Report No. BMI-1324
UC-25 Metallurgy and Ceramics
(TID-4500, 14th Ed.)

Contract No. W-7405-eng-92

PROGRESS RELATING TO CIVILIAN APPLICATIONS DURING FEBRUARY, 1959

by

Russell W. Dayton
Clyde R. Tipton, Jr.

March 1, 1959

Declassified June 12, 1959

BATTELLE MEMORIAL INSTITUTE
505 King Avenue
Columbus 1, Ohio

DISCLAIMER

TABLE OF CONTENTS

TABLE OF CONTENTS
(Continued)

Page

REPORTS RELATING TO CIVILIAN APPLICATIONS ISSUED DURING FEBRUARY, 1959

BMI-1312	"Gas-Pressure Bonding of Flat-Plate Fuel Assemblies", by Stan J. Paprocki, Edwin S. Hodge, Charles B. Boyer, and Ralph W. Getz.
BMI-1313	"Further Studies of Sintered Refractory Uranium Compounds", by Arch B. Tripler, Jr., M. Jack Snyder, and Winston H. Duckworth.
BMI-1315	"Progress Relating to Civilian Applications During January, 1959", by Russell W. Dayton and Clyde R. Tipton, Jr.

A. ASSISTANCE TO HAPO

F. R. Shober

Measurements of the thermal conductivity of unirradiated uranium and UO_2 have been completed. Preparations are being made for tests on irradiated material at the hot-cell facilities. The creep properties of 15 per cent cold-worked Zircaloy-2 are being investigated over the 290 to 400 C temperature range under both constant- and cyclic-temperature conditions. Tests of both types are in progress. The preparation of single crystals of molybdenum from a second lot of material has not been successful, and efforts to find a satisfactory starting material are continuing.

The physical distortion of graphite resulting from irradiation is being investigated. A sink-float density-measurement technique has been developed to measure the density distribution of graphite before and after irradiation. Improvements in the reproducibility of the technique have been achieved. The first case to be computed for in the program to evaluate possible loss-of-coolant incidents is that for a double 14-in. top break. Initial conditions for this case have been compiled and computations are under way. A program to investigate methods for end capping coextruded fuel pins has been initiated.

Thermal Conductivity of Uranium and UO_2

H. W. Deem and C. F. Lucks

Uranium

Thermal-conductivity measurements on unirradiated unclad natural uranium have been completed. Additional measurements made during February confirm the tentative values presented last month. These values will be used as a basis for measuring the effect of irradiation on the thermal conductivity of uranium.

During March, preparations will be continued for making thermal-conductivity measurements on irradiated uranium. It is planned to make the measurements on uranium irradiated at three exposure levels.

Uranium Oxide

The apparatus used in measuring the thermal conductivity of UO_2 was described in BMI-1301. During February, measurements on Specimen 68, a medium-density (91.9 per cent of theoretical) UO_2 sample, were completed. Table A-1 shows tentative interpolated thermal-conductivity values. Values adjusted to 100 per cent of theoretical density are also shown.

Six irradiated UO_2 specimens were received from HAPO in February. They have not yet been examined.

During the next period, work will continue on setting up the apparatus at the hot cell in preparation for measurements on the irradiated UO_2 specimens. The high-density specimen will be measured first.

TABLE A-1. TENTATIVE THERMAL-CONDUCTIVITY VALUES FOR UO_2 SPECIMEN 68[(a)]

Specimen density is 91.9 per cent of theoretical

Temperature, C	Thermal Conductivity, w/(cm)(C) d = 91.9[(b)]	d = 100[(c)]
100	0.071	0.077
200	0.057	0.062
300	0.048	0.052
400	0.042	0.046
500	0.037	0.040
600	0.033	0.036

(a) Specimen description:

Weight	24.2611 g
Diameter	0.250 in.
Length	3.001 in.
Density	10.07 g per cm^3

(b) d ≈ per cent of theoretical density.

(c) Thermal conductivity adjusted to 100 per cent of theoretical density by linear extrapolation.

Mechanical Properties of Zirconium Alloys

F. R. Shober and J. A. VanEcho

An evaluation is being made of the creep properties of annealed and 15 per cent cold-worked Zircaloy-2 at elevated temperatures in order to determine the suitability of Zircaloy-2 for process tubing in the NPR. The investigation requires creep tests both at constant temperature and under cyclic-temperature conditions. Constant-temperature tests will be conducted in the 290 to 400 C range. Cyclic temperatures are from room temperature to either 290 or 345 C. Tests on the cold-worked material are expected to continue for 15,000 hr.

Eighteen tests are in progress; 12 long-time (15,000 hr) tests are in progress on 15 per cent cold-worked material at constant temperature and 6 shorter (3000 hr) tests are in progress on annealed and cold-worked Zircaloy-2 at cyclic temperatures. The cyclic-temperature tests are intended to evaluate the creep properties of Zircaloy-2 under conditions representing anticipated reactor operation. Cyclic-temperature conditions appear to accelerate creep.

A-3

Preparation of Molybdenum Single Crystals

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

Single crystals of high-purity molybdenum are being grown for irradiation studies at another laboratory. These crystals are being grown in a modified Andrade furnace which produces grain growth in a molybdenum rod by utilizing a steep temperature gradient along the length of the specimen. The temperature gradient is obtained by resistance heating the molybdenum rod and water cooling the ends of the rod. The requirements for single-crystal growth in 1/8-in.-diameter rod have been established as an amperage of approximately 240 at 2.5 to 3.0 v for 5 to 6 hr. Six molybdenum single crystals have been grown. During the course of this investigation it has been noted that single crystals are grown only in rods from certain lots of molybdenum. Apparently either the purity or the fabrication history of the molybdenum influences the growth of single crystals.

A new lot of molybdenum rod has been received and attempts to grow single crystals resumed. At the present time no crystals have been grown from the new molybdenum rods. These attempts will continue until a satisfactory starting material is found.

Upon completion of six more molybdenum single crystals, attempts to grow single crystals of alpha zirconium will be made. Two slightly different approaches to this problem will be attempted. One method will be to anneal zirconium rod in the high-beta region, 1800 C, followed by an anneal at 850 C in the high-alpha region. The time required for this heat treatment has not been determined. The second method will be to heat treat in the high-alpha range, 825 to 850 C for several hours. A dynamic vacuum of 10^{-5} mm of mercury will be necessary for this study.

Physical Distortion of Graphite

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

Research to identify factors affecting irradiation-induced volume changes in graphite by means of sink-float density measurement was continued. Reproducibility of the experimental method was materially improved by substituting conical centrifuge tubes and coarse-porosity fritted-glass filters for the round-bottomed tubes and medium-porosity filters previously used.

The density distribution of samples of several size ranges from Specimens 127-86 and 127-96 were measured. The results were compared with the density distributions of the same size ranges from Specimen 127-33. There was a definite difference in the density distributions of the larger-particle-size ranges. However, within the accuracy of the measurement, identical results were obtained in the minus 270-mesh size range. Therefore, irradiated samples that have not been measured prior to irradiation will be sized to minus 270 mesh to facilitate comparison with unirradiated samples.

An irradiated bar of TS-GBF graphite, designated as N19-232, was received from HAPO. This bar was irradiated for 1105 MWD/T at about 450 C. A portion of the bar was crushed and the density distribution of the minus 270-mesh size range will be measured.

Unirradiated samples whose sink-float densities have been measured will be shipped to HAPO for irradiation during March.

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.

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

Modification of the existing digital program for application to the PRTR has been completed, and the various initial conditions needed for computation of the first case have been compiled.

The first case to be computed is that of a double 14-in. top break. A double break indicates that the pipe is sheared off and displaced so that discharge can come from both open ends of the loop. The element upon which the computation will be made will be one from the 6-Rod Ring. Although the flux is slightly higher in the 12-Rod Ring, calculations indicate that much more coolant flow will be available for the 12-Rod Ring; hence, the 6-Rod Ring becomes the most critical case during the blowdown phase where coolant is flowing through the core.

During the next month, it is planned to complete the computations for this first case and begin evaluating conditions for the second case.

End-Capping Methods for Coextruded Fuel Pins

R. M. Evans and J. J. Vagi

The objective of this program is to find end-sealing methods which will minimize the amount of core material that might be exposed to the reactor coolant if a leak should develop in the seal.

To accomplish the objective, a literature survey and analysis is planned, initially. This will be fortified by discussion of the subject with appropriate manufacturers of fuel elements. It is anticipated that this survey will show several possible sealing methods that are applicable to the problem. These will then be evaluated by laboratory fabrication of experimental seals.

Work on the literature has begun. It is planned that this work plus subject discussions will be completed during March.

B. DEVELOPMENTS FOR ALUMINUM-CLAD FUEL ELEMENTS

Preparation of Aluminum-Uranium Alloys

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

Hollow cylindrical tubes of aluminum-uranium alloys clad inside and outside with aluminum are desired as fuel elements for certain reactors operating at low temperatures. Fuel elements containing 20 w/o or less of uranium have been cast and fabricated successfully. It is desirable to increase the uranium content of the alloys. To accomplish this it is necessary to develop casting techniques for the production of hollow cylindrical alloy billets containing high concentrations of uranium. It is necessary also that the billets produced be amenable to coextrusion with aluminum. A program of research concerned with the development of casting techniques and with the improvement of the fabricating characteristics of these alloys is in progress.

Aluminum-35 w/o uranium extrusion billets up to 26 in. long with good interior surfaces have been produced by centrifugal-casting techniques. The removal of less than 0.075 in. of material from the inside diameter was sufficient to produce a clean surface on the inside of the billets. It was the objective of this series of castings to demonstrate the feasibility of a casting technique employing a bottom-pour crucible, a horizontal mold rotating about its axis, and a movable pouring spout. An aluminum-35 w/o uranium melt was poured at 2400 F into a pouring cup and spout heated to approximately 1400 F. The pouring cup and spout were withdrawn from the mold at a rate calculated to deposit the desired quantity of metal along the length of the mold. Pouring time (and spout-retraction time) was approximately 19 sec.

After turning off 0.075 in. from the inside surface of the castings produced by the above technique, the castings were sectioned longitudinally and transversely. Sections from one casting have been radiographed and metallographic specimens have been prepared. Radiographic examination of 3/8-in.-thick rings obtained from each end and the center of the casting revealed sound material from end to end and inside to outside. As would be expected, an area between the inside and outside surfaces contained the greatest concentration of uranium. The uranium content at the outer wall was near nominal. The uranium content increased in the center area and then decreased toward the inner wall. The evaluation of other ingots will include radiography of sections and metallography and chemical analyses of samples taken from areas selected after radiographic examinations.

Since it is desirable to improve the fabricability of the alloys and since it has been shown that certain ternary additions increase the volume of the aluminum matrix through the retention of UAl_3 instead of UAl_4, the effects of these additions on the castability and the extrudability of the binary aluminum-35 w/o uranium alloy are being investigated. A series of 3-in.-diameter castings 12 in. long containing ternary additions of germanium, lead, niobium, palladium, tin, silicon, and zirconium has been poured. From these castings, specimens for radiographic examination, metallography, extrusion, and tensile tests have been obtained. Preliminary metallographic examination of the as-cast material indicates that the structures of the alloys are the same as those found in 1-in.-diameter castings. However, radiographic examination indicates that both longitudinal and transverse segregation is more severe in the 3-in.-diameter castings. The alloys containing lead, palladium, and tin exhibit very little segregation in either the 1-in.-diameter or the 3-in.-diameter castings. The lower cooling rate of the large castings is thought responsible for the increased segregation noted. A comparison of the extent of segregation occurring in the castings containing ternary additions and those containing only aluminum and uranium will be made. Extrudability tests are being conducted at 800 F utilizing a flat-face die. Control alloys of aluminum-25 w/o uranium and aluminum-35 w/o uranium have been extruded and examined visually and by metallographic techniques. These control alloys are to serve as a basis for evaluating the fabricability of the alloys containing the ternary additions. Tensile specimens of all alloys are being prepared. Tensile tests will be conducted at two different strain rates to determine the effects of the additions on the strain-rate sensitivity of the alloys.

Future studies concerned with the effects of ternary additions will be confined to the evaluation of the castings produced as outlined above and with the production of full-size extrusion billets of these alloys. The casting studies will be terminated upon the evaluation of the ingots produced and with the production of ingots to be forwarded to another site for further studies.

C. RADIOISOTOPE AND RADIATION APPLICATIONS

P. Schall

The effort on the development of radioisotope quality-control techniques in the cement industry has been devoted to two areas during the past month: (1) analysis for calcium using a radiometric end point for an EDTA titration and (2) determination of the effect of magnesium concentration on the accuracy of the analytical technique developed previously for this constituent.

The use of a radiometric end point continues to look promising although the desired accuracy has not yet been achieved in the calcium analysis. The accuracy of the magnesium analysis, by titration with phosphorus-32-labeled ammonium phosphate, is poor at low magnesium concentrations, possibly due to the slight solubility of the precipitate.

The review of various processes which might utilize intrinsic radioisotopes as a means of process control is nearing completion. A detailed study of a process for nickel refining has been completed.

In the study of the radiation chemistry of inclusion compounds, additional samples of urea-hydrocarbon complex are being irradiated and will be used for a more detailed analysis of the chemical products formed. Samples of the ammonium nickel cyanide-benzene complex have been prepared, and irradiation of a batch of this material is scheduled for March.

Development of Radioactive-Tracer Quality-Control Systems

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

A radiometric method for the determination of calcium is under study. Evaluation of the precision of the method is not complete, but results accurate to within 2 per cent of the amount of calcium present have been achieved. In the experimental procedure calcium is titrated with 0.05 molar ethylenediaminetetracetic acid (EDTA) in a buffered, chloride-free solution. Insoluble silver iodate labeled with silver-110 is present in the solution as the end-point indicator. While being less stable than the calcium-EDTA complex, the silver complex is sufficiently stable to cause solution of the $AgIO_3$. In the course of the titration the silver-EDTA complex does not form until all the calcium has been complexed. At this point the activity in the solution increases rapidly. A plot of the radioactivity of the solution as a function of the EDTA volume may be resolved into two straight lines, the intersection of which is the end point of the titration. Radioassay is performed by drawing samples of the solution into a chamber surrounding a Geiger tube. A glass frit prevents passage of the undissolved silver iodate particles into the counting chamber.

