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ANNUAL PROGRESS REPORT ON FUEL ELEMENT DEVELOPMENT

FOR FY 1963

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SUMMARY

This report summarizes progress in Fuels and Materials Development performed under Budgeted Activity 04400209 during the period July, 1962 through June, 1963. Program objectives for FY 63 have included a materials study involving (1) development of new fuel compositions, (2) testing of newly developed aluminum alloys at high temperatures, and (3) determining damage thresholds in beryllium. Also included have been prototype studies involving hydraulic, heat-transfer, and irradiation testing of improved fuel element geometries.

Irradiation tests on powdered UAl_3 intermetallic compounds demonstrated good stability and fission gas retention capabilities. Developmental aluminum powder metal products showed good corrosion resistance at high temperatures while retaining excellent high temperature strength. All of the fuel compositions tested (UO_2 , U_3O_8 and UAl_3 in aluminum matrices) exhibited density decreases under irradiation. Tensile tests on sandwich-type fuel plates at elevated temperatures indicate that the fuel plate strength is strongly influenced by the core material rather than dependent primarily on the cladding material as has been found true of lower (MTR) temperatures.

Work for FY 64 will include assessing the engineering feasibility of UAl_3 fuels through establishing a commercial supply for this material and by fabricating and irradiating in the MTR and ETR several "Production" elements, fuelled with UAl_3 . Other fuel materials will be investigated for elevated temperature potential. This work will continue the effort already in progress on fuel irradiations and cladding corrosion tests. Analyses of fuel matrix reactions induced by heat and/or irradiation will be continued using conventional electron microscopy and X-ray diffraction techniques as well as the more recent electron microprobe facility. Beryllium damage investigations will be resumed with modified capsule design. This work will include a determination of mechanical and physical property changes of beryllium under high temperature irradiation and will study the effect of helium gas formation on the stress-strain behavior of beryllium as a function of temperature.

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I. INTRODUCTION

This report summarizes the progress in Reactor Fuels and Materials Development at the MTR-ETR for FY 63. Previous work performed under this program has been reported in IDO-16574(1), 16727(2), and 16799(3). The objectives of the program have included the improvement of fuel elements for high flux and high power density reactors through the development of new fuel compositions, distributions and geometries. An associated project on In-Pile Fatigue Testing which is included under the same budgeted activity is reported separately.

Specific goals in Fuels and Materials Development for FY 63 have included (1) developing uranium-aluminum intermetallic dispersions and comparing the irradiation performance of these materials at elevated temperature with that of uranium oxide cermets, (2) testing newly developed aluminum powder metallurgy alloys as fuel claddings at high temperatures, (3) determining damage thresholds in beryllium, and (4) performing prototype hydraulic, heat transfer, and irradiation tests on improved fuel element geometries. With the exception of item (3), where some delays were encountered, all of the above goals have been accomplished. Because this type of development work is necessarily of a continuing nature, several of these areas of activity will be continued into FY 64 with the addition of new materials and higher operating limits.

During the year a facility for conducting irradiation experiments in high temperature high pressure water was made available for use in this program. Consequently, many of the compositions were fabricated for testing in this loop. The facility was initially used by the Argonne National Laboratory and is located in the HB-2 position of the MTR. It can be operated at temperatures up to approximately 240°C. Irradiation testing of fuel plate compositions has also been continued in the L-51 position of the MTR core. The effects of irradiation on microstructure and mechanical and physical properties of the fuels have been examined.

TABLE I
SAMPLE FUEL PLATE COMPOSITIONS

Bearing Material	Core Material		Cladding Material
	Uranium*	wt % Uranium Additions	
U in Al Alloy	34	1 w/o Si	APM-M-769 Al
U in Al Alloy	34	1 w/o Si	2219-T62 Al
UO ₂ in Al Cermet	20		X8001 Al with 20% Cold Work
UO ₂ in Al Cermet	30		APM-M-768 Al
UO ₂ in Al Cermet	30		APM-M-769 Al
UO ₂ in Al Cermet	30		APM-M-770 Al
UO ₂ in Al Cermet	36	0.19 w/o B ₄ C + X8001 Al	6061-T6 Al
U ₃ O ₈ in Al Cermet	19		X8001 Al 16% Cold Work
U ₃ O ₈ in Al Cermet	30		X8001 Al with 10% Cold Work
U ₃ O ₈ in Al Cermet	37	0.19 w/o B ₄ C + X8001 Al	6061-T6 Al
UAl ₃ in Al Cermet	22		1100-H-14 Al
UAl ₃ in Al Cermet	35	0.19 w/o B ₄ C	X8001 Al** XAP001 Al
UAl ₃ in Al Cermet	35	0.19 w/o B ₄ C	XAP001 Al
UAl ₃ in Al Cermet	35	0.15 w/o B ₄ C + X8001 Al	X8001 with 16% Cold Work
UAl ₃ in Al Cermet	35	0.15 w/o B ₄ C + 6061 Al	6061-T6
UAl ₃ in Al Cermet	36	0.19 w/o B ₄ C + X8001 Al	6061-T6

* All Uranium is fully enriched

** Duplex Cladding

II. MATERIALS

The fuel plate compositions made for the Fuel Element Development Program during FY 63 are shown in Table I. Considerable effort was devoted to fabrication and testing of high weight-percent alloy and dispersion fuel cores clad with several aluminum materials.

A. Fabrication (G. W. Gibson)

1. Core Materials

The procedures used for fabrication of fuel plates containing cores of uranium-aluminum alloys or cermet of uranium oxide dispersed in aluminum have been given in previous reports^(1,2,3). During the past year additional work has been performed on the development of fabrication techniques for plates containing high weight-percent U-Al alloy powders.

The use of powdered uranium-aluminum alloys offers an alternate to uranium oxide for use of high fuel densities while retaining a workability of the core material not achievable through the normal alloy route. Because of the possibility of a violent exothermic reaction between uranium oxide and aluminum⁽⁴⁾, dispersion fuels of this type are potentially dangerous in nuclear reactors. This hazard does not exist when uranium aluminum alloys are used. Moreover, a burnable poison, such as boron, can be dispersed as easily in fuel plate cores with uranium-aluminum powdered fuel materials as in those using uranium oxides.

A comparison of some of the properties of the uranium-aluminum intermetallic compounds with uranium and uranium oxide is shown in Table II.

TABLE II
PROPERTIES OF URANIUM COMPOUNDS

Compound	Density g/cm ³	Uranium Density g/cm ³	Melting Point °C	Thermal	Lattice Structure
				Conductivity 200°C watt/cm-°C	
U	18.9	18.9	1133	0.29	Orthorhombic
UAl ₂	8.1	6.6	1590	0.50*	FCC, Mg Cu ₂
UAl ₃	6.8	5.1	1350	0.63*	FCC, Cu ₃ Au
UAl ₄	5.7	3.9	730	0.72*	Orthorhombic
UO ₂	10.9	9.6	2750	0.08	FCC, Cu F ₂

* Estimated

From the data presented in Table II, it can be seen that:

- (1) The uranium density of the U-Al intermetallics is comparatively low with UAl_4 having the lowest density.
- (2) The melting points of UAl_2 and UAl_3 are higher than that of pure uranium, or of those of the aluminum alloys used for claddings. The oxides are even more refractory materials.
- (3) The estimated thermal conductivities of the intermetallics are about 8 times those of the oxide. These estimates of the thermal conductivities of the intermetallics were made by extrapolation of the data for uranium-aluminum alloys and pure uranium metal.
- (4) UAl_2 , UAl_3 , and UO_2 , all having face centered cubic structures, would tend to be the most irradiation resistant.

When these factors are considered, as well as the fact that there is no possibility of a thermit type reaction between the intermetallics and aluminum, UAl_2 , UAl_3 and UAl_4 appear to be attractive as the fissile materials in dispersion type fuel elements. Indeed, these materials have been used in the wrought form in existing reactors.

A flow sheet showing a method used for the preparation of uranium-aluminum intermetallic powders is given in Figure 1.

