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PROGRESS RELATING TO CIVILIAN APPLICATIONS
DURING MARCH, 1959

by

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REPORTS RELATING TO CIVILIAN APPLICATIONS
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- BMI- 1317 "Oxidation and Contamination Reactions of Niobium and Niobium Alloys",
by William D. Klopp, Daniel J. Maykuth, Chester F. Sims, and
Robert I. Jaffee.
- BMI- 1319 "Postirradiation Examination and Evaluation of an OMRE Fuel Assembly",
by Richard J. Burian and John E. Gates.
- BMI- 1321 "Alumina-Clad UO_2 for Fuel Applications", by Allison K. Smalley,
William C. Riley, and Winston H. Duckworth.
- BMI- 1324 "Progress Relating to Civilian Applications During February, 1959",
by Russell W. Dayton and Clyde R. Tipton, Jr.
- BMI- 1326 "High-Temperature Mechanical Properties of Tantalum", by
David C. Drennen, M. Eugene Langston, Charles J. Slunder, and
Joseph G. Dunleavy.

A-1

A. ASSISTANCE TO HAPO

F. R. Shober

The thermal conductivities of irradiated uranium and UO_2 are to be measured. Modifications to adapt the thermal-conductivity measuring unit for hot-cell use have been made. A series of 15,000-hr creep tests is being performed at 290, 345, and 400 C on 15 per cent cold-worked Zircaloy-2. Shorter term tests are being conducted on annealed and cold-worked Zircaloy-2 under cyclic-temperature conditions. The preparation of molybdenum single crystals has not been successful on the present lot of material.

Development of a method of sink-float density measurement has continued. This technique is being used to identify and study factors affecting irradiation-induced volume changes in graphite. Preliminary results indicate that an irradiated sample contains less high-density material than a similar sample of unirradiated TS-GBF graphite. An evaluation of possible loss-of-coolant incidents in the PRTR by means of simulation on a digital computer has continued. The case of a double 14-in. top break has been run. Computation indicates that, for the time between rupture and when the element temperature has been reduced to coolant-water temperature, the release of fission products is not probable. A literature survey of methods for end capping coextruded fuel pins is being made as part of a program to evaluate end-closure techniques and brazing alloys.

Thermal Conductivity of Uranium and UO_2

C. F. Lucks and H. W. Deem

Uranium

The irradiated natural uranium specimens have been received at the Battelle's Hot-Cell Facility, but they have not been removed from the cask. Preparations for making thermal-conductivity measurements on the irradiated uranium are proceeding concurrently with those for the UO_2 measurements.

Uranium Oxide

Work has continued on changing the apparatus for hot-cell use. Most of the construction work is completed, but some fitting remains to be done.

During this work period, the irradiated specimens were received, and they appear to be sound. Specimens were weighed, and lengths and diameters were measured.

During April, the apparatus will be set up in the hot cells and thermal-conductivity measurements will be begun on the irradiated specimen with highest density.

Mechanical Properties of Zirconium Alloys

F. R. Shober and J. A. VanEcho

The creep properties of annealed and 15 per cent cold-worked Zircaloy-2 are being determined at elevated temperatures. Long-time creep tests (15,000 hr) are being conducted at 290, 345, and 400 C on 15 per cent cold-worked material. Both annealed and 15 per cent cold-worked Zircaloy-2 are also being tested under cyclic-temperature conditions. These conditions are obtained by maintaining a maximum temperature of either 290 or 345 C for 6 days and then reducing the temperature to room temperature for 1 day. This cycle is repeated until a total of 3000 hr has been attained. A third series of tests is being conducted on annealed Zircaloy-2 at 290, 345, and 400 C to compare the properties of annealed Zircaloy-2 with cold-worked material from the same lot and to compare the properties of two different lots of annealed material.

Twenty vacuum-creep machines are being utilized on this program. A comparison made between two tests conducted at 25,000 psi and 345 C, one on annealed and one on 15 per cent cold-worked material, indicates that cold work increases the creep strength of Zircaloy-2 considerably. The annealed material failed in about 860 hr, while the cold worked material has not failed in 12,000 hr. Cyclic-temperature conditions appear to accelerate the process of creep especially in the early stages of creep.

Preparation of Molybdenum Single Crystals

J. A. DeMastry, F. R. Shober, and R. F. Dickerson

Single crystals of high-purity molybdenum are being grown in a modified Andrade furnace for irradiation studies at HAPO. Growth of a single crystal is obtained through use of a steep temperature gradient along the length of the specimen. This temperature gradient is obtained by resistance heating the molybdenum rod and water cooling the ends of the rod. Six single crystals of molybdenum have been grown in 1/8-in.-diameter rod at a power input of 240 amp at approximately 3.0 v for 5 to 6 hr. It has been noted that single crystals are grown only from rods from certain lots of molybdenum. Apparently either the purity or the fabrication history of molybdenum influences the growth of single crystals. A new lot of molybdenum rods has been received, but conditions for growth of single crystals from this lot have not been established. Several unsuccessful runs have been made. Samples from four rods remain to be tested. If none of the four remaining rods shows satisfactory crystal growth, one or two specimens will be elongated about 2 per cent in a tensile machine in an attempt to induce a more favorable growth condition. Additional molybdenum rod has been ordered.

Upon completion of six more molybdenum single crystals, attempts to grow single crystals of alpha zirconium will be made.

A-3

Physical Distortion of Graphite

J. Koretzky, W. C. Riley, and W. H. Duckworth

Research to develop a method of sink-float density measurement to identify factors affecting irradiation-induced volume changes in graphite was continued.

During this month, an effort was made to refine the density-distribution curve by increasing the number of densities at which separations were made. Using minus 325-mesh graphite, density-distribution curves were not reproducible. Apparently, when a very small weight per cent of the sample should float, the fraction that is trapped by the large percentage of sink material becomes significant. Test results indicate that, for minus 325-mesh graphite, a number of separations of a given sample at a given liquid density are required to separate all of the float material.

The density distribution of a minus 270-mesh sample of irradiated TS-GBF graphite, designated as N19-232, was measured. Preliminary results indicate that the irradiated sample contains less high-density material than a similar sample of unirradiated TS-GBF graphite. This sample expanded slightly on irradiation.

Evaluation of Possible Loss-of-Coolant Incidents
in the Plutonium Recycle Test Reactor

C. A. Alexander, L. E. Hulbert, A. W. Lemmon, Jr.,
and R. B. Filbert, Jr.

The program to evaluate possible loss-of-coolant incidents in the PRTR by means of simulation on a digital computer was continued.

The first case, that of a double 14-in. top break, has been run from the time of rupture till the time when the fuel elements are essentially at the temperature of the coolant water. From the computations, it is considered that at no time during this (blowdown) phase is there any danger of release of fission products for either the uranium dioxide fuel element or the enriched plutonium-aluminum fuel element.

In order to complete the injection phase of this case, it is necessary to extend the blowdown prediction for the time required for the pressure to decay to at least 100 psia, since injection of emergency coolant cannot begin until the system pressure is below this value. An engineering analysis of this pressure decay is currently under way.

During the next month it is planned to complete the first two cases and begin the sensitivity analysis on the reaction rate of the steam-metal reaction.

End-Capping Methods for Coextruded Fuel Pins

R. M. Evans and J. J. Vagi

In the survey of end-capping methods for coextruded fuel pins, the literature search has been completed. The analysis of the literature and programming of techniques to be evaluated in the laboratory are in progress. The analysis of literature plus some discussions with appropriate processors should be finished early in April. After this, laboratory work will begin.

B-1

B. DEVELOPMENTS FOR ALUMINUM-CLAD FUEL ELEMENTS

N. E. Daniel and R. J. Carlson

Preparation of Aluminum-Uranium Alloys

N. E. Daniel, E. L. Foster, and R. F. Dickerson

Interest in aluminum-uranium alloys is reflected in current efforts to prepare more dense and improved fuels of this type. These alloys are usually utilized in the form of flat-plate fuel elements containing 20 w/o or less uranium clad in aluminum by the conventional picture-frame roll-cladding technique. However, certain reactor designers contemplate using tubular fuel elements of aluminum-uranium alloys containing greater concentrations of uranium. For this purpose it is necessary that the alloys be cast in relatively homogeneous hollow cylindrical tubes containing a minimum of porosity and suitable for extrusion cladding with aluminum. A program of research concerned with the development of casting techniques for the production of satisfactory extrusion billets is in progress. Another major portion of the program has been directed toward the development of aluminum-uranium alloys containing small additions of third elements which will enhance both the castability and the fabricability of the alloys.

Centrifugal-casting techniques have been utilized to produce sound extrusion billets up to 26 in. long from the aluminum-35 w/o uranium alloy. The evaluation of these castings is still in progress. Chemical-analysis samples have been obtained at points along the length of the castings and from material removed during machining operations. These analyses are necessary to aid in evaluating the material balance which has been obtained on the melts.

That portion of the program concerned with the improvement of casting and fabricating characteristics through the use of ternary additions has received the greatest emphasis during the past month. Extrusion studies and tensile tests on these alloys have been completed. Metallographic examination of the extruded material revealed that the alloys containing tin and zirconium had little or no microporosity as a result of fractures in the compound particles or in areas adjacent to the compounds. This was not true of the binary uranium-aluminum alloys, which exhibited microporosity from both types of fracturing. An examination of the results obtained from the tensile specimens revealed that silicon, tin, and zirconium increased both the elongation and the strength of the uranium-aluminum alloy. The greatest increase in elongation and reduction in area was noted in specimens containing 3 w/o tin. A visual examination of the fractures indicated that the binary alloys fractured in a manner typical of brittle materials with insignificant reductions in areas. Relatively large cleavage planes were evident in these specimens.

Full-size extrusion billets 26 in. long have been cast from the aluminum-35 w/o uranium-3 w/o tin and the aluminum-35 w/o uranium-3 w/o zirconium alloys. The extrusion billet containing the tin addition has very good external and internal surfaces with no visual evidence of porosity. These castings will be evaluated on the same basis as similar castings made from the binary alloys.

B-2

Future work will be confined to completing the above studies and evaluating the data obtained.

C-1

C. RADIOISOTOPE AND RADIATION APPLICATIONS

P. Schall

In the program on the use of radioisotopes for quality control in the cement industry, experiments were performed on the determination of calcium by EDTA titration using a radiometric endpoint and on sulfur analysis by radiometric titration with barium-140 as BaCl_2 . Modifications of the procedure for calcium analysis have yielded improved results but the precision of the method still requires further improvement. In the work on sulfur analysis emphasis is being placed on the separation of the lanthanum-140 daughter activity from the barium activity.

Work on the survey and evaluation of various industrial processes which might utilize intrinsic radioisotopes in process control has been completed. A list of processes has been prepared from which specific processes will be selected for laboratory development.

Irradiation and analysis of inclusion compounds of urea-hydrocarbon and ammonia nickel cyanide-benzene complexes are continuing.

A program on radiation-induced graft polymerization was begun. Initial experiments on the study of factors affecting the formation of grafting sites were completed and further work along these lines is planned for next month.

Development of Radioactive-Tracer
Quality-Control Systems

J. E. Howes, T. S. Elleman, C. T. Brown,
D. N. Sunderman, and M. Pobereskin

The study of the radiometric analysis of calcium based upon EDTA titration with a precipitate of tagged $\text{Ag}^*\text{IO}_3^{(a)}$ as indicator has been continued. Better precision has been obtained through restandardization of the reagents and correction of the titration plots for small amounts of silver-110 complexed before the calcium endpoint is reached. The results of four analyses for calcium are summarized in Table C-1.

TABLE C-1. PRECISION OF CALCIUM ANALYSIS USING A RADIOMETRIC ENDPOINT

Calcium Concentration, mg per ml				Absolute Error, per cent
Theoretical	Experimental	Experimental Average	Standard Deviation	
9.90	9.70			
	10.00			
	9.80			
	10.02	9.88	± 0.160	0.20

(a) In the nomenclature used herein the tagged (*) element in the compound is silver, hence tagged silver iodate is Ag^*IO_3 .

Although the accuracy of the average value is good the precision of the method is in need of improvement.

Work has begun on the determination of sulfate by precipitation as BaSO_4 with BaCl_2 solution labeled with barium-140. The separation of the lanthanum-140 daughter activity is essential since the BaCl_2 solution must be standardized in terms of the barium-140 activity alone. The lanthanum-140 activity will remain in solution as the BaSO_4 is precipitated. A pure barium-140 assay was attempted by precipitating BaSO_4 from a solution of BaCl_2 containing some $\text{La}(\text{NO}_3)_3$ holdback carrier and redissolving the BaSO_4 in alkaline EDTA solution. Only a partial separation was effected in this manner.

Continuous-separation procedures are being investigated in which a small amount of ion-exchange resin would be incorporated in the titration burette. After equilibrium with barium and lanthanum on the resin has been reached, continuous lanthanum separation during the actual radiometric titration should be possible.

Other future work will include further studies on calcium analysis, conclusion of the precision and interference-element studies in the magnesium analysis, and standardization of the activation-analysis technique. Activation analyses of cement raw materials such as limestone and clay will also be performed.

Use of Intrinsic Radioactive Tracers for Process Control

J. L. McFarling, R. B. Price, R. H. Barnes,
D. N. Sunderman, and M. Pobereskin

During the current report period the evaluation of candidate processes for intrinsic radiotracer control has been essentially completed.

Based on the concept of a generalized "ideal" process, the criteria used to evaluate the various industrial processes under consideration are the following:

- (1) The product made in the process should be valuable and commercially important.
- (2) The nature of the process is such that suitable radiotracers can be used to control it.
- (3) The process is difficult or expensive to control by conventional means and the use of radiotracer control would be economically advantageous.
- (4) Application of radioisotopes will involve no undue hazards to operating personnel.
- (5) The product does not become excessively contaminated with radioactivity. In every case the amount of radioactivity in the product is well below

C-3

accepted tolerance levels; and, in the case of direct consumer use, essentially no radioactivity remains in the product.

The aim of the initial phase of this program has been to find a process application which fulfills these criteria. Several processes show promise at this stage of the evaluation, although there is no obvious best choice. The reason for this lies in the impossibility of closely evaluating certain economic and technical suspects of the various processes at this time.

A list of processes has been made up consisting of representative types of radiotracer applications having some chance of success. These process applications are as follows:

- (1) Use of radiocopper in the separation of copper from nickel in a hydro-metallurgical nickel-refining process
- (2) Use of radiosodium to control the chemical recovery in the sulfate pulp process
- (3) Use of a suitable tracer such as sodium-24 to control uniformity and thickness of a physical coating process such as corrugated paperboard adhesive
- (4) Use of radiochlorine to control production and reactions of chlorinated hydrocarbon intermediates
- (5) Use of radiosulfur to control production and reactions of sulfonated hydrocarbon intermediates
- (6) Use of intrinsically labeled impurities in purification processes such as removal of metal-catalyst residue from polymers
- (7) Use of radiosilver in the manufacture of phosphors
- (8) Use of radiosulfur in fuel-gas purification
- (9) Use of suitable tracers in waste streams for automatic and continuous monitoring
- (10) Use of radiocarbon in manufacture of nitrated or oxygenated hydrocarbons such as formic acid derivatives
- (11) Use of radiocarbon to separate closely similar hydrocarbon fractions on a fractionation column.

It is expected that the final decision regarding a process to be studied further will be decided in the near future. Following this, the bench-scale study of the process chosen will begin.

Radiation Chemistry of Inclusion Compounds

M. J. Oestmann, M. J. Landon, E. J. Kahler,
and W. S. Diethorn

Irradiated urea complexes of dodecane and octane are being examined in a continuing study of the radiation chemistry of urea-hydrocarbon complexes.

Chemical analysis of a 400-g sample of urea-dodecane complex irradiated to 2×10^8 rads is in progress. Analytical results will be reported as soon as they become available. A 300-g sample of urea-octane complex has been prepared and is now being irradiated in the Battelle Cobalt-60 Facility.

A small sample of the nickel complex, described last month, was irradiated to 1×10^8 rads, and a cursory examination of the radiolysis gas was completed. Mass-spectrometric analysis of the radiolysis gas shows 56 volume per cent hydrogen, 13 volume per cent methane, 10 volume per cent methyl alcohol, 6 volume per cent acetone, 4 volume per cent alcohol, 3 volume per cent benzene, and a few per cent of nitrogen, oxygen, carbon dioxide, and ethylene. No hydrogen cyanide, cyanogen, or ammonia was found. During the synthesis of the complex, alcohol and acetone were used to wash the sample and this is the source of these two contaminants. The irradiated complex is being analyzed and results will be reported next month. Analytical techniques developed with this small sample will be used to analyze a larger sample which is currently being irradiated.

Graft-Polymerization Studies

I. S. Ungar, J. F. Kircher, R. A. Markle,
and R. I. Leininger

This is the first monthly report on the project entitled "A Study of Radiation-Induced Changes in Polymers Leading to Graft Copolymerization as Influenced by Structural Factors", which was initiated in late February.

The objective of the research is to determine the structural and compositional factors influencing (1) radiation effects in polymer systems, (2) mechanisms of polymerization reactions, and (3) the products of polymerization reactions.

Initial work has been directed toward assembling the necessary apparatus and performing preliminary irradiation experiments in the Battelle Cobalt-60 Facility. Initial experiments on the frozen free-radical method of grafting showed that the equilibrium concentration of free radicals was attained in polymethyl methacrylate at an irradiation period of about 2 hr at room temperature and dose rate of 3.5×10^5 rads per hr. The equilibrium concentration for a prototype polymer with a molecular weight of approximately 100,000 was only about seven free radicals formed per polymer molecule. This is not a large enough free-radical concentration to provide the desired flexibility in this work.

C-5 and C-6

Further experiments are planned under conditions which are expected to increase the free-radical concentration. If this is not successful, work will begin on the peroxy method wherein oxygen is passed through a polymer solution during irradiation. This method is expected to form a much larger concentration of polymerization sites because the greater stability of the peroxide sites will result in a higher equilibrium concentration. Longer irradiation may be required to reach this higher concentration. The peroxide groups formed are thought to provide polymerization sites just as do the frozen free radicals.