An EDTA method for the determination of iron using yttrium-91 oxalate as the end-point indicator will be investigated during the next report period. A method for sulfate determination by precipitating barium-140 sulfate will also be studied.

The evaluation of the radiometric method for magnesium continues. Its precision has been determined at magnesium oxide concentrations of 1 mg per 100 ml and 5 mg per 100 ml. The results are shown in Table C-1.

Low results at the nominal 1 mg per 100 ml magnesium oxide concentration may be attributed to the solubility of magnesium ammonium phosphate in the ammonium hydroxide solution. Additional determinations of the precision of this method will be made at a nominal magnesium oxide concentration of 20 mg per 100 ml. The effect of interfering elements will also be determined during the next report period.

TABLE C-1. PRECISION OF MAGNESIUM ANALYSIS BY RADIOMETRIC METHOD

Concentration of MgO, mg per 100 ml				
Actual	Experimentally Determined	Average Experimentally Determined	Standard Deviation	Error, per cent
0.97	0.90	- -	- -	- -
	0.95	- -	- -	- -
	0.89	0.91	±0.026	6.2
4.85	4.86	- -	- -	- -
	4.88	- -	- -	- -
	4.84	- -	- -	- -
	4.82	4.85	±0.022	0.0

No experiments in activation analysis were performed during the present report period. Future work will include the comparison of various irradiated standards in order to evaluate any intrinsic error in the activation-analysis technique. Raw materials used in the manufacture of cement such as clay and slag will be irradiated in order to evaluate the activation-analysis technique on less complex mixtures than finished cement.

Use of Intrinsic Radioactive Tracers for Process Control

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

During February progress continued along the lines indicated in last month's report. A survey of the unit chemical process which will aid in making the choice of a specific process for further study was completed.

As an example of a possible radioactive-tracer application, a detailed study was made of a hydrometallurgical process for nickel refining. This study served to pinpoint areas of interest in this particular process, as well as to aid in the evaluation of other candidate processes which may be amenable to radiotracer control.

The task of reviewing a wide variety of processes for application of tracers is nearing completion. For these evaluations, the background of Battelle technologists familiar with each process is being utilized.

The final choice of process, subject to AEC approval, will be made early in the next report period. It is expected that the experimental phase of the evaluation will begin as soon as possible following this decision.

Radiation Chemistry of Inclusion Compounds

M. J. Oestmann, R. J. Jakobsen, E. J. Kahler,
and W. S. Diethorn

Investigation of two typical urea-hydrocarbon complexes, dodecane and octane, is being continued in order to fill in the details of the general picture of the radiation chemistry of urea-hydrocarbon complexes outlined last month.

A 400-g sample of the dodecane complex was irradiated to a dose of 2×10^8 rads during February. Next month the radiolysis products will be analyzed. A large sample of the octane complex will also be prepared and irradiated as soon as a sufficient supply of this hydrocarbon is available.

As reported last month, an irradiated complex of benzene and ammonia nickel cyanide will be analyzed for benzene derivatives containing the CN or NH_3 group. Small samples of the complex were prepared in February. These samples contain 1/3 mole of benzene for every mole of $Ni(CN)_2(NH_3)$ in agreement with several of the conflicting literature sources describing the composition of the complex. A larger batch of this complex will be prepared for an irradiation experiment in the Battelle Cobalt-60 Facility.

D. PROCESSING OF FEED MATERIALS

E. L. Foster

Uranium fuel slugs are fabricated from uranium ingots. Therefore, the qualities of the fuel slugs are dependent, to some extent, upon the qualities of the uranium ingot. National Lead Company of Ohio, as operator of the Feed Material Production Center, has a real and continuing interest in these qualities and the processes that produce the fuel slugs.

A program now in progress at Battelle is seeking a convenient method of examining the factors that are present in the casting step of the fabrication procedure. Improvements in the present process and future casting needs are desired.

Solidification of Uranium

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

This program is an investigation of the mechanism of solidification of uranium metal in cylindrical graphite molds. Heat-flow equations have been formulated and a theoretical model incorporating the characteristics of solidifying uranium has been developed to predict the transfer of heat from the ingot and mold during the freezing period. Physical properties of the materials involved and the change of these properties with temperature have been determined experimentally. These property values will be used in the analytic solutions of the solidification processes. Experiments designed to study the physical events occurring at the mold-metal interface as cooling progresses have also been made. By casting 100-lb ingots and using an electrical-probe network in the mold, indications of gap formations between the surfaces were observed shortly after pouring. Melts are also being poured to measure the temperature of the ingot, mold, and furnace from the time of pouring. The thermal data, so obtained, will be used to refine and check the data computed from the theoretical model.

A series of ingots 3 in. in diameter by 20 in. long has been cast to record temperature histories of the mold and ingot. The metal was melted and poured into graphite molds coated with $MgO \cdot ZrO_2$ wash. Each melt was heated to a temperature of 2550 F and held for 5 min prior to pouring. The procedure used to obtain temperature histories was to place thermocouples in the mold in such a manner as to measure temperatures in the metal at horizontal positions 0.25 in., 0.75 in., and 1.50 in. from the ingot surface and vertically on the outside mold wall at intervals of 5 in. One thermocouple was placed flush with the inside wall of the mold. In addition, thermocouples were placed on the inner wall of the furnace jacket and at positions above and below the mold. The thermocouples in these positions were intended to measure the temperatures in the immediate surroundings of the mold and ingot.

The data from these experiments are being evaluated. The results will be used to further refine the mathematical model.

After the conclusion of these studies, a similar program is planned for production-size castings weighing about 1300 lb.

E. SPRAY DEPOSITION OF CALCIUM METAL ON NICKEL OR INCONEL

R. M. Evans and R. E. Monroe

This is the second monthly report on a feasibility study to evaluate arc spraying as a method for coating nickel with calcium.

Work during the month was devoted to the construction of an arc-spraying device. Using existing equipment a device is now available with which spraying can be accomplished in an argon atmosphere but with the auxiliary equipment outside for better mechanical control. "Dry runs" with aluminum have been made, and it is expected that calcium spraying will begin early in March.

Calcium wire, nickel foil, and Inconel foil for use in this program were received during February.

F. RESEARCH FOR AEC REACTOR DEVELOPMENT DIVISION PROGRAM

S. J. Paprocki and R. F. Dickerson

REACTOR MATERIALS AND COMPONENTS

R. F. Dickerson

In an attempt to reduce the amount of trivalent La_2O_3 or Y_2O_3 needed to stabilize UO_2, ternary additions of CaO have been used. The UO_2-Y_2O_3-CaO bodies were poor from the point of view of stabilization. The UO_2-La_2O_3-CaO system seems more favorable. Early results indicate that a composition of UO_2-30 mole per cent La_2O_3-20 mole per cent CaO is competitive with the binary systems. Work has been continued on extending the pressure-temperature phase diagram of U_3O_8 to higher pressures and temperatures than have been previously reported.

The phase diagram for the zirconium-uranium alloy hydrides is nearing completion. X-ray diffraction patterns have been completed and the final hydrogen-absorption isotherm was measured. Irradiation of the zirconium hydride and zirconium-2 w/o uranium alloy hydride is progressing satisfactorily. One of the two test capsules will be irradiated for one more MTR cycle and the other for nine cycles.

The eight capsules containing Type 347 stainless steel are now in the K-7 position of the ETR. The fluxes in this position are much lower than expected and much lower than necessary in order to maintain the necessary lead time for the in-pile loop for which they are to provide damage information. The capsules will be repositioned in the best possible location. Two additional gamma-heat capsules, one with leads and one without leads, are being prepared.

The niobium-base alloys to be investigated as possible cladding materials for EBR advanced fuel elements have been fabricated with little or no difficulty. Specimens are being obtained for chemical analysis, and hardness and cold fabricability tests. A program concerned with the development of niobium-base alloys resistant to corrosion by high temperatures will be reported in this section. The results of work to date indicate that a binary alloy containing 45 a/o zirconium and a ternary alloy containing 25.5 a/o titanium and 6 a/o chromium continue to exhibit adherent tarnish films and low weight changes after 84 days in 680 F water. These alloys were prepared from commercial-purity niobium. Corrosion studies have been initiated on binary 7.5 a/o molybdenum and 12.5 a/o vanadium alloys prepared from high-purity niobium.

F-2

Valence Effects of Oxide Additions to Uranium Dioxide

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

An investigation is under way to determine the feasibility of reducing the amount of additive required to achieve a stable high-temperature nuclear fuel material. Previous work has shown that when certain trivalent additives are reacted with UO_2, a solid solution results which may be oxidized and reduced with only a change in lattice parameter. The present work has been directed toward partial substitution of CaO for either La_2O_3 or Y_2O_3. The CaO substitution was made to reduce the 60 mole per cent of La_2O_3 or Y_2O_3 required to produce a satisfactory stabilized material.

The results obtained to date indicate that the general effect of CaO substitution has been to stabilize the rare-earth "c" structure in preference to the UO_2 fluorite structure. The UO_2-Y_2O_3-CaO bodies are very poor from the stabilization standpoint. Bodies of these materials exhibit relatively high vapor pressure and rapid decomposition to powders with 3000 F oxidation.

If materials competitive to the UO_2-La_2O_3 or UO_2-Y_2O_3 binary systems are possible through this approach, it would appear that the UO_2-La_2O_3-CaO system is the more favorable. Early results indicated that a composition of UO_2-30 mole per cent La_2O_3-20 mole per cent CaO was competitive to the binary systems previously studied.

Recent work has indicated that more severe oxidation, resulting from the use of forced rather than static air oxidation, caused this composition to crack and to show marked volatility. A weight gain and better characteristics were noted for a composition of UO_2-40 mole per cent La_2O_3-20 mole per cent CaO, however. These results indicate that CaO may possibly be used, at least in small amounts, to obtain a partial reduction of the amount of trivalent additive necessary. It is not anticipated that any improvement from the standpoint of reduction of vapor pressure will result from this approach.

Work is under way to directly compare the performance of the best compositions of the binary systems UO_2-La_2O_3 and UO_2-Y_2O_3 with the best UO_2-La_2O_3-CaO composition. New compositions of the latter system will be prepared and evaluated to determine if anomalous results were obtained during the last oxidation test. The UO_2-20 mole per cent La_2O_3 composition has been prepared and will be used for electrical studies.

High-Pressure High-Temperature Solid-State Studies

W. B. Wilson and C. M. Schwartz

An investigation is being made to determine the feasibility of using combined ultrahigh pressure and high temperature as a means of producing new materials, specifically those of interest as high-temperature reactor components. Pressure, as a degree of freedom in the phase rule, may be capable of producing new materials with improved physical properties. Such materials may possibly result through the use of pressure to produce transformations to new structures or to catalyze reactions between substances which are normally nonreactive.

Minor modifications to the new high-pressure die were made to permit use in a larger press. Experimental work was continued to extend the pressure-temperature phase diagram of U_3O_8 to higher pressures and temperatures than have been previously reported.

Work will continue to extend the pressure and temperature range of the new die through use of the larger press. Studies of both reactions and the U_3O_8 pressure-temperature phase diagram will be extended to the higher pressures and temperatures obtained.

Fueled Zirconium Hydride Moderator

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

The investigation of the structure of uranium-zirconium alloy hydrides for potential use in fueled moderators is nearing completion. The final hydrogen-absorption isotherm planned for the zirconium-25 w/o uranium alloy was determined, and the high-temperature X-ray diffraction patterns of hydrides of this alloy were taken. Specimens of the 2 w/o uranium alloy which are undergoing radiation-damage studies are now in their seventh cycle in the MTR.

Structure and Pressure-Composition-Temperature Studies

The final hydrogen-absorption isotherm in the zirconium-25 w/o uranium alloy study was measured at 572 C, and the phase-boundary limits were obtained in the usual manner from Sieverts plots. These boundaries are defined by data given in Table F-1. Only two phase boundaries were found. This result was expected as interpolation of previous data indicated that only two phase boundaries should exist below 601 C.

TABLE F-1. PHASE BOUNDARIES IN THE ZIRCONIUM-25 w/o URANIUM ALLOY HYDRIDE SYSTEM AT 572 C

Phase Boundary	Hydrogen Absorption, cm^3 per g	Pressure, cm of mercury
1	23.2	0.024
4	116.5	0.024

X-ray diffraction patterns of the hydrided zirconium-25 w/o uranium alloy were completed during this report period. Analysis of the data remains to be completed in March.

Radiation-Damage Studies

Capsule BMI-20-1 was relocated during February in an effort to achieve a radiation temperature higher than that of the previous month. However, no increase in temperature resulted, and the average capsule temperatures continued as before: Thermocouple 1 – 1235 F and Thermocouple 5 – 1150 F.

Capsule BMI-20-2 continued at the same average temperatures as recorded previously: Thermocouple 1 – 1000 F and Thermocouple 5 – 1170 F.

Both capsules are now in their seventh cycle in the MTR. Capsule BMI-20-1 is scheduled to be irradiated for eight cycles, and Capsule BMI-20-2 for nine cycles.

Irradiation Surveillance Program on Type 347 Stainless Steel

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

The changes produced in mechanical properties of AISI Type 347 stainless steel as the result of exposure to a fast-neutron flux (neutrons having energies greater than 1 Mev) at 120 and 600 F are of interest to those concerned with operation of loops in the ETR. In view of this a program is in progress concerned with the determination of tensile, cyclic strain fatigue, and impact properties and the evaluation of these properties as measures of changes in the material after periods of exposure representing 6 months of service in the F-10 position of the ETR. A lead time of 6 months is necessary to provide sufficient time to determine the properties and to replace the in-pile-loop tube if the properties indicate possible unsafe operation. Exposure to a total flux representing 3 years of exposure in the F-10 position of the ETR is the maximum expected (approximately 1.1×10^{22} n per cm^2 total neutron exposure based on fast-flux measurements during Cycle 12).

