Report No. BMI-1391 (Rev.)

UC-25 Metallurgy and Ceramics
(TID-4500, 15th Ed.)

Contract No. W-7405-eng-92

PROGRESS RELATING TO CIVILIAN APPLICATIONS DURING OCTOBER, 1959

by

Russell W. Dayton
Clyde R. Tipton, Jr.

November 1, 1959

BATTELLE MEMORIAL INSTITUTE
505 King Avenue
Columbus 1, Ohio

DISCLAIMER

TABLE OF CONTENTS

TABLE OF CONTENTS
(Continued)

REPORTS RELATING TO CIVILIAN APPLICATIONS ISSUED DURING OCTOBER, 1959

BMI-1356 "Core-Temperature Excursions Following a Piping Failure in the Plutonium Recycle Test Reactor", by Alexis W. Lemmon, Jr., Carl A. Alexander, Lewis E. Hulbert, and Robert B. Filbert, Jr.

BMI-1370 "Progress on the Development of Uranium Carbide-Type Fuels", edited by Frank A. Rough and Walston Chubb.

BMI-1381 "Progress Relating to Civilian Applications During September, 1959", by Russell W. Dayton and Clyde R. Tipton, Jr.

A-1

A. ASSISTANCE TO HAPO

F. R. Shober

The creep properties of 15 per cent cold-worked Zircaloy-2, as evident from results being obtained, are superior to those of the annealed Zircaloy-2 at test temperatures of 290, 345, and 400 C. Specimens of cold-worked material in test for periods of time as long as 12,000 hr have shown no severe loss of strength resulting from the long-time annealing at test temperature. The development of a fuel-element leak detector has continued with studies to determine the effect of AgBr concentration and the flow rate of the iodine-131 solution on the exchange between solid AgBr and the iodine. The useful life of the AgBr exchange material is being investigated. Several exchange columns are being constructed to be used in an ETR loop experiment in which ruptured fuel elements will be placed in the loop and the rate of fuel released monitored.

Modification and improvement of a thermal-neutron-flux monitoring system for application to the Hanford reactors has continued. The composition range of ceramic tubes having the desired electrical resistivity, UO_2 content, and fabrication characteristics is being investigated. Search for a metal to form a brazed integral junction between the ceramic and the metal was made, and Kovar was selected. Preliminary tests indicate the electrical characteristics of the junction are reproducible at power levels greater than that required in service. Alloys to be used in a development program for corrosion-resistant welding alloys with Hastelloy F have been melted and fabricated. The niobium and titanium added as stabilizing elements were beneficial for hot working. Heat-treatment studies of the alloys were made. Corrosion studies will be inititated.

Mechanical Properties of Zirconium Alloys

L. P. Rice and J. A. VanEcho

This is a program concerned with the determination of the creep and stress-rupture properties of Zircaloy-2 sheet in both the annealed and cold-worked (15 per cent) conditions at temperatures of 290, 345, and 400 C. All tests are conducted in vacuum. Since test times of from 3000 to 15,000 hr are involved for this alloy, new data for each month are somewhat limited.

Up to the present time, considerable data have been accumulated from regular creep tests, and information from a series of thermal-cycling creep tests that have been in progress on both annealed and cold-worked material is now being obtained.

The most important general conclusions from the data obtained to date are as follows:

(1) Creep tests of annealed Zircaloy sheet show that, at 290 and 345 C, very little creep occurs even at loads approaching the short-time rupture strength. Load-on deformation of over 4 per cent may occur followed by a creep rate of 0.0001 per cent per hr. At 400 C, creep deformation becomes significant.

(2) Cold-worked Zircaloy-2 is considerably stronger than the annealed material at all temperatures, and, as might be expected, the prior cold work results in much less deformation on loading. For equivalent stresses, the average creep rate of the cold-worked Zircaloy is about one-tenth that of the annealed material at temperatures of 290, 345, and 400 C.

(3) Cycling Zircaloy-2 between room temperature and 290 and 345 C once each week during creep testing results in more creep deformation than is produced in noncycled tests at equivalent stresses. The effect is more pronounced at 345 C, and appears to be stress dependent at this temperature but not at 290 C.

A number of tests are still in progress, including long-term ordinary creep tests as well as cyclic-temperature creep tests which will have a duration of about 3000 hr.

Development of a Fuel-Element Leak Detector

J. E. Howes, Jr., T. S. Elleman, and D. N. Sunderman

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

During the past month, the exchange between solid AgBr and iodine-131 in solution was studied as a function of AgBr concentration and flow rate through the AgBr bed. In these experiments, iodine-131 solution was passed through 1-in.-high by 1-in.-diameter and 1-in.-high by 2-in.-diameter columns of 16 to 50-mesh fused AgBr at flow rates of 0.2, 0.5, 1.0, and 1.5 gal per min. Exchange of the iodine-131 with the bromide in AgBr was measured by removing the AgBr from the column and radioassaying it in a scintillation counter.

The efficiency of removal of iodine-131 from the stream under the various column size-flow conditions was found to vary from about 40 to 60 per cent.

Four columns containing AgBr have been sent to Hanford for attachment to an effluent reactor-coolant stream. These columns will be monitored periodically to assist Battelle in determining the buildup of fission products on the bed and the lifetime of the AgBr exchange material.

Several exchange columns are presently being constructed for use in the ETR loop experiment. In this study, ruptured fuel elements will be placed in the loop and the rate of fuel release to the coolant will be monitored by exchange of the halide fission products with bromide in the AgBr columns. The suitability of AgBr columns for leak detection will be evaluated under these conditions.

Studies are in progress to determine gross fission-product contamination as a function of flow rate and AgBr column size. The data from these studies and the exchange studies will permit the selection of the optimum column size and flow conditions for the leak-detection application.

A-3

Thermal-Neutron-Flux Monitoring System

P. M. Steinback, J. W. Lennon, M. J. Snyder, and D. R. Grieser

A thermal-neutron-flux measuring device was previously developed at Battelle. This device is currently being modified and improved for use in a thermal-neutron-flux monitoring system being developed for Hanford reactors.

Operation of the device is based on the measurement of the electrical power required to maintain the temperature of a ceramic tube, containing UO_2 (depleted), $MoSi_2$, and Al_2O_3, at the temperature of a similar ceramic tube which is heated by uranium-235 fission in enriched UO_2. Electric power, then, can be directly correlated with thermal-neutron flux.

Investigations were continued to determine the composition range in which ceramic tubes having the desired electrical resistivity and UO_2 content could be fabricated by extrusion. Compositions in the following range were found satisfactory on the basis of tests to date:

Material	Amount, w/o
$MoSi_2$	47.0-49.0
Al_2O_3	35.2-33.4
UO_2	17.8-17.6

In an attempt to decrease the initially high contact resistance on the ceramic surface, a mixture of finely divided $MoSi_2$ and graphite was painted on the ends of the ceramic tubes before they were fired. On firing, a highly conductive film was formed on the ends which appeared to be bonded to the ceramic fairly well.

A parallel study on a brazed integral junction has led to the choice of Kovar as the junction metal. Its use in the form of a narrow sleeve brazed to the circumference at each end of the ceramic tube should provide a junction which is (1) mechanically stronger, (2) electrically larger, (3) capable of a more uniform distribution of excess heat developed at the junctions, reducing thermal stresses in the ceramic, and (4) more nearly matched to the ceramic so far as thermal expansion is concerned. Although this junction has not yet been thoroughly studied, it has shown, in preliminary tests, reproducible electrical characteristics at power levels from two to ten times greater than required (up to 200 w).

In the coming month, variations of the junction described above, possibly combined with the ceramic tubes tipped with $MoSi_2$ and graphite, will be tested for long-term electrical and mechanical stability when cycled over the anticipated design power and temperature range.

Other promising junction designs will be investigated. Over-all design parameters will receive continued consideration, leading toward fabrication of the first prototype probe and circuitry.

A-4

As part of the continued study of raw-material variations in the ceramic, a method is being sought for characterizing various samples of UO_2. In the initial study of these methods, the sintering characteristics of several UO_2 samples are being determined. One of the samples contains 93 per cent enriched UO_2 and will be used in extruding the fission-heated ceramic tubes.

Development of Corrosion-Resistant Welding Alloys for Use With Hastelloy F to Contain Decladding Solutions

M. E. Langston, R. E. Monroe, C. L. Peterson, and W. K. Boyd

The objective of this program is to develop corrosion-resistant welding alloys for use with vacuum-melted low-carbon Hastelloy F when used as a container material for spent fuel-element decladding solutions. The current phase of the program is concerned with the preparation and the evaluation of forming, welding, and corrosion resistance of 12 experimental nickel-base alloys.

Vacuum-induction melting of 15-lb heats and subsequent fabrication of ingots to 0.125-in.-thick annealed strips were completed. Preliminary studies conducted on pilot ingots of the 12 alloys led to the selection of temperatures of 2000 to 2100 F for forging and 2100 F for hot rolling. As indicated by the observations given in Table A-1, the forging and hot-rolling behavior of the titanium-containing alloys was superior to that of the niobium-containing alloys. The poorest formability was demonstrated by the alloy containing only molybdenum and copper additions to the base composition (10B), which suggests that the stabilizing elements, niobium and titanium, were beneficial to hot working.

Studies were made on specimens of hot-rolled strip to determine the optimum solution-annealing temperature for each of the 12 alloys. The object of this study was to find a suitable annealing temperature for each alloy which would result in the solution of minor phases without excessive grain growth and oxidation. On the basis of the hardness data given in Table A-2, as well as a metallographic examination of the alloy structures, three annealing temperatures were chosen: 1950 F for Alloys 5B and 6B; 2050 F for Alloys 3B, 4B, 9B, 10B, 11B, and 12B; and 2150 F for Alloys 1B, 2B, 7B, and 8B. The hot-rolled strip was annealed for 1 hr at the appropriate temperature and water quenched.

Corrosion evaluations of these alloys, both by themselves and as welding materials for vacuum-melted low-carbon Hastelloy F, will be started. Welded and unwelded coupons will be evaluated by exposing some of each material for five 24-hr periods to boiling Niflex (1M HNO_3, 2M HF) and others to boiling Sulfex (3.5 M H_2SO_4 in which 20 g per liter of Type 304L stainless steel is dissolved) solutions.

A-5 and A-6

TABLE A-1. EFFECT OF COMPOSITION ON FORMING AND ROLLING BEHAVIOR OF VACUUM-MELTED NICKEL-CHROMIUM-IRON EXPERIMENTAL WELDING ALLOYS CONTAINING VARIOUS ADDITIONS OF COPPER, MOLYBDENUM, NIOBIUM, AND TITANIUM

Alloy	Addition[a], w/o				Relative Formability	
	Mo	Cu	Nb	Ti	Forging[b]	Hot Rolling[c]
1B	6	1	2	--	Good	Good
2B	6	2	2	--	Fair	Fair
3B	6	-	-	0.5	Very good	Good
4B	6	-	-	1	Ditto	Very good
5B	3	2	-	0.5	Ditto	Very good
6B	3	2	-	1	Ditto	Good
7B	6	3	2	--	Fair	Fair
8B	9	1	2	--	Fair	Fair
9B	3	1	2	--	Fair	Good
10B	6	2	-	--	Poor	Poor
11B	6	2	-	1	Very good	Very good
12B	3	2	2	--	Good	Very good

(a) Base composition (w/o): 45 nickel-22 chromium-0.02 carbon 0.6 manganese-0.4 silicon-20 to 26.5 iron.

(b) Soaked 1 hr at 2000 to 2100 F prior to forging.

(c) Reheated to 2100 F after each roll pass.

TABLE A-2. EFFECT OF ANNEALING TEMPERATURE ON HARDNESS OF HOT-ROLLED EXPERIMENTAL WELDING ALLOYS

Alloy	Addition[a], w/o				Rockwell A Hardness							Optimum Annealing Temperature, F
					Hot	After Annealing at Temperature Shown[b]						
	Mo	Cu	Nb	Ti	Rolled	1700 F	1800 F	1900 F	2000 F	2100 F	2200 F	
1B	6	1	2	--	63.0	60.5	57.0	54.5	50.0	49.5	48.0	2150
2B	6	2	2	--	63.5	59.5	56.5	54.5	51.0	50.0	48.0	2150
3B	6	-	-	0.5	59.0	53.5	48.5	49.5	48.0	46.5	44.5	2050
4B	6	-	-	1	60.0	56.5	50.0	49.5	49.0	46.5	45.0	2050
5B	3	2	-	0.5	58.5	47.0	45.5	46.0	44.0	43.5	41.0	1950
6B	3	2	-	1	58.5	46.5	46.0	47.0	45.0	44.0	41.5	1950
7B	6	3	2	--	64.0	60.0	57.0	57.0	50.5	49.5	47.0	2150
8B	9	1	2	--	67.5	62.5	61.0	60.0	54.5	52.5	50.5	2150
9B	3	1	2	--	60.0	54.5	50.0	50.5	49.5	47.5	45.0	2050
10B	6	2	-	--	62.5	51.0	49.5	49.0	47.0	46.0	45.0	2050
11B	6	2	-	1	57.5	56.0	49.5	49.5	48.0	46.0	44.5	2050
12B	3	2	2	--	59.5	51.0	50.5	49.5	47.5	46.5	44.5	2050

(a) Base composition (w/o): 45 nickel-22 chromium-0.02 carbon-0.6 manganese-0.4 silicon-20 to 26.5 iron.

(b) Annealed 30 min at indicated temperature and then air cooled.

B-1

B. DEVELOPMENTS FOR ALUMINUM-CLAD FUEL ELEMENTS

N. E. Daniel and R. J. Carlson

Aluminum-uranium alloys containing 0.5 to 3.0 w/o tin or zirconium are being evaluated for possible use as reactor fuels. Metallographic examination of the alloys containing tin indicated that the extent of the UAl_3-to-UAl_4 transformation is inversely proportional to the tin content of the alloys. Those alloys containing up to 2.0 w/o zirconium exhibited no microstructural changes that could be attributed to the ternary additions. The alloy containing 3 w/o zirconium exhibited a marked change in microstructure.

Preparation of Aluminum-Uranium Alloys

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

Aluminum-uranium alloys containing up to 20 w/o uranium have found wide-spread acceptance as fuels for low-temperature water-cooled water-moderated reactors. For certain applications, aluminum-uranium alloys containing up to 35 w/o uranium are desired in the form of tubular fuel elements. Previous studies have indicated that casting and fabricating difficulties may be alleviated by the judicious use of ternary additions such as tin or zirconium. The improved casting characteristics are believed to be the result of the increased fluidity of the alloys containing these additions; whereas, the improved fabricating characteristics are attributed to the retention of UAl_3 in the cast shapes, thereby increasing the proportion of the ductile aluminum-rich matrix.

Although it is known that tin and zirconium will inhibit the UAl_3-UAl_4 reaction, the effects of these elements on the properties of the aluminum-35 w/o uranium alloys have not been investigated fully. For this purpose two series of aluminum-35 w/o uranium alloys containing from 0.5 to 3.0 w/o tin or zirconium were air melted and cast into 3-in.-diameter by 10-in.-long ingots. After casting, 1/2-in.-thick slices were removed from the bottom and from the section immediately below the primary pipe of each casting for evaluation. Hardness tests and radiographic and metallographic examinations were made. The radiographic examinations revealed that the initial air-melted alloys containing zirconium were extremely porous. However, when the five alloys (0.5, 1.0, 1.5, 2.0, and 3.0 w/o zirconium in aluminum-35 w/o uranium) were remelted in air, a decrease in porosity was noted. In addition, some alloys containing zirconium were vacuum melted. They were found to be sound and showed little or no porosity. All of the alloys containing tin exhibited negligible porosity.

The metallographic examination of the alloys containing less than 3.0 w/o tin revealed well-defined reaction areas on the periphery of the primary compound particles. The extent of this reaction zone appears to be inversely proportional to the tin content of the alloy. The 3.0 w/o tin alloy exhibited little or no reaction zone surrounding the particles.

B-2

The metallographic examination of the zirconium alloys revealed very little difference in the compositions containing 0.5 to 2.0 w/o zirconium. However, the 3.0 w/o zirconium alloy exhibited a marked change in microstructure as compared to the former alloys. The 3 w/o zirconium alloy contained a very small quantity of compound in the matrix of aluminum. Further metallographic studies of the as-cast, extruded, and heat-treated specimens are in progress. Future work will be concerned with obtaining the tensile strength, the creep and stress-rupture properties, and the corrosion resistance of the extruded material. Casting studies preparatory to making enriched castings of the alloys in the form of hollow cylindrical extrusion billets will be initiated during the next month.

C-1

C. RADIOISOTOPE AND RADIATION APPLICATIONS

D. N. Sunderman

The four research programs undertaken for the Office of Isotopes Development on radioisotope applications and radiation chemistry are continuing. These programs consist of the application of radiotracer techniques to industrial quality and process control and the use of ionizing radiation in graft polymerization and the nitration of hydrocarbons.

Industrial quality-control methods utilizing radiotracer techniques are under development for the cement industry. Radiometric analyses for iron and aluminum appear very promising. These methods are titrimetric, employing EDTA, with a radioactive insoluble salt as endpoint indicator. In the case of iron, $Y_2^{91}(C_2O_4)_3$ is used, while $Ag^{110}IO_3$ is used for aluminum. Preliminary experiments indicate that pH is the most significant process variable, and the study of its effect is continuing.

The application of intrinsic tracers to the control of the iron-removal process is progressing. Experimental studies of radioassay efficiency indicate that concentrations of iron-59 will be well below maximum permissible concentrations as specified in NBS Handbook 69. The handbook values do not refer to concentrations allowed in a consumer product, but are the basis for such concentration values which are presently under consideration by the licensing branch of the AEC. Work is continuing toward an engineering demonstration of the applicability of this technique.

Radiation-induced graft polymerization of polymethylmethacrylate and vinylpyrrolidone is under study. Electron paramagnetic resonance is used to evaluate the production of reactive sites. Differences between EPR spectra of PMMA obtained on this program and those published in the literature have been shown to be the result of the presence of monomer in material studied by previous investigators. Equipment has been developed which will allow the irradiation of this material in vacuum and its EPR spectrum determined. Other methacrylates have been prepared, polymerized, and deactivated, and will be studied in the coming month.

Eight experimental runs were made to evaluate the effect of phase on radiation-induced nitration of cyclohexane. The temperature range from 60 to 140 C and radiation dose range from 4 to 17 x 10^6 rads were covered in these experiments. The materials are being analyzed by gas chromatographic and infrared spectrometric techniques. Identification work on the previously reported polynitro compound is continuing.

Development of Radioactive-Tracer Quality-Control Systems

C. W. Townley, C. T. Brown, and D. N. Sunderman

Additional activation analyses have been performed on three samples of cement raw materials. Analyses for manganese and sodium were completed. Analysis for sodium produced by the $Al^{27}(n,\alpha)Na^{24}$ reaction in the aluminum metal standard was also performed in order to determine the fast flux. In this manner it is possible to correct the sodium results in the unknown samples due to the (n,α) reaction on the aluminum present in each of the samples.

C-2

In each case the results were corrected for thermal-flux variations by arranging the samples in a fixed bundle and placing dosimeter wires on the outer and inner surfaces of each sample tube in the bundle. The flux dose range was 8.7×10^{11} to 1.1×10^{12} nvt. Corrections for sodium-24 produced by the (n,α) reaction on aluminum-27 only amounted to 0.001 mg Na_2O produced per mg of Al_2O_3 present as 0.1 w/o. The corrected results are shown in Table C-1.

TABLE C-1. ACTIVATION ANALYSIS OF CEMENT RAW MATERIALS FOR MANGANESE AND SODIUM

Sample	Na_2 Content, w/o		Mn_2O_3 Content, w/o	
	Calculated	Experimental	Calculated	Experimental
Slag (19459)	0.12	0.20	0.23	0.25
Slag (19466)	0.14	0.31	1.61	1.33
Slag (19469)	0.11	0.23	0.31	0.24

Work on the radiometric analysis of iron and aluminum in portland cement continued during this report period. The optimum conditions have been investigated for the EDTA titration of aluminum using $Ag^{110}IO_3$ as an endpoint indicator and for the EDTA titration of iron using $Y_2^{91}(C_2O_4)_3$ as an indicator.

The work thus far has been carried out with the iron and aluminum in separate solutions. In the case of the aluminum titration, the best results have been obtained with the pH maintained at 8 or more. Tartaric acid has been used to prevent the coagulation of the aluminum at this pH. It has been found that below a pH of 8 the formation of the EDTA complex of silver is not sufficient to indicate the endpoint of the titration. Heating the solution to 45 to 50 C does not improve the results when the pH is maintained at 8 or more. The error in the results has ranged from 1 to 2 per cent.

The EDTA titration of iron must be performed at a pH below 7 because the iron complex hydrolyzes in basic solution. Trials made thus far at a pH ranging from 3 to 4 have been unsuccessful due to a large fluctuation in the amount of yttrium-91 in solution before the endpoint is reached. This fluctuation has been consistent with the fluctuation in pH.

During the next report period the study of the conditions for the titrations of iron and aluminum will be continued. In the case of the iron determination the pH will have to be maintained more constant, and in the aluminum titrations the errors will have to be reduced. When the conditions for the two titrations are determined, the titrations will be attempted with the iron and aluminum in the same solution.

One or two more activation analyses will be performed in order to determine aluminum and calcium in cement raw materials. The effect of the $Fe^{56}(n,p)Mn^{56}$, $Mg^{24}(n,p)Na^{24}$, and the $Si^{28}(n,p)Al^{28}$ reactions will also be determined by irradiating samples of iron, magnesium, and silicon of predetermined purity.

C-3

Use of Intrinsic Radioactive Tracers for Process Control

J. L. McFarling, J. F. Kircher, and D. N. Sunderman

Current work on the intrinsic-radiotracer process-control application has dealt with the problem of iron removal from refinery streams. The experimental loops described in the report for September have been used to study the effect of various process parameters on the detection system. A study has been made of the relationship between pH of solution and iron-59 activity as a direct measure of iron precipitation. A preliminary summary of radioisotope requirements and activity levels to be expected in various phases of the process was assembled and compared with the accepted continuous exposure levels for iron-59.

