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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

FEBRUARY 1963

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OAK RIDGE NATIONAL LABORATORY

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Oak Ridge, Tennessee
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CONTENTS

REACTOR DEVELOPMENT PROGRAM..... 1
GAS-COOLED REACTOR PROGRAM..... 1
EGCR Fission-Gas-Release Activity..... 1
Fast-Neutron Exposure of EGCR Graphite..... 1
Analysis of Irradiated EGCR Prototype Fuel Elements..... 2
Operation of EGCR with Advanced Fuel Elements..... 2
GCR-ORR Loop No. 1..... 2
GCR-ORR Loop No. 2..... 3
LITR Capsule Irradiations..... 3
ORR Capsule Irradiations..... 3
Advanced Gas-Cooled Reactors with Clad Fuel Elements..... 3
Fabrication Studies of Fueled Graphite..... 4
Advanced Fuel-Element Mass-Transfer and Pressure-Drop Experiment..... 4
Convergence of Transport Theory Calculations..... 4
Large One-Region Reactors..... 4
AVR Fuel Loading..... 5
FUNDAMENTAL HEAT TRANSFER AND FLUID DYNAMICS..... 5
Reynolds Stress Measurements..... 5
Transient Heat Transfer..... 5
Boiling-Potassium Studies..... 6
Boiling-Water Studies..... 6
POWER REACTOR FUEL PROCESSING..... 7
Shear-Leach Process..... 7
Processing of Breeder Reactor Fuels..... 7
Hydrochlorination of Uranium-Aluminum Alloy Fuel..... 8
Chlorination of Uranium Carbide—Thorium Carbide—Graphite Fuels..... 8
Solvent Extraction Flowsheet Development for UC Fuel..... 8
Protactinium Adsorption..... 9
Chemical Analysis of Advanced Reactor Fuels..... 10
THORIUM UTILIZATION PROGRAM..... 10
Thorium Materials Development..... 10
Fuel Cycle Development: Chemistry of Solid Fuel Element Materials..... 10
Fuel Cycle Development: Fuel Preparation..... 11
Fuel Cycle Development: Fuel Fabrication Studies..... 11
MOLTEN-SALT REACTOR PROGRAM..... 12
Molten-Salt Reactor Experiment..... 12
Reactor Analysis..... 12
Development of Components and Systems..... 12
MSRE Pump Program..... 13
Instrumentation Development..... 13
NUCLEAR SAFETY..... 14
Release of Fission Products on Out-of-Pile Melting of Reactor Fuels..... 14

[redacted]

[redacted] [redacted]

Release of Fission Products on In-Pile Melting of Reactor Fuels 15
Characterization and Control of Accident-Released Fission Products 16
Nuclear Safety Pilot Plant 16
Reactor Containment Handbook 16
WASTE DISPOSAL DEVELOPMENT 16
Clinch River Study 16
Mineral Exchange Studies 17
Disposal in Deep Wells 17
High-Activity Waste Calcination 17
Low-Activity Wastes 18
Foam Separation Studies 18
Disposal in Natural Salt Formations 19
Engineering, Economic, and Safety Evaluation 19

PHYSICAL RESEARCH PROGRAM 20
PHYSICS AND MATHEMATICS RESEARCH 20
High-Energy Electronuclear Machines 20
Medium-Energy Nuclear Physics: The 86-Inch Cyclotron 20
High-Voltage Experimental Program: Nuclear Resonance Effects in Coulomb Excitation 20
High-Voltage Experimental Program: Resonant Neutron Capture Spectra in $Fe^{54,56}$ and $Ca^{40,42,44}$ 21
Electronuclear Machines: The Isochronous Cyclotron (ORIC) 21
Stable Isotopes Development 22
Special Separations 22
CHEMISTRY RESEARCH 23
Protactinium Chemistry 23
Pyrolysis and Radiolysis of Organic Liquids 23
Analysis of Molten Salts by Electrochemical Methods 23
Radiation Chemistry: Radiation-Induced Surface Oxidation of Hydrocarbons 23
Radiation Chemistry: Retention of Br^{82} Recoils by Crystalline Alkali-Metal and Alkaline-Earth Bromates 24
High-Temperature and Structural Chemistry 25
Corrosion Tests for the TRU Facility 25
Target Preparation 25
Transuranium Element Processing 26
Analytical Chemistry Development 26
Corrosion Studies 26
Design of Experimental Facilities 26
HFIR Target Development 27
Chemical Process Development 27
Process Equipment Development 28
Transuranium Facility 28
METALLURGY AND MATERIALS RESEARCH 29
Preparation of Pure Materials 29

[redacted] [redacted]

[redacted]

Spectroscopy of Ionic Media..... 29
Fundamental Investigation of Radiation Effects in Solids: Calculations of Some Ground-State Properties of F Centers..... 29
Fundamental Investigation of Radiation Effects in Solids: Electron Structure of the M Center..... 30
CONTROLLED THERMONUCLEAR RESEARCH..... 30
Plasma Physics and Arc Research..... 30
DCX-1 Facility..... 30
DCX-2 Facility..... 31
Vacuum..... 32

BIOLOGY AND MEDICINE PROGRAM..... 32
MOLECULAR- AND CELLULAR-LEVEL STUDIES..... 32
Physics of Tissue Damage..... 32
TERRESTRIAL AND FRESH-WATER ECOLOGY..... 33
Evaluation of Fission Product Distribution and Movement in White Oak Drainage System..... 33
Glass-Rod Dosimetry in Environmental Studies..... 33
Seasonal Changes in the Blood of Rodents on Radioactive and Nonradioactive Areas..... 33
Removal of Sr^{90} from the Clinch River by Emergent Insects..... 33
Removal of Sr^{90} from the Tennessee River in Clam Shells..... 34
Soil Microbiological Studies on Oak Ridge Reservation..... 34
Transfer of Co^{60} and Ru^{106} from Oak and Ash Leaves to Several Forest Soils..... 34
RADIOLOGICAL PHYSICS, HEALTH PHYSICS, AND INSTRUMENTATION.... 35
Radiation Physics and Dosimetry: Time-of-Flight Electron Beam Monochromator..... 35
Radiation Physics and Dosimetry: Electron Slowing-Down Spectra in Copper..... 35
Radiation Physics and Dosimetry: Measurement of Spectral Distribution of Positron Flux in an Infinite Copper Medium Containing Cu^{64}..... 36
Radiation Physics and Dosimetry: Investigation of Collective Effects in the Absorption of Radiation in Matter..... 36

ISOTOPE DEVELOPMENT PROGRAM..... 37
Nuclear Analysis Research and Development: Use of a 14-Mev Neutron Generator..... 37
Nuclear Analysis Research and Development: Individual Characterization by Hair Analysis..... 37
Nuclear Analysis Research and Development: Enforcement of the Narcotics Laws..... 37
Curium-242 Preparation..... 37
Radioisotope Research and Development..... 37
Radioisotope and Stable Isotope Production..... 38

OPEN LITERATURE PUBLICATIONS..... 39
VISITORS..... 41

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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

FEBRUARY 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 Fission-Gas-Release Activity. — Calculations to update the predicted fission product activity release from the EGCR fuel pellets were completed. Based on the latest in-pile data on the thermal conductivity of UO_2 and the results of fission-gas-release tests on actual EGCR fuel pellets, the expected equilibrium activity release from the UO_2 in the maximum-power element is about 0.5 curie, and that from the entire core is about 380 curies. These figures represent a reduction by a factor of about 200 in the initial activity estimates. The UO_2 tested was virgin fine-grain-structure material.

Fast-Neutron Exposure of EGCR Graphite. — The calculations of graphite exposure in the EGCR were reviewed to take into account the more recently obtained conversion factor of 1 Mwd/adjacent ton = 1.27×10^{17} neutrons/cm^2 for neutron energies greater than 0.18 Mev. The use of this more accurate conversion factor gave a reduction by a factor of 1.46 in the graphite exposure. The accumulated 20-year exposure in a core column at the average axial position for an average power channel will be 42,900 Mwd/adjacent ton, and the peak exposure accumulated in the core over this period will be ~93,000 Mwd/adjacent ton. Both values are based on 20 years of continuous full-power operation, and, for a duty factor of ~0.8, the maximum anticipated exposure is ~75,000 Mwd/adjacent ton.

The following correlation between exposure and temperature was made, based on published values for the axial temperature variation and 20 years of continuous full-power operation:

[redacted]

Graphite Temperature (°F)	Twenty-Year Graphite Exposure (Mwd/adjacent ton)
510	7,000
550	64,000
600	84,000
680	93,000
750	87,000
800	76,000
900	37,000
1000	6,000

Analysis of Irradiated EGCR Prototype Fuel Elements. – Data obtained for four irradiated capsules containing EGCR prototype fuel were analyzed with the PANDORA code. The measured and calculated central temperatures are compared below:

Capsule	Heat Rating (Btu hr^{-1} ft^{-1})	Measured Cladding Temperature (°F)	Measured Central Temperature (°F)	Calculated Central Temperature (°F)	Calculated Fission-Gas Release (%)
01A-3	24,000	1429	1750	2118	0.19
01C-4	30,000	1480	2080	2366	0.45
08B-4	30,000	1456	2320	2342	0.44
08-3	14,900	1300	1750	1678	0.06

The measured central temperature for capsule 01A-3 was known to be low. In view of the uncertainties associated with temperature measurements in in-pile capsules, the agreement between the measured and calculated central temperatures is felt to be excellent.

Operation of EGCR with Advanced Fuel Elements. – A series of calculations was undertaken to examine the operational problems that might result if advanced fuel elements were installed in certain individual channels in the EGCR in place of the regular fuel elements. It seems that advanced elements with about 5% enriched uranium could be substituted for the regular elements (2.46% enrichment) in four symmetrically located channels without introducing significant control problems.

GCR-ORR Loop No. 1. – GCR-ORR loop No. 1 is operating satisfactorily with a fuel element that has a roughened surface. There is no evidence of deterioration of the element, except the loss of one thermocouple for measuring the cladding temperature. Performance measurements for this element continue to indicate a significant improvement over previous elements having smooth surfaces.

GCR-ORR Loop No. 2. – The in-pile portion of GCR-ORR loop No. 2 was installed during the January ORR shutdown and has been operating smoothly during flux-plotting experiments. The maximum flux, as determined by argon activation, is 5.7×10^{12} neutrons cm^{-2} sec^{-1}. High-temperature operational tests with electrical heat were started late in the month. Construction of the fuel-holder assembly that provides for testing of four fueled-graphite balls 1-1/2 in. in diameter is under way. The elements will be ready for insertion in the loop during the May shutdown of the ORR, as scheduled.

LITR Capsule Irradiations. – The fourth capsule in the high-temperature UO_2-irradiation test series continues to operate satisfactorily after approximately five weeks of a scheduled three-month irradiation. The present measured operating temperature is 3360°F.

ORR Capsule Irradiations. – Three of the group VI capsules containing vendor-made EGCR fuel were removed for hot-cell examination, and a second experimental assembly containing fueled-graphite balls was installed. This assembly contains four 1-1/2-in.-diam balls consisting of coated UC_2 particles uniformly distributed in a graphite matrix. There is no unfueled shell on these balls, which were made by the National Carbon Company and General Atomic. There has been no detectable fission-gas release from these fuel elements during the first few days of irradiation. Fractional fission-gas-release rates for the fueled-graphite balls having cylindrical inserts containing columnar-type pyrolytic-carbon-coated UC_2 particles increased from 1×10^{-5} to 5×10^{-5} during the past month.

Advanced Gas-Cooled Reactors with Clad Fuel Elements. – The steam-power cycle proposed in the GCR-3 advanced gas-cooled reactor design study employs regenerative feedwater heating to obtain a feedwater temperature of 450°F. Lower feedwater temperatures would result in lower pumping power requirements in the primary-coolant-gas system. A study of the effects of varying the number of heaters in the feedwater system of the GCR-3 shows that the overall thermodynamic efficiency is maximum for a feedwater temperature of 500°F, with constant system energy. The economic optimum is found to be at a feedwater temperature of approximately 470°F.

A parametric study of combined gas-turbine- and steam-power cycles for high-temperature gas-cooled reactors was completed at Westinghouse Electric Company for the Laboratory. The gas-turbine power is used to drive the compressors circulating gas through the reactor and steam generators and to generate variable amounts of electric power, depending on the temperature of gas at the reactor exit. The reactor-outlet temperature was varied from 1240 to 1500°F with compressor-suction temperatures from 375 to 550°F. The overall efficiency varied from 39.8 to 46.2%, the maximum corresponding to a reactor-outlet temperature of 1475°F and a compressor-suction temperature of 470°F. The turbine-exhaust temperature was held constant at 1150°F to provide an adequate temperature difference for the steam generator.

Fabrication Studies of Fueled Graphite. – Developmental work was initiated on a method for preferentially damaging pyrolytic-carbon-coated fuel particles at or near the surface of fueled-graphite spheres without unfueled shells. Damage of this type and removal of surface-contained uranium and thorium is considered to be necessary to preclude damage to surface particles during service and the subsequent contamination of the coolant and downstream piping.

An attempt to fracture fueled particles at the surface with isostatic pressure was made. The fueled graphite spheres were coated with a silicone encapsulating compound that was heavily loaded with angular Al_2O_3 grit. The coated sphere was isostatically pressed first at 25,000 psi and later at 50,000 psi. Subsequent microscopic examination revealed an almost complete lack of damage to the surface particles. Several variations of this method made it evident that a slow and steady buildup of isostatic pressure would not damage the surface fuel particles in the desired manner. It appears that a method using shock or impact will be needed. To this end a device by which air-driven shot will be used to peen the sphere surface, with the intent of damaging the individual surface fuel particles by impact, is being designed.

Advanced Fuel-Element Mass-Transfer and Pressure-Drop Experiment. – Pressure-loss coefficients were determined in a Reynolds modulus range of 2×10^3 to 2×10^5 with bare rods and with all rods spirally wrapped with 0.006-in.-diam wire on a 0.036-in. pitch (pitch-to-height ratio of 6:1). At low Reynolds moduli, approaching laminar flow, both configurations exhibit similar pressure-drop characteristics, whereas at the higher Reynolds moduli, the roughened surface produces greater loss coefficients. For $N_{Re} > 10^5$, the pressure-loss coefficients of the wire-wrapped surface are approximately twice those of the smooth rod.