The eight capsules being irradiated at process-water temperature, approximately 120 F, are now in the K-7 position of the ETR. Four capsules have been inserted in K7-NW and four in K7-SE in the same relative positions as they were in the F-10 position. That is, those that were in the top location in the F-10 position are in the top location of the K-7 position. These capsules were exposed in K-7 during ETR Cycle 12, which resulted in 7 days of exposure to a fast-neutron flux of 1.2×10^{14} n/(cm^2)(sec) in the vicinity of the reactor midplane, 2×10^{13} n/(cm^2)(sec) at the top, and 8×10^{13} n/(cm^2)(sec) at the bottom of the test position. The fluxes were lower than expected, and are much lower than needed to maintain the necessary lead time on the in-pile tube. It was expected that the flux in the K-7 position would be in excess to 2.8×10^{14} n/(cm^2)(sec). A test position of a higher fast flux will be secured and the capsules repositioned such that maximum use may be made of the available flux.

F-5

Five dosimeter assemblies exposed at the ETR in the F-10 and K-7 positions during Cycles 10, 11, and 12 have been returned to Battelle. The assemblies consist of thin-walled aluminum tubing into which have been placed short nickel wires and crystals of ammonium sulfate in individual quartz vials. Three assemblies containing nickel wire at the midplane were exposed in F-10-N, -S, and -W positions during Cycle 10. One containing eight nickel wires, spaced such that they covered the entire length of the test position, was also exposed in F-10-E during Cycle 10. This one was not returned to Battelle. One assembly containing eight nickel and eight ammonium sulfate dosimeters was exposed in F-10-E during Cycle 11. One other assembly containing eight nickel wires was exposed in the K-7 position center during Cycle 12. The dosimeters will be analyzed. Calibration will be performed by methods used by Phillips Petroleum Co. to correlate the fast-flux data from both sites. When the new test position is obtained, fast-flux mapping of this new position will be in order.

The duplicate of Lead Capsule BMI-24-17 is being assembled at BMI. It will be shipped with a nonlead capsule to the ETR. The lead capsule will be used at the ETR in an attempt to correlate gamma flux with temperatures obtained during irradiation in order to monitor those "hot capsules" which were designed to reach 600 F during irradiation. The nonlead capsule is a replacement for BMI-24-1.

Development of Niobium-Base Alloys

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

Advanced design features of the EBR include increased service temperatures of the reactor cladding material. The need for refractory reactor cladding materials has motivated a study of niobium and niobium-base alloys. The fabricability, weldability, thermal conductivity, and the mechanical strength of the alloys will be examined. Studies have commenced with the melting and hot fabrication of unalloyed niobium, vanadium-10 w/o titanium-1 w/o niobium (for comparison with niobium alloys), 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 alloys.

The niobium, niobium-base alloys, and vanadium-base alloy were prepared as 1 to 2-kg heats by consumable-electrode arc melting in water-cooled copper crucibles. The alloys were melted twice to insure homogeneity. Chromium losses by volatilization during melting were expected, so chromium was overcharged up to 100 per cent of the required alloy addition. The ingots were cropped and lathe-turned to remove surface imperfections. A small pipe was observed in the alloys containing 20 and 40 w/o titanium. The remaining ingots appeared dense and sound.

The ingots were encapsulated in molybdenum, soaked for 2 to 3 hr at 1370 C, and upset hammer forged out of the furnace at 1370 C. The ingots were annealed after each 10 per cent reduction, and were forged to a total reduction of approximately 80 per cent. Niobium-1 w/o chromium, niobium-2 w/o chromium, and niobium-10 w/o tantalum-2 w/o chromium alloys showed a number of edge cracks after removal from the molybdenum capsules. The other alloys exhibited no cracking.

Specimens for chemical analysis and hardness and cold-fabricability tests will be prepared from the fabricated alloys. Alloys which are cold-fabricable will be subjected to heat-treating studies.

Development of Corrosion-Resistant Niobium Alloys

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

The development of a strong low-cross-section corrosion-resistant niobium alloy for service in pressurized-water reactors was continued. This study was initiated by screening a number of commercial-purity niobium-base alloys for resistance to corrosion by 680 F water. On the basis of this work, a series of alloys is being prepared, using high-purity materials, to obtain mechanical-property data as well as more complete corrosion data on alloy compositions of interest.

The trends in corrosion behavior for the commercial-purity niobium alloys after 84 days of exposure in 680 F water are the same as those reported in BMI-1316 for 56 days of exposure. The data for 84 days of exposure are summarized in Table F-2. The 45 a/o zirconium binary and the 25.5 a/o titanium-6 a/o chromium ternary alloys continue to exhibit adherent tarnish films and low weight changes, and appear to be the most corrosion resistant of the alloys tested.

Corrosion studies have been started on binary 7.5 a/o molybdenum and 12.5 a/o vanadium alloys prepared by consumable-electrode arc melting with high-purity niobium. Unalloyed niobium samples from the commercial-purity base described above and from the high-purity base are being studied for comparison purposes. The corrosion results for exposure times of up to 14 days in 600 and 680 F water and 750 F steam are presented in Table F-3. These data duplicate results obtained with comparable commercial-base alloys after similar exposure times. Both the high-purity and commercial-purity unalloyed niobium are beginning to flake at 680 and 750 F. The 12.5 a/o vanadium alloy exhibits an adherent straw tarnish film and the 7.5 a/o molybdenum alloy is covered with a dark gray film at all test temperatures.

Vacuum creep testing of high-purity (electron-beam-melted) niobium and two alloys made with this metal were continued. These data are summarized in Table F-4.

Unalloyed niobium is very weak at 1200 F. Thus, one sample exposed to a 6000-psi stress underwent a total deformation of 5.8 per cent in 262 hr. Also, a sample loaded to 8000 psi failed in rupture, with an elongation of 17.9 per cent, after only 10 hr.

Alloying with either 12.58 a/o vanadium or 7.18 a/o molybdenum results in a considerable improvement in the creep resistance of niobium. This is apparent from the creep data given for these alloys in Table F-4. All of the 1200 F tests have been discontinued. These samples are being checked to ascertain freedom from oxygen contamination during the creep exposures. Testing of the 12.58 a/o vanadium alloy at

TABLE F-2. CORROSION DATA FOR COMMERCIAL-PURITY NIOBIUM-BASE ALLOYS

Alloy Content (Balance Niobium), a/o		Cross Section[a], barns per atom	Total Weight Change[b], mg per cm^2	Appearance of Corrosion Film
Nominal	Actual			
Unalloyed	--	1.15	--	Thin specimen completely oxidized at 42 days of exposure; corrosion rate at 28 days, -0.21 mg/(cm^2)(hr)
10 Zr	10.5 Zr	1.05	0.22	Dark gray
25 Zr	26.1 Zr	0.91	0.20	Dark gray, white streaks on one face
35 Zr	35.7 Zr	0.81	0.45	Dark gray
45 Zr	45.7 Zr	0.72	0.30	Shiny black
1 W	1.08 W	1.2	0.13	Dark gray, flaking
5 W	4.67 W	2.0	-11.5	Dark gray, flaking
10 W	9.56 W	2.9	--	Specimen cracked, off test 14 days
2.5 Mo	2.45 Mo	1.18	-1.51	Dark gray, flaking
5 Mo	5.20 Mo	1.21	0.40	Dark gray
7.5 Mo	7.40 Mo	1.25	0.34	Dark gray
5 V	4.42 V	1.31	0.03	Dark gray to brown
7.5 V	6.59 V	1.41	0.21	Dark gray to brown
10 V	8.93 V	1.50	0.17	Dark gray to brown
12.5 V	10.7 V	1.62	0.37	Dark gray to brown
17.5 V	13.7 V	1.80	0.06	Dark gray to brown
25 V	24.2 V	2.10	0.04	Dark gray to brown
5 Fe	4.90 Fe	1.2	0.11	Dark gray
10 Ti	9.41 Ti	1.6	0.12	Dark gray
20 Ti	18.8 Ti	2.1	0.07	Dark gray
25 Ti	24.3 Ti	2.3	0.15	Dark gray
30 Ti	30.5 Ti	2.6	0.18	Dark gray
35 Ti	33.8 Ti	2.8	0.19	Partly iridescent
7.5 Ti-0.5 Cr	12.0 Ti-0.5 Cr	1.54	0.25	Dark gray
19.5 Ti-2.5 Cr	20.2 Ti-2.1 Cr	2.11	0.20	Partly iridescent
25.5 Ti-6 Cr	28.2 Ti-6.1 Cr	2.44	0.06	Bright iridescent
10.5 Ti-3 Mo	12.0 Ti-4.2 Mo	1.69	0.28	Dark gray
16.5 Ti-4.5 Mo	17.4 Ti-6.2 Mo	2.00	0.18	Dark gray
21.5 Ti-6 Mo	23.1 Ti-7.8 Mo	2.24	0.13	Dark gray
10.5 Ti-3 V	10.4 Ti-5.0 V	1.72	0.22	Dark gray
17.5 Ti-6.5 V	16.1 Ti-8.4 V	2.14	0.15	Dark gray
24 Ti-11 V	22.6 Ti-11.0 V	2.67	0.28	Dark gray

(a) Estimated graphically on basis of nominal composition.

(b) One specimen for each alloy composition.

TABLE F-3. CORROSION DATA FOR ALLOYS PREPARED FROM ELECTRON-BEAM-MELTED NIOBIUM

	Total Weight Change After Exposure Shown, mg per cm^2		
	Water		Steam
Alloy Content (Balance Niobium)[a], a/o	600 F, 14 Days	680 F, 14 Days	750 F, 7 Days
Unalloyed, commercial Nb	1.93	0.17	0.52
	1.96	0.66	1.99
Unalloyed Nb, as electron-beam melted	1.57	0.77	-1.29
	1.90	0.82	-1.12
7.18 Mo[b]	0.11	0.22	0.56
	0.11	0.23	0.53
12.58 V[b]	0.07	0.09	0.22
	0.05	0.11	0.22

(a) Data are for duplicate specimens.

(b) Annealed 1 hr at 1500 C prior to corrosion testing.

TABLE F-4. SUMMARIZED CREEP BEHAVIOR OF HIGH-PURITY NIOBIUM AND NIOBIUM-BASE ALLOYS TESTED IN VACUUM AFTER ANNEALING 1 HR AT 1500 C (2730 F)

Alloy	Alloy Content (Balance Niobium), a/o	Temperature, F	Stress, psi	Time, hr	Total Deformation, per cent	Remarks
TP-37	100 Nb	1200	6,000	0.1	0.52	
				0.6	1.14	
				1.0	1.38	
				5.3	2.34	
				11.3	2.28	
				54.2	4.55	
				94.9	5.04	
				143.4	5.40	
				190	5.65	
				262	5.81	Minimum creep rate of 0.0018 per cent per hr
		70	0	--	5.88	Deformation cold, after unloading
NL-2	12.58 V	1200	20,000	0.1	0.124	
				0.4	0.165	
				501	0.165	
			30,000	502	0.240	Stress increased to 30,000 psi
				574	0.247	
				600	0.247	Minimum creep rate of 0.00005 per cent per hr
			40,000	600.1	0.290	Stress increased to 40,000 psi
				651	0.295	
				670	0.316	
				742	0.355	Minimum creep rate of 0.00054 per cent per hr
		70	0	--	0.252	Deformation cold, after unloading
		1380	35,000	0.1	0.316	
				0.2	0.305	
				0.9	0.325	
				65.7	0.508	Minimum creep rate of 0.0028 per cent per hr
NL-2	7.18 Mo	1200	20,000	--	0.111	Deformation on loading
				0.3	0.150	
				2	0.115	
				501	0.115	
			30,000	501.1	0.250	Stress increased to 30,000 psi
				506	0.265	
				525	0.282	
				555	0.300	
				595	0.300	
				669	0.300	Test discontinued
		70	0	--	0.181	Deformation cold, after unloading

1380 F is being continued and an additional sample of the 7.18 a/o molybdenum alloy has just been placed on test at 1380 F under a 30,000-psi stress.

Stainless steel pack rolling of the three ingot slabs listed below was carried out at 1800 F.

Ingot	Nominal Alloy Content (Balance Niobium), a/o
NL-4	45 Zr-5 Ti
NL-6	17.8 Ti-8.9 Mo
NL-7	10.5 Ti-3 Mo

These slabs were salvaged from ingots which had previously cracked during attempted hammer forging in air at 2200 F. A fourth ingot, containing 45 a/o zirconium and 3 a/o vanadium, was too badly cracked after forging to withstand rolling, and, hence, will be reprepared. Excellent-quality sheet was obtained from the other three ingot slabs. These are being pickled, ground, and annealed prior to evaluation for corrosion behavior in hot water and for mechanical properties at room and elevated temperatures.

Three additional scale-up alloys (containing, nominally, 10 a/o zirconium plus 10 a/o vanadium, 6.25 a/o zirconium plus 12.5 a/o vanadium, and 10 a/o titanium plus 5 a/o chromium) are being prepared by double consumable-electrode melting. In addition, arc melting of 40 binary and ternary 50-g niobium-base-alloy ingots is in process. These will be used to screen other alloy compositions of interest for resistance to corrosion by 680 F water and for hot strength.

STUDIES OF ALLOY FUELS

R. F. Dickerson

Specimens for thermal-conductivity, thermal-expansion, and electrical-resistivity measurements have been machined from wrought binary niobium-base alloys containing 10 and 20 w/o uranium. Since the alloys containing 60, 50, and 40 w/o uranium have not been successfully fabricated, specimens for the various property tests are being obtained from cast material at the same time preparations are being made to fabricate these particular alloys at higher temperatures. The niobium-30 w/o uranium alloy is being fabricated for specimen preparation.

Recrystallization studies of binary thorium-uranium alloys are in process, and preliminary results indicate that binary alloys containing 5, 10, 15, and 20 w/o uranium that have been reduced 60 per cent and heat treated for as long as 40 min do not recrystallize. Thermal analyses of the ternary alloy containing thorium-25 w/o uranium-25 w/o zirconium show that beta thorium exists as a single phase at temperatures above 1000 C. This phase is retained by a rapid cool. This is encouraging because the beta phase has a much higher solubility for alloying elements than does the alpha; therefore, the opportunity for improving corrosion resistance by alloying is expanded.

F-11

Development of Niobium-Uranium Alloys

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

Niobium-rich uranium alloys, potential high-temperature reactor fuel materials, have not been utilized due to lack of information available concerning their properties, fabricability, and irradiation stability. A study of the fabrication characteristics and mechanical and physical properties is being made.

