After 45 days of operation the average neutralized waste temperature was 72.2°C and approaching equilibrium. At 8 ft from the edge of the cavity, the temperature was 29.7°C and also approaching equilibrium. At 30 ft from the cavity the temperature rise above ambient (~ 21°C) was less than 0.5°. After 33 days of operation the acid waste temperature was 62.3°C, and the salt temperature rise was following closely that of the neutralized experiment and predicted by the theoretical calculations. (AEC Activity 4452)

Waste Disposal Research and Engineering: Disposal of ORNL Radioactive Wastes. — A series of auger holes, ranging in depth from 5 to 20 ft, has been completed in and around the Melton Valley burial ground (recently abandoned) to determine more specifically the geology, hydrology, and movement of radionuclides in the area.

Water-level measurements, taken about once each week since August 1959, indicate that the water table fluctuates between 2 to 3 ft below the surface in low areas of the burial ground and 10 to 15 ft beneath higher elevations. Thus solid waste is in contact with ground water during most periods of the year.

Activity has been detected in water samples from 8 of the 16 wells located within the burial ground. Radionuclides have also been found in seeps and streams in the area. Identifiable contaminants include Ru^{103}, Ru^{106}, Co^{60}, Cs^{137}, Sr^{89}, Sr^{90}, rare earths, and Po^{210}. Most of the activity occurs in wells and seeps that lie in low areas. Soluble waste material is transported by ground water through the weathered soil to points of discharge in or near surface streams.

Since all drainage from the area is into White Oak Creek, temporary monitor stations have been installed immediately above and below the burial ground to determine the amount, if any, of activity contributed by it. (AEC Activity 4452)

Waste Disposal Research and Engineering: Deep Well Disposal. — At the "Four Acre Site," the location of the first field experiment in waste disposal by hydraulic fracturing in shale, cores drilled northeast of the injection well at distances of 37, 200, and 275 ft were found to contain grout. No grout was found at 310 ft. The thickness of the grout sheet ranged from 0.25 in. at 37 ft to 0.08 in. at 275 ft, and the thickness correlated well with the magnitude of the response obtained by radioactivity logging. The magnitude of the response also correlated well with the response obtained in laboratory tests with sheets of known thickness and composition. In those cases where no grout was found in the cores its absence was positively confirmed by radioactivity logging, which completely justifies the use of the radioactive tracer.

Drilling to the southwest did not reveal any grout 300 ft from the point of injection, and coring is now in progress at 250 ft. It will take about eight more wells to define the extent and thickness of the grout sheet. (AEC Activity 4452)

Waste Disposal Research and Engineering: Process Waste Water Treatment Plant Studies. — Preliminary investigations on the distribution of radioactivity in the effluent from the process waste water treatment plant showed that approximately 50% of the outgoing activity is associated with the suspended solids in the effluent. If the amount of suspended solids present in the effluent could be reduced, it would then be possible to increase the efficiency of the treatment plant.

Several coagulant aids were tested in the laboratory by the conventional jar test technique, under varying conditions of coagulant aid concentration, mode and time of addition, agitation time and speed sequences, temperature, and presence of complexing agents. A combination of Hagan aids Nos. 50 and 18 was found to give fairly good results under most conditions; however, a six-week trial on a plant scale showed that slightly higher concentrations of the aids than those used in the jar tests would be required to get good results. Also, the concentration of the feed solutions and the mode and point of addition are factors influencing the coagulation. While the combination of the aids used gives a good rapidly settling floc, a drop in temperature of the waste water results in an increase in the amount of very fine particles, which have a slower settling rate. Minor modifications to the plant equipment will be necessary to get the best results. The coagulant aids do not work well if the system contains complexing agents, such as Versene, polyphosphates, etc. (AEC Activity 4452)

Waste Treatment and Disposal. — In laboratory studies the addition of calcium hydroxide to synthetic Purex (nitrate) waste decreased the volatilization of sulfate during calcination, but the calcium sulfate precipitate caused bumping and foaming. Addition of sodium nitrate or commercial colloidal calcium silicate both prevented sulfate volatilization up to 900°C and decreased ruthenium volatility. Synthetic Purex waste containing 0.8 to 24 ml of tributyl phosphate per liter was evaporated and calcined without violent reaction, but frothing and bumping were severe.

In an 8-in.-dia by 84-in.-high vessel, synthetic Purex waste to which calcium hydroxide had been added was evaporated and calcined with no bumping but with some frothing. The product was a cake having a density of 3 g/cc and a thermal conductivity at room temperature of 0.66 $Btu \cdot hr^{-1} \cdot ft^{-2} \cdot (°F/ft)^{-1}$. The volume was decreased 240-fold, which is equivalent to decreasing to 5 gal the waste from processing of 1 ton of uranium. Waste was fed to the evaporator continuously at a rate of 20 liters/hr, the rate of waste production in a plant processing 12 tons of uranium per day. Eighty per cent of the nitrate was recovered from the off-gas by a downdraft condenser. In tracer-level studies the calciner off-gas was decontaminated by a factor of 5000 by passage through a 3-in.-dia by 10-ft-high packed column.