Difficulties encountered in installing and removing the cluster from the channel (necessitating appreciable time delays) prevented the collection of satisfactory mass-transfer data for the bare rods. Since allowable exposure times are much less for the wire-wrapped rods (before mass removal causes the wire coil to collapse), it is not anticipated that satisfactory mass-transfer data can be obtained without extensive modification of the apparatus. Accordingly, this phase of the experiment was abandoned.

Convergence of Transport Theory Calculations. – In analyzing certain gas-cooled reactor problems by use of the DSN and TDC transport equation codes, difficulty was experienced in obtaining proper convergence of the solution in a reasonable calculation time. Experimental modifications to these codes were made, and it was found that substituting a form of Liebmann extrapolation for the scale-factoring procedure of the codes markedly accelerated convergence of some two-group problems.

Large One-Region Reactors. – A parametric study is being made of large one-region gas-cooled reactors utilizing unclad fuel. The results indicate an important dependence of fuel-cycle costs on neutron leakage from the core. At a carbon-to-uranium ratio of 2000 and a total fabrication and reprocessing cost of $100 per kilogram of heavy metal, the fuel-cycle cost for 5% leakage was a minimum of 0.33 mill/kwhr (thermal).

In contrast, the minimum cost with 2% leakage was 0.29 mill/kwhr (thermal) with the same unit costs. The optimum power density was found to be 10 w/cm^3 for both 2 and 5% core leakage.

AVR Fuel Loading. – Calculations are under way to determine the fuel composition for the initial AVR (German pebble-bed reactor) core loading. One basic criterion is that there be no reactivity rise should any amount of steam enter the core. In all cases the fueled balls were assumed to contain 5 wt % heavy elements. It was found that the above criterion was met if thorium-to-uranium ratios as high as 7.8 for a hot-clean core and 9.4 for a cold-clean core were used in a core in which one-third of the graphite balls were fueled. If one-fourth of the balls were fueled, the thorium-to-uranium ratio was 5.5 for a hot-clean core.

Fueling schemes using these three compositions are being investigated. One scheme provides for maintaining criticality initially by removing borated balls. When these balls were all removed, fresh balls containing uranium would be added. The initial conversion ratios and the estimated final burnup at the end of two years of operation for the three cases are listed below:

Th-U Ratio	Conversion Ratio	Burnup (Mwd/ton)
5.5	0.143	87,000
7.8	0.197	71,000
9.4	0.207	63,000

FUNDAMENTAL HEAT TRANSFER AND FLUID DYNAMICS

Reynolds Stress Measurements. – Calculation of Reynolds stresses from raw hot-wire data, a process which had previously taken approximately one week per run, was programmed for the IBM 7090 computer. A data run at a Reynolds modulus of 2.2×10^5, the results of which are not yet available, was completed. A second run at a Reynolds modulus of 1.5×10^5 is in progress. The entire loop-anemometer system is functioning properly, with signal drift as low as 2% per hour.

Transient Heat Transfer. – Machining of a new test section was essentially completed. The holes for the temperature and velocity probes were machined and the various probe-positioning subassemblies were fabricated. Tests were run to determine the best surface treatment, flux, brazing alloy, temperature cycle, etc., for the brazing of the Chromalox heaters to the test section. These tests were completed, and the results are being evaluated.

The 200-gpm water deaerating system was installed and leak-checked under pressure. The interior of the tank will be given a rust-inhibiting treatment before the unit is put into operation.

A detailed calibration procedure for the integrated instrumentation system (amplifiers, tape recorder, spectral analyzer) was worked out. The procedure utilizes a precision voltage calibrator (1000 cps), counter-timer (10^6 cps), sine-wave generator, and oscilloscope. This relatively complex system is required to provide the accuracy in amplitude-frequency calibration considered to be necessary for the ultimate application to spectral analysis of turbulence.

Boiling-Potassium Studies. – The copper-jacketed stainless steel high-flux boiler, which was removed from the boiling-potassium heat transfer loop because of the progressive deterioration of the copper-to-stainless steel braze and the failure of a large number of thermocouples, showed no evidence of scale buildup upon visual examination of the inner surface. Metallurgical examination of the brazed regions has not been carried out.

A new high-flux boiler section was assembled and is ready for the furnace-brazing operation. This boiler will provide, in addition to local wall temperatures and heat fluxes, means for determination of fluid temperature at three axial positions within the boiler and for measurement of fluid pressure at the mid-axial position. These additional measurements will permit more accurate correlations of boiling heat-transfer coefficients and two-phase pressure drop. The loop piping, heater location, and instrumentation are being modified slightly to accommodate this new boiler.

A tentative explanation was advanced for the severe pressure and temperature instabilities found with potassium boiling in capsules and in forced- and natural-circulation loops. Based on the analysis of Ellion and the confirming experimental results of Wallis and Griffith, it is estimated that very high wall superheat may be required to initiate potassium boiling. This fact, combined with the high thermal diffusivity of liquid potassium, indicates that, when a vapor bubble is formed at the heated surface, the energy of superheat in a relatively large volume of liquid may be dissipated in such a short time that an "explosive" surge is generated. Further study of this phenomenon is planned.

Boiling-Water Studies. – A series of 127 additional critical-flux (ϕ_{crit}) determinations was made to complete an experimental investigation of inherent uncertainty under conditions of saturated pool boiling. For saturated water at an average pressure of 14.4 psia, the results may be summarized as follows:

Test Section	Number of Tests	ϕ_{crit} (Btu hr^{-1} ft^{-2})	
		Range	Average
		$\times 10^6$	$\times 10^6$
Tube 9	50	0.413–0.498	0.457
Tube 10	54	0.299–0.450	0.353
Tube 11	23	0.201–0.369	0.280

The conclusions previously reached are unchanged. For the complete series of 233 tests, the average liquid-film superheat at the critical heat flux was 40°F. The overall range in ϕ_{crit} of 0.201 to 0.596 × 10^6 Btu hr^{-1} ft^{-2} compares well with the range, obtained from 22 literature references, of 0.200 to 0.651 × 10^6.

The average power-rise rate in these tests immediately preceding attainment of the critical heat flux was ~1.0% of ϕ_{crit} per second. Comparison of two data groups with rise rates ≧1.0%/sec and <1.0%/sec showed the critical fluxes attained at the lower rates to average ~14% higher than those of the other group.

A few supplementary tests were made to determine the effect of liquid level on the saturated pool-boiling critical heat flux for a horizontal, 1/4-in.-OD tube, with the following results:

Liquid Level (in. below top of tube)	ϕ_{crit} (Btu hr^{-1} ft^{-2})
	× 10^6
0.100	0.450
0.125	0.375
0.172	0.275
0.235	0.180
0.250	0.060

A plot of these data indicates that a maximum heat flux of ~125,000 Btu hr^{-1} ft^{-2} can be transferred, even when the water level is barely touching the lower tube surface.

POWER REACTOR FUEL PROCESSING

Shear-Leach Process. — Bench-scale, batch leaching tests are being made with sheared pieces of stainless-steel-jacketed UO_2-ThO_2 fuel to develop a method that ensures good contact between the leachant (nitric acid) and the UO_2-ThO_2 fines produced during shearing. In an initial test, fines were suspended successfully by discharging air through a draft tube; the sheared pieces were leached at a satisfactory rate. Leaching of the UO_2-ThO_2 was 99.95% complete in 14 hr of contact with boiling (117°C) recirculating 12 M HNO_3–0.04 M F–0.10 M Al. The gross recirculation rate in the 18-liter system was about 40 liters/min.

Processing of Breeder Reactor Fuels. — The experimentally determined instantaneous dissolution rate of high-density (85 to 93% of theoretical) UO_2-PuO_2 pellets in nitric acid or in mixtures of nitric and hydrofluoric acids agreed closely with values calculated from the rates for UO_2 pellets

and PuO_2 pellets of the same theoretical density. The addition of oxidizing agents to nitric acid did not significantly increase the dissolution rate of PuO_2 pellets.

Multiple-extraction and Martin-technique experiments indicate that not more than 1% of Pa^{233} tracer is unextractable into 30% tributyl phosphate in dodecane from 0.5 M $Al(NO_3)_3$ containing 2 or 5 N HNO_3 and that the distribution coefficients are about 2 and 6 respectively.

Hydrochlorination of Uranium-Aluminum Alloy Fuel. — Calculations with the GHTR code (using the IBM 7090 computer) to determine maximum temperatures obtainable in the hydrochlorination of U-Al alloy MTR fuel were continued. In these calculations it was assumed that all heat is lost by radiant heat transfer. The maximum steady-state temperature decreased from 1073°C at a hydrochlorination rate of 12 mils/hr to 549°C at 6 mils/hr. At an average rate of 6 mils/hr, the 60-mil inside plates and the 180-mil side plates could be completely hydrochlorinated in 10 and 30 hr respectively. The additional small contribution to heat removal by the heat capacity and convection of the gases was not considered. These results indicate that it will be possible to hydrochlorinate MTR fuel elements at rather low rates without exceeding the melting temperature of the fuel, about 650°C.

Chlorination of Uranium Carbide—Thorium Carbide—Graphite Fuels. — Three 10-g samples of UC-ThC_2-graphite fuel containing 1.5% U and 7% Th were burned in oxygen at 800°C in 2.5 and 3.5 hr. The ash so produced was then chlorinated in 2 hr with 15% CCl_4—Cl_2 at 500 and 550°C for two of the 10-g samples and with phosgene at 500°C for the third. Complete chlorination was achieved in all three cases, and the nonvolatile products were completely soluble in water. The respective amounts of uranium volatilized from the combustion residues during chlorination represented 74, 84, and 64% of those in the residues. This volatilized uranium chloride was collected on the cool exit of the reaction tube.

These experiments indicate that it is possible to process this type of fuel in a nickel vessel, yielding an aqueous product solution containing both the uranium and the thorium. The complete separation of uranium and thorium might be made possible by using higher temperatures in order to completely volatilize the uranium as uranium chloride, but ceramic vessels would have to be used because metals corrode too rapidly.

Solvent Extraction Flowsheet Development for UC Fuel. — Two alternative aqueous head-end procedures are being developed for sheared uranium carbide fuel: (1) hydrolysis with water at 80 to 100°C to form hydrated UO_2, followed by dissolution in nitric acid, or (2) direct dissolution of the UC in nitric acid. Further investigation of both procedures is required to determine whether they will yield a feed solution suitable for processing by conventional solvent extraction methods.

Hydrolysis of 20- to 60-g specimens of impure UC (about 5.1% C), containing some dicarbide, at 90°C for 2 hr followed by a 2-hr dissolution-reflux in 4 M HNO_3 yielded a raw feed solution containing 120 g of uranium per liter in 3 M HNO_3, and waxy, yellow-brown solids. Nearly all the carbon from the UC was hydrolyzed to volatile paraffinic hydrocarbons. The water-washed, air-dried, amorphous organic solids (about 0.3 wt % of

the UC) retained 0.003% U. These solids dissolved readily in acetone, though not in dodecane. Infrared spectroscopy indicated a very complex mixture of organic acids. Further analysis suggested the empirical formula $(C_2H_2O_2 \cdot NO_3)_x$.

During exhaustive extraction of filtered and unfiltered raw feed solutions with 15% tributyl phosphate in Adakane, followed by exhaustive stripping, stable emulsions and interfacial matter were generated and persisted throughout these operations. Uranium recovery, however, was normal: 99.98%.

Pretreatment methods in which the organic acids may be chemically degraded to some form that does not interfere with solvent extraction are being sought. Scouting tests were conducted with unfiltered raw feed solutions treated with 0.1 M $KMnO_4$, $Ce(SO_4)_2$, $Na_2Cr_2O_7$, $NaNO_2$, and $(NH_2OH)Cl$, followed by 1 hr of reflux and then filtration. The filtrates were subjected to exhaustive extraction and stripping. Following the permanganate treatment, no emulsification or interfacial matter was observed during extraction or stripping. Coalescence time was typical for a pure $UO_2(NO_3)_2$–HNO_3–tributyl phosphate system, being 20 sec or less. Uranium extraction and stripping losses were 0.22 and 0.01% respectively. Treatment with the other reagents appeared ineffective, compared with the permanganate treatment, suggesting that only partial degradation of the organic acids was achieved.

Protactinium Adsorption. – Studies on the adsorption of protactinium on unfired Vycor and silica gel were continued. A column of 80- to 100-mesh unfired Vycor, 1 cm in diameter and 8.6 cm long, adsorbed about 7.8 mg of Pa^{231} per gram of glass, or about 85% of the protactinium from 870 ml of a solution that was 0.5 M in thorium, 11.1 N in HNO_3, 0.074 M in aluminum, and about 0.03 M in fluoride. The Pa^{231} concentration was 54 mg/liter. The solution was passed through the column at a flow rate of 0.22 ml cm^{-2} min^{-1}.

During the run, the protactinium concentration in the emerging raffinate rose from 4 to 46% of the initial feed concentration. The 4% loss was constant for the first 400 ml of raffinate and was equivalent to a loading value of about 2 mg of Pa^{231} per gram of glass. The protactinium in this raffinate was not adsorbed during passage through a fresh column of unfired Vycor.

The loaded column was washed with 80 ml of 11 M HNO_3, which removed 13% of the adsorbed protactinium, and then the column was stripped with 0.5 M oxalic acid. The first 5 ml of oxalic acid removed 95% of the remaining protactinium, yielding a solution that contained 6.4 mg of Pa^{231} per milliliter, or about 120 times the concentration of the Pa^{231} in the feed solution. More than 99% of the protactinium had been removed after a total of 30 ml of strip solution had passed through the column.

Chemical Analysis of Advanced Reactor Fuels. — Performance of the application of controlled-potential coulometry to the determination of U(VI) at the microgram level is being studied. A micro coulometric titration vessel designed for the analysis of approximately 1-ml samples and the high-sensitivity, controlled-potential, coulometric titrator are used.