Process-solution temperature variations might introduce errors in the observed counting rate from the process-stream detector. Cold-water coils are used with the detection system to cool both the photomultiplier tube and NaI(Tl) crystal. Experimental results showing the variation of the background count of the detection system over a temperature range from 40 to 86 C are presented in Table C-2. This is the temperature range expected in the process under study. These measurements indicate that no appreciable temperature effects are present. Other measurements of added iron-59 radioactivity also indicated no significant effect of solution temperature or counting rate.

TABLE C-2. TEMPERATURE DEPENDENCE OF DIP-COUNTER DETECTION SYSTEM

Elapsed Time, min	Solution Temperature, C	Crystal Temperature, C	Background Count[(a)], cpm ± σ
0	44	30	452 ± 6.7
30	49	31	457 ± 6.7
60	55	32.5	446 ± 6.7
90	66	34.5	452 ± 6.7
120	73	36	457 ± 6.7
150	76	38	454 ± 6.7
180	82	39	451 ± 6.7
210	86	40	448 ± 6.7
230	54	34	443 ± 6.7
260	40	31	449 ± 6.7

(a) Average count ± σ_x = 451 ± 4.3.

C-4

Other loop experiments have been conducted to study the relationship between iron-59 tracer concentration and observed counting rate. This should be a linear relationship, and the preliminary data shown in Table C-3 demonstrate this. The range of iron-59 concentration used is approximately that to be expected in an actual process-control application. The average efficiency of iron-59 radioassay in this experimental system, defined as $\frac{\text{cpm}}{\text{dpm/liter}} \times 100$, is shown to be 17.15 ± 0.35 per cent.

TABLE C-3. OBSERVED COUNTING RATE AS A FUNCTION OF TRACER CONCENTRATION

Iron-59 Activity Added[a], dpm/liter	Total Iron-59 Activity in Solution[a], dpm/liter	Gross Counting Rate, cpm ± σ	Net Counting Rate[b], cpm ± σ	Radioassay Efficiency[a], $\frac{\text{cpm}}{\text{dpm/liter}} \times 100$
905	905	603 ± 7.75	153 ± 10.5	16.9
905	1810	765 ± 8.75	315 ± 11.1	17.4
915	2715	929 ± 9.65	479 ± 11.7	17.5
915	3630	1060 ± 10.6	610 ± 12.3	16.8
915	4545	1237 ± 11.3	787 ± 13.0	17.3
915	5460	1395 ± 11.8	945 ± 13.6	17.0

(a) Iron-59 is assumed to emit 1γ per disintegration in these calculations.
(b) cpm $\pm\sigma_X$ = gross count − background $\pm \sqrt{\sigma_T^2 + \sigma_B^2}$.

Laboratory studies were undertaken to establish the relationship between solution pH and dissolved iron by measuring the relative activity of iron-59 in solution. The results of an experimental run carried out at 50 C are shown in Table C-4. The purpose of the precipitation experiments is to demonstrate that the reaction can be followed in this manner and to establish a basis of comparison for testing substitute tracers.

TABLE C-4. IRON-59 SOLUTION RADIOACTIVITY AS A FUNCTION OF pH WITH AN INITIAL IRON CONCENTRATION OF 0.75 G PER LITER AT 50 C

pH of Solution	Gross Activity in Solution, cpm	Net Activity in Solution, cpm $\pm\sigma_X$	Precipitation, per cent
2.1	5730	5155 ± 25	0
2.5			
2.75	3640	3065 ± 21	40.5
2.90	1605	1030 ± 15	80.0
3.14	930	355 ± 12	93.1 ± 0.2
3.41	785	210 ± 12	95.9 ± 0.2
3.82	730	155 ± 11	97.0 ± 0.2
5.10	645	70 ± 11	98.65 ± 0.2
7.0	590	15 ± 11	99.71 ± 0.2

C-5

Experimental work with the circulating loops, although preliminary in nature, indicates that the simple detection system using a water-cooled dip counter would be reliable and sensitive enough for some industrial applications. More sensitive detection instruments would be desirable to achieve greater accuracy and smaller tracer requirements. Recent discussions with W. H. Johnston Laboratory personnel indicate that they are developing detection instruments for the AEC Office of Isotopes Development which should be useful for these applications. Such instruments would have higher sensitivity than the present detection system.

Table C-5 summarizes the accuracy of radioassay for different counting times and efficiencies. This summary is for an iron-removal operation which effects a 100:1 reduction in iron concentration. The 30 per cent efficiency in the table could be achieved with the present detection system by using a counting volume of about 30 liters. The high efficiencies might well be attainable by instruments currently under development. The per cent accuracy of measurement, at the 95 per cent confidence level, is calculated from the following formula:

$$A = \pm\left[\frac{2 \quad \text{total count + background count}}{\text{total count - background count}} \times \frac{100}{\text{counting time in min}}\right].$$

The background of 600 cpm is the approximate background expected with the proposed system.

The radioisotope requirements and tracer activity levels to be expected in a continuously operating process have been defined. These data, which are applicable to the present process, are shown in Table C-6. Reference to Table C-4 will give the accuracy of measurement at these particular activity levels. This example assumes the following process parameters:

Flow rate	5 gal per min
Iron reduction ratio at precipitator	100:1
Over-all iron reduction (due to iron hydroxide recycle)	25:1
Iron content of filter cake	15 w/o
Iron concentration in iron-rich solution	0.80 g per liter
Iron concentration in iron-free solution	0.008 g per liter
Cobalt concentration in iron-free solution	30 g per liter
Tailings discarded per day	210 tons

It is believed that the levels of tracer injection covered in this example are in the range of practical interest for process-control schemes. A comparison of these activity levels with the maximum permissible concentrations of iron-59 can be made by referring to Table C-7. It is evident that the highest activity level present in the process is in the iron hydroxide filter cake. This level is equivalent to the maximum permissible concentration for an exposure of 40 hr per week. Actual exposure at this maximum level would be only a few hours a week. The enclosed, dust-free nature of the process together with the low tracer volatility precludes excessive air-borne activity. The product activity levels shown in Table C-6 are also well below the level of 1.4×10^{-4} μc per g suggested by the AEC as a permissible concentration of iron-59 in product material. It would appear, therefore, that the proposed application involves no large-scale contamination hazards.

TABLE C-5. ACCURACY OF MEASURING RADIOACTIVE IRON-59 SOLUTIONS AS A FUNCTION OF TRACER CONCENTRATION, DETECTION EFFICIENCY, AND COUNTING TIME

Iron-59 Detection Efficiency in Iron-Free Solution, $\frac{\text{cpm}}{\text{dpm/liter}} \times 100$	Iron-59 Activity Injected Into Iron-Containing Solution, 10^{-7} μc per ml	Iron-59 Activity Remaining in Iron-Free Solution		Counting Rate Above Background From Iron-Free Solution, cpm	Statistical Accuracy of Radioassay With 600-CPM Background (At 95 Per Cent Confidence Level), per cent		
		10^{-7} μc per Ml	Dpm/Liter		1-Min Count	5-Min Count	10-Min Count
30	2.25	2.25	500	150	49	22	15.5
	4.5	4.5	1000	300	26	12.9	8.2
	9	9	2000	600	14.2	6.4	4.5
	18	18	4000	1200	8.2	3.7	2.6
100	1.13	1.13	250	250	30.5	13.6	9.7
	2.25	2.25	500	500	16.6	7.4	5.2
	4.5	4.5	1000	1000	9.4	4.2	3.0
200	0.57	0.57	125	250	30.5	13.6	9.7
	1.13	1.13	250	500	16.6	7.4	5.2
	2.25	2.25	500	1000	9.4	4.2	3.0

TABLE C-6. RADIOISOTOPE REQUIREMENTS AND ACTIVITY LEVELS IN INDUSTRIAL IRON-REMOVAL PROCESS FOR DIFFERENT AMOUNTS OF INJECTED TRACER

Approximate Iron-59 Activity Required Per Week, mc	Tracer Activity Level in Iron-Containing Solution, 10^{-5} μc per ml	Tracer Activity Level in Iron-Free Solution, 10^{-7} μc per ml	Tracer Activity Level in Iron Hydroxide Filter Cake, 10^{-3} μc per g	Tracer Activity Level in Iron Hydroxide Recycle Slurry, 10^{-6} μc per ml	Maximum Tracer Activity Remaining in Metal (Cobalt) Product, 10^{-6} μc per g	Maximum Tracer Activity Discharged to Tailings, 10^{-7} μc per g
0.33	0.57	0.57	1	0.5	1.8	2
0.65	1.13	1.13	2	1	3.5	4
1.25	2.25	2.25	4	2	7	8
2.5	4.5	4.5	8	4	14	16
5	9	9	16	8	28	32
10	18	18	32	16	56	64

TABLE C-7. MAXIMUM PERMISSIBLE CONCENTRATIONS OF IRON-59 FOR CONTINUOUS OCCUPATIONAL EXPOSURE(a)

Solubility of Radioisotope	Organ of Reference	Maximum Permissible Concentrations, μc per ml			
		For 40-Hr Week		For 168-Hr Week	
		In Water	In Air	In Water	In Air
Soluble	GI tract	2×10^{-3}	4×10^{-7}	6×10^{-4}	1×10^{-7}
	Spleen	4×10^{-3}	1×10^{-7}	1×10^{-3}	5×10^{-8}
	Total body	5×10^{-3}	2×10^{-7}	2×10^{-3}	7×10^{-8}
	Liver	6×10^{-3}	2×10^{-7}	2×10^{-3}	7×10^{-8}
	Lung	0.02	8×10^{-7}	7×10^{-3}	3×10^{-7}
	Bone	0.03	1×10^{-6}	0.01	4×10^{-7}
Insoluble	Lung	--	5×10^{-8}	--	2×10^{-8}
	GI tract	2×10^{-3}	3×10^{-7}	5×10^{-4}	9×10^{-8}

(a) NBS Handbook 69, pages 30, 31.

Further loop experiments are planned for the purpose of confirming the preliminary data reported here as well as to determine effects of other process parameters. Several radioisotopes have been obtained which will be tested as iron-59 substitutes during the coming month. A preliminary design study to scale up the laboratory experiments to a continuously operating system will also begin.

Graft-Polymerization Studies

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

It was reported in BMI-1381 that irradiated pure polymethylmethacrylate (PMMA) does not produce the electron paramagnetic resonance (EPR) spectrum which has often been published in the literature. Further purification of the polymer by multiple reprecipitation resulted in no spectrum change. However, commercial PMMA and PMMA contaminated with methylmethacrylate have been found upon irradiation to produce the so-called typical spectrum. The effect of monomer contamination was also investigated by the repetition of the work described above using polyethylmethacrylate. Once again the pure irradiated polymer did not produce the spectrum reported in the literature. The addition of a fraction of a per cent of monomer to the polymer changed the spectrum, after irradiation, from a relatively simple structure to the complex form found by many investigators. These two experiments indicate that the spectra reported for polymethyl- and polyethylmethacrylate were due to contamination with monomer.

In order to further explore this phenomenon a piece of apparatus was built which will permit a polymer sample to be irradiated in vacuum and its EPR spectrum determined. After the addition of monomer to the irradiated sample in a vacuum a second spectrum will be run. This procedure may elucidate the interaction between monomer and polymer which produces the complex spectra in commercial and contaminated methacrylates.

In addition to the study reported above, preparations were made to work with other methacrylates. Polypropyl, polyisopropyl, polybutyl, and polytertiary butylmethacrylates were prepared by irradiation polymerization. These methacrylates will also be radiation grafted with vinylpyrrolidone.

During the coming month work will proceed as indicated above.

Nitration of Hydrocarbons

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

During October eight thermal and irradiation runs with the nitric acid-cyclohexane system were completed. A 10-to-1 mole ratio of hydrocarbon to nitric acid was used in these runs. In the irradiation work runs were made at temperatures from 60 to 140 C. Reaction times were from 20 to about 117 hr, corresponding to doses of 4.0×10^6 to 1.7×10^7 rads. Thermal runs in the absence of radiation were also carried out at each temperature.

C-10

In a study of the effect of phase on the nitration reactions, several of the runs were made in both the liquid and vapor phases. These runs were made at identical temperatures and charge compositions. Only the total charge mass was changed. The charge mass was varied such that at a given temperature the charge was either essentially all vaporized or all in the liquid phase.

Samples from the eight runs are being analyzed by gas-chromatography and infrared techniques. Results will be reported next month.

Work on the identification of the polynitro compound is continuing. Infrared, gas chromatography, elemental analysis, and molecular-weight determinations will be made when a sufficient amount of this compound can be isolated.

During November, thermal and irradiation runs will be continued.

D-1 and D-2

D. VARIABLE-MODERATOR REACTOR CRITICAL-ASSEMBLY STUDIES

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

As part of the program to develop boiling-water power reactors, the Advanced Technology Laboratories (ATL) of American-Standard Corporation is engaged in a long-range program to develop a method of control based on varying the moderator level in a boiling-water power reactor. The system is termed the Variable Moderator Reactor (VMR). As a part of this program, ATL is preparing a digital computer code, PUREE, for use in analyzing and designing the VMR. Critical-assembly studies are planned to verify the assumptions and methods applied in the development of this code.

During the past month, the critical-assembly program was concerned with designing critical-assembly components and selecting experimental conditions.

Advanced Technology Laboratories and Battelle personnel have been working together to develop the program. The conceptual designs have been completed on major components, and final design work has been initiated. A proposed experimental program was drawn up and submitted to both BMI and ATL personnel for further consideration.

The preparation of a Procedures Manual on Fuel Handling and a Hazards Summary Report will be the major tasks during November. Construction will begin on the reactor components associated with the core structure. The experimental program will be worked out in more detail.

F-1

F. RESEARCH FOR AEC REACTOR DEVELOPMENT DIVISION PROGRAM

S. J. Paprocki and R. F. Dickerson

REACTOR MATERIALS AND COMPONENTS

R. F. Dickerson

Since prior studies have shown that UO_2 can be stabilized to prevent oxidation and volatilization in high-temperature air by binary additions of La_2O_3 or Y_2O_3, research has been in progress to determine whether or not ternary additions can be used to reduce the total additive required for stabilization. A divalent oxide, CaO, has been used as the ternary addition. The choice of this addition was based on the valence-compensation theory. Test results indicate that the over-all amount of additive can be reduced and that the ternary additions also result in significant reduction in thermal-neutron cross section. The investigation of the effects of ultrahigh pressure and high temperature on U_3O_8 is being continued, as has been the study of the UO_2-Sc_2O_3 reactions under similar conditions.

The irradiation of 11 capsules containing Type 347 stainless steel specimens has been continued in the ETR. As of October 1, 1959, some of the capsules had been exposed to a total of over 3.6×10^{21} nvt (fast). The series of gamma-heat capsules has not been inserted in the ETR because of space shortage, and negotiations with commercial test-reactor facilities are in progress.

Niobium-base alloys which are being considered as potential cladding material for the EBR exhibit exceptional strength at 800 C (1470 F). As an example, both a niobium-1.84 w/o chromium and a niobium-4.33 w/o zirconium alloy have yield strengths of about 69,000 psi at 800 C. (Type 347 stainless steel has a yield strength of about 20,000 psi at 800 C.) These alloys have been successfully fabricated. The evaluation of selected niobium-base alloys for use as cladding in pressurized-water reactors has shown that a binary alloy containing 12.6 a/o vanadium has a high degree of corrosion resistance in hot water and steam and has more than adequate creep strength at the temperatures of interest. Further screening of additional alloys is in progress.

The design of the apparatus needed to stress Zircaloy-2 in an irradiation field at 600 to 700 F is in progress. The test capsule will now accommodate flat test specimens, and loading will be accomplished by attaching the specimen ends to bellows actuated by gas pressure. The Zircaloy-2 base material for use in the study of strain aging has been prepared. Preliminary internal-friction measurements have been made, and the data obtained indicate that strain aging may occur. Other work concerned with the determination of the presence or absence of strain aging is in progress. Electron-transmission studies of thin films of zirconium and Zircaloy-2 are in progress, and initial results indicate that this technique has promise as a means of studying dislocation networks on prismatic planes.

F-2

Valence Effects of Oxide Additions to Uranium Dioxide

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

An investigation is being conducted on the effects of oxide additions to uranium oxide, relative to stabilization toward oxidation and volatilization. Previous research has shown that stable oxide materials may be obtained by the incorporation of La_2O_3 or Y_2O_3 into solid solution with uranium oxide. An operative mechanism based upon the concept of valence compensation has been proposed for the stabilizing influence of such additions.

Current work has been directed toward a reduction of the amount of additive required for stable solid solutions. Using the valence-compensation hypothesis, a partial substitution of a divalent oxide, CaO, for La_2O_3 or Y_2O_3 has been employed for this purpose. Solid solutions of UO_2-La_2O_3-CaO and UO_2-Y_2O_3-CaO were fabricated and air oxidized for periods up to 20 hr at 1750 C. The intended composition and weight-change data are shown in Table F-1. Chemical analyses, before and after air oxidation, are given in Table F-2.

The results contained in Tables F-1 and F-2, together with earlier results, show that the solid solutions containing the larger amount of additive, corresponding more nearly to the completely compensated condition, are competitive in stability to the binary systems evaluated previously. Thus, it appears feasible to reduce the amount of additive required for stabilization by use of the valence-compensation principle. The incorporation of divalent additives, in partial substitution for trivalent additions, also results in a significant reduction in neutron cross section.

The electrical characteristics of the ternary stabilized oxides are currently being studied. Fabrication of fully oxidized solid solutions of U_3O_8 containing 40, 50, and 60 mole per cent La_2O_3 is under way. These samples are being prepared to complete the study of the electrical characteristics of this binary system and also to evaluate the thermal conductivity of the 50 mole per cent composition.

High-Pressure High-Temperature Solid-State Studies

W. B. Wilson and C. M. Schwartz

An exploratory investigation is being conducted on the effects of ultrahigh pressure and high temperature on the uranium-oxygen system and on reactions of uranium oxides with certain other oxides. Work was continued on the investigation of the effect of pressure on U_3O_8 at pressures to 100,000 atm with temperatures to 1600 C. Study of the reaction of UO_2 with Sc_2O_3 was continued. Results will be reported when available.

TABLE F-1. SUMMARY OF WEIGHT CHANGES THAT OCCURRED IN SINTERED TERNARY OXIDE COMPACTS DURING AIR FIRING AT 1750 C

Identification	Intended Composition, mole per cent				Weight Loss[a] After Heating at 1750 C in Dry Flowing Tank Air for Indicated Time, per cent		
	UO_2	La_2O_3	Y_2O_3	CaO	1 Hr	5 Hr	20 Hr
14795-67	50.0	25.0	--	25.0	+1.67	+1.35	+0.10
14795-75	60.0	20.0	--	20.0	0.03	1.58	5.96
14795-83	60.0	--	20.0	20.0	+1.66	0.05	3.95
14795-85	50.0	--	25.0	25.0	+2.44	+2.36	+0.59

(a) Plus (+) sign indicates weight gain.

TABLE F-2. ANALYSIS OF SINTERED CERAMIC TERNARY BODIES AFTER DOUBLE VACUUM SINTERING AND ALSO AFTER HEATING IN AIR AT 1750 C FOR 20 HR

Identification	Intended Composition, mole per cent				Analysis, mole per cent							
					After Vacuum Sintering[a]				After Air Firing at 1750 C			
	UO_2	La_2O_3	Y_2O_3	CaO	UO_2	La_2O_3	Y_2O_3	CaO	UO_2	La_2O_3	Y_2O_3	CaO
14795-67	50	25	--	25	54.5	25.6	--	19.9	54.4	25.2	--	20.4
14795-75	60	20	--	20	63.1	20.0	--	16.9	62.4	21.6	--	16.0
14795-83	60	--	20	20	61.9	--	20.3	17.9	63.0	--	20.6	16.4
14795-85	50	--	25	25	52.4	--	25.5	22.1	55.8	--	24.4	19.8

(a) Binary-oxide powder mixtures (UO_2-R_2O_3 and UO_2-CaO) first were compacted hydrostatically at 100,000 psi. Compacts were vacuum sintered at 1650 C for 3 hr. Resulting compacts were crushed in argon-filled dry box. Binary oxide powders were mixed, compacted, and then sintered in vacuum at 1650 C for 4 hr.

F-4

Irradiation-Surveillance Program on Type 347 Stainless Steel

W. E. Murr, F. R. Shober, J. E. Howes, and J. F. Lagedrost

This surveillance program is concerned with the effects of irradiation on the mechanical properties of AISI Type 347 stainless steel. The program has two specific purposes: (1) to provide data to insure safe operation of the KAPL C-33 loop, and (2) to provide a comparison of the mechanical properties of stainless steel irradiated at process-water temperatures (120 F) with those of stainless steel irradiated at 600 F and with those of stainless steel irradiated at 120 F and then annealed at 600 F. A series of capsules containing tensile, cyclic-strain fatigue, and impact specimens is being exposed to fast-neutron irradiation (neutrons having energies greater than 1 Mev) at a low temperature (120 F). Postirradiation testing of the specimens will serve to determine any changes in the mechanical properties of the Type 347 stainless steel. The observed changes in mechanical properties will provide the necessary information to assure that the KAPL C-33 loop and other loops constructed of Type 347 stainless steel are properly designed for continued in-pile operation. On the basis of considerations other than irradiation damage, it is estimated that the optimum design life of the KAPL C-33 loop is approximately 3 years, at which time the loop will have received an integrated fast-neutron exposure of between 1.4 and 1.8×10^{22} nvt.

No data are available for the mechanical properties of Type 347 stainless steel at exposures greater than 3.76×10^{21} nvt, and only limited data are available on the effects of postirradiation annealing on Type 347 stainless steel. The program is expected to provide information on the behavior of stainless steel at exposures between 3.76×10^{21} and 1.6×10^{22} nvt on specimens irradiated at 120 F, specimens irradiated at 600 F, and on specimens irradiated at 120 F and annealed at 600 F.

Eight capsules containing Type 347 stainless steel specimens have been irradiated at 120 F in the ETR since June, 1958. The total fast-neutron exposure of these capsules is given in Table F-3. The first capsule of this series to be discharged will be removed from the reactor for examination and testing of the specimens after an exposure of approximately 5.5×10^{21} nvt. The other capsules of the series will be removed from the reactor and examined at greater total neutron exposures. The additional periods of exposure are equivalent to periods of 6 months of operation of the C-33 loop.