When 406 volumes of synthetic process-water waste spiked with mixed fission products was passed over one volume of hydroxyl-form TAD-1 anion resin at a flow rate of 0.5 ml/min per milliliter of resin, 72% of the gross gamma activity was removed (decontamination factor, 3.6). The column effluent, adjusted to pH 11.3, was passed over a sodium-form phenolic cation resin at a flow rate of 0.7 ml/min per milliliter of resin to remove 95% of the remaining activity (decontamination factor, 20). The over-all volume reduction factor was 1867. (AEC Activity 4452)

PROGRAM 5000 – PHYSICAL RESEARCH

ISOTOPE PRODUCTION

Radioisotope and Stable Isotope Production. – The collection of the isotopes of samarium was completed with ion currents equivalent to 110 g. The collection of the isotopes of indium, barium, and calcium is now in progress.

Charge materials were produced for the separation of the isotopes of Sm, In, Ni, Ba, Pb, and Sn.

New lots of copper, nickel, and ruthenium were prepared for inventory following spectrochemical and mass analyses.

Special services included the following conversions: $H_3B^{10}O_3$ to B_2O_3, providing the $B_2^{10}O_3$ in a D_2O solution of known concentration; Li^6 metal (96.1% Li^6) to Li_2SO_4 (two requests); Li^6 metal (99.3% Li^6) to Li_2SO_4; Ru^{102} and Ru^{104} in the metallic form to $RuO_2 \cdot xH_2O$; and elemental Se^{76} to SeO_2. Target preparation included 18 conversions and 97 targets.

Radioisotope Research and Development. – Previous experiments with single-stage bubbler contactors indicated that water is relatively ineffective and that NaOH solutions above 1 N are very effective for scrubbing $Ru^{106}O_4$ from an air stream. Results obtained from scale-model runs in a 4-in. glass column with 1 ft of 1/2-in. Berl saddle packing indicate that 99.9% of the ruthenium can be removed from the Fission Product Pilot Plant off-gas air stream in either 2.5 M or 10 M NaOH solution. The presence of RuO_2 was indicated by the removal of 90% of the remaining activity in the air stream on a glass-wool filter following the scrubber.

A stainless steel prefilter of the impact type has been selected for installation in the Fission Product Pilot Plant cell ventilation and off-gas filter boxes on the basis of tests which indicate that it is as efficient as the previously used glass fiber prefilters. For installation in air streams bearing highly radioactive particles the stainless steel filter has the advantage of being semipermanently installed and washable by a water spray when it is loaded, thus obviating the necessity of periodic changes of very radioactive filters.

The final design of containment modifications for the Fission Product Pilot Plant is 80% complete, and a preliminary study of containment for the radioisotope area buildings has been completed. The title I design of the new Radioisotope Development Building was revised to include provisions for containment. The title II design for this building is in progress.

A batch of 23.9 g of technetium was purified by processing a Fission Product Pilot Plant concentrate by pyridine extraction and NH_4TcO_4 crystallization. A 10.5-g quantity of technetium metal was prepared by hydrogen reduction of the NH_4TcO_4. Tests of organic extractants other than pyridine for technetium are in progress.

Preliminary design is in progress on a thermal diffusion system for the enrichment of C^{13}, with methane as the operating gas. (AEC Activity 5100)

Stable Isotope Development. – The accumulation of sputtering data involving 5-kev boron ions was stopped temporarily to permit increased emphasis on specific isotope collector designs. As a result of this effort, the last separation of chlorine isotopes yielded a substantial increase in the purity of Cl^{35} and Cl^{37}. A similar type of bromine separation is in progress.

Experimental data acquired to date on osmium are being compiled for review. The results obtained may indicate the feasibility of a calutron separation of osmium isotopes. (AEC Activity 5100)

PHYSICS

High-Voltage Experimental Program: Total Reaction Cross Sections for Alpha Particles on Ni^{58}. – Thick-target yields have been measured for the (α,γ) reaction from 4.9 to 6.1 Mev, for the (α,p) reaction from 4.9 to 11.1 Mev, and for the (α,n) reaction from threshold (10.49 ± 0.03 Mev) to 11.3 Mev by using isotopically enriched Ni^{58} targets. The (α,γ) and (α,p) reactions were identified by measuring the coincident annihilation radiation from the induced β^+ activities. The absolute thick-target yields, which were measured at approximately 100-kev energy intervals, have an accuracy of ±6% (aside from possible uncertainties in the branching ratio β^+/ϵ for the Zn^{62} and Cu^{61} activities). The partial reaction cross sections were obtained to an accuracy of ±15% by differentiation of the smooth thick-target yield curves. Over the measured energy range, the cross sections increase from 1 to 12 microbarns for the (α,γ) reaction and from 0.6 microbarn to 316 millibarns for the (α,p) reaction. For the (α,n) reaction, the cross section increases from 7.7 to 16.7 millibarns between alpha energies of 10.8 and 11.3 Mev. At alpha energies of 5 and 10 Mev the reaction cross section is 7 and 1.5 times smaller than that predicted by a calculation [Phys. Rev. 115, 1665 (1959)] based on an optical-model potential obtained from the analysis of elastic scattering data. (AEC Activity 5220)