The procedure included (1) prereduction of the mercury at −0.250 v vs the S.C.E., (2) prereduction of the sample at +0.075 v vs the S.C.E. and reduction at −0.250 v vs the S.C.E., and (3) correction of total coulombs consumed for "charging" and "faradaic" backgrounds. Samples were taken by pipeting aliquots of known dilutions of a standard solution containing 12 mg of U(VI) per ml. Results are shown below:

Number of Determinations	U(VI) Taken (μg)	U(VI) Found (μg)	Standard Deviation (μg)
8	62.9	62.6	0.16
14	31.5	31.6	0.14
10	12.6	12.8	0.09
7	2.52	2.72	0.08

At present, standard solutions made to contain about 50 μg of U(VI) per g are being titrated. The necessary aliquots are taken by using a weight buret. This method of preparing the original standard and of taking the sample should give a better estimate of the actual amount of uranium taken than the method used in obtaining the results shown above.

THORIUM UTILIZATION PROGRAM

Thorium Materials Development. — Successful extrusions of hot-pressed compacts of thorium containing 10% ThO_2 as a dispersoid were made. Hot-hardness tests suggest that this material should be significantly stronger, at temperatures up to 500°F, than the best thorium alloys that have been developed. At 600°C, the hardnesses are nearly equal. Creep tests, which are a more accurate indication of resistance to swelling under irradiation, will be conducted.

Dispersion strengthening of thorium-zirconium alloys is being attempted. Co-reduction of thorium-zirconium oxides is favored as more economical than the hydriding technique (ORNL-3377, p 12). The pyrophoricity of the metal powders has been minimized by using surfactants, such as stearic acid and sodium potassium tartrate.

Fuel Cycle Development: Chemistry of Solid Fuel Element Materials. — A preliminary study of phase equilibria in the system UO_2-ThO_2-O_2 was completed, and the results are summarized herein. In the presence of 1 atm of air (0.21 atm oxygen), compositions falling on the boundary line

between the single-phase fluorite solid-solution region and regions containing more than one phase are expressed by the following values of temperature, thorium-uranium percentage ratio, and oxygen-metal atomic ratio: 400°C, 42 Th–58 U, 2.32; 600°C, 40 Th–60 U, 2.32; 800°C, 38 Th–62 U, 2.31; 1000°C, 36 Th–64 U, 2.30; 1200°C, 33 Th–67 U, 2.29. At each of these temperatures, compositions containing a higher percentage of thorium than that listed were found as a single-phase solid solution with an oxygen-metal atomic ratio lower than that listed. Experiments to identify the various phases present at equilibrium in the multiphase regions are continuing.

Fuel Cycle Development: Fuel Preparation. – The sol-gel process was modified to prepare rounded particles of thorium dicarbide and thorium-uranium dicarbide. Sols of thorium oxide alone and sols containing thorium oxide and up to 10 at. % uranium oxide, 2 M in metal, were prepared. High-surface-area carbon (from 4 to 6 g-atoms of carbon per g-atom of uranium plus thorium) was mixed into the sol. The sol was evaporated to a gel, and the gel was fired at 1580°C for uranium-thorium dicarbide and up to 1775°C for thorium dicarbide. Better than 99.8% conversions to dicarbides were obtained, with free-carbon residues of 4 to 7 wt % and unconverted oxide residues equivalent to 215 to 260 ppm oxygen.

Rounded particles were prepared by dispersing the sol in carbon tetrachloride–isopropyl alcohol mixtures, drying gently, then firing. Particles 50 to 250 μ in diameter, which kept their shape during firing, were prepared.

Uranyl nitrate was greater than 99% denitrated by steam at 180 to 250°C in 4 to 6 hr in a rotary calciner, producing α-UO_3 having crystallite sizes of 400 to 600 A and surface areas of 0.5 to 3 m^2/g. The nitrate was recovered entirely as nitric acid. The higher-surface-area UO_3 was reduced in the rotary calciner (in the presence of formic acid and hydrogen in a molar ratio of 2:1 and palladium-impregnated thoria), at 100 to 400°C, to UO_2 that had an average crystallite size of 127 A.

Two 22-in.-long irradiation capsules containing sol-gel-produced ThO_2-UO_2 (5 wt % enriched uranium) were put into the MTR. The capsules are scheduled to be moved to higher fluxes three times to accumulate an average burnup of 105,000 Mwd/metric ton. About 4.5 kg of 5.0 wt % enriched uranium in sol-gel-produced ThO_2-UO_2 was prepared for irradiation specimens in the MTR. An operating procedure was written for the rotary denitrator, and successful routine operation was started.

Fuel Cycle Development: Fuel Fabrication Studies. – Installation of the fabrication equipment in the Kilorod Facility is complete; a final operational check, using cold oxides, will be initiated next month.

Research to try to establish the important parameters in producing high density and uniform fuel distribution by vibratory compaction is continuing. Electrodynamic vibrating equipment is being used to explore the effects of frequency and acceleration on density. The initial experiments seem to indicate that when frequency is held constant, changes in acceleration have little effect. Conversely, when acceleration is held constant, varying the frequency has a marked effect.

Examination of many of the first irradiation experiments, using sol-gel ThO_2-UO_2, is nearing completion. No sintering or other

physical changes in the oxides have been observed. This is to be expected, since the heat ratings of these capsules have been modest and the total burnup has been low. One interesting result is the direct comparison of fission gas released from tests using sol-gel-produced oxides with that from tests using arc-fused oxides. The percent Kr^{85} released was 0.5 for sol-gel and 2.4 for arc-fused material. One reasonable explanation for this difference is the greater percentage of fine particles present after irradiation in the capsules containing arc-fused oxide. It is thought that during vibratory compaction, considerably more breakdown in particle size occurs with the friable arc-fused material than with the tough sol-gel oxide.

MOLTEN-SALT REACTOR PROGRAM

Molten-Salt Reactor Experiment. – A complete inspection of the tube bundle for the MSRE heat exchanger revealed no defects; the tube bundle is being assembled into the shell. Fabrication of the fuel drain tanks continued; one tank is nearing completion. Five of the ten tube banks for the radiator were welded to the main headers.

Installation was started in Building 7503. Stairs and ladders were installed in the drain-tank cell. Scaffolding was installed in the reactor cell. Installation of pipe supports, structural supports, and the thermal shield was started.

Procurement of all MSRE materials and components is approximately 80% complete.

Reactor Analysis. – The 34-group cross-section library, used for MSRE nuclear calculations, was revised and extended. The General Atomic program GAM-1, with its 68-energy-group library, was used to generate resonance and fast cross sections appropriate for core compositions which would be encountered with various types of fuel in the MSRE. Thermal-group cross sections were calculated by using the Wilkins "heavy-gas" model to represent the neutron spectrum in the homogenized graphite fuel mixture. The validity of this model was spot-checked by the THERMOS lattice thermalization program. It was found that in the MSRE core lattice, space and energy distributions could be calculated adequately with a one-group P_3 model and the space-independent heavy-gas model.

Development of Components and Systems. – Following 1500 hr of operation with flush salt and graphite in the engineering test loop, treatment with HF in the drain tank removed 79 ppm oxygen from 240 kg of salt. Chemical analysis of samples taken before this treatment indicated an oxygen content of 240 ppm. Failure of efforts to reduce this apparent discrepancy by improving the sampling or analysis techniques has forced acceptance of the poor resolution in the oxide analyses of the salt.

A total of 350 hr of treatment with HF in the Inconel drain tank of the engineering test loop resulted in a removal of chromium from the wetted surface to an equivalent depth of 1.5 mils. Removal of 70% of the chromium occurred in the first 73 hr of the first treatment, which indicates a desirable pretreatment effect. These removal rates are considered acceptable for these short-term treatments.

Installation of the mockup of the MSRE sampler-enricher into the engineering test loop was completed, and the checkout of the various component systems was begun.

The insulated heater for 5-in. pipe, which had failed by short circuit, was returned to 1350°F service after corrections were made to prevent recurrence of the failure. The heat loss was 5% more than that measured during previous operation – apparently a result of damage associated with the failure. This increase would not be large enough to require replacement of a reactor heater.

The size of the frozen zone was difficult to control in preliminary tests of a mockup of the reactor drain-line freeze valve. This resulted in considerable variation in the time required to open the valve. Additional work is being done to improve the control.

Alterations to the MSRE maintenance mockup were completed preparatory to the testing of the special tools required in the crowded area at the freeze flange in the pipe connecting the reactor and the pump. The special tools are under construction.

Three versions of flexible electrical power leads for use with the electrical disconnects were evaluated, and a selection was made. The originally specified flexible lead was too stiff for use with the electrical disconnects.

MSRE Pump Program. – The Inconel centrifugal pump (PKP type) has accumulated 9860 hr of operation in an endurance test with molten salt $LiF-BeF_2-ThF_4-UF_4$ (65-30-4-1 mole %) at 1225°F, 1950 rpm, and 510 gpm.

The MSRE prototype fuel pump continued in operation with molten salt $LiF-BeF_2-ZrF_4-ThF_4-UF_4$ (70-23-5-1-1) at 1200°F, 1200 rpm, and 1200 gpm. After 2150 hr of operation, the test was interrupted by a short circuit that developed in the motor-generator power supply for the pump motor. The test was resumed with the pump motor supplied directly from a 60-cps source. The pump has since operated for 650 hr.

Back-diffusion of Kr^{85} against the flow of helium sweep gas in the annulus between the pump shaft and the radiation shield plug is being measured in the prototype pump. The helium flow has been varied between 1000 and 300 cc/min with no detectable diffusion of Kr^{85} up the pump shaft annulus. Other tests are being conducted to determine the concentration of undissolved gas that circulates with the pumped molten salt. A radioactive source and detector are being used to measure the change in density at constant temperature of the molten salt as the pump is started and brought to design speed.

Fabrication of the drive motor vessels was continued, and additional fabrication problems developed in weld buildup regions. This problem is being resolved.

Instrumentation Development. – Results of the long-term testing of thermocouples continue to be favorable. Thermocouple drift remains less than 2°F.

The ball-float level transmitter has now accumulated one year of operation at design temperature. Performance of the device continues to be satisfactory. Preparations are being made to install a new unit on the prototype pump.

Preliminary tests were made on a Stereo-Tronic single-channel stereo television system with promising results. The only major difficulties

were experienced in adapting the Stereo-Tronic stereo converter to existing television cameras and lenses.

NUCLEAR SAFETY

Release of Fission Products on Out-of-Pile Melting of Reactor Fuels. — A new experimental technique for the large-scale melting of irradiated UO_2 by using multiple cored-fuel-pellet pins, with a central tungsten resistance heater in each pin, was demonstrated. An unclad seven-pin array of 21 cored pellets was successfully melted in the initial trial, conducted to permit a direct observation of the high-temperature plateout effect on surrounding fuel.

Power was supplied to the six outer pins and the center pin through two parallel circuits from a 25-kw low-voltage saturable reactor. The center pin, only, was irradiated for a fission product source and was mounted on a tungsten rod somewhat longer than those used in the peripheral pins. This resulted in a slightly lower power delivery to the center pin; nonetheless, partial melting was accomplished as planned, since radiant loss from this member was minimized.

Analytical results are still incomplete; however, a rough comparison of the gross gamma release was made and is shown below.

Percentages of Total Gamma Activity Released in Single-Pin and Multiple-Pin UO_2 Melting Experiments

	Single Pin (Bare UO_2)		Single Pin (Zircaloy Clad)		Multiple Pin (Bare UO_2)	
Charcoal trap	0.09		0.05		0	
Gas outlet tube	1.15		0.15		0	
Filters	1.09		0.23		0.004	
Total transported release[a]		2.34		0.43		0.004
Reflective shields	1.15		3.80		0.77	
Base plate	2.58		0.50		1.57	
Total deposited release[b]		3.75		4.30		2.34
Rare gases	0.15		0.21		0.17	
Total overall release	6.22		4.94		2.52	

[a]External, outside the tungsten furnace liner.

[b]Internal, inside the tungsten furnace liner.

Encouraging results may be anticipated from this comparison. For example, no significant amount of particulate activity escaped the immediate furnace liner. The most striking reduction is in the transported release, which is less by a factor of 100 in the multiple-pin experiment than in the single-pin experiments. These preliminary results confirm the expectation that a peripheral ring of partially melted UO_2 offers a suitable plateout surface for released fission products. Further experiments to justify this simulation of the loss-of-coolant accident are being conducted.

Release of Fission Products on In-Pile Melting of Reactor Fuels. — Radiochemical analyses of the ninth experiment on the in-pile melting of miniature UO_2 fuel elements were completed, and the results are shown below:

Material Released by the Ninth In-Pile UO_2 Melting Experiment (%)

Origin of Release	Sr^{89}	Zr^{95}	Ru^{106}	I^{131}	Te^{132}	Cs^{137}	Ba^{140}	Ce^{144}	UO_2
Fuel[a]	71.3	71.5	93.1	99.7	99.1	98.8	71.2	70.2	70.5
High-temperature zone of furnace[b]	0.974	0.123	4.02	79.7	89.3	58.1	1.12	0.302	0.0128

[a]Includes miniature fuel element and holder.
[b]Includes fuel and thermal insulation; minimum temperature, 1000°C.

Thermodynamic calculations indicate that the chemical form of the isotopes released during reactor accidents would be greatly affected by the presence of vaporized stainless steel cladding, and that Sr, Ru, Ba, and Ce would be released in metallic form. Fractionation studies of fission product distributions in the entire UO_2 melting series indicate that strontium and barium may be released differently from each other, but that once released, they are transported in the same manner. Similar information is being obtained for Zr, Ru, Cs, I, Te, Ce, and U.

The second experiment on the in-pile burning of a miniature fuel element composed of a graphite matrix containing spheroidal uranium carbide coated with pyrolytic carbon was performed and is being analyzed. This experiment was a duplicate of the first.

The F-3 facility, in which fuel specimens for future meltdown experiments will be preirradiated to obtain high burnup, has been installed in the ORR, and two specimens are being irradiated.

Characterization and Control of Accident-Released Fission Products. – Radioactive aerosols are being studied by a method which determines their distribution vs depth in fibrous filters under carefully controlled conditions. An aerosol, containing Zn^{65} particles 40 to 300 A in diameter, is generated by passing a stream of air over a Tesla-coil spark gap between two irradiated zinc foil electrodes, and is then transported through a series of uniform (11-μ fiber) Dacron filters at linear flow rates from 0.4 to 88 fpm.

