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PROGRESS RELATING TO CIVILIAN APPLICATIONS DURING JUNE, 1959

by

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A. ASSISTANCE TO HAPO

F. R. Shober

The thermal conductivities of irradiated uranium and UO_2 have been measured at room and elevated temperatures. In the study of the creep properties of Zircaloy-2, both annealed and 15 per cent cold-worked materials are being investigated at elevated temperatures. The study also includes the effect of cyclic test temperatures as well as constant test temperature. High stresses under cyclic temperature conditions, room temperature to 345 C to room temperature, produce greater deformation than the same high stresses under constant temperature, 345 C.

The density distributions of a minus 40 plus 60-mesh and a minus 270-mesh sample of irradiated graphite were measured. Irradiation appears to have caused a reduction in the amount of both high- and low-density material. The amount of intermediate-density material increased. Work was completed in the program to simulate conditions for a loss-of-coolant incident in the PRTR. The two final cases considered were a postulated rupture in a top jumper tube and a bottom jumper tube, respectively.

The corrosive action of high-temperature water on defected Zircaloy-2-clad uranium specimens is being investigated. Studies have been made at 200, 225, 250, 300, and 345 C. In the development of a prototype detection system to indicate defected fuel elements in a pressurized-water reactor, work is being directed toward the determination of the rate of dissolution of silver chloride at 180 F, determination of the optimum form of exchange material, and determination of the optimum exchange-column design. In the development of a thermal-neutron-flux monitoring system, work is in progress to develop stable ceramic elements for the flux sensor.

An additional study has been made of UO_2. Samples of 1-μ UO_2 were passed through a plasma-jet flame in an attempt to spheroidize the powder particles. The products of some experiments were composed largely of isotropic spheres.

Thermal Conductivity of Uranium and UO_2

H. W. Deem and C. F. Lucks

Thermal-conductivity measurements have been made on unirradiated and irradiated uranium and UO_2 to determine the effect of irradiation on thermal conductivity. While the data have not been reduced for inclusion in this progress report, a topical report is being prepared.

Provisional thermal-conductivity data for Zircaloy-2 given in BMI-1152 are superseded by those given in BMI-1273.

Mechanical Properties of Zirconium Alloys

F. R. Shober and J. A. VanEcho

The study of the creep properties of annealed and of 15 per cent cold-worked Zircaloy-2 at 290, 345, and 400 C is being continued. This investigation includes long-term creep tests (10,000 to 15,000 hr) on cold-worked material, and stress-rupture and long-term creep tests on annealed material, and a series of cyclic-temperature tests on both annealed and cold-worked Zircaloy-2.

A comparison of data from cold-worked Zircaloy-2 in cyclic-temperature tests and in constant-temperature tests and at comparable stresses has shown that only a slight increase in total creep deformation can be expected as a result of cycling between 290 C and room temperature. However, tests involving cycling from 345 C to room temperature produced data indicating that the difference in total creep deformation between the cyclic- and constant-temperature tests is a function of the stress. High stresses under cyclic-temperature conditions produce greater deformation than the same high stresses under constant-temperature conditions.

Annealed Zircaloy-2 at 290 and 345 C is characterized by a large total deformation upon loading, 3 to 4 per cent, followed by a rapidly decreasing creep rate, decreasing to less than 0.00001 per cent per hr after 1000 hr. A mechanism greatly increasing the creep resistance appears to be operative at these temperatures. This type of creep curve has not been observed for tests at 400 C.

Additional tests will be started on annealed Zircaloy-2.

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.

The density distributions of a minus 40 plus 60-mesh and a minus 270-mesh sample of irradiated TSGBF graphite were measured. The results were compared with the density distributions of unirradiated TSGBF samples of the same particle-size ranges. Analysis of results indicated that irradiation caused a decrease in the amount of both high- and low-density material. The amount of intermediate-density material, between 2.05 and 2.18 g per cm^3, increased.

This research will be recessed pending the completion of irradiation of samples of known pre-exposure sink-float density distribution.

A-3

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

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

The program to simulate conditions after a postulated loss-of-coolant incident within the PRTR was completed this month. The final two cases have been computed on the IBM-653 digital computer, and the results of the machine computation were analyzed.

During the month Case III, that of a postulated rupture in a top jumper tube connecting a process tube to the upper ring header, was considered first. The machine computation indicated that critical flow of steam through the rupture might limit the rate at which the emergency injection coolant could flow into the reactor after injection had been initiated unless heat could be absorbed at some point within the reactor. Fortunately, a heat balance on the water which would be injected into the top ring header indicated that sufficient heat capacity would exist to condense all the steam generated in the core. Hence, it would appear that the reactor could be filled with injection coolant in this case.

After the core would be refilled, it is evident that little circulation of the injection coolant through the process tubes could occur due to the fact that injection of coolant would be made into both ring headers. Although the process tubes initially would be filled with water, the possibility would still exist for the tubes to become steam filled by steam generation within a tube. However, a tube that filled with steam would refill with water as the steam inside it condensed by coming in contact with the cooler water which would enter the process tube from the ring headers. This process would be cyclical and with a period sufficiently short to prevent significant heating of the fuel elements.

Case IV concerns a postulated bottom-jumper rupture. The situation within the core would be slightly more critical in this case because of a slower pressure decay during blowdown. This, combined with other factors, made this case less amenable to machine computation than Case III. The slower pressure decay would result in emergency coolant being injected at a longer time after the incident. Once the injection started it would have to fill the inlet pipe and upper ring header and then spill down the top jumpers onto the fuel element before cooling could be effected. Thus, before the coolant reached the fuel elements there would be a considerable period during which the core would be steam bound and the temperature of the fuel would rise above 1000 F.

As the emergency coolant would begin to overflow and come in contact with the hot fuel elements, it would form a considerable volume of steam. The pressure would quickly rise to a value of 100 psi. At this pressure, injection would stop until the steam would be condensed by coming in contact with the cooler water in the two ring headers. As the steam would condense, the pressure would drop and the injection coolant could again spill down onto the fuel and repeat the cycle. Fortunately, the machine computation indicates that heat would be removed by this process faster than it would be generated, and cooling would occur.

All of the cases considered in this study, believed to be representative of the postulated potentially dangerous ruptures, proved to be capable of control by the planned emergency coolant system.

This concludes the program and a topical report is being prepared. It contains the computations of the various cases, the analysis of the computations, recommendations, and the computer program which was used in the investigation.

A Photographic Study of the Corrosion of Defected Fuel Elements in High-Temperature Water

E. F. Stephan and F. W. Fink

Lapsed-time motion pictures are being made through a windowed autoclave of the corrosive action of high-temperature water on defected Zircaloy-2-clad uranium specimens. Seven defected specimens have been studied. Each one consists of a natural-uranium core extrusion clad with Zircaloy-2. The defect is formed by a 25-mil-diameter hole drilled through the 10-, 20-, or 30-mil cladding. Studies were made at 200, 225, 250, 300, and 345 C. Records of the increase in pressure were kept for use in the rough computations of the corrosion rates.

Table A-1 summarizes some of the data. The first evidence of attack is a swelling of the cladding around the defect. This swelling continues to enlarge and eventually the cladding ruptures. Except for the 10-mil clad specimen, which ruptured through the defect, the first rupture of the cladding occurs about 1/8 in. from the defect. No gassing or corrosion product is observed until this initial rupture occurs. As the reaction proceeds with time, the cladding tends to rupture at other sites. The UO_2 corrosion product begins to darken the water shortly after the initial rupture.

TABLE A-1. CORROSION IN HIGH-TEMPERATURE WATER OF DEFECTED[(a)] NATURAL URANIUM FUEL CORES EXTRUSION CLAD WITH ZIRCALOY-2

Water Temperature, C	Time to First Noticeable Swelling at Defect, hr	Time to First Gas Evolution, hr	Time to Complete Reaction, hr	Uranium Dissolved at Complete Reaction, g
200	8	15	24	136.95
225	6	7.5	13	135.20
250	1.7	2.5	10	132.28
300	1.0	1.5	5	119.63
300(b)	0.75	1.25	6.25	134.45
300(c)	0.75	1.1	6.1	135.91
345	0.4	0.8	3.17	134.55

(a) The defect consisted of a 25-mil-diameter hole drilled through the nominal 30-mil-thick cladding.
(b) Cladding nominally 20 mils.
(c) Cladding nominally 10 mils.

An element containing a uranium-2 w/o zirconium core and two tubular-type fuel elements are to be studied next.

A-5

Development of a Fuel-Element Leak Detector

J. E. Howes, Jr., T. S. Elleman, and M. Pobereskin

This report summarizes the progress to date on the development of an isotopic-exchange leak-detection system.

Work has been initiated on the following experiments:

(1) Determination of the rate of dissolution of silver chloride at 180 F

(2) Determination of the optimum form of the exchange material

(3) Determination of the optimum exchange-column design.

A closed loop is being built to determine the rate of dissolution of AgCl in a dynamic system. Water at 180 F will be cycled through a column of AgCl tagged with silver-110. The silver-110 ions from the dissolved $Ag^{110}Cl$ will be separated from the water by a cation-exchange column in the system. Radiation monitoring of the ion-exchange column will indicate the relative rate of dissolution of AgCl. The absolute rate of dissolution will be determined by eluting the silver-110 from the ion-exchange column and radioassaying the effluent.

After completion of the dissolution experiment, the loop will be used to evaluate various parameters affecting the isotopic-exchange reaction. These parameters include column size, particle size, flow rate, and column design. The experiments are designed to determine the conditions which will give optimum halide exchange together with high fission-product decontamination.

A second loop is being built to evaluate, simultaneously, the physical properties of various forms of exchange material. Water at 180 F will be circulated through columns of the various forms of exchange media. The columns will be examined visually to note any change in physical properties. Changes in particle size due to erosion or particle breakup will be determined periodically by measuring the pressure drop across the column.

Some of the exchange materials which will be considered are fused silver chloride, silver chloride on silver shot, and silver chloride on porous alumina.

The results of the experiments which are planned should give the information necessary to develop a prototype leak-detection system.

Thermal-Neutron-Flux Monitoring System

C. V. Weaver, J. W. Lennon, and M. J. Snyder

The objective of this program is to develop a thermal-neutron-flux monitoring system for the Hanford reactors. The basic unit for the proposed system is a flux sensor

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previously developed at Battelle. It is based on the principle of balancing the temperatures of a fission-heated uranium-fueled sensing element and a nearly identical electrically heated balancing element.

Work is in progress to develop stable ceramic elements with reproducible characteristics for the flux sensor. Compositions in the system $MoSi_2$-Al_2O_3-UO_2 are being investigated first since these have proven satisfactory in past developments. The first phase of the study concerns the effects of variations in the materials and processing conditions on electrical resistivity. Several of the compositions studied so far have had satisfactory physical properties. Bench tests are now in progress to determine their stability as a function of time and temperature. (A 5000-hr minimum life for the flux side is specified.)

During the coming month, techniques for fabricating elements of the required size and shape as well as with the required physical properties will be investigated.

Developments in the mechanical design of the flux sensor have resulted in a brazing technique for making an integral low-resistance junction between the ceramic elements and the electrical leads. The stability of the connection is being tested with the sample ceramic elements described above.

Spheroidizing Uranium Oxide Powder Particles

T. E. Cook, G. H. Kesler, and J. M. Blocher, Jr.

Samples of 1-μ UO_2 powder were passed through a plasma-jet flame in an attempt to spheroidize the powder particles. The powders were introduced as gas-borne suspensions into helium and argon plasmas. Powders were fed either to an external nozzle or into the working gas of the plasma apparatus.

Photomicrographs of the sectioned powder particles taken with polarized light showed that the product of some experiments was largely composed of isotropic spheres. No evidence of these spheres was found in the original UO_2 powder.

While this concludes the study, additional work would be necessary to relate the extent of spheroidization to the process variables.

B. DEVELOPMENTS FOR ALUMINUM-CLAD FUEL ELEMENTS

R. J. Carlson and N. E. Daniel

The program concerned with the preparation of aluminum-35 w/o uranium billets in the form of hollow cylinders and with improving the casting and fabrication characteristics of the alloy by the addition of a third element has been concluded. Problems which have arisen in fabrication of aluminum-clad uranium fuel elements have indicated that determination of the location of the eutectic in the aluminum-uranium-nickel system is necessary and a program for the location of this eutectic has been initiated.

Aluminum-Uranium-Nickel Eutectic Alloys

V. W. Storhok, A. A. Bauer, and R. F. Dickerson

A project is being conducted to determine the temperature and approximate composition of the ternary eutectic in the aluminum-uranium-nickel system. This study was prompted by a desire to determine the amount by which the binary high aluminum-uranium and high aluminum-nickel eutectic temperatures are lowered in ternary combinations.

A number of alloys have been prepared by induction-melting techniques:

Alloy	Composition, w/o
1	Aluminum-13.3 uranium
2	Aluminum-5.9 nickel
3	Aluminum-21 uranium-3.1 nickel
4	Aluminum-18.9 uranium-1.4 nickel
5	Aluminum-15 uranium-3 nickel
6	Aluminum-13.1 uranium-1.9 nickel
7	Aluminum-8 uranium-2.4 nickel
8	Aluminum-8 uranium-4 nickel
9	Aluminum-7.9 uranium-5.5 nickel

While it is unlikely that any of the above alloys are at the ternary eutectic composition, it is felt that at least an indication of the correct composition can be gained through these alloys.

From results of initial metallographic examination, it appears that Alloys 3, 4, and 5 contain an excess of uranium, as rather large amounts of compound (probably peritectic UAl_4) are present. The other ternary alloys exhibit varying amounts of free aluminum and appear to be low in either uranium or nickel. On the basis of these results additional alloys containing increased amounts of nickel, are being prepared.

Differential thermal analysis of all the alloys is being conducted. To date, confirmations of the binary eutectic temperatures at 640 C in both the aluminum-uranium and aluminum-nickel systems are the only data obtained.

C. RADIOISOTOPE AND RADIATION APPLICATIONS

P. Schall

Progress made on programs on the use of tracers in cement-quality control, use of intrinsic tracers for process control, radiation chemistry of inclusion compounds, radiation-induced graft polymerization, and radiation-induced nitration reactions is discussed in this section.

Several experiments on the effect of other elements on the radiometric analysis for magnesium in cement were conducted. These experiments conclude the program on the development of the technique. Work is continuing on activation analysis and radiometric methods for calcium and sulfate analysis.

Laboratory-scale experiments on the use of an intrinsic tracer to control the copper-stripping operation in the ammonia-leach nickel-refining process is continuing. Discussion with representatives of a company utilizing this process has indicated industrial interest in development of the tracer control technique.

Analysis of the chemical products formed in the radiation-induced reactions in urea-dodecane and dicyanoammine-nickel (II) complex of benzene is completed. Promising reactions for commercial radiation processing have been found.

The study of the effect of structure of polymethacrylates on the ability to form graft-polymerization sites is continuing. Preparation of a series of polyesters to be used in further study of structural effects has been initiated.

Construction of apparatus for the nitration studies is completed and development of analytical methods is continuing.

Development of Radioactive-Tracer Quality-Control Systems

C. T. Brown, D. N. Sunderman, and M. Pobereskin

The investigation of the radiometric method for the analysis of magnesium in cement has been concluded. A study of the effect of 4-mg quantities of calcium or manganese per 10 mg of magnesium on the accuracy of the method has shown that calcium yields results 8 per cent high and manganese yields results 3 per cent high. These results confirm the previous results, i.e., chemical separation of calcium must be performed before magnesium can be determined in cement.

The standard procedure as developed in this laboratory is as follows:

To a 10-ml aliquot of the magnesium sample containing 5 to 15 mg of MgO, add about a 25 per cent by weight excess of $(NH_4)_2HP^*O_4$. Dilute to 100 ml and add

3 drops methyl red indicator. The initial radioassay of this solution when drawn into the 10-ml counting chamber should be about 8000 cpm. After the initial radioassay, $MgNH_4P^*O_4$ is precipitated at <4 C with stirring by adding concentrated NH_4OH to the solution drop by drop to the methyl red color change, and then adding 10 ml excess NH_4OH. The solution is then radioassayed as before. For best results the radioactivity present in the solution after precipitation should be 50 to 75 per cent less than the initial radioactivity. The decrease in activity before and after precipitation is a direct measure of the amount of magnesium precipitated as $MgNH_4P^*O_4$.

Neutron-activation analyses of cement raw materials such as limestone, clay, slag, and fly ash are in progress. A series of six irradiations of 16 analyzed samples will be performed. The calibration studies of the analytical procedure have shown that samples must be assayed at comparable distances from the scintillation crystal, and at instrument dead times below 30 per cent for accurate quantitative results. The effect of the following fast-neutron reactions must also be taken into consideration: $Al^{27}(n,\alpha)Na^{24}$, $Al^{27}(n,p)Mg^{27}$, $Si^{28}(n,p)Al^{28}$, $Si^{30}(n,\alpha)Mg^{27}$, $Mg^{24}(n,p)Na^{24}$, and $Fe^{56}(n,p)Mn^{56}$. Fast-flux dosimeters are being used to determine the magnitude of the fast-neutron flux.

During the next month the activation analyses already in progress will be continued. Work on the calcium analysis using a radiometric end point will be concluded. Work will be initiated on the sulfate analysis using barium-140 tracer. Preliminary work on this method will be concerned with a method for continuous separation of the barium-140 and lanthanum-140 activities.

Use of Intrinsic Radioactive Tracers for Process Control

J. L. McFarling, H. B. Brugger, D. N. Sunderman, and M. Pobereskin

During this report period, laboratory study of the copper-stripping operation of the ammonia-leach nickel-refining process has continued. The use of a NaI(TI) crystal in the measurement of the low gamma-activity levels to be expected in process-control applications is currently being investigated.

During the past month, the Sherritt-Gordon nickel refinery at Ft. Saskatchewan, Alberta, was visited Discussions were held with the management, operating, and research staffs at the plant. They expressed a great deal of interest in the radioisotope-application program and expressed interest in cooperating in a plant-demonstration experiment.

Sherritt-Gordon personnel agreed that control of the copper-stripping operation would probably be a good radiotracer control application. In addition, they suggested using an intrinsic radiotracer to control removal of iron from the mixed nickel-cobalt stream. Technically similar to the copper-strip application, radioisotope control of iron removal should have wide application in nonferrous-metals refining.

The decision was made to evaluate experimentally both the copper-removal step and the iron-removal step of the process to determine the advantages and disadvantages of each for a radiotracer control demonstration. This laboratory evaluation will proceed immediately with Sherritt-Gordon being kept informed of the progress of the work. At some convenient later date, arrangements for a plant demonstration are expected to be made with the Sherritt-Gordon refinery.

Radiation Chemistry of Inclusion Compounds

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

During June work continued on the analyses of the urea-dodecane complex and the dicyanoammine-nickel (II) complex of benzene. Evaluation on the latter has been completed but major unknowns in the study of the urea-dodecane complex still remain. Final summaries of the work on both complexes are presented below.

Urea-Dodecane Complex

A satisfactory scheme of separation and analysis has not been developed for the urea-dodecane complex, and the major product in the irradiated complex has not been positively identified. This product is the result of a host-guest reaction and may contain several different compounds. The product amounts to about 16 w/o of the hydrocarbon phase separated from the irradiated complex. Molecular-weight determinations, elemental analyses, spot tests, infrared examinations, and gas chromatography analyses suggest that the product contains substituted ureas, amines, and possibly amides. Large amounts of amide believed to be the major product of the host-guest reaction and reported in BMI-1324 report were not verified.

There is no evidence that the host-guest reaction is specific and the polar product of this reaction may be a spectrum of compounds containing carbon, hydrogen, nitrogen, and oxygen. Failure to positively identify the polar product does not alter previous conclusions regarding the low radiation yield. G-values for the formation of the polar material certainly do not exceed 5.

Dicyanoammine-Nickel (II) Complex of Benzene

About 130 g of the nickel cyanide complex of benzene was irradiated to 2 x 10^8 rads. rads. Infrared analyses of the benzene extracted after solution of the irradiated complex in concentrated ammonia showed the presence of only a trace of benzonitrile and no aniline. This trace amounts to less than 0.02 w/o of the benzene in the complex. The G-value for benzonitrile formation is less than 0.01.

Future Program

Continuation of the program along the present lines is not recommended because no promising results of direct interest to the field of radiation processing have been obtained.

Graft-Polymerization Studies

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

This project is concerned with studies of radiation-induced changes in polymers leading to graft copolymerization. Work has continued in two areas. Several methacrylate esters have been polymerized by radiation for later use in the study of structural factors. In addition, the method of analysis for free-radical sites and the state of the ungrafted polymers have received detailed attention.

It has been found that the unirradiated polymers contain measurable concentrations of peroxy and/or free-radical sites. The number of the free-radical sites can be reduced by prior heating of the samples under vacuum. These sites, however, lead to apparent grafting of the polymers when vinyl pyrrolidone is added under the usual conditions. The extent of this grafting is being studied in order to determine how it will affect the interpretation of the previous work and the course of the future studies.

Concurrent with this work, the syntheses of a number of polyesters have begun. These polyesters will have controlled structural variations to allow study of the effects of factors such as unsaturation, structure of main chains, and aromaticity.

Nitration of Hydrocarbons

M. J. Oestmann, R. E. Fulmer, J. F. Kircher, and G. A. Lutz

During the month, most of the irradiation apparatus was completed. Preliminary testing of the equipment is under way.

Work on the development of analytical methods is continuing. Gas chromatography is being explored in this phase of the program.

Next month, construction and testing of the irradiation apparatus will be completed. Additional thermal runs will also be programmed in July.

D. PROCESSING OF FEED MATERIALS

E. L. Foster

Work on a program for National Lead Company of Ohio aimed at finding a convenient method of describing the solidification of uranium castings and predicting the effects of variations in the casting procedure has been concluded. Through theoretical study and experimental observation, metal-to-mold heat transfer was related to the solidification process, and a mathematical model to predict heat flow and metal solidification under various casting procedures was developed. Correlations of the results predicted by the mathematical model with data obtained during casting of production-size ingots have shown good agreement.

Solidification of Uranium

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

This program has dealt with a study of the solidification of uranium castings in cylindrical graphite molds. The program embraced two phases of study: a theoretical study of the transfer of heat from the metal and mold and its relationship to the solidification processes of the casting; and an experimental phase to substantiate the validity of assumptions used in the theoretical phase, and to provide data to establish the degree of correlation that exists between the theoretical results and experimental observations.

A mathematical model, consisting of 75 cells and representing a pie-shaped section of the mold and ingot, was developed and used to predict the heat flow as solidification occurs. Variables of temperature, time, mass, and physical properties were incorporated into the model and heat-flow rates were calculated. In experimental work, 100- and 1200-lb ingots were cast in molds instrumented to record the time-temperature relationships in the mold and metal from time of pour through the freezing period. Results of temperature measurements obtained were used to develop cooling curves and temperature gradients existing throughout the system. Other ingots of the same sizes were cast to study the physical events taking place at the mold-metal interface. These were investigated by using a network of electrical probes positioned at the inside wall of the mold. The sequence of gap formation was detected at various times along the mold-metal surfaces with operation-type recorders.

Recent activities have been concerned with obtaining correlation of the calculated results with the thermocouple data obtained from 1200-lb experimental castings. The calculated results have been found to be in good agreement with the experiments, and on this basis the mathematical model is believed to be satisfactory for predicting the effects of changes in casting parameters. Computer runs have been completed using variations in the superheat, mold preheat, time of pour, and the emissivity of the outer mold wall. The significance of these alterations in changing solidification patterns is being evaluated. This concludes the current study.

E. SPRAY DEPOSITION OF CALCIUM METAL ON NICKEL OR INCONEL

A. F. Haskins and R. M. Evans

This is the sixth monthly report on a feasibility study to determine if calcium coatings can be successfully put on nickel by arc-spraying methods.

Spectrographic analysis of the white and dark-gray vapors encountered during spraying operations disclosed that both were calcium materials which were very similar. With calcium as the major element, impurities included those elements found in Domal Grade 2 special-purity calcium. These vapors are thought to be caused by a combination of unsatisfactory atmosphere and moisture on the calcium wire itself.

Both helium and argon were used as spraying atmospheres with little difference in the quantity of vapor produced. However, the arc was more stable in a helium atmosphere.

The adherence of the initial coating of calcium on nickel is influenced by both the white and the dark-gray vapors. Adherence appears good if no trouble is encountered during initiation of the arc. Coatings from one to four passes of noninterrupted spraying were obtained this month. The four-pass coating required 2 min of continuous operation, during which the chamber filled with a large quantity of vapor, indicating that if the initial coating can be applied, the vapors have but little effect on subsequent layers of calcium and that the process could be developed for continuous operation.

Variables studied during the period included current input, wire-feed rate, arc-to-base metal distance, wire and base-metal preparation prior to spraying, and drying of gases. Drying the helium or the argon before spraying reduced the quantity of vapor slightly. Drying the calcium wire at 900 F after abrasion cleaning and before spraying, not only reduced the quantity of vapor, but caused it to form as a fine mist rather than as a flocculent material. The most acceptable coating was produced with a single-pass spray in a dried argon atmosphere.