The principal problem encountered in the preparation of the intermetallic compound by vacuum induction melting and casting has been a metallurgical recovery of only 85%-90%; 10 to 15% of the material has been absorbed by the crucible. The production of the powders from the casting has presented two problems. First, the compounds are extremely friable so that they are easily over-ground. Second, powdered UAl_2 is very pyrophoric and must be handled in an inert atmosphere. UAl_3 and UAl_4 , while somewhat pyrophoric, can be treated by conventional powder metallurgical techniques.

Figure 1 - Flow Sheet of a Method Used for Production of U-Al Inter-metallic Powders.

The advantages in using UAl_2 instead of UAl_3 or UAl_4 did not seem to be great enough to justify the added expense required for safe handling of this material. Consequently, very little work has been done with this compound. Due to the lower uranium density and melting point, UAl_4 is of less interest than UAl_3 .

In order to determine the compatibility of the intermetallic compound with the matrix material, particularly during the hot rolling operation, several samples were subjected to a thermal treatment.

850X CHROMIC & ACETIC ETCH

AS CAST

HEAT TREATED

Figure 2 - Microstructure of a 68 w/o U-Al Alloy Before and After Heat Treating 16 Hours at 600°C

In Figure 2 can be seen the microstructure of a high weight-percent uranium-aluminum alloy in the "as cast" condition and after heat treating for 16 hours at 600°C.

By means of X-ray diffraction, it was determined that the rectangular shaped particles in the "as cast" sample are UAl_3 and the matrix material is aluminum. The dark rod-shaped particles have not been identified as yet. Heat-treating this material causes the UAl_3 to react with the aluminum to form UAl_4 as can be seen in the microstructure of the heat-treated specimen. The rods of unidentified compound are now coalesced into the dark areas.

The uranium-aluminum powders with small additions of B or B_4C were blended with X8001, 6061 aluminum alloy powders, or 99% pure aluminum powders and compacted at 30 tsi. The green core compacts were assembled with picture frames and cover plates of aluminum alloy materials similar to the powders blended into the fuel powders. The assemblies were then hot-rolled at 590°C into fuel plates. Both full size MTR-type fuel plates as well as sample fuel plates containing up to 35 w/o U with 0.19 w/o B_4C have been successfully manufactured using this technique.

The microstructure of a typical fuel plate core made by this procedure is seen in Figure 3. The UAl_3 and aluminum material used in

650X

Electrolytic Etch

Figure 3 - Microstructure of Fuel Plate Core Containing Powdered U-Al Alloy - as Fabricated.

this plate was ground to a powder from the "as cast" condition without heat-treating. In the photomicrograph, it can be seen that the UAl_3 has reacted to a considerable extent during fabrication to form UAl_4 . The reaction $UAl_3 + Al \rightarrow UAl_4$ can be suppressed by addition of small quantities of silicon, or tin. Since the presence of UAl_4 caused no problems in fabrication it is not believed to be undesirable. In fact, since UAl_4 is more stable than UAl_3 , a small amount of UAl_4 in the core may be an advantage.

The results of testing of fuel plates containing these core materials are given in other portions of this report.

2. Cladding Materials

The chemical composition of cladding materials fabricated into fuel plates in FY 63 is given in Table III.

TABLE III
NOMINAL CHEMICAL COMPOSITION OF CLADDING MATERIALS

Material	Element or Compound Present - Weight-Percent											
	Al	Al_2O_3	Cu	Si	Mg	Ni	Cr	Fe	Mo	Zr	Ti	V
2219	Bal.		6.3		0.3					0.18	0.06	0.10
6061	"		0.25	0.6	1.0		0.25					
X8001	"					1.0		0.5			0.1	
XAP001	"	6.0										
APM-M768	"	0.5					0.2	2.0	2.0	2.0	0.2	0.2
APM-M769	"	0.5					0.2	2.0	6.0	0.2	0.2	0.2
APM-M770	"	0.5					0.2	4.0	4.0	0.2	0.2	0.2

The first three materials listed in the table are alloys while the balance of the compositions are products of powder metallurgy.

a. Aluminum Alloys

Alloy 2219 is an aluminum-copper alloy which was originally developed for elevated temperature applications. In the solution heat-treated and artificially aged temper, typical mechanical properties of 2219 in the 260-315°C temperature range are superior to those of any other commercially available alloy. In addition, corrosion testing at Argonne National Laboratory indicated that the corrosion rates of alloy 2219 were almost the same as alloy X8001⁽⁵⁾.

Alloy 6061 is an intermediate strength, heat-treatable aluminum alloy which was developed for structural applications requiring a relatively high yield strength in the fully heat-treated temper. Alloy X8001 is a nickel-aluminum alloy for which there is considerable corrosion data and this material therefore forms a good standard of comparison for other developmental alloys.

b. Aluminum Powder Metallurgy Materials (APM's)

The XAPO01, APM-M768, APM-M769 and APM-M770 materials listed in Tables I and III are aluminum powder metallurgy materials of two basic types. The XAPO01, formerly called APM-M257, is made from a milled powder, the particles of which have a flake-like shape, and contains 6% aluminum oxide. The excellent elevated temperature strength is due to a fine dispersion of sub-microscopic aluminum oxide particles throughout the aluminum matrix. No alloying elements have been added, and this material has no better corrosion resistance to high temperature water than does commercially pure aluminum. This type was developed and patented by a Swiss Company, Aluminum-Industrie-Aktiengesellschaft (AIAG). The other type of APM product, developed by the Aluminum Company of America, is made from atomized alloy powder and contains only a small amount of oxide (approximately 0.5%). By judicious choice of alloying elements in the powders it is possible to produce a material with good high temperature strength and, it is believed, good corrosion resistance to high temperature water.

B. Irradiation Experience (G. W. Gibson and W. P. Scarrah)

1. In MTR Process Water

a. High Weight-Percent Uranium-Aluminum Alloy Powder

By July 1, 1963 thirty-one sample fuel plates with cores containing high weight-percent uranium-aluminum alloy powders dispersed in aluminum had been irradiated or were being irradiated in the L-51 position of the MTR core. The sample size and irradiation procedures used have been previously described (1, 2).

Twenty plates with cores containing uranium-aluminum alloy powders were irradiated in a similar manner in the lattice of the ETR for comparison with U₃O₈-aluminum (ATR) fuel cores.

These plates had fuel contents ranging from 22 w/o to 36 w/o uranium. Powders containing UAl₃ as the principal crystalline substance, as determined by X-ray diffraction were used as the fuel materials.

In Table IV, the pre- and post-irradiation thickness measurements of four plates which have been examined in the hot cell are compared. In two plates X8001 aluminum was used as the cladding and matrix material. In the others 6061 aluminum was used for the cladding with either X8001 or 6061 as the matrix.

From these data it is apparent that the aluminum alloys used for claddings and matrix materials have had little effect on the results as the dimensional changes in the plates were so nearly the same. The principal change which has taken place is in the thickness of the plates. This thickening is similar to that observed in other sample fuel plates, containing either alloy or cermet cores, which have been irradiated to similar levels under comparable conditions and apparently

TABLE IV

EFFECTS OF IRRADIATION ON THICKNESS OF SAMPLE FUEL PLATES USING POWDERED UAl_3 CORES

Sample Fuel Plate Composition		% Burn Up	Plate Thickness mm(mils)			Core Thickness mm(mils)		
Core	Cladding		Before	After	Change	Before	After	Change
50 w/o UAl_3 , U Fully Enriched + 0.19 w/o B_4C + 6061 Al Powder	6061-T6 Al	40	1.27 (50)	1.35 (53)	0.08 (3)	0.533 (21)	0.559 (22)	0.026 (1)
50 w/o UAl_3 , U Fully Enriched + 0.18 w/o B_4C + X8001 Al Powder	X8001-Al	25	1.30 (51)	1.33 (52.5)	0.03 (1.5)	0.533 (21)	0.544 (21.4)	0.011 (0.4)
	with 20% Cold Work	40	1.27 (50)	1.35 (53)	0.08 (3)	0.533 (21)	0.559 (22)	0.026 (1)
40 w/o UAl_3 , U Fully Enriched + 0.19 w/o B_4C + X8001 Al Powder	6061-O Al	24	1.30 (51)	1.30 (51)	0 (0)	0.555 (22)	0.555 (22)	0 (0)

is caused by the formation or enlargement of voids due to fission gas and helium buildup.