High-Voltage Experimental Program: Asymmetry of Mott Scattering by Foils of Finite Thickness. – Transversely polarized electrons can be scattered by a thin foil (Mott scattering). The resulting left-right asymmetry $(PS)_t$ depends upon the degree of polarization P, Sherman's function S, and the foil thickness t. Single scattering is always accompanied by plural and small-angle scattering. Expansion of the asymmetry in terms of the foil thickness, $(PS)_t = (PS)_0 + (PS)_1 t + \ldots$, gives the desired single-scattering term $(PS)_0$, the influence of small-angle and double scattering $(PS)_1 t$, etc. The double-scattering term diverges logarithmically either if one of the two scattering angles goes to zero or if the first scattering occurs in a direction perpendicular to the normal of the foil. Both divergences vanish after applying the Bothe-Fermi theory of small-angle scattering to these regions. The straightforward calculations were done on the Oracle. A comparison of the results with more approximate calculations [Z. Physik 151, 252 (1958)] shows that the latter underestimated the double scattering in

the regions between the divergences. The Oracle results agree well with experimental data obtained at Oak Ridge and Erlangen. (AEC Activity 5220)

High-Voltage Experimental Program: Neutron Yield from the Proton Bombardment of C^{14}. – Measurement of the yield of neutrons from the proton bombardment of C^{14} has been extended to 5 Mev with good resolution and with targets of high isotopic enrichment. Measurements were made at three angles with the long counter and at one angle with a proton recoil counter. The regions of the thresholds for the second and third neutron groups were investigated with a lithium iodide detector. The very narrow level at 13.15 Mev excitation did not appear, but the 6-kev-wide level just above threshold was observed and a level (13.36 Mev) was resolved on the low-energy side of the previously observed level at a proton energy of 3.41 Mev. In the previously unobserved region above E_p = 3.41 Mev, anomalies were found in the excitation curve at E_p = 3.63 (Γ = 13 kev); 3.89 Mev (Γ = 35 kev); 4.19 Mev (Γ = 112 kev); 4.24 Mev (Γ = 27 kev); 4.60 Mev (Γ = 140 kev); and 4.93 Mev (Γ = 106 kev). There was some indication that the anomaly at 4.2 Mev may be double. The threshold for the third group of neutrons was observed at 4.910 ± 0.008 Mev, in good agreement with the known energy of the second excited state in N^{14}. (AEC Activity 5220)

Heavy-Particle Physics. – Work is continuing on the inelastic scattering of N^{14} from the 4.43-Mev state in carbon. A decrease in the cross section in the forward direction is interpreted as indicating that the mechanism responsible for this reaction is Coulomb excitation. Certainly no simple direct-interaction picture seems to explain the data.

Thin (0.6 mg/cm^2) Cymel plastic foils, containing 25% nitrogen by weight, were successfully used to study the angular distribution from the reaction $N^{14} + N^{14} \rightarrow N^{13} + N^{15}$. Both reaction products are in the ground state. The radioactive N^{13} atoms were caught on circular aluminum strips placed behind appropriate absorbers. The angular distribution exhibits a pronounced maximum at 31° in the center-of-mass system. It was also found that, within the errors of the measurement, the entire cross section for this reaction is contained in the observed peak; excited states in N^{15} contribute very little. This is not unexpected, since the first excited state in N^{15} is at 5.28 Mev and transfer reaction cross sections show a strong dependence on the Q value. (AEC Activity 5220)

The 86-Inch Cyclotron: Nuclear Physics. – Recent experiments with electron-capturing Re^{181} (half life 20 hr) have shown 75 internally converted transitions with energies up to 1.5 Mev. A partial decay scheme for W^{181} based on these data indicates nine intrinsic orbitals, four of which exhibit rotational excitation. It has been previously suggested that the intense 365-kev transition de-excites a (5/2 – [512]) state to ground (9/2 + [624]). A sequence of three levels may well be the expected low-spin states: 385 kev (1/2 – [510]), 560 kev (3/2 – [512]), and 746 kev (1/2 – [521]). A number of intrinsic states of high spin are possible at 1469 kev: (9/2 – [505]), 953 kev (7/2 + [633]), 807 kev (7/2 – [503]), and 409 kev (7/2 – [514]). (AEC Activity 5220)

The 86-Inch Cyclotron: Applied Physics. — The isotopes Be^7, V^{48}, Co^{57}, As^{74}, and Tl^{202} were produced as service irradiations for outside customers, and Tc^{95m} was produced for the Chemical Technology Division. These irradiations represented 52.8 hr of bombardment time. The Tl^{202} was produced by the bombardment of $HgCl_2$ (free of Tl) contained in a 2.5- by 3-in. nickel tube with the ends crimped. The target withstood 30 μa for an 18-min run. Two old experimental targets which contained Bi^{207} were made available to outside customers. (AEC Activity 5220)

Electronuclear Machines. — The Cyclotron Analogue II, an electron cyclotron which will have a maximum energy of about 450 kev, is being built to study the dynamics of the proposed 850-Mev fixed-frequency proton cyclotron, particularly with respect to beam extraction. The design of the sector coils is essentially complete; the final magnetic field calculations are now approximately 60% complete. Components of a test sector coil were received from the fabricator and were thoroughly inspected for dimensional accuracy. The results were gratifying in that the dimensions in the coil shape were within the allowed ±0.001 in. tolerance. The deviations from flatness exceeded the tolerance (±0.0005 in.), and to correct this some redesign of the holding fixtures will be required. Fabrication of the vacuum tank and procurement of components for the vacuum system are proceeding according to schedule. (AEC Activity 5240)

Relativistic Isochronous Cyclotron (ORIC). — Construction of the ORIC building (Building 6000) is proceeding as rapidly as weather will permit. The reinforced concrete walls of the basement are in place, and the concrete shielding walls are being poured.