D-1

D. PROCESSING OF FEED MATERIALS

E. L. Foster

National Lead Company of Ohio (NLO), as the operator of the Feed Materials Production Center, is concerned with the quality of uranium fuel slugs. One of the processing steps that contributes to the qualities of the fuel slugs is the vacuum remelt and casting operation. Because direct experiments with the large ingots presently produced would be cumbersome and costly, NLO is seeking a convenient method of evaluating the factors present in the solidification of these uranium ingots. It is anticipated that if such a method is found it will be applicable to future casting needs.

Solidification of Uranium

E. L. Foster, C. K. Franklin, B. L. Fletcher,
A. B. Pritsker, and R. F. Dickerson

A study of the solidification of uranium cast in cylindrical graphite molds is being made. A mathematical model has been formulated to compute the movement of isotherms with time and predict the progress of solidification within the casting. In the development of the model, some of the thermal properties of the materials involved and their changes with temperature were determined experimentally. A 100-lb casting has been poured with a network of electrical probes in the mold to study the physical events occurring at the mold-metal interface as the ingot cools. Indications of gap formation between the surfaces were observed shortly after pour. Melts were also poured to measure the temperatures of the mold, metal, and furnace from time of pour. The thermal data obtained experimentally will be used to refine and check the data computed from the mathematical model.

Ingots 3 in. in diameter by 20 in. long were cast to obtain temperature histories of the mold and ingot. Using vacuum-induction procedures, the metal was melted and poured into graphite molds coated with $\text{MgO} \cdot \text{ZrO}_2$ wash. A pouring temperature of 2550 F was used and each melt was held at temperature for 5 min prior to pour. The temperature measurements show that the ingot completely solidifies 60 sec after the initiation of pour. The maximum temperature recorded along the center of the ingot was 2300 F at 10 sec after start of pour, indicating that some loss of heat to the surroundings occurs as the metal fills the mold. The maximum temperature drop across the ingot at any time was 750 F. This maximum occurred at about 12 sec from pour, indicating that superheat was being removed rapidly. Corresponding mold temperatures were 1043 F at the inner surface and 380 F at the outside surface. Virtually no rise in the furnace-jacket temperature was observed during the freezing time.

Two preliminary computer runs have been made to attempt correlation with these small-scale casting experiments. In the first computer run, complete solidification was indicated at 118 sec, a time considerably greater than the value of 60 sec obtained from experimental data. A second computer run contained an arbitrary doubling of the rate of heat loss by radiation from the casting wall. This was done to see what the effect

would be of adopting a rate of heat transfer somewhere between that of pure conduction and radiation. The over-all solidification time was observed to decrease to 83 sec.

The mathematical model used in these two runs assumes gap formation to occur when the solid-metal wall thickness reaches a value of 0.16 in. This value was arrived at by sizing the cells to permit the use of the greatest possible time interval in determining the incremental changes of temperature; this is essential for reducing computer running time. The first opinion adopted as to the actual sequence of events was that "virtual" separation of metal and mold would occur so fast after initial contact that the time during which this contact existed would be insignificant in comparison with the over-all solidification time. Although this assumption may be true for a larger ingot, it appears that at least a partial contact with the mold wall existed in 3-in.-diameter by 20-in.-long ingots until a greater depth was solidified. Also, the wall thickness of solidified metal required for holding the casting away from the mold wall is probably a function of the distance from the top because of the pressure exerted by the liquid metal. This is not considered in the present mathematical model.

The next computer runs to be made will be made assuming a greater wall thickness. Difficulties with this procedure may be experienced in the form of excessively long computer running times, because alterations in cell sizes may adversely affect the time interval required for meeting convergence and stability criteria. The wall thickness picked will be determined by relating the gap-formation times and the thermocouple data at a point 10 in. from the bottom of the experimental ingots. Unfortunately, the temperature response of the thermocouples masks the sequence of events during the early part of the solidification process near the outer wall of the casting; and this implies some severe approximations in estimating the wall thickness at the time that separation occurs.

Another undetermined factor of interest is the thermal conductivity of the liquid metal. Determination of this quantity is being undertaken at present, and future computer analyses are to utilize this value. If this value is below that of the solid metal at 1133 C, it will have an adverse effect on the over-all solidification time (it will increase), but if the value is above that for the solid, it will influence the time in the correct direction.

A program similar to the small pilot-ingot studies is to be conducted in the near future on production-size castings.

E-1 and E-2

E. SPRAY DEPOSITION OF CALCIUM METAL ON NICKEL OR INCONEL

R. M. Evans and R. E. Monroe

The feasibility of arc spraying calcium to form a coating on nickel and nickel-base alloys is being studied.

Spraying of calcium was accomplished during this report period. A number of difficulties were encountered, however, before this was done. The major problem was attributed to oxidation of the wire before spraying. Oxide on the wire contributes to a very unstable arc and also prevents good electrical contact with the power source. The wire is now abrasive cleaned and kept in an argon atmosphere at all times. During successful spraying, considerable white "oxide" dust also causes difficulty. The coating obtained is metallic but is not considered satisfactory for the intended purpose.

Work during the next period will include improvement of the technique and development of suitable coatings of controlled thickness.

F-1

F. RESEARCH FOR AEC REACTOR DEVELOPMENT DIVISION PROGRAM

S. J. Paprocki and R. F. Dickerson

REACTOR MATERIALS AND COMPONENTS

R. F. Dickerson

Studies aimed at reducing the amount of additive needed to stabilize UO_2 have been continued. Additions of 30 mole per cent La_2O_3 and 10 mole per cent CaO result in a reasonably stable body after 20 hr at 3200 F; however, this composition exhibits a weight loss not evident with the binary additions of Y_2O_3 or La_2O_3 . More detailed study of the complex structures of UO_2 - La_2O_3 - CaO and UO_2 - Y_2O_3 - CaO combinations are in process. Calibration of the high-temperature high-pressure die is in progress. Bismuth and thallium transitions at 25,000 atm and 44,000 atm, respectively, have been used for the low pressure range. The barium transition will be used for slightly higher pressure calibration at 80,000 atm. Higher pressures were obtained during the calibration runs.

The first of two capsules containing zirconium-2 w/o uranium alloy hydride specimens has been discharged from the reactor. This capsule is being stored at the reactor until the second capsule is discharged at the end of April. The first capsule was irradiated at temperatures slightly lower than the design temperature of 1500 F because of lower-than-estimated flux levels. The second capsule has operated close to its design temperature of 1100 F.

The eight capsules containing Type 347 stainless steel which are being irradiated in a fast-neutron flux at the ETR have been moved to a specially prepared position which should produce the fast-neutron flux of about $3.0 \times 10^{14} \text{ n}/(\text{cm}^2)(\text{sec})$ needed to maintain capsule lead time over the test loops. Nickel dosimeters exposed during previous cycles have been analyzed. A lead capsule designed for gamma heat has been constructed and sent to the ETR for the purpose of correlating capsule temperatures during irradiation with the gamma flux.

Alloys of niobium-1 w/o chromium, niobium-2 w/o chromium, niobium-4.5 w/o zirconium, niobium-10 w/o tantalum-2 w/o chromium, niobium-20 w/o titanium-1.5 w/o chromium, and niobium-40 w/o titanium-10 w/o aluminum are being investigated as cladding materials for possible use in future core designs of the EBR. All alloys except the niobium-2 w/o chromium alloy have been successfully fabricated. Examinations of the microstructures indicate that the niobium-chromium alloys may be precipitation hardenable.

The program concerned with the evaluation of selected niobium-base alloys for possible service in pressurized water was continued. Of the alloys tested, the 45 a/o zirconium binary and the 25.5 a/o titanium-6 a/o chromium ternary continue to be the most corrosion resistant of the alloys tested. Corrosion studies are under way on alloys

prepared from high-purity niobium, and tensile properties of ternary alloys are being determined. Large-scale heats of some alloys have been prepared, and a number of other alloys have been prepared in 40 to 50-g ingots.

The program concerned with investigation and wear behavior of rubbing surfaces lubricated by high-temperature sodium has been resumed after a brief discontinuance. The first phase of the work resulted in the design and construction of a special friction apparatus, which was used to obtain the coefficient of friction for several materials. These initial tests indicated the need to study surface films and their effect; therefore, this portion of the program will be devoted to such a study. Friction experiments will continue, and attempts will be made to identify the oxide films which may have formed on the molybdenum by electron-diffraction techniques.

Valence Effects of Oxide Additions to Uranium Dioxide

W. B. Wilson, A. F. Gerdts, and C. M. Schwartz

An investigation is being made of the possibility of reducing the large amount of additive required to achieve stabilization of uranium oxide. Previous work had shown that approximately 60 mole per cent of La_2O_3 or Y_2O_3 was required to form solid solutions which were stable and had low volatility in high-temperature air. The present approach has been to substitute CaO in part for La_2O_3 or Y_2O_3 in order to effect an over-all reduction in total amount of additive.

A direct test was performed to evaluate the relative performance of UO_2 -55 mole per cent La_2O_3 , UO_2 -60 mole per cent Y_2O_3 , and UO_2 -30 mole per cent La_2O_3 -10 mole per cent CaO . These materials were placed on disks of BeO and air fired to 3200 F for periods ranging from 1 to 20 hr. All of these specimens showed little tendency to fracture or powder even during the 20-hr exposure at 3200 F, although other compositions were pyrophoric even at room temperature. The relative weight changes of these compositions are tabulated as follows:

Nominal Addition to UO_2 , mole per cent	Total Weight Gain ^(a) at Indicated Time, w/o		
	1 Hr	5 Hr	20 Hr
53.3 La_2O_3	2.22	1.17	6.8
60.0 Y_2O_3	1.22	1.8	1.8
30 La_2O_3 -10 CaO	-0.17	-3.99	-7.1

(a) Minus sign indicates weight loss.

The tabulation shows that the 30 mole per cent La_2O_3 -10 mole per cent CaO composition exhibited fairly large weight loss by comparison with the La_2O_3 and Y_2O_3 materials, suggesting that more additive is required. The weight gain exhibited by the Y_2O_3 and La_2O_3 materials generally results when the weight pickup due to oxidation exceeds the loss due to volatility.

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More detailed structural analysis of the $\text{UO}_2\text{-La}_2\text{O}_3\text{-CaO}$ and $\text{UO}_2\text{-Y}_2\text{O}_3\text{-CaO}$ compositions indicates that a greater complexity exists than previously reported in the preliminary work. Discussion of the X-ray diffraction analysis and the relationship between the structure and oxidation behavior will, however, be deferred until reliable information is available.

High-Pressure High-Temperature Solid-State Studies

W. B. Wilson and C. M. Schwartz

The purpose of this program is to investigate the feasibility of synthesizing new materials through the combined use of ultrahigh pressure and high temperature. Accomplishment of the objectives requires the development of equipment, as well as the use of such equipment in materials studies.

A new die has been developed which had been previously tested and used routinely to 60,000 atm at temperatures to 1500 C with platinum heaters. This unit was moved to a larger press in order to determine its pressure capabilities. The previous calibration work had repeatedly detected the bismuth and thallium transitions at 25,000 and 44,000 atm, respectively. It was desired to obtain the barium transition at 80,000 atm. At the same time, strain gages were installed to provide information as to when significant stresses were developed.

A number of samples have been tested using lava, pyrophyllite, and silver chloride as pressure-transmitting media. The effects of sample and insulation height were also examined, and the electrical resistance of barium was measured to determine if the transition occurred. The sample compression was measured through the use of a dial indicator.

The experimental results obtained indicate that in an internally heated die a composite resistance is measured. Under ordinary circumstances the resistance of barium increases with pressure. At 80,000 atm a slight, discontinuous increase is observed, followed by a more moderate increase beyond 80,000 atm. In the present die this behavior is somewhat altered since the barium resistance is paralleled with the die insulation resistance, which also varies under pressure. In nearly all samples tested an increase in resistance was observed to occur with a discontinuous break at a press force of 192 tons, followed by a decrease.

When the barium, bismuth, and thallium transistors are plotted against press force a linear line is obtained to relate press force to pressure. In addition, by extrapolating the Bridgman data for the compression of AgCl to 80,000 atm a correlation may be obtained with the change in internal volume of the die as indicated by the dial indicator. The agreement between the measured and computed volume change and the transition data is entirely satisfactory.

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Pressures beyond 125,000 atm are believed to have been obtained during calibrating. This was accomplished using 266 tons of press force. The unit is estimated to be capable of withstanding 400 tons of press force. The stresses in the die as indicated by the strain gages were relatively low.

Work will continue to study, at higher pressures and temperature, the reactions and the U_3O_8 phase diagram previously discussed. The satisfactory results obtained in the use of the die indicate that further die-development work may be sharply curtailed or discontinued on this project.

Fueled Zirconium Hydride Moderator

H. E. Bigony, A. K. Hopkins, and H. H. Krause

The experimental investigation of the phase diagrams of the uranium-zirconium alloy hydrides containing 1 w/o and 25 w/o uranium has been completed. One of the capsules containing 2 w/o uranium alloy hydride has been removed from the MTR, and the second capsule will be removed during April. Effects of the irradiation on the specimens will then be determined.

Structure and Pressure-Composition-Temperature Studies

The hydrogen-absorption isotherms required to characterize the uranium-zirconium alloy hydrides containing 1 w/o and 25 w/o uranium have been completed, and the data obtained have been processed to establish the location of phase boundaries in these systems.

Identification of the phases present has been made by means of high-temperature X-ray diffraction. Analysis of the X-ray data is very nearly completed.

Radiation-Damage Studies

Capsule BMI-20-1 was discharged from the MTR during Cycle 119 shutdown. Table F-1 presents a condensed irradiation history for this capsule, which was designed to maintain 2 w/o uranium-zirconium alloy hydride specimens at a temperature of 1500 F in an unperturbed thermal-neutron flux of 0.92×10^{14} nv. While the recorded specimen temperatures display some anomalies on the basis of reactor-quoted fluxes, it appears generally that the capsule was irradiated in lower-than-specified fluxes, and that this is the principal reason the specimens did not achieve design temperature.

Capsule BMI-20-2, which is identical in design to BMI-20-1, will be discharged from the reactor during Cycle 121 shutdown (April 27, 1959). It is estimated that, at discharge, the fission burnup levels will be as follows: 20.8 per cent uranium-235 for the bottom specimen and 14.7 per cent for the top specimen. Temperatures in Capsule BMI-20-2 have been consistent during recent cycles, with the bottom specimen at about

TABLE F-1. IRRADIATION HISTORY FOR CAPSULE BMI 20-1^(a)

(Capsule was located in MTR Position A30SE)

MTR Cycle	Capsule Position From Top of Reflector, in.		Reactor-Estimated Flux at Center Plane of Specimens, 10^{14} nv		Specimen-Temperature Range During Major Part of Cycle, F	
	Capsule Bottom	Capsule Top	Bottom Specimen	Top Specimen	Bottom Specimen	Top Specimen
112	15	2	Not available		1430-1370	1060-960
113	15	2	0.73	0.45	1320-1320	980-1030
114	18	5	0.82	0.65	1360-1340	1120-1150
115	15	2	0.53	0.36	1300-1280	1180-1210
116	16-1/2	3-1/2	0.65	0.45	1240-1100	1260-1240
117	16-1/2	3-1/2	0.65	0.45	1080-1050	1240-1250
118(b)	25-1/2	12-1/2	0.78	0.70	1025-1030(c)	1190-1200(c)

(a) This capsule contains three cylindrical specimens (3/16 in. in diameter by 1 in. long) aligned along the capsule center line. The top and bottom specimens are hollowed out along a portion of their length and the temperatures listed are those maintained by thermocouples inserted directly into the hollows.

(b) Estimated fission burnups at discharge (end of Cycle 118) are as follows: 18.4 per cent uranium-235 for the bottom specimen and 13.7 per cent for the top specimen.

(c) Spot checks as reported from MTR. Entire temperature chart for Cycle 118 not yet available at Battelle.

980 F and the top specimen at about 1150 F. The design temperature for this capsule is 1100 F in an unperturbed flux of 0.73×10^{14} nv. The peak reactor-quoted flux for Position A43SE, which has been used for this irradiation, is quite close to this specified level.

Capsule BMI-20-1 is being stored at the reactor pending discharge of the second capsule. They will be returned at the same time to the Battelle Hot-Cell Facility for postirradiation examinations.

Irradiation Surveillance Program on Type 347 Stainless Steel

J. E. Howes, Jr., J. F. Lagedrost, and F. R. Shober

Investigation of the changes in the mechanical properties of AISI Type 347 stainless steel as affected by exposure to fast-neutron flux (neutrons have energies greater than 1 Mev) at 120 and 600 F is being conducted. These changes are being determined and evaluated in support of the safe operation of loops containing Type 347 stainless steel in the ETR. The tensile, cyclic-strain fatigue, and impact properties are to be measured after each period of exposure representing 6 months of service in the F-10 position of the ETR. Exposure to a total flux comparable to 3 years of exposure of a tube in the F-10 position is the maximum expected, approximately 1.2 to 1.6×10^{22} n per cm^2 , total neutron exposure.

The "cold" capsules being exposed at ETR process-water temperatures have been transferred from the K-7 to the K-8 and L-8 positions. The nickel dosimeter wire exposed in the F-10 position during Cycles 10 and 11 and those exposed in K-7 during Cycle 12 have been analyzed. A thermocouple lead capsule, BMI-24-19, to be irradiated at approximately 600 F, and a nonlead capsule, BMI-24-21, have been prepared and shipped to the ETR.

Because of the low fast flux in Position K-7, 1.2×10^{14} n/(cm^2)(sec), the eight "cold" capsules being irradiated at process-water temperature have been transferred and are now located as follows: K-8-NW contains BMI-24-14 and BMI-24-16, K-8-SE contains BMI-24-4, BMI-24-6, and BMI-24-10, K-8-NE contains BMI-24-2 and BMI-24-6, and L-8-SE has BMI-24-12. These capsules have been loaded to take advantage of the highest fast flux available in the positions. Fuel has been loaded into the L-7 and M-7 positions, and it is expected that a flux somewhat greater than 1.2×10^{14} n/(cm^2)(sec) will be obtained. It has been calculated that a fast flux of approximately 3.0×10^{14} n/(cm^2)(sec) will be necessary to maintain sufficient lead over the in-pile tube. The total accumulated fast-flux exposures to date for the eight cold capsules are given in Table F-2, along with approximate discharge dates.

