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FABRICATION OF HIGH-URANIUM-LOADED U_3O_8-Al DEVELOPMENTAL FUEL PLATES*

G. L. Copeland and M. M. Martin

Metals and Ceramics Division
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37830

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CONTENTS

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ABSTRACT

A common plate-type fuel for research and test reactors is U_3O_8 dispersed in aluminum and clad with an aluminum alloy. There is an impetus to reduce the ^{235}U enrichment from above 90% to below 20% for these fuels to lessen the risk of diversion of the uranium for nonpeaceful uses. Thus, the uranium content of the fuel plates has to be increased to maintain the performance of the reactors. This paper describes work at ORNL to determine the maximal uranium loading for these fuels that can be fabricated with commercially proven materials and techniques and that can be expected to perform satisfactorily in service.

We fabricated developmental fuel plates with cores containing from 60 to 100 wt % U_3O_8 in aluminum encapsulated in 6061 aluminum alloy and evaluated them for aspects of fabricability, nondestructive testing, and expected performance. We recommend 75 wt % U_3O_8-Al (3.1 Mg U/m^3) as the highest loading in the initial irradiation test. This upper limit is based on a qualitative assessment of the mechanical integrity of the core made by using current fabrication techniques and materials. As the oxide loading is increased beyond this point, planar areas and extensive stringers of oxide and voids develop, which leave little strength in the thickness direction. Fuel plates may then blister over these areas as fission gases collect during irradiation.

Current size plates are easily fabricable to the 75 wt % U_3O_8-Al core loading by current fabrication techniques. Dogboning is a potential problem at this loading for some applications; however, this can be easily solved by using tapered compact ends. Current nondestructive radiography and transmission x-ray scanning are applicable to the highly loaded plates. Ultrasonic testing for nonbonds is marginal because of the abrupt change in conductance at the cladding-core interface. Plate thickness can be increased if desired; we fabricated 75 wt % plates with cores up to 1.52 mm (60 mils) thick. We successfully formed a radius of curvature of 84 mm (3.3 in.) in 75 wt % plates with core thicknesses up to 0.89 mm (35 mils). This is a sharper radius than is required for most research reactor elements.

Void contents of the high-uranium-loaded plates agree well with earlier data and should serve to accommodate fission products. Thermal conductivity measurements indicate that opeating temperatures for the cores will be within acceptable limits. Measurements of the energy releases from the thermite reaction show that the higher levels of U_3O_8 do not add a significant chemical reaction hazard to the other considerations of

safe reactor operation. Thus, assuming satisfactory performance in irradiation tests to the required burnup, we anticipate being able to increase the uranium loading in U_3O_8-Al dispersions to the 3.1 Mg U/m^3 level.

INTRODUCTION

A dispersion of uranium oxide (U_3O_8) in aluminum and clad in aluminum alloy is one of three fuel materials commonly used in plate-type research and test reactors. The other two materials in general use are uranium aluminide (UAl_x with $x \approx 3$) dispersed in aluminum and uranium-aluminum alloy, both clad in aluminum alloy. These fuels have generally used uranium enriched to 93% in ^{235}U to obtain high neutron flux and specific power and/or extended fuel life, while maintaining low volume fractions of fueled phases. Recent emphasis on reducing the risk of diversion of enriched uranium for nonpeaceful uses has provided an impetus for reducing the enrichment level of these research and test reactors to less than 20% ^{235}U. The lowered enrichment requires an increase in the total uranium loading to maintain the performance of the reactors. This report presents the results of work at ORNL to determine the maximal uranium loading for the U_3O_8-Al dispersion that can (1) be fabricated by using essentially the commercially proven materials and techniques and (2) be expected to perform satisfactorily in reactor service. A concurrent program for the UAl_x is being conducted at the Idaho National Engineering Laboratory and for higher density fuel compounds (e.g., U_3Si) at the Argonne National Laboratory.