Precise magnetic field measurements are now being made on the quarter-scale magnet and evaluated with special codes developed for use with the IBM-704 computer. The over-all reproducibility of the measurements, including probe positioning and magnet excitation drift, is 0.02% of the maximum field. A polar plot of 2400 points is completed in less than 12 hr. Any given magnetic field configuration is measured and the evaluation is completed in 36 hr. Precision equipment is being assembled for measuring the field of the full-scale magnet.

Design and fabrication of cyclotron components are on schedule. Components of the full-scale r-f model are fabricated; the assembly should be completed and ready for testing within a week. The dee stem and associated structures are designed and the low bid is being reviewed. The RCA-6949 tube was selected; the power supply is designed and specifications are being prepared. The shorting bar is designed and a model is ready for testing.

The vacuum tank and valley coils are being designed. The source and probe, vacuum manifold, and the transporter for the dee and the dee stem structure are being checked before release for bids. The pole-tip sectors and pole-base disks are being fabricated.

Special Separations. — Plutonium and uranium operations have been suspended during this period for a continued review of hazards associated with the Special Separations program in Building 9204-3. During this interim, certain facility equipment items and services have been cleaned for re-use or have been removed.

The following shipments were made: uranium, 4; thorium, 1; and neptunium, 1. (AEC Activity 5250)

CHEMISTRY

Thorium Oxide. — A slurry of 1600°C-fired thorium oxide at a concentration of 100 g of Th per kg of H_2O and containing a sol-prepared palladium-thoria catalyst at a concentration of 117 parts of Pd per million parts of Th gave catalyst performance indices for H_2 and O_2 recombination of 0.3 and 0.9 w/ml at 250 and 280°C, respectively. Adding fresh thoria to the original suspension in increments of 100 g of Th per kg of H_2O to a final concentration of 400 g/kg did not significantly change the catalytic activity of the slurry.

Thoria powders irradiated for 16 months in the LITR were recovered for examination. The total number of fissions in the powders, estimated from the Cs^{137} activities, varied from 3×10^{18} to 7×10^{18} per gram of thorium. Samples that had been originally fired at 650, 800, and 900°C were red, had agglomerated into a few large pieces, and showed marked decrease in surface area. Samples fired at 1100 and 1500°C were recovered as gray and blue powders, respectively, without a significant change in surface area. (AEC Activity 5310)

Surface Chemistry. — Electrokinetic transport properties of several thorium oxide powders in dilute solutions of HCl, NH_4OH, KCl, HNO_3, Na_2CO_3, and Na_2SO_4 were determined at 25°C. The concentrations of the electrolytes were varied stepwise from 10^{-6} to 1 N. The zeta potential, the surface conductance, and the permeability were calculated from the data by classical theory. Equilibration rates with all the electrolytes except sodium carbonate were reasonably fast, and reproducible data were obtained with comparative ease. In the case of the carbonate, results were erratic when the electrolyte concentration was decreased, thus indicating the difficulty of removing adsorbed carbonate from the thorium oxide surface. No trouble was experienced when the electrolyte concentration was increased, except at high dilutions. (AEC Activity 5310)

Inorganic and Physical Chemistry: Inorganic Solution Chemistry. — Activity coefficients of silicotungstic acid, a 4-1 polymeric electrolyte, in aqueous solution were measured by equilibrium ultracentrifugation and were found to be in agreement with the Debye-Hückel theory, including an ionic distance of closest approach factor. Light-scattering measurements on the same two-component system could be correlated with the ultracentrifugation results. These observations are of particular interest because they support the thermodynamic aspects of light scattering of electrolytes and make the postulation of an "ordering" effect, proposed by some for these solutions on the basis of turbidity data, unnecessary.

Density measurements of anion exchangers of various degrees of cross-linking and of model electrolyte solutions were carried out in two-component and multicomponent systems. The apparent molal volumes computed from these measurements appear additive, as anticipated from volume measurements on simpler systems, such as alkali halide solutions.

Equilibrium constants were determined up to 200°C for the following reactions, based on solubility data for Ag_2SO_4 in UO_2SO_4 aqueous solutions:

$$UO_2^{++} + SO_4^{--} = UO_2SO_4$$

$$UO_2SO_4 + SO_4^{--} = UO_2(SO_4)_2^{--}$$

$$2UO_2^{++} + H_2O = U_2O_5^{++} + 2H^+$$

By the use of these constants and their extrapolated values, and T. F. Young's relation for the bisulfate acid constant vs temperature, the concentrations of UO_2^{++}, UO_2SO_4, $UO_2(SO_4)_2^{--}$, $U_2O_5^{++}$, H^+, HSO_4^-, and SO_4^{--} were calculated up to 225°C in solutions containing various amounts of UO_2SO_4, H_2SO_4, alkali sulfates, and di-divalent sulfates. (AEC Activity 5330)

Organic Chemistry. — In connection with a mass-spectrometric study of ion-molecule reactions in formic acid vapor, the ratio of the ionization constants of formic acid and formic-d acid (DCOOH) in aqueous solution was measured. The ratio found at 24.5°C was $K_a(H)/K_a(D) = 1.06 \pm 0.03$ (95% confidence interval). This result fails to confirm a suggestion published by Halevi and Nussim in 1956 that the effect of deuterium on the ionization constant of phenylacetic acid, which is of similar magnitude in spite of the larger distance of the deuterium from the carboxyl group, is exerted mostly through an inductive mechanism.