Five dosimeter assemblies exposed at the ETR in the F-10 and K-7 positions during Cycles 10, 11, and 12 have been analyzed to determine the fast flux in those positions. Only the nickel wires have been analyzed. Decomposition of the ammonium sulfate during irradiation has delayed analysis of these dosimeters. Evaluation of these dosimeters will be made at a later date. The results obtained from the nickel wires indicate a

TABLE F-2. ACCUMULATIVE FAST FLUX ON "COLD" CAPSULES OF THE 347 STAINLESS STEEL IRRADIATION DAMAGE PROGRAM

Capsule	Capsule Type	Approximate Removal Date(a)	Approximate Exposure at Time of Removal(b), 10^{22} nvt	Total Exposure as of March 13, 1959, 10^{21} nvt	
				Top	Bottom
BMI-24-2	Tensile and fatigue	January, 1962	1.31	1.447	2.225
BMI-24-4	Tensile and fatigue	January, 1963	1.78	0.422	1.477
BMI-24-6	Tensile and fatigue	June, 1961	1.08	2.303	1.658
BMI-24-8	Tensile and fatigue	June, 1962	1.54	0.508	1.588
BMI-24-10	Tensile and fatigue	January, 1961	0.84	1.651	2.229
BMI-24-12	Tensile and fatigue	June, 1960	0.61	2.337	1.634
BMI-24-14	Impact	June, 1962	1.54	2.216	2.256
BMI-24-16	Impact	June, 1960	0.61	2.229	2.293

(a) Based on 6-month lead on loop, plus 2 months for tests.

(b) Based on maximum fast flux at tube of 1.7×10^{14} nv for 8-month periods, 6-month lead time on tube and 2-month time for examination.

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somewhat lower value for the fast flux (greater than 1 Mev) than that reported previously for Cycle 10. Two samples have been sent to the ETR for analysis there.

Dosimetry planned for Cycle 13 includes eight nickel wires in one face of the core-filler piece containing the cold capsules in the K-8 position and one each at the midplane on the other three faces. The wires will be returned to Battelle for analysis after the completion of Cycle 13. It is also planned to expose a full-length nickel wire in the center hole. This will be scanned at the ETR. A thermocouple lead capsule, BMI-34-19, has been completed and shipped to the ETR. It is planned to insert this capsule in a position having a measured gamma flux of approximately 20 w per g and to attempt to correlate temperatures of the capsule during irradiation with the gamma flux. This is necessary to establish the temperature at which the "hot" capsules will be irradiated. The lead capsule has been designed to operate at 600 ± 100 F in a gamma flux of 25 w per g. Irradiation at 600 F more realistically represents the condition under which the tube is exposed. It is thought that some damage caused by irradiation may be annealed out at 600 F, and the ductility of the Type 347 stainless steel would not be impaired to as great an extent during irradiation at this temperature. Changes of mechanical properties under such conditions of irradiation would be more meaningful than those changes measured after irradiation at 120 F. It has also been suggested that a postirradiation anneal at 600 F of material irradiated at 120 F would produce a material more closely representing that of the tube after irradiation.

Development of Niobium-Base Alloys

E. Jablonowski, F. R. Shober, and R. F. Dickerson

Utilization of a niobium-base alloy as an alternate cladding material in future core designs of the EBR has prompted a study of the fabricability, weldability, thermal conductivity, and mechanical strength of several niobium-base alloys. Alloys selected for study include niobium-1 w/o chromium, niobium-2 w/o chromium, niobium-4.5 w/o zirconium, niobium-10 w/o tantalum-2 w/o chromium, niobium-20 w/o titanium-1.5 w/o chromium, niobium-40 w/o titanium-10 w/o aluminum, unalloyed niobium, and a comparison alloy of vanadium-10 w/o titanium-1 w/o niobium. Melting and hot fabrication of the alloys was concluded during the month. The cold fabricability, metallography, and heat treatability of the alloys are being

A small rectangular specimen of each alloy was cut from ingots which were upset hammer forged at 1370 C. The resultant specimens were flat rolled at room temperature. The niobium-2 w/o chromium and niobium-tantalum-chromium specimens were completely broken up after 5 per cent reduction. The remaining alloys were reduced 70 to 80 per cent to sound 0.035-in. sheet with a minimum amount of edge cracking.

The microstructure of the hot-worked alloys was observed at 500X. A finely dispersed second phase was found in the niobium-zirconium alloy and in the vanadium-titanium-niobium alloy. Niobium and the remaining alloys exhibited very clean single-phase microstructures. An indication of the recrystallization temperature of the alloys was given by their relative grain size; the niobium-titanium-aluminum and niobium-titanium-chromium alloys possessed the largest grain size, while the niobium-tantalum-chromium and niobium-2 w/o chromium alloys showed a very fine grain size.

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The niobium-chromium equilibrium phase diagram supports the presence of two room-temperature phases, niobium-chromium solid solution phase and NbCr_2 , in four of the experimental alloys containing chromium. The absence of NbCr_2 in metallographic examinations suggested that chromium had been retained in solid solution because of air quenching the hot-forged alloys. This desirable effect may be exploited with respect to the development of high-strength precipitation-hardenable alloys. Specimens of the alloys containing chromium were solution heat treated at 1500 C for 1/2 hr and rapidly cooled. It is expected that precipitation of NbCr_2 can be promoted by annealing at 900 to 1000 C. The hardness of solution-treated and annealed alloys will be determined and used as a measure of the precipitation-hardening characteristics of the niobium-chromium alloys. Precipitation-hardening anneals of the alloys at 900 C are being performed.

Chemical analyses of all fabricated alloys will be reported next month. Tensile and thermal-conductivity specimens will also be prepared.

Development of Corrosion-Resistant Niobium Alloys

D. J. Maykuth, W. D. Klopp, R. I. Jaffee, W. E. Berry, and F. W. Fink

The evaluation of selected niobium-base alloys for possible service in pressurized-water reactors was continued. Mechanical-property data are being accumulated on high-purity base alloys which were chosen on the basis of hot-water corrosion tests of a series of commercial-purity niobium-base alloys.

The commercial-purity niobium alloys have been exposed a total of 112 days in 680 F water. The trends in corrosion behavior are the same as those reported in detail in BMI-1324 for 84 days of exposure. These trends are:

- (1) Unalloyed niobium initially gains weight; begins to lose weight after 7 days of exposure, and subsequently exhibits a linear weight loss of $0.2 \text{ mg}/(\text{cm}^2)(\text{hr})$.
- (2) Binary alloys containing 2.5 a/o molybdenum and 5 or 10 a/o tungsten also possess adherent films initially and then begin to lose weight. The 10 a/o tungsten alloy failed by cracking after 14 days of exposure. The linear weight losses for the 2.5 a/o molybdenum and 5 a/o tungsten alloys after 112 days of exposure are 0.0012 and $0.0067 \text{ mg}/(\text{cm}^2)(\text{hr})$, respectively.
- (3) A large number of alloys exhibit relatively adherent dark gray films and total weight changes ranging from -0.5 to $+0.5 \text{ mg per cm}^2$ after 112 days of exposure. Alloys in this group are:
 - (a) Binaries
 - 10, 25, and 35 a/o zirconium
 - 5 and 7.5 a/o molybdenum

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5, 7.5, 10, 12.5, 17.5, and 25 a/o vanadium

5 a/o iron

10, 20, 25, 30, and 35 a/o titanium

(b) Ternaries

7.5 a/o titanium -0.5 a/o chromium

19.5 a/o titanium -2.5 a/o chromium

10.5 a/o titanium -3 a/o molybdenum

16.5 a/o titanium -4.5 a/o molybdenum

21.5 a/o titanium -6 a/o molybdenum

10.5 a/o titanium -3 a/o vanadium

17 a/o titanium -6.5 a/o vanadium

24 a/o titanium -11 a/o vanadium

- (4) The 45 a/o zirconium binary and 25.5 a/o titanium-6 a/o chromium ternary continue to be the most corrosion resistant of the alloys tested, exhibiting adherent tarnish films and total weight gains of 0.37 and 0.04 mg per cm², respectively.

Corrosion studies are now under way on alloys prepared from high-purity niobium. Binary alloys containing 7.18 a/o molybdenum and 12.58 a/o vanadium and ternary alloys containing zirconium-titanium and titanium-molybdenum are being tested in 600 and 680 F water and 750 F 1500-psi steam. The results after exposure times up to 42 days are presented in Table F-3. To date, the 12.58 a/o vanadium alloy and all the ternary alloys exhibit adherent tarnish films at all test temperatures. The reaction rates of the 12.58 a/o vanadium alloy at the three test temperatures appear to follow the cubic rate law, while those of the 7.18 a/o molybdenum alloy follow the parabolic rate law. There are not sufficient data as yet to establish the reaction rate for the other alloy samples.

Room- and elevated-temperature tensile properties of ternary alloys containing 45 a/o zirconium-5 a/o titanium and 10 a/o titanium-3 a/o molybdenum were determined as shown in Table F-4. As might be expected, the more highly alloyed zirconium-titanium alloy is considerably stronger and less ductile at all temperatures than the titanium-molybdenum alloy.

The results of vacuum creep tests at 1200 and 1380 F on these and other alloys are summarized in Table F-5. These data indicate that the 12.58 a/o vanadium alloy is still the most creep resistant of all the alloys thus far tested. The 10 a/o titanium-3 a/o molybdenum alloy creep-test sample was loaded to 30,000 psi, a stress considerably in excess of its 1200 F yield strength, before the tensile data were available. As a result, a total deformation of 2.42 per cent occurred in this alloy sample on loading. It is of interest to note, however, that after this initial deformation, substantially no further deformation occurred in 210 hr at 30,000 psi. Similarly, after increasing the load on this sample from 30,000 to 35,000 psi, negligible creep has occurred after 50 hr. These

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TABLE F-3. CORROSION DATA FOR ALLOYS PREPARED FROM HIGH-PURITY NIOBIUM

Nominal Alloy Content (Balance Niobium), a/o	Total Weight Change(a), mg per cm ²		
	Water		750 F Steam
	600 F	680 F	
	42 Days of Exposure		28 Days of Exposure
Unalloyed commercial Nb	+3.84	-46.8	(b)
Unalloyed high-purity Nb	+2.08	+1.07	-3.33
7.18 Mo(c)	+0.10	+0.37	-0.13
12.58 V(c)	+0.10	+0.12	+0.34
	14 Days of Exposure		
45 Zr-5 Ti	+0.05	+0.19	+0.51
17.8 Ti-8.9	+0.10	+0.10	+0.24
10 Ti-3 Mo	+0.10	+0.15	+0.31

(a) Average of two specimens.

(b) Completely oxidized after 28 days of exposure.

(c) Compositions determined by chemical analysis.

TABLE F-4. TENSILE PROPERTIES OF HIGH-PURITY NIOBIUM-BASE ALLOYS AS ANNEALED 1 HR AT 1500 C (2730 F)

	Test Temperature, F(a)		
	70	600	1200
Alloy NL-4, Niobium-45 a/o Zirconium-5 a/o Titanium			
0.2 Per Cent Offset Yield Strength, psi	124,000	87,000	--
Ultimate Strength, psi	128,000	101,000	78,000
Elongation, per cent	8	6	3
Alloy NL-7, Niobium-10 a/o Titanium-3 a/o Molybdenum			
0.2 Per Cent Offset Yield Strength, psi	57,000	31,000	22,000
Ultimate Strength, psi	67,000	41,000	39,000
Elongation, per cent	35	32	21

(a) All samples tested with a crosshead speed of 0.02 in. per min. Air atmosphere used at 70 and 600 F and helium used at 1200 F.

TABLE F-5. CREEP DATA ON HIGH-PURITY NIOBIUM-BASE ALLOYS TESTED IN VACUUM AFTER VACUUM ANNEALING 1 HR AT 1500 C (2730 F)

Alloy	Nominal Alloy Content (Balance Niobium), a/o	Test Temperature, F	Load, psi	Total Deformation at Indicated Time, per cent								Average Creep Rate, per cent per hr	Remarks
				On Loading	1 Hr	2 Hr	10 Hr	50 Hr	100 Hr	150 Hr	200 Hr		
NL-2	12.58 V ^(a)	1380	40,000	0.36	0.36	--	0.38	0.47	0.59	0.67	0.78	0.0017	Test discontinued after 307 hr
NL-3	7.18 Mo ^(a)	1380	30,000	0.24	0.4	0.5	1.0	3.9	7.6	11.6	--	0.066	Ruptured at 177 hr; 24 per cent elongation
NL-4	45 Zr-5 Ti	1200	40,000	0.39	0.39	0.45	--	--	--	--	--	0.176	Fractured in shoulder at 7.7 hr
NL-7	10 Ti-3 Mo	1200	30,000	2.42	2.40	2.40	2.40	2.40	2.40	2.40	2.41	0.00005	Stress increased to 35,000 psi at 210 hr;
			35,000 ^(b)	3.56	3.82	3.82	3.82	3.82	--	--	--	--	test still in process

(a) Compositions determined by chemical analyses.

(b) Deformation values at 35,000 psi include 2.41 per cent total deformation incurred during 30,000-psi exposure.

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results indicate that the creep strength of this alloy can be substantially improved by cold working.

Double consumable-electrode melting was completed on four scale-up alloys containing, nominally, 45 a/o zirconium-3 a/o vanadium, 10 a/o zirconium-3 a/o vanadium, 6.25 a/o zirconium, 12.5 a/o vanadium, and 10 a/o titanium-5 a/o chromium. These ingots have been cut into slabs preparatory to rolling, in stainless steel packs, to sheet. The material obtained will be used to determine the tensile and creep properties of the alloys as well as their resistance to corrosion in 600 and 680 F water and in 750 F steam.

Inert-electrode arc melting was also completed on a series of forty 50-g ingots. These include binary alloys containing up to 45 a/o zirconium, 25 a/o titanium, 5 a/o chromium, and 2.5 a/o vanadium, and ternary niobium-zirconium (2.5 a/o to 45 a/o) base alloys containing additions of aluminum, chromium, iron, molybdenum, titanium, or vanadium in amounts up to 15 a/o. Sheet prepared from these ingots will primarily be used to screen the alloys for resistance to corrosion in 680 F water.

Rubbing Surfaces in Sodium Environments

J. W. Kissel, C. M. Allen, and W. A. Glaeser

During Fiscal Year 1958, a program of research to investigate the friction and wear behavior of rubbing surfaces lubricated by high-temperature (above 1000 F) sodium was initiated. Primary objectives were to secure a basic understanding of why friction and wear vary in molten sodium, and, from this information, to establish principles and recommendations that could be applied in the design of sodium-cooled-reactor components. In the past, specific wear and friction problems in liquid-sodium environments have been resolved by empirical methods using component-simulation devices. Little or no effort has been made to obtain knowledge concerning the rubbing behavior of a wide selection of materials over a variety of conditions. The need for information, useful for the synthesis of new high-temperature materials as well as for the selection of those presently available, is becoming increasingly urgent as reactor temperatures are pushed upward to more efficient operating levels.

A low-speed "stick-slip" friction apparatus has been developed and successfully operated with liquid-sodium lubricant. The apparatus provides a record of the frictional force between a solid ball and a lapped flat plate which is moved horizontally at a uniform speed. Provisions have been made to operate the specimens at a controlled temperature (up to about 1350 F) in a controlled environment (vacuum or inert gas) with molten sodium present between the rubbing surfaces. Coefficient-of-friction data and observations of wear have been obtained for several combinations of materials rubbing together in molten sodium. Two of these evaluations provided results exhibiting an interesting contrast. A cermet, WC + 20 w/o cobalt binder, when rubbed against itself, demonstrated higher friction when molten sodium was introduced between the sliding surfaces. However, when molybdenum was rubbed against molybdenum under the same conditions, the friction was lower when wetted by sodium as compared to dry surfaces at the same temperatures. This behavior is believed to result from the presence of surface films which become altered chemically and/or physically when changes are made in the

environmental conditions. From these results it was believed imperative that additional work be directed toward answering such questions as: What are the physical and chemical characteristics of the surface films present on rubbing surfaces in molten sodium? How can these properties be altered? And by what mechanism do these films influence friction and wear?

During the interim period between the end of Fiscal Year 1958 and the resumption of the research program in March, 1959, a number of contacts were made with sodium-system users in order to become familiar with the current problems. By this means, a rather extensive need for knowledge was found to exist concerning basic principles which influence rubbing-surface performance and for design data to cover a variety of conditions. It was also learned that current problems in this field are being solved by trial-and-error modification of existing components - a practice which admittedly is involved and costly. For example, shaft seals for pumps and valves have been moved from sodium-immersed locations to inert-gas-filled thimbles; valve seats have been located in positions where temperatures do not exceed 1200 F; and secondary inert-gas thimbles have been used to back up seals where leakage cannot be prevented. On the other hand, fuel-rod and control-rod mechanisms, which operate immersed in molten alkali metals, cannot be moved or "designed around". In this case, welding and galling of the contacting surfaces lead to premature failure. From the foregoing considerations, it was concluded that the basic study of surface films, referred to previously, would be continued throughout the current program.

The primary objective of the program throughout the remainder of Fiscal Year 1959 will be to acquire information concerning the properties of molybdenum surfaces and of the films which occur thereon under special conditions. Toward this end, techniques of electron and X-ray diffraction, electron and light microscopy, static and low-speed kinetic friction, and others, will be tried and evaluated. The condition and history of preparation of the molybdenum surfaces, as well as temperature and atmosphere conditions, will be the parameters of interest. Molybdenum has been selected as the primary evaluation material for the following reasons:

- (1) Molybdenum has recognized high-temperature strength and hardness.
- (2) It has good high-temperature corrosion resistance to molten alkali metal.
- (3) It has demonstrated relatively little tendency to self-weld when immersed in high-temperature alkali-metal for long periods.
- (4) The friction and wear between molybdenum rubbing surfaces was shown during the preceding testing program to be much lower when sodium was present than when the surfaces were dry.
- (5) The variation in sodium-lubricated friction over a large temperature range, presented a rather complex picture; this suggested a change in the surface-film situation.