Coatings produced with dried gas and dried calcium wire indicate that arc spraying has possibilities as a method for coating nickel with calcium. Unsatisfactory adherence between calcium and nickel is, at present, the major deterrent to the process.

Experimental work for the next period will be directed toward improving the calcium-to-nickel bond, further interpretation of the cause of the vapors, and continued attempts to reduce these vapors. A topical report will be prepared during the latter part of the period.

F. RESEARCH FOR AEC REACTOR DEVELOPMENT DIVISION PROGRAM

S. J. Paprocki and R. F. Dickerson

REACTOR MATERIALS AND COMPONENTS

R. F. Dickerson

The study of the effect of additions in stabilizing the fluorite structure of UO_2 has been continued. Current work is being devoted to reduction of the amount of additive required for stabilization. Previous experiments have shown that U_3O_8 transforms to a new crystal structure above 400 C when subjected to pressures above 15,000 atm. More complete studies show that this "gamma" U_3O_8 occurs at pressures of 25,000 to 37,500 atm and at temperatures between 400 and 600 C. Data obtained also indicate that at pressures above 60,000 atm the U_3O_8 transforms to a cubic structure.

The irradiated zirconium-2 w/o uranium alloy hydride has been examined in the hot cell. The results of this examination were encouraging since the material showed little or no effect of the irradiation.

Irradiation of the capsules containing tensile, cyclic-strain fatigue, and impact specimens has been continued through ETR Cycles 16 and 17. Three additional capsules to be irradiated at process-water temperatures for postirradiation-annealing studies have been shipped to the ETR. Because of uncertainties as to the availability of reactor space in the near future, no definite plan has been evolved for the irradiation of the capsules designed to allow the specimens to gamma heat to about 600 F.

The niobium alloys being studied for possible application as alternate cladding materials in future EBR cores have been cold rolled preparatory to machining tensile specimens. This sheet will also be used for weldability studies. As a part of the program concerned with the development of water-corrosion-resistant niobium alloys, hot-hardness data have been obtained that indicate that ternary alloys containing 2.5 a/o vanadium-2.5 a/o iron, 2.5 a/o zirconium-5 a/o vanadium, and 10 a/o zirconium-5 a/o iron might be considered most attractive. Corrosion tests were initiated on 32 binary and ternary screening alloys prepared from high-purity niobium. After 7 days of exposure in 680 F water, it appears that specimens containing more than 35 a/o zirconium were best. The commercial-purity niobium-base alloys which were exposed 196 days in 680 F water have been removed from test.

Friction between a molybdenum ball and a molybdenum flat in a sodium environment was further investigated. The data indicate that frictional resistance decreases as the oxide surface layers are removed by exposure to high temperatures and moderate vacuum. The surface film alters from MoO_3 to MoO_2. Differential thermal analysis indicates a reaction between MoO_2 and molten sodium at temperatures around 750 F. Studies of electron micrographs show that dry friction produces much more extensive wear than does sodium-lubricated friction.

F-2

Valence Effects of Oxide Additions to Uranium Dioxide

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

An investigation is being made of the valence effects of oxide additions to UO_2 in stabilizing the fluorite structure under both oxidizing and reducing conditions. Previous research has shown that when ion size and relative valence are favorable, extensive solid solutions are possible between UO_2 and sesquioxides such as La_2O_3. A "valence compensation" mechanism appears to be operative in such solid solutions. Thus, UO_2 containing 50 mole per cent La_2O_3 reacts, under reducing conditions, to form an oxygen-defect solid solution. Oxidation tends to produce a more nearly stoichiometric material of fluorite structure and reduced lattice parameter. The resulting material is stable at high temperatures under oxidizing conditions.

Current work has been devoted to reduction of the amount of additive required for stabilization. Attempts are being made to substitute in part CaO for La_2O_3 since divalent additives should be more effective in valence compensation. A new series of materials has been prepared and is currently being evaluated. The compositions of these materials in mole per cent are:

UO_2-20 La_2O_3-20 CaO

UO_2-25 La_2O_3-25 CaO

UO_2-20 Y_2O_3-20 CaO

UO_2-25 Y_2O_3-25 CaO.

High-Pressure High-Temperature Solid-State Studies

W. B. Wilson and C. M. Schwartz

An investigation is being made of the effects of combined high pressure and high temperature on uranium oxide. A part of the current investigation has been devoted to study of the U_3O_8 pressure-temperature phase diagram. Early results had disclosed that U_3O_8 transforms to a new crystal structure above 400 C when subjected to pressures above 15,000 atm. This transformation has been repeatedly observed in several types of ultrahigh-pressure apparatus. Microbalance analyses of the new U_3O_8 indicated that its composition of $UO_{2.65}$ is the same as the material from which it is produced and is not the result of chemical decomposition of U_3O_8 under pressure. The new modification, tentatively designated gamma U_3O_8, has been observed at 25,000 and 37,500 atm at temperatures between 400 and 600 C.

Recent experiments using a Bridgman-anvil apparatus have shown that at a 60,000-atm pressure at 400 and 500 C the gamma U_3O_8 coexists with another phase having a cubic fluorite structure with a lattice parameter the same as that of U_4O_9. Microbalance analyses of similar samples (produced at 100,000 atm in an internally heated die) have indicated that, although the structure is that of U_4O_9, its chemical composition is very close to that of U_3O_8. These results indicate that the effect of pressure is to transform U_3O_8 to a cubic structure at pressures greater than 60,000 atm and suggest that at pressures greater than 100,000 atm the phase field between UO_2 and U_3O_8 is entirely of cubic structure. These high-pressure forms were observed to revert to normal U_3O_8 when fired in air at 750 C for microbalance analyses.

The results obtained on U_3O_8 using an internally heated die are less satisfactory than for the Bridgman anvil owing to larger uncertainties of both temperature and pressure. In nearly all instances, however, a cubic structure is found which may also coexist with any of the other oxides of uranium. This suggests that there is a cubic phase field, a gamma phase field, an alpha-plus-cubic phase field, and a gamma-plus-cubic phase field as a function of both temperature and pressure. The cubic U_4O_9 structure in those samples analyzed to date has contained more oxygen than is observed at normal pressure. In some instances, reduction has occurred, however, and is believed to occur when the platinum heater has melted or ruptured, allowing oxygen to escape from the samples.

Modifications on the girdle-type die allowing full support of all parts has been completed. Methods to eliminate the temperature gradient in the heater have been devised. Improved temperature measurement by direct insertion of the thermocouple into the U_3O_8 sample material has been utilized. Work will continue to characterize the U_3O_8 pressure-temperature diagram and to study high-pressure reactions of uranium oxides.

Fueled Zirconium Hydride Moderator

G. E. Lamale, A. W. Hare, and H. H. Krause

Postirradiation examination of the specimens of hydrided zirconium-2 w/o uranium is nearing completion. Changes in physical properties from the preirradiation condition were slight.

Structure and Pressure-Composition-Temperature Studies

This portion of the program has been completed and a report will issue soon.

Radiation-Damage Studies

The postirradiation examination of Capsules BMI-20-1 and BMI-20-2, which contained specimens of zirconium hydride and zirconium hydride-2 w/o enriched uranium, is being completed at the BMI Hot-Cell Facility. Both capsules were irradiated at the MTR to burnups of about 20 a/o of the uranium. Both the fueled cylinders and the nonfueled cylinders were irradiated in an atmosphere of hydrogen. The recorded center-line temperatures of the specimens during the irradiation varied from about 1000 to 1400 F for both capsules.

Tables F-1 and F-2 are summaries of the postirradiation dimensions. Each length was measured twice, and the diameter was measured at eight different sections. The fueled specimens decreased in length but increased slightly in diameter. The nonfueled specimens did not show any consistent increase or decrease, although the dimensional changes appeared to be somewhat larger than for the fueled specimens.

The percentage change in density of the fueled specimens is based on a comparison with some unirradiated $ZrH_{1.65}$-2 w/o uranium, and similarly, the percentage change of the nonfueled specimen is based on a comparison with unirradiated $ZrH_{1.65}$. In both cases, the changes in density between the preirradiation and postirradiation condition are negligible.

Metallography has been completed on the fueled specimens. All four samples had similar appearances. They exhibited a mixture of grain structures from small-equiaxed grains to large columnar grains. There was considerable cracking in all four specimens, but this could have been caused by the shock from the cutting with the dry cutoff wheel.

All of the four samples selected for metallography were adjacent to portions of the fueled material selected for radiochemical burnup analyses.

Irradiation-Surveillance Program on Type 347 Stainless Steel

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

An evaluation of the effect of irradiation on the mechanical properties of AISI Type 347 stainless steel is being conducted as a surveillance-type program. Changes in mechanical properties resulting from fast-neutron irradiation (neutrons having energies greater than 1 Mev) at 120 and 600 F are being investigated in support of the KAPL-C-33 loop and other loops using Type 347 stainless steel in the ETR. The tensile, cyclic-strain fatigue, and impact properties are to be measured after intervals of exposure comparable to 6-month periods of exposure of the KAPL-C-33 loop. Since fast-neutron damage data are available for this material for exposures up to 3.74×10^{21} n per cm^2, the 6-month periods will be measured starting with that accumulated exposure as a base. It is expected that the total fast-neutron exposure will be approximately 1.6×10^{22} n per cm^2. A series of capsules containing tensile and cyclic-strain fatigue specimens will be used in postirradiation-annealing studies to supplement data obtained from specimens exposed at 600 F.

Irradiation of eight capsules containing tensile, cyclic-strain fatigue, and impact specimens at process-water temperature, "cold" capsules, has continued through ETR Cycles 16 and 17. Total exposure for the period starting May 21 through June 14, 1959, represents approximately 15 days at full reactor power. The capsules are in the K-8 and the L-8 positions. Three additional capsules to be irradiated also at process-water temperatures for postirradiation-annealing studies have been fabricated, assembled, and shipped to the ETR.

TABLE F-1. POSTIRRADIATION DATA OF IRRADIATED FUELED SPECIMENS OF ZrH$_{1.65}$-2 w/o URANIUM

Capsule	Specimen	Location in Capsule	Dimensional Change, per cent		Instantaneous Flux, 10^{13} n/(cm^2)(sec)	Temperature, F	Uranium Burnup, per cent	Irradiation Time, days
			Length	Diameter				
BMI-20-1	1-9[(a)]	Top	0.0	0.60	3.94	1140	25.0	100
	1-13	Middle	0.0	0.30	4.38	--	--	100
	1-11	Bottom	-0.08	0.00	4.51	1200	23.2	100
BMI-20-2	1-12[(a)]	Top	-0.31	0.0	2.44	978	13.9	134
	1-16	Middle	-0.13	0.40	2.75	--	--	134
	1-14	Bottom	-0.18	0.0	3.16	1152	13.6	134

(a) Metallography and burnup analyses performed on these specimens.

TABLE F-2. POSTIRRADIATION DATA OF ZrH$_{1.65}$ SPECIMENS

Capsule	Specimen	Location	Dimensional Change, per cent		Instantaneous Flux, 10^{13} n/(cm^2)(sec)
			Length	Diameter	
BMI-20-1	S-3	Top	0.48	0.45	4.38
	S-6	Bottom	-0.36	0.15	4.38
BMI-20-2	S-10	Top	0.0	1.3	2.75
	S-2	Bottom	0.07	-0.49	2.75

Operation of the ETR with fuel in the L-7 and M-7 positions has continued to give high fast fluxes in the vicinity of the experiment. Exposure during the period May 21 to June 14, 1959, is equivalent to 15.1 days at full power. The total accumulated fast flux for each capsule calculated on that basis is shown in Table F-3.

TABLE F-3. DATA ON "COLD" CAPSULES OF THE TYPE 347 STAINLESS STEEL IRRADIATION-DAMAGE PROGRAM

Capsule	Type of Specimen in Capsule	Approximate Removal Date(a)	Approximate Exposure at Time of Removal(b), 10^{22} nvt	Estimated Total Fast-Flux Exposure, as of June 14, 1959, 10^{21} nvt: Top	Bottom
BMI-24-2	Tensile and fatigue	Jan., 1962	1.31	2.260	3.120
BMI-24-4	Tensile and fatigue	Jan., 1963	1.78	1.327	2.468
BMI-24-6	Tensile and fatigue	June, 1961	1.08	3.198	2.434
BMI-24-8	Tensile and fatigue	June, 1962	1.54	1.499	2.477
BMI-24-10	Tensile and fatigue	Jan., 1961	0.84	2.490	2.840
BMI-24-12	Tensile and fatigue	June, 1960	0.61	3.389	2.722
BMI-24-14	Impact	June, 1962	1.54	2.994	3.079
BMI-24-16	Impact	June, 1960	0.61	3.052	2.886

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

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

Fast flux is to be determined from five nickel dosimeters placed in the four faces and the center hole of the core-filler piece. These were inserted at the beginning of Cycle 15 and will remain until approximately an exposure of 5000 megawatt-days has been accumulated. It now appears that this will be accomplished during Cycle 18.

Plans are incomplete with regard to procedures to determine and monitor the temperature of the capsules containing tensile, cyclic-strain fatigue, and impact specimens to be irradiated at 600 F. It had been planned to heat these specimens by gamma heating; hence, some relationship between existing gamma flux in the test position and specimen temperature needs to be established. It is expected that some difficulty will be experienced in securing a position having a high enough fast flux for a period sufficiently long to establish effects of damage occurring at 600 F.

Development of Niobium-Base Alloys

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

Several niobium-base alloys for possible application as alternate cladding materials in future EBR cores are being evaluated. The alloys currently being studied are niobium-1 w/o chromium, niobium-2 w/o chromium, niobium-4.5 w/o zirconium, niobium-10 w/o tantalum-2 w/o chromium, niobium-20 w/o titanium-1.5 w/o chromium, niobium-40 w/o titanium-10 w/o aluminum, unalloyed niobium, and a comparison alloy of vanadium-10 w/o titanium-1 w/o niobium.

Melting, evaluations of hot and cold fabricability, metallographic examinations, chemical analyses, and a short heat-treatment study have been completed.

Edge cracking occurred during cold rolling of the large hot-forged slabs, including the unalloyed niobium. This indicated that the alloys probably needed to be annealed to relieve stresses developed during forging and cooling. The slabs have been encapsulated in stainless steel evacuated cans and annealed at 1050 C for 1/2 hr. The cracks will be removed and cold rolling continued.

Tensile specimens of each alloy will be prepared for tests at 650 and 800 C. The thermal conductivity will be determined on the three alloys having the highest tensile strengths at 800 C. In the meantime, as soon as sheet material has been obtained, sheet-to-sheet weldability of the cold-fabricable alloys will be studied.

Development of Corrosion-Resistant Niobium Alloys

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

The evaluation of selected niobium-base alloys for possible service in pressurized-water reactors was continued. Elevated-temperature property data are being obtained on high-purity alloys prepared on the basis of water-corrosion-test results conducted on a series of commercial-purity niobium-base alloys. An additional series of screening alloys has been prepared to help optimize new compositions of interest.

Hot-hardness data on 24 niobium-base alloys were obtained (in a vacuum atmosphere) and are listed in Table F-4. Of these alloys, the behavior of those ternaries containing 2.5 a/o vanadium-2.5 a/o iron, 2.5 a/o zirconium-5 a/o vanadium, and 10 a/o zirconium-5 a/o iron appears most attractive. An anomalously large increase in hardness was observed for some alloys (the binary 5 a/o and 10 a/o zirconium alloys and the ternary alloys containing 10 a/o zirconium with 5 a/o each of titanium, molybdenum, and chromium) at 1400 and 1650 F. This was interpreted as the result of contamination during testing. Since other alloys tested simultaneously with these samples did not show this behavior, it is apparent that niobium-base alloys containing 5 to 10 a/o zirconium are exceedingly susceptible toward contamination hardening.

Fabrication of two binary alloys containing 5 a/o and 10 a/o iron and two ternary alloys containing 2.5 a/o vanadium-2.5 a/o zirconium and 2.5 a/o vanadium-2.5 a/o nickel was carried out by pack rolling in stainless steel at 1800 F. Good-quality strip was obtained from each alloy. These have been vacuum annealed at 2190 F (1200 C) and samples are being prepared for water-corrosion and hot-hardness testing.

Corrosion tests were started on 32 binary and ternary screening alloys prepared from high-purity niobium. Results after 7 days of exposure in 680 F water for these alloys are summarized in Table F-5 along with those for all alloys tested to date. Specimens containing more than 35 a/o zirconium exhibited black, shiny tarnish films, while all other alloys were covered with dull dark-gray films.

TABLE F-4. HOT-HARDNESS DATA FOR HIGH-PURITY NIOBIUM-BASE ALLOYS

Alloy	Nominal Alloy Content (Balance Niobium), a/o	Annealing Temperature, F	Hardness, VHN, at Temperature Shown						
			75 F	600 F	900 F	1200 F	1400 F	1650 F	75 F(a)
TP37	100 Nb	(b)	81	78	56	37	26	21	82
N-2	1 Zr	2190	108	75	71	75	87	84	123
N-4	5 Zr	2730	152	101	97	99	109	184	179
N-5	10 Zr	2730	187	142	133	133	158	267	238
K-1	0.6 V(c)	2910	111	75	84	77	74	64	119
K-2	1.2 V(c)	2910	110	67	73	74	73	68	122
K-3	2.0 V(c)	2910	127	79	79	79	76	71	137
N-16	2.5 V	2190	103	67	65	66	63	58	114
K-4	3.5 V(c)	2910	139	95	90	84	81	73	147
NL-2	12.6 V	2730	246	164	155	153	153	149	259
N-11	2.5 Ti	2190	97	52	49	45	42	41	109
N-12	10 Ti	2190	121	69	63	58	57	66	141
N-13	25 Ti	2190	152	84	75	68	67	77	194
N-45	1 Fe	2190	98	72	76	60	60	52	97
N-48	10 Cr(d)	2190	120	82	78	77	77	64	123
N-49	2.5 V-2.5 Ti	2190	119	75	66	68	71	69	129
N-50	2.5 V-2.5 Mo	2190	134	83	77	71	69	77	134
N-51	2.5 V-2.5 Fe	2190	143	104	101	103	104	85	149
N-53	2.5 V-2.5 Cr	2190	112	80	65	66	65	61	111
N-54	2.5 V-2.5 Al	2190	104	73	65	63	69	58	112
N-17	2.5 Zr-5 V	2190	159	117	115	106	119	145	179
N-21	10 Zr-5 Ti	2730	207	135	130	135	151	223	239
N-29	10 Zr-5 Mo	2730	238	146	148	152	178	246	268
N-35	10 Zr-5 Cr	2730	208	173	169	166	169	221	236
N-38	10 Zr-5 Fe	2730	322	237	247	237	249	168	338

(a) Determined after heating to 1650 F.

(b) Tested in as-cast condition; values previously reported.

(c) Prepared at KAPL.

(d) High weight losses in melting indicate the actual chromium content is considerably less than 10 a/o.

TABLE F-5. SUMMARY OF CORROSION RESULTS OBTAINED ON NIOBIUM ALLOYS EXPOSED IN HIGH-TEMPERATURE WATER AND STEAM

	600 F Water		680 F Water		750 F Steam	
Alloy Addition (Balance Niobium), a/o	Exposure Time, days	Total Weight Change, mg per cm^2	Exposure Time, days	Total Weight Change, mg per cm^2	Exposure Time, days	Total Weight Change, mg per cm^2
			Commercial Niobium, Rocking-Hearth Melts			
Unalloyed Nb	112	-1.47	42	Disintegrated	28	Disintegrated
10.5 Zr	--	--	196	0.67	28	0.69
26.1 Zr	--	--	196	0.67	--	--
35.7 Zr	--	--	196	0.66	--	--
45.7 Zr	--	--	196	0.55	--	--
1.08 W	--	--	196	-2.60	28	0.97
4.67 W	--	--	196	-29.3	--	--
9.56 W	--	--	7	Cracked	--	--
2.45 Mo	--	--	196	-7.10	28	0.72
5.20 Mo	--	--	196	-1.30	28	0.72
7.40 Mo	--	--	196	0.62	--	--
4.42 V	--	--	196	0.42	28	0.69
6.59 V	--	--	196	0.73	28	0.55
8.93 V	--	--	196	0.59	28	0.61
10.7 V	--	--	196	0.78	--	--
13.7 V	--	--	196	0.50	--	--
24.2 V	--	--	196	0	--	--
4.90 Fe	--	--	196	0.10	28	1.49
9.41 Ti	--	--	196	0.65	28	0.59
18.8 Ti	--	--	196	0.48	--	--
24.3 Ti	--	--	196	0.52	--	--
40.5 Ti	--	--	196	0.40	--	--
33.8 Ti	--	--	196	0.33	--	--
12.0 Ti-0.5 Cr	--	--	196	0.66	--	--
20.2 Ti-2.1 Cr	--	--	196	0.39	--	--
28.2 Ti-6.1 Cr	--	--	196	0.20	--	--
12.0 Ti-4.2 Mo	--	--	196	0.64	--	--
17.4 Ti-6.2 Mo	--	--	196	0.54	--	--
23.1 Ti-7.8 Mo	--	--	196	0.45	--	--
10.4 Ti-5.0 V	--	--	196	0.56	--	--
16.1 Ti-8.4 V	--	--	196	0.40	--	--
22.6 Ti-1.0 V	--	--	196	0.48	--	--

TABLE F-5. (Continued)

Alloy Addition (Balance Niobium), a/o	600 F Water		680 F Water		750 F Steam	
	Exposure Time, days	Total Weight Change, mg per cm^2	Exposure Time, days	Total Weight Change, mg per cm^2	Exposure Time, days	Total Weight Change, mg per cm^2
High-Purity Niobium, Consumable-Electrode Melts						
Unalloyed Nb	112	0.63	112	0.91	28	Disintegrated
7.18 Mo	112	0.46	112	0.55	70	Cracked
12.5 V	112	0.32	112	0.50	112	0.67
46.8 Zr-5.06 Ti	84	0.24	84	0.84	84	-1.52
11.2 Ti-3.2 Mo	84	0.37	84	0.53	84	0.76
18.8 Ti-8.7 Mo	84	0.23	84	0.38	84	0.47
9.9 Zr-9.4 V	7	0.21	7	0.24	7	0.31
5.7 Zr-11.4 V	7	0.18	7	0.20	7	0.24
9.1 Ti-6.3 Cr	7	0.16	7	0.17	7	0.18
High-Purity Niobium, Rocking-Hearth Melts						
Unalloyed Nb	--	--	28	1.08	--	--
Unalloyed Nb	--	--	7	-1.50	--	--
	--	--	7	0.66	--	--
1 Zr	--	--	28	-22.2	--	--
5 Zr	--	--	7	0.45	--	--
10 Zr	--	--	7	-0.10	--	--
40 Zr	--	--	7	0.12	--	--
65 Zr	--	--	7	0.21	--	--
75 Zr	--	--	7	0.38	--	--
90 Zr	--	--	7	0.18	--	--
2.5 Ti	--	--	28	0.42	--	--
10.0 Ti	--	--	28	0.37	--	--
25.0 Ti	--	--	28	0.19	--	--
1 Cr	--	--	28	0.91	--	--
5 Cr	--	--	28	0.54	--	--
10 Cr	--	--	7	0.34	--	--
1 Fe	--	--	7	0.58	--	--
10 Zr-5 Ti	--	--	7	0.18	--	--
25 Zr-5 Ti	--	--	7	0.34	--	--
25 Zr-15 Ti	--	--	7	0.22	--	--
25 Zr-25 Ti	--	--	7	0.07	--	--
35 Zr-5 Ti	--	--	7	0.15	--	--
35 Zr-15 Ti	--	--	7	0.13	--	--
45 Zr-5 Ti	--	--	7	0.08	--	--

TABLE F-5. (Continued)

Alloy Addition (Balance Niobium), a/o	600 F Water		680 F Water		750 F Steam	
	Exposure Time, days	Total Weight Change, mg per cm^2	Exposure Time, days	Total Weight Change, mg per cm^2	Exposure Time, days	Total Weight Change, mg per cm^2
High-Purity Niobium, Rocking-Hearth Melts (Continued)						
10 Zr-5 Mo	--	--	7	0.02	--	--
35 Zr-5 Mo	--	--	7	0.21	--	--
45 Zr-5 Mo	--	--	7	0.05	--	--
35 Zr-5 Al	--	--	7	0.24	--	--
45 Zr-5 Al	--	--	7	0.13	--	--
10 Zr-5 Cr	--	--	7	0.17	--	--
45 Zr-5 Cr	--	--	7	0.10	--	--
10 Zr-5 Fe	--	--	7	0.13	--	--
2.5 V	--	--	28	0.57	--	--
2.5 V-2.5 Ti	--	--	7	0.15	--	--
2.5 V-2.5 Mo	--	--	7	0.18	--	--
2.5 V-2.5 Fe	--	--	7	0.32	--	--
2.5 V-2.5 Cr	--	--	7	0.41	--	--
2.5 V-2.5 Al	--	--	7	0.32	--	--
5 V-2.5 Zr	--	--	7	0.09	--	--
5 V-25 Zr	--	--	7	0.34	--	--
5 V-35 Zr	--	--	7	0.29	--	--
5 V-45 Zr	--	--	7	0.02	--	--

The data presented in Table F-5 are for exposures ranging up to 196 days and continue to indicate that a number of alloying elements improve the corrosion resistance of niobium significantly. Alloys which possess adherent tarnish films and appear most corrosion resistant are a ternary alloy containing 28.2 a/o titanium-6.1 a/o chromium and those which contain more than 35 a/o zirconium.