In Figure 4 the effects of irradiation on the microstructure of the cores of two of the plates are shown. As would be expected from the fact that the fuel plates had thickened, a number of voids are to be seen in the cores. Nearly all of the voids are associated with a fuel particle, but not all of the fuel particles show voids.

In the pre-irradiation microstructures, some porosity is to be seen. The cores of the plates had densities which were 91% of theoretical.

The microstructures of the core before and after irradiation are shown in Figure 5 in the etched condition. While edges of the fuel particles have rounded off slightly during irradiation very little reaction between the fuel particles and the 6061 aluminum alloy matrix can be detected.

Examinations made in the MTR and ETR canal of the other 47 plates which are undergoing testing have shown no defects.

b. Metallurgical Examination of Temperature Transition Zones of Aluminum-Uranium Alloy Fuel Element Meltdowns (M. J. Graber)

An objective of any fuel development program could well include the extension of the operating limits of the fuel material to the point of failure. Obviously only by so doing can the capabilities of the particular fuel be properly evaluated. Since such testing may result in some fuel meltdown it is important to study this mechanism of failure and the consequences to the test material and the test vehicle. Extrapolation of the results also provides valuable information for reactor safety analysis. This program therefore took advantage of an incident in the MTR which resulted in localized melting in a fuel element and this has been compared with fuel melting obtained in a SPERT test⁽⁶⁾.

A fuel plate in an MTR fuel element, J385, had undergone localized melting due to inadvertent partial blocking of the coolant flow channels during MTR Cycle 182. The plate, an 18 w/o aluminum-uranium alloy core clad with 1100 aluminum, was sectioned through the heat-affected area for metallographic examination. An interesting comparison can be made between this meltdown and one that occurred in a similar fuel plate, an 11.7 w/o aluminum-uranium core clad with 6061 aluminum.

Intergranular melting and cracking of the 6061 cladding occurred at a temperature below the melting point of the core, Figure 6. Cross-sections through the heat-affected area, show intergranular separation of the cladding in Figure 7 where partial melting of the core occurred and in Figure 8 where the core had not melted.

X8001 Al MATRIX

200X As Polished

6061 Al MATRIX

BEFORE AND AFTER IRRADIATION

BEFORE AND AFTER IRRADIATION

Figure 4 - Microstructure of Fuel Plate Cores Containing 50 w/o UAl_3 + 0.19 w/o B_4C
in Two Aluminum Alloy Matrix Materials

500X CHROMIC & ACETIC ETCH

500X H.F. ETCH

Before Irradiation

After Irradiation

Figure 5 - Microstructure of Core of Fuel Plates Containing UAl_3

2X

Figure 6 - A Meltdown of an 11.7 w/o Aluminum-Uranium Alloy Fuel Plate Clad with 6061 Al

Etched

50X

Figure 7 - Intergranular Cracking of 6061 Al Cladding and Melted U-Al Core

Etched

100X

Figure 8 - Intergranular Cracking of 6061-Al Cladding
Adjacent to Main Fission Break

The 1100 aluminum cladding, on the other hand, remained intact for a period of time after the core had melted, Figures 9 and 10. This can be explained by comparing the solidus temperatures of 582°C for 6061, 640°C for the aluminum-uranium alloys and 643°C for 1100 aluminum.

Since an extensive metallographic study was made on the 1100-clad plate, the heat-affected area can be divided into 6 zones, starting where microstructures appeared normal and progressing to the melted edge, totaling approximately 0.46 cm (See Figure 10 and Table V). The temperatures of the various zones were estimated from the melting point of the microconstituents.

The higher magnification core microstructure from Zone 1, Figure 11, appears normal. Even the small intermetallic particles in the hypereutectic matrix did not melt. This fact plus heat-transfer considerations indicate the temperature was less than 550°C.

In Zone 2, Figure 12, the core temperature was still less than, but approaching 640°C, the melting point of the core matrix. The strength of the core at this temperature was insufficient to prevent the formation of small voids around the fuel particles (the U-235 burnup at this point was estimated to be 18%).

Figure 9 - A Meltdown of an 18 w/o Aluminum-Uranium Alloy Fuel Plate Clad with 1100-Al
(Both Sides of Plate Shown)

Figure 10 - A Cross-Section of the Heat Affected Transition for the 18 w/o U-Al Fuel Plate Clad with 1100-Al

Zone		6	5	4	3	2	1
Temp °C	Clad	643-657	≈ 643	< 643	+640 -730	< 640	< 550
	Core		> 730	≈ 730			
Hardness	Clad	21-27	24-26	22-23		22	21-25
	Matrix		16-22*	19-30		32-38	58-82
DPN	Particle						131-162
Typical 500X Micro. Fig.		16	15	14	13	12	11

*Portion fused to outside of clad

TABLE V

TEMPERATURE, HARDNESS, AND 500X PHOTOMICROGRAPH CODE FOR THE VARIOUS ZONES OF MELTDOWN HEAT AFFECTED TRANSITION

M-1244

Etched

500X

Figure 11 - Normal Appearing Microstructure of 1100 Al
Clad Plate, Typical of Zone 1 of the
Heat-Affected Transition

M-1243

Etched

500X

Figure 12 - Voids Formed Around the Fuel Particles in
Melted Plate at Temperatures Below 640°C

The core temperature in Zone 3, Figure 13, was between 640°C and 730°C. The matrix had melted but UAl_4 particles remained partially intact. A sharp division in the grain size of the hypoeutectic matrix structure (now hypoeutectic because there is less than 13% uranium dissolved) came at a transition in void size. The fine grains indicate a more rapid cooling than the coarse grain area. It is believed, the larger voids provided an insulating effect.

The UAl_4 particles in the core at Zone 4, Figure 14, lost their shape and appear to have melted. Under non-equilibrium conditions (it is estimated the temperature surge lasted 10 seconds) there was insufficient time for diffusion and solution by the matrix. The temperature was at least 730°C to melt the UAl_4 particles.

In Zone 5 (see Figure 10) the core became completely fluid and thinned the cladding by melting and dissolving. The core temperature was in excess of 730°C (the maximum temperature cannot be determined from the microstructure). The cladding in this zone began to show intergranular melting, indicating a temperature of at least 643°C. Also in Zone 5 (and extending back to Zone 4) there is a solidified deposit of once molten core attached to the outside of the cladding. Figure 15 shows the outside surface of the cladding where the core deposit is attached. Pieces of cladding entrapped in this deposit indicate portions of the cladding failed by intergranular separation rather than complete melting. The temperature of the attached portion could not have been above 657°C (liquidus temperature of 1100) for an extended period.

The cladding in Zone 6 has cracks and dislodged grains due to intergranular melting. Figure 16 shows the outside surface where a layer of melted core was forced between the grains. Cladding, matrix and particle DPN hardnesses were taken through the heat-affected transition (See Table V). There were no significant hardness transitions in the cladding. The matrix showed a decrease in hardness with an increase in temperature. A DPN of 32 to 38 was found in Zone 2 where the void formation began. The fuel particles also showed a hardness transition in Zone 1, changing from 131 to 162 over a distance of about 0.07 cm. The softening in both the particles and matrix is attributed to annealing out of radiation or fission effects. The hardness for a normal fuel plate of this burnup is in the range of the maximum readings, i.e., DPN 162 for the fuel particles, 82 for the matrix and 27 for the clad.