Earlier experiments had indicated that diffusion is the controlling mechanism of filtration at the lowest linear flow velocities and that interception is the primary mechanism in the intermediate range of flow. Collection efficiency increases with decreasing velocity in the diffusion range but is essentially independent of velocity in the interception range. However, at the highest flow rates, the efficiency appears to increase with velocity, indicating the edge of the area in which the inertial mechanism begins to take effect with particles in this size range. These results are in accord with theory over the whole range of flow velocities tested and indicate that the techniques employed will provide valuable information on particle size and filter performance.

Nuclear Safety Pilot Plant. – The tests of foam suppression of activity in air, conducted during the past two months in cooperation with personnel from the Harvard Air Cleaning Laboratory, were completed. Seven tests were performed: five in which ~10 curies of I^{131} was vaporized into the 6000-ft^3 cell volume, and two in which ~10 curies of mixed fission products was released by the oxidation of slightly irradiated uranium foil. The data obtained are being analyzed.

The model containment vessel for the Nuclear Safety Pilot Plant was received and installation will begin soon. Design and fabrication of remote-handling and viewing equipment, as well as of transfer casks and portable shields, were initiated. Arrangements were made for handling radioactive samples in hot cells and analytical laboratories.

Reactor Containment Handbook. – Chapter IV, "Energy Sources," will be issued shortly for review as an internal memorandum; Chapter II has been edited. Chapter VIII, "Specific Containment Systems," was compiled and a draft is being prepared; Chapter V, "Analytical Techniques," is being compiled.

Of the four chapters under subcontract to Bechtel Corporation, Chapter XI, "Economics," is complete in draft form. Agreement on the information to be included in Chapter VII, "Containment Proof Tests," and Chapter IX, "Material Properties," was reached and these chapters are being written.

WASTE DISPOSAL DEVELOPMENT

Clinch River Study. – The vertical distribution of radioactivity in bottom-sediment cores from three sections in the lower Clinch River was determined by gross-gamma counting. These measurements indicate that coring penetrated through the radioactive zone of the bottom sediments.

The distribution pattern shows two zones of relatively high radioactivity, one of which is near the top of the core. These zones are being examined by gamma spectrometry to determine the major radionuclide contributing to these high levels.

Mineral Exchange Studies. – Comparison of sorption by a hydrobiotite (approximately 50% vermiculite and 50% biotite) in solutions whose potassium or cesium concentrations exceeded the exchange capacity of the mineral revealed that the mineral sorbed more potassium than cesium. In a solution containing 1.5 times more potassium or cesium ions than the total exchange capacity, the hydrobiotite was saturated with potassium ions (70 meq/100 g) after 24-hr contact with the solution, but only 70% saturated with cesium ions (49 meq/100 g). Desorption of the alkali ions with calcium showed that 75% (52 meq) of the potassium ions and 90% (44 meq) of the cesium ions remained fixed. Exchange-capacity determinations showed that 95% of the total exchange capacity is accountable by the sum of the fixed and "non-fixed" capacities in the potassium-treated hydrobiotite, but only 70% of the total capacity is accountable in the cesium-treated sample. It is believed that the 21-meq capacity in the cesium sample which is unaccountable by the conventional ion exchange determination is due to "trapping" of the original sodium ions in interlayer positions during the fixation process.

Disposal in Deep Wells. – Geothermal measurements in Joy No. 1 well and well 400S, made by the U.S. Geological Survey, were compared with the thermal conductivity of core samples, and a provisional rate of heat flow from the earth's interior (0.73 ± 0.04 $\mu cal/cm^2$) was computed. A surprising discovery of abnormally low fluid pressure in the lower Chickamauga and upper Knox geologic formations was observed during the temperature survey in the cased Joy No. 1 well.

Mineralogical analyses of 25 samples from the 3263-ft Joy well indicate a considerable mineralogical difference in the samples from the several possible fracturing zones.

Unexplained anomalies in the pumping time of several mixes have been reported by Westco; the cause is being investigated. It has been shown that even under adverse conditions the bulk of the cesium and almost all of the strontium will be retained by the set grout.

High-Activity Waste Calcination. – Hard, microcrystalline solids incorporating up to 45% Purex-waste oxides were prepared from simulated high-sulfate and 1965 FTW (formaldehyde-treated waste) solution by the addition of phosphite or phosphate and sodium, calcium, and/or magnesium. On a semiengineering scale, simulated Darex waste was fixed into a very hard, strong product that incorporated 25% waste oxides and represented a volume reduction of 5.17 from the waste solution. The feasibility of removing mercury from 1965 FTW by amalgamation with copper was demonstrated on a laboratory scale.

Type 304L stainless steel coupled to titanium 45A showed weight losses amounting to a maximum corrosion rate of 0.41 mil/month for 192 hr of exposure to the vapor above refluxing Hanford FTW (Purex waste) solution. Rates for specimens exposed to the solution and at the interface were slightly lower. Titanium 45A showed slight weight gains for coupled and uncoupled specimens under these conditions.

Low-Activity Wastes. – The scavenging-precipitation ion exchange process employs a sodium hydroxide (0.01 M) precipitation followed by clarification and filtration as the head-end treatment. Phosphates in the waste stream hinder complete precipitation, producing a stabilized supersaturation of hardness ions. A carboxylic resin, Amberlite IRC-50, effectively removed this residual hardness prior to secondary decontamination with the phenolic ion exchange resin Duolite CS-100. Under the worst conditions, the hardness (70 ppm) was removed by the primary column from about 1200 bed volumes of treated waste, postponing the breakthrough of the secondary column to the 1600-to-1800 bed volumes normally achieved when there is less than 10 ppm of hardness in the treated waste containing 0.01 M Na^+. The Amberlite IRC-50 treatment is superior to the one that removes hardness by means of the addition of sodium carbonate, because the sodium carbonate reduces the loading of cesium on the CS-100 resin due to the competition between the added sodium and the cesium. A residual hardness of 40 ppm caused a 0.1% cesium breakthrough of the CS-100 resin at about 570 bed volumes in the presence of the usual 0.01 M Na^+ concentration. The addition of 0.005 M Na_2CO_3 (0.01 M Na^+) reduced the residual hardness to less than 10 ppm but doubled the sodium concentration to 0.02 M. This increase in sodium concentration resulted in a 0.1% cesium breakthrough at about 900 bed volumes on the secondary (CS-100 resin) column. The Amberlite IRC-50 resin, like Duolite CS-100, can be regenerated with 0.5 M HNO_3.

Foam Separation Studies. – The removal of strontium from water by foam separation is much more efficient if the calcium concentration is in the 5-ppm range (calculated as calcium carbonate), rather than in the 65-ppm range, as it is in ORNL tap water or low-radioactivity-level waste. Accordingly, studies of a head-end calcium precipitation are continuing, and a 9-in.-diam sludge column was used with ORNL process-waste water as feed. The unit was operated continuously for two 20-day periods at flow rates from 10 to 60 gal ft^{-2} hr^{-1}. The precipitation of calcium and magnesium was induced by making the water 0.005 M each in NaOH and Na_2CO_3; ferric iron (2 ppm) was used as coagulant. The ratio of sludge-bed height to column diameter was in the range 0.5 to 1. Operation was excellent at all flow rates. Dissolved hardness of the effluent water was 2.0 to 4.5 ppm; turbidities ranged from 0 to 5 ppm. Radiochemical analyses showed Sr^{90}, Co^{60}, Cs^{137}, Ru^{106}, and Ce^{144} decontamination factors of 10–15, 30, 1.1, 2.0, and 40 respectively. Chemical analyses showed the alkyl-benzene sulfonate and phosphate concentrations in the waste water to be 0.75 and 1–2 ppm respectively.

Subsequently, the effluent from this head-end treatment was tested for strontium removal by the foam separation process; a 6-in.-diam column was used. The flow rate was 40 gal ft^{-2} hr^{-1}; the gas-to-liquid flow-rate ratio was 10; the dodecylbenzene sulfonate (DBS) surfactant concentration was 275 ppm. A decontamination factor of 27 was obtained for strontium under these unoptimized conditions. The strontium distribution factors of (0.6 to 2.4) $\times 10^{-3}$ cm, calculated from column runs

with clarifier effluent, agree with values from laboratory studies on solutions that contained about 5 ppm calcium.

Engineering studies with 6-in.-diam foam columns and with DBS as the surfactant were continued. Screen-lined, perforated centrifuge bowls were tried as foam breakers, with the following results: The efficiency of foam condensation is greater for dry foam at a low flow rate than for wet foam at a high rate. Screens of 100 or 120 mesh break foam more efficiently than do screens of 40 or 200 mesh. The capacity of the centrifuge increases as the rotational speed increases. However, the foam-condensing efficiency increases as rotational speed decreases, provided that the capacity of the rotating screen is not exceeded.

Disposal in Natural Salt Formations. — Inspection of the wall heater hole after its removal from the structure as part of a large block showed the shattered zone to extend out to a radius of 6 to 7 in. from the original center of the 10-in.-diam hole. These measurements confirm 290°C as the minimum shattering temperature for Hutchinson salt. Measurements of the hole diameter at 210°C, before shattering, showed a maximum diameter decrease of 4-1/2%. Extrapolation of measurements from unfractured areas to the peak salt-temperature region, 350°C at the center of the heater, indicates a diameter decrease of 15%. In the floor experiments, shale and anhydrite bands altered the closure pattern and prevented correlation of closure measurements. Tests showed that no dome-salt samples shattered and that three of four bedded-salt samples shattered.

Engineering, Economic, and Safety Evaluation. — The safety analysis of tank storage assumes the same nuclear complex previously assumed for all the economic studies and a tank capacity of 4×10^5 gal filled in 5.33 years. At the time of filling, there are 4×10^9 curies in the tank. Of the fission product activity, 50% is Ce^{144}-Pr^{144}; 14% is Pm^{147}; 14% is strontium and its daughter products; and 13% is the Cs^{137} chain. The initial transuranic content is 1.2×10^6 curies, with the isotopes of americium contributing 52% of the total transuranic activity. For the first 400 years, fission product activity exceeds the transuranic activity.

PHYSICAL RESEARCH PROGRAM

PHYSICS AND MATHEMATICS RESEARCH

High-Energy Electronuclear Machines. — An innovation in cyclotron design, designated the "separated-orbit cyclotron," was conceived, and a preliminary study is being made. The machine incorporates the advantages of strong focusing, along with those of fixed-frequency fixed-field operation. The orbit is designed to move axially, as well as radially, in a spiral helix. The beehive-like structure would be composed of stacked strong-focusing magnet blocks intersected by radially placed accelerating cavities. The ions would pass through any given magnetic field only once, but would spiral repeatedly through each cavity. The significance of this new concept is difficult to assess at this early stage, but it is believed that, with additional development, the machine may emerge as an important new type of accelerator for the 0.5- to 10-Bev energy range.

Medium-Energy Nuclear Physics: The 86-Inch Cyclotron. — Experiments with Ir^{187} activity indicate the existence of at least 65 transitions of energies less than 1 Mev populating levels in Os^{187}. All of these stronger radiations up to 400 kev are classified as M1, E2, or a mixture of these. The proposed $1/2^-$ [510] ground-state configuration, consisting of rotational states at 9.8 ($3/2^-$), 75.0 ($5/2^-$), and 100.5 ($7/2^-$) kev, is based on five intraband transitions and twenty-one interband transitions populating the levels. The orbital assignment of $3/2^-$ [512] for the 74.3-kev intrinsic state is consistent with the associated rotational excitations at 187.5 kev ($5/2^-$) and 351.0 kev ($7/2^-$) which characterize this configuration. The Coriolis interaction is apparently displacing the energy levels of the close-lying $K = 1/2$ and $K = 3/2$ band structures. A knowledge of the multipolarity of six transitions de-exciting the 501.4-kev state establishes the spin and parity of the level as $3/2^-$. A level at 987.8 kev ($3/2^-$) is depopulated by M1 transitions to odd-parity states of spin 1/2, 3/2, and 5/2.

High-Voltage Experimental Program: Nuclear Resonance Effects in Coulomb Excitation. — The well-resolved resonances in the $Na^{23}(p,p')$ reaction at 0.871 Mev and in the $F^{19}(p,p')$ reaction at 0.780 Mev are situated upon continua which have been fitted with an E2 Coulomb excitation curve [Phys. Rev. 104, 989 (1956); 97, 1226 (1955)]. Therefore these reactions would appear to be favorable cases for the observation of the interference effects between Coulomb excitation and compound nuclear inelastic scattering suggested by several authors [Nucl. Phys. 32, 273 (1962); abstract 134, International Symposium on Direct Interactions and Nuclear Reaction Mechanisms, Padua, Italy, 1962].

The total cross section for the $Na^{23}(p,p')$ reaction has been measured between proton energies of 0.840 and 1.100 Mev by detecting the 0.439-Mev gamma radiation emitted from the first excited state of Na^{23} in a 3- by 3-in. NaI crystal mounted in essentially 2π geometry. The yield of the 0.198-Mev gamma ray emitted from the second excited state of F^{19} was measured in the $F^{19}(p,p')$ reaction. Primarily due to the low counting rate,

the present $Na^{23}(p,p')$ data are not sufficiently accurate to measure the estimated asymmetry of 10% introduced in the total cross section by the interference term.

The measurement of the shape of the resonance for the $F^{19}(p,p')$ reaction is facilitated by a greatly increased counting rate. However, in this case the effect of the nearby resonance at 0.831 Mev makes an unambiguous interpretation of the shape difficult.