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The program now under way is expected to proceed as follows:

- (1) Molybdenum versus itself friction experiments will be run. The parameters are: temperature, sodium absent and present, surface (of flat specimen) lapped and/or etched, original molybdenum rolled or forged.
- (2) Concurrently, the attempts to identify oxide films which form on molybdenum at various temperatures will be made by electron diffraction.
- (3) The stability of the oxide films will be ascertained by heating in an inert atmosphere.
- (4) The effect of molten alkali metal at various controlled temperatures on the known surface films will be investigated by encapsulation studies.
- (5) The friction between specimens which have surface films of known composition will be determined.

This program is expected to provide a beginning to a better understanding of what is taking place during sliding in liquid sodium and to developing the techniques which are best suited to the investigation of wear and friction mechanisms in this environment.

During March the friction apparatus was reinstalled and placed in operating condition. Flat specimens are being made from two types of molybdenum: hot rolled, and forged and annealed. Microscopic examination revealed the rolled material contained a much lower amount of oxide-phase inclusions; therefore, it has been chosen for initial experimentation. Arrangements have also been made to begin the surface-film studies.

STUDIES OF ALLOY FUELS

R. F. Dickerson

Microstructures of niobium-30 w/o uranium alloys fabricated at 3000 F show what appears to be a cold-worked structure. During fabrication, these alloys appeared to be harder than the molybdenum canning material. Since it is difficult to conceive of this alloy cold working at 3000 F, further investigation is in progress. Corrosion-test coupons of all alloys being investigated have been prepared, and 1000-hr corrosion tests are being initiated. Corrosion tests in air at 300, 350, 400, and 500 C are in progress.

Studies of binary thorium-uranium alloys verify the fact that carbon is a hardener. The results also indicate that alloys prepared by induction melting in zirconia crucibles are slightly softer than similar alloys prepared by arc melting. Recrystallization studies of alloys containing 5, 10, 15, and 20 w/o uranium, reduced 80 per cent, show that after 10 min at 600 C softening occurs. Additional studies are in progress. Cold rolling at reductions of 60 per cent elongate the uranium particles, whereas hot rolling

does not have this effect. Thorium-uranium alloys to which cerium, lanthanum, ruthenium, titanium, and yttrium have been added have been corrosion tested in 200 C water. The results indicate that protective oxide films are not formed.

Development of Niobium-Uranium Alloys

J. A. DeMastry, F. R. Shoher, and R. F. Dickerson

A study of the fabrication characteristics, and the mechanical and physical properties of niobium-rich uranium alloys is being made. These alloys are potential high-temperature reactor fuel materials, and it is necessary to know these properties to better evaluate the alloy.

The effect of impurities is being studied by using three grades of niobium as base materials in the preparation of these alloys. The three grades being used are: one containing 0.7 w/o zirconium and 600 ppm oxygen, a second containing 0.17 w/o zirconium and 700 ppm oxygen, and a third containing less than 200 ppm zirconium and less than 300 ppm oxygen. The alloys being studied are uranium-40, -50, -60, -70, -80, and -90 a/o niobium; each of these alloys has been prepared using all three grades of niobium. The uranium 80 and 90 w/o niobium alloys have been successfully cast as consumable-electrode arc-melted ingots and fabricated into slab material.

Table F-6 shows the results of the attempted fabrication of the uranium-50, -60, and -70 w/o niobium alloys. From previous results it appeared that the uranium-70 w/o niobium alloys would fabricate at 3000 F. However, examinations of the microstructure of these alloys indicate that at a temperature of 3000 F these alloys are cold fabricated. This must be investigated further. In fabrication of the uranium-70 w/o niobium alloys it was noted that the alloys appeared to be harder than the molybdenum canning material. In order to evaluate the strength of this alloy, bars are being cast which will be machined into tensile specimens. Up to the present time no effects of the grade of starting stock has been noted with respect to casting or fabrication characteristics.

Blanks for water-corrosion tests have been machined, and tests for 1000 hr in 600 and 680 F water and 750 F steam will be started. Corrosion testing in NaK at the boiling point is to be initiated as soon as encapsulation is completed. Specimens for corrosion testing in carbon dioxide at elevated temperatures have been machined and are being weighed for testing. The effects of corrosion testing in air at 300, 350, 400, and 500 C are shown in Table F-7. All of the alloys studied are to be tested for oxidation resistance in air at 300, 350 and 400 C.

No results have been obtained from electrical-resistivity, thermal-conductivity, and thermal-expansion measurements. Machining of tensile specimens is nearly complete, and tensile strengths of the uranium-70, -80, and -90 w/o niobium alloys will be obtained.

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TABLE F-6. RESULTS OF HAMMER FORGING 2-IN. -DIAMETER NIOBIUM-URANIUM INGOTS IN MOLYBDENUM PACKS AT 3000 F FROM A HYDROGEN-ATMOSPHERE FURNACE

Nominal Composition (Balance Uranium), w/o	Results of Fabrication Attempts
70 Nb(a)	Pack broke open; it appears that alloy was not reducing; cracks present in alloy
70 Nb(b)	Alloy reduced 70 per cent, but was badly cracked
60 Nb(a)	No fabrication attempted
50 Nb(a)	Alloy reduced about 70 per cent; however, alloy was broken and badly cracked
40 Nb(a)	No fabrication attempted

(a) Niobium used contains less than 300 ppm oxygen and less than 200 ppm zirconium.

(b) Niobium used contains 600 ppm oxygen and 0.7 w/o zirconium.

TABLE F-7. CORROSION DATA FOR URANIUM-NIOBIUM ALLOY IN AIR

Alloy Content (Balance Uranium), w/o	Time on Test, hr	Total Weight Gain(a) After Exposure At Temperature Shown, mg per dm ²			
		300 C	350 C	400 C	500 C
90 Nb(b)	48	13.4	83.8	2458	--
90 Nb(c)	24	--	--	--	9650
90 Nb(c)	100	--	--	2576	--
90 Nb(c)	304	15.8	--	--	--
80 Nb(d)	24	--	--	--	11,400
80 Nb(d)	100	--	--	2664	--
80 Nb(d)	304	16.7	--	--	--
70 Nb(c)	24	--	--	--	10,000
	100	--	--	12,450	--
	304	16.0	--	--	--

(a) Average of two specimens.

(b) Niobium contains less than 300 ppm oxygen and less than 200 ppm zirconium. The surface condition of the alloy following hot rolling was poor, all other alloys had good hot-rolled surfaces.

(c) Niobium contains 700 ppm oxygen and 0.17 w/o zirconium.

(d) Niobium contains 600 ppm oxygen and 0.7 w/o zirconium.

Development of Thorium-Uranium Alloys

M. S. Farkas, A. A. Bauer, and R. F. Dickerson

Thorium-uranium alloys are being investigated with the objective of improving corrosion resistance and irradiation stability by means of alloying and control of processing variables. Ternary and quaternary thorium-uranium alloys (formed by the addition of molybdenum, niobium, or zirconium) are being investigated, the additions being made for the purpose of strengthening the thorium matrix and stabilizing the gamma-uranium phase.

Studies of binary thorium-uranium alloys in the as-cast, hot-rolled, and cold-reduced conditions to determine the variation of structure with fabrication are being carried out. The effect of cold work on hardness is presented in Table F-8. It is evident that the alloys containing Ames thorium are harder than those containing crystal bar-thorium, at all reductions. This is due to the higher amounts of carbon present in the Ames material, carbon being a potent hardener of thorium. The induction-melted alloys are slightly softer than the arc-melted alloys containing the same material. Although the hardness differences may be insignificant, it is thought that the high cooling rates associated with arc melting allow the retention of more uranium in solution than is obtained by induction melting, thus producing an alloy which is slightly harder. Also, induction-melted alloys pick up zirconium from the zirconia crucibles. This zirconium forms zirconium carbide, which lowers the effective carbon content of the alloy and thereby softens it. Of significance, too, is the fact that the rate of work hardening of these alloys increases with increasing uranium content.

Recrystallization studies of thorium-5, -10, -15, and -20 w/o uranium alloys, reduced 80 per cent, show that after 10 min at 600 C a slight softening occurs; however, additional time at temperature causes only minor variations in hardness. It is not known whether or not actual recrystallization takes place since hardness may also be responsive to precipitation of uranium which is soluble at elevated temperatures. Further recrystallization studies at higher temperatures are being performed with material that is 60, 80, and 90 per cent reduced.

Metallographic examinations of hot-rolled and cold-rolled specimens showed the uranium particles to be elongated in the direction of rolling. After 60 per cent cold reduction, thorium-10 w/o uranium alloys exhibited microstructures in which the uranium particles showed elongations of several times their width, whereas the hot-rolled and 20 per cent cold-reduced alloys exhibited microstructures which were relatively unaffected by working. In general the 15 and 20 w/o uranium alloys showed more pronounced stringing at all reductions.

Density determinations were performed on 80 per cent cold-reduced rod. The results are presented in Table F-9. A plot of these densities versus composition for the crystal-bar thorium alloys is linear and extrapolates to a thorium density of 11.64 g per cm³. The density data for the Ames thorium alloys when plotted is too scattered to allow accurate extrapolation to pure thorium. The density of the Ames alloys is, in general, lower than that of the crystal-bar material, which can be accounted for on the basis of the higher oxygen content of the Ames thorium, which leads to the formation of ThO₂.

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TABLE F-8. THE EFFECT OF COLD WORK ON HARDNESS OF THORIUM-URANIUM ALLOYS

Uranium Content (Balance Thorium), w/o	Type of Thorium	Melting Method	Vickers Hardness (10-Kg Load)			
			After Hot Rolling	Cold Reduced Percentage Shown		
				20	60	80
10	Crystal bar	Arc	103	111	130	135
10	Crystal bar	Induction	94.1	109	121	118
10	Ames	Arc	122	142	151	159
10	Ames	Induction	123	132	153	159
5	Crystal bar	Arc	107	114	123	123
15	Crystal bar	Arc	98	117	138	138
20	Crystal bar	Arc	105	132	141	150

TABLE F-9. DENSITIES OF ARC-MELTED AND COLD-SWAGED THORIUM-URANIUM ALLOYS

Uranium Content (Balance Thorium), w/o	Type of Thorium	Density, g per cm ³
5	Crystal bar	11.88
5	Ames	11.79
10	Crystal bar	12.10
10	Ames	12.01
15	Crystal bar	12.33
15	Ames	12.30
20	Crystal bar	12.58
20	Ames	12.61

X-ray examinations of the thorium-25 w/o uranium-25 w/o zirconium and thorium-20 w/o uranium-20 w/o zirconium alloys, quenched from 1000 C, show that alpha thorium and beta thorium are present in both alloys. Previous metallographic examinations had indicated the presence of only one phase, beta thorium. These alloys are of interest because of their comparatively good corrosion performance in 200 C water when quenched from 1000 C.

Thorium-uranium alloys containing small additions of niobium and molybdenum are being studied for the purpose of obtaining a dispersion of stabilized gamma-uranium particles in a thorium matrix. Metallographic examinations of these cast alloys indicate that the desired structure was obtained. However, after heat treating 24 hr at 800 C and furnace cooling, the thorium-10 w/o uranium-1.5 w/o molybdenum contains uranium particles that are rimmed. It is believed that these rims are indicative of possible transformation to delta U_2Mo . X-ray diffraction analysis is being used to determine the nature of the rimmed particles. Further studies on these alloys are being directed toward the detection of transformation in the stabilized gamma particles.

Corrosion tests, in 200 C water, on thorium-10 w/o uranium and thorium-10 w/o uranium-10 w/o zirconium alloys with small additions of cerium, lanthanum, ruthenium, titanium, and yttrium have been made. The results indicate that these additions are harmful to corrosion resistance when compared to the behavior of a thorium-10 w/o uranium-10 w/o zirconium alloy.

FISSION-GAS RELEASE FROM REFRACTORY FUELS

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D. N. Sunderman, S. D. Beck, and F. A. Rough

The objective of this research program is the development of a physical model and mathematical theory to describe the release of fission gases from sintered fuel materials. The scope of the problem is limited to study of the gas-release phenomenon in sintered uranium dioxide. The study of in-pile and postirradiation gas release is being conducted in conjunction with experiments to characterize the structure and properties of UO_2 , fission-gas diffusion coefficient determinations in fused or 100 per cent dense oxide, and development of a mathematical gas-release model.

Examination of Fused Uranium Dioxide

The primary effort in the examination of fused UO_2 obtained from the Norton Company has been in the determination of its physical characteristics and chemical composition and in establishing procedures for preparing specimens for use in irradiation studies. The February progress report stated that surface and internal contamination was present. Various tests have failed to identify this material with certainty. On the basis of X-ray diffraction measurements, the surface and internal contaminants were

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reported as UN_2 and uranium, respectively. However, more recent evidence casts some doubts on the X-ray diffraction results. The suspected uranium phase gives negative response to chemical etchants, atmospheric corrosion conditions, boiling water, and scratch hardness tests. The following in-purity levels were measured:

Nitrogen	20 ± 10 ppm
Carbon	30 ± 10 ppm
All other metals	200 ppm

The oxygen/uranium ratio is 1.99 ± 0.01 . Metallographic examinations showed no voids, inclusions, or imperfections other than the above-mentioned contamination.

Samples were heat treated in vacuum and in tank hydrogen at 1500 C for 3 hr and were compared with unheated controls. Results were inconclusive and more precise experiments will be conducted. However, present observations are that the hydrogen anneal apparently removed the contaminant completely. During the past month, a thin ($\sim 30 \mu$) section was made of one promising portion of one crystal. This section was cut nearly parallel with the (111) planes of the crystal. Examination of the thin section by transmitted light revealed traces of slip or deformation bands. Where these intersect, angles of 60 or 120 deg are formed which would suggest that slip occurred along the (111) plane. Since the number of these traces was very high at the thinner edges of the section, they were probably formed during cutting and polishing. Although the bulk crystal transmitted red light, numerous small spherical spots were not transparent. These could be inclusions of metal, oxygen-rich oxide, other impurities, or voids. Their distribution throughout the thin section could be recognized by adjusting the plane of focus. X-ray diffraction examination of this thin section has not revealed their identity. Their presence, however, might be sinks for the retention of fission products. There was no visible evidence of connection between the opaque spheres when viewed at 500X.

In the event that the present fused oxide is not usable, a substitute material must be obtained. Therefore, several techniques are being surveyed for the preparation of very-high-density sintered UO_2 . Oxide pellets sintered to 97 per cent of theoretical density at 1750 C will be resintered at 2000 C for further densification. High-pressure hot-pressing techniques will also be investigated. Uranium-dioxide powder has been compacted at 50,000 and 120,000 atm and simultaneously heated to 550 and 1100 C, respectively. These specimens are being examined metallographically to determine the nature of their porosity. Electron-beam melting of UO_2 is being considered as a method of avoiding contamination or reduction of the oxide during fusion.

Characterization of Sintered Uranium Dioxide

The shortage of UO_2 powder for specimen preparation has prevented a comprehensive study of pore structure and microstructure of sintered UO_2 . However, preliminary studies of two sintered specimens have been continuing. One specimen was sintered at 1400 C for 3 hr to 94 per cent of theoretical density; the other was sintered at 1700 C.

In addition to grain-size differences, the most obvious variation is in pore shape and size; pores in the specimen sintered at 1400 C are more irregular and are smaller than pores found in the high-temperature-sintered material. Sintering at 1700 C resulted in pore surfaces which correspond to crystallographic planes.

Sufficient UO_2 has been obtained for the preparation of sintered specimens for a comprehensive study of the effects of various factors on pore size, shape, and distribution. The same type of oxide will be used in subsequent irradiation and fission-gas-release experiments.

Determination of Diffusion Coefficients and Preparation
for In-Pile Study of Gas Release

The detailed design and assembly of apparatus to study the release of fission gas during irradiation in-pile and during postirradiation heat treatment are continuing. It is anticipated that the design and construction of the out-of-pile apparatus will be completed by May.

The first attempt to produce the desired specimen temperature of 3000 F by use of induction heating was made in March. It was intended that the experiment answer some immediate questions of interest and great importance in guiding the system design efforts. The objectives of the experiment were to determine:

- (1) Practicality of induction heating with coil leads 12 ft long.
 - (a) Power loss or demand
 - (b) Arc over to surroundings
- (2) Time to achieve specimen temperature of 3000 F
- (3) Compatibility of materials used at 3000 F
- (4) Strength of materials at operation temperature of specimen
- (5) Susceptor-design and -fabrication problems
- (6) Heated-region temperature profile
- (7) Reliability of platinum-platinum-10 w/o rhodium thermocouples in apparatus and at operating specimen temperature.

The UO_2 specimen, a molybdenum crimped-seam susceptor, and a Norton RA-98 Alundum sleeve were enclosed in one chamber of a dumbbell-shaped Vycor vessel with a helium atmosphere. The empty cold chamber of the vessel was provided for gas expansion. Power was delivered by 12-ft-long copper leads to an aluminum coil which surrounded the vessel.

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The conclusions which were drawn from the experiment are:

- (1) It is possible to supply the required power to heat the specimen by use of a 3-kw induction heating unit with the coil 12 ft from the control console.
- (2) Lavite may not be used in the system in the high-temperature regions.
- (3) The Vycor did not heat up to anything near its softening point. This was a point in question prior to the experiment.
- (4) A high alumina content (<97 w/o) ceramic tube will be required to separate the susceptor from the Vycor. Lower alumina content (~ 90 w/o) ceramics will not hold up at desired temperatures.
- (5) The specimen will have to be heated slowly to avoid thermal fracture.
- (6) Platinum-platinum 10 w/o rhodium thermocouples may be used but thermocouples designed for higher temperatures should be considered.
- (7) Slight quantities of oxygen and moisture are intolerable.

Further experimenting is planned wherein the above factors are being recognized. A tantalum susceptor is going to be used instead of the molybdenum. Welding of the seam in the tantalum susceptor will be attempted, thereby avoiding preferential heating in crimp or overlap seams. Seamless tantalum tubing has been ordered for use in the event that use of the welded material is unsuccessful in coping with the seam problem.