The series of capsules scheduled for process-water-temperature irradiation and subsequent postirradiation heat treatments prior to testing have been in the ETR since Cycle 19. These capsules, BMI-24-18, BMI-24-20, and BMI-24-22, were loaded on top of the cold capsules in Positions K-8-NW, K-8-NE, and L-8-SE, respectively. The fast-neutron accumulations achieved by these capsules are also reported in Table F-3.

The series of capsules that is to be irradiated at temperatures near 600 F has not been placed into the ETR due to lack of suitable space. Since prospects for future installation of these capsules into the ETR look very unfavorable, negotiations are in progress to obtain irradiation space in an alternate test reactor, the WTR. The WTR is expected to have a high fast flux (above 3.0×10^{14} nv) and a gamma-heating rate (over 18 w per g) sufficient to satisfy both the flux and thermal-heating requirements of the capsules.

TABLE F-3. CAPSULES PREPARED FOR THE TYPE 347 STAINLESS STEEL IRRADIATION-SURVEILLANCE PROGRAM

Capsule	Type of Specimens in Capsule	Proposed Irradiation Temperature, F	Approximate Removal Date[a]	Approximate Exposure at Time of Removal[b], nvt	Total Exposure as of September 29, 1959, nvt		Location	Remarks
					Top	Bottom		
BMI-24-1	Tensile and fatigue	600	January, 1959	1.55×10^{20}	--	--	BMI	Examined at BMI Hot-Cell Facility for melting
BMI-24-2	Tensile and fatigue	120	January, 1962	1.31×10^{22}	2.904×10^{21}	3.757×10^{21}	ETR K-8-NE	Being irradiated
BMI-24-3	Tensile and fatigue	600	--	--	--	--	ETR	To be irradiated
BMI-24-4	Tensile and fatigue	120	January, 1963	1.78×10^{22}	1.953×10^{21}	3.224×10^{21}	ETR K-8-SE	Being irradiated
BMI-24-5	Tensile and fatigue	600	--	--	--	--	ETR	To be irradiated
BMI-24-6	Tensile and fatigue	120	June, 1961	1.08×10^{22}	3.867×10^{21}	3.000×10^{21}	ETR K-8-NE	Being irradiated
BMI-24-7	Tensile and fatigue	600	--	--	--	--	ETR	To be irradiated
BMI-24-8	Tensile and fatigue	120	June, 1962	1.54×10^{22}	2.225×10^{21}	3.170×10^{21}	ETR K-8-SE	Being irradiated
BMI-24-9	Tensile and fatigue	600	--	--	--	--	ETR	To be irradiated
BMI-24-10	Tensile and fatigue	120	January, 1961	0.84×10^{22}	3.183×10^{21}	3.305×10^{21}	ETR K-8-SE	Being irradiated
BMI-24-11[c]	Tensile and fatigue	600	--	--	--	--	ETR	Damaged at ETR
BMI-24-12	Tensile and fatigue	120	June, 1960	0.61×10^{22}	4.170×10^{21}	3.554×10^{21}	ETR L-8-SE	Being irradiated
BMI-24-13	Impact	600	--	--	--	--	ETR	To be irradiated
BMI-24-14	Impact	120	June, 1962	1.54×10^{22}	3.631×10^{21}	3.706×10^{21}	ETR K-8-NW	Being irradiated
BMI-24-15	Impact	600	--	--	--	--	ETR	To be irradiated

TABLE F-3. (Continued)

Capsule	Type of Specimens in Capsule	Proposed Irradiation Temperature, F	Approximate Removal Date[a]	Approximate Exposure at Time of Removal[b], nvt	Total Exposure as of September 29, 1959, nvt		Location	Remarks
					Top	Bottom		
BMI-24-16	Impact	120	June, 1960	0.61×10^{22}	3.679×10^{21}	3.364×10^{21}	ETR K-8-NW	Being irradiated
BMI-24-17[c]	Tensile and fatigue	600	October, 1958	3.25×10^{20}	--	--	BMI	Examined at BMI Hot-Cell Facility after high temperature observed
BMI-24-18	Tensile and fatigue	120	--	--	3.09×10^{20}	4.85×10^{20}	ETR	Being irradiated for post-irradiation annealing studies
BMI-24-19[c]	Tensile and fatigue	600	--	--	--	--	ETR	Fabricated to replace BMI-24-17
BMI-24-20	Tensile and fatigue	120	--	--	2.99×10^{20}	5.5×10^{20}	ETR	Being irradiated for post-irradiation annealing studies
BMI-24-21	Tensile and fatigue	600	--	--	--	--	ETR	Fabricated to replace BMI-24-1
BMI-24-22	Tensile and fatigue	120	--	--	3.95×10^{20}	6.56×10^{20}	ETR	Being irradiated for post-irradiation annealing studies

(a) Based on 6-month lead on loop, plus 2 months for examination.
(b) Based on maximum fast flux at tube of 1.7×10^{14} nv for 6-month periods.
(c) Thermocouple lead capsule.

F-7

Development of Niobium-Base Alloys

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

Evaluation is being made of several niobium-base alloys which should possess better properties than a vanadium-10 w/o titanium-1 w/o niobium alloy (an acceptable cladding material for the EBR). Niobium alloys possess high strength and are relatively unaffected by sodium at 800 C. Materials currently being investigated include niobium-1.84 w/o chromium, niobium-4.33 w/o zirconium, niobium-9.95 w/o tantalum-3.31 w/o chromium, niobium-39.8 w/o titanium-10.6 w/o aluminum, and niobium-20.5 w/o titanium-4.28 w/o chromium alloys. Heats of unalloyed niobium and vanadium-11.7 w/o titanium-2.6 w/o niobium alloy are being used for comparison purposes.

Melting, hot- and cold-fabricability studies, metallographic examinations, chemical analyses, and a short heat-treatment study have been completed. Ingots ranging from 3 to 5 lb of all of the above-mentioned alloys were forged at 2500 F while protected from the atmosphere by molybdenum jackets. The niobium-1.84 w/o chromium, niobium-4.33 w/o zirconium, niobium-20.5 w/o titanium-4.28 w/o chromium, and vanadium alloys were then rolled at room temperature to 0.035-in. sheet. This sheet exhibited severe edge cracking. The remaining alloys, including the unalloyed niobium, failed to cold roll. Tensile and corrosion specimens were prepared from the fabricated sheet. The corrosion specimens are to be tested in sodium at ANL.

Tensile testing of the cold-worked alloys has been completed. Both the niobium-1.84 w/o chromium and niobium-4.33 w/o zirconium alloys possess exceptional strength at 800 C (1470 F). Both have a yield strength of approximately 69,000 psi, which is much higher than the yield strength of Type 347 stainless steel (approximately 20,000 psi at 1500 F). Results of the tensile tests are shown in Table F-4.

Chemical analyses of the unalloyed niobium revealed that an increase in oxygen content from 200 to 820 ppm had occurred; this could account for the poor fabrication characteristics of the unalloyed niobium. The oxygen content of the remaining alloys did not increase, which indicates that the poor cold-rolling results obtained with these alloys may have been caused by a need for higher rolling temperatures. Table F-5 shows the results of chemical analysis of all alloys for carbon and nitrogen. Based on the impurity content of unmelted niobium (see Table F-5), there does not appear to be any increase in carbon or nitrogen content in the alloys as a result of the melting operation.

Portions of the material as forged to slabs at 2500 F were retained for further testing. These slab sections (approximately 1 by 3/4 by 4 in.) have been rolled at 800 F from a helium-atmosphere furnace to 0.150-in. sheet. Sound sheet material, having very little edge cracking, was obtained from all alloys with the exception of the unalloyed niobium, niobium-9.95 w/o tantalum-3.31 w/o chromium and niobium-39.8 w/o titanium-10.6 w/o aluminum alloys. The unalloyed niobium exhibited surface cracking, while the niobium-tantalum-chromium and niobium-titanium-aluminum alloys fractured. Pieces from the warm-rolled sheet were then rolled at room temperature to 0.021-in.-thick sheet. The sheet obtained after warm rolling the alloys at 800 F was far superior to the sheet obtained after cold rolling the alloys. Results of this fabrication are shown in Table F-6.

F-8

TABLE F-4. TENSILE DATA FOR NIOBIUM- AND VANADIUM-BASE ALLOYS[a] IN THE AS-WROUGHT CONDITION

Analyzed Composition, w/o	0.2 Per Cent Offset Yield Strength, psi	Tensile Strength, psi	Elongation in 1 In., per cent	Reduction in Area, per cent
	Tested at 650 C (1200 F)			
Nb-1.84 Cr	107,000	115,000	4	5
Nb-4.33 Zr	--	82,400	4	7
Nb-20.5 Ti-4.28 Cr	75,500	92,200	11	25
V-11.7 Ti-2.6 Nb	76,600	88,500	9	21
Type 347 stainless steel	41,000	51,200	46	71
	Tested at 800 C (1470 F)			
Nb-1.84 Cr	68,700	81,600	14	37
Nb-4.33 Zr	69,000	76,400	4	3
Nb-20.5 Ti-4.28 Cr	30,800	39,700	40	100
V-11.7 Ti-2.6 Nb	34,900	45,700	36	67

(a) Average of duplicate specimens.

TABLE F-5. CHEMICAL ANALYSIS OF NIOBIUM-BASE ALLOYS

Analyzed Composition, w/o	Analysis, ppm	
	Carbon	Nitrogen
Niobium (unmelted stock)	250	300
Niobium (melted)	300	110
Nb-1.84 Cr	120	140
Nb-3.21 Cr	110	150
Nb-4.33 Zr	110	220
Nb-9.95 Ta-3.31 Cr	80	130
Nb-39.8 Ti-10.6 Al	80	120
Nb-20.5 Ti-4.28 Cr	120	110
Vanadium (unmelted stock)	610	500
V-11.7 Ti-2.6 Nb	330	400

TABLE F-6. FABRICATION DATA FOR NIOBIUM-BASE ALLOYS

From Slabs Forged at 2500 F

Analyzed Composition, w/o	Fabrication Data			
	Rolled at 800 F		Rolled at 75 F	
	Reduction, per cent	Remarks	Reduction[a], per cent	Remarks
Niobium	67	Surface cracks	80	Surface cracks
Nb-1.84 Cr	70	Sound sheet	80	No edge cracks
Nb-3.21 Cr	77	Slight edge cracks	80	Edge cracks
Nb-4.33 Zr	74	Sound sheet	81	No edge cracks
Nb-9.95 Ta-3.31 Cr	Fractured on first pass		--	--
Nb-39.8 Ti-10.6 Al	Fractured on first pass		--	--
Nb-20.5 Ti-4.28 Cr	80	--	82	No edge cracks
V-11.7 Ti-2.6 Nb	73	--	80	Edge cracks

(a) This reduction is based upon measurements of sheet rolled at 800 F (for example: the niobium-3.21 w/o chromium alloy was reduced 77 per cent and then 80 per cent - the total reduction from forged ingot was 96 per cent).

F-9

Additional ingots are being prepared for further evaluation of fabrication techniques. The compositions of these ingots will be the same as those already prepared.

Development of Corrosion-Resistant Niobium Alloys

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

The evaluation of selected niobium-base alloys for service in pressurized-water reactors was continued. Work so far has shown that a binary 12.6 a/o vanadium alloy, which has a high degree of corrosion resistance to hot water and steam and more than adequate creep strength, is well suited for this application. Currently, vanadium-containing ternary alloys of lower total alloy content are being evaluated. It is hoped that such alloys will have improved fabricability along with adequate corrosion resistance.

The corrosion results obtained to date in 600 and 680 F water and 750 F 1500-psi steam continue to indicate that:

(1) Unalloyed niobium does not possess adequate corrosion resistance.

(2) Additions of titanium, vanadium, or zirconium markedly improve the corrosion resistance of niobium.

(3) The most corrosion-resistant alloys are those containing more than 40 a/o zirconium or a ternary alloy containing 28 a/o titanium-6 a/o chromium.

(4) Binary niobium alloys containing 7 to 12 a/o vanadium offer the optimum combination of low neutron cross section, high-temperature strength, and corrosion resistance.

(5) The addition of a ternary alloying agent to the 2.5 or the 5.0 a/o vanadium alloys does not improve their corrosion resistance.

The corrosion results for all alloys tested to date are summarized in Table F-7.

The cooperative corrosion testing program with BAPD and KAPL is continuing. Replicate specimens have been exposed 77 days to 680 F water and 750 F 1500-psi steam. The average of the weight gains in 680 F water is 100 mg per dm^2. Specimens are slowly losing weight, having exhibited their maximum weight gain of 121 mg per dm^2 at 42 days of exposure. Weight losses of specimens exposed to 750 F steam average 790 mg per dm^2. The latter are losing weight at irregular intervals rather than at a constant rate.

Corrosion tests are being continued on all specimens except those noted in Table F-7.

F-10

TABLE F-7. SUMMARY OF CORROSION RESULTS OBTAINED ON NIOBIUM ALLOYS EXPOSED TO HIGH-TEMPERATURE WATER AND STEAM

Alloy Addition (Balance Niobium), a/o	Tested in 600 F Water		Tested in 680 F Water		Tested in 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
Commercial Niobium, Rocking-Hearth Melts						
Unalloyed Nb	224	-25.9	42[a]	Disintegrated	28[a]	Disintegrated
10.5 Zr	--	--	196[a]	0.67	126	-18.3
26.1 Zr	--	--	196[a]	0.07	--	--
35.7 Zr	--	--	196[a]	0.66	--	--
45.7 Zr	--	--	196[a]	0.55	--	--
1.08 W	--	--	196[a]	-2.60	126	-28.0
4.67 W	--	--	196[a]	-29.3	--	--
9.56 W	--	--	7[a]	Cracked	--	--
2.45 Mo	--	--	196[a]	-7.10	98	Disintegrated in 98 days
5.20 Mo	--	--	196[a]	-1.30	126	-73.0
7.40 Mo	--	--	196[a]	0.62	--	--
4.42 V	--	--	196[a]	0.42	126	-2.79
6.59 V	--	--	196[a]	0.73	126	1.26
8.93 V	--	--	196[a]	0.59	126	1.07
10.7 V	--	--	196[a]	0.78	--	--
13.7 V	--	--	196[a]	0.50	--	--
24.2 V	--	--	196[a]	0	--	--
4.90 Fe	--	--	196[a]	0.10	98	Disintegrated in 98 days
9.41 Ti	--	--	196[a]	0.65	126	1.34
18.8 Ti	--	--	196[a]	0.48	--	--
24.3 Ti	--	--	196[a]	0.52	--	--
30.5 Ti	--	--	196[a]	0.40	--	--
33.8 Ti	--	--	196[a]	0.33	--	--
12.0 Ti-0.5 Cr	--	--	196[a]	0.66	--	--
20.2 Ti-2.1 Cr	--	--	196[a]	0.39	--	--
28.2 Ti-6.1 Cr	--	--	196[a]	0.20	--	--
12.0 Ti-4.2 Mo	--	--	196[a]	0.64	--	--
17.4 Ti-6.2 Mo	--	--	196[a]	0.54	--	--
23.1 Ti-7.8 Mo	--	--	196[a]	0.45	--	--
10.4 Ti-5.0 V	--	--	196[a]	0.56	--	--
16.1 Ti-8.4 V	--	--	196[a]	0.40	--	--
22.6 Ti-11.0 V	--	--	196[a]	0.48	--	--

F-11

TABLE F-7. (Continued)

Alloy Addition (Balance Niobium), a/o	Tested in 600 F Water: Exposure Time, days	Tested in 600 F Water: Total Weight Change, mg per cm^2	Tested in 680 F Water: Exposure Time, days	Tested in 680 F Water: Total Weight Change, mg per cm^2	Tested in 750 F Steam: Exposure Time, days	Tested in 750 F Steam: Total Weight Change, mg per cm^2
High-Purity Niobium, Consumable-Electrode Melts						
Unalloyed Nb	224	0.73	196	-4.88	210	-45.7
7.18 Mo	224	0.63	196	-0.03	70[a]	Cracked
12.6 V	224	0.32	196	0.58	210	0.88
46.8 Zr-5.06 Ti	196	0.21	168	0.90	182	2.52
11.2 Ti-3.2 Mo	196	0.33	168	0.57	182	0.56
18.8 Ti-8.7 Mo	196	0.45	168	0.46	182	0.58
9.9 Zr-9.4 V	84	0.25	84	0.46	98	-2.63
5.7 Zr-11.4 V	84	0.22	84	0.33	98	-0.60
9.1 Ti-6.3 Cr	84	0.25	84	0.36	98	-2.38
High-Purity Niobium, Rocking-Hearth Melts						
Unalloyed Nb	--	--	112	1.27	--	--
Unalloyed Nb	--	--	84[a]	Disintegrated in 84 days	--	--
Unalloyed Nb	--	--	84	0.79	--	--
1.1 Zr	--	--	112	-137.0	--	--
2.2 Zr	--	--	28	-10.0	--	--
5 Zr	--	--	84	-3.35	--	--
10.2 Zr	--	--	84	-0.04	--	--
40 Zr	--	--	84	0.52	--	--
65 Zr	--	--	84	0.75	--	--
75 Zr	--	--	84	0.94	--	--
90 Zr	--	--	84	0.85	--	--
3.2 Ti	--	--	112	-1.06	--	--
10.5 Ti	--	--	112	0.57	--	--
25.0 Ti	--	--	112	0.38	--	--
<0.02 Cr]	--	--	112	1.02	--	--
0.5 Cr	--	--	112	0.54[b]	--	--
0.5 Cr	--	--	84	-0.72	--	--
<0.08 Fe	--	--	84	-0.22	--	--
0.3 Fe	--	--	28	-0.95	--	--
10 Fe	--	--	28[a]	-20.0	--	--
10.9 Zr-5.1 Ti	--	--	84	0.50	--	--
25 Zr-5 Ti	--	--	84	0.31	--	--
25 Zr-15 Ti	--	--	84	0.57	--	--

F-12

TABLE F-7. (Continued)

Alloy Addition (Balance Niobium), a/o	Tested in 600 F Water: Exposure Time, days	Tested in 600 F Water: Total Weight Change, mg per cm^2	Tested in 680 F Water: Exposure Time, days	Tested in 680 F Water: Total Weight Change, mg per cm^2	Tested in 750 F Steam: Exposure Time, days	Tested in 750 F Steam: Total Weight Change, mg per cm^2
High-Purity Niobium, Rocking-Hearth Melts (Continued)						
25 Zr-25 Ti	--	--	84	0.57	--	--
35 Zr-5 Ti	--	--	84	0.41	--	--
35 Zr-15 Ti	--	--	84	0.47	--	--
45 Zr-5 Ti	--	--	84	0.58	--	--
10 Zr-5 Mo	--	--	84	0.32	--	--
35 Zr-5 Mo	--	--	84	0.51	--	--
45 Zr-5 Mo	--	--	84	0.52	--	--
35 Zr-5 Al	--	--	84	0.57	--	--
45 Zr-5 Al	--	--	84	0.49	--	--
10 Zr-5 Cr	--	--	84	0.47	--	--
45 Zr-5 Cr	--	--	84	0.41	--	--
10 Zr-5 Fe	--	--	84	0.36	--	--
1.6 V	--	--	112	0.68	--	--
2 V-2.5 Ti	--	--	84	0.67	--	--
2 V-2.3 Mo	--	--	84	0.77	--	--
2.2 V-0.54 Fe	--	--	84	0.74	--	--
1.8 V-<0.02 Cr	--	--	84	0.54	--	--
1.8 V-0.14 Al	--	--	84	0.82	--	--
2.5 V-2.5 Zr	--	--	28	-0.05	--	--
2.2 V-0.87 Ni	--	--	28[a]	-1.00	--	--
4 V-2.3 Zr	--	--	84	0.38	--	--
5 V-25 Zr	--	--	84	0.20	--	--
2.5 V-35 Zr	--	--	84	0.58	--	--
5 V-45 Zr	--	--	84	0.40	--	--

(a) Off test.
(b) Losing weight.

F-13

Additional hot-hardness and tensile-test data were obtained on selected high-purity niobium-base screening alloys. These data are given in Tables F-8 and F-9, respectively. Of the binary alloys tested at 1500 F, the best combination of strength and ductility was obtained in the 1.6 a/o vanadium alloy. While the 2.2 a/o zirconium alloy showed both higher yield and ultimate strengths, its hot ductility was considerably lower. The titanium additions, on the other hand, gave no strengthening and appeared, in fact, to lower the ductility of the base slightly.

The same effects of these three elements appear to be carried over into ternary combinations of these metals with niobium. As shown in Table F-9, the best combinations of 1500 F strength and ductility occur in the ternary niobium-vanadium-base alloys, especially those containing small additions of nickel or iron.

Melting was completed and fabrication started of another group of twenty-five 50-g screening-alloy ingots. These alloys have binary additions of cerium, nickel, palladium, and yttrium and low-level ternary combinations of vanadium with dispersion-hardening additions of titanium or zirconium carbide or oxide. These alloys will primarily be screened for their resistance to corrosion by 680 F water.

In addition, melting is in process on four 2-1/2-lb ingots containing the alloy additions listed below:

(1) 7.5 a/o vanadium

(2) 7.5 a/o vanadium-2.5 a/o titanium

(3) 7.5 a/o vanadium-2.5 w/o molybdenum

(4) 7.5 a/o vanadium-0.19 a/o nitrogen (equivalent to 0.03 w/o nitrogen).

These alloys will be corrosion tested in 600 and 680 F water and 750 F steam and will be evaluated in room- and elevated-temperature tensile and creep tests as well.