High-Voltage Experimental Program: Resonant Neutron Capture Spectra in $Fe^{54,56}$ and $Ca^{40,42,44}$. – By employing time-of-flight and a crude two-dimensional analyzer in the manner described previously [Bull. Am. Phys. Soc. 8, 81 (1963)], measurements were made on the neutron capture gamma-ray spectra for the cases of target nuclei Ca^{40}, Ca^{42}, Ca^{44}, Fe^{54}, and Fe^{56}. The time resolution was only 100 nanoseconds/m, but changes of spectra with energy were clearly evident. It is supposed that the capture was dominated by the resonances at 52 and 74 kev in the isotopes Fe^{54} and Fe^{56} respectively. Transitions were observed to the ground, 0.413-, 0.93-, 1.413-, 2.058-, 2.47-, and 2.94-Mev states in Fe^{55}, and to the ground, 1.20-, 1.62-, 2.51-, and 2.83-Mev states in Fe^{57}. Hence these spectra differ appreciably from the corresponding thermal spectra. The relative intensities of the transitions fluctuated considerably with neutron energy. A target of natural calcium exhibits capture resonances attributable to Ca^{40} in the neutron energy region being investigated. Sufficiently good resolution cannot be attained on Ca^{42} and Ca^{44} targets, because of size, to ascertain the character of the capture in these cases. Principally of interest in the case of the spectra observed in the calcium isotopes is the indication of two states between ground and the 1.95-Mev level in Ca^{41}, and a level between the 0.59 and 0.99 levels in Ca^{43}. A need for further investigation is indicated.

Electronuclear Machines: The Isochronous Cyclotron (ORIC). – Installation of the full-power rf system was completed, and it was test-operated up to the design level of 100 kv on the dee. The variable-frequency system was test-operated over the range 7.3 to 20.5 Mc. The range of mechanical adjustments is being altered to extend operation to the full design frequency of 21.5 Mc. The cyclotron was test-operated at proton energies of 8, 12, and 30 Mev at the design radius, 31.5 in.

Emphasis in test operation at 8 Mev has been on understanding the orbit dynamics of the beam in relation to the beam deflection and ion optics systems. Beam attenuation is less than 10% out to the design radius of 31.5 in. The electrostatic channel, now in operation, moves the beam about 1 in. away from this orbit and provides space for a coaxial magnetic channel. This channel, just installed, will direct the beam into a second channel, now being fabricated. The two-beam positioning magnet, now ready to be installed, will then bring the beam to a horizontal position 48 in. above the floor.

The beam switching magnet is in place, and various components, such as beam pipes, vacuum stations, and shield plugs, are being prepared for delivering the beam to the experiment rooms. Facilities are being installed in the small experiment room; completion of this phase awaits the scheduled delivery of quadrupole magnets in March.

Stable Isotopes Development. — Efforts to increase calutron ion output through the application of radio-frequency potentials in the arc region showed that up to 10% increase in output can be achieved but at the expense of beam focus. The method is not directly applicable in the separations program but provides data useful in understanding the ion extraction process.

Cathodic sputtering is being reexamined as a method for introducing feeds of the platinum elements into the ionization region. The element is placed so that the discharge is directed parallel to the magnetic field. Ion currents of 10 ma of Cu^+ have been achieved.

Thorium-232 was deposited by direct deposition on various types of graphite, and the amount and uniformity of the deposit were determined. Fifty-four samples, each having an area of ~0.8 cm^2, were prepared simultaneously by rotating a supporting wheel through the calutron beam. Uniformity of the deposit was excellent, and the quantity retained varied from 7 to 60 $\mu g/cm^2$, depending on the type of graphite and the length of the bombardment.

Special Separations. — Twenty-six plutonium separation runs have been completed since reactivation of the program in September 1962. Estimated weights and purity ranges are as follows:

Isotope	Estimated Weight for Period (g)*	Accumulative Estimated Weight (g)*	Purity Range (%)
Pu^{239}	5.4563	16.4356	94.88–94.96
Pu^{240}	9.7434	27.3333	96.63–98.78
Pu^{241}	2.4935	6.9848	88.61–94.50
Pu^{242}	1.2660	3.3689	84.22–91.19
	18.9592	54.1226	

*Based on monitored ion currents.

Charge material for ten plutonium separation runs was prepared. Seventeen receiver pockets were received, and processing was completed on six Pu^{240} pockets and six Pu^{241} pockets. Eight of the Pu^{239} and five of the Pu^{242} pockets were scraped and ignited. Since two large carbon burners have been placed in operation, the time required to ignite the backlog of graphite source components has been reduced to ~2 weeks.

A total of 310 g of plutonium was recovered from calutron components and purified; an additional 150 g is being processed. Three returned samples of plutonium are also being processed.

CHEMISTRY RESEARCH

Protactinium Chemistry. — Careful measurements of protactinium distribution between 5 N sulfuric acid and Dowex 1 resin showed that at Pa^{231} concentrations greater than 2×10^5 counts min^{-1} ml^{-1} (0.004 mg/ml) in the acid, the concentration of protactinium in the resin increases with approximately the fifth (±1) power of the protactinium concentration in the acid. In these tests, protactinium concentrations varied from 3×10^6 counts/min per gram of resin (0.06 mg/g) to 1.3×10^8 counts/min per gram (2.6 mg/g). At protactinium concentrations below 2×10^5 counts min^{-1} ml^{-1} in the aqueous phase and 3×10^6 counts/min per gram in the resin, the concentrations in the two phases were approximately linearly related, although scatter in the data prohibited a definite conclusion.

The extraction of protactinium from sulfuric acid into 0.03 N trilauryl amine—diethylbenzene solution showed a similar large increase in protactinium concentration in the solvent as the concentration of the protactinium in the aqueous phase was increased. Preliminary results indicate this to be a third- to fourth-power dependence. In both systems at least several days were required to reach equilibrium.

Pyrolysis and Radiolysis of Organic Liquids. — Gas chromatographic calibration curves for the analysis of the low-molecular-weight pyrolysis products of biphenyl were completed. Liquid- and vapor-phase pyrolyses of equal amounts of biphenyl have been completed, using identical conditions of time and temperature. The yield of benzene, the principal decomposition product, was only half as great for the liquid-phase pyrolysis as for the vapor-phase decomposition. When the glass surface was increased by a factor of 50 upon the addition of borosilicate glass wool, the yield of benzene from the liquid-phase pyrolysis of biphenyl was decreased by a factor of 7.

Analysis of Molten Salts by Electrochemical Methods. — A study was continued on the voltammetry of Fe(II) (previously reported in ORNL-3402, p 26). This study revealed that the current-voltage curves for Fe(II) correspond to the cathodic reduction of Fe(II) to the metal and the anodic oxidation of Fe(II) to Fe(III). From the analysis of the waves, it was established that the electrode reactions proceed reversibly. The limiting current (wave height) of the cathodic wave is proportional to the concentration of iron in the melt over the concentration range investigated (approximately 2 to 10 millimolar). It was established that the limiting currents can be measured with a coefficient of variation of about 10%; these measurements therefore appear to have analytical applications. An activation energy of about 12 kcal/mole was calculated for the limiting-current process which corresponds to the reduction of Fe(II) to the metal.

Radiation Chemistry: Radiation-Induced Surface Oxidation of Hydrocarbons. — When hydrocarbons adsorbed on suitable high-area solids are irradiated with gamma rays, much of the energy absorbed by the solid causes decomposition of the hydrocarbon. It was recently discovered that with oxygen present the hydrocarbon is oxidized, even though with H_2 present unsaturated hydrocarbons are not hydrogenated. So far, the oxidation

of 13 hydrocarbons (methane, ethane, propane, n-hexane, hexene-1, cyclohexane, cyclohexene, toluene, o-xylene, biphenyl, naphthalene, anthracene, and phenanthrene) on each of three adsorbents (silica gel, γ-alumina, and silica alumina) has been studied.

Silica gel is usually the least efficient of the three. Unsaturated hydrocarbons are oxidized more readily than the corresponding saturates, and aliphatics more readily than aromatics. The $G(-O_2)$ values, determined from the decrease of oxygen pressure during irradiation, vary from about 1 for methane, biphenyl, or o-xylene on silica gel to much higher values for o-xylene on alumina ($G = 40$), cyclohexene on alumina ($G = 54$) and on silica alumina ($G = 78$), and hexene-1 on silica alumina ($G = 110$).

Some of the products from toluene oxidation on alumina were extracted and were identified by gas chromatography. The main product was benzyl alcohol, with about one-tenth as much each of o-cresol, p-cresol, and phenol. A trace of m-cresol was found.

In addition to the radiation-induced reaction, there was also a post-irradiation catalytic oxidation. This was evident from the continuing drop in oxygen pressure in the radiation ampoule after irradiation. That this is a true catalytic reaction and not a slow combination of long-lived radiation-produced fragments of the hydrocarbon was shown by irradiating the adsorbent in vacuo, adding the hydrocarbon and oxygen, and observing the pressure decrease.

Radiation Chemistry: Retention of Br^{82} Recoils by Crystalline Alkali-Metal and Alkaline-Earth Bromates. — One of the problems in the preparation of high-specific-activity radioisotopes by the Szilard-Chalmers reaction has been the loss of the separable radioactive atoms by their recombination to the same state of chemical binding that they possessed before neutron capture. Investigations with the alkali-metal bromates, for example, have shown that the recovery of the 35.9-hr Br^{82} recoils is dependent on the nature of the crystalline environment and on the extent to which this environment is damaged by nuclear radiation. Thus the retention of Br^{82} was significantly greater in $LiBrO_3$ than in Li^7BrO_3 because of the extensive decomposition of the former compound by Li^6 fission. If the neutron-irradiated crystals were radiolyzed additionally by Co^{60} gamma rays, the retention could be increased until more than three-fourths of the Br^{82} was recombined. This increase was strongly temperature-dependent; at −196 and −78°C there was no radiation-induced recombination; at 0.0°C it was small.

The Br^{82} recoils also could be recombined by heating the irradiated compounds: Thermal annealing of $LiBrO_3$ for 1 hr at 95, 150, 200, and 240°C increased the retention from an initial 30.5% to 34, 43, 60, and 91% respectively. Melting the crystals and holding them for 30 min slightly above the melting point (254°C) caused 97% of the Br^{82} to re-form into bromate. When small amounts of KBr labeled with Br^{82} were mixed with inactive $KBrO_3$ and held at the melting point of the latter compound (396°C) for 30 min, 40% of the Br^{82} was combined as BrO_3^-.

The Br^{82} recombination reactions can be understood qualitatively if it is supposed that the alkali-metal bromate crystals are decomposed by heat and/or radiation to give mobile oxygen atoms which reoxidize the radiobromine to bromate. The occurrence of such a back-reaction, which

occurs in the presence of a large decomposition of BrO_3^-, suggests (but does not prove) that the radiolysis of bromate ion may be at least partially reversible.

High-Temperature and Structural Chemistry. — The respective electrical conductivities of the solutions of the rare-earth metals La, Ce, Pr, and Nd in their molten tribromides were found to exhibit behavior similar to that of the solutions of these metals in their trichlorides and triiodides. The conductivity rises with metal concentration most sharply with La and Ce and least with Nd; Pr shows again intermediate behavior. The $NdBr_2$-$NdBr_3$ system again exhibits a maximum indicating an electron exchange process between Nd^{2+} and Nd^{3+} as in the iodide system.

The solutions of Gd, Dy, Ho, and Er metals in their trichlorides gave irregular results quite different from those expected on the basis of the measurements with the lighter rare-earth series. The solubility of the metal in the molten trichloride rose from 1.6 mole % in $GdCl_3$ to 28 mole % in $DyCl_3$, only to drop again to 18.5 mole % in $HoCl_3$ and to 6 mole % in $ErCl_3$, in disagreement with the expectation that the stability of the M^{2+} ion, and thus the solubility, would increase in this series. The conductivity showed similar irregularities. The maximum characteristic of M^{2+}-M^{3+} electron exchange was exhibited by Dy-$DyCl_3$.

Corrosion Tests for the TRU Facility. — Hydrogen embrittlement was observed in specimens of zirconium grades 11 and 12 after 1000 hr at atmospheric boiling in 6 M HCl—0.0023 M $LaCl_3$ solution containing hydrogen peroxide. Specimens of both materials were exposed in the solution and the vapor. The hydrogen content of the grade 11 solution-exposed specimen was 1200 ppm, while that of the grade 12 solution-exposed specimen was 1600 ppm. The unexposed control materials contained approximately 22 ppm hydrogen. The vapor-phase specimens of both grades, which were not analyzed for hydrogen content, underwent brittle fracture during a bend test, indicative of substantial hydrogen pickup. An unexposed control did not crack during bending.

Target Preparation. — Research investigations were initiated to develop an explanation for the apparent cessation of work hardening in metals which were cold-rolled to thicknesses of < 0.001 in. Preliminary electron microscopic examinations of rolled nickel foils showed that, upon initial rolling, the number of crystal dislocations was increased over that in the annealed metal, but that the dislocations became concentrated near the edges of the crystallites as rolling continued. This movement of dislocations caused the central bulk of each crystallite to be nearly void of discontinuities.

By vacuum evaporation and subsequent condensation of metal vapors onto single-crystal substrates, it is hoped that single-crystal films of the condensed metals might be achieved. These films would be crystallographically oriented to correspond to the substrate crystal face exposed to the condensing vapor. Cleaved and polished crystals of NaCl, KCl, and LiF are being used as substrate materials, and the conditions of vaporization and condensation are being studied to achieve 1:0:0 and 1:1:0 cubic, crystal-face orientations for silver metal.

Transuranium Element Processing

Analytical Chemistry Development. — A computer program was written for the analysis of absorption spectrophotometric data for systems containing a number of absorbing species that may or may not be at equilibrium. The program was tested by using synthesized spectrophotometric absorption data for two parallel first-order reactions involving three components. The results were excellent.

The investigation of various methods for the determination of zirconium in various Tramex solutions was started. At least four methods seem to be applicable to the microgram quantities of zirconium that are present and also to hot-cell manipulation. These will be evaluated in terms of the effect of high radiation (both alpha and gamma), interfering ions, and sensitivity.

Both the 400-channel analyzer and the automatic alpha counter are being inspected and assembled and should be operational by February 15.