Components for the gas-sampling system associated with the in-pile studies are being collected for assembly. Assembly of this system will follow that of the post-irradiation-heating apparatus and should be completed by June.

Mathematical Analysis of Gas Release

There may be several mechanisms which contribute to fission-gas release from UO_2 fuel elements. The purpose of the mathematical phase of the program is to attempt to formulate one or more models by which the gas release may be explained and predicted.

One mechanism which may have an important role is that of volume diffusion. Many investigators utilize the concept, in the case of porous materials, of small characteristic spheres of homogeneous and isotropic solid. The gas release from the fuel element is described in terms of that from this characteristic sphere. Release by volume diffusion may also be described in terms of other geometric models. In each case, however, the assumption that Fick's law holds is implicit in the analysis.

The volume diffusion model will be examined to ascertain whether it is adequate to explain fission-gas release. It may prove necessary to develop a more sophisticated model to accurately describe the release phenomenon.

GENERAL FUEL-ELEMENT DEVELOPMENT

S. J. Paprocki

The effort on the development of cermet materials containing 60 to 90 volume per cent of UO_2 , UN, or UC has been primarily devoted to the fabrication of niobium-coated UO_2 . Cold-pressed compacts of niobium-coated UO_2 powders were densified to a 90 per cent of theoretical density by gas-pressure bonding 3 hr at 2300 F with a helium gas pressure of 10,000 psi. Excellent structures were obtained with each UO_2 particle enveloped by niobium.

The gas-pressure-bonding process is being utilized to clad and bond niobium- and molybdenum-base fuel materials. The niobium-base cermets are being fabricated into niobium-clad fuel elements. The ultimate objective is to pressure bond integral assemblies in a one-step operation.

Stainless-clad stainless-24 w/o UN and UC dispersions have been irradiated and are being evaluated. The irradiation stability of these materials will be compared to stainless- UO_2 dispersions irradiated under comparable conditions. It is anticipated that these materials will be superior to stainless- UO_2 because of improved structure and properties resulting from a lower volume loading of dispersed material for an equivalent uranium-235 content.

A hot-pressing technique is being utilized to study the mechanism and kinetics of solid-state bonding. It is anticipated that the results from this study will be directly applicable to the gas-pressure-bonding process.

Fabrication of Cermet Fuel Elements

S. J. Paprocki, D. L. Keller, G. W. Cunningham, and D. E. Kizer

Techniques for producing cermets of 90 per cent of theoretical density or better containing 60 to 90 volume per cent fuel dispersed in a metal matrix are being investigated. Evaluations of the cermets are being made on the basis of microstructures and physical and mechanical properties.

Uranium dioxide powder has been coated with niobium by the vapor-deposition method. This coated powder, when compacted to a high density, offers the advantage over standard UO_2 -metal mixtures of having each UO_2 particle discrete in that it is surrounded by niobium metal. A pellet containing a mixture of 85 w/o niobium-coated minus 100 plus 140-mesh UO_2 powder and 15 w/o minus 325-mesh uncoated UO_2 powder was compacted at 40 tsi. This mixture resulted in a pellet of 84.8 volume per cent UO_2 -niobium. The 0.5-in. -diameter pellet was fitted into a stainless steel tube which was sealed by welding in end plugs, one of which incorporated an evacuation tube. The assembly was evacuated subsequent to hydrostatic pressing for 3 hr and 20 min at 2200 F under a helium gas pressure of 10,000 psi. This operation yielded a measured core density of 88.6 per cent of theoretical.

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A similar assembly containing a core of 80 w/o niobium-coated minus 100 plus 140-mesh UO_2 powder and 20 w/o minus 325-mesh uncoated UO_2 powder was hydrostatically pressed for 3 hr at 2300 F under a helium gas pressure of 10,000 psi. The mixture resulted in a core of 86 volume per cent UO_2 -niobium. The core density was increased to 90 per cent of theoretical. A similar core is being prepared for hydrostatic pressing for possible use as a thermal-conductivity specimen.

Microscopic examination of a 80 volume per cent UN-niobium cermet hot-press forged at 2100 F indicated that a reaction occurred between the fuel particles and impurities in the metal matrix. Examination of a sample of the cermet after a 3-hr vacuum anneal at 2300 F indicated that the UN gettered oxygen to form a layer of UO_2 up to 0.0005 in. thick between the UN and niobium.

Methods of producing fuel elements of shapes other than flat plates or rods are also being investigated using UO_2 -niobium and UO_2 -molybdenum. Specimens are being made to determine the feasibility of pressure bonding green-pressed cermets to a high density by fitting green-pressed rings into the annulus of a hollow-walled metal tube prior to sealing and pressure bonding.

Other materials being prepared for hot-press-forging studies include cores of 80 volume per cent UO_2 dispersed in chromium and 80 volume per cent UC dispersed in chromium, molybdenum, and niobium.

Gas-Pressure Bonding of Molybdenum- and
Niobium-Clad Fuel Elements

S. J. Paprocki, E. S. Hodge, C. B. Boyer, and R. W. Getz

Molybdenum and niobium retain their strength at elevated temperatures and possess favorable nuclear properties, which makes them potential cladding and structural materials for application in high-temperature reactors. In inert gas-cooled or some liquid-metal-cooled reactors, the niobium and molybdenum can be utilized without benefit of a protective coating. The gas-pressure-bonding process is being investigated as a method of cladding of ceramic and cermet-type fuels with molybdenum and niobium.

A compartmented niobium-clad uranium dioxide fuel plate was bonded at 2200 F for 4 hr at 10,000 psi. The four-compartment element was prepared from niobium piece components. Niobium cladding 0.010 in. thick was employed in the preparation of the 0.060 by 1.5 by 5.8-in. fuel plate. This plate contained 0.040-in.-thick core inserts that were approximately 1 in. square. Good bonds were obtained, as revealed by the metallographic and peel tests of the niobium interfaces after pressure bonding. The niobium surfaces were conditioned with a Cr_2O_3 acid solution subsequent to a treatment in a sodium hydroxide bath. After washing, drying, and assembly of the niobium fuel plate, it was inserted into a Ti-Namel can with graphite-coated molybdenum spacers between the can and the fuel plate. The coated spacers were used to reduce any possible can-to-fuel plate reaction. The bonded element had less ductility after

bonding; consequently, either the spacer was not completely effective or the specimen was contaminated in some manner. The specimen is being analyzed for carbon, hydrogen, molybdenum, and oxygen. It is not expected that the carbon is diffusing into the niobium in a sufficient amount to account for the loss in ductility during bonding. Specimens of niobium that were coated with carbon were heated through the bonding cycle employed in the autoclave without any significant loss in ductility. Additional niobium fuel plates have been prepared with carbon-coated Ti-Namel spacers between the cladding and container for bonding at 2200 F.

A limited number of molybdenum-clad fuel plates and surface-preparation specimens have been bonded. Bonding of molybdenum-clad elements was limited to 4 hr at 2450 F at 10,000 psi with the available temperature-measurement thermocouples contained in the autoclave closures. New thermocouples have been prepared and assembled into the equipment for the bonding studies at higher temperatures planned for molybdenum-clad specimens. A single-compartment fuel plate consisting of molybdenum-clad uranium dioxide was prepared from pieced components and bonded at 2450 F for 4 hr at 10,000 psi. The cladding cover plates were cold rolled prior to degreasing and washing for pressure bonding. The results obtained with this specimen were encouraging; however, the frame material was not cold worked prior to bonding and the bond properties were not consistent along the length of the specimen.

Molybdenum surface-preparation specimens were degreased in kerosene and acetone prior to bonding at 2450 F for 4 hr at 10,000 psi. The plates utilized in this study were subjected to cold reductions of 50 per cent prior to the degreasing operation. The bonds obtained were strong, and complete grain growth had occurred in many areas of the specimen. Partial grain growth was achieved in all areas. The amount of complete grain growth across the interface was determined by a Hurlbutt counter to be 51 per cent. The improved bonds obtained appeared to be significantly influenced by the prior cold work put into the molybdenum. Additional cold-worked molybdenum specimens will be bonded at higher temperatures in the altered bonding equipment. If satisfactory bonding is achieved, a four-compartment fuel plate will be clad with molybdenum at 2500 to 2600 F.

The Irradiation of UC- and UN-Stainless Steel Dispersion-Type Fuel Elements

A. W. Hare and D. L. Keller

The purpose of this program is to irradiate stainless-clad stainless-24 w/o UN and UC dispersions at elevated temperatures and to compare the results with dispersion fuel elements consisting of UO_2 dispersed in stainless steel.

Twelve test specimens, six containing UN as fuel and six containing UC as fuel are being investigated. Four specimens (two UN and two UC) each were irradiated in a single irradiation capsule. Two of the capsules have been irradiated and returned to BMI for postirradiation examination and evaluation. The dimensional measurements before and after irradiation were reported last month.

F-27

A metallographic examination of the irradiated fuel specimens is currently in progress.

Dosimetry data from both capsules, BMI-18-1 and BMI-18-2, were determined and are tabulated and compared with estimated burnups in Tables F-10 and F-11.

It is interesting to note that the reactor-quoted fluxes agree within 15 per cent if the Battelle experience factor is used. These results will be compared with radiochemical burnup analyses of the specimens from both capsules. The radiochemical analyses are currently in progress.

The four remaining specimens contained in Capsule BMI-18-3 have been in the MTR A38NE position for two complete cycles (Cycles 116 and 117), and thermocouple data indicate that the fuel core temperatures have been in the range of 1540 to 1650 F. It is planned to discharge these specimens at the end of Cycle 119.

Factors Affecting Pressure Bonding

G. W. Cunningham and J. W. Spretnak

A hot-pressing technique is being used to investigate the mechanism and kinetics of solid-state bonding of metals by the application of heat and pressure. Although true hydrostatic pressure is not obtained in the hot-press die, any theories which are developed will be evaluated in gas-pressure-bonding autoclaves.

In the work with copper specimens, pressure-bonded specimens are being annealed in an effort to gain information on the nature of the interface formed by pressing the pieces of metal together. Examination of specimens pressed at 1150 F and 10,000 psi for 10 min and then annealed 2 hr at 1800 F indicates that the interface may be changing to a structure similar to that of a grain boundary with void areas dispersed along the interface, and, in some cases, there is evidence that the boundaries gradually disappear after longer annealing times. An accurate evaluation has not been possible because of the presence of a copper-CuO₂ eutectic structure in the electrolytic tough-pitch copper being used. OFHC copper has been ordered, and several procedures are being examined as possible means of removing oxygen from the tough-pitch copper. Hydrogen annealing at 1400 to 1600 F will clean the surfaces and cleaner interfaces are obtained in the body of the specimens. Hydrogen atmospheres have also been used while hot pressing the specimens, and, although the atmosphere is beneficial in keeping the surfaces clean, very little purification occurs during the run. Metallographic examination indicates that oxygen-free copper can be produced by melting copper shot in hydrogen and allowing the melt to furnace cool. Ingots prepared by this method are being rolled to strip and will be machined into hot-press specimens.

For the study on the closure of macroscopic voids between the two metal surfaces by hot pressing, it would be desirable to have some sort of markers whereby deformation could be followed. As previously described, the specimens are prepared by bonding an 8 rms ground surface to a 500 rms shaped surface. In one specimen, instead

TABLE F-10. BURNUP ESTIMATES BASED ON REACTOR-QUOTED FLUX

Capsule	Cycle	Position	Distance of Capsule from Top of Reflector, in.		Reactor-Quoted Fluxes, Fuel Center Lines, 10^{14} nv		Estimated Specimen Uranium-235 Burnup Based on Reactor-Quoted Fluxes and Standard Empirical Correlation, per cent	
			Bottom	Top	Bottom Specimen	Top Specimen	Bottom Specimen	Top Specimen
BMI-18-1	113	A38NE	20	7	1.55	1.13	8.6	6.3
	114	A38NE	19	6	1.50	1.00	--	--
BMI-18-2	115	A38NE	17-1/2	4-1/2	1.35	0.85	3.9	2.4

TABLE F-11. BURNUP AND FLUX ESTIMATES BASED ON DOSIMETER DATA

Capsule	Unperturbed Reactor Flux, 10 ¹⁴ nv		Estimated Specimen Uranium-235 Burnup Based on Estimated Fluxes and Standard Empirical Correlations
	Based on Dosimeter Data	Estimate Based on Dosimeter Data, F Component of Brad Lewis Correlation, and 0.8 Experience Factor	
BMI-18-1	0.59 (facing top specimen)	0.95	5.4
	0.603 (facing 2nd specimen)	0.97	5.6
	0.842 (facing 3rd specimen)	1.36	7.7
	0.861 (facing bottom specimen)	1.39	7.8
BMI-18-2	0.442 (facing 2nd specimen)	0.71	2.1
	0.456 (facing top specimen)	0.73	2.2
	0.719 (facing bottom specimen)	1.16	3.4

F-29 and F-30

of a 0.100-in. plate with an 8 rms surface, a stack of 0.002- and 0.005-in. sheets was used. The specimen bonded in a manner similar to previously examined standard specimens with the teeth of the 500 rms surface penetrating into the flat surface. However, there appeared to be more extrusion of the flat surface into the roots of the teeth. This result may have been caused by the decrease in resistance to lateral slip which would be predicted for the initially unbonded layers of metal. Nevertheless, examination of the specimen does indicate that the expansion of hemispherical shells which gradually expand into a flat surface occurs, with sheets 0.010 in. from the interface being essentially unaffected. Refinement of this model and possible inclusion of grid marker wires in the specimen are being considered for further study.

Metallographic examination of a zirconium specimen bonded at 1250 F and 10,000 psi for 10 min indicates that the same mechanism of macroscopic void closure as previously described for copper, i. e., penetration of the teeth of the coarse surface into the flat surface applies. Annealing studies have not been completed on the zirconium.

G-1 and G-2

G. FATIGUE STUDIES OF INCONEL AND INOR-8

R. G. Carlson

Fatigue Studies of Inconel

The objectives of this program are to obtain basic fatigue information on Inconel and to establish quantitative relationships for Inconel among the variables of temperature, stress, strain, time, and cyclic frequency. The current phase of the program is concerned with measuring and recording strain in Inconel specimens under elevated-temperature fully alternating stress-cycling conditions.

During March, stress-cycling tests were continued for the 1500 F and 6-cpm condition. This work will be continued in April. In addition, specimens will be evaluated for the 1100 F and 6-cpm condition. This condition will be the last to be evaluated and testing should therefore be concluded in April.

Fatigue Studies of INOR-8

This program is concerned with the investigation of temperature and frequency dependence of fatigue properties of INOR-8 alloy.

During March, rotating-beam tests of INOR-8 were concluded at 1100 F, with cycling at 100 and 3000 rpm. The material is presently being evaluated at 1300 F, with cycling at 100 and 3000 rpm.

Tests will be continued during April for the 100- and 3000-rpm cyclings at 1300 F. In addition, an intermediate frequency will be evaluated for temperatures of 1300 and 1500 F.

H-1

H. PHYSICAL RESEARCH

F. A. Rough

Progress on two of the projects supported by the AEC Division of Research, is described. The first, the preparation of sintered uranium monocarbide specimens for study of irradiation effects is continuing, and initial work on the preparation of uranium nitride specimens is under way. The second, a fundamental study of the mechanism of thermal migration of hydrogen in zirconium hydride, presently involves experiments to obtain necessary data for comparison of actual calculated migration of hydrogen in zirconium.

Preparation and Properties of Refractory
Uranium Compounds

A. B. Tripler, Jr., M. Jack Snyder, and W. H. Duckworth

In this study, a correlation between fission-product release and selected physical properties of some sintered refractory uranium compounds is being sought. Physical properties such as porosity, grain size, and bulk density are varied by varying initial particle size, compacting pressure, temperature of sintering, and time at temperature. The sintered compacts will be subjected to short-time low-temperature low-level irradiation in the Battelle Research Reactor, after which fission-product release will be studied.

The effect of forming pressure and initial particle size on the final grain size and porosity has been studied for uranium monocarbide specimens sintered at 1600 C for 1 hr. These data are given in Table H-1.

TABLE H-1. EFFECTS OF FORMING PRESSURE AND INITIAL PARTICLE SIZE ON THE POROSITY AND GRAIN SIZE
OF UC COMPACTS SINTERED AT 1600 C FOR 1 HR

	Formed at 50,000 PSI		Formed at 100,000 PSI	
	6.2- Material ^(a)	14.7- Material ^(a)	6.2- Material ^(a)	14.2- Material ^(a)
Specimen	14335-72B	14335-82A	14335-63B	14335-63A
Average Void Volume, per cent	24.4	33.5	28.8	29.3
Average Grain Size,	22.0	43.0	22.0	43.0

(a) Average particle size for powders used.

H-2

The average grain size was determined by use of a Hurlbert counter. The average porosity values are estimates also determined by use of the Hurlbert counter. The Hurlbert counter consists of a motor-driven mechanical stage attached to a microscope. Associated with the stage is a counter which is activated by the operator. As the specimen traverses the field of the microscope at a constant speed, the operator presses the appropriate key of the counter, thus recording, for example, a void or solid material, as the case may be.

As shown in Table H-1, the average grain sizes of the sintered specimens correlate with the initial particle size. It appears that forming pressure has no effect on the grain size. With one exception, the percentage of voids correlates with forming pressure and possibly with initial particle size. The anomaly occurs with Specimen 14335-72B where the percentage of voids appears to be low. The determination of voids will be rechecked. The bulk-density measurements will also serve as a check.

At the 100,000-psi forming pressure only a small difference was measured in the percentage of voids with the two particle sizes; the difference may not be significant.

No UC₂ was observed in any of the specimens. Occasionally small amounts of uranium were observed at the grain boundaries. The total amount at the grain boundaries is very small.

The UC series will be continued with specimens prepared for longer times at 1600 and 1800 C.

A kilogram of U₂N₃ has been prepared. This will be denitrided to form high-purity UN. The UN will be ground so as to give two batches, each having a different average particle size, and experiments similar to those with UC will be performed on the UN.

When both UC and UN specimens are finished, they will be subjected to irradiation.