Investigation of the Creep Properties of Zircaloy-2 During Irradiation at Elevated Temperatures

F. R. Shober, P. B. Shumaker, A. P. Young,
M. F. Amateau, and R. F. Dickerson

The creep properties of Zircaloy-2 at elevated temperatures used by reactor designers have been based on tests of unirradiated material, and the effect of reactor environment on the creep properties of this material has not been taken into consideration. Increased operating temperatures for reactors have imposed more stringent requirements on the creep properties of materials. Hence, the influence of fast-neutron irradiation on creep properties at elevated temperatures is of more importance than formerly. The purpose of this investigation is to compare the creep deformation obtained in Zircaloy-2 under irradiation with the deformation obtained from Zircaloy-2 under out-of-reactor conditions at the same stresses. The technique employed should give a measure of the degree of acceleration or deceleration of creep resulting during

TABLE F-8. HOT-HARDNESS DATA FOR HIGH-PURITY NIOBIUM-BASE ALLOYS AT VARIOUS TEMPERATURES

Alloy	Alloy Content (Balance Niobium), a/o	Hardness[a], VHN, at Temperature Shown						
		75 F	600 F	900 F	1200 F	1400 F	1650 F	75 F[b]
N40	100 Nb	97	95	74	65	55	47	93
N3	2.2 Zr	130	116	87	84	100	99	126
N46	0.3 Fe	106	123	95	92	89	71	110
N47	10 Fe[c]	251	182	187	182	172	119	235
N52	2.2 V-0.87 Ni	168	139	135	128	124	94	171
N55	2.5 V-2.5 Zr[c]	141	114	103	100	113	115	144

(a) All alloys vacuum annealed 1 hr at 2190 F before testing.
(b) Determined after heating samples to 1650 F.
(c) Nominal composition.

TABLE F-9. 1500 F TENSILE PROPERTIES OF SELECTED HIGH-PURITY NIOBIUM-BASE ALLOYS

Alloy	Alloy Content, (Balance Niobium), a/o	Annealing Temperature, F	Tensile Properties[a]		
			0.2 Per Cent Offset Yield Strength, psi	Ultimate Strength, psi	Elongation in 1 In., per cent
N40	100 Nb	2190	16,000	21,000	18
N3	2.2 Zr	2190	30,000	32,000	5
N16	1.6 V	2190	16,500	28,500	30
N11	3.2 Ti	2190	14,000	17,000	11
N12	10.5 Ti	2190	12,000	20,000	12
N53	1.8 V-<0.02 Cr	2190	17,000	31,000	51
N54	1.8 V-0.14 Al	2190	16,000	30,000	38
N49	2 V-2.5 Ti[b]	2190	19,000	35,000	18
N50	2 V-2.3 Mo	2190	21,000	34,000	29
N51	2.2 V-0.54 Fe	2190	25,000	43,000	31
N52	2.2 V-0.87 Ni	2190	38,000	47,000	52
N55	2.5 V-2.5 Zr[c]	2190	--	38,000	1
N17	4 V-2.3 Zr[b]	2190	30,000	46,000	9
N21	10 Zr-5 Ti[c]	2730	34,000	37,000	2

(a) All tests conducted in helium atmosphere.
(b) Values previously reported.
(c) Nominal composition.

irradiation. The temperature range of interest, 600 to 700 F, encompasses that temperature range at which strain aging has been reported to occur. Tensile tests and internal-friction tests at room and elevated temperatures are planned as part of the program to detect and describe the strain-aging phenomenon.

The design of an instrumented capsule has been undertaken to carry out the in-reactor portion of the Zircaloy-2 creep tests. The concept of the internally pressurized tubular specimen, as originally planned for the in-pile creep test, was changed to a design involving a 0.03-in.-thick flat sheet. This change was necessary in order to obtain a more favorable geometry for heat transfer. It also was found that the internal pressure necessary to obtain stresses to produce creep could not be obtained without special pressurizing equipment. It had previously been thought that the pressure of gas in commercial cylinders would be sufficient. It now is planned to load the flat sheet-type specimen by attaching the ends to a bellows which will be compressed by introducing gas pressure to the inside of the capsule. It appears that a 500-psi gas pressure will be sufficient on the exposed cross-sectional area of the bellows to produce a status of 25,000 psi in a specimen 5/16 in. wide. Specimens having several gage sections of different widths can be used to obtain several different stress levels. Auxiliary heaters will surround the specimens at their gage sections. Individually controlled circuits will regulate the heat to the individual gage sections. Guard heaters will be placed in the vicinity of the grip ends to prevent heat losses from the specimen through the bellows.

Zircaloy-2 to be used in the study of strain aging has been fabricated, and chemical analyses have been made. Swaged wire for internal-friction studies has been prepared from the same ingot. Flat-sheet-type tensile specimens have been prepared and will be used for a series of tensile tests at room and elevated temperatures. It is planned to plastically strain these specimens approximately 5 per cent and then age them for 1 hr in vacuum at 450, 550, 650, or 750 F. These then will be tested at room temperature, and if a major increase of the yield strength is observed as a function of annealing temperature it can be assumed to be related to a strain-aging mechanism. The appearance of a yield point in any of the stress-strain curves from the above specimens would be further evidence of strain-aging.

Preliminary internal-friction measurements have been made on swaged Zircaloy-2 wire to detect any evidence of strain aging. The specimens were cut from swaged wire, 0.050 in. in diameter, heat treated for 1 hr at 750 C in vacuum, and furnace cooled. Data were taken in a torsional pendulum-type apparatus at frequencies of 1 to 2 cps over a temperature range of -70 to 500 C. Two small internal-friction peaks were observed, one at -40 C and another at 290 C. The specimens subsequently were strained approximately 2 per cent in tension, and the internal-friction values were remeasured. The damping capacity was increased slightly, but no change was observed in the -40 C internal-friction peak. The 290 C peak, however, disappeared. This may be an indication of strain aging and will be investigated further. If these internal-friction peaks are associated with solute atoms of an element for which the solubility limit has been exceeded, then the size of the peaks possibly could be increased by quenching the specimens to produce a supersaturated solid solution. Such an experiment will be undertaken.

Studies by electron transmission of thin films of zirconium and Zircaloy-2 have been initiated. The first work has been done on 1-mil zirconium foil. The foil has been examined in the as-rolled condition, as-annealed condition, and as annealed and reduced slightly by cold rolling. As might be expected, annealing followed by rolling

has yielded the most interesting results. These specimens had a very strong texture, with the basal plane nearly parallel with the rolling plane. Since slip occurs mainly on prismatic planes, the slip planes can be viewed only in a section on the basal plane. In order to see dislocation networks on prismatic planes, thin sections will have to be prepared from bulk material. Future plans involve improving the polishing technique to obtain thinner sections, preparation of thin sections with various orientations, and preparation of thin sections from Zircaloy-2.

The specific information sought from the thin-film investigation is the nature of the dislocation networks on the slip planes, the interaction between neighboring dislocations and between dislocations and other obstacles, such as grain boundaries, twin boundaries, precipitate particles, and so on. Also, thin sections of Zircaloy-2 will be irradiated to study the effects of radiation on the fine structure.

Determination of Oxygen in Sodium at Concentrations Below 10 PPM

D. R. Grieser and W. H. Goldthwaite

Because of the great susceptibility of such important reactor materials as beryllium, niobium, and zirconium to oxygen pickup and corrosion in sodium-cooled systems, a need has arisen for a fast and sensitive means of monitoring the oxygen level in a reactor. Although several techniques have been developed for the determination of oxygen in sodium, none combine high sensitivity with speed of analysis. The goal of the present program is to evaluate and develop techniques which show ±1 ppm sensitivity at oxygen concentrations of 10 ppm and below, and are adaptable to continuous or, at least, very rapid oxygen-level determination.

Five promising approaches are being investigated at present. These include the application of mass spectrography, the techniques of polarography and ellipsometry, the measurement of electrical resistivity, and extension of the plugging-indicator technique. Preliminary investigations are to be conducted with sodium containing relatively large amounts of oxygen, 20 to 100 ppm, since it is more readily produced, measured, and handled in this concentration range. Those techniques which promise the required sensitivity at lower concentrations will then be selected for advanced study.

The construction of a system for producing and measuring sodium containing specific oxygen concentrations is being completed. Sodium specimens will be available for the various analytical phases after proof test runs have been made. Equipment for handling sodium for the various feasibility studies is being designed and constructed, and the appropriate measuring equipment is being adapted for use on the present program.

F-17

STUDIES OF ALLOY FUELS

R. F. Dickerson

Niobium-uranium alloys containing 10, 20, 30, 40, 50, and 60 w/o uranium have been exposed to 600 and 680 F water for 140 days. At 600 F, all alloys have an adherent black oxide; however, although most of them show a net weight gain, they are beginning to corrode with a weight loss. One-hundred-hour creep-rupture tests are in progress on the 10 and 20 w/o uranium alloys, and fabrication studies on the other alloys are continuing. Studies of the effect of oxygen on the composition limits of the gamma loop are continuing.

Thorium-uranium alloys containing ternary additions can be reduced about 40 per cent by hot rolling without stringering the uranium-rich phase. This is good because calculations indicate that the size of the uranium-rich particle in the thorium matrix is important from the point of view of irradiation stability. Tensile and creep properties of a number binary and ternary alloys are being determined with the objective of attempting to predict irradiation behavior on the basis of strength. An alloy of thorium and nitrogen has been prepared by arc melting thorium in a dynamic atmosphere of nitrogen at a pressure of about 1/3 atm. The material analyzed at $ThN_{0.71}$. Further attempts to obtain stoichiometric ThN will be made using greater pressures of nitrogen during melting.

With the near completion of the BMI plutonium facility, a program designed to investigate several plutonium alloys is being planned. The initial investigation will be concerned with the niobium-plutonium alloy system. The extent of this particular study will be entirely dependent on the extent of the solubility of plutonium in niobium. If there is no solubility, the investigation will be discontinued and another system studied. In addition, it is planned to investigate the thorium-plutonium-alloy system to complement the thorium-alloy study now in progress.

Development of Niobium-Uranium Alloys

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

An investigation of the fabrication characteristics, mechanical and physical properties, and corrosion behavior in various media of niobium-uranium alloys is being made to determine their applicability as reactor fuels.

Alloys containing from 10 to 60 w/o uranium were prepared using niobium containing from 300 to 700 ppm oxygen and from 0.02 to 0.74 w/o zirconium. The fabrication characteristics, tensile properties, and corrosion behavior in NaK, water, steam, air, and CO_2 were determined using the above material. No effects were found on these properties as a result of the variations in oxygen and zirconium contents of alloys examined in the study.

The effects of oxygen on the composition limits of the gamma loop in the niobium-uranium system is also being determined. Alloys in wire-bar form are being homogenized for 8 hr at temperatures between 1400 and 1500 C for alloys containing 40 to 58 w/o niobium and for 24 hr at 1200 C for alloys containing 10 to 30 w/o niobium.

F-18

Uranium-rich ternary uranium-niobium-zirconium alloys are being prepared for a study of the gamma-immiscibility loop in the ternary system. The purpose of this investigation is to determine if single-phase ternary alloys can be prepared containing greater concentrations of uranium than is possible in binary alloys. Alloys to be considered include niobium-60 w/o uranium-5, -10, and -20 w/o zirconium and niobium-50 w/o uranium-5, -10, and -20 w/o zirconium.

Niobium-10 and -20 w/o uranium alloys have been successfully fabricated by forging at 2500 F and rolling at 1800 F. Fabrication of alloys containing 30 w/o uranium or more has been unsuccessful to date. Ingots of niobium-30 and -10 w/o uranium have been prepared for fabrication at 3000 F.

Corrosion testing of niobium-uranium alloys in water at 600 and 680 F has continued through 140 days. The test data are shown in Table F-10. At 600 F all alloys have an adherent black oxide. Most of these alloys still show a net weight gain, but are corroding with a weight loss. The alloys at 680 F have been removed from test after 140 days for hydrogen and oxygen analyses. Most of these alloys are covered with a brown oxide which appears to be spalling slightly.

Specimens now being corrosion tested in NaK at 1600 F will be taken off test after 1000 hr and machined to remove any corrosion products present. After machining the specimens will be returned for further tests in sodium at 1500 F.

Chemical analyses for carbon, hydrogen, nitrogen, and oxygen on material being used in corrosion testing have been completed. Alloys rolled in air at 1800 F appear to have picked up both oxygen and nitrogen. However, the contamination does not appear to have an adverse effect on corrosion life in 600 F water or in 1600 F NaK. The results of these analyses are shown in Table F-11.

Creep test specimens of niobium-10 and -20 w/o uranium alloys have been prepared, and 100-hr stress-rupture tests are in progress.

Development of Thorium-Uranium Alloys

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

Thorium-uranium and thorium-uranium-base alloys are being investigated with the aim of improving their irradiation stability and corrosion resistance. Variables such as purity, casting technique, fabrication history, and alloy content are being studied with regard to their effects on the size and mode of occurrence of the uranium-rich phase in the thorium-rich constituent. Thorium-5 to 25 w/o uranium-base ternary alloys that have been and are now being studied contain either 5 to 25 w/o zirconium or small amounts of molybdenum or niobium. Some quaternary alloys contain niobium and zirconium additions. An investigation of the preparation and properties of arc-melted thorium and thorium-uranium carbides and nitrides is also being performed.

Metallographic examination of ternary alloys reduced approximately 40 per cent by hot rolling indicates that only minor stringering of the uranium-rich phase occurred.

TABLE F-10. CORROSION DATA FOR NIOBIUM-URANIUM ALLOYS[a] IN WATER AFTER 140 DAYS

Alloy Content (Balance Niobium), w/o	Impurity Content		Specimen Condition	Total Weight Change After 140 Days, mg per cm^2	
	Oxygen, ppm	Zirconium, w/o		In 600 F Water	In 680 F Water
10 U	600	0.74	Fabricated	0.61	-1.66
	700	0.17	Fabricated	0.60	-17.7
	300	0.02	Fabricated	0.38	-48.2
20 U	600	0.74	Fabricated	1.14	-0.58
	700	0.17	Fabricated	1.35	-10.0
	300	0.02	Fabricated	0.71	0.05
30 U	600	0.74	As cast	0.74	-1.16
	700	0.17	Fabricated	-0.25	-1.66
	300	0.02	As cast	1.07	-2.74
40 U	600	0.74	As cast	0.49	1.01
	700	0.17	As cast	0.47	0.67
	300	0.02	As cast	0.77	-2.66
50 U	600	0.74	As cast	0.87	-2.66
	700	0.17	As cast	0.66	-2.81
	300	0.02	As cast	0.38	-8.18
30 U	600	0.74	As cast	-2.12	--
	700	0.17	As cast	-1.32	-17.4
	300	0.02	As cast	-0.18	-12.4
Zircaloy-2	--	--	--	0.27	0.48

(a) Average of duplicate specimens.

TABLE F-11. CHEMICAL ANALYSES OF NIOBIUM-URANIUM ALLOYS

Alloy Content (Balance Niobium), w/o	Nominal Impurity Content (Base Material), ppm				Condition	Analyzed Impurity Content, ppm			
	Oxygen	Hydrogen	Nitrogen	Carbon		Oxygen	Hydrogen	Nitrogen	Carbon
10 U	600	--	580	280	Fabricated	680	9	900	340
	700	--	600	270	Fabricated	1190	3	780	340
	300	--	300	100	Fabricated	3170	7	780	80
20 U	600	--	580	280	Fabricated	458	7	1600	370
	700	--	600	270	Fabricated	523	10	1000	330
	300	--	300	100	Fabricated	198	19	340	60
30 U	600	--	580	280	As cast	586	62	850	420
	700	--	600	270	Fabricated	669	12	410	250
	300	--	300	100	As cast	165	39	120	50
40 U	600	--	580	280	As cast	661	7	320	310
	700	--	600	270	As cast	579	2	330	230
	300	--	300	100	As cast	261	0.7	130	20
50 U	600	--	580	280	As cast	375	7	270	260
	700	--	600	270	As cast	334	4	260	280
	300	--	300	100	As cast	271	2	130	300
60 U	600	--	580	280	As cast	471	9	230	190
	700	--	600	270	As cast	273	3	200	190
	300	--	300	100	As cast	192	3	120	80

F-21

Corrosion tests of thorium-uranium-zirconium alloys in 200 C water are now being performed. These alloys were furnace cooled or water quenched from 1000 C prior to testing. It is hoped that corrosion resistance will be improved if a microstructure can be developed that exhibits a continuous grain-boundary network of the delta uranium-zirconium phase. The thorium-rich grains would then be protected or shielded from the corrosive media, and corrosion should proceed at a rate determined mainly by that of the delta phase, which is superior to alpha thorium in resistance to corrosion by 200 C water.

Alloys of thorium-5, -10, and -20 w/o uranium, thorium-10 w/o uranium-1.5 w/o molybdenum, -2 w/o niobium, or -10 w/o zirconium, and thorium-10 w/o uranium-10 w/o zirconium-2 w/o niobium have been prepared for tensile and creep testing at 600 and 700 C. These tests will be performed to obtain quantitative strength data which will permit an evaluation of the resistance of these alloys to radiation swelling. The ternary and quaternary alloys of this group are known to have good high-temperature-strength characteristics from hot-hardness measurements and have been selected on this basis.

Further studies of the above-mentioned alloys will include determinations of the effect of heat treatment on microstructure. A study of the recrystallization behavior of several of the ternary alloys will be undertaken. Also, alloys having high strengths at 600 C will be reprepared with deliberate carbon additions in an attempt to produce alloys of still greater hot strength.

An alloy of thorium and nitrogen has been prepared as part of an attempt to prepare thorium-uranium nitrides. Arc melting of thorium in a dynamic atmosphere of nitrogen maintained at a pressure of about 1/3 atm resulted in an alloy containing 4.2 w/o nitrogen, equivalent to $ThN_{0.71}$. The microstructure of this specimen consists of ThN particles surrounded by thorium. Further attempts to obtain the stoichiometric ThN composition will be made using greater pressures of nitrogen during melting.

The Development of Plutonium Fuel Alloys

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

A program aimed at investigating plutonium-alloy systems for possible fuel materials is being planned.

In an attempt to develop alloys for application at reasonably high temperatures, zirconium-niobium-plutonium and uranium-niobium-plutonium alloys will also be investigated. Alloys will be prepared by arc-melting techniques and examined by metallography and X-ray diffraction. Mechanical and physical properties of promising alloys will be determined.

As part of a thorium-alloy program, thorium-plutonium alloys are to be studied also. As a result of solid-solution hardening resulting from the rather extensive solid solubility of plutonium in alpha thorium, these alloys are expected to exhibit improved

mechanical properties. Consequently, irradiation properties also may be improved. Thorium-5, -10, and -30 w/o plutonium alloys will be prepared by arc-melting techniques. Initial investigations of these alloys will include metallography, hardness testing, and age-hardening and recrystallization studies. In addition, a thorium-10 w/o zirconium-10 w/o plutonium alloy will be prepared and studied. This alloy may also be subjected to boiling-water corrosion tests.

Investigations of titanium-base plutonium alloys, zirconium-base plutonium alloys, and uranium-molybdenum-plutonium alloys are planned also. All alloys will be carefully screened by the above-mentioned tests, and any alloy showing outstanding properties will be considered for possible irradiation testing.

Experimental work on the program will begin upon completion of the Battelle Plutonium Facility.

FISSION-GAS RELEASE FROM REFRACTORY FUELS

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

Preparations for study of the important mechanisms of gas release in UO_2 are in progress. This study will include both a determination of diffusion coefficients in UO_2 specimens of known geometry (single crystals) and use of these data in an in-pile study of gas release from sintered UO_2.

Characterization of Sintered UO_2 and Model of Gas Release

Specific structural characteristics of sintered uranium dioxide bodies are being studied in order to evaluate the open and closed porosity of various preparations and to aid in developing an improved model for fission-gas release. Present studies are concerned with interpreting the relationship between surface-area measurements determined by gas adsorption and the pore structure and microstructure of sintered UO_2 bodies. In addition, the effect of irradiation upon the structure of these bodies is also of concern.

During the past month, the distribution of pores in various preparations of sintered UO_2 was investigated by light and electron microscopy. The specimens examined included hydrogen-, vacuum-, and argon-sintered UO_2 bodies. Pycnometric densities on the specimens studied were 95 per cent of theoretical for the hydrogen-sintered bodies, 96 per cent for the vacuum-sintered body, and 93 per cent, 98 per cent, and 99 per cent for the argon-sintered bodies. The pore distribution appears to be somewhat related to the atmosphere used in sintering. In the case of hydrogen-sintered bodies, there was a concentration of large pores in the grain boundaries. The intergranular pores were, in general, much smaller (approximately one-eighth to one-fourth the diameter of the grain-boundary pores) and were present in higher concentrations at

a uniform distance from the grain boundaries. This would suggest that pores near the grain boundaries collected and moved with the grain boundaries during crystal growth. The size of the grain-boundary pores indicated a coalescence of these pores in the hydrogen-sintered bodies. A similar distribution of pores was observed in the vacuum-sintered body. However, the grain-boundary and intragranular pores were essentially the same size, but these pores were significantly smaller than any other specimen examined. The grain size of the vacuum-sintered body was less than one-half that of the hydrogen- or argon-sintered materials. It is not expected that the 50 C lower sintering temperature of the vacuum-sintered body would have accounted for this difference. In the case of the argon-sintered bodies, a rather distinct difference in the pore distribution was observed. There was little or no evidence for pore concentration or coalescence at the grain boundaries. However, a slight deficiency in pore concentration adjacent to the boundaries was observed. The distribution of the pores in the argon-sintered bodies (93 to 99 per cent of theoretical density) was essentially identical. The size of the pores in the 99 per cent and 98 per cent bodies was less than that of those in the body with a density of 93 per cent theoretical.

A limited study of the effect of irradiation and fission upon the pore distribution and the microstructure of UO_2-sintered bodies and single crystals has been initiated. The surfaces of several metallographic specimens have been characterized by light and electron microscopy prior to irradiation. These are being irradiated in BRR and will be re-examined after various irradiations. Postirradiation examination of specimens having 1 per cent atom displacement should be completed during the next period. Successive irradiations are planned to give 10, 50, and 100 per cent atom displacements by fission fragments.

Diffusion in UO_2

The assembly of the apparatus to study the release of fission gas from refractory fuel specimens in the radioisotope laboratory during postirradiation heat treating is in its final stages. The apparatus is scheduled to be in operation in November, and a series of fission-gas-release measurements will be initiated using fused uranium dioxide crystals.

To date, characterization of the series of fused UO_2 spheres and plates for diffusion studies has consisted of measurements of gross physical dimensions, density and weight, determination of the crystallographic orientation of the plates, and preliminary efforts at surface-area determination by the BET microweighing technique. No effort was made to control crystal orientation of the plate specimens during the machining and polishing stages. It has been established that all plate and spherical fused specimens are single crystals. Surface-area measurements on the fused UO_2 are of questionable significance. Surface areas of two plate specimens of 1 cm^2 nominal area were less than the limit of sensitivity of the analytic apparatus (25 cm^2). However, a sphere approximately 0.8 cm^2 in nominal surface area had a BET surface area of 100 cm^2. Additional determinations on the fused UO_2 are necessary to resolve these surface-area variations.