In summary, a study of the heat-affected transition indicates the following effects as the temperature increased: (1) At about 500°C the radiation or fission hardening began to anneal. (2) At about 600°C voids formed around the fuel particles due to decrease in strength of the matrix. The voids had an insulating effect causing the particles to further increase in temperature. (3) Between 640 and 730°C the core matrix melted and the voids around the fuel particles increased in size. The plate began to swell. (4) Around 730°C the fuel particles melted and alloyed with the matrix improving heat transfer somewhat. However, large voids which were formed had an insulating effect. (5) Above 730°C the core became fluid and forced its way through the cladding intergranularly until the cladding collapsed. This released the pressure in the core allowing it to expand and flow out of the plate. The water pressure forced the molten core along the outside surface of the plate.

M-1242

Etched

500X

Figure 13 - An Area from Zone 3 of Melted Plate. Note the Sharp Change of Grain Size in the Re-solidified Core. Some of the Fuel Particles have Retained Their Shape.

M-1245

Etched

500X

Figure 14 - A Typical Area from Zone 4 of Melted Plate. The Fuel Particles Have Lost Their Shape and Appear as Uranium-Rich Areas at the Edge of Voids.

M-2446

Etched

500X

Figure 15 - Zone 5 of Melted Plate.
A Portion of Melted Core Attached Itself to the Outside
of the Cladding. Note the Entrapped Cladding, Upper Left.

M-1248

Etched

500X

Figure 16 - Zone 6 of Melted Plate.
A Layer of Melted Core on the Outside Surface of the
Cladding was Forced Between the Cladding Grains.

c. The Effects of Radiation and Elevated Temperature on
Tensile Properties of Aluminum Fuel Plates (M. J. Graber)

Most radiation damage work evaluating aluminum alloys has been confined to ambient temperature testing of non-fuel-containing specimens. Designs for fuel elements, of necessity, have been based on these radiation effects data. In addition to radiation damage, fission damage occurs, as shown in IDO-16799(3) and IDO-16727(2), by large increases in hardness in aluminum fuel plate matrices.

A number of sample fuel plates and cladding materials, with various neutron exposures, were tensile-tested at ambient temperatures and at short-time temperatures of 93°C and 204°C (Table VI and VII) to study the effects of radiation, temperature, type of fuel, weight per cent of fuel, and type of cladding. The core materials used were aluminum-uranium alloys and powder metallurgy compacts of 1100 aluminum combined with either UO₂ or U₃O₈. The fuel plates studied were clad with 1100, X8001 or 6061 aluminum. Non-fueled aluminum materials tested were 1100, 5052-O, 6061-O, 6061-T4, 6061-T6, and XAP001.

The plates were irradiated in the L-51 position of the MTR in a holder specially designed for test plate irradiation. The fast (> 1 Mev) flux was approximately 1×10^{13} n/cm²/sec and the surface temperature of the plates was approximately 63°C. After irradiation and a suitable cooling period in the MTR canal, the plates were machined to a subsized of 1" x 1/4" x (plate thickness) in the gauge area plus tabs for gripping. Specimens were at test temperature from 10 to 30 minutes when tested.

Room temperature and 93°C tests show that the increase in fuel plate strength (or the slope of the curves in Figures 17 and 18) due to radiation (or fission) is dependent, primarily, on the type of cladding material. But, at the 204°C test temperature, Figure 19, the increase (or decrease) in the fuel plate strength is dependent on the type of fuel and the fuel loading as well as the type of cladding.

The effect of temperature on unirradiated fuel plates, Figure 20, as might be expected, is to lower the plate strength. This occurs to a greater degree on the irradiated plates, Figure 21. At 204°C and about 28% burnup the plate strengths for the 1100 clad and X8001 clad plates are about equal to their unirradiated room temperature strengths.

Plates clad with X8001 react to irradiation and temperature similarly to those clad with 1100 Al. With the exception of plates having higher fuel loadings, strengths were increased by radiation and decreased by elevating the test temperature. After irradiation, and for a given fuel loading (in the range of 18 to 32 w/o), alloys are stronger than oxides (Figure 21). Radiation has a greater strengthening effect on UO₂ plates than on U₃O₈ plates (compare Figure 20 and 21).

For a given type of fuel and cladding material the effect of increased fuel content is to lower the initial plate strength.

TABLE VI
FUEL PLATE TENSILE PROPERTIES

Plate Composition	Comp. No.	Room Temperature					Plate No.	93°C					Plate No.	204°C				
		NVT >1 Mev X10 ⁻¹⁹	% Burnup	T.S. kg/cm ² (10 ³ psi)	Y.S. kg/cm ² (10 ³ psi)	Elong- ation in 1"		NVT >1 Mev X10 ⁻¹⁹	% Burnup	T.S. kg/cm ² (10 ³ psi)	Y.S. kg/cm ² (10 ³ psi)	Elong- ation in 1"		NVT >1 Mev X10 ⁻¹⁹	% Burnup	T.S. kg/cm ² (10 ³ psi)	Y.S. kg/cm ² (10 ³ psi)	Elong- ation in 1"
45 wt% U, 93% Enriched +3% Si + Al Alloy Core 6061 T Al Clad	A	0 2.82 20.36	-- 18 75	(17.1) (25.2) (30.0)	(12.4) (21.1) (22.9)	8.6 3.9 12.6												
18 wt% U, 93.5% Enriched in Al Alloy core	B	0 1.33	-- 9	1180 (16.8) (27.5)	650 (9.3) (22.3)	24.3 3.2	100	0	--	970 (13.9)	550 (7.8)	20.8	98	0	--	760 (10.8)	470 (6.7)	21.1
1100 Al Clad		5.10	29	2270 (32.4)	2070 (29.6)	1.4							44	5.10	29	1320 (18.8)	1020 (14.6)	3.4
25 wt% U, 20% Enriched + Al Alloy Core	C	0 1.33	-- 9	1140 (16.3) (26.6)	650 (9.3) (19.5)	18.4 5.8	99	0	--	960 (13.7)	620 (8.8)	15	100	0	--	710 (10.2)	400 (5.7)	?
1100 Al Clad		9.70	49	2040 (29.1)	1640 (23.5)	4.1	7	10.58	52	1770 (25.3)	1040 (14.9)	7.7	9	21.86	78	1560 (22.3)	1130 (16.1)	6.5
22.3 wt% U ₃ O ₈ 93% Enriched +Al Cermet Core,	D	0 5.82	-- 33	1300 (18.5) (22.6)	1190 (17.0) (18.1)	1.4 2.0	5	0	--	1080 (15.4)	970 (13.9)	1.8	6	0	--	850 (12.1)	680 (9.7)	2.3
1100 Al Clad				1220 (17.5)	1220 (17.5)		5	16.73	69	--	--	--	14	22.62	79	940 (13.4)	880 (12.5)	.6
22.7 wt% UO ₂ 93.5% Enriched in Al Matrix + .123 wt% B 1100 Al Clad	E	0 1.29	-- 8	1130 (16.1) (21.4)	1030 (14.7) (14.7)	4.1 5.5	98	0	--	1070 (15.3)	1000 (14.3)	2.5	100	0	--	790 (11.3)	670 (9.6)	2.4
							7	22.52	79	1550 (22.2)	--	--	7	22.52	79	1120 (16.0)	--	--
36 wt% UO ₂ 93.5% Enriched in Al Cermet Core	F	0 1.77	-- 11	1020 (14.6) (19.4)	880 (12.6) (17.4)	1.8 1.2	2	0	--	780 (11.2)	680 (9.7)	1.6	5	0	--	620 (8.8)	590 (8.4)	1.1
1100 Al Clad				1220 (17.4)	1220 (17.4)		7	2.48	16	1280 (18.3)	890 (12.7)	2.3	7	2.48	16	880 (12.6)	--	--
							8	6.89	38	1510 (21.6)	1250 (17.9)	1.4	15	7.11	39	1190 (17.0)	1030 (14.7)	1.0
54 wt% UO ₂ , 20% Enriched + Al Cermet Core 1100 Al Clad	G	0 1.19	-- 8	900 (12.7) (19.9)	890 (12.7) (18.0)	.6 1.3	100	0	--	850 (12.1)	840 (12.0)	.5	99	0	--	700 (10.0)	660 (9.4)	.8
				1260 (18.0)	1260 (18.0)		20	5.42	31	1180 (16.8)	1070 (15.3)	1.0						
32 wt% U, 93% Enriched in Al Alloy Core	H	0	--	1100 (15.7)	930 (13.3)	5.6	100	0	--	930 (13.3)	670 (9.6)	3.2	99	0	--	700 (10.0)	600 (8.5)	2.9
X8001 Al Clad							2	38		490 (7.00)	1580 (22.5)	--	7	11.51	55	1050 (15.0)	--	--
							6	59		850 (12.2)	1420 (20.3)	--	6	12.2	59	1020 (14.6)	--	--
50 wt% UAl ₃ (69 wt% U, Fully Enriched) +.19 B ₄ C + 6061-T4 Al Powder Cermet 6061-T4 Al Clad	I	0	--	2240 (32.0)	2170 (31.0)	1.0	1	0	--	2100 (30.0)	2010 (28.7)	.9	3	0	--	1720 (24.5)	1600 (22.8)	1.6
													11	8.09	43	1410 (20.2)	--	--
	J	0		1090 (15.6)	--	--	97	0	--	1050 (15.0)	1040 (14.9)	.6	98	0	--	790 (11.3)	740 (10.5)	.8
													4	8.09	--	700 (10.0)	--	--