Thermal Migration of Hydrogen in Zirconium

J. W. Droege, W. M. Albrecht, W. D. Goode, and H. H. Krause

Fabrication and construction of the apparatus designed to study the migration of hydrogen in zirconium hydride under the influence of a thermal gradient have continued. The effect of hydrogen concentration on the diffusion coefficient in the delta phase of zirconium hydride has been investigated.

Thermal Diffusion

The thermal-diffusion cell, incorporating a specimen of beta-phase zirconium hydride, has been completed. Most of the components of the vacuum system in which the cell will operate have been built and assembled. Preliminary experiments should be

H-3 and H-4

under way soon, in which the disk of zirconium hydride will be maintained in a carefully controlled one-dimensional thermal gradient. The equilibrium hydrogen pressures will be measured at each face of the disk, and from these data it will be possible to determine the concentration at the two faces. Because of the short migration path involved ($1/4$ in.), it should be possible to arrive at steady-state conditions in a short time, so that measurements can conveniently be made at many temperatures and compositions. Since measurements of hydrogen pressure can be made continuously, it will be possible to determine whether the steady state has been attained, a determination which has not been possible with the methods reported in the past. Measurements are planned in all three major phases of the zirconium-hydrogen system, and perhaps also across phase boundaries. Within each phase, measurements will be made at various temperatures, concentrations, and thermal gradients.

Diffusion Coefficients

Permeation rates for hydrogen in delta zirconium hydride have been measured at 600 and 700 C. The diffusion coefficients showed a considerable increase with increasing hydrogen concentration. At 600 C the values varied from 8.8×10^{-7} to 2.5×10^{-4} cm^2 per sec in the concentration range 62.1 to 66.0 a/o hydrogen. At 700 C the diffusion coefficients varied from 1.1×10^{-6} to 2.5×10^{-4} cm^2 per sec in the range 60.3 to 63.8 a/o hydrogen. Further permeation experiments will be made at 400 to 700 C.

Diffusion coefficients for hydrogen in beta zirconium in the range 650 to 850 C have been determined in this program and reported previously. The data indicated that there may be a slight change in diffusion rates with composition. In view of the large concentration effect noted in the delta-phase hydride, further measurements will be made in the beta phase, using permeation membranes of differing thicknesses, in order to establish whether or not the diffusion rates vary with concentration of hydrogen in beta zirconium.

I-1

I. SOLID HOMOGENEOUS FUELED REACTORS

W. H. Goldthwaite

A radiation-effects study of spherical (1-1/2 in. in diameter) fueled-graphite fuel elements is in progress as a part of the Pebble-Bed Reactor program. The release of gaseous fission products from selected fuel-element types is being studied by two experimental techniques: neutron activation followed by laboratory heat treatment in flowing helium, and high-level high-temperature irradiation in flowing helium. In both types of experiments, the helium stream is sampled for fission gases released from the specimens.

Measurements of Fission-Gas Release During Postirradiation
Heating of Fueled-Graphite Balls

R. Lieberman, P. Gluck, D. N. Sunderman,
and M. Pobereskin

During the past month, work has continued on the postirradiation study of fission-gas release from uranium oxide-graphite spheres at elevated temperatures.

A graphite sphere impregnated with UO_2 was irradiated in the BRR for 2 min at a neutron flux of 2.6×10^{10} nv. A week after irradiation the sphere was heated at three temperatures and the fission gas which was released was collected on activated charcoal. Helium, flowing at 3 liters per hr, was used as a carrier gas. The fission gas released was measured by radioassay of individual traps for xenon-133 by means of gamma-ray spectrometry. The data from this experiment are shown in Table I-1.

TABLE I-1. FISSION GAS RELEASE FROM UO_2 -GRAPHITE SPHERE E-12 AS A FUNCTION
OF TIME AND TEMPERATURE OF POSTIRRADIATION HEAT TREATMENT

Temperature, F	Time at Temperature, hr	Fraction of Total Present Xenon-133 Found(a)
1000	2	0.0361
	4	0.0390
1250	2	0.0095
	4	0.0120
1500	2	0.0208
	4	0.0251

(a) Cumulative value for time at temperature. The total amount of xenon present was calculated from reactor-flux and irradiation-time data.

Sphere 90, an admixture-type graphite-enriched UO_2 specimen, was irradiated at a neutron flux of 6.2×10^9 nv for 2 min. Two weeks after irradiation the sphere was placed in a preheated furnace and the xenon-133 released was measured at three temperatures. The sphere was allowed to cool to room temperature between heat treatments. The results of this experiment are given in Table I-2.

A sphere coated with SiC-silicon and containing normal UC was irradiated at a neutron flux of 8.7×10^{11} nv for 2 min. Two weeks after irradiation the sphere was heated using the same procedure and cycle described in the previous experiment. No xenon-133 was detected in any of the sample tubes. The minimum detection limit for this experiment was calculated to be 0.02 per cent based on the xenon-133 activity present in the sphere at the time of heating, and the minimum activity of the gas which could be measured on the gamma-ray spectrometer in a single charcoal trap. This specimen will be reactivated to a higher level and reheated at 1500 F in order to verify the above observation with increased sensitivity of detection of xenon-133.

The interpretation of the data has brought to the fore several factors which appear significant to the experimental program. First, the total quantity of fission gas released from the admixture sphere is small compared to the 10 per cent which would be expected to be recoiled from the fuel particles. Second, the initial release rate (first 30 min of heating) is larger than the long-term release rate. Since it is important to be able to predict the extent of coolant contamination due to long-lived daughters of the short-lived fission-gas isotopes, this initial release rate may be of more significance than the long-term one. Third, since the precursors of the more troublesome cooling-system contaminants are short lived, and released to only a small degree, a more significant contribution to the contamination of the coolant may be the diffusion of the long-lived daughters from the graphite matrix.

Further experimentation should be directed to the evaluation of the factors mentioned above. This would require a fairly detailed analysis of the microstructure of the graphite diluent. The pore-size distribution, extent of closed porosity, extent of surface diffusion versus free-atom diffusion in pores of various sizes and surface-area measurements would be of value. The resting places of fission-product recoils from the fuel particle should be determined to evaluate the fraction of fission-gas atoms immediately available for release by different mechanisms. The study of such parameters would yield a more sound basis for a description of the gas-release process, and also point the way to producing materials of higher integrity.

Work has been initiated on the measurement of fission-gas release from additional coated and uncoated spheres.

Fission-Product Release During Irradiation of Uranium Oxide-Graphite Spheres

G. E. Raines and W. H. Goldthwaite

Experimental operation of the sweep-gas capsule designed to study the fission-product-release behavior of spherical fueled-graphite specimens was initiated on March 1. Activity release during the initial operation from the impregnated specimens

TABLE I-2. FISSION-GAS RELEASE FROM UO_2 -GRAPHITE SPHERE 90 AS A FUNCTION OF TIME AND TEMPERATURE OF POSTIRRADIATION HEAT TREATMENT

Temperature, F	Time at Temperature, min	Temperature Change During Time, F	Fraction of Total Present Xenon-133 Found(a)
1000	10	<100 to 600	0.00064
	20	600 to 1000	0.00177
	40	At 1000	0.00290
	60	At 1000	0.00315
	120	At 1000	0.00361
	300	At 1000	0.00424
	390	At 1000	0.00455
1250	10	<100 to 1100	<0.0002
	20	1100 to 1250	0.00105
	40	At 1250	0.00248
	60	At 1250	0.00311
	120	At 1250	0.00431
	240	At 1250	0.00623
	360	At 1250	0.00761
1500	10	<100 to 1350	<0.0002
	20	1350 to 1500	0.00179
	40	At 1500	0.00516
	60	At 1500	0.00683
	120	At 1500	0.00908
	240	At 1500	0.01182
	360	At 1500	0.01298

(a) Cumulative value for time at temperature.

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(fuel-particle size probably as small as 1μ) was so great (as indicated from radiation levels from lines, etc.) that it was decided to isolate the inner capsule containing these specimens and concentrate on the admixture specimens (particle-size average about 130μ), from which the activity release was much smaller. Another attempt later in the month to operate the impregnated specimen capsule yielded the same type of behavior.

Two direct volume samples of the gas were obtained before the Battelle Research Reactor was shut down on March 9 for conversion to 2 megawatts. The results of the analyses of these samples are shown in Table I-3. Release rate was obtained by dividing the sample concentration by the volumetric flow rate which was 15.5 cm^3 per sec for Sample 1 and 13.7 cm^3 per sec for Sample 2. An immediate gamma scan of Sample 2 revealed the presence of quantities of krypton-87, krypton-88, and possibly krypton-83m and xenon-135m in addition to the three isotopes listed in the table.

TABLE I-3. RESULTS OF ANALYSES OF SAMPLES OF RELEASED FISSION GASES FROM ADMIXTURE-TYPE SPECIMENS

Isotope	Concentration in Sample, atoms per cm^3		Release Rate, atoms per sec		Ratio of Release Rate to Equilibrium Production Rate	
	Sample 1	Sample 2	Sample 1	Sample 2	Sample 1	Sample 2
Xenon-133	4.87×10^{11}	2.48×10^{12}	3.14×10^{10}	1.81×10^{11}	(a)	(a)
Xenon-135	1.47×10^{11}	3.48×10^{11}	9.47×10^9	2.54×10^{10}	0.006	0.017
Krypton-85m	3.53×10^{11}	3.83×10^{11}	2.28×10^{10}	2.80×10^{10}	0.052	0.064

(a) No values are given for these ratios for xenon-133 because the reactor had not been operating long enough for equilibrium production of this relatively long-half-life isotope to be achieved.

While the reactor was down, the capsule was moved from an in-core lattice position to a core-face position to allow operation at 2 megawatts, which was started March 24. About 48 hr of continuous operation of the admixture-specimen capsule was achieved, although plugging of all but one of the final activity-removal traps was encountered. After 48 hr, this final trap was not removing the activity and gas flow was stopped and the admixture-specimen capsule was then closed in. No samples were taken during this period because equilibrium operation was not achieved.

The plugging difficulties may have arisen from water vapor in the helium being frozen out in the liquid-nitrogen-cooled traps or may be due to a gradual movement of the fines present in the charcoal particles with flow to the bottom of the traps. A moisture trap has been installed in the helium inlet line and new revised activity-removal traps are being fabricated.

Operation, when possible, will be continued during early April with a second capsule of this type containing silicon-SiC-coated and SiC-coated specimens fabricated by two different vendors.

I-5 and I-6

Preirradiation Evaluation of Fueled-Graphite Spheres

J. F. Lynch, W. C. Riley, and W. H. Duckworth

Laboratory measurements of physical properties of fueled-graphite spheres are being made.

Six types of coated or impregnated fueled-graphite spheres from various vendors were subjected to a qualitative test for gas permeability. Spheres were placed in a bath of silicone oil heated to 375 F. The presence of streams of bubbles emanating from the surface of a sphere indicated that internally held gases were being emitted and, therefore, that the surface was not impervious. On the basis of this test, some spheres with a SiC-silicon coating and some of the SiC-coated spheres appeared to be impervious. In the other four types tested, no spheres appeared to be impervious.

Mechanical-property tests on the two types of spheres with impervious coatings revealed that spheres with SiC-silicon coatings were more resistant to abrasion and impact and had considerably higher compressive strengths than the SiC-coated spheres.

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J. PROBLEMS ASSOCIATED WITH THE RECOVERY OF SPENT REACTOR FUEL ELEMENTS

C. L. Peterson, P. D. Miller, O. M. Stewart, J. D. Jackson
W. C. Baytos, W. E. Bresler, T. E. Snoddy, and F. W. Fink

CORROSION STUDIES

The evaluation of materials of construction for the Darex, Sulfex-Thorex, Darex-Thorex, Zirflex, and Fluoride-Volatility processes has been continued. Titanium appears to withstand Darex dissolver solutions even under conditions of heat transfer to the liquid. The boiling Darex fission-product-recovery solution is too aggressive for handling with either Types 304 ELC or 347 stainless steel. The optimum heat-treating temperature found for Ni-0-nel exposed to boiling Initial Thorex solution is 1950 F. Hexavalent chromium in this solution leads to severe intergranular attack. Titanium shows promise for handling a combined Darex-Thorex dissolution. The corrosiveness of the NaF-LiF salt is greatly reduced in the presence of hydrofluorination of zirconium.

The Darex Process

Several materials of construction have been evaluated for the Darex process. Titanium appears to be the material of choice, with respect to corrosion, for the construction of critical components such as the dissolver vessel. In this vessel, stainless-clad or -bearing fuel elements are dissolved in dilute aqua regia. Following the dissolution, the chlorides are stripped from the solution and, after suitable adjustment, uranium is recovered by solvent extraction. The corrosiveness of the solution at several points in the circuit is under study.

Dissolver Studies With Titanium

Metallographic examination of the titanium steam tubes exposed to various boiling Darex dissolver solutions has been completed. Corrosion of the tube exposed for 2010 hr to the Initial Solution (2 M HCl, 5 M HNO₃) was estimated to range from 1.5 to 2.0 mils per month. Hairline cracks were observed at the 1000-hr inspection at one site on this tube. They did not appear on new sections replaced at the interface zone. It is now thought that these cracks were local imperfections present in the tubing before the start of the exposure.

A lower range of corrosion rates, 0.5 to 1 mil per month, was estimated for the tube exposed for 2049 hr to the Beginning Solution (Initial Solution acid plus small amounts of dissolved stainless steel and uranium). Crevices were observed in the

immersed weldment of this tube similar to crevices observed after 1000 hr. Sectioning and metallography showed these crevices to be reasonably shallow and blunt at the bottom. They do not appear to be a serious problem.

Little, if any, loss in thickness could be measured on the tube exposed for 2000 hr to the Darex dissolver solution. No localized attack was observed.

Thus, it is concluded that titanium can be expected to withstand all the normal operating conditions of the Darex dissolver without undergoing excessive attack. Attention is called to the well-known fact that titanium should never be used under conditions where only hydrochloric acid is present.

Fission-Product-Recovery Solutions

No stress-corrosion cracking was observed on stressed specimens of either Type 304 ELC or Type 347 stainless steel upon exposure for over 900 hr to boiling 3 M HNO_3 containing 60 g per liter of dissolved stainless steel, added as salts, and up to 800 ppm Cl^- . However, heavy attack occurred on the surfaces of all the specimens independent of the chloride content of the solutions. Knife-line attack was pronounced next to the weldments of Type 347 specimens. Intergranular attack was abundant, being more localized in the heat-affected areas of Type 347 and general over the surfaces of Type 304 ELC specimens. Very little hexavalent chromium could be found in these solutions, so this does not account for the high corrosion.

Stress-corrosion cracking does not seem to be a problem with the boiling fission-product-recovery solution. However, the other forms of corrosion are too excessive to allow either of these steels to be recommended for use. Lower temperatures might be expected to reduce the over-all corrosion but, on the other hand, might increase the susceptibility to cracking. Further study of this system will be postponed until temperature limits for operation have been established.

Darex Transfer Lines

Welded specimens of Type 304 ELC stainless steel are being stirred at 3 and 6 linear ft per sec in a final composition of the Darex dissolver held at 125 F. The objective of this study is to determine if existing stainless piping at ORNL can be used for transfer lines.

The Sulfex-Thorex Process

Dejacketing of stainless-clad thoria fuel elements using boiling 6 M H_2SO_4 constitutes the Sulfex process. The thoria is then dissolved with a boiling 13 M HNO_3 , 0.04 M F^- solution containing sufficient aluminum to mitigate corrosion.

J-3

Experiments With Ni-o-nel

Ni-o-nel specimens given various heat treatments after welding have been exposed for 1000 hr to boiling 13.0 M HNO_3 , 0.05 M F^- , 0.20 M Al^{+3} solutions. The heat treatments studied were: 1850 F, 0.5 hr, air or water quench; 1850 F, 2.25 hr, air or water quench; 1850 F, 10.0 hr, air quench; and 1950 F, 2.0 hr, air quench.

The difference in corrosion rates for these various heat treatments was not great. The lowest rate for air-quenched specimens was measured on those treated at 1950 F and was approximately 1.8 mils per month in the absence of hexavalent chromium. This treatment was used on the bench-scale Ni-o-nel dissolver.

During the study, one set of these specimens was exposed under conditions which allowed a large portion of the chromium from the corrosion products to be oxidized to the hexavalent state. The other duplicate set was so exposed that the chromium remained almost completely in the trivalent state. In the presence of hexavalent chromium, intergranular attack was excessive over the surfaces of the specimens and corrosion rates were increasing with continued exposure. With trivalent chromium, the rates remained fairly constant and little localized attack occurred, with the exception of a fibrous pitting in the weldment. Hexavalent chromium is not expected to form in the presence of the radioactivity of actual processing solutions.

Specimens of Ni-o-nel have been exposed to boiling 3.5 M H_2SO_4 containing 50 g per liter of dissolved stainless steel added as iron, chromium, and nickel salts. After 312 hr, the rates are in the 0.3 to 0.4-mil per month range, but intergranular attack is occurring. The chromium was added in the hexavalent state. These studies will be repeated with solutions prepared by steel dissolution rather than salt addition.

Experiments With Titanium

Consideration has been given to the possibility of conducting Thorex dissolutions in titanium equipment. If this proves feasible, fuel elements of the Consolidated Edison type might be de jacketed by the Darex process and core dissolution by Thorex could be carried out in the same vessel, thus eliminating the necessity for a Sulfex dissolver.

Titanium 75A specimens have been exposed to boiling 13 M HNO_3 , 0.04 M F^- solutions with no additions and with various concentrations of aluminum or thorium. The results have been very encouraging. Without additions, corrosion rates are in the 8 to 10-mil per month range after 240 hr. Aluminum additions decrease this to 1 to 2 mils per month. Thorium additions also help but do not appear to be quite so effective. No severe localized attack has been noted.

Bench Dissolution of Consolidated Edison Fuel Pins

Three bench-scale dissolvers have been constructed from Titanium 75A, stabilized Ni-o-nel, and Carpenter 20 Cb. Dissolution of Consolidated Edison fuel pins has been started in the titanium and Ni-o-nel dissolvers using the Darex-Thorex and

Sulfex-Thorex processes, respectively. After dissolution of about 25 pins in each dissolver, the vessel will be sectioned and studied metallographically for corrosion. Coupons of similar metal, inserted in each dissolver, will be examined periodically. Dissolutions in the Carpenter 20 Cb dissolver are being postponed.