F-24

An alternate route is being taken in the preparation of fused UO_2. In this case, the fused material consists of spherical powder particles. The process consists of transporting ball-milled UO_2 powders (1 to 100-μ particle size) through a helium plasma jet capable of attaining temperatures to 11,000 K. Results of two fusion experiments are encouraging. Substantial amounts (about 80 per cent) of the powder particles were fused into spherical forms 0.5 to 30 μ in diameter. The major process problems to be solved involve increasing the efficiency to obtain a greater percentage of spherical particles and preventing contamination (partial oxidation to U_3O_8 and U_4O_9 has occurred either because of impurities in the helium-carrier gas or because of adsorbed gases or water vapor on the starting powders).

Preparation for In-Pile Study

Construction of the in-pile fission-gas trapping train and the California hood enclosure containing the gas-collection system is in the final stages of completion. These components have been installed at the BRR. Tasks that remain include completion of the carrier-gas purification system, checkout and calibration of equipment for temperature and vacuum control, and installation of radiation-monitoring equipment and associated radiation-warning circuits.

For this in-pile study, an extensive survey of the effects of powder preparation and pressing and sintering conditions on the chemical, physical, and microstructural properties of sintered material has been completed. The parameters considered were ball-milling procedures (time and ball-milling medium), pressing pressure, sintering time, temperature, and atmosphere, and initial oxygen-to-uranium ratio of the oxide powder.

The resulting materials occupied a broad properties spectrum, and, from this spectrum, a few sintered conditions have been selected for further study. Materials of the different densities (90 and 95 per cent of theoretical density) are now under consideration for both the in-pile and postirradiation experiments. The most reliable estimates indicate that these materials will possess sufficient internal surface area and interconnected porosity to minimize the confusion between the recoil and diffusion-release mechanisms at temperatures about 1000 C. These specimen materials will differ in one other basic property: pore distribution. One group of specimens with a density of 90 per cent of theoretical will have essentially the same pore structure as the 95 per cent dense sintered bodies with pores distributed both intergranularly and also within sintered grains. A second group of 90 per cent dense specimens was prepared in such a manner that porosity is situated in grain boundaries only. The sintered oxygen-to-uranium ratio of these materials has been determined to be 2.01, within the limits of analytic precision. Limited surface-area measurements by the BET microweighing technique reveal that the surface area of the 95 per cent dense specimens is about 2200 cm^2 per cm^3. Additional surface-area determinations will be carried out on the 95 per cent dense material and also on the sintered bodies having a density of 90 per cent of theoretical.

F-25

GENERAL FUEL-ELEMENT DEVELOPMENT

S. J. Paprocki

The preparation of cermets containing 60 to 90 volume per cent of fuel dispersed in metal matrices is being studied. The prime objective of this study is the development of cermet structures possessing optimum physical and mechanical properties, with particular emphasis on the improvement of thermal conductivity.

The gas-pressure-bonding process is being utilized as a method for the bonding of niobium and molybdenum fuel elements and assemblies. Techniques have been developed for producing ductile bonds possessing grain growth across the bond interface. The current studies have been concerned with the investigation of spacer materials in an effort to develop unreactive materials that can be utilized as fuel-element separators during multiple plate bonding or as channel spacers in the bonding of subassemblies.

The kinetics of solid-phase bonding occurring during a bonding process involving temperature and pressure is being investigated. It is anticipated that the results from this study will have direct practical application to the gas-pressure-bonding process.

Fabrication of Cermet Fuel Elements

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

Evaluations of fabrication techniques for producing cermets of 90 per cent of theoretical density or better containing 60 to 90 volume per cent fuel are being made. Microstructures and physical and mechanical properties of the fabricated cores are being evaluated.

Two cermet cores (1 by 1 by 0.100 in.) of 80 volume per cent UO_2-molybdenum were pressure bonded for 3 hr at 2200 and 2400 F under a helium gas pressure of 10,000 psi. The green density of each core was approximately 70 per cent of theoretical. The core densities obtained during pressure bonding were 82 and 87 per cent of theoretical, with the higher density being obtained at 2400 F.

A similar result was obtained with a core of 80 volume per cent UO_2-niobium, which, when pressure bonded at 2200 F for 3 hr, increased in density from about 70 to 83 per cent of theoretical.

Additional cores of each matrix material (some incorporating spherical UO_2) will be prepared to further evaluate thermal conductivities and moduli of rupture by comparison with previously reported values.

Recent studies indicate that slip casting appears promising as a means of preparing cermet cores of a size larger than is feasible for preparation in conventional presses. The slip-cast cores show considerable green strength and can be handled

easily. Several castings of 80 volume per cent UO_2-stainless steel were prepared from slips of various pH values. The slip was prepared by dissolving 0.35 w/o ammonium alginate and 0.25 w/o polyvinyl alcohol in 24.85 w/o distilled water. After the solution was obtained, approximately 75 w/o of the 80 volume per cent UO_2-stainless steel powder mix was added and stirred in. The pH of the slips was varied over a range of 4 to 11, by adding HNO_3 or NaOH to each mixture before casting into plaster of Paris molds. The castings were allowed to dry overnight and then removed from the mold and dried for 3 hr at 200 F in a drying oven. A section was cut from the bottom of each casting, and a density measurement was obtained. The highest density (48.8 per cent of theoretical) was obtained on the casting with a pH of 11. After compacting this core at 50 tsi in a powder-compacting die, a density of 82 per cent of theoretical was obtained. Additional slips of high pH as well as slips in which the water content is varied are being prepared.

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

S. J. Paprocki, E. S. Hodge, and P. J. Gripshover

Molybdenum and niobium retain their strength at elevated temperatures and possess favorable nuclear properties. These characteristics make them potential cladding and structural materials for high-temperature-reactor applications. The gas-pressure-bonding technique is being studied as a possible method for cladding ceramic and cermet-type fuels with these metals.

Examination of earlier niobium-clad fuel plates containing UO_2-Al_2O_3 core inserts revealed that some void space could be observed at the core corners after bonding at 2100 F at 10,000 psi for 3 hr. Since these bonding conditions close all void space between two flat plates of niobium, it was necessary to determine if the void space observed resulted from insufficient flow of the niobium at these bonding conditions. A specimen was prepared which contained controlled amounts of void space. This void space was formed by machining slots of varying widths and depths into a flat plate of niobium. A matching plate was placed on the slotted piece of niobium and sealed in a stainless steel bonding container. Bonding was accomplished at 2100 F and 10,000 psi for 3 hr. Examination of this specimen revealed that all void space had been closed in each of the slots. Good bonding was evident in most areas of this specimen except within the machined slots. Earlier studies have shown that machined surfaces do not produce satisfactory bonds. However, this specimen did show that flow is sufficient at 2100 F and 10,000 psi to close void space within a fuel-plate specimen. It is believed that the void space in the first fuel plates prepared resulted from outgassing of the UO_2-Al_2O_3 cores used in these specimens during bonding. To determine if this assumption is correct, several fuel plates are being prepared at the present time which contain UO_2-Al_2O_3 cores which have been outgassed at 1650 F in vacuum prior to bonding. These specimens will be pressure bonded at 2100 F and 10,000 psi for 3 hr.

Earlier niobium specimens which utilized graphite as a barrier between the specimen and the bonding container exhibited extremely brittle properties after pressure bonding. It has been determined, however, that this brittleness was due to hydrogen pickup during pressure bonding and not due to the slight increase in carbon

content. For this reason, a specimen has been prepared incorporating graphite-coated Ti-Namel spacers. These spacers have the advantage that they can be pickled away from the niobium specimen after pressure bonding. After pressure bonding at 2100 F and 10,000 psi for 3 hr, there was no alloying of the spacer with the specimen. The ductility of this specimen very nearly approached that of the starting material. Metallographic examination indicated that some carbide formation had occurred at the surface of the niobium; however, the encouraging results of these specimens warrant further investigation.

A pickling solution of concentrated nitric acid under a layer of kerosene has proven to be the most satisfactory surface preparation for molybdenum specimens to date. However, this surface preparation does not produce satisfactory bonds consistently. For this reason, efforts have been made to devise a more satisfactory pickling solution for future molybdenum specimens. Results of these studies indicate that pickling in solutions of 2 parts nitric acid-1 part water and 3 parts nitric acid-1 part water produces a surface which is free of apparent oxide and stain. Several specimens have been prepared from molybdenum plates pickled in these solutions. Evaluation of these specimens after pressure bonding will determine the merits of these surface preparations.

Two edge-welded molybdenum specimens have been successfully pressure bonded. Bend tests showed these specimens to have a ductility equal to that of the starting material. Metallographic examination revealed numerous voids were present along the original bond interface. Since these specimens were fusion welded in a helium atmosphere, it is probable that the voids were caused by entrapped helium. This difficulty would be overcome by electron-beam welding in an evacuated chamber.

Factors Affecting Pressure Bonding

G. W. Cunningham and J. W. Spretnak

In an effort to obtain information on the kinetics of solid-phase bonding of metals by the application of heat and pressure, the process has been arbitrarily divided into three stages for detailed study. The stages may be briefly described as follows: (1) formation of a bond interface by placing the metal surfaces in intimate contact, (2) removal of excess voids or vacancies from the bond line, and (3) grain-boundary movement and grain growth across the original interface. At present, OFHC copper is being used in these studies, but later work will involve the use of iron, nickel, and zirconium.

In the studies on the first stage, specimens are bonded by pressing for periods of 5 min at various temperatures and pressures. Under these conditions, it should be possible to establish a relationship among pressure, temperature, and area of contact. It is then believed that a relationship can be established between some property of the metal, such as hot hardness, and the area of contact. Although the specimens in this study are of a simple configuration (a row of teeth resting on a flat surface), surface roughness can be expected to affect any general relationship and will have to be considered at a later date. The first specimens evaluated were bonded at 1000 F for 5 min at

pressures of 2,000, 4,000, 8,000, and 12,000 psi. A plot of pressure versus area of contact gives a smooth curve which is almost linear at high pressures. Other specimens are being prepared at 600, 800, and 1200 F. Hot-hardness measurements have been obtained over the range from room temperature to 1600 F. In addition to these studies on plastic flow and closure of the voids, annealing studies may provide information as to the rate of vacancy diffusion along the bond line, since specimens prepared as described above vary in size and opening of large voids. Thus, considering the voids as free surfaces, one can compute the distance over which vacancies will be eliminated from the bond interface in a specified time.

Copper specimens have been vacuum annealed at 1800 F to form very large grains and will be bonded and used to study the effect of grain orientation on bonding.

FF-1

FF. FUEL-CYCLE PROGRAM STUDIES

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

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

The objective of this program is to develop a fabrication technique which will maintain or improve the quality of ceramic, cermet, and dispersion fuel elements, while reducing manufacturing costs. The most promising method of fabrication for achieving these objectives appears to be the gas-pressure-bonding technique. The study is therefore directed toward the refinement and further development of the gas-pressure-bonding process to accomplish simultaneous densification and cladding of ceramic, cermet, and dispersion fuels with stainless steel.

Initial studies are concerned with the development of high-density green-compacted UO_2 ceramic cores of specific densities which will achieve a desired range of densities during pressure bonding. These UO_2 ceramic cores have been utilized for the development of Type 304 stainless steel-clad elements and assemblies of various shapes. The techniques developed for the UO_2 ceramic fuel can be applied to cermet and dispersion systems with a minimum of additional development work.

As reported previously, mixtures of several grades of UO_2 powder yielded cold-pressed compacts of up to 83 per cent of theoretical density. To further investigate powder variables a cursory study of the effects on the cold-compacted and pressure-bonded densities of UO_2 of varying the particle-size distribution and type of binder was initiated. Using pressures of 55 and 50 tsi, Spencer fused UO_2 with a particle distribution of 92.50 w/o minus 60 plus 80 mesh, 6.25 w/o minus 150 plus 200 mesh, and 1.25 w/o minus 325 mesh powders exhibited cold-pressed densities of 86.5 and 86.4 per cent of theoretical. These compacts, which contained Ceremul "C" as an internal binder, were strong and easy to load. Compacts of Spencer fused UO_2 of minus 20-mesh particles were cold pressed using a Carbowax 6000 and methyl alcohol binder. At a 55-tsi pressure a density of 85.9 per cent of theoretical was realized. Densities of 84 to 85 per cent were prevalent through a pressure range from 25 to 55 tsi. Pressures as low as 10 tsi were used to give a substantially sound compact, which is significant when compared with the 20 to 25-tsi minimum compacting pressure required in previous work for other powder mixtures using other binders. Carbowax 6000-methyl alcohol solutions gave superior loading and handling characteristics when compared with all other binders. The density resulting from pressure bonding these compacts has not yet been determined.

Details for a series of pressure-bonded stainless steel-clad UO_2 assemblies are reported in Table FF-1. Assemblies PB-1, PB-2, and ODID-1 (tubes) along with Assembly PB-7 (flat plate) contained tap-packed UO_2 cores ranging from about 64 to 70 per cent of theoretical density. The remaining assemblies, all tubes, contained cold-compacted pellets with densities ranging from 77.8 to 86.5 per cent of theoretical.

TABLE FF-1. DENSIFICATION OF UO_2 BY PRESSURE BONDING[a]

Assembly	Description of UO_2	Compact	Compacting Pressure, tsi	Height, in.	Green Density		Pressure-Bonded Density[b]	
					G per Cm^3	Per Cent of Theoretical	G per Cm^3	Per Cent of Theoretical
ODID-1	MCW special dense (minus 20 mesh)	--	Tap packed	1.650	7.03	64.1	8.60	78.5
PB1	75 w/o MCW special dense (plus 325 mesh) and 25 w/o MCW ceramic grade (minus 325 mesh)	--	Tap packed	3.370	7.04	64.1	10.41	95.1
PB2	Same	--	Tap packed	3.030	7.05	64.3	10.45	95.4
PB7	Spencer fused (minus 20 mesh) (square plate 1.19 by 2.13 in.)	--	Tap packed	--	7.69	70.0	9.93	90.6
28[c]	MCW pellets	U-1175	(d)	0.513	8.54	77.9	10.65	97.2
		U-1176	(d)	0.508	8.59	78.4		
		U-1177	(d)	0.515	8.53	77.9		
		U-1178	(d)	0.514	8.52	77.8		
30	Spencer fused as a mixture of 1.25 w/o minus 325-mesh, 6.25 w/o minus 150 plus 200-mesh, and 92.50 w/o minus 60 plus 80-mesh powder	U-1208	55	0.422	9.46	86.5	9.60[e]	87.6[e]
		U-1209	50	0.423	9.45	86.4		
31	MCW special dense as a mixture of 1.25 w/o minus 325-mesh, 6.25 w/o minus 150 plus 200-mesh, and 92.50 w/o minus 60 plus 80-mesh powder	U-1213	55	0.453	8.84	80.6	9.00	82.0
		U-1214	50	0.450	8.81	80.4		
		U-1215	45	0.455	8.74	79.8	9.19	83.7
		U-1216	40	0.456	8.79	80.2		

(a) All assemblies pressure bonded at 2100 F for 3 hr at 10,000 psi except where indicated.
(b) Densities determined from measurements of assembly and original weight of UO_2.
(c) Pressure bonded at 2300 F for 2 hr at 10,000 psi.
(d) Pellets received from MCW - unknown history.
(e) Void space in tube - densification may be affected by nonuniform end pressure.

Pressure-bonded UO_2 densities of up to 95.4 per cent of theoretical for the tap-packed powders and up to 97.2 per cent of theoretical for the compacted pellets were attained. These density results are being verified by pycnometer measurements. Visual examination of the flat-plate assembly revealed a uniform thickness with little distortion. Two tap-packed tubes, Assemblies PB-1 and PB-2, exhibited warpage and wrinkling of the stainless steel cladding. Assembly 28 showed a very uniform surface with no wrinkling, whereas Assemblies 30 and 31, containing stainless steel spacers, exhibited a nonuniform diameter. These latter two assemblies contained pellets that were compacted from a powder mixture with a particle-size distribution that, as mentioned earlier, produced a high green density. These pellets exhibited little densification on pressure bonding.

The effect of different surface preparations on the quality of Type 304 stainless steel bonds resulting from pressure bonding is being investigated. Specimens are being evaluated by metallographic observations and bend tests. Samples prepared thus far include as-rolled, belt-abraded, and vacuum-annealed surfaces. These have been gas-pressure bonded for periods of 3 hr at 2000 and 2100 F at 10,000 psi. The as-rolled specimens have been evaluated. Surface preparation of the as-rolled material included a 2-min pickle of the components in an aqueous solution of 10 parts nitric acid and 2 parts hydrofluoric acid heated to 120 F. The components were then processed through a decrease-and-wash cycle. The bonds obtained under both sets of pressure-bonding conditions were shown adequate in both bend and peel tests. However, metallographic examination revealed that there was approximately 25 per cent grain growth across the interface of the specimens bonded at 2000 F, while the interface of the specimen bonded at 2100 F exhibited 70 to 80 per cent grain growth.

The fabrication of small rod, tube and flat-plate elements, and flat-plate assemblies of stainless steel-clad UO_2 has been accomplished by gas-pressure bonding. By utilizing high-density green-pressed UO_2 cores to minimize densification effects, the problems of dimensional tolerances, shrinkage, warpage, and deformation of the cladding are being given extensive consideration. Methods of improving dimensional properties of the clad elements by varying container designs and using compacted stainless steel end plugs and spacers with densities equivalent to the core are being investigated.

DEVELOPMENT OF URANIUM CARBIDE-TYPE FUEL MATERIALS

F. A. Rough and W. Chubb

The uranium carbides and, particularly, uranium monocarbide are being developed for use as power-reactor fuels under the sponsorship of the AEC Fuel-Cycle Development Program. This type of reactor fuel appears to have promise as a low-cost, long-life, and easily reprocessable fuel for nuclear reactors using sodium, organic liquids, or inert gases as coolants. This integrated program of research is designed to eliminate most of the technological uncertainties which have prevented the use of this new and relatively unknown material. The research is directed toward developing techniques for preparing and shaping the carbides both by powder metallurgical and by ordinary melting and casting techniques. It will also produce information with respect to the engineering

properties of uranium carbides, including their physical properties, their mechanical properties, their resistance to chemical attack, and their resistance to damage by neutron irradiation.

Information obtained during the first 6 months of this program has been collected and issued in the form of a report.* During the last few weeks, additional information has been obtained. Last month, it was reported that 2-in.-diameter castings had been prepared, that the carbides react with ferrous alloys at temperatures as low as 1000 C, that the activation energy for interdiffusion in uranium carbides, where carbide layers are formed between graphite and carbon-saturated liquid uranium, is about 79 kcal per mole, and that metallographic structures of the carbide suggest that minor revisions in the published uranium-carbon constitutional diagram may be necessary.

During the last month, uranium carbide castings, weighing from 2 to 6 lb, have been made in the form of hexagonal, rectangular, and cylindrical rods by inert-electrode arc-melting and casting techniques. Hardness and transverse-rupture-strength data obtained on carbides, both in the as-cast condition and as annealed at 1550 C, indicate that maximum hardness and strength in the uranium-carbon system occurs near the composition of the sesquicarbide (7 w/o carbon). Corrosion rates of the uranium carbides in various media appear to vary erratically over wide ranges without any apparent reason. Corrosion rates for alloys containing from 4.6 to 9.2 w/o carbon vary from 30 to 1600 mg/(cm^2)(hr) in water at 60 C, from 0.03 to 2 mg/(cm^2)(hr) in ethylene glycol at 150 C, and from 0.2 to 6 mg/(cm^2)(hr) in Santowax R at 350 C.

Alternate Fabrication Methods for UC

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

The primary objectives of this program are to develop techniques for preparing UC powder and to determine the conditions required to prepare high-density (95 per cent of theoretical or greater) UC bodies by powder-metallurgy processes.

During the past month, the reaction of uranium metal with methane was investigated. Also, sintering studies were conducted on arc-cast and crushed UC powder in an attempt to evaluate the effect of time and temperature on sintered densities.

An analysis has been obtained on UC powder prepared by reacting uranium-metal powder with methane gas at temperatures ranging from 650 to 700 C. A chemical analysis resulted in a total carbon content of 5.95 to 6.01 w/o and a "free carbon" content of 0.14 to 0.17 w/o. An X-ray diffraction analysis showed very strong UC, strong UC_2, faint UO_2, no uranium-metal or graphite lines, and two sets of faint unknown lines. In addition, initial studies were conducted leading to the possible formation of UC powder by the reaction of uranium metal and propane gas.

*Rough, F. A., and Chubb, W., "Progress on the Development of Uranium Carbide-Type Fuels", BMI-1370 (August 21, 1959).

FF-5

Powder was prepared by crushing arc-cast UC buttons of near stoichiometric composition for sintering studies. Green-pressed cores of uranium-4.79 w/o carbon were vacuum sintered 1/2 hr at 1680 C. Sintered densities of 65.8 and 67.9 per cent of theoretical were measured, while corresponding green densities were 61.9 and 67.0 per cent of theoretical. Additional cores are being prepared for sintering at higher temperatures for various lengths of time.

Melting and Casting Techniques for Uranium-Carbon Alloys

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

Reliable techniques for the production of high-quality cast shapes of uranium carbide are being developed. Relatively large castings of varied shapes have been produced during the past month by inert-electrode skull-type arc-melting techniques. In addition, an investigation of the effects of alloying upon the castability of uranium monocarbide has been initiated.

The inert-electrode skull-type arc-melting technique was successfully employed in the casting of elongated shapes with round, square, and hexagonal sections. The cross-sectional areas of these shapes are approximately 2 sq in., and the castings are as long as 8 in. Graphite molds were used in preparing these castings, and it was noted that surface quality improved with increased mold temperature up to the highest temperature imposed (2400 F). No attack or erosion of the mold was observed at these temperatures.