The maximal uranium loading in aluminum-based fuel tested and qualified as reactor fuel is the 55 wt % U (65 wt % U_3O_8—35 wt % Al) dispersions developed at ORNL for the Puerto Rico Nuclear Center (PRNC) Reactor.[1] The PRNC reactor operated satisfactorily with a core loading of the U_3O_8 fuel with the uranium enriched to the 20% level. Thus, this point serves as a base for further increased loading. Even higher loaded test plates of up to 100 wt % U_3O_8 have been fabricated at ORNL for certain experiments, but were not fully evaluated.[2,3]

The technical issues that require evaluation at the higher loadings include those concerning fabricability, nondestructive evaluation, and expected performance in-reactor. The fabricability issues with increased loading are increased core end thickening (dogboning), increased void fraction, potential difficulty in forming (curving the plate is required for most element designs), mechanical integrity of the core during fabrication, and the ability to fabricate plates with thicker cores as a means of increasing total uranium loading. The nondestructive examinations sensitive to the higher loadings are radiographing and x-ray scanning for homogeneity and ultrasonic testing for bonding evaluation. The expected performance in-reactor as the oxide content is increased and the aluminum is decreased depends primarily on the mechanical integrity of the core, the thermal conductivity, and the increased potential for $Al-U_3O_8$ thermite reactions in core meltdown accidents.

FABRICATION PROCEDURES

The fabrication techniques used in this investigation were essentially those used for years in both developmental work and commercial fabrication of aluminum-base dispersion fuel plates.[4-6] The procedures consist of

1. weighing and blending the component powders for each fuel compact,
2. cold pressing at a gage pressure of 414 MPa (30 tsi) to form the green compacts,
3. degassing the compacts at 590°C at an absolute pressure less than 7 Pa (0.05 torr) for 1 h to remove pressing lubricants and absorbed gases,
4. cleaning the frames and cover plates for roll bonding — this consists of caustic etching for 6061-6061 aluminum alloy cladding bonds,
5. assembling the degassed compacts into frames and welding on the cover plates to form the rolling billet,
6. bonding the cladding to the frame and compact by hot rolling at 490°C (914°F) to a reduction in thickness of 85%,
7. annealing at 490°C for 1 h to soften and also to test for blistering,
8. cold rolling to a reduction in thickness of 10% (total reduction in thickness of 86.5%),

9. heat treating to the "O" temper for 6061 alloy, and
10. finishing operations — core location, plate shearing, machining, cleaning, and inspections for dimensional requirements, fuel homogeneity, and ultrasonic nonbond indications.

Three sizes of plates with several different core thicknesses were fabricated during this work. In terms of widths and lengths the miniplates were either 31.8 by 73.1 mm (1.25 × 2.88 in.) with 28 by 69-mm (1.1 × 2.7-in.) core or 50.8 by 114.3 mm (2.00 × 4.50 in.) with 45 by 99-mm (1.8 × 3.9-in.) core. Full-sized test reactor plates 71.1 by 624.8 mm (2.80 × 24.60 in.) with 61 by 597-mm (2.4 × 23.5-in.) core were fabricated for the forming demonstration.

NONDESTRUCTIVE TESTING

Radiography was used to examine the plates for core configuration, core and edge border dimensions, high-density inclusions, fuel particles in the edge and end borders, and qualitative assessment of fuel homogeneity. Radiographs suitable for these purposes were obtained with no difficulty at loadings up to 100 wt % U_3O_8. Two radiographs of different intensities are required: one suitable for examining the edge borders and one suitable for examining the core.

Transmission x-ray scanning was used to evaluate the homogeneity of the fueled region of the plates. This technique enables quantitative measurement of the fuel loading over the entire plate, including the dogbone region. With suitable calibration standards, good results were obtained up to the highest areal uranium loading examined, which was 3.2 kg/m^2 for a 75 wt % U_3O_8-Al core of 0.89 mm (35 mils) thickness.

Ultrasonic testing was used to examine the plates for unbonded regions. The drastic change in conductance at the cladding-core interface makes this test difficult for the highly loaded plates. However, ultrasonic testing did prove to be a screening technique that was used to select the better plates from several otherwise identical plates. Several plates with known blisters were tested, and the blistered areas showed in the trace in each instance.

DOGBONING

Dogboning is the core thickening that occurs at the ends of the core during roll bonding. This core thickening is of concern for two reasons. First is the thinning of the cladding over the dogbone. Secondly, the higher fuel concentration could result in excessive temperature during irradiation. The temperature concern is more of a problem in irradiation tests where the end of the test plate may be in the high-flux region of the reactor core. In full-sized plates, the core ends are usually in the lower flux regions of the core.

For fuel loadings in use up to now, dogboning has not been a problem for U_3O_8-Al dispersions. However, dogboning increases with U_3O_8 content and could potentially limit the loading for some applications. The dogboning produced by typical fabrication sequences for several core thicknesses, cladding thicknesses, and fuel loadings is shown in Table 1. Dogboning does increase with U_3O_8 content and decrease with increasing core thickness. For core thicknesses of 0.51 mm (20 mils), U_3O_8 contents up to 75 wt % should always exhibit dogboning less than 40%.