The Zirflex Process

Zirflex processing of Zircaloy-2-clad fuel elements involves a dejacketing step using boiling 6 M NH_4F , 1 M NH_4NO_3 solution. This is followed by core dissolution in 10 M HNO_3 containing 0.05 M HF .

Scouting studies have shown that some corrosion inhibition is afforded Ni-o-nel and Type 309S niobium-stabilized stainless steel specimens exposed to the dejacketing solution in the presence of small additions of silicon and boron. Corrosion is reduced in these solutions by about 50 per cent.

The Fluoride-Volatility Process

INOR-8 specimens were evaluated as a construction material for the Fluoride-Volatility hydrofluorinator during an actual hydrofluorination of Zircaloy-2. The bath of molten 43 NaF-57 LiF (mole per cent) salt containing the Zircaloy-2 slugs was sparged with HF at a flow rate of 10 g per hr.

Dissolution was continued for 214 hr at 700 C and over 1900 g of Zircaloy-2 was dissolved, resulting in an increase in the salt volume of about 90 per cent. The inspection of the corrosion specimens has not been completed, but corrosion was much less severe than in previous runs in which no dissolution of zirconium was made. There appear to be a few areas of localized attack on some of the INOR-8 dip tubes. These will be examined metallographically.

STUDY OF THE EFFECT OF IRRADIATION ON CLADDING- AND CORE-DISSOLUTION PROCESSES

R. A. Ewing, D. K. Dieterly, and M. Pobereskin

The first irradiated fuel pins for study in the investigation to determine the effects of irradiation upon decladding and core dissolution are to be the Consolidated Edison type ($\text{ThO}_2\text{-UO}_2$; stainless steel cladding). The initial samples, now being irradiated, are expected to be available about May 1.

J-5 and J-6

Design of the remotely operated dissolution equipment is proceeding meanwhile, and most of the special equipment items needed are on order. Several preliminary dissolutions of stainless steel by the Sulfex process have been made to clarify design questions and to provide typical solutions for analytical-method development.

Selection and standardization of analytical procedures is under way and it is expected that this work will be completed (for the Sulfex process) during the next month. Fabrication of the dissolution equipment can begin as soon as the necessary items are received. It is expected that this can be done and preliminary dissolution experiments with "cold" fuel pins initiated during April.

K-1

K. DEVELOPMENTS FOR SRE, OMRE, AND OMR

F. A. Rough and J. E. Gates

EVALUATION OF URANIUM MONOCARBIDE AS A REACTOR FUEL

F. A. Rough

In studies of cast uranium monocarbide present emphasis is directed toward (1) irradiations in the MTR and (2) the preparation of a series of slugs for irradiation in the SRE.

Irradiation data on UC specimens containing about 5.0 w/o carbon are encouraging for exposures up to 5700 MWD/T at mean center temperatures of up to 1376 F. Irradiations of four capsules are in progress or scheduled for insertion in the MTR.

The preparation of a series of UC slugs, 0.645 in. in diameter, for irradiation in the SRE is described.

Irradiation of Uranium Monocarbide

D. Stahl and W. H. Goldthwaite

The irradiation of Capsules BMI-23-3 and BMI-23-4, containing uranium monocarbide specimens, is continuing in the MTR. Capsules BMI-23-5 and BMI-23-6 will follow BMI-23-3 and BMI-23-4, respectively, as shown by the schedule and other information listed in Table K-1.

During Cycle 117, Capsule BMI-23-3 operated with a central core temperature range of 1400 to 1550 F, while BMI-23-4, (Cycle 117) which is in a lower flux position, operated with a fairly stable core temperature of about 1230 F. In an attempt to avoid a midcycle refueling, a new reactor core loading was used for Cycle 118; the decreased neutron-flux density which resulted lowered the core temperatures of BMI-23-3 and BMI-23-4 to 1200 and 1000 F, respectively, at the start of the cycle. However, these temperatures increased steadily during Cycle 118 and continued to increase during Cycle 119 to a level at the end of March of 1390 and 1175 F, respectively. A summary of maximum, minimum, and mean core and surface temperatures for Capsules BMI-23-3 and BMI-23-4 is given in Table K-2.

According to reactor-quoted flux data, the top specimen in BMI-23-3 was in a flux of 1×10^{14} nv at the time it operated with a central core temperature of about 1500 F. These parameters, an unperturbed flux of 1×10^{14} nv and 1500 F core temperatures, comprise the original capsule design specifications. Thus, on this basis, there is reasonable assurance that the target burnup for the peak-flux specimen in this capsule can be accumulated during the cycles specified. BMI-23-4 is located in a reported flux field of about 0.8×10^{14} nv and is operating with a central core temperature lower than the 1500 F design point. These observations indicate that it will

TABLE K-1. MTR IRRADIATION SCHEDULE OF BMI-23 SERIES CAPSULES CONTAINING UC SPECIMENS

Capsule ^(a)	Carbon, w/o	Uranium Burnup Desired, MWD/T	Number of Cycles Required Based on an Unperturbed Flux of 1×10^{14} nv	MTR Position	Insertion	
					Cycle	Date
BMI-23-3	5.0	10,000	12	A28NE	116	1-12-59
BMI-23-4	5.0	7,500	9	A27SE	115	12-22-58
BMI-23-5	4.6	3,000	4	A28NE	To follow BMI-23-3	
BMI-23-6	4.8	5,000	6	A27SE	To follow BMI-23-4	

(a) Capsules BMI-23-1 and BMI-23-2, containing 5.0 w/o carbon, were irradiated for one and six cycles and were discharged from the MTR at the end of Cycle 107 and 115, respectively.

TABLE K-2. SUMMARIZED TEMPERATURE DATA FOR CAPSULES BMI-23-3 AND BMI-23-4 FROM INSERTION THROUGH CYCLE 117

Capsule	Cycle	Core Temperature of Top Specimen ^(a) , F			Surface Temperature, F					
		Maximum	Minimum	Mean	Top Specimen			Bottom Specimen		
					Maximum	Minimum	Mean	Maximum	Minimum	Mean
BMI-23-3	116	1360	900	1120	800	550	720	710	520	580
	117	1550	1400	1460	890	840	860	680	660	670
BMI-23-4	115	1320	1190	1250	790	750	760	620	570	590
	116	1280	1120	1190	740	690	720	670	510	590
	117	1260	1200	1230	750	710	730	580	510	530

(a) Specimens are 2 in. long and are aligned along the axis of the capsule. The core temperature of the top specimen is obtained with a thermocouple located in the hollow of that specimen. Surface temperatures are estimated from the readings of the thermocouples located near the surfaces of both top and bottom specimens.

K-3

be necessary to irradiate this capsule one or two cycles longer than the nine cycles now specified to reach the burnup desired.

From temperature-gradient data obtained from thermocouples located at varying distances from the center of the top specimen, heat-generation rates are being obtained, and from these estimates are being made of effective thermal conductivity of the UC specimens during irradiation. In both BMI-23-3 and BMI-23-4, the conductivity appears to be varying from about 5.0 to 6.1 Btu/(hr)(ft)(F), as compared with the corresponding out-of-pile conductivity of about 14.0 Btu/(hr)(ft)(F). This difference between in-pile and out-of-pile conductivities was also observed from data obtained during the irradiation of BMI-23-1 and BMI-23-2. It may be attributable to very early neutron damage, although this cannot be ascertained from the temperature data.

Postirradiation Examination of Uranium Monocarbide

S. Alfant, A. W. Hare, F. A. Rough, and R. F. Dickerson

A program is currently being conducted to determine the changes in the properties of uranium monocarbide produced by irradiation in a test reactor. In order to study these changes, specimens for seven irradiation capsules were prepared. One capsule containing twelve specimens was irradiated at the BRR, and six capsules, each containing five specimens, were sent to the MTR for irradiation.

At this time, three capsules have been irradiated, one capsule in the BRR and two in the MTR. Data compiled after examination of capsule BMI-23-1 and the BRR capsule showed that irradiation produced minor changes in density and diameter, indicating no severe effects on the dimensional stability of UC.

Capsule BMI-23-2 was discharged from the MTR and returned to the BMI Hot-Cell Facility after being irradiated in a thermal-neutron flux of approximately 1×10^{14} nv for six cycles. This capsule was irradiated to an estimated burnup of 5400 MWD/T or 0.7 a/o of the uranium at a maximum core temperature of 1830 F.

During the past month, postirradiation examinations of the specimens taken from capsule BMI-23-2 were completed. These examinations included dimensional and density measurements and macroscopic and microscopic examination of samples taken from each specimen. The release of krypton-85 was measured, and at this time a burnup determination based on an evaluation of the flux data obtained by dosimetry analysis is being completed. Burnup data from chemical analysis of the irradiated UC have not been obtained as yet.

Tables K-3, K-4, and K-5 list the preirradiation and postirradiation data for the specimens from Capsule BMI-23-2.

An examination of the data given in Tables K-3, K-4, and K-5 shows that UC appears to behave very well at the temperatures and estimated burnup levels which it experienced in Capsule BMI-23-2. The generally small changes in the measurements

K-4

TABLE K-3. DENSITY OF SPECIMENS FROM CAPSULE BMI-23-2 BEFORE AND AFTER IRRADIATION

Description	Specimen	Density, g per cm ³		Density Change, per cent
		Before Irradiation	After Irradiation	
Large pieces approximately 1 in. long	AI-37	13.428	13.18	2.5
	AI-26	13.3439	13.09	2.0
Small fragments	AI-37	13.428	13.19	2.5
	AI-26	13.3439	13.26	0.6

TABLE K-4. DIAMETER MEASUREMENTS OF SPECIMENS FROM CAPSULE BMI-23-2
BEFORE AND AFTER IRRADIATION

Specimen	Diameter, in.		Diameter Change, per cent
	Before Irradiation	After Irradiation	
AI-37	0.3732	0.3769	0.9
AI-26	0.3737	0.3773	0.9

TABLE K-5. MEASURED AND CALCULATED RELEASE OF KRYPTON-85 FROM
CAPSULE BMI-23-2

Vial	Krypton-85 Released, cm ³		
	Calculated Total	Measured	Calculated Release Due to Recoil
157	2.1 x 10 ⁻¹	9.7 x 10 ⁻⁵	8.8 x 10 ⁻⁵
155		24.1 x 10 ⁻⁵	

K-5

of diameter seem to show that the irradiation produced no severe effects on the dimensional stability of UC. However, consideration must be given to the fact that both visual and microscopic examinations revealed evidences of radial cracking and the presence of large voids in the structures of both specimens. Due to the cracking and breaking off of pieces from the ends of the specimens, it was not possible to make accurate measurements of length. It is evident that both specimens had decreased in density as a result of the irradiation they received. It is interesting to note the differences in the density changes recorded for the large and small piece taken from Specimen AI-26. At this time, no explanation can be given for this apparent divergence in results. However, due to the fact that pieces had fallen off all the specimens in the capsule, the possibility exists that the smaller piece may have come from another specimen.

It is of interest to calculate the approximate amount of krypton-85 released due to recoil, and then attempt to compare this amount to the measured amount of krypton-85 that could be formed with a burnup of 0.7 a/o uranium be $2.1 \times 10^{-1} \text{ cm}^3$. As can be seen in Table K-5, there are two sets of data for the measured release of krypton-85. These results are based on measurements made with two vials out of a total of six vials that were used to contain the released fission gas. Because of this difference in the measured amount of krypton-85 in both vials, it was decided to take a gamma count of the four remaining vials, and then evaluate and determine the correct measurement. Therefore, a comparison of the measured and calculated release of krypton-85 would not be feasible at this time.

A complete discussion and evaluation of the burnup of specimens from Capsule BM1-23-2 will be forthcoming in the future, when burnup data based on dosimetry and radiochemical burnup analyses are compiled and completed. It is felt that a comparison of the several methods used in estimating the burnup will serve to give a clearer picture of UC as a fuel material.

Preparation of UC Pins for Irradiation in the SRE

E. L. Foster and R. F. Dickerson

As part of a broad program to evaluate advanced fuel materials Atomics International is interested in the behavior of an appreciable quantity of UC cast shapes under operating power-reactor conditions. While results from cast shapes of UC exposed in the BRR and MTR appeared promising, confirmation is desired on fuel elements of the shape, size, and qualities that would be used in operating power reactors. Battelle is participating in part of the program to the extent of preparing the as-cast shapes and evaluating their preirradiation qualities.

The arc-melting and -casting technique is to be employed in preparing a quantity of shapes. Several factors determined the shape, size, and qualities of the specimens to be prepared. Among these were: the limitation of the specific casting technique, the qualities of the cast shapes themselves in terms of surface condition and internal soundness, and the quantity of material available (the weight of uranium at the enrichment desired). It is also desired that the specimens total at least 90 in. in length.

However, because of the total material limitation, this length is dependent upon the other factors to a great extent.

Since the diameter of the cast shapes was to be in the neighborhood of 0.600 in. as compared with the 0.4-in. diameter of previously prepared specimens, it was necessary to re-establish specific casting techniques. This has been done and the qualities of the shapes, so produced, defined.

The specimens will be cast in slightly oversized graphite thimble molds, and the diameter of the castings will be 0.645 in. A maximum of 0.035 in. on the diameter of these specimens will be removed by grinding to give a final specimen diameter of 0.610 in. At this diameter, and with this cleanup, it is likely that some casting surface imperfections will remain. The specimens will be cast in molds 2-3/8 in. long. At this length primary pipe will limit the specimen lengths to some degree. Small internal shrink voids have also been noted, usually occurring when the primary pipe is very slight. If the total of 90 in. of specimen length is to be obtained from the limited quantity of enriched uranium available, it is likely that specimens with some surface imperfections and internal shrink voids will be included in the groups to be irradiated. Since the effect of irradiation on all specimens with and without defects will be noted, some quantitative information of the effect of these imperfections will be forthcoming.

Routinely, complete notes of the qualities of the specimens will be recorded. Each specimen will be identified, radiographed, and photographed, and its density will be recorded. Metallographic examination and chemical analyses of selected specimens will also be made.

Since the specific techniques have been established for the larger diameter specimens and the evaluations in terms of recovery and specimen qualities have been completed, the preparation of the enriched specimens will begin as soon as the enriched uranium has been received.

POSTIRRADIATION STUDIES OF SRE, OMRE, AND OMR FUEL MATERIALS

J. E. Gates

An examination of OMR-type fuel plates has been completed and the data are being evaluated. An examination of OMRE-type fuel plates has been partially completed. Radiochemical burnup analyses and metallographic examinations of plate sections are still in progress.

The linear thermal expansion of two additional thorium-11 w/o uranium specimens is being measured.

K-7

OMR Fuel Plates

D. K. Dieterly

The examination of three aluminum-clad uranium-3.5 w/o molybdenum fuel plates from an OMR-type subassembly has been completed. The plates were gamma scanned, analyzed for burnup, and examined metallographically.

The results of the radiochemical burnup analyses are included in Table K-6. These data are being correlated with the gamma scans to obtain a burnup profile of each plate. The results of the metallographic examinations are being evaluated.

TABLE K-6. BURNUP OF OMR FUEL PLATES 341-1, 348-3, AND 347-1

Plate	Location of Burnup Specimen(a)	Uranium Burnup(b), a/o
341-1	2 in. from tongue end	0.122
	2 in. from groove end	0.172
348-3	2 in. from tongue end	0.087
	2 in. from groove end	0.121
347-1	2 in. from tongue end	0.069
	2 in. from groove end	0.087

(a) All specimens were about 1/4 by 1/4 in. and were located on the plate center line.

(b) Based on a 30-year half-life for cesium-137.

OMRE Fuel Elements

D. K. Dieterly

The examination of three stainless steel-clad OMRE fuel plates of UO₂ dispersed in stainless steel has been partially completed. The examination consisted of the preparation of stereomacrographs, optical and vernier-micrometer plate-thickness measurements, gamma scans, radiochemical burnup analyses, and metallographic examinations.

The stereomacrography, gamma scanning, and vernier-micrometer thickness measurements have been completed. The radiochemical burnup analyses and the metallographic examinations are in progress. An analysis of the data will be made as soon as the burnup analyses and metallographic examinations are completed.

K-8

SRE Fuel Materials

J. E. Gates

The unusual heating and cooling curves obtained during measurement of the linear-thermal-expansion coefficients of a thorium-11 w/o uranium specimen have prompted the measurement of expansion coefficients of additional specimens. It is planned to measure the thermal expansion of an unirradiated control specimen and of irradiated Specimen 15-7-F.

The results of the additional measurements will be reported as soon as they are available.

L-1

L. TANTALUM AND TANTALUM-ALLOY STUDIES

J. H. Stang

Battelle's program of assistance on Los Alamos Molten Plutonium Reactor Experiment (LAMPRE) is directed toward (1) developing improved materials for the containment of plutonium-alloy fuel mixtures and (2) investigating the effects of irradiation on the mechanical properties of tantalum. In the former study, metallurgical screening studies were conducted during March on a series of tantalum alloys (based on alloying additions of carbon, rhenium, and yttrium) prior to their shipment to LASL for corrosion exposures. Present findings indicate that the carbon and high-rhenium alloys are unsatisfactory on the basis of marked tendency toward embrittlement; other alloys in the group, however, possess acceptable characteristics such as ductility and a single-phase structure.

As pointed out in past reports, the tantalum-irradiation study has been delayed because of a lack of acceptable specimen material. During March, the appropriate material was received and specimens were prepared. According to the present schedule, specimen encapsulation will be completed by the end of April, and shipment of the capsules to NTRS will follow.

Development of Container Materials for LAMPRE Applications

D. C. Drennan, M. E. Langston, C. J. Slunder, and J. G. Dunleavy

Tantalum-alloy buttons containing 0.025, 0.1, and 0.25 w/o carbon or yttrium, and 1.5, 3, and 6 w/o rhenium, have been prepared by arc casting in a helium atmosphere. Radiographic and metallographic examinations indicate that the alloys are sound and homogeneous. The tantalum-carbon alloys contain a carbide phase throughout the structure and at the grain boundaries, whereas, the tantalum-yttrium and tantalum-rhenium alloys are void of a second phase.