While the emphasis in this work to date has been placed on the development of casting techniques, limited studies have been made with respect to control of composition. The carbon content of different castings of the same nominal composition has varied from 5.5 to 8 w/o. This variation in composition is caused by erosion of the graphite electrode tip and of the graphite crucible liner. To reduce these variations, plans are being made to eliminate one source, the graphite crucible, by melting the UC in direct contact with the water-cooled copper crucible. Recently, the life of the carbon electrode was increased and loss of electrode carbon to the melt was reduced by dipping hot electrode tips into molten uranium. Subsequent heating produced a coating of uranium carbide on the surface of the electrode; however, this coating made the arc more difficult to control. The cause of this difficulty is not understood and further studies are planned.

Inert-electrode drop-casting techniques were employed in an attempt to produce 3/8-in.-diameter castings of uranium monocarbide containing alloy additions of 1, 10, and 50 mole per cent Al_4C, Cr_3C_2, Mo_2C, NbC, TaC, TiC, VC, and ZrC. Very poor surfaces and severe cracking were observed in all specimens containing 50 mole per cent alloy additions. Some cracking and comparatively poor surface characterized the alloys containing 10 mole per cent additions. The surface quality of the alloys containing 1 mole per cent additions was good. The type of imperfections exhibited by some of the alloy specimens are believed to be defects that can be eliminated by changes in the molding procedures. If the properties of the alloy materials are sufficiently promising, further efforts to obtain sound cast shapes of these alloys will be expended.

FF-6

Metallurgical and Engineering Properties of Uranium Monocarbide

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

A study of the properties of uranium-carbon alloys is in progress in an effort to determine and perhaps improve the properties of these alloys. The study is concerned with the effect of such variables as impurity content, carbon content, and heat treatment on density, resistivity, thermal conductivity, and corrosion resistance.

The effects of carbon content and heat treatment have been investigated by measuring the density, resistivity, transverse rupture strength, and Knoop hardness of cast specimens containing from 2.2 to 9.2 w/o carbon before and after 1-hr anneals at 1550 C. Data from these studies are presented in Table FF-2.

TABLE FF-2. PROPERTIES OF URANIUM-CARBON ALLOYS AS A FUNCTION OF HEAT TREATMENT AND CARBON CONTENT

		Properties As Cast				Properties After Annealing		
Specimen	Carbon Content (Balance Uranium), w/o	Density, g per cm^3	Resistivity, microhm-cm	Knoop Hardness, kg per cm^2	Transverse Rupture Strength, psi	Density, g per cm^3	Knoop Hardness, kg per cm^2	Transverse Rupture Strength, psi
37	2.2	15.4	46.7	541	20,000	--	--	--
38	4.6	13.7	44.2	896	13,000	13.6	644	9,600
39	5.0	13.4	43.2	--	13,500	--	--	--
40	5.7	13.1	54.8	787	14,200	13.2	758	12,200
41	6.6	12.5	52.4	886	15,200	13.0	1,167	13,500
42	7.0	12.4	57.9	819	15,500	12.7	1,077	13,200
43	7.8	11.9	72.2	766	8,300	12.3	1,051	9,300
44	8.5	11.8	77.1	--	8,800	11.7	461	5,300
45	9.2	10.6	124.0	249	12,000	11.5	511	6,400

Maximum strength and hardness were evidenced before and after annealing by material containing about 7.0 w/o carbon. The rupture stress values were considerably below those previously obtained from specimens annealed for 1 hr at 1500 C, and further heat-treatment studies at 1400, 1600, and 1800 C are planned on material near the 4.8 and 7.0 w/o compositions. Metallographic examinations of the annealed alloys indicate that the 7.0 w/o carbon alloy is approximately 95 per cent U_2C_3.

Preliminary corrosion tests have been completed on specimens in water, Santowax R, and ethylene glycol. The corrosion rates observed are presented in Table FF-3 and indicate that annealing has little effect on corrosion rates.

Alloy additions of 1, 10, and 50 mole per cent Al_4C, Cr_3C_2, Mo_2C, NbC, TaC, TiC, VC, and ZrC have been made to uranium monocarbide. Castings 1/4 in. in diameter and 2 in. long have been produced by drop-casting techniques. Castings containing 10 and 50 mole per cent alloy additions had poor surfaces, while the 1 mole per cent additions had no observable effect on surface quality.

TABLE FF-3. EFFECT OF CARBON CONTENT AND HEAT TREATMENT ON THE CORROSION RATE OF URANIUM-CARBON ALLOYS IN VARIOUS MEDIA

Specimen	Carbon Content (Balance Uranium), w/o	Corrosion Rate in Medium Shown, mg/(cm^2)(hr)				
		As-Cast Material		Annealed Material		
		Water at 60 C	Santowax R at 350 C	Water at 60 C	Santowax R at 350 C	Ethylene Glycol at 150 C
38	4.6	414	--	118	(a)	0.32
39	5.0	(b)	6.5	--	--	--
40	5.7	819	--	1,100	4.5	0.37
41	6.6	28.3	0.20	384	0.31	0.03
42	7.0	139	4.3	70.8	1.6	0.51
43	7.8	80.8	--	230	0.31	0.48
44	8.5	1,623	0.35	918	0.40	2.1
45	9.2	488	0.70	441	2.6	0.55

(a) Specimen completely disintegrated in 1 week.
(b) Specimen completely disintegrated in 1 hr.

A combination of low strength and high resistivity was obtained for many of the alloys. It is believed that this is the result of internal cracking in the specimens, and is not necessarily characteristic of the material. Additional work is planned to recheck the values obtained and to determine if the alloy additives have any effect on the corrosion resistance of the UC base material.

Uranium Monocarbide Diffusion Studies

W. Chubb, R. W. Getz, and F. A. Rough

Diffusion rates in uranium monocarbide are of interest in predicting the elevated-temperature behavior of this material. The rate of interdiffusion of uranium and carbon has been determined by measuring the rate of growth of the carbide layers formed between molten uranium and graphite at temperatures from 1600 to 2000 C. The results of this investigation were reported last month. Currently, the rate of self-diffusion of uranium in uranium monocarbide is being investigated using a tracer technique.

Mechanical and physical techniques for determining the rate of self-diffusion of uranium in uranium monocarbide are being developed. Depleted uranium (0.04 per cent uranium-235) has been fabricated into a suitable form for the melting and casting of 1/2-in.-diameter rods of uranium monocarbide. A 0.001-in.-thick foil of enriched uranium metal (93 per cent uranium-235) will be placed between two 1/4-in. lengths of depleted uranium monocarbide rods to form diffusion couples. The couples will be bonded in a graphite jig at 1400 C or above. Bonding has been achieved using this technique on natural uranium samples, but the bonds appear to be contaminated with a small amount of UO_2. Additional couples have been prepared, and metallographic examination indicates that satisfactory bonds can be achieved, if special precautions are taken to exclude oxygen.

FF-8

Bonded diffusion couples will be heated at a series of temperatures between 1600 and 2000 C. After suitable diffusion heat treatments the couples will be mounted in Bakelite, and analytical samples will be obtained by grinding the couple on a silicon carbide block using nitric acid to dissolve the uranium carbide. Samples obtained in this manner have been analyzed for uranium content. The analytical results have demonstrated the suitability of the sampling technique, but have indicated two areas where further refinements are needed. Changes in the procedure have been made, and additional samples have been prepared and submitted for analysis. The purpose of these preliminary tests is to determine if all the uranium in a measured length of sample ground from the couple is recovered in the nitric acid solution.

Two hundred and forty samples, each representing a 1-mil length of uranium monocarbide, will be taken from each diffusion couple by the grinding technique described. The samples of interest, containing the enriched uranium diffusion zone, will be identified by measuring the gross gamma activity. The samples selected by this technique will be activated in the BRR, and the concentration of the uranium-235 in the original samples will be established more precisely by gamma counting for fission-product activity. From these data, it will be possible to calculate the rate of self-diffusion of uranium in uranium monocarbide.

To date, standards containing various concentrations of the two uranium isotopes have been prepared and irradiated. Plots of gamma activity versus isotope concentration have been prepared.

Future work will include the further development of procedures for the preparation of diffusion couples, the establishment of reliable sampling techniques, the casting of uranium monocarbide rods from depleted uranium, and the preparation of diffusion couples containing enriched uranium.

Irradiation Effects in UR

A. E. Austin and C. M. Schwartz

A study of the effects of irradiation on the structural properties of UC is under way. Preparations are being made to carry out X-ray diffraction examinations. Electron microscopic examination of irradiated specimens is in progress.

The use of low-angle scattering is being investigated for the detection of voids or gas bubbles. For this purpose, solid solutions of ZrC-UC have been made, and thin sections have been prepared for examination.

GG-1 and GG-2

GG. VOID-DISTRIBUTION AND HEAT-TRANSFER STUDIES

D. V. Grillot, R. Wooton, H. M. Epstein, D. A. Dingee,
and J. W. Chastain

Subcooled boiling void-distribution and heat-transfer research is being conducted for the Euratom-U.S. Joint Research and Development Board. During the past month the transformers, electric lines, and controls required for the operation of the loop were installed and tested. The power to the test section is continuously variable from 0 to 150 kw. The power to the preheater can be varied in steps from 0 to 30 kw. A continuous range of inlet temperatures to the test section can be obtained by varying the preheater power and the amount of fluid bypassing the heat exchanger.

Void fraction in the test section versus the counts from a Geiger tube was calibrated using polyethylene to simulate water. A magnet is used to deflect the praseodymium-144 beta particles away from the beam of gamma rays in order to reduce the background. In a channel formed by 0.010-in.-thick stainless steel side plates, a change of 0.00025 in. in the thickness of a 0.050-in.-thick strip of polyethylene can de detected with this system in 5 min of counting. The stability of the system was checked and found satisfactory.

A pressurized ionization chamber which is highly selective against gammas has been designed and will be constructed in an attempt to improve the efficiency of the detecting system. With a more efficient detector, the exposed area of the test section may be reduced, giving more precise information on position.

It is expected that checkouts and improvements on the system will continue through next month.

H-1

H. PHYSICAL RESEARCH

F. A. Rough

Physical research supported by the AEC Division of Research is reported in this section. In the study of niobium-nitrogen reaction kinetics, special attention is being given to the effects of surface condition in order to obtain consistent results in the range of 200 to 650 C. Two new reports devoted to the investigation of methods of preparation of high-purity single crystals of UO_2 are included this month. Two general methods of vapor deposition and fusion are to be studied.

Niobium-Gas Reactions

W. M. Albrecht and W. D. Goode

A fundamental investigation is being made of the reactions of nitrogen with niobium. The reaction kinetics and mechanism are being determined. Reaction rates, diffusion coefficients, and solubility of nitrogen in niobium in the range 675 to 1600 C have been described in a recent topical report.*

Experimental work to determine the reaction rates below 675 C are continuing. The rates determined in the initial experiments were erratic and followed no particular rate law. It was believed that this was due to slight variations in surface condition from sample to sample. In order to obtain uniformly clean surfaces, the reaction system was altered so that a sample could be vacuum annealed at elevated temperatures just prior to the rate determination. For the experiments, the sample was sealed in the reaction tube which was then evacuated to about 10^{-5} mm of mercury for 16 hr. Then the sample was vacuum annealed at 1100 C for 45 min and cooled to the experimental temperature. The reaction was initiated by adding high-purity nitrogen to the reaction system. The reaction rate was determined by following the weight gain on a microbalance. Initial experiments with this method indicate that the reactions follow the logarithmic rate law**,

$$w\ (\mu g/cm^2) = k \log (a\,t + 1)\ .$$

For experiments at 300 to 600 C, the logarithmic rate constant, k, varied from 1.9 to 7.5 μg per cm^2. Further experiments are being made to determine the reaction rates in the range 200 to 650 C. Also, X-ray diffraction patterns are being obtained on the surfaces of reaction samples to identify reaction products.

*Albrecht, W. M., and Goode, W. D., Jr., "Reaction of Nitrogen With Niobium", BMI-1360 (July 6, 1959).
**Kubaschewski, O., and Hopkins, B. E., Oxidation of Metals and Alloys, Academic Press Inc., New York (1953), p 38.

H-2

Growth of UO_2 Crystals From the Vapor Phase

C. A. Alexander and R. B. Filbert, Jr.

A program has been initiated this month to produce UO_2 crystals by the process of vaporization. Vaporization in the UO_2 phase region can occur by either direct sublimation as $UO_2(g)$ or by oxidation to UO_3 and evaporation as $UO_3(g)$. Calculations indicate that at temperatures below 2000 K and with oxygen pressures of the order of 10^{-4} to 10^{-8} atm, the vapor above UO_{2+x} would be almost entirely UO_3. Since UO_3 is not stable in the condensed phase at temperatures greater than a few hundred degrees centigrade, the UO_3 would decompose after condensation. Hence, by suitable control of the experimental conditions, it should be possible to produce crystals of UO_2.

The purpose of this investigation is to determine the experimental conditions necessary to produce large single crystals of UO_2. The use of UO_3 rather than UO_2 as the volatile oxide results in an evaporation rate increase of several orders of magnitude, but at the same time imposes conditions upon the design of the experimental equipment.

Equipment for initial experiments is under construction, and during the next month it is planned to begin experimental work in an effort to produce crystalline UO_2 from vapor-phase deposition of UO_3.

Fusion Methods to Prepare Single Crystals of UO_2

W. P. Allred and B. Paris

The primary purpose of this investigation is to grow monocrystals of UO_2 by use of fusion methods. It is possible that these crystals can be grown by floating-zone melting, using either electron-beam melting or high-frequency induction heating to establish the molten zone. Purification of the UO_2 will also be studied along with the growing of the crystals. As soon as crystals become available, studies on the composition of the crystals will be made to determine how near the material is to stoichiometric UO_2.

Semiconducting materials such as UO_2 decrease in resistivity at high temperatures. It is therefore essential to heat the UO_2 to a high temperature before inductive coupling is possible. The UO_2 to be zone refined will be held in a vertical position by molybdenum holders. Heating will be achieved by passing an a-c current through the sample.

For preliminary studies, silver paste was painted on the ends of the UO_2 to lower the contact resistance with the molybdenum holders. After other problems are solved, the ends of the UO_2 will be coated with uranium to reduce this resistance.

Initial experimental work has included the following: an induction-heated zone refiner has been modified for preliminary studies with UO_2. With the changes in equipment experiments can now proceed to establish conditions necessary for growing of

single crystals of UO_2. One sample was heated to approximately 1800 C by resistance heating, and it became obvious that a current-limiting device is necessary. This is now under construction.

It is also planned to use the electron-beam zone refiner as a possible method of growing and purifying the UO_2 crystals. The equipment is being modified such that the UO_2 specimen can be heated by an a-c current. This heating is necessary to raise the entire specimen to a high temperature in order to eliminate thermal shock. It is possible that, since electron-beam melting is done in a high vacuum, this method of melting will be impractical because of the vapor pressure of UO_2. However, preliminary studies of UO_2 in this system indicate that there is a possibility that the vapor pressure is low enough to allow electron-beam melting. If this is the case, the problems of zone refining and crystal growing will be greatly reduced.

Studies on both electron-beam melting and induction melting will be continued during November.

I. SOLID HOMOGENEOUS FUELED REACTORS

W. S. Diethorn and W. H. Goldthwaite

As part of the Pebble-Bed Reactor program, Battelle is investigating the fission-gas retention and mechanical and physical properties of fueled-graphite spheres supplied by commercial vendors.

LABORATORY EVALUATIONS OF FUELED-GRAPHITE SPHERES

J. F. Lynch, M. C. Brockway, S. Rubin,
W. C. Riley, and W. H. Duckworth

Tests of the strength, impact resistance, and coating integrity of coated fueled-graphite spheres, described in previous monthly reports, are continuing.

A self-welding test of two SiC-coated spheres of the same type reported last month was completed. With the exception of temperature, 2000 F, this test was carried out under the same conditions as the test reported in BMI-1381. The spheres did not adhere to each other. The coating at the contact point of the two spheres did not leak in a hot-oil test.

Apparatus for the study of the gas permeability of sphere coatings at high temperature was completed this month.

Laboratory work is under way on the development of apparatus for the investigation of sphere coating integrity at high internal-sphere gas pressures. Gas pressure will be applied to the coating of a fueled-graphite sphere by drilling a radial hole to the center of the sphere. Methods of sealing the gas supply to the cored sphere are under development.

EVALUATION OF METAL-COATED UO_2 PARTICLES

A. F. Gerds, J. Koretzky, and F. W. Boulger

As reported last month, UO_2 particles coated with either nickel, nickel-chromium, or niobium react with carbon at 1700 F. The investigation of alumina-coated particles is continuing.

I-2

FABRICATION DEVELOPMENT OF Al_2O_3-CLAD UO_2 FUEL PARTICLES

A. K. Smalley and W. H. Duckworth

FISSION-PRODUCT RELEASE FROM FUELED-GRAPHITE SPHERES

W. S. Diethorn

Fission-gas retention and the effect of radiation on fueled-graphite spheres is being investigated in neutron-activation experiments and in sweep-gas and static irradiation capsules.

Neutron-Activation Studies

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

Neutron-activation experiments completed this month are reported in Table I-1. Sphere FA-14 was fueled with a 11:1 weight ratio of ThO_2-UO_2. Sphere FI-2 contained a slug of UC-UC_2 fuel in a cored and plugged hole in the sphere. Prior to neutron activation this sphere leaked in a hot-oil test. The release observed with this sphere agrees very well with a calculated value of 0.025 per cent based on complete postirradiation release of fission gas recoiled from the slug.

The bare UO_2 particles were prepared by a method similar to that used to prepare the UO_2 cores in the Al_2O_3-clad UO_2 particles, neutron activated and reported last month (BMI-1381). Particle diameter is 600 to 900 μ. The cumulative release from these bare particles is about 2300 times greater than the cumulative release from clad particles reported in BMI-1381.

TABLE I-1. NEUTRON-ACTIVATION RESULTS[a]

Specimen	Coating	Postirradiation Heat-Treatment Temperature[b], F	Time at Temperature[c], min	Fission-Gas Release[d], per cent of xenon-133
FA-14, 373	None	800	15	0.1
		1500	158	0.2
F1-2, 1	ZrC, 10 mils	1500	30	0.03
UO_2 particles	None	1600	15	0.3
		1800	15	Nil
		2500 (1 day)	30	2.0

(a) Each specimen irradiated once.
(b) Time in parentheses is delay time between this heat treatment and prior heat treatment.
(c) Zero time begins when heat is applied to specimen. Heatup time is 5 to 10 min for all specimens.
(d) Number of atoms released divided by number of atoms present at beginning of heat treatment, times 100.

In-Pile Capsule Experiments

D. B. Hamilton, D. Stahl, G. E. Raines, R. J. Burian,
and W. H. Goldthwaite

SP-3

Metallographic examination of Coated Spheres FA-6, 18E and FA-8, E4 is nearly complete. Results will be available next month.

SP-4

No significant change in the operating performance of this capsule has occurred since the last monthly report. This BRR experiment has been rescheduled and will continue to January, 1960.

SP-5 and SPH-1

The rate of effort on these two capsules was low in October. There is nothing new to report.

SPF-1

The objective of the low-flux SPF capsule series was described in BMI-1377.

I-4

The first capsule in this series, SPF-1, was inserted in the BRR on October 14. The effective thermal-neutron flux in the sphere is 3×10^{10} nv, estimated from cobalt-wire measurements at the outer capsule wall. This capsule contains a fueled-graphite sphere, FA-20, coated with a 5-mil layer of dense carbon. The fuel is 100 to 500-μ-OD UO_2 dispersed uniformly in the graphite matrix. Capsule performance is good, the sphere temperature running about 1500 F.

In this experiment helium flows over the heated sphere and into an out-of-pile refrigerated charcoal trap where the fission gases are removed. Fission gases are identified and assayed with a gamma spectrometer. The trap is removed for analysis after 30 min of helium flow. The trap is replenished with fresh charcoal after each analysis. Fission-gas-release data are summarized in Table I-2.

TABLE I-2. SUMMARY OF SPF-1 FISSION-GAS-RELEASE DATA

Sample	Collection Date (1959)	R/B[b]				
		Krypton-87	Krypton-88	Krypton-85m	Xenon-135	Xenon-133
1[a]	10-14	8.6×10^{-3}	1.7×10^{-3}	27×10^{-3}	24×10^{-3}	145×10^{-3}
2	10-15	9.7×10^{-3}	1.3×10^{-3}	41×10^{-3}	19×10^{-3}	17×10^{-3}
3	10-16	8.5×10^{-3}	0.8×10^{-3}	12×10^{-3}	16×10^{-3}	40×10^{-3}
4	10-18	(c)	(c)	26×10^{-3}	28×10^{-3}	26×10^{-3}
5	10-19	8.8×10^{-3}	1.5×10^{-3}	66×10^{-3}	63×10^{-3}	39×10^{-3}

(a) Collected 9 hr after capsule startup.
(b) R = the number of atoms released per second from the sphere.
B = the number of atoms generated per second in the sphere.
(c) Delay between sample collection and analysis prevented analysis.

With the exception of xenon-133, R/B for each species changes very little over this 5-day period. This capsule experiment will be continued, and the effect of temperature will be studied at one lower temperature.

SPF-2

A SiC-coated fueled sphere has been selected for the second capsule in the SPF series. Irradiation of this capsule will be postponed until the SPF-1 experiment is completed.

J-1

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

CORROSION STUDIES OF THE FLUORIDE-VOLATILITY PROCESS

C. L. Peterson, P. D. Miller, W. N. Stiegelmeyer, and F. W. Fink

The evaluation of the corrosion of various articles of process equipment from the ORNL Volatility Pilot Plant is being carried out as a program of assistance to the Chemical Technology Division.

The evaluation of the Unit Operations INOR-8 hydrofluorinator has been continued. Intergranular attack of about 8 mils is visible on the inner walls of the shell in the vicinity of the interface. Intergranular penetration as deep as 13 mils occurred on the inside of the draft tube. Heavy pitting was observed on the inner wall of the shell. Most of this occurred in the interface region with a maximum pit depth of 72 mils. Pitting was observed in other submerged areas and on both sides of the draft tube. The greatest metal loss or penetration, which occurred in the pitted area around the interface, was in the neighborhood of 70 to 80 mils. Neglecting the penetration by the pitting, which actually occurred during the final INOR-8-10 run in which zirconium was absent, the general metal penetration was much less. The evaluation of this vessel is being continued.