The commercial-purity niobium-base alloys which have been exposed 196 days in 680 F water have been removed from test. These specimens will be examined for: (1) nature and composition of the oxide film, (2) oxygen contamination as measured by hardness traverses, and (3) hydrogen absorption. Corrosion testing of the remaining alloys described in Table F-5 is being continued.

Rubbing Surfaces in Sodium Environments

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

This study of friction and wear of materials immersed in molten sodium is aimed at (1) increasing understanding of the physical and chemical phenomena involved, and (2) discovering materials and environmental conditions which improve operating characteristics. During June, friction between a molybdenum ball and flat was further investigated.

A series of curves was obtained by plotting the average coefficient of kinetic friction, measured from friction recordings, versus successively higher temperatures over the range of 80 to about 1350 F. These indicate that the frictional resistance decreases as the oxide surface layers are removed by exposure to high temperatures and moderate vacuum. Electron-diffraction analysis showed that the surface film alters from MoO_3 to MoO_2 under the conditions described above. Differential thermal analysis indicates the presence of a reaction between MoO_2 and molten sodium at temperatures around 750 F. Another differential thermal analysis between sodium and molybdenum chips gave no indication of a "wetting" reaction over the temperature range of 200 to about 1400 F. Electron micrographs were obtained for lapped and for etched molybdenum surfaces, and for wear tracks across these surfaces. From these, numerous stress cracks were observed to exist throughout the base metal; and dry friction was observed to produce much more extensive wear than sodium-lubricated friction.

STUDIES OF ALLOY FUELS

R. F. Dickerson

Various properties of binary alloys of niobium and 10, 20, 30, 40, 50, and 60 w/o uranium are being studied. After a 50-hr exposure to CO_2 at 600 F, reasonable resistance to corrosion was noted. A black adherent film formed in all cases. A series of alloys was also exposed to NaK at 1600 F. In most cases a black adherent oxide film formed; however, in the case of the 10 w/o uranium alloy, weight-change measurements showed that the corrosion film spalled away slightly. Corrosion tests in 600 and 680 F water performed to date (42- and 56-day exposures) show that, with two exceptions, the alloys are as good as or better than unalloyed niobium. There were one 10 w/o uranium alloy and one 60 w/o uranium alloy. Initial tensile tests show that the 20 w/o uranium alloy exhibits exceptionally high strength at room temperature and at 1600 F.

Thorium-uranium alloys can be strengthened by additions of zirconium and zirconium-niobium. At 600 C the hardness is increased by a factor of two by using these additions. Additions of zirconium and niobium reduce the water-corrosion rate of thorium by about one-half. The recrystallization temperature for binary alloys is about 100 C higher than for thorium itself.

Development of Niobium-Uranium Alloys

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

Niobium-rich niobium-uranium alloys have not received much consideration as metallic fuel materials because of a lack of information concerning their properties. Previous work done on the uranium-rich uranium-niobium alloys indicated that their properties are influenced by impurity content. The major impurities in niobium are oxygen and zirconium. The effect of the above-mentioned impurities on the fabricability, mechanical and physical properties, and compatibility in various environments is being studied.

Three grades of niobium were used in alloy preparation: one containing 0.74 w/o zirconium and 600 ppm oxygen, a second containing 0.17 w/o zirconium and 700 ppm oxygen, and a third containing 0.02 w/o zirconium and 300 ppm oxygen. Niobium-10 and -20 w/o uranium alloys have been fabricated to slab material. No effect of impurity content has been noted during fabrication.

Fabrication of alloys containing more than 20 w/o uranium has been unsuccessful at 3000 F. Additional alloys (niobium-30 and -40 w/o uranium) have been prepared and will be fabricated from a high-temperature furnace (3200 F). Fabrication of these alloys by extrusion will be attempted since the niobium-40 w/o uranium alloy has been extruded successfully.

Table F-6 lists additional results of oxidation of alloys in air at 572, 662, and 752 F. Good corrosion behavior was exhibited by all alloys at 572 F; at this temperature for at least 500 hr, a black adherent oxide was formed. In contrast, a brown spalling oxide was formed after 40 hr of testing at 662 and 752 F. No effects due to impurity levels of the base niobium were noted in corrosion testing.

Corrosion data for niobium-uranium alloys in CO_2 at 600 F are listed in Table F-7. All alloys formed a black adherent oxide similar to that formed at 572 F in air.

TABLE F-6. CORROSION DATA FOR NIOBIUM-URANIUM ALLOYS IN AIR(a)

	Impurity Content			Total Weight Change, mg per cm^2		
					After 100 Hr	
Nominal Alloy Content (Balance Niobium), w/o	Oxygen, ppm	Zirconium, w/o	Specimen Condition	After 500 Hr at 572 F	At 662 F	At 752 F
10 uranium	700	0.17	Fabricated	0.35	--	25.7
	300	0.02	Fabricated	0.80	1.07	2.34
20 uranium	600	0.74	Fabricated	0.40	--	44.0
30 uranium	600	0.74	As cast	0.17	25.2	164.0
	700	0.17	Fabricated	0.29	20.0	27.6
	300	0.02	As cast	--	160.0	288.0
40 uranium	600	0.74	As cast	0.08	--	--
50 uranium	600	0.74	As cast	0.28	--	--
	700	0.17	As cast	0.31	--	--
	300	0.02	As cast	0.18	--	--
60 uranium	600	0.74	As cast	1.36	--	--
	700	0.17	As cast	0.43	--	--
	300	0.02	As cast	0.40	--	--

(a) Average of two specimens.

TABLE F-7. CORROSION DATA FOR NIOBIUM-URANIUM ALLOYS IN CO_2 AT 600 F(a)

	Impurity Content			
Nominal Alloy Content (Balance Niobium), w/o	Oxygen, ppm	Zirconium, w/o	Specimen Condition	Total Weight Change After 50 Hr in 600 F CO_2, mg per cm^2
10 uranium	600	0.74	Fabricated	0.17
	700	0.17	Fabricated	0.28
	300	0.02	Fabricated	0.23
20 uranium	600	0.74	Fabricated	0.43
	700	0.17	Fabricated	0.12
	300	0.02	Fabricated	0.13
30 uranium	600	0.74	As cast	0.92
	700	0.17	Fabricated	0.65
	300	0.02	As cast	0.51
40 uranium	600	0.74	As cast	0.44
	600	0.17	As cast	0.36
	300	0.02	As cast	0.22
50 uranium	700	0.74	As cast	0.32
	600	0.17	As cast	0.28
	300	0.02	As cast	0.33
60 uranium	600	0.74	As cast	0.34
	700	0.17	As cast	0.39
	300	0.02	As cast	0.33

(a) Average of two specimens.

Corrosion data for selected niobium-uranium alloys in NaK at 1600 F for 156 hr are shown in Table F-8. A black oxide was formed which was not adherent in all cases, as shown by a loss of weight in the niobium-10 w/o uranium specimens.

The results of testing in water at 600 and 680 F after 42 and 56 days are shown in Table F-9. These alloys are exhibiting a corrosion life comparable to or better than unalloyed niobium at both 600 and 680 F. In 750 F steam (Table F-10), all alloys are losing weight but none has disintegrated, as did unalloyed niobium at 28 days. All of the niobium-uranium alloys formed a black oxide which spalled from the surface, and a new oxide layer formed. In general, no effect on the corrosion life of the alloys tested due to differences in impurity levels of the niobium melting stock can be detected.

Room-temperature tensile properties are listed in Table F-11. The niobium-20 w/o uranium alloys exhibited exceptional strength.

The tensile strengths of the niobium-20 w/o uranium alloys at 1600 F (Table F-12) are quite high, indicating that this alloy should perform well for high-temperature application.

Suitable specimens have been prepared for thermal-conductivity, electrical-resistivity, and thermal-expansion measurements.

Construction of a high-temperature furnace for future experiments is nearly complete.

Development of Thorium-Uranium Alloys

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

Development of thorium-uranium alloys with improved irradiation resistance and corrosion life is being sought by means of process control and ternary or quaternary alloying. Effects of various fabrication variables on the size and distribution of uranium particles within the thorium binary alloys are being investigated. Additions of molybdenum, niobium, or zirconium are being studied; these elements are added for the purpose of strengthening the thorium matrix and/or stabilizing the gamma-uranium phase.

Small uranium particle size and a random distribution of particles are considered desirable for optimum radiation behavior. In studies of thorium-uranium alloys, it was found that, if the uranium content exceeds about 15 w/o, the particle sizes tend to be large and concentration of uranium in grain boundaries occurs. Also, if induction-melting instead of arc-melting techniques are used greater grain-boundary concentrations of uranium are found. However, if these induction-melted alloys are rapidly cooled from 1000 C, the uranium is more evenly distributed. Subsequent working of these alloys shows that no significant change in as-cast structure occurs until about a 60 per cent reduction is effected. Then the structures show definite evidences of stringering.

TABLE F-8. CORROSION DATA FOR NIOBIUM-URANIUM ALLOYS IN NaK AT 1600 F[(a)]

Nominal Alloy Content (Balance Niobium), w/o	Impurity Content: Oxygen, ppm	Impurity Content: Zirconium, w/o	Specimen Condition	Total Weight Change After 156 Hr in 1600 F NaK, mg per cm^2
10 uranium	600	0.74	Fabricated	0
	700	0.17	Fabricated	-0.66
	300	0.02	Fabricated	-0.33
20 uranium	600	0.74	Fabricated	0.92
	700	0.17	Fabricated	0.67
	300	0.02	Fabricated	0.31
30 uranium	300	0.02	As cast	0.93
40 uranium	300	0.02	As cast	1.48
50 uranium	300	0.02	As cast	0.96
60 uranium	300	0.02	As cast	1.36

(a) Average of two specimens.

TABLE F-9. CORROSION DATA FOR NIOBIUM-URANIUM ALLOYS IN 600 AND 680 F WATER AFTER 42 AND 56 DAYS[(a)]

Nominal Alloy Content (Balance Niobium), w/o	Impurity Content: Oxygen, ppm	Impurity Content: Zirconium, w/o	Specimen Condition	Total Weight Change, mg per cm^2: In 600 F Water, 42 Days	In 600 F Water, 56 Days	In 680 F Water, 42 Days	In 680 F Water, 56 Days
100 niobium	131	0.05	Fabricated	2.08	--	1.07	--
100 niobium	500	0.03	Fabricated	3.84	--	-46.8	--
10 uranium	600	0.74	Fabricated	0.31	0.40	0.23	0.16
	700	0.17	Fabricated	0.54	0.74	-5.66	-7.67
	300	0.02	Fabricated	0.63	0.82	-31.8	-38.5
20 uranium	600	0.74	Fabricated	0.49	0.64	0.74	0.91
	700	0.17	Fabricated	0.64	0.78	1.20	1.58
	300	0.02	Fabricated	0.39	0.51	0.66	0.89
30 uranium	600	0.74	As cast	0.63	0.78	0.21	-0.84
	700	0.17	Fabricated	0.50	0.76	-0.43	-0.75
	300	0.02	As cast	0.76	1.03	-0.82	-1.67
40 uranium	600	0.74	As cast	0.99	1.03	1.08	1.41
	700	0.17	As cast	0.51	0.63	0.29	0.51
	300	0.02	As cast	0.51	0.61	1.05	1.30
50 uranium	600	0.74	As cast	0.51	0.68	0.29	0.39
	700	0.17	As cast	0.48	0.62	0.23	0.09
	300	0.02	As cast	0.47	0.55	-0.12	-1.92
60 uranium	600	0.74	As cast	0.15	0.18	-39.0	-57.6
	700	0.17	As cast	0.45	0.62	-4.29	5.95
	300	0.02	As cast	0.30	0.46	-2.41	3.50
Zircaloy-2	--	--	--	0.16	--	0.23	--

(a) Average of two specimens.

TABLE F-10. CORROSION DATA FOR NIOBIUM-URANIUM ALLOYS IN 750 F STEAM[(a)]

Nominal Alloy Content (Balance Niobium), w/o	Impurity Content		Specimen Condition	Total Weight Change in 750 F Steam After Time Shown, mg per cm^2		
	Oxygen, ppm	Zirconium, w/o		28 Days	35 Days	42 Days
10 uranium	600	0.74	Fabricated	-0.09	-0.42	-0.94
	700	0.17	Fabricated	-9.34	-11.6	-13.3
	300	0.02	Fabricated	-21.7	-24.3	-26.3
20 uranium	600	0.74	Fabricated	-5.06	-12.2	-17.0
	700	0.17	Fabricated	-8.53	-15.0	-18.2
	300	0.02	Fabricated	-0.91	-1.60	-2.45
30 uranium	600	0.74	As cast	-2.27	-2.41	-2.80
	700	0.17	Fabricated	-3.34	-3.31	-3.75
	300	0.02	As cast	-5.29	-5.90	-6.00
40 uranium	600	0.74	As cast	-0.50	-0.76	-1.16
	700	0.17	As cast	-0.57	-1.02	-1.67
	300	0.02	As cast	0.68	0.37	-0.71
50 uranium	600	0.74	As cast	-8.70	-10.8	-13.2
	700	0.17	As cast	-5.45	-12.6	-29.7
	300	0.02	As cast	-7.37	-8.70	-9.7
60 uranium	600	0.74	As cast	-16.0	-19.8	-23.1
	700	0.17	As cast	-11.1	-13.6	-16.1
	300	0.02	As cast	-9.0	-11.6	-13.7
Zircaloy-2	--	--	--	--	0.42	0.55

(a) Average of two specimens.

TABLE F-11. ROOM-TEMPERATURE TENSILE PROPERTIES OF NIOBIUM-URANIUM ALLOYS AS FABRICATED AT 2550 F[(a)]

Nominal Alloy Content (Balance Niobium), w/o	Impurity Content: Oxygen, ppm	Impurity Content: Zirconium, w/o	0.1 Per Cent Offset Yield Strength, psi	0.2 Per Cent Offset Yield Strength, psi	Ultimate Tensile Strength, psi	Elongation, per cent	Reduction in Area, per cent	Modulus of Elasticity, 10^6 psi
10 uranium	700	0.17	69,300	(b)	70,000	3	11	18.6
	300	0.02	76,800	(b)	77,900	3	16	15.5
20 uranium	600	0.74	--	84,000	95,500	6	19	18.1
	300	0.02	--	93,200	102,000	7	21	20.3

(a) Average of two specimens.

(b) Strain gages failed before 0.2 per cent offset was reached.

TABLE F-12. TENSILE PROPERTIES AT 1600 F OF WROUGHT NIOBIUM-URANIUM ALLOYS AS FABRICATED AT 2550 F[(a)]

Nominal Alloy Content (Balance Niobium), w/o	Impurity Content: Oxygen, ppm	Impurity Content: Zirconium, w/o	0.2 Per Cent Offset Yield Strength, psi	Ultimate Tensile Strength, psi	Elongation, per cent	Reduction in Area, per cent
10 uranium	600	0.74	33,900	49,500	13	31.3
	700	0.17	(b)	51,000	1	12.0
20 uranium	600	0.74	58,600	71,000	2	16.0
	700	0.17	60,000	71,600	10	37.0
	300	0.02	58,100	68,400	--	0
30 uranium	600	0.74	(b)	71,200	2	0

(a) Average of two specimens.

(b) Extensometer failed before 0.2 per cent offset was reached.

Hot-hardness measurements show that zirconium and zirconium-niobium are potent strengtheners of thorium-uranium alloys. Hot hardness increases by up to a factor of two at 600 C with these additions.

The corrosion rates of some of the ternary and quaternary alloys studied were as little as half that of thorium. This improvement was obtained with alloys of thorium-10 to 25 w/o uranium by adding 10 to 25 w/o of zirconium and in some cases 2 to 4 w/o of niobium also.

Recrystallization of 80 and 90 per cent cold-reduced thorium-5 to 20 w/o uranium alloys is characterized by an initial softening, followed by a leveling off of hardness values. At 600 C the hardness decrease with time at temperature is insignificant, whereas at 725 C it is very prominent and occurs within the first 2-1/2 min for 90 per cent reduced material and within 5 min for the 80 per cent reduced material. Studies conducted on iodide thorium-10 w/o uranium indicate that 90 per cent cold-reduced material is fully recrystallized after 2-1/2 min at 725 C, 10 min at 700 C, and 20 min at 675 C. These results indicate that the recrystallization temperature for binary alloys is about 100 C higher than for thorium itself.

FISSION-GAS RELEASE FROM REFRACTORY FUELS

J. B. Melehan, D. A. Vaughan, R. H. Barnes,
S. D. Beck, and F. A. Rough

Research on various parts of the program to study fission-gas release from UO_2 is reported in this section. Routine progress was made during June on the preparation of specimens for study of diffusion coefficients and for in-pile experiments. Considerable effort was also devoted to construction of apparatus for both types of experiments.

Diffusion in UO_2

A series of thin plates of Norton Company single-crystal UO_2 has been prepared for irradiation and study of fission-gas diffusion by subsequent heating. The specimens were sectioned by an ultrasonic grinding method and were abraded to final size. The specimens were then heat treated for 3 hr at 1800 C in high vaccum to remove a nitride second phase from at least a surface layer. Preirradiation examination of these specimens is now being conducted. Small single crystals of UO_2 supplied by MCW and produced by the Verneuil process are being prepared for similar study.

Characterization of Sintered UO_2 and Model of Gas Release

Sintered UO_2 specimens prepared from two batches of UO_2 powder are being evaluated. After preliminary study of this sintered stock, conditions will be established for preparation of samples for in-pile study of gas release. Details of this work will be reported in the future.

One of the objectives of the diffusion study is to determine whether fission-gas release occurs by a volume diffusion mechanism. In support of this objective and to provide a method of characterizing UO_2 having various structural characteristics, thin-section techniques are being developed. Thus, optical transmission through thin sections is being developed as a potential means of observing lattice defects and of obtaining a three-dimensional view of crystallite boundaries and pore structure. Once the techniques are better developed, it is hoped to correlate these observations of defects with metallographic observations of etched specimens.

Preparation for In-Pile Study

Construction of apparatus for in-pile study of gas release is under way. The apparatus is scheduled for insertion into the BRR in August.

GENERAL FUEL-ELEMENT DEVELOPMENT

S. J. Paprocki

Techniques are being developed for the preparation and cladding of cermets containing 60 to 90 volume per cent of fuel dispersed in metal matrices of niobium, molybdenum, chromium, and stainless steel. These cermets are being prepared by conventional mixing of fuel and matrix powders and by the use of metal-coated UO_2 particles.

Gas-pressure bonding is being investigated as a method for the cladding and bonding of niobium and molybdenum fuel elements and assemblies. Techniques for the surface preparation and bonding of niobium have been developed that produce ductile metallurgical bonds with complete grain growth across the interface.

Stainless-UN and -UC dispersion fuel elements were irradiated at temperatures of 1390 to 1800 F to nominal burnups from 2.5 to 10 a/o of the uranium. The objective of the study is to relate the burnup potential of these dispersions with stainless-UO_2 dispersions containing an equivalent uranium-235 content. It was anticipated that these dispersions, especially the stainless-UN, would be superior to stainless-UO_2 because of the higher concentration of uranium in UN and, consequently, lower volume loading required. Preliminary evaluations indicate that stainless-UN is a very promising fuel material for high-temperature service, and from an irradiation standpoint it may be significantly superior to stainless-UO_2.

A study is being conducted to obtain a better understanding of the solid-state bonding mechanism occurring during a pressure-bonding operation. An attempt is being made to evaluate the results and correlate them with established sintering phenomena and theories, and, if the results warrant, to also develop independent explanations and theories regarding the kinetics of solid-state bonding.

Fabrication of Cermet Fuel Elements

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

Techniques of producing cermets of 90 per cent of theoretical density containing 60 to 90 volume per cent fuel include hot-press forging and densification by use of gas pressure at elevated temperatures. Cermets are being evaluated on the basis of microstructures and physical and mechanical properties. Thermal-conductivity measurements are now being made.

Cermets containing 80 volume per cent UO_2 in chromium have been hot-press forged at 1900 F to 96.5 per cent of theoretical density. The cermets were prepared by green pressing the powder mixture in a 1-in.-diameter die at 40 tsi prior to assembly in a 2-in.-square Type 304 stainless steel frame 0.250 in. thick. Green pellet densities ranged from 75 to 76 per cent of theoretical. Hot-press forging from a furnace at 1900 F to obtain reductions in thickness of 36 per cent resulted in a density of 83.5 per cent of theoretical. Other 80 volume per cent UO_2-chromium pellets hot-press forged at 1900 F resulted in densities of 95.4 and 96.5 per cent of theoretical when reduced 46 and 52 per cent in thickness, respectively. However, surface cracks could be detected on those cermets reduced more than 30 per cent in thickness.

Also, 80 volume per cent UO_2-niobium powder pellets were hot-press forged at 1900 and 2000 F. The green pellets were prepared and assembled in the same manner as the UO_2-chromium cermets described above. However, heating of the sealed packs containing UO_2-niobium green-pressed pellets resulted in swelling of the cover plates; therefore, the packs were fitted with evacuation tubes and evacuated at 2000 F prior to sealing and hot-press forging. These sealed packs were pressed to obtain reductions of 40 to 62 per cent in thickness. All the cermets were cracked in a direction perpendicular to the direction of hot-press forging except the core reduced 62 per cent in thickness. Its resulting density was 87.1 per cent of theoretical. All green cermet densities were from 62 to 65 per cent of theoretical.

Pellets of 80 volume per cent UO_2-molybdenum powders have been green pressed in a 1-in.-diameter die at 40 tsi and assembled in 2-in.-square Type 304 stainless steel hot-pressing packs prior to hot pressing at 1900 F. Green pellets ranged from 75 to 76 per cent of theoretical density. Reductions of 30 to 53 per cent of thickness were achieved. A reduction of 30 per cent in thickness resulted in a cermet processing surface cracks and a density of 85.2 per cent of theoretical. A reduction in thickness of 45 per cent resulted in a density of 94 per cent of theoretical; but, surface cracks were visible. The remainder of the cermets were broken while removing from the hot-press-forging frames.

Thermal-conductivity measurements have been made on a 70 volume per cent UO_2-Type 302B stainless steel rod of 97 per cent of theoretical density. The cermet rod was fabricated by green pressing the 70 volume per cent UO_2-stainless powder mixture and hydrostatic pressing for 3 hr at 2300 F under a helium-gas pressure of 10,000 psi. Thermal-conductivity values of 0.0787, 0.0897, and 0.1307 w/(cm)(C) were, respectively, measured at 100, 400, and 800 C.

The thermal conductivity of a hydrostatically pressed 70 volume per cent UO_2-molybdenum cermet is presently being measured. In addition, hydrostatic pressing of flat-plate cermets to 90 per cent of theoretical density or greater is being investigated.

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

S. J. Paprocki, E. S. Hodge, R. W. Getz, and P. J. Gripshover

The gas-pressure-bonding process is being investigated as a method of fabricating fuel elements clad with molybdenum or niobium. These metals retain their strength at elevated temperatures and possess favorable nuclear properties which make them desirable cladding materials for high-temperature reactor application.

An optimum surface preparation has been developed for niobium which results in bonds of complete integrity during gas-pressure bonding. A number of specimens consisting of two flat plates of niobium were subjected to various surface preparations subsequent to bonding at 2100 F at 10,000 psi for 3 hr. The mating surfaces of these specimens were surface conditioned by end milling, belt abrading, pickling in a solution of 30 parts lactic acid-10 parts nitric acid-10 parts hydrofluoric acid, and pickling in a solution of 65 parts nitric acid-35 parts hydrofluoric acid. Evaluation of these specimens has shown that end milling and belt abrading are not satisfactory surface preparations. Specimens with surfaces prepared by pickling in the 65 nitric acid-35 hydrofluoric acid solution possessed excellent bonds with high strengths and complete bond integrity. High-quality bonds were also obtained using the alternate pickling solution containing lactic acid; however, the bonds obtained with this surface preparation evidenced small voids or dots of light contamination along the original bond interface even though complete grain growth had occurred during bonding. The nitric acid-hydrofluoric acid solution will be used as a standard method of surface conditioning for all future planned niobium-clad specimens.

Ductile niobium-clad specimens have been prepared by pressure bonding by utilizing an improved gettering system in the pressure-bonding autoclave atmosphere and an inner can of niobium inside of the stainless steel envelope. A number of bonded specimens display ductility equal to that of the as-received material. Most of these specimens were enclosed in a Type 304 stainless steel can with an inner can of niobium foil separating the can from the specimens. An additional method of preparing ductile specimens consisted of bonding in a can at low temperatures, employing a zirconium braze, followed by decanning and bonding at high temperatures to achieve complete bonding of the niobium-to-niobium interfaces. This specimen consisted of two flat plates of niobium with a 0.020-in. groove milled in the mating surfaces around the periphery of each plate into which strips of zirconium were placed. The resultant specimen was inserted into a stainless steel can and bonded at 1650 F for 4 hr at 10,000 psi. A strong bond between the niobium and the zirconium was achieved, resulting in a gastight specimen. After the specimen was stripped from the can, it was treated at 2100 F at 10,000 psi for 3 hr. Preliminary results indicate a well-bonded specimen was achieved. This second method of producing ductile niobium-clad specimens permits the elimination of impurities that would be introduced if the specimen were fusion edge welded, and eliminates the need for a barrier layer as the bonding accomplished at the high temperatures does not require a pressure-bonding envelope.