TABLE VII

CLADDING MATERIAL TENSILE PROPERTIES

Cladding Material	Room Temperature				Plate No.	93°C				Plate No.	204°C			
	NVT >1 Mev $\times 10^{-19}$	T.S. kg/cm ² (10 ³ psi)	Y.S. kg/cm ² (10 ³ psi)	Elong- ation in 1"		NVT >1 Mev $\times 10^{-19}$	T.S. kg/cm ² (10 ³ psi)	Y.S. kg/cm ² (10 ³ psi)	Elong- ation in 1"		NVT >1 Mev $\times 10^{-19}$	T.S. kg/cm ² (10 ³ psi)	Y.S. kg/cm ² (10 ³ psi)	Elong- ation in 1"
1100 - 0 HT 316°C	0	930 (13.3)	410 (5.8)	37.9		0	760 (10.8)	410 (5.9)	37.9		0	530 (7.6)	290 (4.2)	37
1100 - 0 HT 538°C	0	940 (13.5)	460 (6.6)	35.6		0	890 (12.7)	(8.0)	33.0		0	670 (9.6)	430 (6.2)	31.3
6061-T6	0	3480 (49.7)	3090 (44.2)	13.1		0	3180 (45.5)	2880 (41.2)	11.1		0	2620 (37.4)	2440 (34.8)	11.2
					2	4.60	2990 (42.7)	2130 (30.4)	22.2	4	5.20	2370 (33.9)	1840 (26.3)	18.5
6061-T4					2	5.15	2670 (38.1)	1760 (25.2)	21.2	3	4.95	2110 (30.2)	1530 (21.8)	19.0
6061-0					2	5.15	2030 (29.0)	1420 (20.3)	9.7	3	4.80	1550 (22.1)	1230 (17.6)	10.4
APM-257	0	3030 (43.3)	2570 (36.7)	6.5		0	2600 (37.2)	2130 (30.4)	3.2		0	1870 (26.7)	1490 (21.3)	4.8
	4.90	2680 (38.3)	2420 (34.5)	6.2	2	4.80	2550 (36.4)	2300 (32.8)	5.3	3	5.20	2170 (31.0)	1900 (27.2)	5.1
APM-583					2	4.80	2210 (31.6)	2050 (29.3)	7.5	4	5.20	1960 (28.0)	1770 (25.3)	6.7
5052-0					4	4.80	1970 (27.5)	850 (12.1)	26.6	5	5.10	1580 (22.6)	880 (12.6)	36.9
5052-H18					3	4.80	2840 (40.5)	2450 (35.0)	5.1	4	4.80	2430 (34.7)	2230 (31.8)	8.7
8001										3	5.00	740 (10.6)	460 (6.5)	33.4
8002					2	4.90	1710 (24.2)	1280 (18.3)	11.2	4	5.00	1170 (16.7)	920 (13.2)	17.8
8003										3	5.00	1660 (23.7)	1440 (20.5)	12.2

Figure 17 - The Effect of Radiation on the Ambient Temperature Strength of Fuel Plates and Cladding Materials.

Figure 18 - The Effect of Radiation on the Short-Time Elevated Temperature Strength (< 30 min at 93°C) of Fuel Plates and Cladding Materials.

Figure 19 - The Effect of Radiation on the Short-Time Elevated Temperature Strength (< 30 min at 204°C) of Fuel Plates and Cladding Materials.

Figure 20 - The Effect of Temperature on the Strength of Unirradiated Fuel Plates and Cladding Materials

Figure 21 - The Effect of Temperature on the Strength of Fuel Plates and Cladding Materials Irradiated to 5×10^{19} Fast (> 1 Mev) Neutron Exposure, n/cm^2

The fuel plates generally showed a decrease in ductility due to increases in temperature and increases in radiation exposure.

d. Effect of Irradiation on the Densities of Aluminum Clad Sample Fuel Plates (G. W. Gibson)

As part of the studies being made to evaluate the effects of irradiation on aluminum-base fuel-containing plates, density determinations were made on a number of irradiated sample fuel plates. Standard sample fuel plates, fabricated in the normal manner, and irradiated at ambient conditions in the L-51 position of the MTR(1,2,3) were used for these determinations. To date, plates containing cores of wrought U-Al alloys, powdered UAl₃ compound, UO₂ dispersed in aluminum and U₃O₈ dispersed in aluminum have been tested in this manner. The results of these tests are shown in Figure 22.

Figure 22 - Effect of Irradiation on the Densities of Aluminum Clad Sample Fuel Plates with Different Core Materials

All of the fuel used in these specimens was fully enriched uranium. The irradiation exposures represent burnups of up to 80% of the original U-235 atoms present in the samples.

As can be seen in the Figure all of the samples experienced a decrease in density during irradiation. This decrease can readily be accounted for by the increase in porosity seen in the microstructure of the plates during irradiation^(1,2,3).

ORNL investigators⁽⁷⁾ reported an increase in the irradiated density of U_3O_8 dispersed in aluminum while other materials reported by the same investigators showed a decrease. The uranium used in the Oak Ridge U_3O_8 plates was 20% enriched.

The density changes produced by the irradiation of uranium-aluminum alloy-cored plates are in good agreement with the Oak Ridge results. The density change appears to be independent of whether the core material is a wrought alloy of uranium and aluminum or a powdered alloy of these materials.

Calculations of the core density changes which would occur when a thermit type reaction such as $3 U_3O_8 + 52 Al \longrightarrow 9 UAl_4 + 8 Al_2O_3$ indicate that the density of the core material would increase. The formation of gaseous elements due to the fissioning of U-235 would tend to decrease the density of the cores. In a fuel plate containing 20% enriched uranium the density increase due to the chemical reaction is apparently greater than the density decrease due to fission gas formation. However, when fully enriched uranium is used the effect of fission gas is greater than that of the chemical reaction.

2. High Temperature Irradiation

a. Description of the HB-2 Loop (W. P. Scarrah)

At the beginning of the fiscal year, this program assumed responsibility for the HB-2 facility. This loop had previously been used by the Argonne National Laboratory for aluminum corrosion studies.

The HB-2 facility (Figure 23) is a high pressure (1500 psi, 105 kg/cm²), high temperature (260°C), circulating water loop located in the MTR. The in-pile facility is in a horizontal tube having its tip adjacent to the center of the north face of the reactor core. Holders (Figure 24) were used to irradiate up to twelve sample fuel plates (5.55 cm x 7.30 cm x 0.127 cm). This design enabled existing plate fabrication equipment to be used.