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

R. A. Ewing, H. B. Brugger, and D. N. Sunderman

Sulfex Process

One "cold" Sulfex run has been conducted at the hot cell, primarily to check the performance of the dissolution apparatus when remotely operated. The desirability of several minor changes, which have now been made, were indicated by this test. Unlike the laboratory tests, there was a substantial carry-over of sulfate ion to the core dissolution in this test. As a result heavy crystallization of thorium sulfate occurred in the core solutions, and dissolution of the core was incomplete (3.0 g residue).

Attempts to develop a lanthanum fluoride carrier-precipitation method for separation of thorium from sulfate cladding solutions have been abandoned. A solvent-extraction method in which thorium is extracted by triisooctylamine is now being used.

Some analytical results (Table J-1) are now available on the first five exploratory "cold" dissolutions of 7-in. prototype Consolidated Edison pins.

J-2

TABLE J-1. ANALYSES OF SOLUTIONS FROM DISSOLUTIONS OF CONSOLIDATED EDISON FUEL PINS

	Run				
	Sulfex C-1	Sulfex C-2	Sulfex C-5	Darex C-3	Darex C-4
Cladding Solution					
Volume, ml	300	312	~280	465[a]	580
Uranium					
Concentration, g per liter	0.0195	0.0166	--	0.023	0.0067
Total, g	0.0059	0.0052	--	0.0107	0.0039
Loss, w/o	0.36	0.31	--	0.60	0.20
Thorium					
Concentration, g per liter	0.035	0.055	0.062	0.074	0.065
Total, g	0.0102	0.017	0.017	0.034	0.038
Loss, w/o	0.027	0.044	~0.045	0.087	0.095
Iron					
Concentration, g per liter	46.0	52.0	46.0	31.5	21.2
Total, g	13.8	16.2	12.9	14.7	12.3
End-Cap Residue, g	2.52	2.26	2.32	3.34	0.15
Core Solutions					
First Core Solution					
Volume, ml	166	160	~125	160	165
Thorium					
Concentration, g per liter	196	195	169	231	204
Total, g	32.6	31.2	21.1	36.9	34.7
Second Core Solution					
Volume, ml	42	94	100	50	50
Thorium					
Concentration, g per liter	83	64.5	104	31.5	105
Total, g	3.5	6.1	10.4	1.6	5.2
Total Thorium, g	36.1	37.3	31.5	38.5	39.9
Composite Core Solution[b]					
Volume, ml	305	254	~225	255	260
Thorium					
Concentration, g per liter	122	152	146	154	154
Total, g	37.2	38.6	32.7	39.2	40.1
Uranium					
Concentration, g per liter	5.31	6.68	7.50	6.78	7.58
Total, g	1.62	1.70	1.68	1.77	1.97
Sulfate, g per liter	1.6	2	12	--	--
Iron, g per liter	0.32	0.33	2.7	0.49	0.26
Free Acidity, N	7.8	7.9	8.0	7.8	7.8
Insoluble Residue, g	0.33	0.26	3.0	0.09	0.12

(a) 400 ml of Darex solution instead of usual 500 ml.

(b) Prepared by taking 1/5 aliquots of core and wash solutions.

J-3 and J-4

Darex Process

One "cold" familiarization Darex run is scheduled at the Battelle Hot-Cell Facility before commencing dissolution tests of irradiated pins. Although scheduling difficulties have delayed this run, it is expected that it will be possible to conduct it shortly. Following this run, dissolution tests of the Consolidated Edison pins supplied by ORNL will begin. First dissolutions are to be via the Sulfex process; subsequently, the Darex process will be employed.

Several CETR pins, irradiated 2 cycles at the MTR, are scheduled to be subjected to dissolution studies following the ORNL pins. In connection with other work at Battelle, burnup analyses are being performed on these pins. No difficulties were encountered in dissolution of a number of earlier samples from a loop test. However, the first core sample of a later series, from a static capsule experiment, has been found to be insoluble in either 13 M HNO_3, 0.04 M NaF or 15 M NHO_3, 0.10 M NaF. A small (~0.4 g) core cross-section sample appeared to be virtually unaffected after approximately 20 hr of exposure to these boiling solutions. In contrast to the results from this solid chunk, approximately 1 g of small chips from the same specimen was dissolved in about 6 hr by standard techniques. The reported composition of this core is 93.5 w/o ThO_2-6.5 w/o UO_2 (92.7 per cent enriched) at approximately 92 per cent of theoretical density.

Several factors may be contributing to the observed insolubility. Among them are radiation effects, temperature history, and density. Which of these may be most significant is not yet known. However, if these preliminary indications are confirmed, some process modifications may be necessary before dissolution tests are conducted on these pins.

K-1

K. DEVELOPMENTS FOR SRE, OMRE, AND OMR

EVALUATION OF URANIUM MONOCARBIDE AS A REACTOR FUEL

F. A. Rough

Irradiation of uranium-carbon specimens having compositions in the vicinity of uranium monocarbide are continuing for Atomics International. Early results of irradiations have been satisfactory. Two additional capsules of specimens, one having been irradiated to 10,000 MWD/T of uranium carbide and one to 4000 MWD/T of carbide, are to be examined during November. The preparation of enriched specimens for irradiation in the SRE by Atomics International is also reported.

Irradiation of Uranium Monocarbide

D. Stahl, J. H. Stang, and W. H. Goldthwaite

The MTR irradiation of Capsules BMI-23-4 and BMI-23-6 containing uranium monocarbide specimens is continuing in the A-27-SE and A-13-NE positions, respectively. Capsule BMI-23-4 will be irradiated for 3 more cycles to accumulate a total of 18 MTR cycles; BMI-23-6 will be irradiated for 2 more cycles to accumulate a total of 6 MTR cycles. At the end of the irradiation, burnups for Capsules BMI-23-4 and BMI-23-6 will be approximately 15,000 and 5000 MWD/T of carbide, respectively.

Temperatures during the last two cycles (Cycles 128 and 129) have been fairly steady. The central-core temperatures, as estimated from thermocouples close to the top specimens, were about 1000 F for BMI-23-4 and 1300 F for BMI-26-6.

Postirradiation Examination of Irradiated Uranium Monocarbide

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

In order to study the effects of irradiation on cast uranium monocarbide, a series of six capsules, each containing two specimens, has been or is now being irradiated at the MTR.

To date, irradiation experiments have been completed on three capsules containing specimens of uranium-5.0 w/o carbon. Two additional capsules have been discharged recently from the MTR and have been shipped to the BMI Hot-Cell Facility. These capsules contain specimens of uranium-5.0 w/o carbon and uranium-4.6 w/o carbon. Capsule BMI-23-3, containing the uranium-5.0 w/o carbon specimens, was irradiated to an estimated burnup of 10,000 MWD/T of carbide, and Capsule BMI-23-5 was irradiated to an estimated burnup of 4000 MWD/T of carbide.

K-2

Postirradiation examination of specimens from each capsule will be initiated in November.

Preparation of UC Pins for Irradiation in the SRE

C. K. Franklin, W. J. Hildebrand, E. L. Foster,
and R. F. Dickerson

Ceramic-type fuels are being developed for use as high-temperature fuels in power reactors. The high thermal conductivity, high uranium density, and castability of uranium monocarbide make it attractive for this application. Approximately 90 in. of cast specimens 0.610 in. in diameter and having surfaces representing both the as-cast and machined-ground conditions are being prepared for irradiation in the SRE.

The specimens are being prepared from right-cylinder castings which are poured into graphite molds using drop-casting techniques. From these castings two types of specimens are being produced. Specimens of one type are being cast to a diameter of 0.610 ± 0.002 in. and are to be irradiated in the as-cast condition. Specimens of the second type are being cast to a nominal diameter of 0.650 in. and subsequently are ground to a final diameter of 0.610 ± 0.001 in. At the present moment, 45 in. of surface-ground and 36 in. of as-cast specimens have been completed.

Various physical-property determinations are being conducted on the UC specimens prior to shipment to the SRE. The tests include electrical-resistivity and density measurements. The densities of the specimens were found to be 98 to 99 per cent of the theoretical value (13.65 g per cm^3) based on calculations from the weight difference in air and in carbon tetrachloride. The electrical resistivities of the specimens were found to vary from 30 to 50 microhm-cm.

The balance of the work will be confined to the preparation of the remaining specimens and a compilation of the preirradiation properties of the materials.

L. TANTALUM AND TANTALUM-ALLOY STUDIES

J. H. Stang

This program was recessed during October.

N-1

N. DEVELOPMENTS FOR THE MGCR

W. C. Riley

Research on core materials in support of the MGCR program is in progress at Battelle. The major effort is on the development and evaluation of UO_2 dispersions in BeO and dispersions of UC and UC_2 in graphite, and on the cladding of UO_2 particles with BeO. 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 is in progress.

FABRICATION AND CHARACTERIZATION OF FUEL MATERIALS

A. B. Tripler, Jr.

Attempts to increase the baked density of carbon and graphite fuel-matrix compacts continued. No increase in density was observed in fuel-free carbon compacts baked in an electric furnace with an argon atmosphere where the rate of increase of temperature was lowered to match the rate of a gas-fired furnace. The gas-fired material still had the higher density. The difference in density is now thought to be associated with differences in furnace atmospheres.

Additions of Thermax carbon black resulted in higher baked densities for compacts containing coke filler, and in slightly lower densities for compacts containing skeletal graphite as filler material.

Additional static neutron-activation testing on BeO-UO_2 type pellets was performed. The specimens tested contained fuel introduced as $Be(OH)_3$-UO_2 or UO_3. These fuel forms were used in an attempt to match fuel and matrix shrinkages. Results indicate that fission-product retention was somewhat better where the fuel was introduced as $Be(OH)_3$-UO_2 than where it was originally present as UO_3. Also, higher temperature heat treatment during fabrication resulted in improved-fission product retention.

UO_2 Dispersions in BeO

A. K. Smalley and W. H. Duckworth

Work in this area has been recessed.

N-2

UC and UC_2 Dispersions in Graphite

W. A. Hedden, W. C. Riley, and W. H. Duckworth

During October the work on graphite fuel cores was concerned with (1) the effect on density of the rate of increase of temperature during baking, and (2) the effect of carbon-black additions on the density of compacts having coke filler or skeletal graphite filler. Maximum density of the finished compacts is desired to achieve minimum gas permeability.

In previous work it has been found that compacts baked in a gas-fired furnace at a heating rate of about 5 to 10 F per hr to 1200 F had higher bulk densities than were obtained on identical compacts heated in an electric furnace with an argon atmosphere at a slightly higher heating rate.

In the current work, effort was made to duplicate the densities of the gas-baked compacts by baking in an electrically heated furnace in an argon atmosphere, using the same heating rate as that used for the gas-heated furnace. The bulk densities of the compacts baked in the electric furnace again were somewhat lower than those of compacts baked in the gas-heated furnace. The reason for this result is not fully apparent; however, it appears that the furnace atmosphere during baking may be a controlling factor.

In other work, bulk-density data were obtained on pitch-bonded coke compacts containing 12 to 18 w/o of carbon black, and on skeletal graphite compacts containing 10 to 16 w/o of carbon black. The compacts were baked in argon in an electrically heated furnace. Maximum bulk density (1.63 g per cm^3) was obtained with the compacts composed of 18 w/o Thermax carbon black and 82 w/o Texas 90 coke flour. On the basis of this result, additional mixes containing larger amounts of carbon black would be needed to optimize the composition.

Addition of carbon black to the minus 325-mesh skeletal graphite resulted in a slight decrease in bulk density as compared to compacts containing no carbon black.

No further fabrication work on graphite fuel-element cores is planned.

Cladding of UO_2 Particles With BeO

A. K. Smalley and W. H. Duckworth

Work in this area has been recessed.

N-3

Carburization Studies in the BeO-Graphite System

J. Koretzky, A. J. Roese, S. A. Rubin,
W. C. Riley, and W. H. Duckworth

Work in this area has been recessed.

Preliminary Characterization by Neutron Activation

P. Gluck, R. H. Barnes, and D. N. Sunderman

The investigation of the release of fission products from potential fuel materials for the MGCR was continued in October. This report covers the final series of results on this phase of the program.

Measurements were made of fission-product release from nine 48.5-w/o BeO-UO_2 specimens. Fabrication details are presented in Table N-1, and fission-product-release results in Table N-2.

Fission-product retention was somewhat better in samples in which the fuel was added as $Be(OH)_3$-UO_2 than in those in which it was originally present as UO_3. For both types of samples, higher temperature heat treatment during fabrication resulted in improved fission-product retention.

Only high-purity-grade charcoal was used for the fission-product adsorption. As this work was terminated, no further tests were conducted to ascertain the difference between the adsorptive capacities of the high-purity charcoal and low-purity charcoal which was used in the earlier studies.

STUDIES OF FISSION-GAS RELEASE FROM FUEL MATERIALS

R. H. Barnes

Work on detailed neutron-activation studies and in-pile studies has been recessed.

TABLE N-1. FABRICATION DETAILS FOR BeO-UO_2 FUEL-ELEMENT CORES PREPARED FOR NEUTRON-ACTIVATION TESTING

Starting Fuel Material[a]	Pellet	Sintering Conditions[b]: Maximum Temperature, F	Sintering Conditions[b]: Time at Maximum Temperature, hr	Sintered Bulk Density[c]: G per Cm^3	Sintered Bulk Density[c]: Per Cent of Theoretical
UO_2-10 w/o $Be(OH)_2$	77-C-1	3000	1	4.40	94.8
	77-C-2	3000	1	4.38	94.4
	77-C-3	3000	1	4.37	94.2
	77-D-2	2600	1	4.21	90.7
	77-D-3	2600	1	4.19	90.3
UO_3	79-A-2	2800	1	4.17	89.8
	79-A-3	2800	1	4.15	89.4
	79-B-1	3000	4	4.15	89.4
	79-B-2	3000	4	4.25	91.6

(a) All specimens contained a nominal fuel loading of 20 volume per cent UO_2 in the sintered condition.
(b) In flowing hydrogen.
(c) These data reported previously.

TABLE N-2. FISSION-GAS RELEASE FROM 48.5 w/o BeO-UO_2 DURING POSTIRRADIATION HEAT TREATMENT IN VACUUM AT 1800 F FOR 24 HR

Pellet	Decay Time[b], days	Specimen Weight, g	Thermal-Neutron Flux, 10^{11} nv	Total Uranium Burnup, 10^{-7} a/o	Fission-Gas Release[c], per cent: Xenon-133	Fission-Gas Release[c], per cent: Iodine-131
77-C-1	18.07	0.7548	4.83	7.12	1.15	1.98
77-C-2	18.00	0.7323	4.77	7.04	1.11	2.07
77-C-3	18.04	0.7288	4.91	7.25	1.06	1.87
77-D-2	18.12	0.7469	4.89	7.21	4.38	7.98
77-D-3	18.16	0.7431	4.89	7.21	4.10	8.22
79-A-2	17.85	0.7872	3.50	6.44	5.28	8.52
79-A-3	17.89	0.7839	3.57	6.58	4.95	8.55
79-B-1	17.93	0.7902	3.61	6.65	2.31	4.02
79-B-2	17.96	0.8067	3.64	6.70	2.88	5.18

(a) Pellet designation is 15171.
(b) Radioactive decay time between irradiation and radioassay.
(c) Fission-gas release is the ratio of atoms released divided by atoms present at the beginning of heat treatment multiplied by 100.

N-5

HIGH-BURNUP IRRADIATION EFFECTS IN FUEL MATERIALS

W. E. Murr, N. E. Miller, J. E. Gates, and R. F. Dickerson

A study of the radiation stability of ceramic-type fuels at a specimen-surface temperature of about 1500 F is in progress. The study includes the irradiation, examination, and evaluation of four types of fuel materials: one consisting of about 20 volume per cent uranium dioxide in beryllium oxide (UO_2-BeO), another containing about 20 volume per cent uranium monocarbide in graphite (UC-graphite), a third containing about 20 volume per cent uranium dicarbide in graphite (UC_2-graphite), and a fourth containing about 20 volume per cent UO_2 in Al_2O_3 (UO_2-Al_2O_3).

Three instrumented capsules, each containing two specimens of each of the first three fuel types mentioned above, have been in operation at the MTR since the beginning of MTR Cycle 125 (July 24). The specimens are composed of four cylindrical fuel pellets about 0.222 in. in diameter by 0.250 in. long sealed in Type 316 stainless steel tubing under a helium atmosphere. Table N-3 indicates the temperature and heater power consumption for the MTR capsules during Cycle 129.

TABLE N-3. OPERATION OF MTR CAPSULES DURING CYCLE 129

Capsule	Thermocouple Temperatures(a), F	Heater Power(b) Consumption, w
BMI-31-1	1240-1490	670
BMI-31-2	1380-1495	2200
BMI-31-3	1390-1495	2880

(a) Specimen-surface temperatures are calculated to be 25 to 30 F higher.
(b) Each capsule contains three heaters, rated at 1 kw each.

A comparison of the power requirements of the heaters during Cycles 127 and 129 indicates that more external heater power was required to maintain the capsules near the 1500 F design temperature during Cycle 129. Currently, all three heaters and four of the six thermocouples in each capsule are operating satisfactorily. The heaters in Capsule BMI-31-3 appear to be approaching their maximum capacity.

A calculation of the burnup incurred in the specimens based on thermocouple temperatures and external power inputs is being made, and will serve as a basis for determining the in-pile exposure times for the specimens.

A capsule containing Al_2O_3-UO_2 fuel material is being irradiated in the BRR. The capsule was designed and built by a cooperating laboratory. Battelle has assumed the responsibility of final capsule assembly, hazard evaluation, and conduction of the irradiation experiment.

During the first cycle (2 weeks) the peak fuel cladding temperature was approximately 1500 F. The peak temperature was increased to 1600 F at the beginning of the second cycle. The capsule will be irradiated for four cycles.

N-6

DIFFUSION OF FISSION PRODUCTS IN CLADDING MATERIALS

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

A program is in progress in which the diffusion of fission products in "A" Nickel cladding is being studied. The fission products under consideration were introduced into "A" Nickel foils by irradiation recoil from enriched uranium mechanically bonded to the foils.

Chemical analysis for cesium in layers etched from the "A" Nickel foils is still in progress. No results are available at this time.

CARBON-TRANSPORT CORROSION STUDIES

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

Selected metal and graphite specimens have been exposed to radiation in helium-filled quartz tori designed to promote convective flow of the helium and gaseous impurities past the specimens. The initial results were reported in BMI-1366. Additional metallography and microhardness tests are under way.

O-1 and O-2

O. ENGINEERING ASSISTANCE TO KAISER ENGINEERS

Reactor-Flow Studies

G. R. Whitacre and H. R. Hazard

Studies of flow in a quarter-scale air model of the Partially Enriched Gas-Cooled Power Reactor have been conducted at Battelle to provide design data for the prototype. During October experimental studies were completed.

Work previously reported includes model construction and completion of a program to determine the effect of hole location in the core-support cylinder on core-flow distribution, mixing, and pressure drop. Then, with the final core-support cylinder in place, additional studies were made on the effect of Reynolds number on transition from two-loop to one-loop operation, and on flow patterns in the upper plenum.

In October, complete thermal-shield flow studies were run and velocities in the lower and inlet plenums were measured. The draft of a topical report was also prepared.

This concludes the research under this program.

P-1

P. DEVELOPMENTAL STUDIES FOR THE SM-2

S. J. Paprocki

This work is being conducted in assistance to Alco Products and is concerned with the development of fuel, absorber, and suppressor materials for the SM-2.

Studies are in progress to develop process specifications for the fabrication of full-size reference fuel plates. The reference fuel plate is a stainless-UO_2 dispersion consisting of Mallinckrodt spherical UO_2 fuel and ZrB_2 burnable poison dispersed in Type 347 stainless steel. The fuel cores are sintered in vacuo to prevent oxidation of the ZrB_2 and reaction with the stainless steel matrix. A high-purity fused-ZrB_2 product is being utilized to also prevent reaction of impurities with the steel. A small loss of boron is still being experienced during the rolling operation; however, the results indicate that this loss can be controlled to a ±5 per cent specification.

Three uninstrumented capsules containing eight specimens each are being irradiated in the MTR, and one instrumented capsule containing six specimens is being irradiated in the ETR in a flux with a profile variation of 3 to 5 x 10^{14} nv. The heat generation is calculated to vary from 1,000,000 to 1,750,000 Btu/(ft^2)(hr). The irradiations are being conducted under this high neutron flux in order to obtain a reference peak burnup (2-$in.^2$ area) of 77 per cent of the contained uranium-225 in a relatively short period of time.

Materials Development

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

Fabrication techniques are being evaluated for use in preparing fuel elements, suppressor components, and control rods for the SM-2 reactor. Reference materials have been selected, and specifications will be established on the basis of the fabrication studies.

Fuel Materials

Major emphasis is being placed upon the production of full fuel plates for use in critical-assembly tests and welding studies. Twenty plates containing cores of 26 w/o fully enriched UO_2 and 1.2 w/o ZrB_2 dispersed in Type 347 prealloyed stainless steel (0.030 in. thick) and clad with Type 347 stainless steel (0.005 in. thick) are being prepared for the critical-assembly tests. Forty plates which are similar but which contain depleted UO_2 are being prepared for the welding studies.

As a check on the estimated boron content, small-scale specimens are being sintered and rolled simultaneously with the full-size cores and will be analyzed after

roll cladding. Fabrication data are being correlated with respect to tolerances maintained throughout the process and will be available for use in setting specifications. To date, all plates produced are well within presently specified tolerances. The results indicate that with present specifications, the reject rate will be low.

In addition to the preparation of full-size fuel elements, small-scale specimens are used to study the effect of fabrication variables on core structure, mechanical properties, and boron loss. By the use of a high-purity ZrB_2 and by sintering in vacuo, boron losses during sintering can be maintained well within the range of accuracy for boron analyses. For example, a specimen containing 0.179 w/o boron before sintering contained 0.176 w/o boron after sintering 2 hr at 2150 F in vacuo. However, on specimens roll clad by standard procedures, losses of up to 12 w/o boron have been reported. During the roll-cladding operation, the ZrB_2 particles are placed in intimate contact with the matrix and are thus more likely to react with oxygen and other impurities in the matrix. Work with a 0.461 w/o boron alloy has shown that in 0.010-in. foil, boron losses are rapid when the foil is annealed in hydrogen at 2000 F or higher. A loss of 50 w/o boron was reported after a 2-hr anneal at 2200 F. In the final stages of roll cladding, the cladding is less than 0.010 in. thick, and thus any boron in solution could diffuse through the cladding.