In connection with the solvent extraction program the reactions of tributyl phosphate with nitrates were studied by an adaptation of differential thermal analysis. With tributyl phosphate as the solvent, exothermic reactions were observed in homogeneous phase for nitric acid at 147°C, for uranyl nitrate, thorium nitrate, and ferric nitrate at 160 to 165°C, for aluminum nitrate at 185°C, and for lithium nitrate at 197°C. (AEC Activity 5330)

Radiation Chemistry. — The marked effect of gamma rays upon the catalytic activity of silica gel for hydrogen-deuterium exchange was reported previously. Further experiments showed that the enhanced activity could be produced by 50-kev x rays and even by ultraviolet light. A radiation-produced strong chemisorption of hydrogen was shown to involve the color centers produced by irradiation, but the catalysis is probably not related to these centers in a simple way, since ultraviolet irradiation bleaches the color and decreases the chemisorption while increasing the catalytic activity. (AEC Activity 5330)

Chemical Physics: Molecular Beam Studies. — Molecular association equilibria of seven alkali halides in the gas phase were measured by a direct method [J. Phys. Chem. 63, 938 (1959)] in order to resolve the rather severe discrepancies that have existed between measurements (and calculations) by different groups of investigators. The temperature dependence of the equilibrium constants for dimer dissociation yielded dissociation energies (evaluated at 1300°K) ranging from 48.0 kcal/mole for sodium chloride to 34.7 for cesium chloride, and dissociation entropies

varying from 28.3 e.u. for sodium chloride to 25.0 for potassium iodide. A statistical calculation of the entropy changes based on an ionic model was found to agree well with the experimental values.

A limitation to the sensitivity of the surface ionization gage, used for measuring molecular beams of substances of low ionization potential, is the background of positive ion emission characteristic of tungsten, the usual filament material. It was found that the background consists of bursts of 10^5 to 10^7 alkali metal ions, with rise times in the region of 1 μsec. The frequency of bursts is higher in the presence of oxygen (even at 10^{-6} mm pressure), and is also increased temporarily by stress, introduced either mechanically or by abruptly raising or lowering the temperature of the filament. These characteristics were explained by assuming that alkali impurities concentrate at dislocations and are released rapidly when some stress causes the dislocation to move to the surface. (AEC Activity 5330)

Chemical Physics: Neutron Diffraction. — Potassium acid chloromaleate was found by x-ray diffraction to be orthorhombic, with a unit cell of a = 15.72 Å, b = 10.92 Å, and c = 7.68 Å and the space group D_{2h}^{14} - Pbcn. Work is continuing to determine the complete structure.

The neutron diffractometer installed at the ORR is in working order. The wavelength resolution of this instrument is superior to that of the old one at the Graphite Reactor by a factor of approximately 4, and the intensity by a factor of approximately 10. Programing equipment for automatic data handling is being designed. (AEC Activity 5330)

Chemical Physics: Microwave and Radio-Frequency Spectroscopy. — α,α-Diphenyl-β-picrylhydrazyl, with and without N^{15}, contained in single crystals of the corresponding hydrazine was studied by the paramagnetic resonance method. All resolved hyperfine effects arise from the two hydrazyl nitrogen atoms. The tensors describing the hyperfine interaction for each nitrogen and the directions of the principal axes were deduced. The tensors were interpreted on the basis of s and p orbitals with the following electron densities: for the α nitrogen $a_s^2 = 0.011$ and $a_p^2 = 0.263$, with the two parts having the same sign of spin density; for the β nitrogen $a_s^2 = 0.024$ and $a_p^2 = 0.396$ or $a_s^2 = 0.010$ and $a_p^2 = 0.605$. The two choices for the β nitrogen arise from an ambiguity in interpreting the hyperfine tensor, but with either choice the two parts have the same sign of spin density. The total unpaired electron spin density found on the two nitrogen atoms is 0.694 or 0.889, depending on the choice made for the β nitrogen. (AEC Activity 5330)

High-Temperature and Structural Chemistry. — The parameters of the unit cell of bismuth subchloroaluminate, $Bi_x(AlCl_4)_x$, were found to be a = 12.12 Å and α = 58°23", with the probable rhombohedral space group C_{3v}^6 - R3c. Work is continuing on this structure, which is of interest in the current study of liquid and solid subhalides.

The solubility of lithium metal in molten lithium chloride and in molten lithium iodide from about 650 to 950°C, which was found to be in the range 0.5 to 2.5 mole %, is, as expected, smaller than that of sodium in its halides. (AEC Activity 5330)

Separation of Oxygen Isotopes. – A facility to produce 50% O^{17} and 98% O^{18} is to be constructed, and will consist of two interconnected units, a water distillation cascade and a thermal diffusion cascade. Oxygen gas will be employed in the latter unit. The status of the water distillation cascade is as follows: engineering design, 92% complete; procurement, 45% complete; fabrication, 54% complete. The engineering design of the thermal diffusion unit is 86% complete; procurement is 78% complete. A contract was completed with the H. K. Ferguson Construction Company for the applicable construction phases of the project. (AEC Activity 5340)

Transuranic Studies. – Tests to determine the feasibility of separating rare earths and actinide elements by solvent extraction were not encouraging. The distribution coefficients for curium between 0.5 M mono-2-ethylhexylphosphoric acid in xylene and 10 M LiCl containing from 0.1 to 0.8 M HCl were nearly identical to those for lanthanum. At higher acidities, curium was slightly less extractable than lanthanum; however, a separation factor of only 1.25 was obtained with 10 M LiCl containing 3 M HCl. Higher acid concentrations cannot be used because this is the limit of HCl solubility under these conditions. (AEC Activity 5360)