As the HB-2 is located outside the reactor core, the neutron flux is at a maximum in the tube tip and decreases rapidly away from the tip. Even though the small cross-sectional area of the tube limits the number of plates in the high flux holder, effects due to the rate of irradiation can be investigated by placing identical plates in different holders with significantly different neutron fluxes. The low pH and hydraulic conditions prevailing in the HB-2 are especially

Figure 23 - Isometric Sketch of HB-2 Loop in MTR

Figure 24 - Sample Holder and Samples Used in HB-2 Loop

conducive to the formation and deposition of stainless steel corrosion products (crud) on the sample fuel plates. However, as the temperature of the plates is primarily determined by the coolant temperature, fuel stability experiments are especially suited for this facility.

b. Irradiation Testing

Fuel materials irradiated included UO_2 -Al cermet (23-35 wt-%), U_3O_8 -Al cermet (22-35 wt-%), U-Al alloys (18 wt-%) and UAl_3 intermetallics (50 wt-%). The cladding materials tested have included 2219, 6061, and X8001 aluminum alloys, and APM M-768, APM M-769 and APM M-770 aluminum powder metal products.

Most irradiations were at 230°C with only one loading at 180°C. However, lower temperatures had to be maintained in nearly every cycle due to flow channel blockage by crud.

c. Out-of-Pile Testing in HB-2 Loop (G. W. Gibson, W. P. Scarrah)

The out-of-pile test section of the HB-2 loop has been used to provide comparative corrosion test data on the specimens undergoing irradiation and to test potential cladding materials prior to using these materials on sample fuel plates. Materials tested have been 2219, 6061, and X8001 aluminum alloys and aluminum powder metallurgy product APM M-768, M-769, and M-770. The nominal compositions of these materials are shown in Table III.

The sample holder, made from 6061 aluminum, and sample plates are shown in Figure 24. The same size holder and plates were used in both the in-pile and out-of-pile test sections of the loops. This holder, as can be seen in the picture, held 12 sample plates.

The plates were prepared for exposure in the loop by first cleaning with a detergent solution, then rinsing with demineralized water, washing with acetone, and air drying. The loop conditions during the time the samples were under test are given in Tables VIII and IX.

After exposure in this loop the oxide scale was removed from the plates by an electrolytic descaling technique using boric acid as the electrolyte.

Hot cell examinations had not been completed by the end of the fiscal year on the samples which had been irradiated; however, the examination of samples which had been tested in the out-of-pile section of the loop was completed.

The results of these tests have been plotted in Figure 25. The times at which the samples were at temperatures greater than 150°C have been plotted against the weight loss of the samples. By choosing different temperatures for the cut-off temperature different sets of corrosion rates could be obtained. But, the relative standing of the materials when compared against X8001 aluminum would be the same. One sample of APM M-768 showed a large weight gain during testing and was

TABLE VIII

SUMMARY OF CONDITIONS OF TESTING IN HB-2 LOOP

MTR Cycles	185-186	189-190-191
Total Time in Loop - hrs	887.2	1521.0
Temperature (See Table II)		
Pressure	105 kgs/cm ²	105 kgs/cm ²
Flow Rate	(4.27 m/sec) 14 ft/sec	(4.27 m/sec) 14 ft/sec
Ratio of Minimum Surface Area to Loop Water Vol.	125 $\frac{\text{cm}^2}{\text{liter}}$	150 $\frac{\text{cm}^2}{\text{liter}}$
pH		
Average	6.0	5.7
Maximum	7.5	9.5
Minimum	5.0	3.6
Dissolved Oxygen (ml/liter)	0.01	17
Dissolved Hydrogen		
Average	3.6	3.1
Maximum	8.5	9.9
Minimum	1.4	0.5
Dissolved Nitrogen	0.2	152

TABLE IX

TEMPERATURE CONDITIONS FOR HB-2 LOOP

<u>Temperature of Loop °C</u>		<u>Time at Temperature - Hours</u>	
		<u>185-186</u>	<u>189-190-191</u>
238	249	1.5	--
227	238	276.5	794.0
204	227	16.7	21.6
177	204	6.0	18.3
149	177	58.7	13.9
121	149	96.7	132.0
93	121	16.3	229.8
66	93	286.5	124.8
38	66	114.0	124.8
10	38	<u>14.3</u>	<u>61.8</u>
		887.2	1521.0

Figure 25 - Corrosion Rates of
Aluminum Materials Tested in Out-
of-Pile Section of HB-2 Loop

badly swollen. The descaling procedures would not reduce the sample weight to that of its weight before exposure. The data for this sample have not been plotted in Figure 25.

From these data it would appear that the corrosion resistance of the aluminum powder metallurgy materials under these test conditions is equivalent to that of X8001 aluminum, while that of the commercially available 2219 and 6061 aluminum alloys is slightly worse. All of the materials tested are stronger than X8001.

d. Uranium Oxide Reactions with Aluminum (M. J. Graber)

In our last annual report⁽³⁾ it was stated that a UO_2 -Al dispersion-type fuel plate showed unexpected microstructural changes when irradiated at elevated temperatures in the MTR. Oxide fuel particles, when irradiated at MTR conditions, usually appear as dark gray areas surrounded by light gray reaction zones and almost always include many voids. When irradiated at elevated temperature, the fuel particles became a light gray-to-white and were quite uniform (as observed by optical microscopy). Additional samples of this and other compositions have since been irradiated at elevated temperatures (i.e., 175-200°C) and to higher burnups with similar results.

Table X lists, for comparison, specimens which have been heat-treated without irradiation, which have been irradiated at MTR conditions, and which have been irradiated at elevated temperature. Figure 26 shows an unirradiated UO_2 -Aluminum dispersion plate, after

H-53

As Polished

520X

Figure 26 - Microstructure of 42 w/o UO_2 + Al, Heat-Treated 40 Hours at 600°C

TABLE X

TESTING CONDITIONS FOR URANIUM OXIDE-ALUMINUM CERMET CORED SAMPLE FUEL PLATES

Composition	Uranium Enrichment	nvt (th) 10^{20}	Irradiation Conditions	Microstructure
42 w/o UO_2 + Al	40%**	0	-----*	Light Gray Particles $UA1_3$ - $UA1_4$ Dark Gray Particles UO_2
65 w/o UO_2 + Al	20%**	3.7	MTR	Non-Uniform Partially Reacted Particles
24 w/o UO_2 + Al	full	10.3	MTR	Non-Uniform Partially Reacted Particles
22 w/o U_3O_8 + Al	full	10.6	MTR	Non-Uniform Partially Reacted Particles
65 w/o UO_2 + Al	20%**	17.9	Elevated Temperature in MTR	Three Distinct Particles Dark Gray, light gray porous and light gray non-porous
24 w/o UO_2 + Al	full	2.8	Elevated Temperature in MTR	Uniform Totally Reacted Particles
24 w/o UO_2 + Al	full	20.6	Elevated Temperature in MTR	Uniform Totally Reacted Particles
24 w/o UO_2 + Al	full	26.6	Elevated Temperature in MTR	Uniform Totally Reacted Particles
22 w/o U_3O_8 + Al	full	17.8	Elevated Temperature in MTR	Uniform Totally Reacted Particles

*This specimen was heat treated at 600°C for 40 hours but was not irradiated.

**This enrichment was obtained by blending natural uranium with fully enriched uranium.

heat-treating for 40 hours at 600°C. This shows the usual dark gray UO_2 particles and the light gray UAl_3 - UAl_4 particles. Uranium oxide-aluminum dispersion plates irradiated at MTR conditions are seen to contain the typical, partially reacted, non-uniform particles in Figures 27, 28, and 29.

With the exception of Figure 30, the uranium-oxide particles "reacted" completely under irradiation at elevated temperatures giving a lighter color and more uniform particle, [Figures 31 through 33 ($UO_2 + Al$) and Figure 34 ($U_3O_8 + Al$)]. Figure 30 shows the microstructure of a blend of natural and fully enriched $UO_2 + Al$. Three distinct areas are observed, a light gray with small pores, a light gray-to-white non-porous area and a dark gray "unreacted" area.