The effect of impurities is being studied by using three grades of niobium as base material for the preparation of these alloys. The three grades are: one containing less than 0.17 w/o zirconium and 700 ppm oxygen, a second containing 0.7 w/o zirconium and 600 ppm oxygen, and a third containing less than 100 ppm zirconium and less than 300 ppm oxygen. The alloys of interest are uranium-40, -50, -60, -70, -80, and -90 w/o niobium; each of these alloys has been prepared using the three grades of niobium. The uranium-80 and -90 w/o niobium alloys have been successfully fabricated into slab material.

Table F-5 shows the results of fabrication of the uranium-70, -80, and -90 w/o niobium alloys. It appears that the uranium-70 w/o niobium alloy can be fabricated at 2500 F and three 3-lb ingots of this composition are being readied for fabrication. In order to obtain maximum information on the alloy system being studied, specimens for thermal-expansion, electrical-resistivity, and corrosion tests of the uranium-40, -50, and -60 w/o niobium alloys have been machined from slices taken directly from ingots. Fabrication of the uranium-40, -50, and -60 w/o niobium alloys appears to be dependent upon obtaining higher fabrication temperatures (2800 to 3200 F).

TABLE F-5. RESULTS OF HAMMER FORGING 2-IN.-DIAMETER NIOBIUM-URANIUM INGOTS IN MOLYBDENUM PACKS AT 2500 F FROM A HYDROGEN ATMOSPHERE

Nominal Composition (Balance Uranium), w/o	Thickness, in.		Reduction, per cent	Remarks
	Before Fabrication	After Fabrication		
90 Nb(a)	1.8125	0.420	76	Sound material
80 Nb(b)	2.9375	0.640	78	Sound material
80 Nb(c, d)	2.625	0.625	76	Sound material
70 Nb(c)	0.5625	0.185	67	One crack in material, otherwise sound

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

(b) Niobium used contains less than 300 ppm oxygen and less than 100 ppm zirconium.

(c) Niobium used contains 70 ppm oxygen and 0.17 w/o zirconium.

(d) Ingot was 1.750 in. in diameter.

Specimens for thermal-conductivity, thermal-expansion, and electrical-resistivity measurements have been machined from the uranium-80 and -90 w/o niobium alloys fabricated last month. The remaining slabs of material were then rolled at 1850 F from a helium-atmosphere furnace to 0.050-in. sheet. Blanks are being cut from these sheets. Tensile specimens and corrosion specimens will be machined from these blanks.

The uranium-70 w/o niobium ingots will be enclosed in molybdenum packs and fabricated at 2500 F. The canning operation is now in progress. The preparation of additional tensile specimens from the uranium-80 and -90 w/o niobium alloys which were fabricated this month will be started. Corrosion studies in water at 600 and 680 F and in 750 F steam for 1000 hr are planned. The specimens for corrosion testing of the uranium-70, -80, and -90 w/o niobium alloys in air and carbon dioxide have been prepared, and testing is to start next month. A capsule for corrosion testing in sodium is being designed. As soon as an acceptable design is made specimen preparation will begin.

In order to study the fabrication of the uranium-40, -50, and -60 w/o niobium alloys, higher temperatures (2800 to 3200 F) are needed. A furnace to obtain these temperatures has been designed, and construction of this furnace will start as soon as materials have been obtained.

Development of Thorium-Uranium Alloys

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

Improvement of the irradiation-damage resistance and of the corrosion resistance of thorium-uranium alloys is being sought by means of alloying and process control. Ternary and quaternary additions of molybdenum, niobium, and zirconium have been made for the purpose of stabilizing the gamma-uranium phase and for strengthening the thorium matrix.

Studies of binary thorium-uranium alloys are intended to outline the effects of hot rolling, cold rolling, and swaging on uranium particle distribution. Preparation of specimens for metallographic examination is currently under way. Recrystallization studies on the thorium-uranium binaries have also commenced; alloys of thorium-5, -10, -15, and -20 w/o uranium are being studied. They have been cold rolled to reductions of 20, 60, 80, and 90 per cent, and will be heat treated at several different temperatures for various periods of time. Preliminary results indicate that alloys reduced 60 per cent and heat treated at 600 C for as long as 40 min do not recrystallize. Data for higher temperatures should be available shortly.

Thermal analyses of a thorium-25 w/o uranium-25 w/o zirconium alloy and a thorium-20 w/o uranium-20 w/o zirconium alloy indicate that beta thorium exists as a single phase at temperatures above 1000 C. Results of the thermal analyses and tentative phase changes are listed on the following page.

F-13

Tentative Phase Transformations	Transformation Temperature, C Th-20 w/o U-20 w/o Zr	 Th-25 w/o U-25 w/o Zr
$\alpha Th + UZr_2 \longrightarrow \alpha Th + UZr_2 + \gamma UZr$	552	549
$\alpha Th + UZr_2 + \gamma UZr \longrightarrow \alpha Th + \gamma UZr$	585	587
$\alpha Th + \gamma UZr \longrightarrow \alpha Th + \gamma UZr + \beta Th$	966	990
$\alpha Th + \gamma UZr + \beta Th \longrightarrow \beta Th$	999	1002
$\beta Th \longrightarrow \beta Th$ + liquid	1200	1199
βTh + liquid $\longrightarrow$ liquid	1262	1239

Metallographic examination of these alloys quenched from 1000 C suggests that beta thorium can be retained by a rapid cool. X-ray diffraction examination is planned to substantiate this conclusion. The beta phase has a much higher solubility than the alpha-thorium allotrope for certain alloying elements; therefore, the retention of beta thorium may be considered desirable from the viewpoint of corrosion resistance.

FISSION-GAS RELEASE FROM REFRACTORY FUELS

J. Melehan, D. A. Vaughan, J. C. Smith, R. Barnes,
D. N. Sunderman, and F. A. Rough

Last month the objectives of this program were reviewed in some detail. The immediate objectives embrace the determination of diffusion coefficients for UO_2 of good geometry, use of these diffusion data to evaluate the mathematical model for release, and collection of data to permit prediction of gas release from practical UO_2 elements. Progress in two major areas is reported this month.

Determination of Diffusion Coefficients

In the first area, the determination of diffusion coefficients, an evaluation is being made on a lot of fused single-crystal UO_2 obtained from a commercial producer.

Unfortunately, the UO_2 appeared to have a surface coating of the uranium nitrides UN_2 and $UN_{1.5}$. There were also small particles of uranium metal dispersed throughout the UO_2. The nitride at the surface appeared to vary from practically none at the top to a high concentration at the bottom of the melt. The amount of metal decreased toward the interior of the UO_2 crystals. X-ray diffraction studies showed that the lattice parameter of the UO_2 phase varied considerably throughout the melt. One small area beneath the top crust gave a lattice parameter of 5.4707 A ± 0.0002 A, which approaches the value for stoichiometric UO_2. The lattice parameter of the UO_2 from other areas varied considerably, indicating a range in the oxygen/uranium ratio throughout the melt. The area which appears to be $UO_{2.01}$ constitutes a small portion of the material received, but may be sufficient for the studies of interest provided the microstructure reveals no uranium particles in the center of this section.

Various treatments intended to improve this UO_2 are being planned, and large-crystal UO_2 is being prepared by other methods in order to have an additional source of specimens for this study.

It is expected that once a suitable UO_2 is available, experiments will be initiated to measure gas release after irradiation in the radiochemical laboratory.

The assembly of the out-of-pile gas-collection system and construction of the electrical-resistance furnace will be initiated early in March. It is anticipated that this apparatus will be ready for operation during the following month. Concurrently with this effort, the induction-heating unit will be designed and built, and the components for the in-pile sampling system collected for assembly.

Preparation for In-Pile Study of Gas Release

During February, initial investigation began of various problems associated with the design of an in-pile apparatus for forthcoming studies of the release of fission gases from solid UO_2 specimens maintained at temperatures up to 3000 F. The fission gases are to be swept out of the irradiation zone of the apparatus by a flow of helium into out-of-pile collection traps from which quantitative determinations will be made to establish release rates. The apparatus is to be located in an 8-in. beam-tube facility in the Battelle Research Reactor. The nominal unperturbed thermal-neutron flux in this facility is 10^{12} nv. Each experiment will employ one specimen having a maximum dimension of approximately 1/2 in.; the specimen may be in the form of a sphere, a cylinder, or a cube. Access to the specimen is desired only during reactor downtime when the core is displaced from its operating position.

To achieve specimen temperature and control, induction heating and electrical-resistance heating are being considered. Both are capable of producing the desired specimen temperatures, but application of either to a beam-tube experiment poses unusual problems. Specific advantages of induction heating are:

(1) Broad temperature span available

(2) Simple in-pile components

(3) Rapid specimen heating

(4) Readily disposable in-pile materials.

The leading problem areas with induction heating stem from (1) high power losses in the long (approximately 11 ft) lead lines required, and (2) a limited selection of refractory materials (molybdenum, tantalum, tungsten, and possibly niobium) for the susceptor which is required between the inductor coils and the specimen. These problems are currently receiving attention in preliminary experiments. The use of shaped graphite is being considered for furnace designs based on resistance heating. The power requirements for such systems, shaping of the heating section, and protection and insulation of the graphite are also being investigated in simplified mock-up experiments.

In addition to exploring the characteristics of the above heating system, studies of the general compatibility of materials in the specimen zone are being made in the early laboratory experiments. In addition, problems associated with thermocouples in the 3000 F range will be evaluated.

The design of the gas collection systems and the development of analytical techniques for the in-pile and out-of-pile studies associated with the fission-gas-release program are continuing.

Preliminary designs for both the in-pile and out-of-pile apparatus have been completed. The fission-gas collection systems planned for both the in-pile and out-of-pile experiments are essentially the same. In the case of the in-pile experiments, the fission gas released from a specimen during irradiation will be continuously removed from the temperature-controlled section containing the specimen using helium as a carrier gas. After leaving the in-pile section, the carrier gas containing the fission products will be continuously monitored by a gamma-scintillation counter to follow the release of radioactive fission products as a function of time. The fission gas will then be separated from the carrier gas by collecting it on activated charcoal. Samples of the fission gas taken during various time intervals will be analyzed using a gamma-ray spectrometer to determine the identity and quantity of the fission products that are released. In the out-of-pile phase of the program, irradiated specimens will be heat treated at temperatures ranging to above 2000 C with the released fission gas being handled in the same manner. For temperatures up to 1600 C, electrical-resistance heating will be used. Induction heating will be used for the higher temperatures.

Within the next month, a decision will be reached about the kind of heating unit that can successfully be employed in the prototype apparatus. Also, other problems, particularly those concerned with remote specimen-handling techniques and shielding equipment, will be emphasized. It is expected that the final design phase can be started prior to April 1.

F-16

GENERAL FUEL-ELEMENT DEVELOPMENT

S. J. Paprocki

Cermet fuel materials containing 60 to 90 volume per cent of UO_2, UN, or UC have been fabricated by hot pressing, hot swaging, and gas-pressure bonding. These materials have been prepared by blending powders of the fuel compounds with metal powders and also by the direct fabrication of niobium-coated UO_2 powders. The fabricated fuel specimens are being evaluated on the basis of microstructure and physical and mechanical properties.

The gas-pressure-bonding technique is being investigated as a fabrication method for the solid-phase bonding of niobium and molybdenum fuel elements and assemblies. The feasibility of bonding these refractory elements has been established. Excellent bonds have been achieved with both molybdenum- and niobium-clad UO_2 fuel elements. The results are very promising, as the method is not restricted by the design of the fuel elements or assemblies.

Dispersions of stainless-UN and -UC containing 24 w/o of the fuel compound have been irradiated and are undergoing postirradiation evaluation. The examination of these specimens has not been completed; however, preliminary results indicate that these dispersions may be more resistant to irradiation than dispersions of stainless-UO_2 irradiated under comparable conditions to an equivalent uranium-235 burnup.

A study is being conducted to determine the mechanism of solid-state bonding that occurs during the joining of two metallic surfaces by the application of heat and pressure. The initial experimental results are not in complete agreement with the predicted mechanism.

Fabrication of Cermet Fuel Elements

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

Evaluations of fabrication techniques for producing cermets with densities 90 per cent of theoretical or better containing 60 to 90 volume per cent fuel are being made. Fabrication techniques include hot pressing, warm swaging, and pressure bonding of both hot-pressed and green-pressed cermets. Materials include mixtures of ceramic fuel powder with metal powders as well as niobium-coated UO_2 powders. Fabricated cores are being evaluated on the basis of microstructure and physical and mechanical properties.

Electrical-resistivity measurements are being made on hot-pressed and pressure-bonded cermets composed of UO_2 dispersed in matrix materials of molybdenum, niobium, and stainless steel. Since any directional properties are readily shown by electrical-resistivity measurements, values are being used as a rapid verification of changes in structure. Also, these measurements are being used in an attempt to calculate thermal conductivity. Thermal-conductivity measurements are being made on a rod of 80 volume per cent UO_2-stainless steel fabricated by hydrostatic pressing and on a plate of the same material produced by hot press forging. These measurements will be used as checks for calculated thermal-conductivity values. Calculated and measured values will be reported as they become available.

Cermets composed of 80 volume per cent UO_2-molybdenum have been hot press forged at 1900 and 2100 F to a density of 92 per cent of theoretical. Transverse microstructures indicate a continuous matrix of molybdenum in both cases; however, UO_2 fracture is more pronounced in the core fabricated at 2100 F. Machining of either core to obtain modulus-of-rupture specimens was unsuccessful due to cracking.

Cermets of 80 volume per cent UN dispersed in molybdenum and niobium have been hot press forged at 2100 F to densities of 89 and 90.6 per cent of theoretical, respectively. Metallographic examination of the molybdenum-matrix specimen indicated a continuous matrix comparable to the UO_2-molybdenum cermet. The microstructure of the niobium-matrix cermet showed a somewhat less uniform distribution of metal with less discrete-fuel-particle separation by the metal matrix. Also, indications of a reaction between the UN and niobium were noted. Samples heated in vacuum will also be examined to determine if a reaction has occurred.