Corrosion Studies. — An investigation of the effect of oxidant species on the corrosion of Hastelloy C by 6 M HCl at 35°C was completed. Tests were run with (1) normal atmospheric oxygen in solution, (2) oxygen aeration of the solution, and (3) continuous addition (about 3 ml/hr) of 0.05 M H_2O_2 solution. The peroxide concentration in the test solution never exceeded 0.001 M. The best corrosion resistance was shown by a solution-exposed weld specimen with atmospheric oxygen present; after 1000 hr the rate was 0.7 mil/month. With oxygen aeration and with peroxide addition, the rates for similar specimens increased to 5.2 and 4.0 mils/month, respectively, after 1000 hr. All specimens were preferentially attacked in heat-affected zones adjacent to welds. The severity of the attack was greatest in the oxygen-aeration test, where the measured penetration ranged between 28 and 32 mils. Rates after 1000 hr were nearly identical for unwelded specimens exposed to the vapor in the no-added-oxidant and in the peroxide-added tests, being 1.5 and 1.7 mils/month respectively. The unwelded specimen in the vapor above the oxygen-aerated test corroded at a rate of 3.7 mils/month.

Corrosion data were obtained on welded Zircaloy-2 in mixtures of nitric and hydrochloric acids at 43°C and at boiling temperatures. When the solution was 6 M in nitric acid, with varying concentrations of hydrochloric acid up to 0.5 M, the corrosion rate was consistently less than 0.1 mil/month in the liquid phase. A maximum vapor-phase corrosion rate of 0.2 mil/month was observed above a boiling solution of 6 M HNO_3–0.5 M HCl. There was no indication of preferential attack in the weld areas. However, accelerated corrosion was observed in both 3 and 6 M HCl that was also 0.1 and 0.5 M in nitric acid. In 3 M HCl the 0.5 M HNO_3 produced a corrosion rate of 5.5 mils/month at boiling and 0.7 mil/month at 43°C. In the 6 M HCl solution, 0.1 M HNO_3 produced the highest corrosion rates: 1 mil/month at 43°C and 10.8 mils/month at boiling. Again, no preferential attack in heat-affected zones was noted.

Design of Experimental Facilities. — The design of cell 4 is 98% complete, and the design of the roof-area sample-handling equipment is in the checking stage. The design of prefabricated piping is 75% complete. Design of the sampling system and the shielding for the multichannel gamma spectrometer has not been started.

The construction of the makeup area is complete except for the installation of the glove box for the pump drive. The construction of the operation area is complete except for the installation of the fire protection system and the gamma spectrometer equipment. The equipment for the roof area has not been installed, but all components except the sample removal box are fabricated. All structural supports are in the cell, and installation of the piping was started.

The cell tanks to be plated are now at Parma, Ohio, being prepared for application of the tantalum. A tantalum valve is being made in the ORNL shops, and, when completed, it will be tested for operability and leak-tightness at the TRU mockup.

HFIR Target Development. — Pellet-pressing studies showed that the addition of about 20 vol % of ultrafine aluminum powder to 63 vol % of the 20-μ metal particles produces a continuous-metal matrix when pressed with 17 vol % of oxide. The ultrafine metal powder and the ceramic powders were blended first and then mixed with the coarser metal powder, finally being cold-pressed. A very uniform dispersion of the oxide in the metal matrix was obtained.

The use of a continuous-acting press with rapid load rates produced a superior surface on the aluminum cylinders. The cap powder was also better densified and more resistant to erosion during ultrasonic cleaning. The use of the fast-acting press may permit less stringent dimensional requirements in the fitup between the die and sleeve.

End closures on simulated target capsules were made by using a braze alloy consisting of Al—12% Si. Several advantages can be cited for braze joints, compared with welded joints. Among the advantages are the following: The bond area is ten times larger when the joint is brazed; it is a simple operation that easily produces a good bond; and the joint can be inspected. Several different types of braze-joint designs are being evaluated.

Construction is nearing completion on the area for the TRU mockup assemblies. The plutonium-target line will be the first equipment assembled in this laboratory. Fabrication of the pellet inspection and loading equipment is about 80% complete in the shops at ORGDP. Designs were issued for comments and checked for correctness on the cubicle 3 transfer arm and the Blendor dispenser. Comments were favorable, and the drawings were returned for correction.

Chemical Process Development. — Laboratory tests indicate that the Tramex feed adjustment step can be satisfactorily accomplished by evaporation. Simulated dissolver solutions containing 2 M HCl were evaporated, without aluminum precipitation, to yield a 10 M LiCl—0.2 M $AlCl_3$ solution containing about 0.1 M HCl, which had a boiling point of 129°C. Unfortunately, the depletion of acid from such solutions by radiolysis due to alpha radiation is high. With Cm^{242} at about 2 w/liter, a G value for hydrogen ion depletion of 5.5 was observed for a synthetic feed solution; it was 4.1 for an actual feed. The latter value is equivalent to the destruction of 0.5 mole of HCl per liter per day at a process level of 10 w/liter.

By pressurizing the column to suppress gas-bubble formation, Am^{241} and Cm^{242} were separated by elution from Dowex 1-8X with 4 M $LiNO_3$ at pH 1.5. The single-stage separation factor was 1.7. Use of this procedure

with irradiated Cf^{252}, however, gave almost no separation of californium, einsteinium, and fermium. Single-stage separation factors between adjacent elements were less than 1.1. Almost complete separation of americium and californium was obtained in a test which used preferential stripping from 2-ethylhexyl phenylphosphonic acid into hydrochloric acid solutions. An ion exchanger was prepared for this test by adsorbing the 2-ethylhexyl phenylphosphonic acid on powdered glass and using it as a static exchanger bed. The americium was preferentially stripped with 2 M HCl, and a single-stage separation factor of 18 was obtained.

Tertiary amine extraction was found to be an attractive alternative method for recovering americium and curium from nitrate raffinate solutions of the ion exchange process for the recovery of plutonium. By adjusting the nitrate solution to 3 M $LiNO_3$–1 M $Al(NO_3)_3$ and then extracting with 0.6 M Alamine 336·HNO_3 in diethylbenzene, a distribution coefficient of 8.0 was obtained for americium and 5.5 for curium. By stripping the actinides from the amine with hydrochloric acid, the conversion from the nitrate to chloride systems can be accomplished by this procedure.

Process Equipment Development. – A system composed of three pulse columns was used to study the Tramex process. When the column was fitted with only sieve plates (1/32-in. holes, 5% free area, 1/4-in. spacing), the smallest stage height observed was 28 in. for the scrub section of the process. This stage height was reduced to 18 in. by providing four coalescence sections consisting of 2-in. spaces filled with Teflon Raschig rings (1/4 × 1/8 in.). This stage height is adequate for the proposed TRU pulse column if it can be provided by a packing more radiation resistant than Teflon.

In tests to determine resistance to gamma radiation, 11 protective coatings remained serviceable after exposure to more than 10^9 rads in deionized water. In air, all coatings were more resistant to radiation, and 16 coatings were serviceable after exposure to more than 3×10^9 rads.

Ten of the commercial disconnect clamps were tested for misalignment tendencies. Measurements made at 60 lb-ft of torque gave a misalignment ranging from 1.1 to 1.8°. The acceptable limit is 0.5°. The contact surface of the upper clamp arm was then machined at 1.5°. Subsequent misalignment tests indicated acceptable values for all clamps except one, which gave 0.8° of misalignment.

Five 3/8-in. tantalum disconnects made by forming the cones on the tubing itself leaked excessively during tests. The sealing surfaces of these specimens will be burnished with a rolling device and then retested. This procedure should improve the sealing of the tantalum disconnects.

Transuranium Facility. – The title II design for the Transuranium Facility was completed, and the lump-sum contract portion of the construction job is being submitted for bids. It is expected that construction will begin by July 1, 1963. Procurement of special materials and equipment for the facility by ORNL was begun. Orders were placed for Hastelloy C and stainless steel tube plate, tubing, and fittings. Requests for bids were issued for several other fabricated items.

Design of chemical process equipment was started, and a design of a Zircaloy-2 dissolver was issued for comment. Design is in progress on the following: the arrangement and mounting of disconnects in the tank pit, the hot disconnect well, line bundles and track support, the service plugs for bringing lines into the tank pit, and the in-cell sampler.

Satisfactory bids were received and orders were placed for Zircaloy-2 for the dissolver and one solvent extraction equipment rack.

A test ion exchange column was designed and is being built. The final 15 engineering flowsheets are complete and ready for checking.

METALLURGY AND MATERIALS RESEARCH

Preparation of Pure Materials. — The fourth LiF crystal (ORNL-3402, p 29) was examined under polarized light and infrared radiation. No strains or hydroxyl contamination was detected.

In preparation for growth of the fifth LiF crystal, approximately 300 g of high-purity LiF containing 96.50% Li^7 was synthesized, dehydrated, and melted in the Stockbarger furnace.

Attempts to produce UO_2 crystals (for radiation damage studies) by solution growth from $Na_2B_4O_7$ and electrolytic reduction of UO_2^{2+} from chloride solvents were initiated. Crystals with 1-mm edges were produced by each method. Approximately 100 ppm each of sodium and boron were found in the UO_2 crystals produced from the borate melt. The UO_2-$Na_2B_4O_7$ solution remaining on termination of the cooling cycle was in the form of intensely colored green glass which contained 0.5 wt % UO_2.

Spectroscopy of Ionic Media. — Spectra of Ni^{2+} showed tetrahedral NiX_4^{2-} (X = Cl, Br, I) present in the molten salt solvents KCl and CsCl, for which measurements spanned $^3T_1(F) \rightarrow {}^3T_2(F)$, $^3A_2(F)$, $^3T_1(P)$, and gave $Dq = 370\ cm^{-1}$ at fixed $B = 810\ cm^{-1}$; the spectra also showed NiX_4^{2-} in a large number of organic halides, for which measurements spanned $^3T_1(F) \rightarrow {}^3A_2(F)$, $^3T_1(P)$, a ^1D component, and CT bands. Spectra of NiF_2 in molten LiF-NaF-KF showed octahedral NiF_6^{4-} with $Dq = -660\ cm^{-1}$ and $B = 950\ cm^{-1}$. Spectra of Ni^{2+} in sulfate and nitrate melts showed strong threefold splitting of ^3P, indicating a rhombic distortion. Spectra of Ni^{2+} in molten dimethylsulfone show octahedral solvo-complex formation with spin-orbit splitting of $^3A_{2g} \rightarrow {}^3T_{1g}(F)$ and abnormally low Dq. Additions of halide salts produce tetrahedral halo complexes. Measurement of $A_1' \rightarrow E(\pi \rightarrow \pi^*)$ for $LiNO_3$ in molten $LiClO_4$ gives $\epsilon = 8400$. Spectra of Bi in molten $BiCl_3$ show two solute species with $D_n/D_m = 1/4$. Choosing $4Bi^+ = Bi_4^{4+}$ gives $K_N = 3.8 \times 10^6$ at 264°C. Densities of many fused salts were measured. Numerical tests of the split p-orbital treatment of electron correlation utilizing σ core orbitals for the $1s^2 2s^2 2p_z^2$ nonstationary state of carbon showed the lowest-energy singlet to correspond to a conventional rather than a split p-orbital wave function.

Fundamental Investigation of Radiation Effects in Solids: Calculations of Some Ground-State Properties of F Centers. — Most of the recent calculations of the electronic structure of the F center have been made with a wave function expressed as a linear combination of a function $\psi_0(\bar{r})$, centered at the vacancy, and the occupied orbitals of free, positive, alkali ions centered on the nearest-neighbor sites of the vacancy. The linear coefficients in the wave function are determined by orthogonality and normality conditions. In the present work, various ground-state properties are treated with a wave function of the above type but

which contains, in addition, halide ion orbitals centered on the next-nearest-neighbor sites. The properties considered are the energy, the hyperfine interaction, and the displacement of neighboring ions. The results of calculations on LiF and LiCl show that the hyperfine interaction with the nearest-neighbor lithium ions is reduced by inclusion of the neighboring halide orbitals, which is more in accord with experiment. The ground-state energy is raised relative to the nearest-neighbor approximation.

Fundamental Investigation of Radiation Effects in Solids: Electron Structure of the M Center. — Experimental work of the last few years has fairly conclusively established that the M center in alkali halides may be considered as two F centers bound together at neighboring halide sites. If the F center is considered to be the lattice analogue of the hydrogen atom in free space, the M center can be considered the lattice analogue of the hydrogen molecule. By use of this model, the M center is treated theoretically in the Heitler-London approximation. The F-electron wave functions are of the vacancy-centered type. The finite extension of the neighboring alkali ions is treated, but the point-ion approximation is used for all other ions. The results of calculations on LiF and LiCl show that a satisfactory value for the binding energy ($\approx$0.4 ev) can be obtained with this model. The value of the variation parameter which minimizes the M-center energy is very nearly the same as that for the F center.

CONTROLLED THERMONUCLEAR RESEARCH

Plasma Physics and Arc Research. — The study of the plasma formed between magnetic mirrors by the pressure-gradient arc has continued. The deuterium plasma is of main interest, but argon, nitrogen, helium, and hydrogen have been found to form plasmas with electron temperatures six to ten times greater than the electron accelerating potential. The maximum electron temperature observed was 80 kev, calculated from the x-ray spectrum. The spectrum shows exponentially decreasing abundance, dropping into the noise at about 450 kev. The maximum electron density was about 10^{11} ions/cm^3, calculated from the total emission and the volume of the plasma. This gives a beta ($8\pi nkT_eB^{-2}$) of about 0.1.

DCX-1 Facility. — Experiments were completed in which the contribution to trapped proton density by Lorentz dissociation of H_2^+ beams from the new duo-plasmatron source was measured. The source was designed so as to permit injection of milliampere beams with calculated (i.e., vacuum) fields in the extractor region either significantly above or below the critical field value (about 7×10^4 v/cm) that results in premature dissociation of the high vibrational states of interest. The contribution of Lorentz dissociation relative to that of low-pressure gas dissociation was measured for source operation in both the low- and the high-arc-voltage modes. The contribution of Lorentz dissociation was not apparent in either mode, which indicates that the fraction of the H_2^+ beam trapped via Lorentz dissociation was not greater than about 10^{-7}. In contrast, the calculated Lorentz trapping fractions in DCX-1, based on excited-state

populations calculated or measured elsewhere (no other measurements have been reported on beams from duo-plasmatron sources), range from about 10^{-6} to 10^{-4}.