Materials utilized thus far as barrier layers to prevent reaction between the pressure-bonding envelope and the niobium-clad specimens have included graphite, iron-chromium-aluminum alloys, alumina, alumina with titania additions, and hard lime window glass. Of these materials only hard lime glass has shown any potential as a barrier at 2100 F. This material will be investigated further to determine the feasibility of glass as a barrier layer. In addition, it is planned to bond several specimens at lower temperatures using the optimum surface-preparation condition developed. It is hoped that, if bonding at a lower temperature can be accomplished, a material such as graphite may be a satisfactory barrier.

Molybdenum specimens consisting of two plates enclosed in degassed Type 304 stainless steel cans with an inner molybdenum envelope have been bonded at 2300 F and 10,000 psi for 3 hr. The molybdenum in these specimens was subjected to a 4-to-1 cold reduction prior to assembly for bonding. Metallographic examination indicated specimens pickled in nitric acid under kerosene showed the best molybdenum-molybdenum bonds. This surface preparation consistently yields specimens with high bond quality. No other surface preparations investigated yielded satisfactory bonds that were free of contamination.

Molybdenum-clad specimens bonded to date have been extremely brittle. This embrittlement may be attributed to the reaction of the can with the molybdenum specimen. A number of materials are being evaluated as barrier layers between molybdenum and the pressure-bonding container. Flame-sprayed alumina and hard lime glass have looked promising as barriers. Chemical analysis of specimens bonded with window glass, however, has shown that there is an increase in the iron content of specimens during bonding. Efforts are being made to determine the source of the iron contamination.

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

D. L. Keller, G. Cunningham, and A. W. Hare

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.

Four specimens, two of each type, were irradiated in three capsules, BMI-18-1, BMI-18-2, and BMI-18-3, to burnups of about 2.5, 5, and 10 a/o of the uranium. Radiochemical burnup determinations made on four specimens from the first two capsules, BMI-18-1 and BMI-18-2, showed burnups of 2.09 and 2.77 w/o and 4.54 and 6.98 a/o of the uranium. Based on dosimetry data by comparison with the dosimetry results from Capsules BMI-18-1 and BMI-18-2, the burnup of the specimens from BMI-18-3 should be from about 8 to 11 a/o of the uranium. Estimated core temperatures for the specimens during the last reactor cycle ranged from a minimum of 1390 F to a maximum of 1800 F, with the maximum temperature occurring in two specimens from Capsule BMI-18-1.

Of the 12 specimens which were irradiated, 5 developed blisters during the irradiation period. Of particular note is the fact that only one of the UN-type specimens developed a blister, and this particular specimen was one of the two specimens which reached an estimated central core temperature of 1800 F.

The metallographic examination of the specimens contained in Capsule BMI-18-3 indicates that very little change has occurred in the case of the UN dispersion, while in the case of UC there appears to have been some reaction of the UC with the stainless as well as some gas release and subsequent cracking of the matrix and slight bulging of the specimen.

The UN dispersions appeared to have structures similar to UO_2 dispersions irradiated at low temperatures and low burnups, i.e., any cracks present in the UN prior to irradiation appear to have healed, but internal porosity was produced to a greater extent in the irradiated UN particles than normally produced in UO_2. No change could be detected in the core-matrix structure, and no cracks could be detected in the matrix or the core-to-cladding interface. Some void areas could be seen in the matrix; however, these voids are similar to voids observed in unirradiated specimens fabricated in an identical manner. Although comparison of the control specimen with the irradiated specimen has not been completed, it can be stated that the voids are not cracks. No halo effect, as sometimes observed in UO_2 dispersions, was visible.

A complete evaluation of the UC dispersions cannot be made at this time; however, as in the case of UN, cracks in the UC particles were healed, but internal porosity of the irradiated UC was much greater than that of the UN. The method by which the cracks in the matrix were initiated and propagated cannot be deduced from the metallographic specimen; but, as would be expected, it appears that the cracks are extended by proceeding from one UC particle to the next. The results now available do not show whether the cracks follow metal grain boundaries. It is interesting to note that the cracks propagate in the direction of rolling, as would be expected from available data on UO_2 dispersions. In addition to damage caused by release of fission products, there is some evidence that a chemical reaction occurred to a limited extent between the UC and the stainless. Such reactions are known to occur at 1800 F, but it is doubtful that any portion of the core of the specimens from Capsule BMI-18-3 reached that high a temperature. Additional study of the metallographic specimens will be required to clarify the nature and extent of the reaction.

F-25

Factors Affecting Pressure Bonding

G. W. Cunningham and J. W. Spretnak

Hot-pressing techniques are being used to investigate the kinetics and mechanism of solid-state bonding of metals by the application of heat and pressure. At the present time copper is being used to prepare most of the specimens, and the major effort has been directed toward determination of the nature of the interfacial bond.

In recent years several investigators who are conducting basic studies on sintering have indicated that grain boundaries are very effective sinks for vacancies and have shown that sintering rates are increased in the presence of grain boundaries. It is also thought that, at least in copper, low-angle grain boundaries, dislocations, and twin boundaries are not very effective as vacancy sinks. Although it is difficult to ascertain the exact nature of a pressure-bonded interface, it appears that the interface may contain a large concentration of vacancies, depending upon the procedure used to produce the bond. This opinion is somewhat verified by the observation that voids form in the interface upon annealing at high temperatures while, simultaneously, the interface either becomes more narrow or disappears. It is also interesting to note that thicker interfacial boundaries and, consequently, larger voids upon annealing are produced when a high pressure (10,000 psi) is applied at a high rate than when a slow uniform buildup of pressure is used. Such a result may be due to increased friction in the former case. Nevertheless, if one assumes that the interface contains vacancies, then the disappearance of the interface and subsequent grain growth during high-temperature anneals should occur by a mechanism similar to the final stages of sintering. Evaluation of tests run to date, although by no means complete, indicates that such results can be expected.

In earlier tests, previously reported, on specimens in which a pressure of 10,000 psi was rapidly applied at 1100 F, subsequent annealing for 2 hr at 1900 F resulted in the formation of voids at the interface and partial disappearance of the interface. These specimens were not annealed for long enough periods to completely remove the interface, since the presence of the Cu_2O-copper eutectic interfered with the interpretation of the microstructure. In recent tests, specimens which do not contain any oxygen impurity have been used. However, these specimens before hot pressing had grains five to ten times larger in diameter than the previously described specimens, and, although the orientation was not determined by a series of etches, the etched structure revealed that at the interface most of the boundaries were either twin boundaries or straight boundaries which probably connect grains varying only a few degrees in orientation. During the hot pressing, pressures were applied slowly to a total pressure of 10,000 psi at 1100 F. Examination of the pressed structure indicated that possibly some new grains and annealing twins formed during the bonding, but there was very little change in the average grain size. Also, as indicated above, the bond interface was narrow and resembled an ordinary grain boundary. Very little change could be detected in these specimens after annealing at 1900 F for periods up to 48 hr. In some cases the interface appeared to be thicker. Close examination of these areas indicated that closely spaced small voids had appeared. In other areas no change could be detected. An explanation of this difference in behavior in hot-pressed specimens may lie both in a difference in the nature of the interface and a difference in the number of wide-angle boundaries available as vacancy sinks. Additional experiments will be required, but, because the interface in some areas behaves in a similar manner regardless of the bonding method, it appears that the presence of wide-angle grain boundaries is the most important factor.

Other tests in progress which should be beneficial in determining the nature of the interface include a series of specimens in which silver is being diffused into a bonded specimen. It should be possible in this way to compare diffusion along the interface with diffusion along grain boundaries.

FF-1

FF. FUEL-CYCLE PROGRAM STUDIES

GAS-PRESSURE BONDING OF CERAMIC, CERMET, AND DISPERSION FUEL ELEMENTS

S. J. Paprocki, D. L. Keller, E. S. Hodge, C. B. Boyer,
J. B. Fox, and D. E. Kizer

The prime objective of this program is to utilize the gas-pressure-bonding technique to reduce fuel-element manufacturing costs, while maintaining or improving the quality of the fabricated elements. The study is directed toward the refinement and further development of the gas-pressure-bonding process for the simultaneous densification and cladding with stainless steel of ceramic, cermet, and dispersion fuels.

The fabrication studies are primarily concerned with the development of uranium dioxide fuel material clad with Type 304 stainless steel. Several basic fuel-element designs are being investigated. Uranium dioxide was chosen as it is being considered by more power-reactor programs than any other material and the general results that are obtained with this material should also be applicable to other ceramic fuels. The development of uranium dioxide cermet and dispersion systems will also be investigated to a small degree.

The rod and flat-plate shapes are receiving all of the attention in the initial studies. To avoid excessive distortions accompanying densification during pressure bonding, it is desirable to utilize cold-pressed UO_2 compacts with relatively high density. A base density 70 per cent of theoretical has been arbitrarily selected. However, UO_2 cold pressed to both lower and higher densities will also be investigated.

Compacting studies are being conducted to determine effects of compacting pressure, UO_2 powder particle size, type of UO_2 powder, and type of lubricant on the initial cold-pressed density. These cold pressed compacts are clad with stainless, gas-pressure bonded, and tested for density and examined for structure. The desired characteristics of the initial UO_2 powder and pressing conditions differ from those utilized in the conventional pressing and sintering operation. In the latter process, a high initial cold-pressed density is not significant, and, depending on the type of oxide that is used, it is usually undesirable. For this application, a high initial cold-pressed density is essential.

In the use of a flat-plate UO_2 fuel element, it will be necessary to compartmentalize the UO_2 to prevent excessive bulging from the release of fission gas. It may also be desirable to employ full compartments in the rod-type elements to increase the cladding strength and localize damage in the event of a cladding failure. To do this, it will be necessary to develop proper pressure-bonding parameters that will simultaneously densify the UO_2 and produce a metallurgical bond between the stainless steel components.

The effect of the bonding parameters of time, temperature, and pressure on the bonds obtained with Type 304 stainless steel are being investigated. The establishment of a range of optimum gas-pressure-bonding conditions for obtaining acceptable stainless steel bonds will be obtained from this study. Cylindrical specimens consisting of two 1/2-in.-diameter by 1-3/4-in.-long Type 304 stainless steel rods assembled into a 0.020-in.-wall stainless steel tube are being pressure bonded at different bonding parameters. Two types of specimens are being bonded at the same conditions. In one, the rods are butted directly together in the tube; in the other two 0.005-in. disks of as-rolled Type 304 stainless steel are placed between the two rods. This gives a comparison of machined surfaces versus as-rolled surfaces. The components are placed through a standard wash cycle after being pickled in 10 volume per cent nitric acid -2 volume per cent hydrofluoric acid aqueous solution and before assembly. The tubes are evacuated and sealed. After the gas-pressure-bonding cycle, each bonded assembly is machined into a standard 1/4-in.-round tensile-test specimen. The load is applied along the specimen axis in a screw-type tensile machine. The applied load is gradually increased until the specimen breaks. The ultimate strength of the pressure-bonded Type 304 stainless steel and the strength of the bond are obtained by this test. Tensile-test results as a function of bonding parameters and surface conditions are given in Table FF-1. Additional specimens have been bonded for tensile-test evaluations of bond strength as a function of bonding conditions.

A small stainless-clad UO_2 flat-plate subassembly consisting of five fuel plates approximately 0.055 by 2.5 by 5.0 in. was pressure bonded to determine process feasibility. The UO_2 cores were compartmented and possessed a high initial density. Ti-Namel channel spacers were employed. After bonding, the Ti-Namel was dissolved by acid pickling to form coolant channels. The subassembly was sectioned and examined. The results of the evaluation indicate that the gas-pressure-bonding process can be utilized for the bonding of flat-plate UO_2 assemblies in one operation.

DEVELOPMENT OF URANIUM CARBIDE-TYPE FUEL MATERIALS

F. A. Rough and W. Chubb

As part of the AEC's Fuel-Cycle Development Program, an integrated program of research on the preparation, properties, and irradiation resistance of uranium carbides has been initiated at Battelle. The objective of this program is to develop the technology of uranium carbides, particularly uranium monocarbide, as reactor fuels. These new fuels are being studied because of potential economies in fuel-element production and fuel-element reprocessing for power reactors.

A technique was recently developed at Battelle for casting molten uranium carbide into geometric shapes; and the present program is an attempt to learn more about the properties of this cast product and to further develop fabrication and casting processes for uranium carbides. The research program has been divided into five sections, including preparation and forming by powder-metallurgical techniques, development of improved casting techniques, study of physical and mechanical properties, measurement of diffusion rates in uranium monocarbide, and investigation of the important aspects of irradiation damage in uranium carbides.

TABLE FF-1. RESULTS OF TESTS OF PRESSURE-BONDED TYPE 304 STAINLESS STEEL BOND-TEST TENSILE SPECIMENS

		Pressure-Bonding Conditions					
Specimen	Type and Condition	Time, hr	Temperature, F	Pressure, psi	Approximate Yield Strength, lb	Ultimate Load, lb	Comments
1	Machined from Type 304 stainless steel stock	--	--	--	2000	4460	Used as a comparison for the pressure-bonded specimens
2	Butted rods; 115 to 135-rms surface	3	2100	10,000	1700	4100	Specimen necked down and broke away from the bond; complete grain growth
3	Butted rods; 110 to 130-rms surface	3	2100	10,000	1700	4100	Specimen necked down and broke away from the bond; complete grain growth
4	Butted rods; 120 to 150-rms surface	1-1/2	2100	10,000	1700	4160	Specimen necked down and broke away from the bond; complete grain growth
5	Machined from Type 304 stainless steel stock after pressure bonding	1-1/2	2100	10,000	1700	4100	Used as a comparison for the bond-test specimens
6	Butted rods with 0.005-in. disks in between; 30 to 40-rms surface on rods	3	2000	10,000	2000	4240	Specimen did not neck down all the way but parted between the disk and rod on one end; pieces of the disk were pulled away; complete grain growth across the disk interface and 70 per cent grain growth across the rod-to-disk interfaces
7	Butted rods with 0.005-in. disks in between; 50 to 60-rms surface on rods	3	1900	10,000	1620	3500	Specimen did not neck down all the way but parted between the disk and rod on one end; break along bond line with no metal being torn from either surface; 10 per cent grain growth between the disks
8	Butted rods; 50 to 60-rms surface	3	1900	10,000	1600	3750	Specimen did not neck down all the way but parted at the bond line, with metal torn from both surfaces

In a study of the effect of various impurities upon uranium monocarbide, it was reported last month that their effect upon mechanical properties was generally detrimental, as was an increase in carbon content from stoichiometric UC (4.8 w/o carbon) to 5 w/o carbon. Results obtained during the last month indicate that these same impurities (with the possible exception of oxygen and hydrogen), as well as excess carbon, have a detrimental effect upon the corrosion resistance of carbides in water. Oxygen and hydrogen produced effects upon the corrosion resistance of uranium monocarbide which might be described as beneficial, but it should be noted that it is quite possible that none of the oxygen or hydrogen added to these alloys remained in them. This possibility will be examined.

The density of uranium alloys containing up to 9.2 w/o carbon appears to be a linear function of atomic per cent carbon. The electrical resistivity of uranium alloys containing up to 9.2 w/o carbon appears to be a logarithmic function of atomic per cent carbon.

Attempts to prepare uranium carbide by solid-state reaction of uranium oxide and uranium metal powders with graphite or hydrocarbons have produced some indications of success. Analytical data are needed before the results can be evaluated.

Fabrication of UC by Powder Processes

S. J. Paprocki, D. L. Keller, and D. E. Lozier

Methods of producing dense UC pellets by powder metallurgy are being investigated. Evaluations will be made on the basis of proposed costs for large-quantity production as well as on the quality of the material fabricated. Procedures which are currently being investigated include reactions of U_3O_8 and graphite and uranium metal powder and graphite at temperatures high enough to insure both the completion of the reaction and the formation of dense pellets. In addition, sintering studies are being conducted on the UC compound using materials containing both stoichiometric and nonstoichiometric amounts of carbon. The UC is being produced by reaction of uranium metal with hydrocarbons such as methane or propane.

A pellet compacted at 30 tsi from a stoichiometric mixture of U_3O_8 and graphite powders was reacted at 1982 C. Upon heating considerable degassing occurred in the range 1400 to 1982 C. The pellet was held at temperature until a vacuum of 10 μ was obtained and then furnace cooled. Examination of the furnace after the experiment indicated that two of the insulating materials had reacted. Therefore, it is considered probable that most of the degassing at elevated temperatures was due to the reaction of the refractories. The density of the test specimen was 7.96 g per cm^3 compared to 2.87 g per cm^3 before reaction. The pellet was visibly porous and surface striations present in the cold-pressed compact could still be detected even though a shrinkage of 71 per cent in volume had occurred during reaction. The pellet is being analyzed for free and combined carbon content.

UC powder is being prepared for sintering studies by reaction of uranium metal powder produced from the hydride with methane gas at 649 to 816 C. At these temperatures the reaction is slow, but if carbon analysis is satisfactory the powder will be used for sintering. On the basis of thermodynamic calculations it is expected that more satisfactory results will be obtained with propane or higher alkanes such as octane. Other methods of preparation of UC powder are also being considered.

In addition, compacts of uranium and graphite powders are being prepared for reaction studies at various temperatures.

Physical Characteristics and Properties of Uranium Carbides

W. M. Phillips, E. L. Foster, and R. F. Dickerson

High uranium density, melting point, strength, and thermal conductivity make uranium carbide a potentially desirable fuel material. Further, it has been demonstrated that a convenient method of realizing these inherent qualities in the fabrication of UC fuel elements is by melting and casting.* The studies now in progress are concerned with maintaining or improving these inherent properties through a fuller understanding of the effects of such variables as impurities, carbon content, and heat treatment on density, resistivity, thermal conductivity, and corrosion resistance. The ultimate goal of the program is the development of reliable methods for the production of high-quality cast uranium carbide shapes.

Hydrogen, iron, tungsten, nitrogen, and oxygen were selected as materials which might normally be present in charge materials or available to contaminate UC during melting and casting. Charges containing these impurities at levels from 100 to 800 ppm were melted and cast to a right-cylindrical shape 3/8 in. in diameter by 1-3/4 in. long. A diamond grinding wheel was used to smooth the surfaces of the castings. Density, resistivity, and flexural-strength measurements were made on each specimen. Corrosion testing was done in 60 C water. The observations and data recorded are presented in Table FF-2.

Impurities were found to have no effect on the ease of casting into shapes except in the case of silicon additions, where sound castings were difficult to prepare. Iron and tungsten appear to have an adverse effect on corrosion resistance, while hydrogen and oxygen appear to be beneficial to a small degree. Melting in a hydrogen-containing atmosphere apparently produced a significant increase in corrosion resistance. Analyses of these alloys have not been obtained. The effects of hydrogen, nitrogen, oxygen, and silicon are being rechecked.

Carbon content in excess of stoichiometric (4.8 w/o) was found to have an adverse effect on both strength and corrosion resistance. Data taken on the specimens from the series of alloys shown in Table FF-3 having nominal carbon contents from 0.02 to 9.25 w/o carbon indicate a linear relationship between density and atomic per cent carbon. The change in resistivity with carbon content appears to be a logarithmic function of atomic per cent carbon.

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*Secrest, A. C., Jr., Foster, E. L., and Dickerson, R. F., "Preparation and Properties of UC Castings", BMI-1309 (January 2, 1959).

TABLE FF-2. EFFECT OF CARBON CONTENT AND IMPURITIES ON CORROSION RESISTANCE OF UC IN 60 C WATER

Sepcimen	Nominal Carbon Content, w/o	Impurity Content, ppm	Corrosion Rate at 2 Hr, $mg/(cm^2)(hr)$
1	4.8	100 Fe	(a)
2	5.0	100 Fe	703
3	4.8	300 Fe	(a)
4	5.0	300 Fe	(a)
5	4.8	800 Fe	(a)
6	5.0	800 Fe	(a)
7	4.8	100 Si	(a)
8	5.0	100 Si	(a)
9	4.8	300 Si	(a)
10	5.0	300 Si	616
11	4.8	800 Si	Not tested
12	5.0	800 Si	(a)
13	4.8	100 W	(a)
14	5.0	100 W	755
15	4.8	300 W	(a)
16	5.0	300 W	(a)
17	4.8	800 W	1520
18	5.0	800 W	(a)
19	4.8	100 N	(a)
20	5.0	100 N	631
21	4.8	200 N	514
22	5.0	200 N	501
23	4.8	500 N	1664
24	5.0	500 N	(a)
25	4.8	100 O	(a)
26	5.0	100 O	1581
27	4.8	100 O	(a)
28	5.0	200 O	865
29	4.8	500 O	765
30	5.0	500 O	851
31	4.8	H_2	246
32	4.8	None	638
33	4.8	None	Not tested
34	5.0	None	1050
35	5.0	None	Not tested

(a) Disintegrated, or broke into small pieces.

TABLE FF-3. EFFECTS OF CARBON CONTENT ON DENSITY AND ELECTRICAL RESISTIVITY OF UC

Specimen	Nominal Carbon Content, w/o	Density, g per cm^3	Resistivity, ohm-cm 10^{-6}
36	0.02	18.54	40
37	2.4	15.43	46
38	4.5	13.65	45
32	4.8	13.67	41
34	5.0	13.48	35
39	5.0	13.38	42
40	5.9	13.05	45
41	6.75	12.53	51
42	7.25	12.41	57
43	8.00	11.93	72
44	8.75	11.77	83
45	9.25	10.57	129

Metallographic examination of the above materials was made, and the microstructural effects of impurities upon uranium monocarbide in the as-cast condition may be summarized as follows:

(1) Excess carbon increases the tendency toward the formation of cracks within the specimen, particularly when iron is added as an impurity.

(2) Most ternary additions (iron, oxygen, silicon, and tungsten) produce second phases which are concentrated at the grain boundaries.

(3) Grain size and nature of the columnar structure vary widely but not consistently with impurity additions.

Since apparently improved water-corrosion resistance was possessed by materials containing small additions of oxygen and hydrogen, specimens containing increased quantities of these additions plus silicon and nitrogen are being prepared. The preparation of specimens for compatibility testing and corrosion testing in NaK has been initiated. Compatibility of UC with aluminum, beryllium, copper, magnesium, niobium, zirconium, Inconel, mild steel, and Type 304 stainless steel, will be noted. The effects of high-temperature heat treatment on corrosion resistance, strength, density, resistivity, and microstructure are also being investigated.

Uranium Monocarbide Diffusion Studies

W. Chubb and F. A. Rough

Diffusion-rate measurements are being made on uranium monocarbide in an effort to obtain a basic understanding of the behavior of this material at very high temperatures such as might be generated in the material operating as a fuel in a nuclear reactor. Measurements of the interdiffusion rates of uranium and carbon in the uranium monocarbide-uranium dicarbide portion of the system are in progress and tentative diffusion coefficients were reported last month for temperatures of 1600 and 1980 C.

During the last month four additional melts of carbon-saturated uranium were held for various lengths of time at 1800 and 1900 C in thick-walled graphite crucibles. A visual examination of the carbide layers formed between the uranium and the graphite showed that the thickness of the layers is very nearly that predicted by extrapolation of the diffusion data reported last month. Diffusion coefficients for 1800 and 1900 C will be calculated when these layers have been measured by metallographic techniques. Additional melts will be made at 1600 C, and, if the data appear reasonable, no further effort will be devoted to this technique of measuring diffusion coefficients. The technique provides useful information, but is inexact to the extent that it neglects the fact that uranium monocarbide and uranium dicarbide are not completely soluble in one another.

Materials have been ordered and techniques are being developed for determining the rate of self-diffusion of uranium in uranium monocarbide. It is expected that samples of enriched uranium foil sandwiched between samples of depleted uranium carbide will be diffusion annealed, sectioned, activated, and analyzed by gamma counting. Techniques for doing this are being examined. It has been found that suitable 1/4-in. lengths of uranium carbide can be cut from cast rods using a diamond cutoff wheel. Some improvement in technique is still required to obtain perfectly clean cuts which need no grinding after cutting, but this appears feasible. Graphite pressure-bonding jigs have been machined, and their utility will be investigated in the near future. Sampling techniques have been considered, and a metallographic technique will be investigated as being apparently the most suitable for this type of material.

Determination of the rate of self-diffusion of carbon in uranium monocarbide is complicated by the lack of a suitable technique for obtaining small samples of carbide (or nitride) for analysis for carbon or carbon-14. In the latter case, a serious health hazard is involved. The possibility of using an autoradiographic technique will be investigated.