Analytical Chemistry Research. – A method was developed for the determination of trace amounts of lithium in high-fired, high-density beryllium oxide. After dissolution of the sample in a mixture of hydrofluoric and sulfuric acids, the sulfate solution is neutralized and made 1 M with respect to potassium hydroxide, following which the lithium is extracted with a 0.1 M solution of dipivaloylmethane in ethyl ether. Prior to the addition of potassium hydroxide, potassium fluoride is added to complex the beryllium, thereby preventing its extraction or its precipitation as the hydrous oxide. Lithium is then re-extracted from the ether phase with 1 M HCl, after which it is determined colorimetrically with Thoron as the chromogenic reagent. This method is suitable for the estimation of from 1 to 10 ppm of lithium in beryllium oxide. Within this concentration range the coefficient of variation is 10%. Lithium can also be determined flame photometrically in the acid backwash solution.

METALLURGY AND SOLID STATE PHYSICS

Microstrains in Crystals. – It is a common practice to study thin films after they have been stripped from the metal surface on which they were grown. In order to determine whether the stripping process significantly changes the structure of the film, the x-ray diffraction line profiles from such a film were measured before and after stripping. The measurements indicate that, after stripping, the lattice constants are more nearly those of bulk oxide. The diffraction maxima are also significantly sharper. A quantitative study of the line profiles is in progress to determine whether the sharper lines correspond to an increase in the film thickness. (AEC Activity 5420)

Fundamental Physicometallurgical Research. – Recent studies of the axial ratio of the hexagonal form of zirconium with B-subgroup solutes show that the ratio depends more strongly on zirconium content than on the number of

s-p electrons associated with the valency of each solute. This is apparently a result associated with the discontinuous nature of the incomplete d band in these alloys which, occurring only with the zirconium atoms of the alloy, is diluted when the pure metal is alloyed.

A negative deviation from the ideal entropy of formation in the alpha and beta phases of Cd-Zr has been observed by vapor pressure measurements. In searching for the origin of this deviation, a correlation was noted between the sign and magnitude of the entropy deviation and the heat of formation in a large number of disordered solid and liquid alloy systems reported in the literature. The rule may be formulated that only systems with very small heats of formation have ideal entropy and that negative deviations from ideal entropy, such as occur in Cd-Zr, are usually accompanied by negative heats of formation. Since stronger chemical binding energy is related closely with increased elastic constants, one implication is that the origin of the entropy deviation is thermal, arising in the long-wavelength part of the atomic vibrational spectrum, where increased elastic properties would reduce the specific heat and give negative entropy deviations. (AEC Activity 5420)

Ceramics Research. – Work is in progress on three methods for growing nonmetal single crystals: (1) Hydrothermal – two autoclave and furnace systems are in operation on an exploratory program. Quartz (SiO_2) has been grown, and attention is now directed toward other oxides. (2) Flame fusion – a flame fusion (Verneuil) burner has been completed, and small boules of corundum (Al_2O_3) and bunsenite (NiO) have been grown with an oxyhydrogen flame. (3) Fused salts – schemes for growing oxide crystals from fused halide salts are being explored. Zirconium oxide crystals up to 1 mm on an edge have been grown by the hydrolysis of zirconium fluoride in $NaF-LiF-ZrF_4$ melts. Magnesium oxide crystals up to 0.5 mm on an edge have been grown by the hydrolysis of magnesium chloride melts.

A calorimeter has been designed and constructed for the measurement of the energy stored in metals and alloys during deformation. The operation of the instrument depends on the boiling of a refrigerant such as liquid nitrogen. It may be operated from room temperature to -190°C. The simultaneous measurement of mechanical properties will be accomplished by the operation of the calorimeter in a mechanical testing machine. (AEC Activity 5420)

THERMONUCLEAR PROJECT

DCX-1 and Arc Research. – Pressure characteristics, breakup efficiencies, and containment times have been measured for several gas arcs. The arcs investigated were of the PIG type (with D_2, Ar, He, and N_2 gas feeds) and the d-c type (with D_2 and N_2 gas feeds). The gas flows ranged from 2 to 4 cc/sec (atmospheric pressure), the resulting liner pressures from 2 to 6.5×10^{-5} mm Hg, and the tank pressures from 2×10^{-4} mm to more than 1 μ. The breakup efficiencies ranged from 0.6% for a 2.6-cc/sec deuterium PIG arc (50 amp at 400 v) to about 8.5% for a 2.5-cc/sec argon PIG arc (74 amp at 290 v). The liner pressures were so high that the measured decay times were essentially completely determined by the gas pressure.

Measurements on a carbon arc operated with a tungsten cathode indicated a breakup efficiency of 4% for a 110-amp, 450-v arc.

Similar runs have been made using the deuterium arc with a newly developed baffle. More complicated baffle arrangements resulted in considerably lower liner pressures. For example, a 4-cc/sec (atmospheric pressure), 200-amp, 160-v arc gave a liner pressure of 7×10^{-6} mm Hg and a tank pressure of 4×10^{-4} mm Hg. This same arc had a measured breakup efficiency of about 2%. The containment times for these arcs have not yet been fully assessed, but it is believed that they show no significant improvement over the usual carbon arcs.