Five cermets of 80 volume per cent UN-stainless steel with fuel sizes varied in the range of minus 80 to plus 325 mesh have been hot press forged at 1900 F to densities varying from 86 to 89 per cent of theoretical. The cermets fabricated with minus 80 plus 100-mesh and minus 100 plus 140-mesh UN were not broken when removed from the stainless steel hot-press frames; however, surface cracks developed when grinding modulus-of-rupture specimens. Cores fabricated using UN sizes finer than 140 mesh were cracked and broken when removed from the stainless steel hot-press frame. Microscopic examination of the cores indicated a discontinuous matrix in all cases, becoming more discontinuous when smaller UN fuel sizes were used.

Vapor-deposited niobium-coated UO_2 powder of 14.4 w/o niobium has been green compacted and hot press forged at 2100 F to 92 per cent of theoretical density. Microscopic examination showed a highly continuous matrix of metal with each UO_2 particle discrete as a result of its being surrounded by the niobium coating. Samples from this cermet are being prepared for electrical-resistivity measurements.

A pellet of the vapor-deposited niobium-coated UO_2 powder was green pressed in a 0.5-in.-diameter die and fitted into a stainless steel tube to which end plugs were welded. The assembly was evacuated subsequent to swaging at 1800 F to obtain a 52 per cent cross-sectional-area reduction, resulting in a density of 84.7 per cent of theoretical. Indications are that warm swaging is not a satisfactory method of producing dense rods with this type of material. Additional rod assemblies of this type are being prepared for pressure bonding.

F-18

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

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

The gas-pressure-bonding technique is being investigated as a method for cladding cermet and ceramic-type fuels with molybdenum or niobium. These metals are desirable as cladding materials because they have good high-temperature strength and favorable thermal-neutron-absorption cross sections. In gas-pressure bonding, there is very little deformation required to bring the components into intimate contact for solid-state bonding, and the problems associated with dimensional distortion of fuel elements fabricated by conventional techniques are eliminated. Small-scale fuel plates consisting of molybdenum- and niobium-clad uranium dioxide cores have been prepared by this technique.

Niobium-clad uranium dioxide fuel plates prepared with pieced niobium components have been satisfactorily bonded at 2200 F for 4 hr at 10,000 psi. The pieced niobium receptacle bonded satisfactorily at all of the mating niobium interfaces, and strong cladding-to-receptacle bonds were also obtained. Niobium plates bonded in this manner evidenced less ductility after bonding than before bonding. Portions of these fuel plates were analyzed to determine the cause of the reduced ductility encountered during pressure bonding. In addition, specimens of niobium were subjected to the same heat-treating cycle as that experienced inside of the autoclave to determine if ductility is normally decreased by this time-temperature cycle or if the decrease in ductility is a function of the autoclave atmosphere even though the fuel plates are enclosed in protective cans. A chemical analysis of an as-bonded fuel plate revealed the hydrogen content had increased from 1 ppm in the as-received niobium to 250 ppm in the as-bonded niobium. Iron contamination was also observed to have increased from 100 ppm to 1000 ppm during the bonding treatment; however, samples with an iron diffusion layer were machined to remove the iron-rich layer and no noticeable improvement in ductility was obtained. The specimens subjected to an equivalent heat treatment as that encountered in the autoclave did not evidence any significant decrease in ductility during heat treating; consequently, the embrittlement is introduced during the bonding operation. It has been recently determined that the hydrogen contamination can be removed from the autoclave atmosphere by heat treating the insulating material and furnace that are contained in the autoclave. Also, it is necessary to use gas with a very low dew point to eliminate a source of hydrogen from moisture carried by the high-pressure gas. All of these precautions will be observed before any additional niobium-clad specimens are bonded. The next niobium-clad specimen will consist of a four-compartment receptacle containing uranium dioxide cores. The receptacle will be prepared from pieced niobium components.

Molybdenum surface-preparation specimens were pressure bonded at increased pressure-bonding parameters of 4 hr at 2450 F and 10,000 psi. The specimens employed molybdenum plates which had been cold rolled from 0.060 in. to 0.015 in. in order to increase their initial ductility and increase the surface energy of the molybdenum for subsequent bonding. The best bonds were obtained with plates which were not surface prepared except to degrease in kerosene and alcohol prior to pressure bonding. A molybdenum fuel plate was also bonded at the increased pressure-bonding parameters. The fuel-plate receptacle consisted of a 0.040-in.-thick picture frame fabricated from pieced components from as-received material. The covers were fabricated from 0.060-in.-thick stock which had been cold rolled to 0.015 in. and the core insert was a 0.040-in.-thick by 1-in.-square uranium dioxide compact. The assembled fuel plate was bonded for 4 hr at 2450 F at 10,000 psi. Preliminary evaluation of this sample indicates there was sufficient flow of the pieced components to obtain the intimate contact required for bonding. The specimen appears well bonded in the areas examined and the integrity of the bond achieved is far superior to that obtained with previous specimens. The increased bonding parameters may be responsible for this improvement; however, the more ductile cladding with the cold-rolled surface also may be an important factor. Additional specimens will be prepared to establish the reason for the improved bonding observed in this specimen.

Specimens prepared from molybdenum and niobium which will incorporate multiple core receptacles and pieced components will be emphasized in future investigations. A detailed study of the embrittlement of niobium during pressure bonding is in process.

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

A. W. Hare and D. L. Keller

The high-temperature irradiation behavior of dispersion fuel elements consisting of 24 w/o UN and UC dispersed in stainless steel is being evaluated in this program.

Eight test specimens are presently undergoing hot-cell evaluation. These eight specimens were contained in two capsules (BMI-18-1 and BMI-18-2), and were exposed to either a one- or two-cycle irradiation at the MTR. The four specimens (two UN and two UC) in Capsule BMI-18-1 received a burnup estimated from thermocouple temperatures to be between 7 and 8 per cent of the uranium-235 atoms. The burnup of specimens in Capsule BMI-18-2 has been estimated to be between 2 and 3 per cent of the uranium-235 atoms. These estimated burnups will be revised when dosimeter and burnup analyses become available.

The preirradiation data and initial postirradiation data are given in Table F-6 for the eight specimens contained in Capsules BMI-18-1 and BMI-18-2. In addition to these data, it is worth noting that no cladding failures were observed during the visual examination of these specimens even though in several cases the blistering was quite pronounced.

The initial results of these first two capsule experiments are encouraging when compared to the performance of UO_2-stainless steel dispersions irradiated under similar conditions. However, a more detailed evaluation and interpretation (now in progress) of the irradiation results must be made before their full potential can be estimated.

TABLE F-6. PRE- AND POSTIRRADIATION MEASUREMENTS ON 24 w/o UN- AND UC-STAINLESS STEEL SPECIMENS

Capsule	Specimen	Fuel Compound	Average Dimensions, in.			Density, g per cc	Thickness Increase, in.	Density Decrease, per cent	Blister Height, in.	Estimated Maximum[b] Core Temperature During Last Reactor Cycle, F
			Length	Width	Thickness[a]					
					Preirradiation Measurements					
BMI-18-1	3-E	UN	1.4666	0.6906	0.0456	8.165	--	--	--	--
	4-A	UC	1.4698	0.6895	0.0468	8.085	--	--	--	--
	4-B	UN	1.4655	0.6801	0.0471	8.172	--	--	--	--
	4-B	UC	1.4654	0.6873	0.0473	8.093	--	--	--	--
BMI-18-2	4-C	UN	1.4704	0.6884	0.0465	8.157	--	--	--	--
	4-C	UC	1.4707	0.6861	0.0466	8.116	--	--	--	--
	4-D	UN	1.47702	0.6835	0.0461	8.172	--	--	--	--
	4-E	UC	1.4707	0.6896	0.0473	8.144	--	--	--	--
					Postirradiation Measurements					
BMI-18-1	3-E	UN	1.4703	0.6912	0.0462	8.135	0.0006	0.37	0	1600
	4-A	UC	1.4707	0.6914	0.0476	7.852	0.0008	2.88	0.017	1710
	4-B	UN	1.4641	0.6822	0.0482	7.995	0.0011	2.16	0.030	1800
	4-B	UC	1.4782	0.6892	0.0480	7.599	0.0007	6.12	0.024	1800
BMI-18-2	4-C	UN	1.4774	0.6878	0.0469	8.134	0.0004	0.28	0	1490
	4-C	UC	1.4781	0.6863	0.0468	8.110	0.0002	0.08	0	1600
	4-D	UN	1.4752	0.6838	0.0460	8.173	0.0001	Nil	0	1720
	4-E	UC	1.4739	0.6889	0.0474	8.121	0.0001	0.28	0	1490

(a) Postirradiation thicknesses are averaged over core area but do not include blister.

(b) Core temperatures are estimated from thermocouple temperatures adjacent to each specimen.

Four additional specimens (two UC and two UN dispersions in stainless steel) are presently being irradiated at the MTR at average core temperatures of about 1460 F. It is planned to irradiate this capsule (BMI-18-3) to three of four MTR cycles.

Factors Affecting Pressure Bonding

G. W. Cunningham and J. W. Spretnak

An investigation of the mechanism and kinetics of solid-state bonding of metals by the application of heat and pressure is in progress. The initial work has been concerned mainly with the bonding of copper, but iron, nickel, and zirconium will also be used in the study.

The initial interface between the two copper blocks has been previously described as resembling a series of V-shaped teeth resting on a flat surface. Examination of bonded and partially bonded specimens indicated that the teeth were acting as indentors and were actually being pressed into the flat surface. Therefore, a series of specimens pressed at several pressures at 1100 F and a series pressed for several times at 1150 F were prepared to further examine the mechanism of placing the metals in intimate contact. In the time series, specimens were pressed for periods of 5, 10, 15, 120, and 360 min at 10,000 psi and 1150 F. Very little difference could be detected in the volume of void space at the interface regardless of the time used. Although a series will be repeated at a lower pressure where large void areas can be expected, these results could be predicted from Bridgman's study of the collapse of hollow cylinders under pressure. Bridgman's work was conducted at room temperature where creep rates could be expected to be low, but he found that time at pressure apparently did not affect the final inside diameter of the cylinder.

On the other hand, the final pore size does appear to be directly related to the pressure. In a series of specimens pressed at 5,000, 10,000, and 15,000 psi at 1100 F for 2 hr, the mechanism of closing the pores can be followed qualitatively. (Due to an incorrect application of the load, exact pressures for this series are not known, but the ratio of the applied pressures is correct.) Metallographic examination indicates that the V-shaped teeth are pressed into the flat plate and at the same time the point of the V is flattened. There appears to be a less marked tendency for the flat plate to extrude into the root of the teeth. At the intermediate pressure, sufficient force was exerted at the root of the teeth to actually cause fracture and a partial separation of the teeth. At the higher pressure, no fractures were detected. An approximate measure of the area of the voids indicates that the void area is inversely proportional to the applied pressure. Whether or not this relationship proves to be true, it does not appear that voids in the model used can be closed by the application of pressure to the extent that they cannot be detected at 1000X with the optical microscope. This result appears to conflict with Bridgman's relation

$$\ln \frac{r_o}{r_i} = kP \quad ,$$

where r_o is the outside diameter and r_i is the inside diameter of hollow cylinders to which a hydrostatic pressure P has been applied. The equation indicates that an infinite pressure would be required to completely collapse the cylinder, but Bridgman reports the complete collapse of copper and soft steel cylinders. He noted that at a point where the voids were smaller than the average grain size the voids were no longer circular in shape, and that some other mechanism such as slip within the grains was responsible for the collapse rather than hydrostatic forces alone. It should be noted that at no time do the voids between machined surfaces have a circular shape. In the particular model chosen for this study, the voids resemble a right triangle with the flat machined surface being the hypotenuse.

Although essentially intimate contact can be achieved by the application of pressure at temperatures where the metal is reasonably ductile, the bond can by no means be considered perfect. The nature of the interface has not been established, but is apparently a disturbed layer approximately twice the thickness of an ordinary grain boundary which is produced by friction when the two surfaces are brought in contact. In a series of annealing studies at 1800 F, the interface appears to change to a row of small voids which usually, but not always, are connected by what appears to be ordinary grain boundaries. This change has been detected in specimens hot pressed at 1150 F and 10,000 psi, even when the specimen was subjected to 600,000 psi at room temperature before annealing. In this specimen, severely cold-worked grains, but no interfacial boundary, could be detected before annealing, but after annealing 2 hr at 1800 F a mixture of large and small recrystallized grains and the interfacial boundary with voids could be seen. In this case, small recrystallized grains predominated near the interface.

Additional studies as to the effect of time, temperature, pressure, and annealing after bonding are being conducted with the objective of determining the nature of the interfacial bond.

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 February, stress-cycling tests were concluded at the 1300 F and 1-cps condition. The data obtained are now being analyzed. Alteration of the Krouse fatigue machine, necessary for tests at 6 cpm, was completed during the month. Fatigue tests have now been initiated at 1500 F and 6 cpm.

During March, stress-cycling tests of Inconel will be continued at 1500 F and 6 cpm.

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 February, rotating-beam fatigue tests of INOR-8 were concluded at 1500 F, cycled at 100 and 3000 rpm. Tests are now being carried out at the same frequencies at 1100 F.

Tests will be continued during March to obtain additional data for cyclic frequencies of 100 and 3000 rpm at 1100 F.

H. PHYSICAL RESEARCH

F. A. Rough

Two projects supported by the AEC Division of Research and reported bimonthly are included in this section. Two additional projects are reported on alternate months.

Additional kinetic data on niobium-nitrogen reactions have been obtained at 300 and 1500 C supplementing earlier data at 400 to 1400 C. Additional data on diffusion coefficients of nitrogen in niobium have been obtained at 800 and 1100 C.

In the study of uranium-niobium constitution, high-purity diffusion couples heat treated at temperature are being analyzed by the electron-probe microbeam analyzer. Once the binary system is better established, based upon high-purity material, the effects of important impurities will be considered.

Niobium-Gas Reactions

W. M. Albrecht and W. D. Goode

A fundamental study of the reactions of nitrogen with niobium is being made. The study consists of determinations of reaction kinetics and mechanisms and includes determination of diffusion coefficients and solubility of nitrogen in niobium.

Results of kinetic experiments made at atmospheric pressure in the range 400 to 1400 C have been reported. Additional data have been obtained at 300 and 1500 C. At 300 C the reaction followed a cubic rate law with a rate constant of 1.1×10^{-3} $(\mu g/cm^2)^3/sec$. At 1500 C the reaction fits a parabolic law with a rate constant of 4.9×10^{-3} $(\mu g/cm^2)^2/sec$. A change in the parabolic reaction mechanism has been observed in the range 1000 to 1200 C. Kinetic and X-ray studies are being made in this range in order to determine precisely where change occurs. Also, the variation of the reaction rates with pressure is being investigated.