A report of the X-ray diffraction analysis is given in Table XI, for $UO_2 + Al$ and uranium-aluminum alloy, UAl_4 , irradiated at MTR conditions and for $UO_2 + Al$ irradiated at elevated temperatures. None of the samples after irradiation showed the characteristic peaks for UO_2 , UAl_3 , or UAl_4 . One of the $UO_2 + Al$ samples and the U-Al alloy sample each had unidentified peaks of a dissimilar nature. Some interference was caused by the bakelite mounting material, possibly masking the characteristic lines required for fuel particle identification.

Continued study of the UO_2 -Al reaction under irradiation has not completely defined the mechanism. The loss of crystallinity of UO_2 and Al_2O_3 under long irradiation exposure complicates the detection of these materials. The additional investigations since the last report, however, have not refuted the earlier conclusion that uranium oxide-aluminum fuels irradiated at elevated temperatures show a different microstructure from those irradiated at reactor ambient temperature.

C. Beryllium Program (J. M. Beeston and E. H. Porter)

Several complementary investigations on the effects of irradiation on beryllium are presently underway in Phillips Petroleum Company, Atomic Energy Division. Work on gas release from beryllium by means of post-irradiation annealing has been reported quarterly by R. L. Tromp since the second quarter in 1959(8). An example of the effect of the gas on the microstructure of the beryllium is shown in the electron micrograph reproduced in Figure 35. This electron micrograph is from a palladium-shadowed carbon replica of a freshly fractured surface of irradiated beryllium and shows the porosity within the metal due to the accumulated bubbles of irradiation-produced gas. An MTR beryllium damage investigation was reported for the second quarter 1963(9). An ATR beryllium surveillance study is presently planned with irradiation in the ETR until the ATR start-up. The Fuels and Materials Development Program is concerned with the high temperature irradiation of beryllium in lead capsules. This work has been in progress for some time, but difficulties with the capsules after insertion in the ETR necessitated their removal.

Three lead capsules each containing three beryllium compression samples were inserted in position E-14 in ETR, in order to determine the strength, gas release, and growth during a high temperature (600-800°C)

M-526

As Polished

500X

Figure 27 - Microstructure of 65 w/o UO_2 + Al, 20% Enriched, Irradiated at MTR Conditions

M-833

As Polished

500X

Figure 28 - Microstructure of 24 w/o UO_2 + Al, Fully Enriched, Irradiated at MTR Conditions

M-786

As Polished

500X

Figure 29 - Microstructure of 22 w/o U_3O_8 + Al,
Fully Enriched, Irradiated at MTR Conditions

M-1127

As Polished

500X

Figure 30 - Microstructure of 65 w/o UO_2 + Al, 20%
Enriched Irradiation at Elevated Temperature

M-908

As Polished

500X

Figure 31 - Microstructure of 24 w/o UO_2 + Al, Fully Enriched, Irradiated at Elevated Temperature

M-1128

As Polished

500X

Figure 32 - Microstructure of 24 w/o UO_2 + Al, Fully Enriched, Irradiated (to greater nvt than Figure 31) at Elevated Temperature

M-1126

As Polished

500X

Figure 33 - Microstructure of 24 w/o UO_2 + Al, Fully Enriched, Irradiated (to greater nvt than Figure 32) at Elevated Temperature

M-1125

As Polished

500X

Figure 34 - Microstructure of 22 w/o U_3O_8 , Fully Enriched, Irradiated at Elevated Temperature

TABLE XI

X-RAY DIFFRACTION ANALYSIS OF URANIUM OXIDE-ALUMINUM AND URANIUM-ALUMINUM ALLOY SAMPLE FUEL PLATES

<u>Composition</u>	<u>Uranium Enrichment</u>	<u>nvt (th) 10^{20}</u>	<u>Irradiation Conditions</u>	<u>X-Ray Diffraction</u>
40 w/o UO_2 + Al	60%	17.0	MTR	Unidentified Peaks No UO_2 UAl_3 or UAl_4
40 w/o UO_2 + Al	60%	22.2	MTR	No UO_2 UAl_3 UAl_4
18 w/o U-Al Alloy (UAl_4)	full	2.8	MTR	Unidentified Peaks No UAl_3 or UAl_4
24 w/o UO_2 + Al	full	2.8	Elevated Temperature in MTR	No UO_2 UAl_3 or UAl_4

Figure 35 - Electron Micrograph of Freshly Fractured Surface of Irradiated Beryllium

irradiation. A disassembly view of one of these capsules is shown in Figure 36 and the assembled capsule ready for reactor insertion is shown in Figure 37. The information to be provided by these high temperature in-reactor irradiations is essential to assess the effect of temperature on long term irradiations, and the effect of radiation on the diffusion and agglomeration of the helium gas with subsequent swelling. It has been shown in the beryllium gases experiment that post-annealing of beryllium irradiated to 1×10^{21} n/cm² (> 1 Mev) for 30 hours at 750°C is required for appreciable swelling to occur. Any deviation due to in-reactor conditions needs to be established before the problem of the behavior of helium gas in beryllium is completely defined.

The instrument panel for controlling the temperature of the in-pile lead capsule beryllium as installed on the ETR mezzanine is shown in Figure 38. Control samples of beryllium are heated at equivalent

Figure 36 - Beryllium Irradiation Capsule Disassembly

Figure 37 - Beryllium Irradiation Capsule

Figure 38 - Instrument Panel of Beryllium Capsule Experiment

temperature and similar length of time out-of-pile to assess the effect of the high temperature on the beryllium for these extended periods. A view of the out-of-pile capsule is shown in Figure 39 while furnaces and control instrumentation for this capsule are shown in Figure 40.

Leaks and thermocouple failures developed in the in-pile capsules before appreciable exposure had been achieved and all were removed from the reactor. The capsule design is being re-evaluated to improve the reliability for continuing these irradiations.

Figure 39 - Beryllium Control Capsule

Figure 40 - Furnace and Control Instrumentation
for Out-of-Pile Beryllium Experiment

III. PROTOTYPE STUDIES

A. 32-Plate MTR Fuel Element (M. L. Griebenow)

An MTR fuel⁽³⁾ element employing advanced metallurgical techniques to optimize the hydraulic and heat transfer characteristics was successfully irradiated in the MTR. The fuel element, 32-2, consisted of 32 fuel plates (0.76 cm-0.030" thick) containing 250 grams of U-235 in a U₃O₈-Al dispersion. It was irradiated in positions 11 and 19 for portions of 5 cycles extending from October 8, 1962 to February 27, 1963. The element was in the reactor longer and had a greater total U-235 fissioned (74 grams compared to approximately 60 grams) but was subjected to a slightly lower fission rate than an average MTR element. This element has a capability of increasing the reactor power by 46% or reducing the pumping rate (gpm) by 44% while maintaining the existing maximum surface temperature.

The spent fuel element is being stored in the MTR canal until post-metallurgical examination can be started.

A second element, 32-1, identical to 32-2, will be irradiated when the post-metallurgical examination of the first element is completed.

B. Side Plate-Less Fuel Element Design (G. W. Gibson)

The plate-type fuel elements used in nuclear reactors have the advantages of being a proven design for which fabrication techniques are well developed, and of providing a large amount of heat transfer surface area for a given volume of fuel material. In the conventional design, however, heavy side plates are required to firmly anchor the fuel plates to withstand the hydraulic forces developed in the reactor. The resulting narrow rectangular flow channels produce the undesirable results which are discussed elsewhere in this report.

One means used to alleviate this problem was to develop a design of a plate type fuel element in which side plates were not used. In this type of element streamlined spacers were used instead of side plates to separate and position the fuel. In order that this method of assembly could be used a procedure for leaving areas in the fuel plate cores free of fuel had to be developed. The procedure was to punch holes through the cores of the plates before the cores were assembled with the picture frames and cover plates. The cores were next assembled with picture frames and cover plates after which the plates were rolled in a conventional manner. Figure 41 is a print of a radiograph

Figure 41 - Print of Radiograph of Sample Fuel Plate Showing Area in Core Left for Fuel Plate Spacers

showing the oval-shaped area, free of fuel, produced in a small fuel plate by this technique. The streamlined fuel plate spacers were assembled over these areas.