It is believed that the measured breakup efficiencies are useful for comparing the performance of the various gas arcs, even though in each case there is the possibility of heavy ion contamination from, for example, carbon and various metals utilized in the electrode and baffle structures. (AEC Activity 5540)

Magnetic Studies and Larger Magnetic Facility. — Supporting work for special projects included a continuation of the DCX-2 magnetic field calculations and computation of coil heat transfer data and mechanical forces. Besides other magnet systems, mirror and booster coils have been designed to produce a very homogeneous field of 7.5 kilogauss in the midplane of a 24-in. tank facility, with a maximum field of 30 kilogauss.

Since the existing empirical relations are not satisfactory when applied to actual coil design, hydraulic and heat transfer studies on hollow copper conductors have been continued. In order to achieve high current densities with moderate water pressure, designs with multiple parallel water paths have been developed.

Mathematical work included magnetic field synthesis and the application of zonal harmonics to rotationally symmetric magnetic fields. Simple mechanical devices for designing ion trajectories were completed. (AEC Activity 5540)

Vacuum. — The 32-in. Consolidated Electrodynamics Corporation diffusion pump has thus far attained only half its rated pumping speed and back-streams oil at an intolerable rate.

Adaption of the metal vapor pumping technique to the intermediate vacuum chamber of DCX is in progress. Of three possible methods of titanium vaporization, (1) from a hot filament, (2) electron bombardment, and (3) titanium arc, the titanium-wrapped tantalum filament is the most practical. The evaporation rate is sufficient, and it is not affected by the magnetic field.

A Consolidated mass spectrograph is in operation at pressures of 2×10^{-7} mm Hg and scans from mass 48 down in about 4 min. Adaption to DCX to analyze the background gas is in progress. (AEC Activity 5540)

Chemistry. — Hydrogen pumping speed measurements on a molecular sieve type material were made. The efficiency of pumping appeared to be impaired because of the presence of other gases in the system. Modifications of a test assembly are being made in order to minimize or eliminate this factor. (AEC Activity 5540)

PROGRAM 6000 — BIOLOGY AND MEDICINE

BIOPHYSICS

Waste Disposal Research and Engineering: Evaluation of Soil Disposal. — The use of vermiculite-filled columns for the decontamination of the ORNL intermediate-level waste stream appears promising. The vermiculite has a high selectivity for cesium over sodium, its hydraulic properties are excellent, and it is readily available in large quantities at a low price, so that abandonment of a column after exhaustion would be feasible.

Five columns of vermiculite (1 in. in inside diameter, 20 cm long, and containing 100 g of vermiculite) were run at different flow rates ranging from 3.9 to 55 ml/min. The solution had the same stable-element composition as the present intermediate-level waste stream and contained the mass equivalent of ~ 2 μc of cesium per milliliter. The slope of the breakthrough curve, as expressed by the number of theoretical plates, did not change with changing flow rate. The breakthrough curves obtained were of the type predicted for a process controlled by particle diffusion. Previous studies on the mechanism of cesium sorption by vermiculite had also indicated that particle diffusion is of importance.

The partial cesium capacity of the vermiculite is strongly dependent on the time of contact between the solution and the particle. Data obtained from column studies were in excellent agreement with data previously obtained by slurry techniques. The time dependency of the cesium sorption reaction, which would hinder efficient column operation, is offset by the fact that the selectivity for cesium over sodium increases with decreasing cesium concentration. The net effect on the breakthrough curve is to make the number of effective theoretical plates high at low breakthrough (below ~ 30% breakthrough), with the number of effective theoretical plates decreasing as breakthrough increases. (AEC Activity 6440)

Ecological Studies. — Radioassay of wild passerine birds living on White Oak Lake bed revealed a striking seasonal difference in fission product uptake which illustrated two routes by which body burdens are accumulated in nature. Uptake by birds in summer was largely through the food chain (fruit, insects) with the result that the burden of Sr^{90} in bone was greater than that of Cs^{137} in muscle, even though the concentration of Cs^{137} in the soil was 25 to 75 times greater than that of Sr^{90}. In winter, birds ingested soil while probing for seeds with the result that the body burden of Cs^{137} was much greater than that of Sr^{90}.

Foliage is an important site of entry of radioisotopes from the atmosphere into the biosphere and provides a convenient natural collector for monitoring fallout. Analyses of leaves by gamma spectrometry showed that I^{131} contributed the most radioactivity to foliage of trees and forest ecosystems for several square miles around ORNL (up to 583 μμc per gram dry weight of dogwood leaves in the northeastern part of the Laboratory grounds). In late September 1959, I^{131} was detectable on dogwood 8 miles to the northeast (5 μμc/g) but only 4 miles to the southeast (1 μμc/g). These and other features of the spatial distribution generally fit the pattern that would be predicted from meteorological records of wind direction, frequency,

and velocity. However, some features suggest that releases occurred during brief periods when winds were not representative of the whole week or month before collection.

Preliminary estimates indicate that Ru^{106}, Cs^{137}, Ce^{144}, Zr^{95}-Nb^{95}, and probably Co^{60} locally exceeded the levels that would be expected from weapons fallout. However, immediately outside the AEC Controlled Area the level of contamination was reduced to that expected from weapons fallout. (AEC Activity 6480)

OPEN LITERATURE PUBLICATIONS

Agron, P. A., and R. D. Ellison, "A Structure Proposal for $Na_7Zr_6F_{31}$," J. Phys. Chem. 63, 2076 (1959).