If it is desired to decrease the dogboning, several approaches may be taken. The most effective approach is probably to taper the ends of the fuel compact as it is pressed. This technique has been proven to reduce dogboning effectively with UAl_x-Al dispersions.[7] To determine the effectiveness of the tapered cores in U_3O_8-Al dispersions, we fabricated 75 wt % U_3O_8-Al plates from compacts with a double taper of 0.2 and 1.0 rad (11° and 55°) over 7.6 mm (0.30 in.) as used in the manufacture of the Advanced Test Reactor plates[7] and a single taper of 0.3 rad (18°) over 4.6 mm (0.18 in.). Both types of tapered compacts essentially eliminated dogboning for the 75 wt % U_3O_8-Al plates. The rolled core diminished smoothly from full thickness to about 0.1 mm (4 mils) over a length of about 75 mm (2.9 in.) in the finished plate. Thus, if desired, dogboning could be essentially eliminated for U_3O_8 dispersions up to 75 wt % by use of tapered compacts. For many applications, the level of dogboning obtained by using untapered compacts will not be detrimental to the performance of the fuel plate.

Table 1. Maximal Dogboning and Minimal Cladding Measurements for Miniature U_3O_8-Al Dispersion Fuel Plates

U_3O_8 Content wt %	Thickness, mm			Maximal Dogboning (Core Thickening) Observed, %		Minimal Cladding Thickness Observed (mm)
	Plate	Cladding	Core	by X-ray[a]	by Metallography[b]	
60.0	1.52	0.38	0.76	10	10	0.30
65.0	1.27	0.38	0.51	27	46	0.25
65.0	1.52	0.44	0.64	15	2	0.38
65.0	1.27	0.25	0.76	23	24	0.15
65.0	1.52	0.38	0.76	22	23	0.30
65.0	1.52	0.32	0.89	9	3	0.25
70.0	1.52	0.32	0.89	14	14	0.23
70.0	1.52	0.38	0.76	27	27	0.28
70.0	1.52	0.44	0.64	16	15	0.30
70.0	1.27	0.38	0.51	24	38	0.28
75.0	1.52	0.38	0.76	24	17	0.28
75.0	1.52	0.44	0.64	24	19	0.30
75.0	1.27	0.38	0.51	26	48	0.25
75.0	1.52	0.32	0.89	18	21	0.20
77.5	1.27	0.38	0.51	39	46	0.25
80.0	1.27	0.38	0.51	38	35	0.30
82.5	1.27	0.38	0.51	38	66	0.25
82.5	1.27	0.25	0.76	12	17	0.18
82.5	1.52	0.38	0.76	28	40	0.15
100.0	1.27	0.38	0.51	75	110	0.00
100.0	1.27	0.25	0.76	16	30	0.10
100.0	1.52	0.38	0.76	29	50	0.13

[a] Maximum observed within five plates for transmission x-ray scanning data as correlated with equivalent aluminum step-wedge thickness.

[b] The maximum of a set of five plates (as determined by x-ray scanning) was selected for metallographic examination. Dogbone is determined by measuring the minimal cladding on both sides and subtracting from plate thickness to determine core thickness. The percent dogbone is relative to the nominal core thickness.

MECHANICAL INTEGRITY

The mechanical integrity of clad cores with increasing U_3O_8 content was judged qualitatively by the appearance of the microstructure. Figure 1 shows two polished cross sections of a clad dispersion of 75 wt % U_3O_8-Al. The aluminum matrix is on the verge of becoming the discontinuous phase. At higher volume fractions of voids plus U_3O_8, large planar areas with little or no tensile strength in the thickness direction occurred. These areas could accumulate fission gas during irradiation and develop a blister. Thus, although the exact loading that could lead to failure in-reactor is not clear, we chose to limit the loading to 75 wt % U_3O_8 for the first irradiation experiment. This concentration yields a uranium density in the fabricated core of 3.1 Mg/m^3.

The cause of the planar areas of voids and U_3O_8 is the particle breakup and stringering that occurs during rolling. Previous work[8] showed qualitatively that increasing the particle strength or changing the morphology could delay the onset of particle breakup and thus minimize stringering. Such changes in existing and proven materials are outside the scope of this investigation.