These buttons (weighing 150 and 250 g) have been fabricated into 30-mil-thick strip specimens according to the following procedure:

- (1) Cold pressing with reductions of from 28 to 65 per cent
- (2) Machining the faces of the buttons to obtain a smooth surface and to eliminate surface contaminants
- (3) Cold cross rolling with reductions of from 2 to 15 per cent
- (4) Intermediate annealing at 2600 F for 2 hr to recrystallize the worked structure (portions of three alloys, tantalum-0.025 w/o carbon, tantalum-3 w/o rhenium, and tantalum-0.1 w/o yttrium, were annealed at 4000 F for 2 hr to further homogenize the alloys)

L-2

- (5) Cold cross rolling to 30-mil strip with approximately equal reductions in both directions (80 to 85 per cent total reduction).

Table L-1 presents the hardness and fabricability of these tantalum-base materials; the compositions given in this table are nominal inasmuch as the recovery rates of the alloy additions are not yet known. These data show that the hardness of the tantalum-rhenium alloys increased almost linearly with increasing additions of rhenium. The detrimental effect of carbon is shown by the difficulty encountered in fabricating the alloys containing more than 0.025 w/o carbon. The severe cracking of the tantalum-carbon alloys containing 0.1 and 0.25 w/o carbon is attributed to an excessive amount of carbide phase at the grain boundaries. The hardness data for the tantalum-carbon alloys indicate that carbon additions of 0.025 to 0.25 w/o cause about the same increase in hardness. The high hardness of the tantalum-0.1 w/o carbon alloy is thought to have been caused by gaseous contamination during melting. The addition of yttrium to tantalum apparently did not affect the fabricability. As indicated in the table, the only tantalum-yttrium alloy which showed any hardness effect was the one containing 0.25 w/o yttrium.

TABLE L-1. FABRICABILITY AND HARDNESS OF BINARY TANTALUM ALLOYS

Specimen	Nominal Composition (Balance Tantalum), w/o	As-Cast Hardness, VHN	Edge Cracking Limit, per cent reduction after intermediate anneal
27	Unalloyed Ta	89	73
28	1.5 Re	141	79
29	1.5 Re	140.5	80
30	3 Re	208	74
31	3 Re	197.5	74
32	6 Re	293	25
33	6 Re	271	23
35	0.025 C	152	79
36	0.1 C	267	33(a)
34	0.25 C	142.5	32(a)
43	0.25 C	156.5	32(a)
38	0.025 Y	85.5	78
39	0.1 Y	89.5	79
37	0.25 Y	117	76

(a) These specimens were not given an intermediate anneal because they cracked catastrophically after cold pressing and cold rolling with the indicated per cent reductions.

Some of the tantalum-base alloy specimens have been retained in the as-rolled condition while the others are being annealed at about 2600 F in vacuo. The as-rolled and the annealed specimens will be sent to LASL for plutonium-alloy compatibility testing as soon as the chemical analysis, hardness, and microstructure data are obtained. In addition to these properties, tensile data are being obtained on the tantalum-rhenium alloys.

Five new binary tantalum-alloy systems have been selected for future investigation; these will contain alloying additions of beryllium, hafnium, thorium, titanium, and

L-3 and L-4

zirconium. These addition elements were selected because of their thermodynamic potential for deoxidizing tantalum. In addition, a tantalum-3 w/o tungsten alloy will be prepared, using zone-refined high-purity tungsten melting stock.

Effect of Irradiation on Tantalum

J. A. DeMastry, F. R. Shober, F. A. Rough, and R. F. Dickerson

High-purity sheet tantalum for preparation of tensile specimens to be irradiated at the MTR or the ETR has been received from Los Alamos, and specimen fabrication has been completed. Six such specimens are to be encapsulated in each of two NaK-filled capsules. These capsules will be exposed to obtain approximately 7×10^{20} and 14×10^{20} nvt, respectively; these dosages are necessary for the transmutation of 1.5 and 3.0 w/o tantalum to tungsten. Calculations based on the estimated maximum gamma heating in the test reactors indicate that the specimen temperatures will be 200 ± 40 F, which is within 100 F of reactor process water.

While in anticipated LAMPRE service two types of damage to tantalum, fast-neutron damage and conversion of tantalum to tungsten, are expected, this investigation will concentrate on evaluating the conversion damage only. This will be done by (1) minimizing the effect of fast-neutron damage of the test specimens by irradiating in a position of high thermal flux and relatively low fast flux, if such space is available, and (2) comparing the tensile properties of the irradiated specimens with those of unirradiated tantalum and tantalum-tungsten alloys.

M-1 and M-2

M. DEVELOPMENTAL STUDIES FOR THE PWR

R. W. Dayton

Pressure Bonding of Zircaloy-2-Clad Fuel Elements Containing
Compartmented Oxide Fuel Plates

S. J. Paprocki, E. S. Hodge, D. C. Carmichael, and P. S. Gripshover

During the past month, this study was recessed.

N-1

N. DEVELOPMENT FOR THE MGCR

W. H. Goldthwaite

Research in support of the Maritime Gas-Cooled Reactor Program has continued. The fabrication and irradiation of candidate fuels and the effects of radiation on the corrosion and carbon transport by coolant impurities are being studied.

FABRICATION AND IRRADIATION OF FUEL MATERIALSFabrication of BeO-UO₂ Fuel Elements

A. K. Smalley, W. C. Riley, and W. H. Duckworth

Fuel-element cores containing 20 volume per cent UO₂ of 150- μ particle size dispersed in BeO are being developed.

Samples fabricated to date with these specifications have lower density than desired, and some difficulty in obtaining a uniform density distribution has been encountered. Various processing methods and types of UO₂ grains will be employed in an attempt to achieve maximum bulk density and uniformity. Also, the use of a new grade of BeO powder, UOX, reported to be readily sinterable will be investigated.

Fabrication of Graphite Fuel ElementsW. A. Hedden, A. B. Tripler, Jr., W. C. Riley,
and W. H. Duckworth

In this research program, fabrication of graphite fuel-element cores containing UC or UC₂ fuel particles is being studied.

A study of the effect of varying the binder-filler-fuel ratios in the mix on the fuel content of baked specimens has been completed. Application of the results of this study permit control of the fuel content to ± 0.5 volume per cent in the range of 10 to 30 volume per cent fuel.

A study of the effect of the heat-treatment cycle on the properties of UC-graphite and UC₂-graphite samples has been initiated. Prior work has indicated that, for UC-graphite samples, the maximum processing temperature will be limited to 2400 F because of conversion of UC to UC₂. The effect of this conversion on the properties of the fuel core will be studied. Also, the properties of these samples will be compared with those of UC₂-graphite samples.

Radiation-Effects Study of Potential Fuels

J. E. Gates, C. V. Weaver, and R. F. Dickerson

A capsule has been loaded with pellets of BeO fueled with UO_2 , graphite fueled with UC_2 , and graphite fueled with UC, and is to be irradiated under conditions simulating those occurring in the MGCR. Four pellets of each material were sealed in stainless steel tubes under a helium atmosphere. Three pairs of these specimens were included in the capsule, which was equipped with thermocouples and heaters. The irradiation will be performed in the Battelle Research Reactor.

The capsule, GA-3-1, will be loaded into the BRR on April 6, 1959. The specimens will operate at a surface temperature and heat flux of about 1500 F and 185,000 Btu/(hr)(ft²), respectively. A completion date of June 30, 1959, is still anticipated for the postirradiation examination of the irradiated pellets.

CORE-MATERIALS IRRADIATION PROGRAM

C. V. Weaver, N. E. Miller, and W. S. Diethorn

As part of the MGCR program, Battelle is investigating carbon-transport, metal-oxidation, and metal-carburization problems associated with a helium coolant and unclad graphite moderator.

A BRR irradiation of four quartz tori described in previous monthly reports has been delayed. The tori have been loaded with specimens, and sealed, but assembly of the outer stainless steel capsule has not been completed. It is expected that the capsule will be ready for reactor insertion in April. Reactor schedule permitting, the capsule will be inserted during the current reactor cycle.

The tori specimens reported last month have been changed to nickel, K-Monel, and niobium-1 w/o zirconium.

A brief bench study of the helium-graphite system, designed to support the post-irradiation interpretation of the specimen behavior and helium analysis, was completed in March. In this study AGOT graphite, previously vacuum outgassed for 48 hr at 1000 F, was exposed to commercial helium in a sealed tube. This outgassing procedure is identical to that used for the tori specimens. The concentration of gaseous impurities was followed mass-spectrometrically for several hundred hours in separate experiments at 1500 and 800 F. Significant amounts of impurities, chiefly nitrogen and carbon monoxide, were released at both temperatures. This study shows that graphite outgassing will make an important contribution to the impurity levels in the tori.

O-1 and O-2

O. ENGINEERING ASSISTANCE TO KAISER ENGINEERS

Reactor Flow Studies

L. J. Flanigan and H. R. Hazard

Air-flow studies in a quarter-scale model of the Partially Enriched Gas-Cooled Power Reactor are being conducted at Battelle to provide design data for the prototype. Past work includes construction of the plastic model parts, the model core, and the parts for the test setup.

In March, work was continued on all three phases. The plastic model parts are about 80 per cent complete. At Battelle, assembly of the core is about 90 per cent complete and about 60 per cent of the work on the test setup has been finished.

In April, assembly of the core and the test setup will be completed. Delivery of the plastic model parts is expected in April, following which the model will be assembled and installed in the test setup.

P-1

P. DEVELOPMENTAL STUDIES FOR THE SM-2

S. J. Paprocki

This work is being performed to assist Alco Products in the development of fuel absorber and suppressor materials for the SM-2 (formerly APRR-1B).

Techniques are being developed for fabricating fuel specimens for irradiation testing. The reference fuel-element plate consists of a stainless- UO_2 dispersion clad with stainless steel. Boron is incorporated into the fuel core as a dispersion of ZrB_2 .

Capsules are being designed for the irradiation testing of these fuel specimens. It is planned to conduct the irradiations in the ETR at a flux of 3×10^{14} nv. Because of this high flux, and since a high burnup must also be achieved, the capsule is designed for optimum heat transfer. Auxiliary resistance heaters will be incorporated into the capsules to maintain reference temperatures during the life of the irradiation.

Development of Fuel Materials

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J. M. Fackelmann, D. E. Lozier, and W. M. Pardue

Methods of fabricating stainless steel- UO_2 dispersion fuel elements for the SM-2 reactor core are being developed. The reference fuel element contains approximately 28 w/o UO_2 dispersed in a prealloyed Type 347 stainless matrix and is clad with Type 347 stainless. The fuel element is 0.040 in. thick with a 0.005-in.-thick cladding. A spherical UO_2 powder manufactured by Mallinckrodt is being used as the fuel material. Boron is incorporated into the fuel elements as a burnable poison.

Several methods for incorporating boron into the fuel elements were considered, and after evaluating these, a reference method was chosen. It involves the dispersion of an unreactive boron compound in the core matrix. This method appears to have the following advantages: (1) radiation damage is confined to the dispersed phase and surrounding matrix as with the UO_2 fuel, (2) the boron content can be accurately controlled, (3) boron losses arising during the sintering operation can be minimized or eliminated, and (4) there is no boron diffusion, consequently, problems of cladding corrosion and embrittlement are avoided.

After preliminary compatibility studies, zirconium diboride was chosen as the reference dispersion. This choice was based on previous in-pile tests which revealed that this material was capable of retaining substantial amounts of helium, and also on results of the compatibility studies which indicated that ZrB_2 is compatible with stainless steel up to 2300 F.

Major emphasis is being placed upon the development of fabrication procedures and on the preparation of irradiation specimens. Although ZrB_2 is thought to be stable in pure hydrogen, pure argon, or in vacuo, it will react with the oxygen which is

normally present to a limited degree in commercial hydrogen and argon atmospheres. In addition, commercially available ZrB_2 generally contains excess carbon and nitrogen which cannot be separated from the diboride during normal production operations. The ZrB_2 used in this investigation contains 0.55 w/o carbon and 0.21 w/o nitrogen.

Because of the sensitivity of the ZrB_2 and, consequently, the possibility of boron loss to the sintering atmosphere used, a series of tests has been conducted in which ZrB_2 powders have been fired in hydrogen and in vacuo. The weight losses have been measured, and the boron content was determined by analytical analysis. The results are listed in Table P-1 for ZrB_2 fired in hydrogen. Examination of the data leads to some interesting conclusions. First, the major boron loss and the major impurity loss occur within the first 3 hr at temperature. Although not shown in the table, the carbon content decreased from 0.55 w/o to 0.13 w/o after 3 hr at 2300 F. Second, the total loss is related to the firing temperature. Third, even after long periods of time, the boron loss is greater than the total weight loss due to oxidation of zirconium. Fourth, even though such losses should be predictable, long firing periods will not completely eliminate boron loss upon subsequent firing at the same temperature. One remaining factor for which results are not yet available is the amount of boron loss, if any, which occurs upon subsequent treatment at a lower temperature. Also, it should be noted that at any one temperature, boron loss will vary from run to run. For example, after 3 hr at 2300 F in hydrogen, boron contents of 15.5, 15.7, 16.5, and 16.9 w/o were reported for four different runs on ZrB_2 which initially contained 17.5 w/o boron. The exact conditions causing the difference in boron content have not yet been established.

On the other hand, some of the same lot of ZrB_2 fired for 3 hr at 2300 F in vacuo had a total weight loss of 0.2 per cent and a loss of only 0.57 per cent of the boron. When a good vacuum is maintained, it would be expected that the only boron loss would be due to volatilization of B_2O_3 already present plus an extremely small amount of B_2O_3 which could form during the firing.

TABLE P-1. BORON LOSS IN ZrB_2 POWDER FIRED IN HYDROGEN(a)

Temperature, F	Time, hr	Boron Content, w/o	Boron Loss, per cent	Total Weight Loss, g per 100 g ZrB_2	Total Boron Loss, g per 100 g ZrB_2
--	--	17.5	--	--	--
1600	3	16.1	9.15	1.04	1.6
2300	3	15.7	11.40	1.45	2.0
2300	6	15.2	14.80	2.01	2.6
2300	9	14.3	20.60	2.80	3.6

(a) -100 F entrance dew point, -35 F exit dew point.

Boron loss can be expected during sintering for the same reasons that losses occur upon firing of ZrB_2 powders. Correlation of chemical analyses with metallographic examinations indicates that the degree of boron loss can be predicted by metallographic examination. During the sintering operation, any oxygen present in either the atmosphere or the powder can react with the ZrB_2 to form ZrO_2 , B_2O_3 , or

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a complex oxide. X-ray diffraction indicates the presence of ZrB_2 and ZrO_2 after firing, which suggests that ZrO_2 and B_2O_3 are formed, with B_2O_3 being lost by vaporization. On the other hand, in vacuo, depending upon the pressure and leak rate attained, it may be possible to prevent the formation of additional oxide with boron losses being due only to vaporization of B_2O_3 initially present. The amount of boron loss as determined by chemical analysis appears to be related to the amount of the oxide phase (ZrO_2) present in the specimen. For example, there was a 50 w/o boron loss in one specimen sintered under poor conditions. Metallographic examination indicated that no ZrB_2 was present; only the oxide phase could be detected. In another specimen, roll-clad green but containing ZrB_2 fired at 1600 F for 3 hr, an oxide layer could be detected at the edge of each ZrB_2 particle. A specimen which contained some of the same fired powder but was sintered at 2200 F showed an 11 w/o boron loss and contained a higher percentage of ZrO_2 .

Several series of specimens have been prepared to compare hydrogen and vacuum-sintered compacts. Chemical analyses are not yet available, but comparisons of the microstructures have been made. In the specimens vacuum sintered 2 hr at 2200 F which contained ZrB_2 prefired 3 hr at 2300 in vacuo, little or no oxide could be detected in the individual ZrB_2 particles, but vacuum-sintered specimens containing ZrB_2 fired at 2300 F for 3 hr in hydrogen before use contained a small oxide layer around many of the particles. Specimens containing ZrB_2 fired 3 hr in hydrogen which were sintered at 2200 F in hydrogen contained a somewhat heavier oxide.

Even though presently available results indicate that better and more consistent results are obtained with vacuum-sintered compacts, irradiation specimens are being prepared by both methods and the specimens used will be selected on the basis of boron content and appearance of the microstructure.

Encapsulation Studies

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During the last report period, emphasis was placed on the fabrication of various components for the SM-2 specimen capsules which are to be irradiated at the ETR in an unperturbed thermal-neutron flux of 3×10^{14} nv. In addition, construction of several auxiliaries that must be furnished to the reactor site was initiated. Included are nuclear mock-ups for the ETR critical assembly. Two mock-ups are required, one simulating capsules in which only nominal-composition specimens (clad dispersions of 28 w/o UO_2 in stainless steel) are present and one for capsules which, in addition, contain a Eu_2O_3 specimen. Also being constructed are instrument-panel sets in which controllers and saturable-core reactors (to control the output of supplementary heaters within the capsules) are incorporated, and a three-phase, 100-kva transformer unit for powering the internal heaters.

As discussed in BMI-1324, the capsule design being employed is based on a 650 F nominal specimen-surface temperature and dissipation of the relatively large quantities of fission heat by a NaK-copper block-nickel shell scheme. Since there are a number of components per capsule (six flat-plate specimens, specimen mounts,

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copper blocks, thermocouples, dosimeters and six 2-1/4-kw sheathed resistance heaters in coils) and the available space for them is limited, considerable attention has recently been given to detailed problems such as capsule assembly and specimen mounting. A laboratory mock-up capsule is being used to aid in studying these problems.

At present, it would appear that the seven capsules which are being constructed during this period can be loaded and shipped to the ETR in time for the June 1 loading. Assuming there is no delay in reactor insertion, the irradiation of the five high-burnup capsules (total specimen burnup to 77 per cent uranium-235) in the group can be concluded by spring 1960, which is satisfactory to the over-all schedule. The other capsules in the group contain specimens to be irradiated to 37 a/o burnup, and these will be returned to the hot cell for examination near the end of the year.

RWD:CRT/all