A new record was established for the charge-exchange decay time of the steady-state proton distribution at an (uncorrected) ion-gage pressure of 1×10^{-9} torr. A semilog plot of the decay signal from a centrally located foil-covered neutral-particle detector showed an initial τ of about 20 sec, a time of 53 sec for decay to the 1/e point, and a decay from 50 to 10% of initial amplitude that closely fitted a τ of 80 sec. Other runs were made at low operating pressures (generally $< 5 \times 10^{-9}$ torr), and it was noted that the decay times registered by detectors at progressively larger displacements from the median plane were progressively shorter. The detectors at Z = ±4-1/2 in. typically gave τ's 1/2 to 1/3 those of the central detectors. These observations indicate that the average energies of the proton distributions off the median plane are degraded, as would be expected in an environment where fresh protons are injected in the median plane and the scattering is not catastrophic.

A number of the low-pressure runs were investigations of the effects of stabilization techniques (described in ORNL-3410) on gross plasma properties (generally the steady-state neutral-particle detector signals, the charge-exchange decay times, and the coherent rf emission). Longitudinal electron currents (from 0 to 80 ma of 100-v electrons and 0 to 500 ma of 420-v electrons) were accelerated through the plasma but did not appear to lead to enhanced stability. Radiation at the lower proton cyclotron harmonics could be partially suppressed, but other signals appeared usually at greater than $3\omega_{ci}$ (the range 10 to 100 Mc was surveyed). The neutral-particle-detector signals also showed that the proton energy distributions were considerably degraded with electron currents greater than about 50 ma.

The effects of varying the H_2^+ injection orbit were also studied. The relative amplitudes and the repetition rate of the rf burst activity could be modified by almost any change in this orbit, but the changes in the other gross properties were usually modest and not consistently reproducible. Exceptions were the data obtained by first securing the normal beam trajectory at the normal or less-than-normal value of the confinement field and then operating at 5 to 10% above the normal field value. At the higher fields the repetition rate of the rf bursts was reduced by an order of magnitude, and there was evidence for reduced proton energy degradation.

DCX-2 Facility. — Further measurements with gridded probes in the mirror throat and in the plasma regions confirmed plasma potential estimates given earlier. Gridded probes and calorimetric measurements show energetic ions at greater radial positions than expected, but the ions do not extend out to the liner wall. However, obstacles at the same radial position, but different θ and Z positions, severely affect the energetic ions measured in both ways. Thus permanent obstacles, such as the injector, should also interfere with the ions before they reach the walls.

Correlations in time have been identified on data received by various kinds of detectors in the apparatus. For example, the onset of radiofrequency radiation is correlated with a drastic change in the current of

neutral particles leaving the machine in certain regions. Also, delayed correlations were observed between fast-ion current on certain gridded probes placed radially outside the main plasma region and the onset of radio-frequency radiation. However, phase correlations of the fast-ion current did not indicate rotation of the plasma.

Phase correlations of the radio frequency up to ~40 Mc were observed on three electrostatic probes arranged at 0, 90, and 180° about the axis. The phase shift of the radio frequency was determined and showed approximately 90 and 180° shifts between probes oriented 90 and 180° apart in azimuth.

Vacuum. — Initial efforts to improve the instrumentation and the reliability of data acquired from the getter test facility were begun. Several molybdenum filaments wound on tungsten were tested as a part of the effort to develop reliable methods of evaporating molybdenum. Several additional experimental filaments were designed and constructed.

Design of the small (10-in.) bakeable flange test facility was completed, and the unit is being assembled. Various flange assemblies and gaskets were procured for dependability determinations under conditions of temperature cycling which will simulate normal procedures in the systems which require baking.

A small facility was constructed which will allow base-pressure measurements to be made for various mechanical pump oils. This work and measurements of the integral oil backstreaming rate and pumping speeds from modern diffusion pumps constitute present efforts on component testing.

BIOLOGY AND MEDICINE PROGRAM

MOLECULAR- AND CELLULAR-LEVEL STUDIES

Physics of Tissue Damage. — Predictions about events in irradiated condensed materials are usually made by extrapolating the results of ionization in gaseous systems with appropriate density corrections. Recent theoretical and experimental reports indicate that such simple extrapolations are probably inadequate to account for the spectrum of events produced in condensed systems. In biological systems there is even less agreement, since there are few determinations of the values of energy loss processes in carbonaceous material in the condensed state.

The optical emission from x-irradiated powders of the aromatic amino acids and the enzymes trypsin and ribonuclease has been compared with the data of Augenstein for ultraviolet excitation. The results indicate that: (1) the ratio of the number of photons emitted during fluorescence to the number emitted during phosphorescence from ultraviolet excitation is approximately one order of magnitude greater than the ratio from x irradiation; (2) the decay of the x-irradiation-produced phosphorescence appears to have longer-lived components than that produced by ultraviolet radiation; (3) the emission spectra obtained during x irradiation contain a long-wavelength component not found in the corresponding ultraviolet irradiation.

TERRESTRIAL AND FRESH-WATER ECOLOGY

Evaluation of Fission Product Distribution and Movement in White Oak Drainage System. — A series of 250 core samples was taken in the bed of former White Oak Lake to determine the quantity, type, and distribution of radionuclides within the area. These cores show that the contaminated lacustrine sediment, which was deposited during the 1943–1955 impoundment, is as much as 20 in. thick in the lower part of the lake bed near the dam; however, there is a gradual thinning of the layer upstream and, in general, toward the shoreline.

Glass-Rod Dosimetry in Environmental Studies. — In new dosimetry studies, miniature metaphosphate glass rods are being used to estimate absorbed dose in air and in the plants and soils of White Oak Lake bed. Combined beta and gamma doses in the top inch of mineral soil ranged from 20 to 25 rads/day during November. Absorbed doses in air at the surface of a marsh plant (*Typha latifolia* L.) ranged vertically from 6.2 rads/day at ground level to 2.8 rads/day at 5 ft above the ground. Absorbed doses in stem tissue of the same plant were 23% higher due to absorbed radionuclides. The dose to root systems of these plants was three to seven times higher than the dose to shoot components. These soil dose rates may be high enough to prohibit germination or seedling establishment of radiosensitive plants such as conifers. In other experiments, techniques were developed for insertion of glass rods into acorns to achieve better estimates of dose absorbed by sensitive embryonic tissue in contaminated soil.

Seasonal Changes in the Blood of Rodents on Radioactive and Nonradioactive Areas. — Erythrocyte numbers and hematocrits in free-ranging wild rodents on nonradioactive areas increased 8 to 9% from autumn to winter in white-footed mice, house mice, and pine mice but decreased in rice rats (−11%) and cotton rats (−9%). No change in erythrocyte number and hematocrit was exhibited from autumn to fall in free-ranging rice rats on the radioactive upper White Oak Lake bed, but values for these measurements increased in penned cotton rats on both the lake bed and on uncontaminated area. Total leukocyte counts for the five species showed great variability within species, but no significant differences between species and seasons were found. Average leukocyte count for all species in all seasons was $5.2 \times 10^3/mm^3$.

Removal of Sr^{90} from the Clinch River by Emergent Insects. — The transport of Sr^{90} from the Clinch River by emerging aquatic midges (Chironomidae) was compared with the accrual of Sr^{90} to the river from weapons-testing fallout. Larval development of midges occurs in the contaminated river-bottom sediments. Insect light traps were operated on the river bank at dusk to collect the emerging adults. These were analyzed for Sr^{90} by radiochemical methods. The adults in one large, composite sample collected in 1962 contained 8.76 times as much Sr^{90} [$(3.55 \pm 1.13) \times 10^{-6}$ $\mu c/g$] as did an equal quantity of river-bottom sediment from the same location. Studies elsewhere have shown that as much as 10 g m^{-2} $year^{-1}$ may be contained in the emergent insect biomass. These emergent insects would transport $(3.55 \pm 1.13) \times 10^{-5}$ μc of Sr^{90} per square meter per year

from the river bottom. The average increment of Sr^{90} from fallout in the United States between 1959 and 1960 was 4.2×10^{-3} curie/mile^2 (1.62×10^{-3} $\mu c/m^2$). Thus, fallout entering the river directly would add 45 times as much Sr^{90} as was removed by emerging chironomids.

Removal of Sr^{90} from the Tennessee River in Clam Shells. — Previous studies showed that fresh-water clams concentrated strontium in their shells by factors of 2500 to 9000 times and therefore are useful as indicators of Sr^{90} in the environment. Since there is a commercial clam (mussel) fishery on the Tennessee River in Tennessee and north Alabama, calculations were made to estimate the removal of Sr^{90} from the river in clam shells. The annual harvest of shell ranges from 5000 to 10,000 tons. By using the maximum harvest and the average Sr^{90} concentration in clam shells (7.019×10^{-6} $\mu c/g$), the calculated removal of Sr^{90} was 64 mc/year. If Sr^{90} releases were constant and if the growth of the population was in equilibrium with the harvest, a similar amount of Sr^{90} would be incorporated into the population each year. Other studies suggest that there are about 4.5×10^7 clams in these populations. Assuming an average weight of 250 g each and the same Sr^{90} concentration, the reservoir of Sr^{90} in clams in the river is calculated as 79 mc. The annual removal and reservoir of Sr^{90} in clam shells is small when compared with Laboratory releases of Sr^{90}, which have ranged from 29 to 150 curies/year.

Soil Microbiological Studies on Oak Ridge Reservation. — Estimates of the microflora and its activity on leaf litter of mulberry, redbud, white oak, and pine were made in forest stands of maple, white oak, and pine in Oak Ridge. Measurements of bacteria, fungi, mycelium development, and soil respiration were made biweekly from November 1960 to November 1961. Time and leaf species influenced microbial populations and their development and metabolic activity significantly. The influence of the various stands was only significant for mycelium development. Bacterial and fungal populations on oak and pine litter increased in all stands throughout the year. This increase was most rapid in pine litter (from 2 to 32×10^6/g for bacteria and from 6 to 40×10^4/g for fungi). Bacterial and fungal counts on mulberry litter decreased, and those on redbud did not change considerably with time and decay (from 6500 to 300×10^6/g and from 1000 to 55×10^4/g respectively). Mycelium development was low during winter (28 mm cm^{-2} week^{-1}), increased with increasing decay and temperature in spring and summer (46 mm cm^{-2} week^{-1}), and slowed off in the fall. Mycelium growth increased on litter in the order mulberry, redbud, oak, and pine and was significantly larger in white oak stands (40 mm cm^{-2} week^{-1}) than in pine (34 mm cm^{-2} week^{-1}) and maple (34 mm cm^{-2} week^{-1}) stands. Microbial respiration rates (in μl of CO_2 per g per hr) paralleled temperature throughout the year and decreased on leaf species in the order mulberry (15), redbud (6), white oak (4), pine (3). Rates of weight loss of the various species decreased in the same order (90, 64, 52, and 45%/year respectively). As a first approximation it was shown that bacterial and fungal populations were mainly controlled by leaf species and stage of decay; mycelium growth by decay and temperature; and microbial respiration by temperature.

Transfer of Co^{60} and Ru^{106} from Oak and Ash Leaves to Several Forest Soils. — Leaves of southern red oak (*Quercus falcata*) and white ash (*Fraxinus americana*), tagged with Co^{60} and Ru^{106}, were enclosed in 1-dm^2 bags

of Fiberglas netting and exposed to natural weathering for two years in shortleaf pine, red maple, and white oak forests. Thin (2 to 3 cm) slabs of underlying soil (organic mor for pine forest, A_1 mull horizon for maple stand, and a combination of organic and mineral layers for oak forest) had previously been cut out of the forest floor and inserted in plastic sandwich boxes with Fiberglas window-screen bottoms. Litter bags were placed on the slabs of underlying soil. Both bags and soil were dried, reweighed, and recounted at 0.5-, 1.0-, and 2.0-year intervals.

Both leaf weight and radionuclide content decreased in an exponential manner. The radionuclides lost by leaching were accounted for in the thin slabs of soil, indicating relatively slow leaching of both trace elements through several kinds of forest humus. Regression estimates of rates of loss from oak leaves in oak, maple, and pine forests in the first year (in percent per day) were: weight, 0.226, 0.156, and 0.197; Co^{60}, 0.350, 0.340, and 0.268; Ru^{106}, 0.276, 0.162, and 0.246. Contrary to expectation, losses were slower for ash leaves: weight, 0.118, 0.086, and 0.103; Co^{60}, 0.201, 0.150, and 0.213; Ru^{106}, 0.195, 0.064, and 0.131. These results and the data after two years indicate that cobalt, at least, disappeared faster than total organic matter, in spite of the strong tendency for organic compounds to complex cobalt. Disappearance of Ru^{106} was little, if any, faster than that of litter. This indication of strong retention in dead organic materials contrasts with the rapid elimination of ruthenium from living organisms.

RADIOLOGICAL PHYSICS, HEALTH PHYSICS, AND INSTRUMENTATION

Radiation Physics and Dosimetry: Time-of-Flight Electron Beam Monochromator. — An electron gun, under development, utilizes time-of-flight velocity selection to obtain a monoenergetic electron beam. An avalanche transistor pulser supplies nanosecond gating pulses to a negative grid. Electrons are accelerated from a negatively biased cathode to a grounded plate containing a hole which allows entry into a drift tube aligned with the earth's magnetic field. Tube length is chosen so that electrons of any particular energy will spiral around the direction of the earth's field with the cyclotron frequency and be refocused on an aperture at the tube end. A delay-line-coupled electrostatic or magnetic deflector will eliminate all electrons except those arriving within 600 ± 1 nanoseconds, which should be homogeneous in energy within 0.01 ev. These electrons will then be used in investigations of electronic energy levels in solids. Characteristics of the system have been obtained with a flat collector at the aperture position. Pulses amplified by cascaded distributed amplifiers are delivered to a sampling oscilloscope. Electrons of 1.5 to 15 ev have been employed, with pulse shapes and arrival times indicating energy spreads of the order of 1 ev, as expected from thermal energy of emission from the filament source. The square of the reciprocal of the arrival time is a linear function of electron energy which extrapolates to the origin.