Irradiation Effects in UC

A. E. Austin and C. M. Schwartz

The study of the effects of neutron irradiation and fission in uranium carbide has been started. It is planned to use X-ray diffraction measurements to follow crystal strain and fragmentation. Methods of specimen preparation to yield satisfactory data are being investigated.

An examination by electron microscopy of presently available irradiated arc-cast material has been started. Because of reactions of the carbide with water the conventional techniques used for replication and electron microscopy of metals were not applicable. Suitable procedures have been developed for specimen preparation and replication of arc-cast uranium carbide.

FF-9 and FF-10

Low angle X-ray scattering can detect very small voids or bubbles. It does not appear possible to examine pure uranium carbide because of its strong absorption of usable X-rays. However, an alloy of zirconium and uranium carbides may be usable. Therefore, attempts are being made to prepare and examine a 90 w/o ZrC-10 w/o UC alloy, in order to test the possibility of studying void formation in this material from irradiation.

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 June, the analysis was completed of the data obtained from the stress-cycling fatigue studies of Inconel. The following conditions were used for the experimental program:

Temperature	1300 and 1500 F
Cyclic frequency	6 and 60 cpm
Lifetime range	10^3 to 10^5 cycles

Stress-strain hysteresis curves were drawn for all tests from which stress, total strain, and plastic strain could be measured.

Several interesting phenomena were observed. It was found that fatigue strength, whether measured in stress, total strain, or plastic strain, was a function of both temperature and frequency. It was also found that the stress-strain hysteresis curve varies throughout the lifetime of a specimen, never reaching what might be considered an equilibrium hysteresis curve. In addition, it was observed that strain was not a continuous function of stress. Rather, under certain predictable conditions, strain in the plastic region occurred in a number of sudden steplike increments of alternating elastic and plastic strain.

This concludes the work on this program and a topical report is being prepared.

Fatigue Studies of INOR-8

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

Fatigue properties of INOR-8 for rotating-beam fatigue cycling were studied at temperatures of 1100, 1300, and 1500 F, and for cyclic frequencies of 100, 600, and 3000 cpm. The fatigue strength of INOR-8 decreased with increasing temperature. Frequency was found to affect fatigue strength significantly at 1500 F. The fatigue strength decreased with decreasing frequency. However, there was no effect of frequency at 1100 F and only a slight frequency effect at 1300 F. In addition, it was found that the fine-grained material studied showed greater fatigue strength than the coarse-grained material.

This concludes the work on this program and a topical report is being prepared.

GG. VOID-DISTRIBUTION AND HEAT-TRANSFER STUDIES

R. A. Egen, D. A. Dingee, and J. W. Chastain

A research program is being undertaken for the Euratom-U. S. Joint Research and Development Board to provide information on void-content and heat-transfer relationships in forced-convection subcooled-boiling water. The first phases of the program will involve primarily measurements of void fraction and distribution in light water for experimental conditions chosen to encompass a range of significant parameters. Pressures from 1 to 90 atm and mass flow rates from 0.5×10^5 to 1×10^6 lb/(hr)(ft^2) will be investigated. Heat fluxes will range from 0.5×10^5 to about 2×10^6 Btu/(hr)(ft^2), and the amount of inlet subcooling from 0 to 60 F. Symmetrical test-section shapes will be selected for the studies to facilitate void-distribution measurements.

A high-pressure loop is available and is being reassembled at Battelle for this study. The loop provides a positive-displacement pump (to maintain constant mass flow rates independent of the varying two-phase pressure losses through the test specimens) and low total fluid volumes. The test section is heated electrically by continuously variable power transformers. A preheating section is located prior to the test section and can be separately heated so that any axial region of a long heat-transfer passage can be simulated. Flow rates are measured by an enthalpy balance across the preheater section and by an orifice plate in the cold part of the loop.

During the past month, the program was reviewed to specify test conditions, experimental techniques, and details of loop construction.

The test-section dimensions to be used in these experiments are governed to some extent by the method of void detection. Subcooled boiling occurs at the heated surfaces and bubble formation may be confined largely to this region. Accordingly, test-section shapes and void-detection methods which will provide useful engineering data in subcooled boiling are being investigated. To avoid errors in detecting this preferential void distribution, symmetrical test-section shapes, such as square or circular cross sections, or parallel plates, are being considered. In this latter case, the void content perpendicular to the plates would be measured.

Gamma-ray detection methods which have been used in previous studies at Battelle may not be suited to the present studies wherein low total voids and small test-section dimensions are involved. Improved measurements may be possible using either a neutron, beta-ray, or X-ray source.

The experimental heat-transfer loop used in previous studies is being evaluated as it applies to the present research. It appears that higher flow rates will be desirable and modifications to the present pump are being investigated. Heat requirements for the program are being estimated and appear to be within available capacity.

During the coming month, bench tests of void-detection methods are planned. The heat-transfer loop will be designed and preliminary construction is anticipated.

H. PHYSICAL RESEARCH

F. A. Rough

Two physical-research studies, one of the kinetics of reaction of niobium and nitrogen and the other of the constitution of high-purity uranium-niobium alloys, are reported this month.

In the report of the niobium-nitrogen-reaction studies, kinetics of reaction from 675 to 1600 C and diffusion coefficients and terminal solubilities for nitrogen in niobium from 800 to 1600 C are summarized. Future research will be directed toward understanding the kinetics of reaction at temperatures below 700 C.

The study of the constitution of high-purity uranium-niobium alloys is concluded. Results obtained from diffusion couples are described, giving the limits of the gamma$_1$-plus-gamma$_2$ immiscibility gap and a series of diffusion penetration curves for heat-treated couples.

Niobium-Gas Reactions

W. M. Albrecht and W. D. Goode

Fundamental studies are being made of the reaction of niobium with nitrogen, and the diffusion and terminal solubilities of nitrogen in the metal.

The reaction of nitrogen with niobium at 675 to 1600 C followed a parabolic rate law. A change in the reaction mechanism was indicated between 975 to 1100 C. The activation energies are 24.4 ± 1.0 kcal per mole at 675 to 975 C and 50.0 ± 1.8 kcal per mole at 1100 to 1600 C. X-ray studies indicate that both NbN_2 and NbN were produced below 1000 C while only NbN phases were formed at 1200 C and above. This difference in surface product accounts for the change in the reaction mechanism between 975 and 1100 C.

The diffusion coefficients and terminal solubilities for nitrogen in niobium were determined in the range 800 to 1600 C. The diffusion coefficients were found to follow the relationship,

$$D\ (\mathrm{cm}^2/\mathrm{sec}) = 0.061 \exp\left[-38{,}800/RT\right],$$

where the activation energy is 38.8 ± 0.7 kcal per g-atom. The ΔS for diffusion was calculated to be 6.2 cal/(g-atom)(C), which is consistent with the theory for interstitial diffusion.

The terminal solubilities for nitrogen in niobium were found to have the relationship,

$$C\ (\mathrm{a/o\ N}) = 180 \exp\left[-13{,}700 \pm 200/RT\right].$$

These solubilities were checked by metallographic examination of heat-treated niobium samples containing known concentrations of nitrogen.

A topical report describing the experimental work is being prepared.

Studies on the kinetics of the niobium-nitrogen reaction below 675 C are continuing.

Constitution of Uranium-Niobium Alloys

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

A uranium-niobium constitution study utilizing diffusion couples to establish the composition limits of the gamma-phase immiscibility gap in the system is in progress. High-purity uranium and niobium are being used in these studies. The results are intended to serve as a basis for future studies of the effects of impurities on uranium-niobium-alloy constitution.

Uranium-niobium couples diffusion annealed at 925 C for a total of 500 hr, at 900 C for 168 and 500 hr, and at 700 C for 2500 hr were analyzed by electron-probe microanalyses. Concentration-penetration curves for these couples, and for a couple diffusion annealed at 1000 C for 50 hr which was previously analyzed, are shown in Figures H-1 through H-4. Figure H-1 depicts the data for the couple heated at 1000 C. The curve shown is fairly typical of that expected for a single-phase alloy, except for a slight break in smoothness of the slope at 35 a/o uranium. The curve indicates that 1000 C lies above the two-phase immiscibility gap. Diffusion coefficients have been calculated and reported for this temperature. The concentration-penetration curve for the couple diffusion annealed at 925 C shown in Figure H-2 indicates a two-phase region between 40 and 60 a/o uranium, as indicated by the vertical break between these compositions. The curves for the couples heated at 900 and 700 C also show these breaks, occurring between 35 and 65 a/o uranium at 900 C and between 24 and 80 a/o uranium at 700 C. In addition, the 700 C curve contains a second two-phase boundary occurring between 91 and and about 98.5 a/o uranium, corresponding to the beta-plus-gamma_1 region in the uranium-niobium system.

Figures H-2 through H-4 show anomalous regions of very low slope immediately adjacent to the two-phase boundary below 40 a/o uranium in each curve. These regions are lengthened at lower temperatures and longer annealing times. It is felt that they are caused by clustering of uranium and niobium atoms in the gamma_2 phase, with clustering becoming more pronounced as the gamma_1-plus-gamma_2 loop broadens at the lower temperatures. Because of this phenomenon, meaningful diffusion coefficients cannot be calculated. Measurements of activities across the system are required for interpretation of the concentration-penetration curves obtained and calculation of diffusion coefficients; however, no further experimentation is planned at this time.

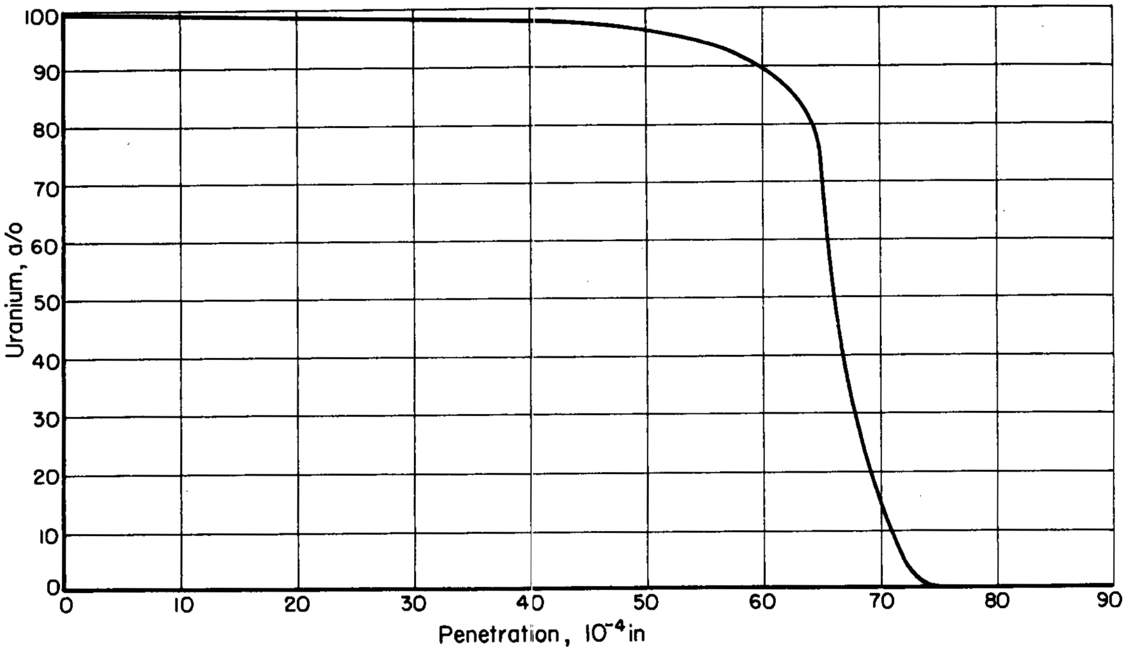

FIGURE H-1. CONCENTRATION-PENETRATION CURVE FOR URANIUM-NIOBIUM COUPLE DIFFUSION ANNEALED AT 1000 C FOR 50 HR

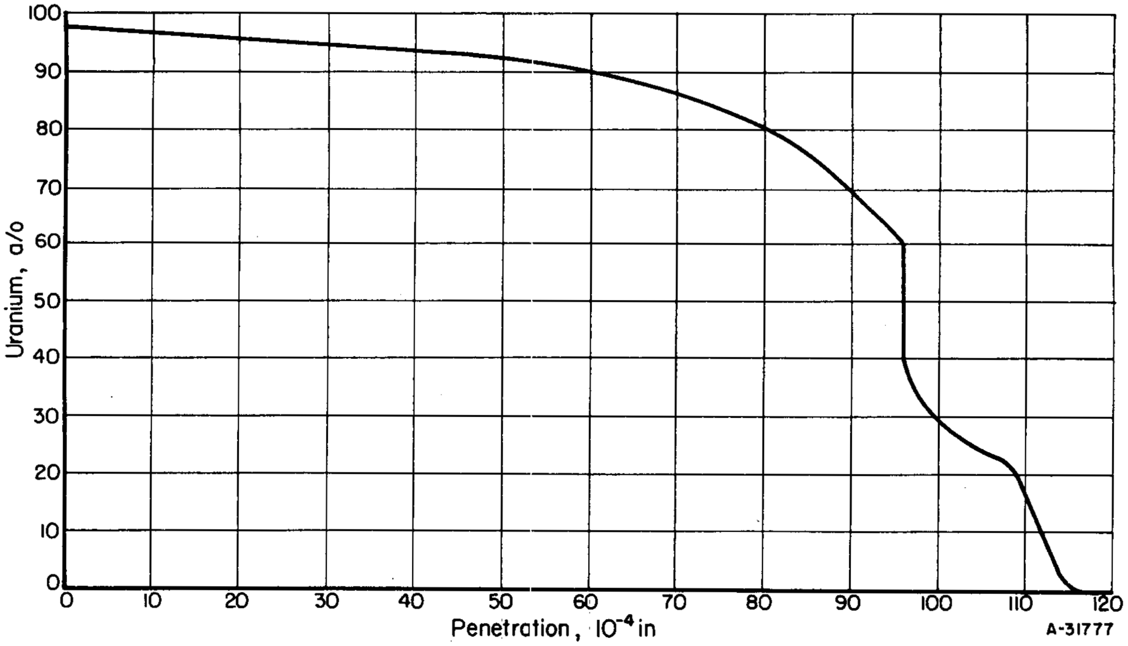

FIGURE H-2. CONCENTRATION-PENETRATION CURVE FOR URANIUM-NIOBIUM COUPLE DIFFUSION ANNEALED AT 925 C FOR 500 HR

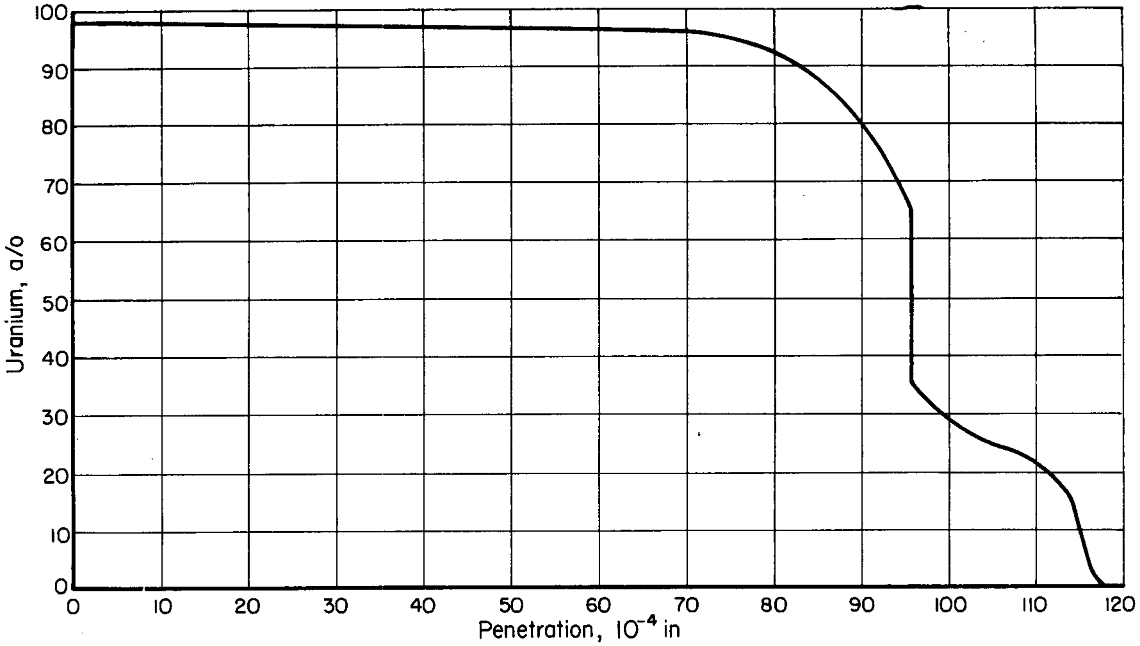

FIGURE H-3. CONCENTRATION-PENETRATION CURVE FOR URANIUM-NIOBIUM COUPLE DIFFUSION ANNEALED AT 900 C FOR 500 HR

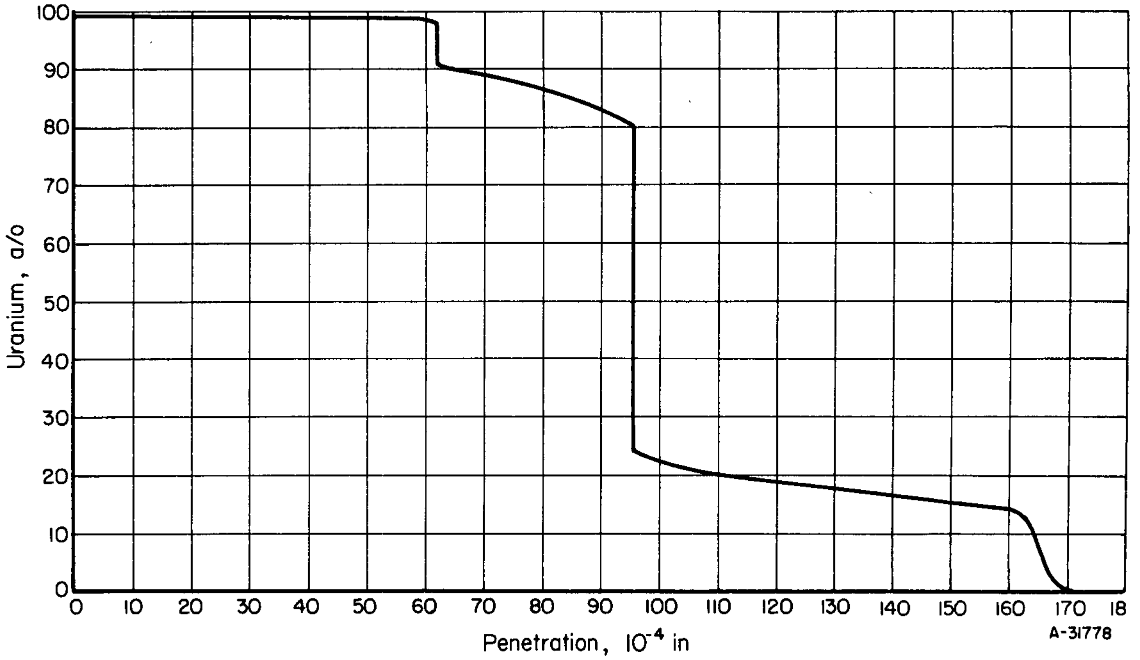

FIGURE H-4. CONCENTRATION-PENETRATION CURVE FOR URANIUM-NIOBIUM COUPLE DIFFUSION ANNEALED AT 700 C FOR 2500 HR

I. SOLID HOMOGENEOUS FUELED REACTORS

D. N. Sunderman and W. H. Goldthwaite

Research in support of the development and evaluation of spherical fueled-graphite fuel elements for the Pebble-Bed Reactor program is in progress at Battelle. Candidate fuel elements, 1-1/2 in. in diameter, prepared by several vendors and varying in composition and method of manufacture, are being evaluated by laboratory tests and irradiations designed to obtain information on their structural integrity and fission-product-retention characteristics.

LABORATORY EVALUATIONS OF FUELED-GRAPHITE SPHERES

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

Selected thermal and mechanical properties of spherical fueled-graphite elements are being measured as a preirradiation evaluation.

During June, apparatus for measuring the permeability up to 2000 F of coatings on graphite elements was designed and construction was started. The permeability test will consist of heating the spheres rapidly by induction in a known gaseous environment and measuring with a mass spectrograph any gases which are forced out of the coated spheres by thermal expansion. Also, the tendency of the coatings to weld together under load at high temperature is being investigated. Initial tests will be made on elements with silicon-silicon carbide coatings under a 50-lb load at 2000 F in an argon atmosphere.

Microstructural examinations of coated and uncoated fueled spheres are being made by metallographic techniques. Particular attention will be given to the particle size and uniformity of dispersion of the fuel and to the coating-graphite interface.

FISSION-PRODUCT RELEASE FROM FUELED-GRAPHITE SPHERES

D. N. Sunderman

The release of fission products from fueled-graphite spheres of various compositions is being studied both by postirradiation heat-treatment experiments (dynamic activation) and by in-pile sweep-gas experiments (dynamic irradiation). The dynamic-activation technique serves to screen materials of potential value as a reactor fuel. The most promising materials are advanced to the dynamic-irradiation experiments.

Dynamic-Activation Experiments

R. Lieberman, H. S. Rosenberg, D. N. Sunderman, and M. Pobereskin

During June, preliminary experiments were started in which a test UO_2-graphite sphere was heated by induction heating. A spark-gap-converter high-frequency generator with a 6-kw maximum input was used. Temperature measurements were made by means of a thermocouple imbedded in the UO_2-graphite sphere. Helium flowed through the system during each heating cycle. In Table I-1, the rate of heating using the sphere as a conductor is compared with the rates of systems using an auxiliary carbon conductor or susceptor and a molybdenum susceptor.

TABLE I-1. TIME-TEMPERATURE RELATIONSHIP USING INDUCTION HEATING ON A UO_2-GRAPHITE SPHERE

Time From Startup, min	Temperature, F		
	Sphere Alone	Carbon Susceptor	Molybdenum Susceptor
0.5	670	550	170
1.0	1020	970	300
1.5	1260	--	550
2.0	1490	1640	740
2.5	1790	1860	1020
3.0	2000	2010	1190
3.5	2080	2110	1420
4.0	2130	2180	1570

Future work will include the statistical evaluation of the postirradiation technique using admixture balls, determination of the leak rates from several coated and reimpregnated specimens, evaluation of leaky coatings, and measurement of gas release from ThO_2/UO_2 elements.

Dynamic-Irradiation Experiments

D. B. Hamilton, G. E. Raines, and W. H. Goldthwaite

During June the gas-holdup tank used in the SP-2 in-pile experiment for admixture specimens was removed to the radioisotope laboratory for identification of the nonvolatile radioisotopes present on the inside surface.

The tank was cut open at the end which was nearest to the reactor. A portion of the inside surface was rinsed with nitric acid-containing barium, cerium, lanthanum, and strontium carriers. A qualitative chemical separation was carried out on the wash solution and the radioisotopes which were identified were barium-140, cerium-141, lanthanum-140, and strontium-89. These isotopes are daughters of rare-gas isotopes. The search for zirconium-95 and ruthenium-103, which do not have gaseous precursors, was unsuccessful. Table I-2 presents the results of the analysis.

TABLE I-2. RESULTS OF RADIOCHEMICAL ANALYSIS OF RINSINGS FROM SP-2 HOLDUP TANK FOR ADMIXTURE SPECIMENS

Isotope	Half-Life, days	Quantity Found, disintegrations per sec
Strontium-89	54	5.0×10^6
Barium-140	12.8	1.4×10^4
(lanthanum-140)		
Cerium-141	32	1.5×10^2

If it is assumed that rinsing removed the same fraction of each isotope as existed on the wall of the tank, and corrections are made for the 85-day period of decay since the gas was passed through the tank, an apparent delay time between formation of the gaseous precursor and deposition of the daughter in the tank can be calculated. A comparison of the activity of barium-140 and cerium-141 with that of strontium-89 indicates a delay between formation of the gas and deposition of the daughter of about 20 sec and 80 sec, respectively. This analysis of the data is not rigorous but is an indication of the holdup of short-lived gases.

Operation of SP-3, a sweep capsule containing pairs of FA-6 (fueled with UC and coated with silicon-silicon carbide) and FA-8 (fueled with UC_2 and coated with silicon carbide) elements, in May indicated that the fission-product release had risen to a level too high to be handled in the gas-sampling train. During the June 2 reactor shutdown, operation at reduced reactor power (100 kw) and reduced element temperature (170 F) was attempted a second time. This time, however, because it was not possible to remove the capsule from the core before the shutdown to provide several days for decay, the xenon-133 activity in the gas sample was too high to permit quantitative measurements on xenon-135 and krypton-85m.