In general, boron losses reported for roll-clad plates are on the order of 5 w/o. Examination of microstructures shows a thin layer of oxide on the surface of the massive ZrB_2 particles which is not visible on sintered compacts and also a small amount of an unknown precipitate around many of the particles. In the reference specimens examined to date, there has been no indication of boron-containing phases in the cladding.

In order to further study losses which occur during rolling, a series of specimens is being prepared which will be used to determine the effect of time and temperature of the various operations on the boron loss. In addition, an attempt is being made to coat ZrB_2 particles with chromium, niobium, and/or tungsten by a fluidized-bed process. These particles will be used to determine whether a protective coating will be beneficial in preventing secondary reactions with impurities in the stainless matrix.

Fabrication variables are also being evaluated by means of transverse tensile tests. Results of recent tests are shown in Table P-1. All plates containing spherical UO_2 possessed higher strength than similar plates containing high-fired UO_2. The results on the high-fired UO_2-containing plates have been previously reported, but increases of as much as 13,000 psi were obtained by using spherical UO_2. Although not shown in the table, another interesting observation was made which showed that a full-sized fuel element containing high-fired UO_2 had a room-temperature transverse tensile strength of 25,100 psi, while a similar small-scale specimen had a transverse tensile strength of 17,000 psi. Additional specimens will be run to determine whether such behavior is consistent.

P-3

TABLE P-1. RESULTS OF TENSILE TESTS IN THE THICKNESS DIRECTION OF FUEL PLATES CONTAINING SPHERICAL UO_2

UO_2, w/o	ZrB_2, w/o	Reduction[a] on First Pass, per cent	Core Thickness, in.	Core Width, in.	Total Reduction Ratio	Test Temperature, F	Ultimate Tensile Strength, psi
26	1.45	12	0.090	1	1.1:1	Room	22,800
26	1.45	12	0.090	1	1.1:1	600	20,300[b]
26	1.45	19	0.092	1	1.2:1	Room	23,700
26	1.45	36	0.097	1	1.6:1	Room	29,500[b]
26	1.45	44	0.100	1	1.8:1	Room	29,700[b]

(a) Plates reduced one pass only.
(b) Average of two tests.

Fabrication of Irradiation Specimens

The specimens for the remaining capsules in the SM-2 irradiation program have been fabricated, drilled, leak tested, and radiographed. A microscopic examination and an anlysis of the boron content is presently being made on representative samples of each specimen type.

Development of Control and Suppressor Materials

A dispersion of Eu_2O_3 in a matrix of stainless steel has been chosen as the reference control material and as a possible flux suppressor in the SM-2 reactor. Corrosion tests made at Alco on Eu_2O_3-stainless steel dispersions fabricated at Battelle produced a swelling of 30 per cent throughout the core. There were no cracks or bond separations visible by metallographic examination.

A comparison study will be made of stainless steel dispersions containing Eu_2O_3 powder produced by different methods. One method was developed at KAPL and was described in BMI-1381. Another process, which originated at ORNL, employs no binder and the particle size is reduced to 44 μ. Specimens containing each type of Eu_2O_3 will be fabricated and evaluated.

A 100-g sample of minus 100-mesh monoclinic Eu_2O_3 has been received from a commercial source. This material is presently being examined by spectrography, petrography, and X-ray diffraction to determine the quality of the powder. This Eu_2O_3 will also be evaluated as a dispersion with stainless steel and compared with the dispersions containing the KAPL- and ORNL-process Eu_2O_3.

P-4

Encapsulation Studies

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

The irradiation behavior of clad specimens of UO_2 dispersed in a Type 347 stainless steel matrix, and containing a boron poison material, is of interest to the SM-2 program. A ten-capsule program designed to test parameters of composition, fabrication technique, and burnup is in progress. Three of the ten capsules contain no instrumentation and are being irradiated in the MTR. Seven of the capsules are highly instrumented and will be irradiated in the ETR. The three noninstrumented capsules, BMI-32-1, BMI-32-2, and BMI-32-3, have been irradiated for three cycles, two cycles, and one cycle, respectively, in unperturbed fluxes of 3 to 5 x 10^{14} nv, as of the end of Cycle 128 (October 6). During Cycle 129, all three capsules were removed from the reactor when the activity-monitoring system indicated that a fission-product break had occurred somewhere in the system, resulting in a high nickel concentration of the coolant water. As the three MTR capsules are constructed with nickel water-contacting shells, they were suspected of contributing to the high nickel activity. However, as Cycle 129 progressed, the activity continued abnormally high and the suspicion that the BMI-32 series capsules were responsible for the high nickel activity was reduced. In the meantime, the three capsules were examined both in the MTR pool and in the NRTS Hot Cell. The only unusual finding reported by the end of October was that BMI-32-3 has a small dent below the weld joining the shell to the bottom header. This type of dent could be produced by dropping the capsule obliquely. As things now stand, BMI-32-1 and BMI-32-2 will be reinserted during Cycle 130 shutdown. However, reactor personnel feel that additional inspections of BMI-32-3 are warranted, and, hence, permission to reinsert this capsule may be withheld.

During October, the temperature levels in ETR capsule BMI-32-4 indicated that peak-flux specimens-surface temperatures in the neighborhood of 600 F were being maintained. At the end of October, a total of 16.5 days of irradiation had been accumulated. It is estimated that the fission burnup is approximately 12 per cent, based on a quoted peak thermal flux of 3.5 x 10^{14} nv.

During mid-October, the six auxiliary sheathed electrical heaters incorporated into capsule BMI-32-4 failed almost simultaneously and without warning. Resistance checks show, in essence, that one leg of each heater is shorted to the capsule body. It is not possible to pinpoint the exact reason for this failure but it is conjectured that the resistance element-to-sheath potting material (a commercially available inorganic cement) used just above the capsule body has broken down, presumably by the intense irradiation it has experienced. Without the auxiliary heat available to compensate for the reduction of fissionable material as the irradiation progresses, a steady decrease in specimen temperature is anticipated.

During November, brief laboratory tests will be conducted to investigate the possibility of spurious thermal breakdown of the suspected potting material. Various other elements in the sheathed heater-to-leadout wire connection system are also to be re-examined in an attempt to bolster potentially weak links. The results of these activities will influence to some degree a decision about incorporating heaters into the six additional capsules scheduled in the program.

Q-1

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

BeO-UO_2 pellets containing 25 volume per cent UO_2 were prepared for capsule-irradiation experiments. The procedures employed for fabricating the pellets with different UO_2 particle sizes are described.

Encapsulation studies were continued of four fuel types in support of the over-all gas-cooled reactor program. Included are solid UO_2, BeO-UO_2, UO_2-graphite, and stainless steel-UO_2.

The hot-cell evaluation of a second series of solid UO_2 and annularly loaded UO_2 specimens contained in Capsule BMI-27-1 was initiated. Also, the second in-pile-loop assembly containing 19 solid UO_2 fuel pins was transferred from the BRR loop to the hot cells for evaluation.

Critical experiments were continued during the past month to aid in an evaluation of the 19-pin UO_2 fuel element for the GCRE-1B and ML-1 reactor. Reactivity worth, intracell thermal-flux distributions, and power distributions were measured for various ratios of enrichments between the 12 outer pins and 7 inner pins of the assembly.

Fabrication of BeO-UO_2 Fuel Pellets

A. K. Smalley and W. H. Duckworth

Pellets consisting of 25 volume per cent UO_2 dispersed in a densely sintered BeO ceramic were prepared for a proposed capsule-irradiation experiment. Two sizes of UO_2 grains were used, about 200 μ or less than 10 μ. Sintered pellets containing the large fuel grains had a bulk density of about 83 per cent of theoretical, and those containing the small grains had a density about 99 per cent of the theoretical value.

Pellets containing 200-μ UO_2 were made by the following procedure:

(1) MCW ceramic-grade fully enriched UO_2 was mixed with water, 10 w/o of $Be(OH)_2$, and 4.5 w/o of polyvinyl alcohol on a Stewart-Bolling roll mixer to leather-hard consistency. The batch was dried at about 120 F, and then crushed and ground to minus 60 plus 70-mesh particles. The particles were rinsed with ethanol to remove UO_2 dust from their surfaces.

(2) A puttylike mixture of LOH-grade BeO, 3 w/o of beeswax, and CCl_4 was prepared. The UO_2 particles were added to the BeO-beeswax paste, and the mass was mixed for about 1 min on the Stewart-Bolling mixer, using a nip clearance several times the thickness of the UO_2 particles. The mixture was stripped from the mixer rolls, granulated while damp through a 20-mesh sieve, and dried at about 120 F.

(3) The dried BeO-UO_2 batch was placed in rubber envelopes and pressed hydrostatically at 50,000 psi to form rods measuring about 0.3 in. in diameter and about 4 in. long.

(4) The compacted rods were sintered in flowing hydrogen at 2800 F for 1 hr. The rods were heated to maximum temperature in about 6 hr and cooled with the furnace.

(5) The sintered rods were ground and cut with diamond wheels to form pellets measuring 0.160 in. in diameter and 0.25 in. long.

Processing steps used to prepare pellets containing UO_2 in particle sizes less than 10 μ were the following:

(1) MCW UO_2, as above, was ball milled in ethanol for 19 hr, using a rubber-lined mill and uranium grinding balls. The mill was opened, LOH-grade BeO was introduced, and milling was continued for 1 hr. The mill was discharged, and the BeO-UO_2-ethanol paste was dried at about 120 F. The dried batch was then granulated through a 20-mesh sieve.

(2) The batch was compacted without binder addition, as described above. The compacted rods were sintered either at 2800 F for 1 hr or at 3200 F for 4 hr in flowing hydrogen. The sintered rods were ground and cut to form pellets having the same dimensions as those described previously.

Metallographic examination of a pellet containing 200-μ UO_2 indicated that substantial porosity existed in and around the UO_2 inclusions. The BeO matrix appeared to be moderately dense, and it consisted of crystals having an average diameter of about 12 μ. Pellets containing the fine-grained UO_2 appeared to be nonporous. The UO_2 phase was very nearly continuous, consisting of lakes and fingers interspersed throughout the

BeO crystals. The BeO crystal size averaged about 7 and 10 μ for pellets sintered at 2800 and 3200 F, respectively.

Encapsulation Studies

D. W. Nicholson, P. B. Shumaker, J. F. Lagedrost, and J. H. Stang

Irradiation of Clad Pin-Type Specimens Containing Dense UO_2

Attempts during October to develop further definitive information relative to the thermal performance of Capsules BMI-27-1 and BMI-27-2 were not successful. As reported previously, several specimens in these capsules were damaged during the exposure, possibly as a result of local overheating of the specimen cladding. Further evaluation will be made.

Irradiation of Specimens Containing UO_2 in Graphite

Capsule BMI-29-1, in which six graphite-8 w/o (highly enriched) UO_2 clad specimens were irradiated in the MTR, was returned to Battelle during October for hot-cell examination. Two each of the specimens were clad with Inconel 702, Hastelloy X, and Carpenter 20 Cb. During the two-cycle irradiation, cladding-surface temperatures were fairly constant (about 1775 F for the peak-temperature specimen and about 1600 F for the lowest temperature specimen). Approximately 1-1/2 kw of electrical heat was required continuously to maintain these temperatures. Based on recorded temperatures and estimated flux levels, fission burnups of 8.0 and 5.7 a/o are anticipated for the highest and lowest temperature specimens, respectively.

Irradiation of Specimens Containing MCW Spherical UO_2 Dispersed in Stainless Steel

Capsules BMI-33-1 and BMI-33-2, which contain stainless steel-clad specimens fueled with MCW spherical UO_2 and with ORNL hydrothermal UO_2 (dispersed in stainless steel), have not yet been inserted in the MTR because of the existing backlog of experiments. It now appears that the irradiation of these capusles will not begin before mid-December.

Irradiation of Specimens Containing UO_2 Dispersed in BeO

During October, this program was concerned with (1) the design and fabrication of an instrumented double-wall helium-annulus capsule for the 1725 F irradiation of six Hastelloy X-clad pins containing UO_2 in BeO and (2) the exposure at the Battelle Research Reactor of a nuclear mock-up of this capusle to obtain flux-perturbation data. The target date for this irradiation, initially November 1, has been moved ahead to about December 15 to permit a change in the technique employed to fabricate the UO_2-BeO pellets.

The nuclear-mock-up experiment was conducted in Position 12 in the BRR core with the reactor operating at approximately 500 w (normal operation is 2 megawatts). The fueled pellets (0.160 in. in diameter in a 1-in. stack) were simulated by cobalt slugs. These slugs were housed in stainless steel shells closely matching the geometry of those anticipated for the irradiation capsule. After a 5-min exposure, the relative fluxes of dosimeter (manganese-copper) wires taped to the slug surfaces were determined using a crystal-scintillation counting technique. According to preliminary analyses of the dosimeter data, the peak specimen-surface flux was 1.9×10^{13} nv, indicating a surface-to-unperturbed attenuation of about 0.41. The peak unperturbed level was obtained by gold-foil exposures in the empty irradiation space. The specimen-effective flux was obtained from these data by P-3 analysis; a specimen effective flux-unperturbed flux ratio of approximately 0.30 was indicated.

It should be mentioned that the actual irradiation specimens will consist of a stack of three 1/4-in.-long pellets fueled with 25 volume per cent UO_2 and, at each end of the stack, a single 1/8-in.-long pellet fueled with approximately 17 volume per cent. The end wafers will be present to decrease the opportunity for higher-than-average heat-release rates at the end zones where self-shielding effects are minimum. In the nuclear mock-up, the cobalt slugs were tailored to this situation by drilling 3/32-in. blind holes into the ends. In a subsequent analysis of the mock-up dosimetry, the wires taped to the cobalt surfaces will be cut into short pieces that will be individually analyzed. This will be done in an attempt to define any flux-profile characteristics along the specimens that are superimposed on the main profile along the capsule axis.

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

Owing to a change in emphasis in the over-all program, the activity in this capsule-irradiation program was curtailed early in October. While the actual irradiation experiment will not be conducted until further notice, a few facets of the capsule-design phase are still being pursued as a minor effort. This includes completing the design and the hazards analysis. This work, which will be completed during the next few weeks, will facilitate fabrication of the system if the program is resumed.

Effects of Irradiation

J. H. Saling, J. E. Gates, and R. F. Dickerson

A study of the radiation stability of fuel-element materials for compact gas-cooled reactors includes (1) the evaluation of in-pile-loop subassemblies containing PWR-type fuel pins of solid UO_2 clad with Inconel, and (2) the evaluation of encapsulated solid and annularly loaded UO_2 specimens clad with Inconel.

Capsule Program

The irradiation of Capsule BMI-27-1, containing four specimens composed of solid UO_2 clad with Inconel and two composed of UO_2 annularly loaded with MgO core and clad

with Inconel, has been completed. The specimens were loaded with the fuel materials under an atmosphere of helium. This capsule was a duplicate of Capsule BMI-27-2, the design of which was described in BMI-1381. The specimens in Capsule BMI-27-1 were irradiated in contact with NaK to burnups of 5 to 6 a/o of the uranium-235 as estimated from MTR operating data.

Specimen temperatures were measured with thermocouples during the irradiation. A preliminary examination of the temperature charts indicates that temperatures near the surfaces of the specimens ranged from approximately 800 to 1600 F, except for the first few (less than 10) hours of the irradiation. During this period, temperatures of 1800 to 1850 F were recorded. The occurrence of temperatures near 1800 F for a short time at such an early point in the irradiation should not cause difficulties if normal heat-transfer conditions were operative. However, upon opening the capsule after irradiation, it was discovered that all six of the specimens had failed.

Capsule BMI-27-1 consisted of a sealed inner chamber containing the specimens in NaK and a sealed outer chamber containing helium. A small amount of NaK was found in the outer chamber during the opening operation. The presence of this NaK in the outer chamber was assumed to be an indication that NaK from the inner capsule had leaked out during the irradiation. The inner chamber was, therefore, not sampled for fission-gas content but was opened for recovery of the specimens. The top specimen and the third and fourth specimens from the top, all originally containing solid UO_2 pellets, appeared to have failed because of melting of the Inconel cladding. The second and fifth specimens from the top, originally containing annularly loaded UO_2, also appeared to have failed in this manner. The bottom specimen, containing solid UO_2 pellets, was ruptured and showed no apparent evidence of melting. In any normal sequence of events, it would be expected that rupture would precede melting because of the helium present inside the specimen cladding.

The temperature history of the specimens gives no indication which would suggest that temperatures near the melting point of Inconel, about 2540 F, were reached, assuming that the thermocouples were recording specimen temperatures. Also, the temperatures obtained from the thermocouples indicated that the bottom specimens operated at higher temperatures than the top specimens in the capsule, yet the cladding on the top specimens melted and cladding on the bottom specimens did not. All of the data presently available concerning capsule design, specimen-fabrication histories, and irradiation-temperature histories, are being reviewed and evaluated in an attempt to logically explain the unexpected behavior of these specimens.

Loop Program

The postirradiation evaluation of the 1B-1βT in-pile-loop subassembly is scheduled to be initiated in November, 1959. This element is similar to the 1B-1αT except that each of the 19 fuel pins is fluted in order to provide better heat transfer. This element was irradiated in the BRR for a period of approximately 35 hr. The irradiation was discontinued because of high fission-product activity in the coolant gas.

Q-6

GCRE Critical-Assembly Experiments

J. Ray, W. S. Hogan, D. A. Dingee, and J. W. Chastain

Experiments were performed during the past month to aid in an evaluation of 19-pin UO_2 fuel elements for the GCRE-1B and ML-1 reactors These experiments use mocked up fuel pins constructed from highly enriched and natural uranium foils inserted in 0.164-in.-ID by 0.030-in.-wall Inconel X tubes. A number of enrichment ratios between the outer 12 pins and inner 7 pins of the assembly are being considered. The ratios investigated were 24/48, 30/93, and 35/70 where the numbers indicate the enrichment in per cent and the first figure applies to the outer ring of pins.

Reactivity worth, intracell thermal-flux distributions, and power distributions have been measured in these studies. The reactivity-worth measurements compared a pin-type element with a specially constructed GCRE-1B mock-up element which had the same active fuel length, 22.75 in. Intracell flux-distribution measurements were made on the outer surfaces and at the center of the pins and in the moderator cell which surrounds the element (19-pin cluster). The power distribution was obtained by counting representative samples of the fuel from the irradiated pins.

The experimental portion of this program was essentially completed during the past month. All of the measurements outlined above were made for the various pin mock-ups. A limited number of measurements are planned for the 35/70 enrichment-ratio case with the natural uranium foil removed, i.e., the uranium-235 content will be identical to the 35/70 enrichment case but the fuel will no longer be diluted with uranium-238.

These experiments will be completed in November, and data reduction and analysis will be started.

IN-PILE-LOOP PROGRAM

G. A. Francis

During October, activity was continued on the two in-pile gas loops being built in support of the gas-cooled reactor program. At the Battelle Research Reactor, the 1B-1βT specimen which was designed by Aerojet General Nucleonics was removed from the loop and preparations were made for future irradiations. Preparation for operation of the in-pile loop at the Engineering Test Reactor was continued. The major activity on this task was in the field of blower study, repair, and testing. The different phases of the loop program are described in the following paragraphs.

Q-7

BRR Loop Program

S. J. Basham and W. H. Goldthwaite

On October 19 the 1B-1βT specimen was removed from the loop. The transfer from the loop to the cask was complicated by mechanical difficulties. These included crimping of the steel tube containing the thermocouple lead wire, interference between the transfer-mechanism chuck and the element top fitting, and failure of the positioning mechanism for the transfer plug.

At one time during the operation, the test specimen was removed from the loop and replaced. The operation showed that it would be possible to remove a specimen for inspection and return the specimen for further irradiation. The specimen was transported to the Battelle Hot-Cell Facility for examination.

Prior to irradiation of the beta specimen, leaks were found in aluminum tubing associated with the loop underwater package. Examination of the tube revealed that the surface silicon content was approximately 2 w/o, while the underlying material contained 0.4 w/o. Holes in the tube were examined microscopically, and the attack was found to be uniform. The exact cause of the failure has not been determined. Present plans call for replacement of the tube in question by stainless steel.

ETR Loop Program

E. O. Fromm and J. V. Baum

The primary problem being encountered during the hot check of the ETR loop is related to blower operation. The three blower units installed at the ETR were repaired during September when new impeller-shaft bearings were installed. Subsequent to the bearing replacement, one unit was run for 274 hr.

During the period covered by this report, the second unit was put into operation. Early in the period, the impeller came in contact with the housing, and the high-speed shaft seized. When operation was shifted back to the first blower, the drive belt failed. This left only one installed blower satisfactory for operation, so auxiliary coolant (plant air) is now being used to cool the in-pile leg. At present four blowers are at Battelle for modification and test. The first unit has been modified and is now being tested in Battelle's laboratory. The modifications which are being made to each unit include the installation of 3/8-in.-diameter coolant lines in the motor housing in order to reduce temperatures in the area adjacent to the bearings. A second modification involves the installation of a bypass line between the blower drive chamber and the impeller housing in order to reduce or eliminate contamination of bearings by dirt. The clearance between the impeller and the housing was also increased. Initially this clearance was between 0.009 and 0.014 in. on both faces. This has been increased to approximately 0.024 in. on both faces. The initial contact between the impeller and housing could have been caused by either bearing failure or the close tolerances which had been established.

Q-8

Another cause of bearing failure may have been excessive axial thrust load. In order to reduce this load, holes were drilled through the impeller to reduce the pressure differential on the two faces. In the test now under way, the pressure differential has been measured at 2.5 psi. The blower now on test has been run for 235 hr at design flow of 0.9 lb per sec and with the blower inlet-gas temperature held at 200 F. There have been no bearing-temperature excursions or belt problems to date. The maximum bearing temperature has been 183 F.

During October approval of a control system based on flow and of test operation with a single blower in operation was obtained.

Plans call for modification of all four blowers and preliminary tests at Battelle before reinstallation in the loop at the ETR. During November the blower modification and test program should be completed, and the units should be shipped back to the ETR for installation and completion of the hot check.

RWD/CRT:all