Using this method of plate spacing a prototype ETR fuel element was fabricated. A view of this element is shown in Figure 42.

Figure 42 - Prototype ETR Fuel Element Made Without Side Plates

In this element dummy fuel plates of 6061-T6 aluminum 1.27 mm thick were used. The element was hydraulically tested to failure which started to occur under the following conditions:

	<u>At Time of Failure</u>	<u>% of Normal</u>
Flow - gpm	775	141
ΔP - psi	81.0	180

In addition to being safer than a plate-type element of conventional design this type of element might offer certain economic advantages. Only simple sheet metal working tools were used to make the prototype element. Consequently, the initial tooling costs could be low. If after final assembly an element was found to not meet specifications, it could be easily disassembled, the problem corrected, and the element reassembled. Thus, the reject rate would be low. Finally, this type element could be easily disassembled after irradiation so that only the fuel plates would have to be reprocessed. This would lower the reprocessing costs.

C. Heat Transfer Tests on Simulated ETR Fuel Geometries (M. L. Griebenow)

Additional ac resistance-heated, ex-reactor, tests simulating ETR conditions have been performed⁽²⁾. All convective heat transfer data fell within $\pm 20\%$ of the Modified Colburn Equation (shaded area in Figure 43) indicating this equation with a 20% safety factor, as currently used by Phillips, is adequate. However, the Dittus-Boelter Equation had a 15% accuracy and followed the data trend much better in the high heat flux range ($1-2 \times 10^6$ Btu/hr-ft). For this reason and since it eliminated the

Figure 43 - Wall Temperature vs Heat Flux for ETR Flow, Pressure, and Channel Dimensions

Figure 44 - Edge Heat Transfer

Figure 45 - Optimized MTR Roughened Channel

need for frequent trial-and-error solution the use of the Dittus-Boelter Equation with a 15% safety factor is recommended for forced convection heat transfer in rectangular channels.

The Jens and Lottes Correlation fits all fully developed nucleate boiling heat transfer data, from the same simulated ETR assembly, to within $\pm 7^\circ\text{C}$. Burnout occurred at 85% of the value predicted by the improved Bernath Correlation justifying the continued use of this equation with a 40% safety factor.

The forced convection heat transfer from the corners and the short walls (simulated side plates) correlated well assuming the velocity was reduced by 43% in these regions (Figure 44).

D. Artificially Roughened Fuel Plates (M. L. Griebenow)

The anticipated advantages of turbulence promoters in the form of a roughened fuel plate surface have been reported⁽²⁾. In attempting to obtain commercial means of roughening the heat transfer surface laboratory experiments were performed on the various shapes of promoter surfaces. It was found that the friction factor for a square edge protrusion (most easily obtained by commercial methods) did not correlate well with ϵ/D (protrusion height/hydraulic diameter).

To check the Reynolds Analogy (i.e., the film heat transfer coefficient is proportional to the friction factor) that was employed in the computer optimization⁽²⁾ a heat transfer test assembly with an MTR cross section and 30.5 cm long was fabricated with the recommended degree of roughness. The roughness shape was that produced by the knurl previously reported in reference 2. The results of the heat transfer test are shown in Figure 45. The data, with one exception, agree well with the values predicted. The data point at 181°C and $9 \times 10^5 \text{ Kcal/m}^2\text{hr}$ appears somewhat low but the Reynolds Analogy appears to hold quite well.

If the existing heat transfer facility capabilities can be expanded to higher power it is planned to test a roughened assembly to burnout as a preliminary to reactor testing of such an element.

IV. CONCLUSIONS

1. The fiscal year 1963 saw the development of a new fuel material, powdered UAl_3 intermetallic compound, offering possibilities of improvements over either the oxide or the alloy fuels.
2. Fuel plate cladding failures due to excessive temperature occur at a lower temperature in 6061 aluminum alloy than in 1100 aluminum alloy. Core failures by the formation of voids which surround the fuel particles will occur at temperature surges between $288^{\circ}C$ ($550^{\circ}F$) and $316^{\circ}C$ ($600^{\circ}F$) in low weight % uranium-aluminum alloy cores.
3. Tensile tests on sandwich-type fuel plates indicate that the strength is dependent primarily on the cladding material at low (MTR conditions) temperatures but at temperatures in the vicinity of $200^{\circ}C$ (ATR conditions) the fuel plate strength is strongly influenced by the core material.
4. All of the highly enriched fuel compositions tested (i.e., UO_2 , U_3O_8 , UAl_3 in aluminum matrices) exhibited density decreases under irradiation.
5. Preliminary work on newly developed (by Alcoa) aluminum powder metal products (APM M-768, 769, and 770) indicates that these materials have corrosion properties equal to X8001 while possessing much superior high temperature strength.
6. Development of plate-type elements without side plates indicates feasibility, but that complete communication between channels will not prevent collapse under certain conditions.
7. Work on roughening heat-transfer surfaces continued to show attractive results but commercial techniques need to be developed for producing suitable surfaces.

V. FUTURE WORK

In FY 64 it is planned to demonstrate the engineering feasibility of the powdered UAl_3 fuel by (1) obtaining sufficient quantities of this material through commercial sources to establish procedures, yields, and realistic costs, and (2) obtaining sufficient numbers (e.g., ten) of MTR and ETR fuel elements made with this fuel to permit statistical evaluation of fabrication properties (i.e., rollability, homogeneity, and formability) and to "proof-test" these production elements in the reactors. Since it is expected that the fully-fuelled elements will attain normal burnup in both reactors, the costs for these elements will be absorbed as part of the fuel for the MTR and ETR.

Other prototype irradiations planned for FY 64 include a fiberglass MTR element fabricated by the Clevite Corporation and the remaining 32-plate high strength fuel element not tested in FY 63.

New fuel materials offering higher temperature potential than those investigated in FY 63 will be studied. Compositions involving beryllium indicate interesting fuel possibilities on the basis of computer studies and preliminary corrosion tests. These materials will be tested as fuel plate samples in the reactor and in out-of-pile corrosion loops.

Additional work is planned for the UAl_3 intermetallics to determine the effects of stabilizing the UAl_3 with silicon and tin. The irradiation stability of the two types of fuels will be compared. Microprobe studies will be conducted on the UAl_3 fuel to determine (1) changes in composition of fuel particles in stabilized and unstabilized conditions and after heating, and (2) diffusion effects as revealed by concentration gradients in the vicinity of the fuel particles. Similar studies will be continued on the uranium-oxide aluminum reaction. The microprobe investigations will be correlated with electron microscopy, electron diffraction, and X-ray diffraction studies.

Work is scheduled to continue on aluminum alloys having good high temperature strength and corrosion resistance. The composition of the matrix and the isolated phases will be studied in Al-alloys before corrosion and in the corrosion film present after corrosion. These studies will be made both on corrosion-resistant alloys and on alloys showing little resistance to corrosion in an effort to determine corrosion mechanisms and to pin-point the factors contributing to the corrosion resistance of these materials.

It is intended that the high temperature irradiation of high strength beryllium (extruded) be continued. It is proposed also that zone refined beryllium be irradiated to help define the position of helium atom entrapment at intermediate temperatures. The helium may be entrapped in the crystal lattice, in irradiation-produced trapping centers, or in the original void spaces left after sintering. Since the effect of the helium on the stress-strain behavior as a function of temperature will be different in each of these cases it is planned to determine the helium atom distribution.

Finally, it is expected that the work begun earlier on non-uniformly distributed fuels containing lumped burnable poisons will be resumed through computer calculations and fabrication feasibility experiments. Critical facility measurements followed by reactor loadings are the ultimate objectives of such a program but are unlikely for FY 64.

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