An analysis of the operation of the gas train has shown that several modifications can and should be made to make it possible to operate continuously at fractional release rates which are too high for the present system but still in the range of interest in evaluating variations in fuel-element design and fabrication. These modifications have been given first priority and are well under way. The modified gas train is scheduled for checkout operation with Capsule SP-3 by the middle of July and for continuous operation with SP-5, the next sweep capsule, by August 12 at the latest. The modifications will include: (1) the addition of a doubly contained bypass line to carry the major fraction of the carrier gas to shielded charcoal traps, (2) the redesign of the gas-sampling line to minimize its volume and, hence, the total activity outside the doubly contained and shielded portion of the system, and (3) the construction of apparatus to provide a continuous and automatic supply of liquid nitrogen to the charcoal safety traps. With these modifications, it should be possible to operate continuously with a fractional release (R/B)* of 10^{-2} or higher. The incorporation of a second bypass which will include a packed column for trapping nonvolatile daughters of short-life fission gases is being considered. This would provide a more direct means of determining the release of the products which present the most troublesome contamination problem.

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*R/B is the release rate in atoms per second divided by the production rate in atoms per second of the same fission-product species.

Several capsules are in various stages of preparation for irradiation. The fabrication of Capsule SP-5 has been initiated. This sweep-gas capsule will be similar to Capsule SP-3, will contain pairs of FA-6 (an improved version) and FA-11 (UO_2 fuel, pyrolitic carbon coating) elements, and will utilize the modified gas system. Reactor insertion will take place either July 29 or August 12.

POSTIRRADIATION EXAMINATIONS OF FUELED-GRAPHITE SPHERES

R. J. Burian and J. E. Gates

The irradiation of Capsule SP-3 is continuing. Capsule SP-3 contains four 1.5-in.-diameter spherical specimens, two FA-8 fueled with UC_2 and clad with silicon carbide and two FA-6 fueled with UC and clad with silicon-silicon carbide. Discharge from the BRR is tentatively scheduled for July 27, 1959. The postirradiation examination will begin shortly after discharge.

The examinations planned for the specimens irradiated in Capsule SP-3 include both nondestructive and destructive tests. As part of the nondestructive portion of the examination, the specimens will be visually examined, pictures of representative and unique areas will be taken, dry weight and dimensional measurements will be made, and the specimens will be subjected to a hot oil bath (approximately 400 F) to determine if any failures occurred in the ceramic cladding. The destructive tests will include drop-weight impact tests followed by immersion in the hot oil bath to determine when a failure occurs in the cladding, and compressive tests while the specimen is immersed in a hot oil bath. Several neutron dosimeters irradiated in the capsule will also be analyzed. The equipment for performing these examinations is being prepared.

The design of the static capsule experiment, SP-4, is being completed. This capsule is to yield information on the conversion of UO_2 to UC_2 and the possibility of shrinkage of the matrix from the ceramic coating at temperatures in the neighborhood of 2000 F. It will be irradiated in Test Cell 12 in the BRR, which has a higher quoted unperturbed flux than Test Cell 45 in which Capsule SP-3 is being irradiated. The increased heat generation due to the higher flux brings up the possibility of boiling of the reactor coolant water at the surface of the capsule. A hydraulic simulation has been prepared and measurements are to be taken to provide a more accurate determination of the coolant flow past the capsule. It is expected that the flow will be great enough to permit use of the present concept of a capsule containing six specimens; if not, the capsule design will have to be modified to space the specimens farther apart and provide sufficient heat-dissipation area to prevent boiling of the coolant. In either case, the capsule is tentatively scheduled for insertion by September 1, 1959.

Preliminary calculations on a high-flux high-burnup capsule (SPH-1) have been made and indicate that a 1600 F specimen-surface temperature is possible using the present basic capsule concept with a heat generation of 6 kw per ball in an unperturbed flux of 1 to 1.5 x 10^{14} nv. Negotiations are being carried out with several test reactors. Either the WTR or MTR would seem to be feasible irradiation sites.

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. N. Stiegelmeyer, W. C. Baytos, and F. W. Fink

The evaluation of materials of construction for several processes for the recovery of spent reactor fuel elements is being conducted as a program of assistance to the Chemical Technology Division of ORNL. Corrosion studies associated with the various head-end aqueous processes are being terminated and will be summarized in a topical report.

The dissolution of 25 fuel pins in a Ni-o-nel dissolver by the Sulfex-Thorex process has demonstrated the necessity of solution annealing all weldments in order that severe intergranular attack be prevented. Even with solution annealing, some intergranular attack occurs and the corrosion rates are sufficiently high to indicate that further study should be conducted before a Ni-o-nel dissolver is constructed. A Titanium 75A dissolver used for the dissolution of 25 fuel pins by the Darex-Thorex process underwent negligible attack except in areas around weldments in the vapor phase. Since not all of the vapor-phase weldments show this attack, weld contamination is suspected. Further study appears to be necessary with this system, also.

The Sulfex-Thorex Process

The proposed Sulfex-Thorex Process combines a decladding step in which stainless cladding is removed by dissolution with boiling 6 M H_2SO_4 with a dissolution step using a boiling solution which is 13 M HNO_3, 0.04 M F^- for the thoria core.

Bench Dissolution of Fuel Pins by the Sulfex-Thorex Process

Twenty-five Consolidated Edison fuel pins were dissolved, one at a time, using the Sulfex-Thorex process in a bench-scale dissolver constructed from Ni-o-nel. The loss in weight of the vessel during the 285.5-hr exposure from the dissolutions was equivalent to about 3 mils per month of corrosion. This rate was higher during dissolution of the latter half of the pins, indicating that corrosion was accelerating, probably as a result of localized attack. Coupon-type specimens in the dissolver lost weight equivalent to 1.5 to 2 mils per month.

A metallographic study was performed on sections cut from the vessel. Two annealed weldments were included in the walls of the dissolver. Around both of these weldments, located in the heat-affected areas at some distance from the weld beads themselves, were bands about 1/8 to 1/4 in. wide in which deep intergranular attack had occurred. The annealed weldments did not show such severe attack. Intergranular attack occurred to some extent on all exposed surfaces. No indication of crevice attack was found anywhere.

The metallographic study indicates that Ni-o-nel must be solution annealed to prevent the occurrence of severe intergranular attack around weldments upon exposure to Sulfex-Thorex solutions. However, the unwelded metal, even after solution anneal, shows some intergranular attack. The intergranular attack occurs both on the submerged and vapor-phase portions. It appears that further investigations will be necessary before the use of Ni-o-nel as a construction material for the Sulfex-Thorex dissolver could be recommended.

The Darex-Thorex Process

The proposed Darex-Thorex Process, an alternate to the Sulfex-Thorex route, is under consideration. In this process, decladding would be accomplished in boiling dilute aqua regia and the core would be dissolved by Thorex solution. It is thought that such a process could be carried out in titanium Darex equipment.

Scouting Experiments With Titanium

The exposure of titanium specimens to chloride-contaminated Initial Thorex solutions was continued for a total of 1000 hr. The corrosion rates remained low (0.3 mil per month, or less) during this exposure. However, a considerable amount of pitting occurred on the specimens. The pits were scattered over the surfaces of the coupons but were most prevalent on cut edges, around the sharp corners raised by imprinting designation numbers, and along weldments. Pitting was more severe on vapor-phase specimens. The average pit was about 20 mils in diameter and about 3 to 5 mils deep. The bottoms of most of the pits were bright gold in color. Sectioning showed this coloration to be due to some sort of a deposit about 0.5 mil thick. The submerged specimens were dull gray-black in color and showed only the isolated pits mentioned. The vapor-phase specimens were tarnished with an iridescent oxide film and their surfaces were covered with numerous small pits in addition to the isolated larger ones. In many cases, the entire surface was roughened by a fine-grained etch.

The solutions were changed weekly, but this was not often enough to insure retaining the desired 100 ppm Cl^-. Analyses of solutions after exposure showed that only 5 to 10 of the original 100 ppm Cl^- remained. Conceivably, the pitting might have been more severe if the chloride concentration had been maintained. Aluminum chloride has been reported to cause pitting of titanium. Further investigations should be conducted in which the chloride concentration is maintained but aluminum additions are omitted. The problem of pitting should be thoroughly resolved before titanium is employed for a Darex-Thorex dissolver.

Bench Dissolution of Fuel Pins by the Darex-Thorex Process

A metallographic study has been completed on the Titanium 75A dissolver in which 25 Consolidated Edison fuel pins were dissolved by the Darex-Thorex process. Negligible attack was found on the side walls of this vessel. No evidence of crevice attack was found either. The only significant corrosion occurred on the top horizontal weldments between the walls and the top flange of the vessel and on the weldment between the flange and condenser tube. A very rough etch took place on these weld beads and their surrounding heat-affected areas.

These weldments were exposed to the vapor. However, vertical weldments in the corners of the vessel also received vapor-phase exposure, and no indication of etching is present on these welds. It may be that the conditions for condensation within the dissolver were such that fluorides, or possibly chlorides, concentrated along the weldments which are etched. However, this attack is more severe than any developed during a similar exposure time in coupon studies with chloride and fluoride contamination. Another possible explanation is that these weldments were contaminated even though precautions were taken during the fabrication of the vessel to prevent this. There is no opportunity to pursue the question further during the present investigation, but it is strongly recommended that the problems associated with weldment corrosion in the vapor above Darex-Thorex solutions either with or without chloride contamination be studied before any full-scale titanium dissolver is constructed.

The Fluoride-Volatility Process

A metallographic study of the corrosion which occurred on the INOR-8 sparge tube and thermocouple well of the laboratory hydrofluorinator at ORNL was completed. Several hydrofluorination runs were made in this unit using a 42 mole per cent NaF-58 mole per cent LiF salt. The only significant corrosion found on these tubes occurred on the sparge tube in the region of the vapor-liquid interface. Since the equipment was run for varying time intervals at temperatures from 500 to 700 C, it is difficult to ascribe a definite corrosion rate to the metal loss that was measured. If, however, most of the attack occurred during the exposure at 700 C, as previous experience with similar systems indicates, the rate may have been as high as 16 mils per month. Calculated over the entire exposure period the rate was 3 to 4.5 mils per month.

No indication of intergranular attack was observed on either tube. A thin scale built up on the tubes and increased the nominal thickness over the original thickness in several areas. This scale varied considerably in thickness and at one area was as much as 6 mils thick. It was metallic appearing and a qualitative spectrographic analysis indicated that it was composed mainly of the components of both INOR-8 and Zircaloy-2. The finding of zirconium and tin indicates that at least part of the scale originated from the molten salt and that the scale is not entirely composed of corrosion products.

A metallographic study of the corrosion which occurred on the Mark II fluorinator and its auxiliary equipment during its pilot operation at ORNL is under way. Following this, a similar study will be made of the Unit Operations INOR-8 dissolver.

J-4

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

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

Work was continued during June on assembly and testing of the glass dissolution equipment to be used for dissolution tests of irradiated fuel pins. Need for further modifications was revealed by this work; these are now completed, and "cold" dissolution tests will be started in July.

If the initial lot of irradiated Consolidated Edison pins from ORNL is received as scheduled, it is expected that hot-cell tests of Sulfex dissolution of these pins can be begun during July.

K. DEVELOPMENTS FOR SRE, OMRE, AND OMR

EVALUATION OF URANIUM MONOCARBIDE AS A REACTOR FUEL

F. A. Rough

Routine details of irradiation of three capsules of uranium carbide specimens in the MTR are reported. Data for three capsules in which the irradiation and examination have been completed have been presented in previous reports. One capsule of uranium carbide specimens, making a total of seven capsules, remains to be inserted in the MTR for irradiation this fall.

Enriched metal has been received for the preparation of a series of carbide specimens to be jacketed by Atomics International for irradiation in the SRE.

Irradiation of Uranium Monocarbide

D. Stahl and W. H. Goldthwaite

The irradiation of Capsules BMI-23-3, BMI-23-4, and BMI-23-5, containing uranium-carbon specimens, is continuing in the A28NE, A27SE, and A30NE positions, respectively, of the MTR.

Capsules BMI-23-3 and BMI-23-4 contain uranium-5 w/o carbon specimens and are being irradiated to 10,000 and 15,000 MWD/T, respectively. Capsule BMI-23-5 contains uranium-4.6 w/o carbon specimens, and is scheduled for a 5000-MWD/T irradiation.

Core temperatures of the top specimens in Capsules BMI-23-4 and BMI-23-5, as indicated by thermocouples located in drilled holes in the top specimens, were fairly constant at 1140 and 1360 F, respectively, for Cycle 122 and the completed portion of Cycle 123. The core thermocouple in BMI-23-3 failed at the end of Cycle 121, after about six cycles. The core temperature for Cycles 122 and 123 for this capsule was estimated to be about 1300 F from the readings of other thermocouples in the capsule.

Summarized maximum, minimum, and mean core and surface temperatures for top and bottom specimens for Capsules BMI-23-3 and BMI-23-4 during Cycles 118, 119, and 120 are given in Table K-1.

TABLE K-1. MAXIMUM, MINIMUM, AND MEAN CORE AND SURFACE TEMPERATURES OF SPECIMENS IN CAPSULES BMI-23-3 AND BMI-23-4 DURING MTR CYCLES 118, 119, AND 120

		Core Temperature, F						Surface Temperature, F					
	MTR	Top Specimen			Bottom Specimen			Top Specimen			Bottom Specimen		
Capsule	Cycle	Max	Min	Mean	Max	Min	Mean	Max	Min	Mean	Max	Min	Mean
BMI-23-3	118	1350	1120	1220	1090	920	1010	840	720	790	640	570	620
	119	1430	1160	1270	1210	1020	1100	870	760	820	660	610	640
	120	1300	610	1170	1120	570	1030	820	450	760	660	410	610
BMI-23-4	118	1030	940	1000	860	770	820	630	570	610	460	440	450
	119	1170	1100	1160	990	910	940	710	660	690	540	500	520
	120	1200	600	1120	980	450	900	710	390	670	530	300	490

Preparation of UC Pins for Irradiation in the SRE

W. M. Phillips, W. J. Hildebrand, E. L. Foster,
and R. F. Dickerson

Atomics International is interested in the radiation behavior of an appreciable quantity of cast UC shapes under SRE operating conditions. Battelle is participating in the program to evaluate this fuel material to the extent of preparing the as-cast shapes and determining their preirradiation qualities.

Specific techniques for the preparation of UC castings approximately 0.6 in. in diameter have been established. The necessary uranium enriched to 8.0 per cent in uranium-235 was received recently and is being fabricated to a size appropriate for alloying purposes. Metallographic and analytical evaluations to establish its quality are also in progress. If the quality is satisfactory, alloying of the materials will begin.

L. TANTALUM AND TANTALUM-ALLOY STUDIES

J. H. Stang

The two investigations reported in this section comprise current research for Los Alamos Scientific Laboratory (LAMPRE program). The study of tantalum binary alloys has the objective of developing a material which is superior to unalloyed tantalum with respect to resistance to corrosion attack by molten plutonium-alloy fuels of interest. During June, a variety of alloy buttons prepared by arc casting were fabricated to strip, which is now being subjected to metallurgical screening studies. Selected samples will be submitted to LASL for corrosion evaluations.

In the tantalum irradiation-damage study, tensile properties of irradiated and unirradiated tantalum will be compared, particularly from the viewpoint of delineating embrittlement owing to irradiation. Encapsulated specimens of tantalum are now being irradiated at the MTR.

Development of Container Materials for LAMPRE Applications

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

A series of binary tantalum alloys containing various amounts of hafnium, thorium, titanium, yttrium, and zirconium as scavenging additions were processed during June to 0.030-in. strip to provide material for investigation; nominal additions of the various elements were listed in BMI-1346. Twenty-five arc-melted buttons, weighing 150 or 250 g, were cold pressed to destroy the cast structure, machined to remove surface impurities, cold rolled to a thickness of about 0.150 in., vacuum annealed for 1 hr at 2600 F, and cold cross rolled to 0.030-in. strip without particular difficulty. Between each of the above steps the material was etched in aqua regia to remove surface contaminants. Currently, specimens cut from each of a total of 25 strips are being given a final anneal, again at 2600 F in vacuum. These, along with specimens in the as-rolled condition, will be sent during July to LASL for tilting-furnace plutonium-alloy compatibility evaluations if chemical analyses and hardness data now being obtained are acceptable.

A supplementary investigation has been initiated to identify minor phases in tantalum (and, perhaps at a later date, in tantalum alloys) by electron microscopy, electron diffraction, and lattice-parameter measurements. It is known that grain-boundary contaminants in tantalum have an adverse effect on its resistance to corrosion attack by molten plutonium-fuel mixtures. If a link between distribution and identity of these contaminants and corrosion behavior can be demonstrated, it will serve as a valuable guidepost for future materials studies. As a starting point, materials representative of both high- and low-purity tantalum are to be investigated during the next few weeks.

L-2

Effect of Irradiation on Tantalum

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

As reported in BMI-1346, two capsules, each containing six sheet tensile specimens, were inserted in the MTR during Cycle 122. It was incorrectly stated, however, that both capsules were located in Position L-53. Actually, Capsule BMI-25-1 is located in Position L-53 while Capsule BMI-25-2 is located in Position L-57. Based on a total perturbation factor of 0.38, estimated effective specimen fluxes are approximately 1.0 to 1.5×10^{14} nv for the former capsule and 1.5 to 2.0×10^{14} for the latter capsule. To achieve a desired transmutation of approximately 1.5 w/o tantalum to tungsten for specimens in Capsule BMI-25-1, exposure during four normal MTR cycles will be required; for specimens in Capsule BMI-25-2, corresponding parameters are 3 w/o conversion and seven normal cycles.

Nickel-cobalt and pure nickel are present in the capsules as dosimeters for determination of fast- and thermal-neutron flux, respectively.

M. DEVELOPMENTAL STUDIES FOR THE PWR

R. W. Dayton

Studies of pressure bonding as a technique for preparing Zircaloy-2-clad compartmented flat-plate fuel elements containing UO_2 cores have continued. Five-hundred and fifty-nine uranium dioxide cores were coated with pyrolytic carbon by cracking methane on UO_2 at 1880 F. Preliminary evaluation indicated that these coatings prevent core-to-cladding reaction and allow good Zircaloy-to-Zircaloy bonding. About 400 cores have been coated with chromium by vacuum evaporation. These coatings also prevented core-to-cladding reaction. Chromium coating also allows the formation of a good Zircaloy-to-Zircaloy bond. After a 1-week exposure in 680 F water, defected test samples were examined. No growth was noted in specimens coated with pyrolytic carbon, graphite, or chromium, and metallographic examination showed no reaction.

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

S. J. Paprocki

The gas-pressure-bonding process is being investigated as a backup effort for the preparation of Zircaloy-2-clad compartmented flat-plate fuel elements containing uranium dioxide cores for the PWR Core 2. In this process the cores and Zircaloy strip components are assembled and fusion edge welded to form a gastight envelope or are inserted in a protective can for bonding. The assembly is pressure bonded at 1525 to 1550 F for 4 hr at 10,000 psi, using helium as a pressure medium to bring the components into intimate contact for solid-state bonding. Previously, the bonded components were subjected to an additional heat treatment of 5 min at 1850 F in a salt bath to improve grain growth across the Zircaloy-to-Zircaloy interfaces. This heat treatment has been discontinued since it is possible to achieve satisfactory bonds without this added treatment.

To prevent reaction between the core and cladding during bonding, the uranium dioxide cores were coated with sprayed graphite, pyrolytic carbon, or vapor-deposited chromium. The sprayed-graphite coatings employed in most of the previous work prevented reaction; however, it was difficult to eliminate the possibility of mechanically contaminating the Zircaloy surfaces during assembly of the components for bonding. Any Zircaloy surfaces contaminated in this manner failed to bond; consequently, failures in burst tests were observed. The failures were between compartments rather than through the cladding which is the normal type of failure. These failures followed no consistent pattern, so it was necessary to evaluate other methods of coating to prevent reaction that would not readily contaminate the Zircaloy surfaces during assembly. An investigation of pyrolytic carbon and vapor-deposited chromium was initiated and studies with sprayed-graphite coatings were continued to serve as a base line for comparison of the effectiveness of the coatings in preventing core-to-cladding reaction.

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Coating of Uranium Dioxide Cores with Pyrolytic Carbon

E. H. Layer, J. D. Foti, and E. Wintucky

During the month of June, 559 uranium dioxide cores were coated with pyrolytic carbon by the cracking of methane on uranium dioxide at 1880 F. Of this total, 486 cores were coated with as-sand blasted surfaces and 73 cores were coated in the as-sintered condition. Fifty of the cores coated in the sand-blasted condition were shipped to Bettis for evaluation studies, while the remaining 375 coated in this manner were prepared for pressure-bonding evaluation studies at Battelle. The 73 cores coated in the as-sintered condition are also being further evaluated by pressure bonding at Battelle.

Thick coatings can be applied by this process to sand-blasted cores with a deposition time of 20 min. These coatings have been evaluated after bonding and were found to be very satisfactory in preventing core-to-cladding reaction. The coatings did not flake or rub off during assembly. The adherence of pyrolytic coatings on as-sintered cores was also investigated in an attempt to eliminate the sand-blasting operation. It was found that the maximum thickness of carbon that could be applied to as-sintered cores corresponded to a deposition time of 2 or 5 min. All films on as-sintered cores produced for pressure-bonding studies were applied in 2 min. Preliminary evaluations of these cores indicated that the coatings were uniform and adherent. The coatings have not been fully evaluated to determine if they are thick enough to prevent core-cladding reaction during bonding.

Preliminary measurements of the thickness of the pyrolytic carbon coatings using a multiple-beam interferometry technique indicate that the previous estimate of several microinches was qualitatively correct. Additional measurements are planned to more accurately determine the thickness of the coatings obtained.

A report is being prepared to cover this phase of the coating program.

Coating of Uranium Dioxide Cores With Vacuum-Evaporated Chromium

E. H. Layer, E. C. Wintucky, and J. D. Foti

Approximately 400 additional cores of uranium dioxide have been coated with chromium deposited by vacuum evaporation. These cores have been prepared for pressure-bonding studies. Coatings were applied to both as-sintered and as-sand blasted cores. Approximately 300 of the cores were sand blasted prior to coating by vapor deposition. The remainder of the cores were coated in the as-sintered condition. All of the coatings were of the quality and thickness found to be satisfactory in preventing reaction between cladding and core during pressure bonding.

The adherence of the chromium on as-sintered cores was investigated and was determined to be equivalent to the coatings obtained on sand-blasted cores based on a

Scotch Tape test. These cores will now be evaluated by scrape tests prior to assembly for pressure bonding.

A report is being prepared concerning this phase of the coating program.

Pressure-Bonding Studies of Zircaloy-2-Clad PWR-Type Fuel Elements

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

The preparation of compartmentalized flat-plate Zircaloy-2-clad fuel elements containing uranium dioxide fuel by gas-pressure bonding is being studied. Elements being pressure bonded contain uranium dioxide cores coated with sprayed and buffed graphite, pyrolytic crystalline carbon, and vacuum-evaporated chromium. The effectiveness of each of these barriers in preventing core-to-cladding reaction and the quality of the Zircaloy-to-Zircaloy bonds in elements containing each of the coatings are being determined. The elements being prepared are assembled using belt-abraded Zircaloy-strip cladding components, which are fusion edge welded, evacuated, and sealed to form a gastight assembly. The edge-welded assembly is gas-pressure bonded at 1550 F and 10,000 psi for 4 hr. The elements currently being prepared are being evaluated in the as-bonded condition only. Previously portions of the specimens were subjected to an additional heat treatment at 1850 F for 5 min subsequent to bonding; however, results have indicated that this extra step is probably not necessary. Specimens already prepared incorporating that heat treatment are being evaluated in both conditions. Evaluation of the elements is based on internal-pressure burst tests, metallographic examination, and intercompartmental leakage and corrosion tests of purposely defected compartments.

Additional tests have been conducted on previously reported specimens containing cores coated with crystalline carbon, graphite, and chromium, portions of which were heat treated after pressure bonding. These elements, prepared using improved sealing, cleaning, and rinsing techniques, have continued to demonstrate strong bonds during burst testing. Burst tests of a total of 52 compartments in these specimens in the as-bonded condition or after an additional 5-min heat treatment in an 1850 F salt bath have resulted in all cases in failure of the cladding at high pressures with normal cladding deflections, rather than failure of the bonds. These included tests of compartments containing each of the three types of core coatings. Metallographic examination of the bonds in these specimens with the various core coatings revealed that there was no contamination of the interfaces and that grain growth had occurred across the original bond interface in all areas. The original specimens of the present program, as previously reported, had contained Zircaloy-to-Zircaloy bonds which were contaminated and revealed in some cases relatively low strength in burst tests, since failure occurred due to rupture of the bond at elevated pressures rather than through the cladding. Because difficulty had been experienced in sealing these specimens and tests showed the possibility of contamination on the Zircaloy bonding surfaces subsequent to cleaning and rinsing, improved cleaning, rinsing, and sealing techniques were used in preparing the later elements.

Thus, the satisfactory results obtained with these specimens show that the cause for the contamination has been eliminated and that uncontaminated bonds with high strength can be consistently produced.