Diffusion coefficients and terminal solubilities for nitrogen in niobium obtained by the concentration-gradient method have been reported for 1200, 1300, and 1400 C. Additional data were obtained at 800 and 1100 C. At 800 C the diffusion coefficient is 6.8×10^{-10} cm^2 per sec and the terminal solubility is 0.28 a/o nitrogen. At 1100 C the diffusion coefficient is 4.1×10^{-8} cm^2 per sec and the terminal solubility is 1.2 a/o nitrogen. A diffusion specimen prepared at 1500 C is being analyzed. Check determinations of terminal solubilities are being made by metallographic examination of structures resulting from direct saturation of niobium with nitrogen. Preliminary results are in agreement with the values obtained from the concentration-gradient experiments. The metallographic studies are continuing.

H-2

Constitution of Uranium-Niobium Alloys

S. G. Epstein, A. A. Bauer, and R. F. Dickerson

An investigation of the constitution of uranium-niobium alloys is in progress. Diffusion couples are being employed in the investigation as a means of establishing the composition limits of the gamma-phase immiscibility gap in the system. Diffusion coefficients are also being determined at several temperatures. These binary alloy studies, which are being performed with high-purity materials, are intended to serve as a basis for the future determination of the effects of impurities on uranium-niobium-alloy constitution.

Diffusion couples have been heat treated at 1000 C for 50 hr, at 925 C for 70 hr, at 900 C for 68 hr, at 800 C for 1000 hr, and at 700 C for 2500 hr. The first three couples have been analyzed by an electron-probe analyzer. At 1000 C a satisfactory diffusion curve was obtained. No evidence of an immiscibility gap at this temperature was obtained. At 900 C a break in the diffusion curve indicated the presence of the immiscibility gap. However, the diffusion zone was not wide enough to permit precise determination of the immiscibility limits. Consequently, this couple has been reheated at 900 C for a total of 500 hr to allow reanalysis of a broader diffusion zone. Analysis of the data obtained from the couple heated at 925 C revealed that an impurity particle, probably a carbide, had been traversed during the analysis. Consequently, this couple will require reanalysis. However, based on the results obtained at 900 C it has been decided to increase the time of heat treatment at 925 C also, in order to broaden the diffusion zone before again analyzing the couple.

The three couples heated at 900, 800, and 700 C are now awaiting analysis.

The diffusion data for the couple heated at 1000 C have been analyzed graphically by the Matano method. The initial results are presented in Table H-1. An intermediate maximum in a plot of diffusion coefficients versus composition is obtained. The data are now being analyzed to determine the effect of experimental error on the calculated coefficients. This analysis may permit a conclusion to be drawn as to whether the intermediate maximum, which occurs at approximately 30 a/o uranium, is real.

TABLE H-1. DIFFUSION COEFFICIENTS FOR THE URANIUM-NIOBIUM SYSTEM AT 1000 C

Uranium Content (Balance Niobium), a/o	Diffusion Coefficient, D, 10^{-8} cm^2 per sec
5	5.15
10	5.99
20	8.06
30	8.26
40	4.20
50	4.18
60	4.23
70	6.64
80	10.87
90	34.47
95	37.45

I. SOLID HOMOGENEOUS FUELED REACTORS

W. H. Goldthwaite

A radiation-effects study of spherical (1-1/2 in. in diameter) fueled-graphite fuel elements in support of the Pebble Bed Reactor program is continuing. Currently, emphasis is being placed on the evaluation of the fission-product-retention characteristics of several types of coated and uncoated fuel elements. Impregnated and admixture types will be studied in experiments designed to reveal the identity and quantity of fission products released from each type.

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

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

A study of postirradiation fission-gas release from uranium oxide-graphite spheres at elevated temperatures has been started. This work is being carried out in conjunction with in-pile studies involving spheres of similar composition.

During February, the apparatus necessary for collecting and measuring released fission gas was constructed and tested.

A trial sphere, A-4 (impregnated), containing 5.1 g of natural uranium as UO_2 was irradiated in the BRR. After a 1-week cooling period, fission-gas release from the irradiated sphere was measured at three temperatures. Helium flowed over the heated sample at the rate of 3 liters per hr. It was found that more than 99 per cent of the xenon-133 released from the sphere was retained in the first of two activated-charcoal traps operated in series in the flow system. Upon completion of the heating cycle, the individual traps were isolated and xenon-133 concentrations were measured by means of gamma-ray spectrometry. Results are shown in Table I-1. This preliminary experiment has served to check out the operation of the apparatus for measurement of fission-gas release.

Work has been initiated on the irradiation and study of gas release of one other sphere of the impregnated type and one of the admixture type which contain enriched uranium. This will be followed by similar studies of coated specimens.

TABLE I-1. FISSION GAS RELEASE FROM UO_2-GRAPHITE SPHERE A-4 (IMPREGNATED) AS A FUNCTION OF TIME AND TEMPERATURE OF POSTIRRADIATION HEAT TREATMENT

Temperature, F	Time at Temperature, hr	Xenon-133 Found(a), fraction of total present(b)
1000	4	0.083
1000	8	0.090
1300	2	0.030
1500	2	0.036

(a) Cumulative value for time at temperature.
(b) Total present based on estimated burnup.

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

G. E. Raines and W. H. Goldthwaite

Final assembly and installation of the gas-flow and trapping apparatus for a study (discussed in BMI-1315) of the fission-product-release behavior during irradiation of spherical fueled-graphite specimens have been completed. The first of two in-pile capsules for this sytem is complete and ready for insertion in the reactor.

Irradiation of this first gas-flow capsule is scheduled to start in early March in a BRR core-lattice position. This capsule will only be operated for a short time (1 week to 10 days) before the BRR will be shut down for conversion to 2-megawatt operation. A new capsule will be designed for operation at a higher neutron flux and probably will be installed in early April. It is planned to utilize the same gas-flow and trapping equipment for both capsules.

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

CORROSION STUDIES

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

Materials of construction for the Darex, Sulfex-Thorex, Zirflex, and Fluoride-Volatility processes are being evaluated. Titanium still appears promising for the construction of the Darex dissolver. Stainless steel is being considered for use in other parts of the system. While no stress-corrosion cracking has been observed as a result of chloride contamination in these areas, the rate of attack is high. Investigations are under way to determine whether titanium vessels could be used to process Consolidated Edison fuel pins. Evaluations of Ni-o-nel and Carpenter 20 Cb for the Sulfex-Thorex process, Type 309S Cb, Ni-o-nel, and Carpenter 20 Cb for the Zirflex process, and INOR-8 for the Fluoride-Volatility process are being continued.

The Darex Process

Studies have been made of construction materials for several phases of the Darex process for fuel elements containing stainless steel as a diluent or cladding. In this process, uranium and stainless steel are dissolved in dilute aqua regia, the chlorides are removed by stripping, and, after suitable adjustment, uranium is recovered from the solution by solvent extraction.

Dissolver Studies With Titanium

Titanium steam tubes exposed to Beginning and Middle Darex solutions for 2000 hr or more have been sectioned. An accurate determination of the corrosion rate is difficult because of the shape of the tubes. Corrosion in the Beginning solution appears to be about 1 mil per month and, in the Middle solution, considerably less. Another tube is still being exposed to Initial Darex acid.

Fission-Product-Recovery Solutions

No stress-corrosion cracking was observed in specimens of Types 304 ELC or 347 stainless steel even after a 1250-hr exposure to boiling 3 M HNO_3 solution containing 60 g per liter dissolved stainless steel and 800 ppm Cl^-. The surfaces of these specimens were badly corroded with heavy intergranular attack. This does not seem to result from the Cl^- content, as other studies with Type 347 specimens with similar chloride-free solutions and in solutions containing 400 ppm Cl^- have produced equally heavy attack.

Darex Transfer Lines

A scouting experiment is planned in which specimens of Type 304 ELC stainless steel will be stirred in cold solution (1.75 M HCl, 1.2 M HNO_3, 6 g per liter uranium, 54 g per liter dissolved stainless steel) to determine whether stainless piping can be used in transferring this cold Darex solution after it leaves the dissolver.

The Sulfex-Thorex Process

In the contemplated Sulfex-Thorex process, stainless-clad fuel elements of thoria would be dejacketed with boiling sulfuric acid. The thoria would then be dissolved in a solution of 13 M HNO_3, 0.04 M F^-, containing sufficient aluminum to mitigate corrosion.

Experiments With Carpenter 20 Cb

A small dissolver has been constructed from Carpenter 20 Cb. It, along with a similar one constructed from Ni-o-nel, will be used to carry out dissolutions of Consolidated Edison fuel pins. These dissolvers will be inspected periodically for corrosion.

Experiments With Ni-o-nel

Metallographic study of Ni-o-nel specimens, heat treated for 0.5 hr at 1850 F, both before and after welding, and then exposed for 1000 hr to boiling 13.0 M HNO_3, 0.05 M F^-, 0.20 M Al^{+3}, has helped substantiate that heat treatment after welding imparts somewhat better corrosion resistance.

Heat treatments for longer periods at 1850 F and at 1950 F were made. These specimens are now being exposed to the boiling solution just mentioned. The exposures have not been long enough to differentiate between the various treatments and are being continued.

Scouting Experiments With Other Metals

Type 319L stainless steel was scouted in boiling 6 M H_2SO_4 and found to have an excessive corrosion rate. Reasonable rates of around 2 mils per month were measured for this material when exposed to boiling 13.0 M HNO_3, 0.05 M F^-, 0.20 M Al^{+3}.

Presently, titanium specimens are being exposed to the nitric-fluoride Thorex solutions containing varying amounts of aluminum and/or thorium. Preliminary results are encouraging. If the thoria dissolution could be carried out in titanium, then the dejacketing of Consolidated Edison fuel pins could be accomplished by means of the Darex process. These experiments will be continued.

The Zirflex Process

In the Zirflex process, Zircaloy-2-clad uranium fuel elements are declad with a boiling 6 M NH_4F, 1 M NH_4NO_3 solution. The core is then dissolved in 10 M HNO_3 containing 0.05 M HF and small amounts of zirconium and aluminum as contaminants.

Continued scouting experiments have shown Type 319L stainless steel to have high corrosion rates in the decladding solution. The attack on titanium was excessive in this solution.

Presently, corrosion rates for Ni-o-nel and Type 309S Cb are being evaluated in the decladding solution to which 0.05 M additions of boron and silicon have been added as possible corrosion inhibitors.

Cyclic studies are planned in which Ni-o-nel, Type 309S Cb, and possibly Carpenter 20 Cb will be exposed to both decladding and core-dissolution cycles. During these experiments, dissolution of zirconium and of UO_2 pellets will be carried out.

The Fluoride-Volatility Process

The investigation of various materials of construction for the hydrofluorinator of the Fluoride-Volatility process has been resolved to a study of INOR-8 for this purpose. In this hydrofluorinator, HF is sparged through a bath of molten fluoride salts to remove zirconium from fuel elements.

A run is nearly ready to start using a 43 NaF-57 LiF (mole per cent) salt at 700 C. Slugs of Zircaloy-2 will be dissolved continuously during this run and the salt will constantly change in composition. Specimens of INOR-8, both electrically connected to and insulated from the dissolving Zircaloy-2, will be used to study possible galvanic protection.

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

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

Currently proposed head-end treatment processes (Zirflex, Sulfex, Darex, etc.) for spent power reactor fuel elements are based primarily on data obtained with unirradiated fuels. Data on the effects of fuel irradiation on the amenability of fuel elements to these processes are quite limited. More specific data on irradiated fuels are needed before fuel-element reprocessing on a large scale can begin.

A study is being initiated to determine the nature and magnitude of the effects of irradiation upon the decladding and core dissolution of fuel elements. Of major concern are the effects of irradiation upon losses of fuel and fertile material in both steps. To be studied are both the direct effect of irradiation of the fuel itself, as a function of burnup, and the secondary effect of radioactivity levels in dissolving solutions.

An experimental program is being planned, and remotely operated dissolution equipment is currently being designed.

K. DEVELOPMENTS FOR SRE, OMRE, AND OMR

EVALUATION OF URANIUM MONOCARBIDE AS A REACTOR FUEL

F. A. Rough

The irradiation of uranium monocarbide (5 w/o carbon) for the Sodium Reactor Experiment has progressed routinely. The third capsule, irradiated to about 5000 MWD/T, operated at temperatures up to a maximum center fuel temperature of 1830 F. Preliminary observations on these specimens, along with details of several other capsules in various stages of progress, are reported in the following sections.

Irradiation of Uranium Monocarbide

D. Stahl and W. H. Goldthwaite

Irradiation of three capsules (one at the BRR and two at the MTR) containing specimens of uranium monocarbide is now complete. Four other MTR capsules are included in the program; two of these are currently being irradiated. The second capsule irradiated at the MTR, BMI-23-2, was removed from the A28NE position after Cycle 115; this capsule, which was irradiated for six cycles, is presently at the Battelle Hot Cell Facility for postirradiation examination. Burnup of specimens in BMI-23-2 is estimated to be about 4600 MWD/T for the top specimen and 5700 MWD/T for the bottom specimen.

Capsule BMI-23-3 was inserted into the A28NE position for Cycle 116 and will be irradiated for 12 cycles; the target burnup is 10,000 MWD/T. At Cycle 117 shutdown the capsule was lowered 2-1/2 in. and it is now operating at the 1500 F design level. Capsule BMI-23-4 is being irradiated in A27SE, a position having a lower flux than A28NE. The peak temperature in this capsule has been about 1250 F consistently.

Capsule BMI-23-5, containing uranium-4.6 w/o carbon, is scheduled for insertion into A27SE when BMI-23-4 is removed during Cycle 120 shutdown. Capsule BMI-23-6, containing 4.8 w/o carbon, which was to have followed BMI-23-3, will be inserted into a position comparable to A28NE if the space is available. Otherwise it will be inserted into A27SE when BMI-23-5 is removed (during Cycle 124 shutdown).