Radiation Physics and Dosimetry: Electron Slowing-Down Spectra in Copper. — Primary- and secondary-electron flux spectra were obtained with

the spherical electrostatic beta spectrometer (Keplertron). The energy calibration of the Keplertron was further checked by Auger lines from relatively thin sources of Cu^{64} and Eu^{152}. Plane and "black body cavity" sources were irradiated in a reactor to produce the effect of "semi-infinite" and "infinite" media respectively. These sources were hydrogen-annealed to remove surface oxide before irradiation. The sources, which had a strength of several curies, were then placed on the inner sphere of the Keplertron, and the spectra were obtained by varying the outer-sphere potential. The electron flux falls with decreasing energy from 40 kev to a broad minimum at about 8 kev and is thought to represent the contribution from the primary beta rays. Below this energy the flux is attributed to secondary electrons and rises rapidly by 5 decades, achieving a maximum at 3.3 ev and declining below this energy. Calculations on low-energy slowing-down spectra are in progress.

Radiation Physics and Dosimetry: Measurement of Spectral Distribution of Positron Flux in an Infinite Copper Medium Containing Cu^{64}. — The spectral distribution of the flux of positrons from a cavity in a beta-radioactive medium was experimentally determined by using an anthracene coincidence scintillation spectrometer. Positrons leaving a copper cavity source containing Cu^{64} were absorbed in a thin anthracene crystal. The light pulse from the latter was recorded in a 256-channel analyzer if an NaI scintillation spectrometer nearby recorded simultaneously a count under the total absorption peak for annihilation radiation. Discrimination against negative and secondary electrons, thus obtained, permitted observation of the primary positron slowing-down flux. Data were corrected for the nonlinear pulse height vs energy relationship in anthracene by a semiempirical theory due to Birks, the validity of which was demonstrated by its use in obtaining a linear Fermi plot of Pm^{147}. Positron spectra were corrected also for Compton absorption of the annihilation radiation in the anthracene. Results were compared with a simplified version of the continuous slowing-down model of Spencer and Fano. Over the energy range measured (20 to 650 kev), it was concluded that this continuous slowing-down model gives the correct shape for the primary slowing-down spectrum.

Radiation Physics and Dosimetry: Investigation of Collective Effects in the Absorption of Radiation in Matter. — The radiation emitted by thin foils of Au, Cu, Ge, Al, and Ag was investigated with an electron accelerator providing energies up to 100 kev and a vacuum ultraviolet spectrometer and photomultiplier for spectral analysis. In addition to the broad continuum exhibited by other metals, silver emits a relatively sharp and intense peak at 3300 A (3.76 ev). Some investigators have taken this as evidence of support of Ferrell's theory, as the energy-loss experiments show a discrete loss at about this value.

While the energy dependence and polarization of this optical peak are in accord with predictions of Ferrell's theory, the broad continuum requires a different explanation. Ritchie's generalization of the theory of transition radiation and his theory of optical bremsstrahlung explain both the continuum and the silver peak.

ISOTOPE DEVELOPMENT PROGRAM

Nuclear Analysis Research and Development: Use of a 14-Mev Neutron Generator. — A 14-Mev neutron generator is being used both for carrying out nuclear analyses and in a research program directed toward the development of nuclear analysis methods and techniques. Oxygen, fluorine, phosphorus, and carbon have been determined in a number of matrices. Neutron-induced reactions in materials composed of low-Z elements are being systematically studied; the information thus acquired will be used to evaluate the sensitivities and potential uses of specific techniques and methods.

Nuclear Analysis Research and Development: Individual Characterization by Hair Analysis. — Factors affecting the utilization of activation analysis of hair in the identification of individuals are under investigation. The effectiveness of various hair-washing methods in the removal of surface contamination was evaluated. It was found that any shampoo removes essentially all surface contamination. Organic solvents are not effective wash solutions for removing inorganic materials from the hair.

Nuclear Analysis Research and Development: Enforcement of the Narcotics Laws. — Over 100 documented samples of raw opium have been irradiated in the ORR to 7×10^{16} nvt, and gamma-ray spectra of these samples have been recorded after several different decay periods. A comparison code for the gamma-ray spectrum is being written for the IBM 7090 computer; it is hoped that the opium spectra can be processed by this code.

Curium-242 Preparation. — The recommended conditions for the Tramex process were modified in three respects to obtain the decontamination necessary for the processing of Cm^{242}: (1) Aluminum chloride was added to the feed to simulate the aluminum used as target material. (2) More-concentrated hydrochloric acid (8 M) was substituted for the stripping solution to improve the zirconium, niobium, and iron decontamination. (3) A scrub of freshly prepared Alamine 336·HNO_2 was added to improve ruthenium decontamination. Ruthenium, zirconium, niobium, and rare-earth decontamination factors were $\geq 10^4$ in laboratory mixer-settlers with simulated feed containing tracers.

Although irradiation tests of 0.6 M Alamine 336·HCl—diethylbenzene indicated no difficulties up to 300 whr/liter, the loss of acid in Tramex feed by radiolysis at high activity levels poses a potentially serious problem. Experimentally determined G values for acid destruction were between 4.0 and 5.6 molecules per 100 ev. Unless methods for averting this acid loss can be found, it will be necessary to add makeup acid to prevent the precipitation of hydroxides.

Stable Tramex feeds of low acidity, which are required for actinide extraction, were prepared on a 1- to 2-liter scale in laboratory glassware by simply distilling off excess acid and water. Further tests are needed, when plant equipment becomes available, to determine whether the configuration of the equipment is important.

Radioisotope Research and Development. — The total combined flow of the organic and aqueous solvent phases at flooding conditions was determined for several solvent systems in a laboratory-scale, air-pulsed mixer-settler. The total flow capacity, V_t (liters/hr), is given by the following equation:

$$V_t = K \frac{\rho_a - \rho_o}{\mu_a} + \frac{\rho_a - \rho_o}{\mu_o} ,$$

where ρ_a and ρ_o are the densities (g/ml) of aqueous and organic phases, μ_a and μ_o are the viscosities (centipoises) of the aqueous and organic phases, and K (= 42) is a constant of proportionality. This equation confirmed the measured value with a standard deviation of ±22%.

The new thermal-conductivity calorimeter for the rapid assay of Cs^{137} products has proved highly satisfactory for routine measurements.

The method of making cesium-glass melt material by crystallization of cesium tetraoxalate and calcination to the acid oxalate was tested and was found to be successful. When the glass was melted into 10 × 2 × 1/4 in. bars, however, some difficulties were encountered due to adhesion of the bar to the mold. Additional work will be required to eliminate this problem. The glass was analyzed as 51% cesium by calorimetry.

Radioisotope and Stable Isotope Production. — Ion currents equivalent to 2575 g of separated isotopes were monitored. Isotopes of Ca, Br, Ta, Yb, Si, Cd, and Pb were separated and collected, using 28 calutrons.

Several kilograms of PbO was prepared as a possible alternate charge to be used in the separation of lead isotopes. Operations with this material will be compared with those with fused anhydrous $PbCl_2$, the preferred charge material.

OPEN LITERATURE PUBLICATIONS

Alexeff, I., and R. V. Neidigh, "Observations of Ionic Sound Waves in Plasmas: Their Properties and Applications," Phys. Rev. 129, 516 (1963).

Ard, W. B., M. C. Becker, R. A. Dandl, H. O. Eason, A. C. England, and R. J. Kerr, "Neutron Production from a Steady-State, Hot-Electron Deuterium Plasma," Phys. Rev. Letters 10(3), 87 (1963).

Atta, Susie E., and J. A. Harvey, "Numerical Estimation of Neutron Resonance Parameters," J. Soc. Ind. Appl. Math. 10, 617 (1962).

Bassel, R. H., G. R. Satchler, R. M. Drisko, and E. Rost, "Analysis of the Inelastic Scattering of Alpha-Particles," Phys. Rev. 128, 2693 (1962).

Cameron, A. E., "The Determination of Atomic Weights by Mass Spectrometry," Anal. Chem. 35, 23A (1963).

Clayton, R. K., and P. S. Rudolph, "The Lethal Effects of Alpha Irradiation on Wild-Type and on a High-Catalase Mutant of Rheudopseudomonas spheroides," Radiation Res. 17, 839 (1962).

Danford, M. D., and H. A. Levy, "The Structure of Water at Room Temperature," J. Am. Chem. Soc. 84, 3965 (1962).

DeVan, J. H., and R. B. Evans III, "Corrosion Behavior of Reactor Materials in Fluoride Salt Mixtures," pp 557–79 in Conference on Corrosion of Reactor Materials, June 4–8, 1962, Proceedings, vol II, International Atomic Energy Agency, Vienna, 1962.

DiStefano, J. R., and E. E. Hoffman, "Relation Between Oxygen Distribution and Corrosion in Some Refractory Metal–Lithium Systems," pp 431–49 in Conference on Corrosion of Reactor Materials, June 4–8, 1962, Proceedings, vol II, International Atomic Energy Agency, Vienna, 1962.

Ernst, W. S., Jr., and R. L. Beatty, "Vibratory Compacted Studies at the Oak Ridge National Laboratory," Symposium on Powder Packed Uranium Dioxide Fuel Elements, November 30 and December 1, 1961, Nuclear Division, Combustion Engineering, Inc., Windsor, Conn., CEND-153, vol I, pp II-D-1–II-D-14 (1962).

Ferguson, D. E., E. P. Arnold, and W. S. Ernst, Jr., "Preparation and Fabrication of ThO_2 Fuels," pp 513–37 in Il Ciclo Combustibile U-Th, VI Congresso Nucleare di Roma, 13–15 giugno 1961, Comitate Nazionale Energia Nucleare, 1962.

Griffin, P. M., J. R. McNally, Jr., and G. K. Werner, "Ion Energy Evaluation from Spectrum Line Contours," pp 651–61 in Temperature – Its Measurement and Control in Science and Industry, vol 3, part 1 (ed. by F. G. Brickwedde), Reinhold, New York, 1962.

Grimes, W. R., G. M. Watson, J. H. DeVan, and R. B. Evans III, "Radio-Tracer Techniques in the Study of Corrosion by Molten Fluorides," pp 559–74 in Conference on the Use of Radioisotopes in the Physical Sciences and Industry, Sept. 6–17, 1960, Proceedings, vol III, International Atomic Energy Agency, Vienna, 1962.

Holz, P. P., C. M. Smith, and L. J. Shersky, "Cutting up the HRE-2 Core Vessel," Nucleonics 21(1), 72 (1963).

Ilakovac, K., L. G. Kuo, M. Petravic, I. Slaus, P. Tomas, and G. R. Satchler, "The Reaction $V^{51}(n,d)Ti^{50}$ at 14.4 Mev," Phys. Rev. 128, 2739 (1962).

Joy, H. W., "Integrals of Products of Associated Legendre Functions," J. Chem. Phys. 37, 3018 (1962).

Koehler, W. C., "A History of Neutron Diffraction," Proceedings of Advanced Course on Neutron Crystal Spectrometry (Joint Netherlands-Norway Reactor School, Kjeller, Norway, June 1962), vol 1, sec 1, Institute for Atomic Energy, Kjeller, Norway, 1962.

Koehler, W. C., "Applications of Neutron Diffraction to Investigations in Magnetism," Proceedings of Advanced Course on Neutron Crystal Spectrometry (Joint Netherlands-Norway Reactor School, Kjeller, Norway, June 1962), vol 1, sec 3, Institute for Atomic Energy, Kjeller, Norway, 1962.

Koehler, W. C., "Magnets and Cryostats," Proceedings of Advanced Course on Neutron Crystal Spectrometry (Joint Netherlands-Norway Reactor School, Kjeller, Norway, June 1962), vol 2, sec 4, Institute for Atomic Energy, Kjeller, Norway, 1962.

Koehler, W. C., "Mechanical Design Considerations for Neutron Diffractometers," Proceedings of Advanced Course on Neutron Crystal Spectrometry (Joint Netherlands-Norway Reactor School, Kjeller, Norway, June 1962), vol 2, sec 1, Institute for Atomic Energy, Kjeller, Norway, 1962.

Leibfried, G., "Higher Order Averages of Primary Recoil Distributions," Z. Physik 171, 1 (1962).

Maddox, W. L., "Remote Decapper," p 335 in Proceedings 10th Conference on Hot Laboratories and Equipment, American Nuclear Society, Chicago, 1962.

Potter, R. A., and L. A. Harris, "The Stability of $Ca_2Be_3O_5$ in the System BeO-CaO," J. Am. Ceram. Soc. 45, 615 (1962).

Roberts, L. D., and J. O. Thomson, "Solid State and Nuclear Results from a Mössbauer Type Study of Au^{197}," Phys. Rev. 129, 664 (1963).

Robinson, M. T., and O. S. Oen, "The Channeling of Energetic Atoms in Crystal Lattices," Appl. Phys. Letters 2, 30 (1963).

Smith, H. G., and H. A. Levy, "Neutron Diffraction Study of Ammonium Perchlorate," Acta Cryst. 15, 1201 (1962).

Snyder, W. S., "Major Sources of Error in Interpreting Urinalysis Data to Estimate the Body Burden of Pu^{239}: A Preliminary Study," Health Phys. 8, 767 (1962).

Snyder, W. S., and M. J. Cook, "Variability of Relative Organ Concentration of Trace Elements in Human Tissue," Health Phys. 9, 57 (1963).

Thurber, W. C., "Compacted Powder Fuel Irradiation Studies at the Oak Ridge National Laboratory," Symposium on Powder Packed Uranium Dioxide Fuel Elements, November 30 and December 1, 1961, Nuclear Division, Combustion Engineering, Inc., Windsor, Conn., CEND-153, vol II, pp IV-C-1–IV-C-27 (1962).

Thurber, W. C., and A. L. Lotts, "Development of Procedures and Equipment for Fabrication of HFIR Target Rods," pp 27–38 in Proceedings of the 10th Conference on Hot Laboratories and Equipment, ANS, Washington, Nov. 26–28, 1962, American Nuclear Society, Chicago, 1962; and Trans. Am. Nucl. Soc. 5, 298 (1962).

Zittel, H. E., and L. B. Dunlap, "Controlled-Potential Coulometric Titration of Uranium(IV) and Uranium(VI) in Sodium Tripolyphosphate Medium," Anal. Chem. 35, 125 (1963).

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