Short-time corrosion tests of purposely defected compartments in 680 F water have been conducted on elements containing cores coated with graphite, crystalline carbon, and chromium in the as-bonded and heat-treated conditions. The graphite coatings had been sprayed on the as-sintered cores, outgassed, and buffed, giving a coating of about 5 mg of graphite per in.^{2} of core surface. The crystalline-carbon coatings were applied to sand-blasted cores by the methane-decomposition technique and were approximately 3 to 5 μin. thick. The vacuum-evaporated chromium coatings, which were applied on sand-blasted cores, had thicknesses in several ranges between the extremes of 10 and 100 μin. The specimens containing cores coated with graphite and crystalline carbon showed no growth during 1 week of exposure; these results are substantiated by metallographic examination which showed no appreciable reaction between the Zircaloy cladding and the cores coated with graphite or crystalline carbon. Specimens containing chromium-coated cores did not show any growth during 1 week of exposure, except one specimen which had chromium coatings only 10 to 12 μin. thick and which had received an additional heat treatment of 5 min at 1850 F. This specimen swelled slightly, increasing in thickness approximately 0.002 to 0.005 in. The specimens with 10 to 12-μin.-thick coatings which were not heat treated and those with coatings greater than 25 μin. thick with and without the additional heat treatment did not reveal any growth during the short-time exposure. Metallographic examination of specimens containing 25 to 100-μin.-thick coatings of chromium indicated no apparent reaction between the cladding and the coated cores in the as-bonded condition and only slight reaction in the heat-treated specimens. The corrosion tests are being continued to determine behavior for longer exposure times.

Elements currently being pressure bonded are being prepared using the improved cleaning, rinsing, and sealing techniques, and are being evaluated in the as-bonded condition only. A set of six specimens has been prepared, one containing graphite-coated cores, two containing chromium-coated cores, and three containing crystalline-carbon-coated cores. Burst tests of 24 compartments in these elements resulted in all cases in failure of the cladding at high pressures and cladding deflections, rather than failure of the bonds. Thus, the 76 consecutive burst tests which have not produced a single bond failure demonstrate that strong Zircaloy-to-Zircaloy bonds are consistently being produced with all three types of core coatings. Metallographic examination and corrosion testing of the six elements are being conducted. Additional elements are being pressure bonded to further investigate the properties and reproducibility of elements containing each of the core coatings. A further set of seven elements has been bonded. This set includes one element containing graphite-coated cores and three elements each containing chromium-coated or crystalline-carbon-coated cores. Burst testing of these specimens is in progress; a limited number of tests of several of these specimens have not resulted in any bond failures. A similar group of seven specimens has been assembled and is being pressure bonded. One of these specimens contains cores which were not sand blasted prior to application of the vacuum-evaporated chromium coatings. Previously, all cores with chromium and crystalline-carbon coatings had been sand blasted prior to coating to roughen the surfaces and improve coating adherence, as discussed in the other sections of this report. A final set of seven elements is planned to be bonded, including two elements containing cores which have not been sand blasted and have been coated with crystalline carbon or vacuum-evaporated chromium.

N. DEVELOPMENTS FOR THE MGCR

W. C. Riley and W. H. Goldthwaite

Research on core materials in support of the MGCR program is in progress at Battelle. Programs on the development and evaluation of UO_2 dispersions in BeO and UC and UC_2 dispersions in graphite, and on the cladding of UO_2 particles with BeO, are in progress. The evaluations include laboratory tests, examinations, and measurements, neutron-activation screening studies of comparative fission-gas-release characteristics, detailed neutron-activation studies of promising developments, in-pile fission-product-release studies, and static capsule irradiations to high burnups.

A study of the diffusion of fission products through fuel-element cladding materials has been initiated.

A study of potential carbon-transport, oxidation, and carburization problems in an unclad-graphite-moderated helium-cooled reactor and of the effect of radiation on these reactions by high-temperature capsule irradiations of graphite and metals in small thermal-convection loops is being concluded.

FABRICATION AND CHARACTERIZATION OF FUEL MATERIALS

A. B. Tripler, Jr.

Current work on fabrication development of fueled-graphite materials is directed mainly toward increasing the density of the matrix and decreasing the permeability. Impregnation with pitch, followed by rebaking at 2300 F, decreased the permeability by a factor of 2.

Progress has been made on the development of a process for applying an impermeable BeO cladding to UO_2 particles. This work is continuing.

Fueled-graphite and fueled-beryllia specimens were prepared for neutron-activation studies. During the initial tests on the fueled-graphite specimens unexpected failure of the quartz tubes occurred, and the work was halted temporarily to determine the cause and to eliminate the difficulty.

UO_2 Dispersions in BeO

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

Fuel-element cores consisting of about 20 volume per cent of UO_2 particles dispersed uniformly in a densely sintered BeO matrix are being developed.

During June, a series of BeO-UO_2 pellets was prepared by the procedure described in BMI-1346 for neutron-activation testing. These bodies contained about 20 volume per cent of natural UO_2. The total porosity of the pellets and the crystal size of the BeO phase were varied in this experiment by varying the conditions under which the pellets were sintered. The sintering conditions, the bulk density of the pellets, and the crystal size of the BeO are shown in Table N-1.

TABLE N-1. DESCRIPTION OF BeO-UO_2 FUEL-ELEMENT CORES PREPARED FOR NEUTRON-ACTIVATION TESTING

Sintering Conditions(a)		Properties of Sintered Pellets		
		Bulk Density(b)		
Maximum Temperature, F	Time at Maximum Temperature, hr	G per Cm^3	Per Cent of Theoretical(c)	BeO Average Crystal Size(d), μ
2600	1	4.27	92.3	5
2800	1	4.41	95.3	18
3000	1	4.54	98.0	37
3000	4	4.51	97.5	37

(a) In flowing hydrogen.
(b) Average of four specimens; determined by xylene absorption and displacement.
(c) Taken as 4.63 g per cm^3 for bodies containing 48.25 w/o UO_2 and balance BeO.
(d) From examination of polished and etched section at 250X.

The bulk densities of four groups of pellets ranged from 92.3 to 98.0 per cent of the calculated theoretical density for this composition. Average crystal size of the BeO phase in the four groups ranged from 5 to 37 μ. Room-temperature permeability measurements were made with a 1-atm differential pressure of argon. Results indicated that the permeability of specimens from all groups was less than 3×10^{-7} darcy.

UC and UC_2 Dispersions in Graphite

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

The objective of this research is to develop improved graphite fuel-element cores containing dispersed UC or UC_2 in an amount equivalent to 20 volume per cent of UO_2.

In previous work it was reported that the optimum pitch-binder content was about 16 w/o for bodies made with pulverized AGOT graphite fillers having two different ranges in particle size, and about 14 w/o for bodies made with a pulverized petroleum-coke filler. During June, data were obtained on the gas permeability of these bodies.

The specimens that were made with minus 325-mesh particles of AGOT graphite filler, bonded with 16 w/o of pitch, showed the lowest permeability. After pitch impregnation and rebaking at about 2300 F, the permeability of the specimens was decreased by approximately a factor of 2.

An investigation of minor additives for aiding densification was initiated. Results to date indicate that a small addition of mineral oil destroys the bonding power of the coal-tar pitch.

Another additive tried was beeswax dispersed in CCl_4. No improvement in density or compressive strength resulted from additions of 1.5 to 3.0 w/o beeswax to a mix containing 16 w/o of coal-tar pitch.

In future work the investigation of minor additives for aiding densification will be continued. The effect of reimpregnation on gas permeability of specimens also will be determined.

Cladding of UO_2 Particles With BeO

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

A process for applying an impermeable BeO cladding to UO_2 particles is being investigated.

Work was continued during June on developing a process for preparing solid, spherical UO_2 particles in the size range of 100 to 500 μ. Principal emphasis was on spray drying. Progress was made, although UO_2 particles meeting the above specifications are not yet available. Work is continuing in this area.

BeO-clad UO_2 pellets in the size range of 1000 to 2000 μ were prepared and tested for cladding integrity during June. The fuel materials used in these experiments were nonspherical grains of either UO_3 or a mixture of UO_2 and $Be(OH)_2$. These were selected on the basis of their high shrinkage resulting from (1) the reduction of UO_3 to UO_2, or (2) the dehydration of $Be(OH)_2$ to BeO during sintering. The sintering shrinkage of such fuel particles would be expected to equal or exceed the shrinkage of the BeO cladding, and thus obviate the use of an intermediate organic burnout layer.

The fuel particles were clad with LOH-grade BeO by a pelletizing technique. The pellets were compacted isostatically at 100,000 psi and then sintered for 1 hr at 2800 F. Chemical analyses of sintered pellets indicated that the batches contained from 20 to 30 w/o of UO_2.

The quality of the claddings was evaluated by heating about 1000 pellets in air at 1200 F and measuring the total weight gain of the pellets resulting from the oxidation of the UO_2 cores to U_3O_8. In these initial trials, from 10 to 20 w/o of the UO_2 present was oxidized during a 100-hr heat treatment. It is probable that the weight gain resulted from complete oxidation in some of the pellets and that no oxidation occurred in some pellets.

Techniques for fabricating BeO-clad UO_2 pellets will be refined in future experiments. Also, a study of the tendency of densely sintered BeO to react with carbon at temperatures up to 3000 F will be undertaken.

N-4

Preliminary Characterization by Neutron Activation

P. Gluck and M. Pobereskin

Specimens of UC-graphite, UC_2-graphite, and BeO-UO_2 were received during the first half of the month and screening studies were initiated.

The UC-graphite and UC_2-graphite specimens were separately irradiated at a neutron flux of 3.5×10^{11} nv for 1 hr and 1-3/4 hr, respectively. After a decay period of 5 days, each of the five UC-graphite specimens, along with 1 cm^3 of activated charcoal, was individually encapsulated in quartz tubing. The tubes were connected to a vacuum system overnight while the charcoal was degassed at a temperature of 500 F. The evacuated tubes were sealed off and were annealed at 2100 F for 24 hr.

Fission-gas-release data were not obtained because during the annealing process excessive pressure built up in the tubes which resulted in leaks or ruptures. Only one of the tubes remained intact, but this, too, was deformed by expansion; the activity in the gas of this tube has not yet been determined. The remaining tubes ruptured and broke up as they cooled. During the cooling process the formation of chalklike layers on the inside surface of the tubes was observed. In the ruptured tubes the specimens were found to have disintegrated or to have swollen appreciably.

As a result of this observation the annealing studies were temporarily suspended in order to investigate the causes of failure in the quartz tubing. It was found in the literature that quartz goes through a structural transformation at temperatures in excess of 1180 C. This transformation is reported to be catalyzed by alkali salts, phosphoric acid, and radioactivity; the transformed quartz is crystalline in nature and has a much lower coefficient of expansion than fused quartz.

Experiments are presently being conducted with nonirradiated specimens – the annealing being conducted at 1800 F. The extent to which the activated charcoal must be preheated and the quartz tube evacuated will be determined before resuming the annealing of the irradiated specimens.

STUDIES OF FISSION-GAS RELEASE FROM FUEL MATERIALS

M. Pobereskin

A program on fission-gas release from MGCR candidate fuel materials is under way. The program includes gas-release measurements by postirradiation heat treatment of low-burnup specimens (neutron-activation studies) and dynamic measurements made during irradiation (sweep-gas capsule studies) on materials judged to be promising in the neutron-activation studies.

Initial efforts have been devoted to design and procurement of the necessary apparatus and to the development of experimental design and data-evaluation techniques.

Detailed Neutron-Activation Studies

P. Gluck and M. Pobereskin

The objective of this study is to investigate fission-product release from specimens that have been subjected to low-exposure low-temperature irradiation and then heat treated at controlled temperatures up to 2800 F. Purified helium will be passed over the specimens. A proportional flow counter will continuously monitor the activity in the gas stream. The fission gases carried in the helium stream will be adsorbed on activated charcoal. The activated charcoal can be removed from the system for measurements in a scintillation counter.

During this month, various components of the apparatus required for detailed neutron-activation studies were ordered and received. Progress was made in assembling and constructing the apparatus for the postirradiation study to be conducted in the radioisotope laboratory. It is now anticipated that preliminary experiments will be initiated during July.

In-Pile Studies

N. E. Miller, P. Gluck, and G. E. Raines

The objective of these studies is to determine the fission-gas release from prospective fuel-element specimens during irradiation under conditions of surface temperatures and heat fluxes comparable to those expected in service. The unclad fuel specimen (0.22 in. in OD and up to 2 in. long) will be contained, with minimum clearances, in a cylinder within an irradiation capsule. During irradiation, helium will flow through the fuel-containing cylinder to carry the released fission gas out to an analytical train for elemental determinations. From preliminary design calculations it appears feasible to operate the fuel over a range of surface heat fluxes from 150,000 to 400,000 Btu/(ft^2)(hr) and surface temperatures from 1500 to 2000 F when irradiated in the BRR core.

The components for this system have been received and assembly has begun.

Experimental Design and Evaluation

A. E. Mace and S. D. Beck

The objective of this study is to develop an experimental design for in-pile gas-release experiments to determine the effect of surface heat flux and temperature on gas release.

The detailed development of the experimental design must take into account the details of the experimental apparatus. When these details are available, experimental design will proceed.

HIGH-BURNUP IRRADIATION EFFECTS IN FUEL MATERIALS

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

The irradiation in the BRR of a Capsule GA-3-1, containing compacts of UO_2 in a BeO matrix, UC in a graphite matrix, and UC_2 in a graphite matrix, has been completed. The objective of the irradiation was to study the stability of these fuels under conditions simulating those of the MGCR design. The capsule contained three pairs of specimens, each specimen composed of four pellets approximately 0.222 in. in diameter by 0.250 in. long. The pellets were sealed into stainless steel tubes under a helium atmosphere.

The capsule operated satisfactorily during all four cycles in the reactor. The calculated surface temperatures of the specimens based on temperatures measured by thermocouples located inside the capsule, are listed in Table N-2 along with the estimated burnups, which were calculated from neutron-flux measurements obtained in the reactor holes prior to insertion of the experiment. These burnups will be compared to burnups calculated from neutron dosimetry, after radiochemical analyses of the dosimeters are completed. The postirradiation examination of the specimens has been initiated in the Battelle Hot-Cell Facility.

The six specimens have been recovered from the irradiation capsules. All specimens appeared in good physical condition with no obvious signs of failure. The specimens were examined macroscopically and were photographed. Measurements of the physical dimensions of each specimen indicated that none of the specimens swelled during irradiation. Three specimens, each containing a different type of fuel, were sampled for fission-gas content, and analyses of the collected gases are in progress.

All of the specimens have been cut open with an abrasive wheel for recovery of the fuel pellets. The pellets contained in the specimens sampled for fission gas were crushed, apparently as a consequence of the puncturing operation. The pellets in the other specimens were intact. Measurements of the physical dimensions of all the intact pellets and the relative density of the BeO-UO_2 pellets are in progress.

TABLE N-2. CALCULATED IRRADIATION TEMPERATURES AND BURNUPS OF SPECIMENS RECOVERED FROM CAPSULE GA-3-1

	Composition[b]					
Specimen[a]	Fuel	Matrix	Calculated Surface Temperature[c], F	Calculated Heat-Generation Rate[c], Btu (ft^2)(hr)	Calculated Burnup of Uranium Atoms, per cent	Calculated Fission Density[d], 10^{19} fissions per cm^3 of fuel
1	UC_2	Graphite	1520	204,000	1.6	9.34
2	UC	Graphite	1560	198,000	1.6	9.24
3	UO_2	BeO	1550	179,000	1.5	8.14
4	UO_2	BeO	1505	165,000	1.4	7.55
5	UC	Graphite	1430	156,000	1.2	6.93
6	UC_2	Graphite	1275	132,000	1.0	5.85

(a) Specimens are numbered from the top of the capsule.

(b) The BeO-matrix fuel specimens contain about 24 volume per cent of 93.2 per cent enriched UO_2. The fuel content of the other specimens was adjusted to match this.

(c) Based on temperatures measured by thermocouples to ± 20 F.

(d) The fission density is based upon the actual volume of the fuel pellets.

In general, the appearance of all pellets, aside from those crushed due to puncturing operations, was good. A few small cracks or flaws were noted on some of the pellets, but these have not as yet been definitely attributed to irradiation.

The metallographic examination of one pellet of each fuel material, and a section of cladding from three specimens, each of which contained a different type of fuel pellet, is in progress. Analyses of neutron-dosimeter wires recovered from the capsule are also in progress. Radiochemical analyses of the pellets for fuel burnup and of the cladding for fission products are planned.

Specimens have been fabricated and loaded into three additional irradiation capsules. These capsules are en route to the MTR for irradiation under conditions of temperature similar to the BRR capsule irradiations. It is currently planned to irradiate one capsule to a burnup of 12 a/o of the uranium-235, and the other two capsules to burnups of about 20 a/o, with the irradiations to begin in July.

DIFFUSION OF FISSION PRODUCTS IN CLADDING MATERIALS

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

The program to study the diffusion of fission products in "A" Nickel cladding material is being continued.

It is planned to impregnate the cladding materials by irradiation recoil. For this purpose sandwiches have been prepared by placing a 1-mil 93 per cent enriched uranium foil 1 cm square between two foils of "A" Nickel, 1 in. square and 1, 5, and 10 mils thick. The uranium was centered between the "A" Nickel foils and the sandwich was spot welded at several places around the edges. Good contact between the uranium and "A" Nickel was provided by compressing the sandwiches under an applied load of 20 tons.

Twenty sandwiches thus prepared for irradiation were contained between two 1/8-in.-thick aluminum plates approximately 6 in. high by 7 in. wide. One of these plates had 20 cavities milled in one face to the depth of the sandwiches. The sandwiches were inserted in the face and the two aluminum plates were welded together in a helium atmosphere. A second containment was made by aluminum channels which were slipped over each edge and welded to each face of the capsule. The capsule was irradiated in the BRR for 3 hr in a flux between about 8 x 10^{12} and 1 x 10^{13} nv. The flux each sandwich was exposed to will be determined by dosimeter wires attached to the capsule. The sandwiches will be allowed to decay for about 1 month before removal from the capsule and subsequent analyses.

In a study of the introduction of gases into metals by cathodic bombardment, two "A" Nickel foils were subjected to argon bombardment for 2 hr at 200 μ of pressure at an applied voltage of 450 v. The foils were surface cleaned and sealed in evacuated Vycor capsules and heated to 900 C for 4 days to drive off any absorbed argon. The capsules will be analyzed for released argon by mass spectroscopic methods.

CARBON-TRANSPORT CORROSION STUDIES

N. E. Miller, D. J. Hammon, J. E. Gates,
and W. S. Diethorn

A study is being made of potential carbon-transport, metal-oxidation, and metal-carburization problems which may be associated with an unclad-graphite-moderated helium-cooled reactor system. Capsule GA-2-2 containing the four quartz tori described in previous reports was irradiated in the BRR core for one cycle (240 hr at 2 megawatts). During this time the specimens in the hot and cold legs of each torus were at 1500 ± 10 F and 960 ± 10 F, respectively. Preliminary hot-cell results from the torus capsule are now available. These results are summarized in Table N-3.

All four tori were broken during removal from the opened capsule. The heat barrier holding the tori could be not be slipped out of the capsule and a strong pull was required to dislodge it. During this operation the tori were broken into several pieces, maintaining, in general, the torus geometry. There was no evidence that torus breakage or fracture had occurred before the pull and it is believed that the tori were intact during the irradiation. Torus breakage, of course, prevented the collection and analysis of torus helium.

There was no evidence of carbon deposition on the few pieces of quartz debris examined. One of the zirconium foils was not recovered. This foil may have become lodged in the capsule when the tori broke.

TABLE N-3. WEIGHT CHANGES OF SPECIMENS FROM CAPSULE GA-2-2

Specimen Type	Code Number	Harp Number	Preirradiation Weight, g	Postirradiation Weight, g	Weight Change[(a)], mg	Temperature, F
Graphite	11	5	5.4896	5.4898	(+0.2)	960
	12	5	5.5260	5.5249	-1.1	1500
	13	6	5.5241	5.5229	-1.2	960
	14	6	5.5149	5.5131	-1.8	1500
	15	7	5.5142	5.5133	-0.9	960
	16	7	5.5034	5.5024	-1.0	1500
	17	8	5.5430	5.5416	-1.4	960
	18	8	5.5316	5.5303	-1.3	1500
Nickel	16	7	0.1248	0.1250	(+0.2)	1500
	17	7	0.1376	0.1294	-8.2	960
	18	8	0.1294	0.1297	(+0.3)	1500
	19	8	0.1377	0.1282	-9.5	960
Monel	26	7	0.1324	0.1326	(+0.2)	1500
	27	7	0.1315	0.1320	+0.5	960
	28	8	0.1321	0.1323	(+0.2)	1500
	29	8	0.1304	0.1305	(+0.1)	960
Niobium-1 w/o zirconium	36	7	0.1370	0.1384	+7.4	1500
	37	7	0.1376	0.1332	-4.4	960
	38	8	0.1372	0.1381	+0.9	1500
	39	8	0.1377	0.1382	+0.5	960
Zirconium foil	1	6	0.2158	Not recovered	--	1500
	2	8	0.2460	0.2470	+1	960

(a) Precision of weight measurements is ±0.1 mg; therefore, weight changes of $\leq$ 0.2 mg (indicated by parentheses) are not significant. Weight losses of the graphite specimens are probably the result of further outgassing during irradiation.

Photomacrographs of the graphite (1X) and the metal specimens (4X) were taken. The graphite specimens were intact after removal from the capsule and were indistinguishable in appearance from unirradiated specimens. The only visual change in the metal specimens was a slight coloration of a few of them relative to unirradiated specimens. There was no visual evidence of severe attack on the metals. Metallography of a few graphite specimens and all of the metal specimens has just been completed, and the results 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 construction of the plastic-model parts, the model core, and the parts for the test setup.

In June, the plastic-model parts were received from the vendor. The model was assembled and installed in the test setup and a preliminary run was made to check general operation and instrumentation. For the several different air-flow rates used, model operation was satisfactory and flow was stable. The 248 flow-measuring orifices, one in each simulated fuel assembly, were calibrated in place after assembly of the model in preparation for core flow-distribution studies.

In July, studies will be made of the effects of size and location of the holes in the core-support cylinder on core-flow distribution, pressure drop, and mixing. Based on the results of these studies, a hole pattern will be selected for use in more detailed studies.

P. DEVELOPMENTAL STUDIES FOR THE SM-2

S. J. Paprocki

The work discussed in this portion of the report is being conducted to assist Alco Products in the development of fuel, absorber, and suppressor materials for the SM-2.

Stainless steel-UO_2 dispersion-type specimens containing both ORNL hydrothermal UO_2 and Mallinckrodt spherical UO_2 have been fabricated for irradiation testing in the ETR and MTR. These specimens embody a burnable poison in the form of ZrB_2 or NbB_2 compound dispersed in the fuel core. Studies have now been initiated to develop techniques for the fabrication of full-size fuel elements incorporating these materials.

Capsules have been designed for the irradiation of SM-2 specimens in both the ETR and MTR. The ETR capsules will be fully instrumented with thermocouples, dosimeters, and auxiliary resistance heaters. The MTR capsules will be uninstrumented as they will be irradiated in MTR core positions which do not accommodate instrumentation leads. Capsule mock-ups are being prepared and will be forwarded to the ETR and MTR for low-power exposure to determine the total capsule perturbation. These data will be utilized to more accurately estimate burnup rates during irradiation of the SM-2 specimens.

Development of Fuel Materials

S. J. Paprocki, D. L. Keller, G. W. Cunningham,
D. E. Lozier, W. M. Pardue, and J. M. Fackelmann

Reference and alternate fuel-element materials have been selected and techniques are being developed for preparing full-size plates. The reference fuel elements contain 0.030-in.-thick cores of 26 w/o UO_2 and 1.1 w/o ZrB_2 dispersed in a prealloyed Type 347 stainless matrix and are clad with 0.005-in.-thick Type 347 stainless. Alternate fuel materials involve a change only in the boron compound, and NbB_2 and BN are being investigated as alternate compounds. Fabrication procedures are being evaluated on the basis of metallographic examination, chemical analysis, and physical and mechanical properties. In addition, a major part of the development program has been directed toward the preparation of irradiation specimens.

Several full-size fuel elements have been fabricated to approximate dimensions, and sufficient data have been collected to design the core-compacting die. The die is being machined and will be used for producing full-size plates for evaluation tests. Other work on the reference plates has been primarily concerned with determining the effect of various impurities on the behavior of ZrB_2 and the effect of impurities and change in shape of UO_2 particles on the quality of the roll-clad dispersion.

As previously reported, under poor sintering conditions, ZrB_2 will oxidize, boron will be lost, and, in some cases, a reaction product may be observed in the matrix.

However, even under these conditions the behavior of individual ZrB_2 particles varies. Thus it appears that impurities or secondary phases in the ZrB_2 may be detrimental. Therefore, some tests are being conducted to determine whether impurities such as carbon, niobium, or silicon, or oxygen-bearing constituents cause reaction when added either to the ZrB_2 or to the metal matrix. Samples containing three grades of ZrB_2 with known impurity contents are being used. In addition, iron-matrix specimens containing 0.1 to 1.0 w/o carbon, 0.5 to 2.0 w/o niobium, or 1 to 3 w/o silicon, 0.5 to 2.0 w/o niobium are being used to evaluate the effect of impurities added to the matrix metal. These specimens are being fabricated but have not been examined.

Preliminary work with UO_2 shot indicates that fracturing and stringering of UO_2 particles is related to the percentage of spherical particles in the shot. Apparently the strength of the particle is closely related to the shape. Additional tests are being planned to establish this relationship.