Postirradiation Examination of Uranium Monocarbide

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

The objective of this program is to determine the changes in the properties of uranium monocarbide produced by irradiation in a test reactor. Data on specimens from two capsules have been evaluated and MWD/T reported. Irradiations up to 1420 MWD/T were encouraging.

K-2

During the past month, Capsule BMI-23-2 was returned to the hot-cell facility for examination. This capsule, containing uranium-5.0 w/o carbon specimens, was irradiated in the A28NE position of the MTR in a thermal-neutron flux of approximately 1 x 10^{14} nv. It was charged in the reactor during Cycle 110 shutdown on September 8, 1958, and was discharged during Cycle 116 shutdown on January 12, 1959, after a six-cycle irradiation.

Upon arrival at the hot-cell facility, the outside dosimeters were removed, the capsule was punctured, and the released fission gas was collected in small glass vials for subsequent analysis. Macroexamination of the specimens after removal from the capsule revealed that all specimens were fractured and cracked to some extent. The specimens taken from this capsule after irradiation to an estimated burnup of 5000 MWD/T showed no evidence of any appreciable change in appearance as compared with earlier specimens irradiated to 1420 MWD/T.

Measurements of density and dimensions are being obtained and other post-irradiation tests are being performed, and the results will be forthcoming in the near future.

POSTIRRADIATION STUDIES OF SRE, OMRE, AND OMR FUEL MATERIALS

J. E. Gates

The examination of the thorium-11 w/o uranium specimens irradiated in Capsule Trains NAA-15-6 and NAA-15-7 has been completed. The results of the linear expansion measurements are discussed in this report.

Three OMR fuel plates have been gamma scanned and sectioned. Radiochemical burnup analyses and metallographic examinations are in progress. The examination of three OMRE fuel plates has not been initiated.

OMR Fuel Plates

D. K. Dieterly

The gamma scanning and sectioning of three plates of aluminum-clad uranium-3.5 w/o molybdenum alloy from an OMR-type fuel element recently removed from the OMRE have been completed. The plates were scanned through a collimating hole 0.25 in. in diameter and 3 ft in length. A shielded NAI(Tl) scintillation crystal and an RIDL Model 2300 spectrometer were used to measure the gamma intensity in the 700 to 750-kev range. Data from the charts of the scanner are presently being evaluated.

The OMR plates were sectioned in the following manner: two pieces about 1/4 in. square were removed from along the center line of each plate for radiochemical burnup analysis. One piece was removed 2 in. from the top and the other was taken 2 in. from the bottom of the plates. Two samples were cut from Plate OMR-347-1 for metallographic examination. Samples to be examined and analyzed at ORNL and CPP were also sectioned from Plate OMR-341-1.

Dissolution of the burnup specimens has been completed, and radiochemical analyses have been initiated. The metallographic examinations have also been initiated.

OMRE Fuel Elements

D. K. Dieterly

The examination of three stainless steel OMRE fuel plates has been rescheduled for March.

SRE Fuel Materials

G. E. Lamale, E. A. Eldridge, and H. W. Deem

The linear thermal expansion of three thorium-11 w/o uranium specimens has been studied. One specimen was an unirradiated control while the other two were irradiated. Table K-1 lists pertinent irradiation data for each specimen.

TABLE K-1. LIST OF THORIUM-11 w/o URANIUM SPECIMENS USED IN LINEAR-THERMAL-EXPANSION MEASUREMENTS

Capsule Train	Specimen	Burnup(a), total a/o	Irradiation Temperature, F
None	Control 1	0	0
NAA-15-7	15-7-B	0.63	950-1400
NAA-15-6	15-6-2	1.1	900-1100

(a) Determined by correlating radiochemically determined burnups with dosimeter data.

The specimens, originally about 1.39 in. long, were prepared for the thermal-expansion measurements by cutting off the end containing a thermocouple well. Specimen 15-7-B was first heated to 425 C and then to 1000 C. The first cycle was terminated due to difficulties with the length recorder. Specimen 15-6-2 and the control specimen were heated to 1000 C. The heating and cooling rate for all tests was about 3 C per min. The mean linear-thermal-expansion coefficients obtained from the heating curves over various temperature ranges are given in Table K-2. It should be noted that these coefficients apply only to the particular specimen measured since the normal expansion of the metal is increased by the presence of fission gas.

TABLE K-2. MEAN LINEAR-THERMAL-EXPANSION COEFFICENTS FOR THE IRRADIATED THORIUM-11 w/o URANIUM SPECIMENS

	Mean Linear-Thermal-Expansion Coefficient Over the Temperature Range Shown, 10^6 per C					
			Specimen 15-7-B			
	Control 1		First Heating	Second Heating	Specimen 15-6-2	
Temperature, C	Heating	Cooling	Cycle	Cycle	Heating	Cooling
20-100	6.9	6.3	9.0	11.4	9.3	10.5
20-200	8.9	7.3	9.9	12.0	9.7	10.7
20-250	9.4	8.3	--	20.9	--	--
20-300	8.9	8.7	10.6	43.9	10.4	11.0
20-350	--	8.9	13.0	--	--	--
20-400	9.8	8.9	30.4	40.8	11.1	11.2
20-500	10.8	9.4	--	35.6	11.9	11.7
20-600	11.8	10.1	--	32.2	12.8	12.1
20-700	12.8	10.8	--	29.8	14.0	12.6
20-800	13.4	11.4	--	28.0	15.3	13.0
20-900	13.0	11.8	--	25.9	17.4	13.2
20-975	--	--	--	27.4	25.6	13.1
20-995	12.0	12.3	--	--	--	--

An unusual phenomenon was noted in the heating and cooling curve for the control specimen and in the heating curve for Specimen 15-7-B. At a temperature between 200 and 400 C, both the curves for the control specimen showed a slight upward bulge. The two heating curves for Specimen 15-7-B showed a rapid increase in the expansion rate between 350 and 400 C in one case, and 250 and 300 C in the other. These aberrations did not appear to be due to instrumentation and did not occur in the heating or cooling curves for the other irradiated specimen or for the stainless steel calibration specimen.

The expansion rates of both irradiated specimens started to increase rapidly at about 900 C. This change was rather sudden for Specimen 15-7-B and gradual for Specimen 15-6-2. This behavior has been observed in other irradiated fuel alloys during expansion measurements and is probably the result of the internal pressure exerted by pockets of fission gas in the metal and a simultaneous decrease in the strength of the metal with increasing temperature. Both irradiated specimens exhibited surface cracks after the expansion measurements. The specimen with the highest burnup, 15-6-2, was in the worst condition. The second heating cycle usually performed on a specimen during expansion measurements was not run because of the condition of the specimens.

No further work is planned.

L. TANTALUM AND TANTALUM-ALLOY STUDIES

J. H. Stang

In the program being conducted to develop container materials for LAMPRE (Los Alamos Scientific Laboratory) applications, strip specimens are being prepared from a variety of tantalum-base alloys for subsequent corrosion studies in plutonium-alloy fuel mixtures at LASL. At present, tantalum-tungsten specimens in both the as-rolled and annealed conditions have been submitted. Emphasis is now being placed on investigating the fabricability of buttons of tantalum containing carbon, rhenium, and yttrium additions. These buttons were produced during February by arc casting.

Development of Container Materials for LAMPRE Applications

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

Specimens for plutonium-alloy compatibility testing (30-mil strip), consisting of tantalum and tantalum-tungsten alloys containing 1.5, 3, and 6 w/o tungsten, have been sent to LASL in the as-rolled and annealed conditions. Chemical-analysis, hardness, and grain-size data for these materials are given in Tables L-1 and L-2.

Observations of particular interest from these data are:

(1) In general, the total gaseous constituents of annealed specimens are higher than those in the cold-rolled specimens. This is somewhat surprising since stringent precautions were taken during the annealing operation. For example, the specimens were individually wrapped in tantalum foil and, during annealing, a furnace vacuum of approximately $2 \times 10^{-6}\ \mu$ was maintained.

(2) Both hardness and grain-size measurements indicate that the cold-rolled material was completely recrystallized during annealing at 2600 F, as expected.

(3) Tungsten additions have a retarding effect on grain growth during annealing. This effect may be of benefit to the behavior of these alloys during fuel-mixture exposures, since prior experience at LASL indicates that intergranular attack is a prime mechanism in the corrosion process.

Three new series of tantalum-base alloys have been prepared by arc melting in a helium atmosphere. One series consists of tantalum plus 1.5, 3, and 6 w/o rhenium, while the other two are intended to contain 0.025, 0.100, and 0.250 w/o additions of carbon or yttrium. An effort is being made to produce a uniform fine-grained structure in the final 30-mil strip specimens of these alloys by processing the buttons according to a schedule which features cold pressing to break up the cast

TABLE L-1. REPRESENTATIVE CHEMICAL ANALYSES AND HARDNESSES OF COLD-ROLLED (90 PER CENT REDUCTION) TANTALUM AND TANTALUM-TUNGSTEN 0.030-IN. STRIP SPECIMENS

Nominal Composition, w/o	Chemical Analysis, ppm				Hardness (5 to 10-Kg Load), VHN
	Oxygen	Hydrogen	Carbon	Nitrogen	
Unalloyed Ta	24	9	40	30	171
Unalloyed Ta	20	8	10	10	166
Unalloyed Ta	--	--	--	--	171
Ta-1 W	30	12	60	20	174
Ta-1.5 W	26	12	40	40	233
Ta-1.5 W	8	8	30	20	203
Ta-3 W	19	14	60	10	254
Ta-3 W	22	9	20	20	266
Ta-6 W	43	8	30	120	306
Ta-6 W	12	8	40	30	297

TABLE L-2. REPRESENTATIVE CHEMICAL ANALYSES, HARDNESSES, AND GRAIN SIZES OF ANNEALED[(a)] TANTALUM AND TANTALUM-TUNGSTEN 0.030-IN. STRIP SPECIMENS

Nominal Composition, w/o	Chemical Analysis, ppm				Hardness (5 to 10-Kg Load), VHN	Average Grain Diameter, mm
	Oxygen	Hydrogen	Carbon	Nitrogen		
Unalloyed Ta	50	1.7	40	50	112	--
Unalloyed Ta	17	0.6	30	30	99.5	0.10
Unalloyed Ta	45	0.4	20	50	111	--
Ta-1 W	46	2.0	50	70	140	0.042
Ta-1.5 W	53	1.2	20	30	145	--
Ta-1.5 W	14	0.4	20	10	134.5	0.060
Ta-3 W	22	1.0	50	50	184	--
Ta-3 W	20	0.5	20	20	175.5	0.036
Ta-6 W	93	0.6	10	140	243	--
Ta-6 W	19	0.1	30	30	222	0.025

(a) Cold rolled (90 per cent reduction) and vacuum annealed at 2600 F for 2 hr.

structure, intermediate and final recrystallization treatments, and cross rolling with equal reductions in both directions. In keeping with the findings at LASL, aqua regia will be substituted for a HF-HNO_3 bright-dip solution previously used as a cleaning agent for tantalum and its alloys.

Irradiation Damage of Tantalum

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

Tantalum sheet stock needed for tensile specimens and for preparation of tantalum-tungsten control specimens has not been received from Los Alamos. Work on this program will resume when materials are available. Recent word from LASL indicates that delivery to Battelle can be anticipated late in March.

M. DEVELOPMENTAL STUDIES FOR THE PWR

R. W. Dayton

The current program of flow studies in the PWR model has been completed, and the results have been included in the report draft.

There has been some further study of previously pressure bonded plate-type oxide fuel elements for the PWR. In these studies, as in earlier ones, it was found that intercompartmental leakage did not occur on samples which had been made with buffed graphite coatings on the UO_2.

Reactor Flow Studies

L. J. Flanigan and H. R. Hazard

Flow studies using air in a quarter-scale model of the PWR are being conducted to determine the effects of lower-plenum geometry on mixing and on flow distribution in Core 2. Previously reported work includes development of an optimum baffling arrangement for the 7.5-ft core design, construction and installation of a quarter-scale model of a similar design suitable for incorporation into the prototype, and initiation of flow studies to verify the design.

In February, the flow studies were completed and a draft of a final report covering all the studies made with the 9-ft core design and the 7.5-ft core design was prepared.

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

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

A flat-plate Zircaloy-2-clad fuel element containing compartmented uranium dioxide fuel is being considered for Core 2 of the PWR. The feasibility of preparing these elements by gas-pressure bonding is being investigated. In this process, the fuel elements are assembled from belt-abraded Zircaloy-strip components, edge welded, evacuated, and sealed to form a gastight assembly which is then gas-pressure bonded at 1550 F for 4 hr using a pressure of 10,000 psi of helium. Graphite coatings are sprayed on the uranium dioxide fuel before bonding in order to minimize reaction with the Zircaloy 2-cladding during bonding. One half of each element is evaluated in the as-bonded condition, and the other half is subjected to heat treatment in an 1850 F salt bath for 5 min prior to evaluation.

Additional compartments have been burst tested in the 0.150 by 4.3 by 15.0-in. specimens which were made using graphite-sprayed cores which were buffed to a high luster prior to assembly for bonding in order to remove loosely adherent graphite. Twenty compartments have been tested in as-bonded and in beta-heat-treated specimens prepared in this manner. No leakage between compartments has been observed during any of the tests. Metallographic examination of these specimens showed that the amount of bond-line contamination was greatly reduced as compared with earlier specimens containing unbuffed graphite-sprayed cores. This contamination appears to be a thin layer of zirconium carbide as indicated by electron-diffraction and electron-microscope observations. In the buffed-graphite specimens, only slight contamination was visible in a few areas, and there was observed to be grain growth across almost the entire original bond interface. Thus, the specimens containing cores with buffed coatings appeared significantly better in metallographic examinations and burst tests than the specimens with unbuffed coatings, which exhibited considerable bond-line contamination and some weak bonds. The weak bonds failed by intercompartmental leakage during burst testing. Also, the buffing operation apparently did not decrease the effectiveness of the graphite coating as a barrier to reaction, since almost no reaction between the cores and cladding was observed in these specimens.

N. DEVELOPMENTS FOR THE MGCR

W. H. Goldthwaite

In research assistance to the Maritime Gas-Cooled Reactor Program the postirradiation examination of two stainless steel-clad UO_2 specimens has been concluded. A program which includes the development of fabrication techniques and the irradiation of three potential fuel materials is in progress. The effect of radiation on the transport of carbon by impurities in helium coolant gas is also being investigated.