Bredig, M. A., and H. R. Bronstein, "Miscibility of Liquid Metals with Salts. IV. The Sodium-Sodium Halide Systems at High Temperature," J. Phys. Chem. 64, 64 (1960).

Bredig, M. A., H. A. Levy, F. J. Keneshea, Jr., and D. Cubicciotti, "The Volume of Dimeric Bismuth Monohalide Dissolved in Molten Bismuth Trihalide," J. Phys. Chem. 64, 191 (1960).

Brolley, J. E., Jr., and J. L. Fowler, "Monoenergetic Neutron Sources: Reactions with Light Nuclei," chap. I.C in Fast Neutron Physics, Part I: Techniques (ed. by J. B. Marion and J. L. Fowler), Interscience, New York, 1960.

Cathcart, J. V., and G. P. Smith, "Oxidation Rates of K and Rb Between -79°C and -20°C," J. Electrochem. Soc. 107, 141 (1960).

Curtis, O. L., Jr., and J. W. Cleland, "Mono-energetic Neutron Irradiation of Germanium," J. Appl. Phys. 31, 423 (1960).

Fulmer, C. B., "Total Reaction and Elastic Scattering Cross Sections for 22.8-Mev Protons on Uranium Isotopes," Phys. Rev. 116, 418 (1959).

Gibbons, J. H., and H. W. Newson, "The $Li^7(p,n)Be^7$ Reaction," chap. I.E in Fast Neutron Physics, Part I: Techniques (ed. by J. B. Marion and J. L. Fowler), Interscience, New York, 1960.

Grindell, A. G., W. F. Boudreau, and H. W. Savage, "Development of Centrifugal Pumps for Operation with Liquid Metals and Molten Salts at 1100-1500°F," Nuclear Sci. and Eng. 7, 83 (1960).

Higgins, C. E., and W. H. Baldwin, "Effect of Centrifugation on Solution Temperature and Solubility of Tributyl Phosphate and Tributyl Phosphine Oxide in Water," Anal. Chem. 32, 236 (1960).

Higgins, C. E., and W. H. Baldwin, "Refractometric Determination of Mutual Solubility as a Function of Temperature. Tributyl Phosphine Oxide and Water," Anal. Chem. 32, 233 (1960).

Johnson, C. H., "Recoil Telescope Detectors," chap. II.C in Fast Neutron Physics, Part I: Techniques (ed. by J. B. Marion and J. L. Fowler), Interscience, New York, 1960.

Ketelle, B. H., and A. R. Brosi, "Radioactive Decay of Dy^{159}," Phys. Rev. 116, 98 (1959).

Lamphere, R. W., "Fission Detectors," chap. III.E in Fast Neutron Physics, Part I: Techniques (ed. by J. B. Marion and J. L. Fowler), Interscience, New York, 1960.

Lee, D. A., "The Enrichment of Lithium Isotopes by Ion Exchange Chromatography. II. The Influence of Temperature on the Separation Factor," J. Phys. Chem. 64, 187 (1960).

Lind, S. C., letter to the editor, Chem. Eng. News 38(5), 8 (1960).

Livingston, R., "Chemical Applications of Pure Quadrupole Spectroscopy," Record Chem. Progr. (Kresge-Hooker Sci. Lib.) 20, 173 (1959).

Livingston, R. S., and F. T. Howard (compiled and edited by), "The Oak Ridge Relativistic Isochronous Cyclotron. Part I. Development of the Isochronous Cyclotron," Nuclear Instr. and Methods 6, 1 (1959).

McHargue, C. J., L. K. Jetter, and J. C. Ogle, "Preferred Orientation in Extruded Aluminum Rod," Trans. Met. Soc. AIME 215, 831 (1959).

Melton, C. E., and P. S. Rudolph, "Negative Ion Mass Spectra of Hydrocarbons and Alcohols," J. Chem. Phys. 31, 1485 (1959).

Neiler, J. H., and W. M. Good, "Time-of-Flight Techniques," chap. IV.A in Fast Neutron Physics, Part I: Techniques (ed. by J. B. Marion and J. L. Fowler), Interscience, New York, 1960.

Nelson, F., "Anion Exchange Studies. XXVIII. Comparison of Diffusion in Anion Exchange Resins and Quaternary Amine Solutions," J. Polymer Sci. 40, 563 (1959).

Nelson, F., R. M. Rush, and K. A. Kraus, "Anion-Exchange Studies. XXVII. Adsorbability of a Number of Elements in HCl-HF Solutions," J. Am. Chem. Soc. 82, 339 (1960).

Stoughton, R. W., and M. H. Lietzke, "The Solubility of Silver Sulfate in Electrolyte Solutions. Part 6. Heats and Entropies of Solution vs. Temperature. Species Present in HNO_3 and H_2SO_4 Media," J. Phys. Chem. 64, 133 (1960).

Thurber, W. C., and C. J. McHargue, "Deformation Textures in Aluminum-Uranium Alloys," Trans. Met. Soc. AIME 218, 141 (1960).

Young, F. W., Jr., and A. T. Gwathmey, "Development of Facets, Spirals, and Etchpits on Copper Crystals by Heating to High Temperatures in High Vacua," J. Appl. Phys. 31, 225 (1960).

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Mitchell, R. L.	McCauley Institute for Soil Research	Great Britain
Miyata, K.	Japanese N. S. S. T.	Japan
Moat, A.	United Kingdom Atomic Energy Authority	United Kingdom

Name	Affiliation	Citizen of
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OAK RIDGE NATIONAL LABORATORY

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