Results of transverse tensile tests are being used to aid in evaluating fabrication procedures. The effect of varying the total reduction is being tested on a series of plates containing 28 w/o minus 100 plus 200-mesh high-fired UO_2, and 1 w/o minus 200 plus 270-mesh ZrB_2 dispersed in a prealloyed Type 347 matrix. The plates were rolled at 2200 F using a 40 per cent reduction in thickness on the first pass followed by 20 per cent reductions for each remaining pass. All plates were cold rolled 15 per cent and annealed at 2050 F before testing. The results of the room-temperature tensile tests are shown in Table P-1. Although there is no marked change in strength over the range of 6:1 to 9:1 total reduction, there is a trend toward decreasing strength at higher reductions.

TABLE P-1. EFFECT OF TOTAL REDUCTION ON TRANSVERSE TENSILE STRENGTH OF 28 w/o UO_2-1 w/o ZrB_2 DISPERSION CORES

Total Reduction in Thickness	Transverse Tensile Strength(a), psi
6:1	18,350
7:1	16,600
8:1	16,950
9:1	15,350

(a) Measured at room temperature – average of two tests.

Work was continued on investigation of alternate poison compounds for the SM-2 as well as on determination of optimum fabrication treatment for the 40 w/o UO_2 specimens. To re-evaluate the previously reported results concerning 1.94 w/o ZrB_2 and 40 w/o UO_2 in elemental matrices, a specimen of this type fabricated under reference conditions (prefiring of ZrB_2 for 2 hr at 2300 F in vacuum, prefiring iron powder for 2 hr at 1400 F in dry hydrogen, vacuum sintering at 2000 F, and hot rolling at 2000 F) was annealed in hydrogen. Annealing was for 2 hr at 2300 F. Maximum variation of specimen temperature was ±10 F as measured by a thermocouple adjacent to the specimen. Metallographic examination showed distinct ZrB_2 particles with a continuous, but slight, oxide layer around the circumferences. Identical specimens have also been prepared with the high loading in a prealloyed matrix. By use of spherical UO_2 a discrete dispersion comparable to the structure obtained with the elemental matrix was observed after hot rolling. Evaluation after cold rolling has not been made, but it appears that good dispersions containing 40 w/o UO_2 can be made with prealloyed matrices.

In order to determine whether boron could be in the matrix but not visible because of the fabrication procedure, a series of roll-clad specimens was run through two cycles of heating for 2 hr at 2300 F in hydrogen followed by furnace cooling overnight. Specimens and results indicate that when ZrB_2 is used no new phases are present after heat treating. A slight precipitate was visible in the case of NbB_2, but 2300 F is considerably higher than the fabrication temperature (2000 F). These results can be correlated with previously reported chemical analyses on microdrillings in a specially prepared matrix.

Boron nitride (BN) has also been investigated in both elemental and prealloyed matrices at sintering temperatures of 2000 and 2200 F for periods of 3 hr, after standard mixing and pressing. Both types of matrix showed reaction precipitates after sintering at 2200 F. After sintering at 2000 F the elemental matrix showed slight edge reaction of the BN, while the structure of the sample containing prealloyed powder compared favorably with NbB_2 fabricated at 2000 F.

The revision of the SM-2 irradiation program is presently in process, and the specimens previously assigned for ETR Capsules 1 through 7 are being reassigned. Six specimens have been assigned to one ETR capsule and are currently in process of being encapsulated. Four specimens are being used in a mock-up capsule and 22 specimens, including 2 double-sized suppressor specimens, have been assigned to the 3 eight-specimen capsules scheduled for irradiation in the MTR core. All of the scheduled specimens have been fabricated with the exception of a pair of SM-1 type specimens which are in process.

No change is planned in loadings previously described for the ETR capsules, but fabrication of the specimens and loading of the capsules can be extended over a longer period of time since the ETR will operate for very short periods of time over the next several months.

Chemical analyses for boron content are listed in Table P-2 for irradiation specimens which have not been previously reported.

Encapsulation Studies

A. K. Hopkins, W. E. Murr, and J. H. Stang

As reported previously, this program has been devoted to the design and construction of thermocoupled and heatered capsules for the irradiation of SM-2 fueled specimens to burnups of 77 and 37 per cent of the fissionable atoms. It was planned to

TABLE P-2. BORON ANALYSES FOR IRRADIATION-TEST SPECIMENS

Description of Specimen	Sintering Temperature (2 Hr in Vacuum), F	Rolling Temperature, F	Boron Composition, w/o		Boron Analysis, w/o	
			Reference	Nominal	Sintered Core Compact	As-Rolled Plate
Hydrate UO_2-NbB_2	2000	2000	0.20	0.22	0.212 (0.219 green)	0.209
	2000	2000	0.20	0.22	--	0.207
Spherical UO_2-NbB_2	2000	2000	0.20	0.22	0.201 (green)	0.204
	2000	2000	0.20	0.22	--	0.202
Boron-alloy frame	2200	2200	--	--	--	0.023
Spherical UO_2-ZrB_2	2200	2200	0.10	0.11	--	0.0925
	2200	2200	0.20	0.22	0.232	0.200
	2200	2200	0.20	0.22	--	0.192
26 w/o UO_2-ZrB_2	Rolled green	2200	0.2	0.22	--	0.238
40 w/o UN-ZrB_2	2000	--	0.348	0.386	0.420	--
40 w/o hydrate UO_2-ZrB_2	2000	--	0.365	0.405	0.343	--
40 w/o spherical UO_2-ZrB_2	2000	--	0.365	00.405	0.406	--

irradiate all capsules in ETR beryllium reflector positions, where quoted thermal fluxes in the neighborhood of 3×10^{14} nv are available. However, as reported in BMI-1336, official information received from the ETR during May indicated that the reactor will operate at only a fraction of the normal on-stream time during the remainder of this calendar year. Since this low duty would make it impossible to achieve the specimen-burnup rates desired, it has been necessary to revise the capsule-irradiation plan originally adopted. The revised schedule, which was formulated early in June, is essentially the following:

(1) Three nonlead capsules, each containing eight specimens, will be inserted in the MTR core as soon as possible. Quoted fluxes in positions of interest (in the L-50 row) range from 3 to 5×10^{14} nv. Without auxiliary heat, the specimen temperature in these capsules, nominally 650 F at the start of irradiation, will decrease steadily as burnup progresses. This characteristic will make it desirable to incorporate suitable heat treatments into the postirradiation specimen-examination schedule.

According to present estimates, these capsules can be readied for shipment to MTR during July.

(2) One thermocoupled and heatered capsule, containing six specimens, will be fabricated immediately for insertion into the beryllium reflector at ETR. It is estimated that this capsule can be assembled and shipped by mid-July.

(3) Six additional ETR beryllium reflector capsules are to follow between now and the end of the calendar year. These will contain specimens of advanced fuels and specimens containing Eu_2O_3.

During June, design studies involving the three MTR capsules were completed by computational and analog procedures. These capsules are designed on the basis of an unperturbed thermal-neutron flux of 5.0×10^{14}, a Brad Lewis-correlation-derived capsule-perturbation factor of 0.38, a gamma-heating rate of 10 w per g, and a specimen-surface temperature of 650 F. The design formulated is similar to that previously developed for the ETR capsules, in that the various elements are selected to dissipate a large quantity of fission heat while maintaining a relatively low specimen temperature. In essence, the specimens are immersed in NaK with a split copper block insert adjacent to but not touching them. These components are sealed into a nickel water-contacting shell. In the heatered capsules, coils of sheathed resistance-wire heaters are wrapped around the copper-block assembly.

Also during June, supplementary capsule-perturbation-factor studies were conducted using a P-3 type analysis with an IBM-650 computer. Results now available for the case of capsules in MTR core positions indicate the total capsule perturbation may be roughly twice as great as predicted from the Brad Lewis analysis. However, since both calculational procedures are based on simplifying assumptions, it appears necessary to conduct actual nuclear-mock-up experiments to obtain concrete data. An appropriately dosimetered capsule mock-up is now being constructed and, early in

July, this mock-up will be shipped to the MTR for a low-power exposure. At the same time, another mock-up will be submitted for an exposure in the ETR beryllium reflector. The immediate use of data derived from these mock-ups will be in estimating burnup rates as the irradiations of the three MTR and the first ETR capsules progress. If the high perturbations predicted by the P-3 calculations are verified, it will probably be desirable to modify the design of the later series of six ETR capsules in the direction of reduced heat-dissipation characteristics.

Q. GAS-COOLED REACTOR PROGRAM

D. L. Keller

Studies for Aerojet-General Nucleonics (AGN) directed toward the development of compact gas-cooled reactors are reported in this section. The activities on the various tasks are reported under "Materials Development Program" and "In-Pile-Loop Program".

MATERIALS DEVELOPMENT PROGRAM

D. L. Keller

Capsule BMI-27-2 was prematurely discharged from the MTR due to higher than planned temperatures. Capsule BMI-27-1 may be relocated after postexamination of BMI-27-2. Capsule BMI-28-1, containing flat-plate specimens of UN and UO_2 dispersions in stainless steel was also prematurely discharged because of high-temperature exposure.

Preparation of the graphite-UO_2 pins was completed and the encapsulated pins were shipped to the MTR.

The preparation of 6500 UO_2 ceramic pellets for in-pile testing has been completed.

The ML-1 reactor was mocked-up for critical studies and criticality was achieved with 59 fuel elements.

Encapsulation Studies

J. C. Smith, P. B. Shumaker, C. V. Weaver,
D. W. Nicholson, J. F. Lagedrost, and J. H. Stang

Irradiation of Clad UN and UO_2 Dispersions

Capsule BMI-28-1 was discharged from the MTR on June 16. This capsule, which contains one UO_2 and three UN (dispersions in stainless steel) clad flat-plate specimens, was inserted in the MTR at the start of Cycle 120, April 10, 1959. It operated at design temperatures (1650 ± 50 F at the specimen surfaces) for three cycles; during this period, it is estimated that fission burnup was accumulating at the rate of approximately 2.4 a/o uranium-235 per cycle.

The capsule was scheduled for removal from the reactor at the end of the fourth cycle of exposure. However, temperatures were, for some unexplained reason, higher than normal during the first portion of Cycle 123, which began on June 12. Temperatures at the topmost specimen, previously around 1600 F, were in the 1750 to 1780 F range. The specimens by this time had accumulated an estimated burnup of approximately 7.2 a/o, which is high enough to be in the range of practical interest. Hence, since it was believed that the Type 318 stainless steel cladding could not withstand these higher temperatures for an extended length of time, a decision was made to remove the capsule prematurely and avoid inordinate specimen damage that would make it impossible to obtain meaningful postirradiation data.

Original planning called for opening of this capsule at the Battelle Hot-Cell Facility during July or early August. As things now stand, the capsule should arrive at Battelle early in July. However, owing to a recent shift of emphasis on the over-all program, it will probably be several months before the hot-cell inspection is performed.

Irradiation of Clad Pin-Type Specimens Containing Dense UO_2

The irradiation of Capsules BMI-27-1 and BMI-27-2, each of which contains six Inconel-clad UO_2-fueled pin-type specimens, continued at the MTR during June. However, Capsule BMI-27-2 was discharged at the end of five MTR cycles (at the conclusion of Cycle 123). It was irradiated in Position A-30-SE at elevations as follows:

(1) An initial position (for three reactor cycles) with the specimen zone located between approximately 15 and 29 in. below the lattice. Thermocouple-monitored temperatures in this position ranged from approximately 1100 to 1650 F, the higher temperatures being maintained at the bottom-specimen locations; specimen-surface temperatures are in the neighborhood of 50 F higher than the thermocouple temperatures.

(2) A second position approximately 8 in. higher than the original. This position was occupied after an unsuccessful attempt to lower the capsule 3 in. As described in BMI-1346, the indicated temperatures after the 3-in. lowering were unexpectedly high during the reactor startup period.

(3) A third position approximately 2 in. higher than the second. This is the position occupied during the last month of irradiation. The repositioning was carried out after an attempt to achieve the original position failed because the indicated temperatures during reactor startup were again unexpectedly high.

In this third position thermocouple temperatures ranged from 1000 to 1200 F.

During June, the decision was made to discharge the capsule sooner than previously anticipated. This decision was based on several factors. In the first place, the thermal behavior of the capsule could not be completely rationalized; MTR personnel were unwilling to attempt to lower the capsule again without complete clarification of this behavior. Secondly, the estimated peak-flux specimen burnups were in the 4 a/o fission-burnup range, which is near the average anticipated for prototype reactor core life. Lastly, it was thought that hot-cell inspections of the capsule might reveal internal conditions that could account for the irregular thermal operation; if so, it would be of advantage to gain this knowledge as soon as possible so that it could be used in formulating a decision regarding relocation of Capsule BMI-27-1 in the reactor.

The latter capsule was positioned approximately 9 in. (midspecimen-zone plane) above the peak-flux zone in Position A-7-NW from Cycle 120 through Cycle 122. Peak-flux specimen (bottom-pair) temperatures during that period were in the 1400 to 1600 F range. At the startup for Cycle 123, the indicated temperatures were higher than expected, and the capsule was subsequently raised about 3 in. At this elevation, bottom-specimen temperatures still appear to be in the 1400 to 1600 F range based on estimates inferred from upper-specimen thermocouples which are registering temperatures in the neighborhood of 500 F; the thermocouples adjacent to the bottom specimens are erratic. While it would be desirable to lower the capsule from the viewpoint of raising the temperatures of the top specimens, plans to do so have currently been postponed since personnel at the MTR believe that such action is undesirable, at least until results of the posttest examinations of Capsule BMI-27-2 as described above are available.

Irradiation of Specimens Containing UO_2 in Graphite

Capsule BMI-29-1 was loaded in mid-June and shipped to the MTR. This capsule contains six metal-clad 1/4-in.-OD pins containing 8 w/o UO_2 (highly enriched) in graphite; cladding materials are Hastelloy X, Inconel 702, and Carpenter 20Cb. According to design specifications, the specimens should operate at 1750 ± 50 F in an unperturbed flux of 1.33×10^{14} nv. The capsule design is based on the double-wall scheme in which the specimens are immersed in NaK contained in an inner sealed shell; a narrow helium annulus separates the shell from the outer water-contacting shell. Auxiliary sheathed resistance-wire heaters and thermocouples are incorporated into the system.

In the flux listed above, it is estimated that the target fission burnup of 15 a/o will be accumulated in three normal MTR cycles.

Irradiation of Specimens Containing MCW Spherical UO_2 Dispersed in Stainless Steel

Two irradiation-capsule experiments designed to compare the irradiation stability of stainless steel-clad dispersions of stainless steel-30 w/o MCW spherical UO_2

(highly enriched) with that of clad dispersions of stainless steel-30 w/o ORNL hydrothermal UO_2 have been initiated. The capsules are designed to maintain the four specimens present in each capsule at a nominal temperature of 1650 F by fission heat; two of the specimens in each capsule will contain the spherical UO_2 and the other two will contain the hydrothermal UO_2. The target burnups are 6 a/o and 9 a/o fission burnup, respectively, in the two capsules.

The capsule design is based on those employed previously for Capsules BMI-11-4 through BMI-11-7 and BMI-18-3. It involves a double-wall configuration in which the specimens are immersed in NaK contained in a finned stainless steel inner shell. The fins provide the thermal contact between the inner shell and the water-contacting outer shell. Thermocouples are present in the NaK space to provide specimen-temperature data.

Fabrication of the capsule components is completed, and assembly is in progress. The specimens have also been fabricated, and preinsertion measurements and inspections have been completed. It is estimated that both capsules will be ready to send to the MTR during the first week in July.

Irradiation of UO_2-Graphite Specimen Assemblies With an Integral Corrosion-Gas-Flow System

A new elevated-temperature capsule-irradiation program involving silicon carbide-coated graphite-UO_2 (8 w/o highly enriched uranium-235) bodies is under way. Three capsules will form a vertical train, which will be positioned adjacent to a core face of the Battelle Research Reactor. Each capsule will contain a specimen consisting of a stack of two fueled and one unfueled graphite disks housed in a close-fitting metal can. The disks will be about 1-3/8 in. in diameter and 1 in. long. The peak unperturbed flux in this irradiation space is approximately 1×10^{13} nv. Evaluation of the resistance of the fueled bodies to irradiation-damage effects will be an important objective of the program. Also of importance will be the evaluation of the specimen-can materials (Inconel 702, Carpenter 20Cb, and Hastelloy X) to corrosion attack by 400-psi GCRE-II reference gas (nitrogen-5 volume per cent oxygen) during the irradiation period (the target exposure period is 5500 hr). This will be done by flowing the gas continuously (but slowly) during the experiment through small-bore tubes welded into each specimen assembly and making appropriate postirradiation examinations of the gas-contacting surfaces. These corrosion-gas-contacting tubes will be 5/16 in. in OD with a 26-mil wall. The design operating temperature for these tubes is 1700 ± 50 F.

Basically, in the capsule design that will be employed for these irradiations, a canned specimen assembly will be housed in a sealed water-contacting stainless steel shell with a narrow inert-gas-filled annulus between. The quantity of heat to be handled is low, being in the neighborhood of 3000 Btu per in. of specimen. Consequently, transfer of heat from the can surface to the shell can be achieved by conduction through the gas which fills the annulus and radiation between the facing walls. Regulation of specimen temperature during irradiation will be achieved by controlling the composition of the annulus gas. Helium and neon (which have a thermal-conductivity ratio of approximately 3-1/2:1) will be the gases utilized. Composition of mixtures of the gases will be altered by external control as the irradiation progresses in accordance with reference temperatures. These will be monitored by thermocouples located adjacent to the corrosion-gas-contacting surfaces.

During the current design phase of this capsule program, a number of leading problems are being studied, both analytically and experimentally. Included here are the following:

(1) Control of heat-transfer-gas mixing and response of specimen temperature to mixing accomplished (a) with individual helium and neon lines leading directly from cylinders to the annular space (over a 25-ft span) and (b) by premixing in a separate chamber.

(2) Axial heat flow in a three-disk stack where heat is being generated only in the top and bottom disks.

(3) Control of corrosion-gas flow at the design pressure of 400 psi (± 5 psi) and at the low flow rates desired.

(4) The magnitude of temperature gradients across the few-mils-thick helium-filled gap between the graphite disks and the inside wall of the can and the effect of these gradients in the over-all heat-transfer and specimen-temperature-level picture. While the machining tolerances for these elements will be close, differential expansion between the graphite and the can will produce the gap referred to at the operating temperature.

(5) Suitable thermocouplry techniques to monitor the temperature of the important corrosion-gas-exposed surfaces.

(6) Flux-perturbation characteristics of the capsule assembly.

Each of these problems (with exception of No. 4) will be investigated in supplementary experiments. Suitable thermal mock-ups, one for heat-transfer studies and one for corrosion-gas-flow studies, have been constructed and initial laboratory data are being obtained. In addition, plans are under way to construct one or more nuclear mock-ups for flux-perturbation studies. Low-power mock-up exposures will be made at the BRR during July. Most of the laboratory work should also be completed during July or early August, with the final capsule-design stage to follow.

Fabrication of UO_2 Pellets

H. D. Sheets and A. G. Allison

The fabrication of enriched UO_2 ceramic pellets for in-pile testing has been completed. The procedure used was as follows:

Q-6

(1) The UO_2 powder received from AGN was crushed to pass a 20-mesh screen and then ball-milled in 100-g lots in a 1-qt natural-rubber-lined mill with Al_2O_3 balls. The 50 per cent enriched powder was ball-milled for 15 hr, the 25 per cent enriched powder for 4 hr.

(2) Three weight per cent of camphor (based on UO_2 weight), dissolved in alcohol, was added to the milled powder. The alcohol was evaporated from the mixture at room temperature.

(3) Pellets were pressed in a hardened steel die at about 20,000 psi. The green densities of the 50 per cent enriched pellets ranged from 5.1 to 5.4 g per cm^3. The 25 per cent enriched UO_2 pellets had green densities of 4.9 to 5.1 g per cm^3. Forming pressure was varied from batch to batch in order to obtain green densities within these ranges.

(4) The pellets were placed on a molybdenum boat and sintered in a hydrogen-atmosphere molybdenum resistance-tube furnace at 3000 F for 1 hr.

About 6500 usable pellets were produced by this method. Yields of acceptable pellets were as follows:

Enrichment, per cent	Outside Diameter, in.	Inside Diameter, in.	Total Length of Pellets Required, in.	Yield of Acceptable Pellets, per cent
25	0.160 ± 0.002	(Solid)	308	93.4
50	0.160 ± 0.002	(Solid)	175	89.5
25	0.160 ± 0.002	0.048 ± 0.004	618	92.5
50	0.160 ± 0.002	0.048 ± 0.004	342	85.1

The sintered densities of the pellets, calculated from weights and dimensions, ranged from 10.0 to 10.4 g per cm^3 (91 to 94 per cent of theoretical).

Random pellets have been chosen for oxygen/uranium ratio determinations, spectrochemical analyses, porosity and surface-area measurements, and microstructure examination.

Q-7

GCRE Critical-Assembly Experiments

R. A. Egen, W. S. Hogan, D. A. Dingee, and J. W. Chastain

Experiments with the GCRE critical assembly were resumed during the month. The fuel elements and reactor components were reassembled to mock up the ML-1 reactor. This configuration resembles the previous Uniform-Radial-Power-Distribution reactor reported in BMI-1306.

GCRE-1 concentric-ring fuel elements were rewrapped to an average fuel loading of 303.4 ± 4 g uranium-235 per element. New fuel-element positioning and support plates were constructed according to a special nonuniform lattice spacing designed to produce a nearly uniform radial power distribution. Sixty-one core positions are available. A 4-in.-thick lead reflector was obtained to completely surround the core region. The lead is 28 in. high and is spaced 1/2 in. from the fuel elements at the core perimeter.

The ML-1 reactor mock-up was critical with 59 fuel elements with approximately 0.4 per cent excess $\Delta k/k$. Following criticality, the worths of critical-assembly control rods and of an additional peripheral fuel element were measured. The relative power distribution was measured using 1/4-in.-wide catcher-foil strips wrapped entirely around each fuel cylinder in a symmetry sector of the core. These data are being analyzed.

During the coming month neutron-flux distribution measurements will be carried out. These measurements will provide data on sub- and epicadmium flux distribution (manganese-wire activation) throughout the core and in detail in and around several fuel elements. It is anticipated that the worth of mock-up ML-1 scissor-action control blades will also be evaluated during the next month.

IN-PILE-LOOP PROGRAM

G. A. Francis

During June, preparation for operation of the ETR in-pile recirculating gas loop was continued. Operator training was completed and the cold check of the system continued. The Battelle Research Reactor loop was put into operation with the irradiation of the 1B-1 αT specimen, an assembly of 19 Inconel-clad UO_2 fuel pins in a concentric-ring arrangement. Activities on the two loop programs are described under the following headings.

Q-8

BRR Loop Program

S. J. Basham and W. H. Goldthwaite

Early in this report period, the 1B-1αT was installed in the loop and significant leakage of the primary coolant was detected. The leakage consisted of a minor leak in the fuel subassembly where thermocouple leads were brazed through the pressure wall. Major leaks were detected near the blower inlet and in the pressure wall between the primary coolant and blower water jacket. Total leakage from the system was approximately 6 standard ft^3 per hr.

Since this leak rate was considered excessive from both experimental and hazard viewpoints, the entire loop assembly was removed from the reactor pool and steps were taken to eliminate leakage. A new blower inlet housing was constructed and installed by welding. The blower cooling-water line was also repaired. Subsequent to these changes, the loop was charged with 200 psig of nitrogen and gas leakage was measured to be 1.1 standard ft^3 per hr.

At this point, the irradiation of the 1B-1αT was started. The specimen and loop performed satisfactorily. The loop modifications have permitted operation of the experiment at design inlet and outlet gas temperatures and design pressure. The element is generating approximately 33 kw at full reactor power of 2 megawatts.

Minor control changes are being made as irradiation continues. Further details on behavior of the experiment will be presented in next month's report.

ETR Loop Program

J. V. Baum, W. J. Stevens, and E. O. Fromm

Activity on this loop program has been confined to the reactor site where Battelle has two cognizant engineers present. The problem of system gas leakage has been solved to the satisfaction of the reactor operator by eliminating minor leaks throughout the system.

During this report period, lectures were given to reactor operating personnel to complete the formal operator training. The cold check was continued. At present, some difficulty is being encountered in checkout of the three loop blowers. The problem of operating centrifugal compression with radial outlets in parallel has caused concern in that an unstable flow condition has been detected. The lack of cooling water at the reactor site made it necessary to suspend operation during the last week of the month. The study of the blower problem will continue when cooling water again becomes available.

Work on the checkout of the removal equipment has not progressed according to plan. There has been a delay of approximately 1 month because of lack of availability of reactor personnel. At present, no specific time has been set for this work.

Q-9 and Q-10

The most recent ETR schedule shows the installation of the BMI-16 in-pile tube to start on July 28. If there is no further change in the insertion schedule, the present lack of reactor personnel and utilities may be the cause of delay on the loop program.

RWD/CRT:all