THE POSTIRRADIATION EXAMINATION OF GA-BNL SINTERED UO_2 FUEL SPECIMENS CLAD WITH STAINLESS STEEL

G. E. Lamale, J. E. Gates, and R. F. Dickerson

The postirradiation examination of UO_2 samples irradiated at Brookhaven National Laboratory has been completed. One of the samples consisted of two pellets of UO_2 sintered to a density of 93 per cent of theoretical and clad with 10 mils of Type 316 stainless steel. This sample was irradiated in a CO_2 atmosphere to an integrated exposure of 5.2×10^{18} nvt at a cladding-surface temperature of 1600 F. The cladding on this sample burst. The second sample consisted of two pellets of UO_2 sintered to a density of 93 per cent of theoretical and clad with 5 mils of Type 316 stainless steel. This sample was irradiated in a helium atmosphere to an exposure of 4.7×10^{19} nvt at a cladding-surface temperature of 1200 F. The cladding of this specimen did not fail.

The failed sample contained two intact pellets. One of these pellets was cut into two sections. The section furthest from the outer end of the pellet was used for burnup analysis. The radiochemical analysis showed a uranium burnup of 0.009 a/o.

The sample whose cladding did not fail showed random cracking in the UO_2 pellets when transverse sections of the capsule were made. A burnup wafer was taken near the middle of this particular capsule. Radiochemical analysis showed a uranium burnup of 0.05 a/o.

No further work is planned.

FABRICATION AND IRRADIATION OF FUEL MATERIALS

Potential fuel materials for the MGCR are being investigated. Procedures for the fabrication of BeO-UO_2, graphite-UC, and graphite-UC_2 fuel specimens clad with Type 316 stainless are being developed. The elements will contain approximately 20 volume per cent of 30 per cent enriched fuel. Specimens 1/4 in. in diameter are to be irradiated to burnups of 3, 12, and 20 per cent of the contained uranium-235 at a heat flux of about 200,000 Btu/(hr)(ft^2) and a surface temperature of 1500 F.

Fabrication of BeO-UO_2 Fuel Elements

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

Techniques for fabrication of ceramic fuel elements consisting of 20 volume per cent of 150-μ UO_2 particles dispersed in a densely sintered BeO matrix are being developed.

During February, BeO-UO_2 pellets, containing fully enriched UO_2, were prepared by the procedure described in BMI-1315. Measurements indicated that the bulk densities of eight pellets, 0.222 in. in diameter by 0.25-in. long, ranged from 89.2 to 90.9 per cent of the calculated theoretical density of the mixture, and that their average bulk density was 90.5 per cent of theoretical. Microscopic examination of a mounted and polished section indicated that the UO_2 grains were fairly uniformly dispersed in the BeO matrix, and that they ranged in size from about 100 to 200 μ. Very little dusting of the UO_2 grains occurred during mixing and handling. The BeO matrix consisted of equiaxial grains whose average diameter was about 16 μ.

Current effort is directed toward increasing the sintered bulk density of the composite bodies by modifications in both the processing method and the type of UO_2 grain used in the mixtures.

Fabrication of Graphite Fuel Elements

W. A. Hedden, A. B. Tripler, Jr., W. C. Riley,
and W. H. Duckworth

Techniques are being studied for fabricating graphite-matrix bodies containing 20 volume per cent of fuel compound in the form of UC or UC_2.

In work reported previously, graphite bodies, containing either UC or UC_2, were fabricated by mixing the fuel compound with graphite filler and standard pitch binder, forming, and baking at 2000 F in argon. No measurable oxidation of either carbide as a result of processing operations was detected.

During this report period, test specimens were fabricated with enriched fuel for irradiation testing. The test specimens were formed from two different mixes, one containing UC and the other containing UC_2. After fabrication of these bodies, it was found that, on a bulk-volume basis, the fuel content was about 15 volume per cent rather than the 20 volume per cent desired. This resulted from an unexpectedly low density in the test specimens. Additional specimens are being fabricated from bodies having adjusted fuel contents calculated to yield the desired 20 volume per cent.

Metallographic examinations were completed on specimens made from bodies containing UC or UC_2 baked at temperatures of about 2350 and 2700 F in a protective atmosphere of argon gas. Examination indicated that samples containing UC_2 were essentially unchanged by these heat treatments. However, for samples originally containing UC, the 2700 F heat treatment converted almost all of the UC to UC_2 and the 2350 F heat treatment converted trace amounts of UC to U_2C_3. No formation of UC_2 was noted at the latter temperature.

Radiation-Effects Study of Potential Fuels

S. Alfant, J. E. Gates, and R. F. Dickerson

Pellets of BeO fueled with UO_2, graphite fueled with UC_2, and graphite fueled with UC will be irradiated under conditions simulating those occurring in the MGCR. The pellets will be loaded into stainless steel tubes, encapsulated, and irradiated to burnups of 3, 12, and 20 per cent of the uranium-235 at a surface temperature of 1500 F.

It was not possible to prepare the first group of specimens to be irradiated in a single capsule to 3 per cent burnup in time for irradiation in the MTR. It is therefore planned to irradiate the first capsule in the BRR starting on March 23, 1959. This should allow a significant burnup of the fuel to be achieved and still permit examination of the irradiated fuel pellets by June 30, 1959.

Production of the pellets and capsules necessary for the remaining irradiations will be initiated in the near future.

CORE-MATERIALS IRRADIATION PROGRAM

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

Work is continuing on the preparation of four quartz capsules for a BRR irradiation of Type 430 stainless, Type 310 stainless, Inconel, and unclad graphite together in high-temperature commercial helium. The purpose of this equipment is to determine the effect of helium impurities on metal corrosion and carbon transport in this metal-graphite system. As reported last month, a 700 F temperature gradient in the torus-shaped capsule will promote convective flow of the helium over the specimens and around the torus.

Capsule design has been completed and construction is under way.

In support of the postirradiation evaluation, a brief bench study of degassed graphite and the reaction of this graphite with commercial helium at 1500 F is being conducted. Results of this study will be reported next month.

O. ENGINEERING ASSISTANCE TO KAISER ENGINEERS

Reactor Flow Studies

L. J. Flanigan and H. R. Hazard

Flow studies using air 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. Previously reported work includes starting construction of the plastic model parts, the model core, and the parts for the test setup.

In February, construction was continued on all three phases. Fabrication of the plastic model parts by a vendor is about 40 per cent complete. At Battelle, all parts for the model core have been completed, and work on parts for the test setup is about 30 per cent complete.

In March, assembly of the model core will be completed and work will continue on the plastic model parts and the parts for the test setup.

P. DEVELOPMENTAL STUDIES FOR THE SM-2

S. J. Paprocki

The work discussed in this portion of the report is being performed to assist Alco Products in the development of the SM-2 (formerly APPR-1B). It is concerned with the development of material specifications for the fuel, absorber, and supressor components.

The proposed reactor core life of the SM-2 is approximately double the life of the SM-1 (formerly APPR-1). From a core-material standpoint, this condition can be achieved by an increase in the amount of burnable poison and by considerable improvement in the over-all performance of the fuel.

Encapsulation Studies

W. E. Murr, S. K. Hopkins, D. B. Hamilton,
and J. H. Stang

During February the design of the capsules (designated as BMI 30 series) containing specimens of clad fuels under development for the SM-2 (formerly APPR-1B) program was fixed. As a result of recent negotiations with NRTS personnel, the capsules will be irradiated at the ETR in an unperturbed thermal-neutron flux of 3×10^{14} nv. Approximately ten capsules will be fabricated and irradiated; the total burnup anticipated for five of these is 77 w/o and is about 37 w/o for the remainder.

As discussed in BMI-1307, a leading design problem in the encapsulation is that of providing enough thermal conductance from the specimens to the water-contacting capsule shell (1-1/8-in. OD) to dissipate the fission heat and yet maintain the specimen surface temperature in the neighborhood of 650 F. In the design now established, the specimens will be immersed in NaK but in a substantial fraction of the remaining inner capsule space a system of copper (OFHC) blocks will replace the NaK to take advantage of the relatively high thermal conductivity of copper. Auxiliary heaters (six 2-kw units per capsule) will be wound around these copper pieces and the entire assembly will be contained in a nickel shell. With such a conductance path, the specimen temperatures in the flux field of 3×10^{14} nv should not exceed 700 F at the start of irradiation. As burnup progresses and fission heat is reduced the reference specimen temperature of 650 F will be maintained by auxiliary heat. Based on the best information available in the literature and Battelle's experience, there will be no corrosion problems in the capsule.

According to the current schedule, fabrication of components for seven capsules will be completed in mid-March. The capsule-loading phase will occupy the remainder of March and part of April.

P-2

Development of Fuel Materials

S. J. Paprocki, D. L. Keller, G. W. Cunningham, J. B. Fox,
A. K. Foulds, D. E. Lozier, and W. M. Pardue

Techniques are being developed for producing stainless steel-UO_2 dispersion fuel elements which contain a boron-bearing burnable poison. The reference fuel element contains UO_2 prepared from UO_3 hydrate, and boron as discrete particles of ZrB_2. As alternates, fuel elements containing other borides (NbB_2, MoB) or having a boron-alloy core matrix are being considered.

Compatibility studies have shown ZrB_2 to be the most promising poison material, and it has been chosen as the reference boron-containing compound. The secondary, or backup, compound will probably be NbB_2.

Zirconium diboride particles have been shown to be completely reaction-free even when sintered in hydrogen, provided a step-sintering process is used. This consists of holding 16 hr at 1600 F and increasing the temperature in increments of 150 F per hr to 2200 F, then holding for 2 hr. Some of the ZrB_2 particles show formation of an oxidized layer inside the original particle, but no signs of melting or reaction. This compound has a strong affinity for oxygen, and, in addition, X-ray diffraction studies show that for the powder being used some ZrO_2 is present in the as-received condition. In specimens sintered at 2400 F, reaction has been detected, but little or no interaction of ZrB_2 with stainless occurs at 2300 F.

Specimens containing 1 w/o NbB_2 which were hot rolled at 2200 F show no apparent reaction between matrix and boride particle but exhibit small unidentified particles randomly distributed in the matrix. Work is now being done to determine if these small particles are fragments of the original NbB_2.

Investigation of the weight loss of various boron compounds during sintering is being studied. The following tabulation shows per cent weight loss of several compounds sintered as loose powders in hydrogen and *in vacuo*.

Compound	Weight Loss, per cent 2270 F in Hydrogen	 2400 F in Vacuum
NbB_2	3.0	0.12
CrB_2	3.72	3.1
CaB_6	11.05	6.6
MoB	12.27	6.6
YB_6	17.15	2.2
ZrB_2	2.83	0.118

This loss is believed to be due to vaporization of B_2O_3. The higher loss in hydrogen is probably due to reaction of the boride with oxygen in the hydrogen atmosphere. Exit dew points of -35 F were measured during these tests. Additional tests are being run to determine whether a pretreatment of the powders in hydrogen or vacuum will prevent loss during sintering or whether additional loss occurs each time the borides are subjected to a high temperature in hydrogen or vacuum.

Analytical chemistry, spectrography, and X-ray diffraction have been used to determine whether the compounds are stoichiometric. With the exception of a small amount of ZrO_2, the X-ray diffraction patterns indicated that the ZrB_2 used in this program is a single-phase boride. Values of 18. 2 w/o boron and 0. 78 w/o carbon were measured by chemical analysis. Spectrographic results indicated that no other major impurities were present. The original NbB_2 used in this work consisted of approximately 50 w/o NbB_2, 40 w/o Nb_3B_4, and less than 10 w/o of NbB plus NbB_2 plus niobium. The only impurities are approximately 0. 25 w/o carbon, 0. 25 w/o oxygen, and 0. 5 w/o iron. The NbB_2 now being used matches the ASTM standard. An X-ray diffraction pattern shows 0. 05 w/o carbon and 0. 1 w/o iron.

Compacts have been prepared using the SM-2 (formerly APPR-1B) reference treatment containing the proper loading of ZrB_2 and NbB_2 and using 28 w/o spherical UO_2 as a fuel. These compacts are now being roll clad. In addition, two packs using no fuel and containing the specified SM-2 content of B_4C are being fabricated for comparison. This compound was sintered in both prealloyed and elemental matrices and step sintered in hydrogen. The prealloyed specimen reacted grossly, but the elemental appeared sound and is in the fabrication process.

Also, a series of compacts with elemental matrices are being fabricated with ZrB_2 and NbB_2. These elemental mixes were prepared with a nickel-chromium alloy to determine the degree of reaction with nickel already in solution. Investigations of elemental matrices indicate that severe reactions between boron and nickel occur readily if the nickel is not in solution before a temperature of approximately 1900 F is reached or if nickel sintering trays are used.

In the fabrication of these fuel elements, it may be necessary to alter the standard fabrication procedure so that there will be no reaction of the boride with the stainless nor will there by any loss of boron during the process. However, the procedure cannot be altered so that excessive stringering and fracturing of UO_2 particles occur.

Variations of the sintering procedure listed above for specimens containing ZrB_2 are being evaluated for producing core compacts. The effects of compacting pressure, time at 1600 F, heating rate, and exit dew points are under study. Packs are being hot rolled at temperatures ranging from 2000 to 2200 F. The change in temperature has very little effect on the structure of cores containing spherical UO_2, but the higher temperatures result in less stringering and fracturing of the hydrothermal UO_2.

Techniques are presently being developed for fabricating the various types of irradiation specimens outlined for the SM-2 (formerlly APPR-1B) core program. In addition to the reference core of ZrB_2 particles dispersed in prealloyed Type 347 stainless steel with 93. 15 per cent enriched hydrate UO_2, specimens will also be prepared to determine the desirability of using UN, spherical UO_2, an alternate boride, and alternate methods of incorporating the boron, such as boron master-alloy powders.

P-4

Calculations have been made to determine the weight, shape, and density of the sintered prerolled core necessary to produce a core 1 by 0.50 by 0.030 in. after a 7.5-to-1 reduction in thickness. These calculations are being verified by preparing dummy specimens containing natural UO_2. Fabrication of the enriched irradiation specimens will be started during the first week in March.

RWD:CRT/all