FABRICATION VOIDS

Voids formed during the rolling of dispersion fuel plates play an important part in the irradiation performance of the plates by allowing room for solid swelling and fission-gas accumulation. Previous work[2,9] has investigated the influence of several parameters on void volume and has shown that oxide concentration has the strongest effect. The void volumes obtained in this study are shown in Fig. 2 and agree well with those reported earlier. The plates with thicker cores have slightly lower void volumes. The void fraction must be considered when core loadings are calculated and when fabricability of the plates is considered. Total oxide plus void volume fraction appears to be the controlling factor in fabricability.

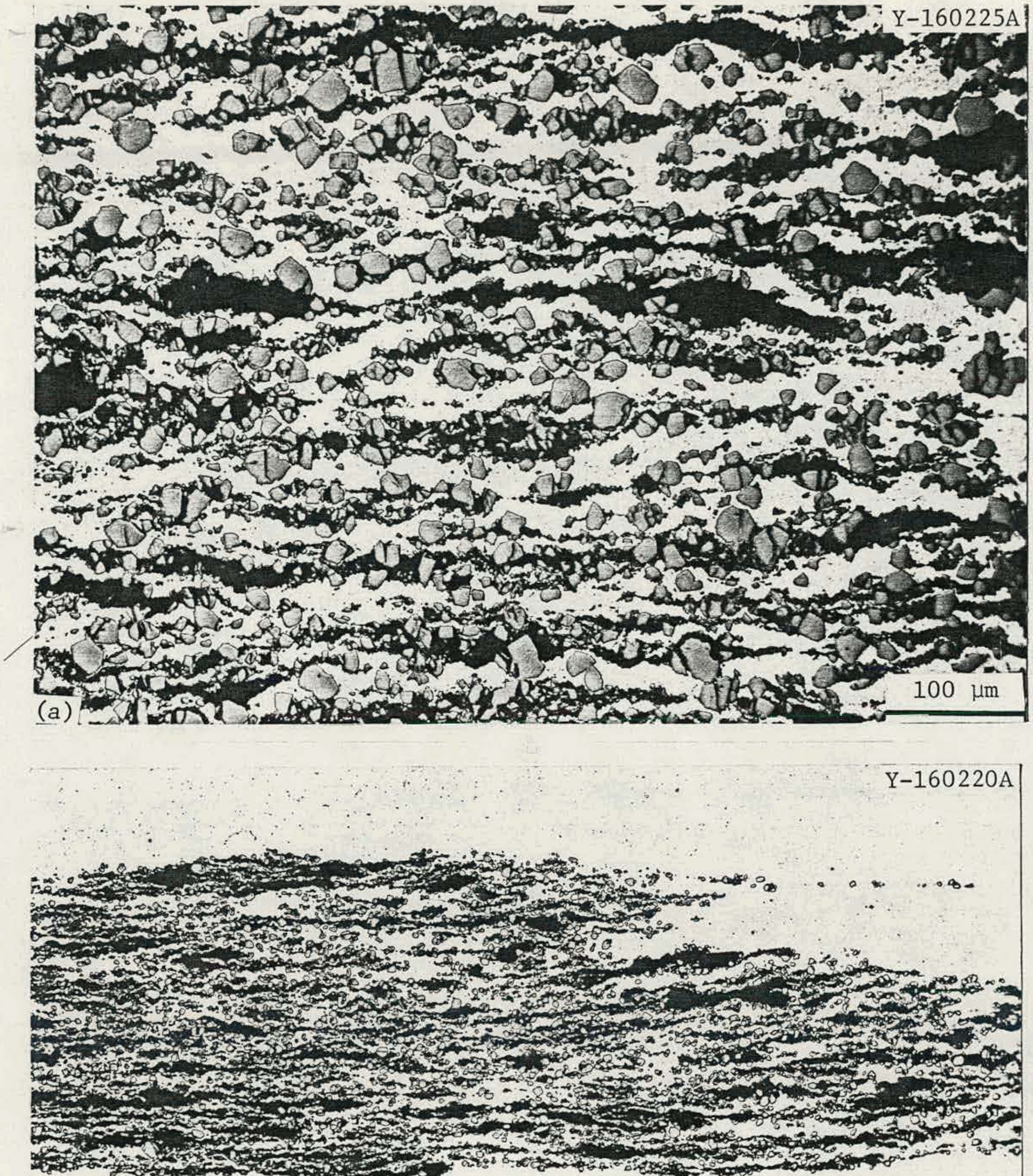

Fig. 1. Microstructure of 75 wt % U_3O_8 in Aluminum Reveals that Aluminum is on the Verge of Becoming Discontinuous. (a) Transverse. (b) Longitudinal.

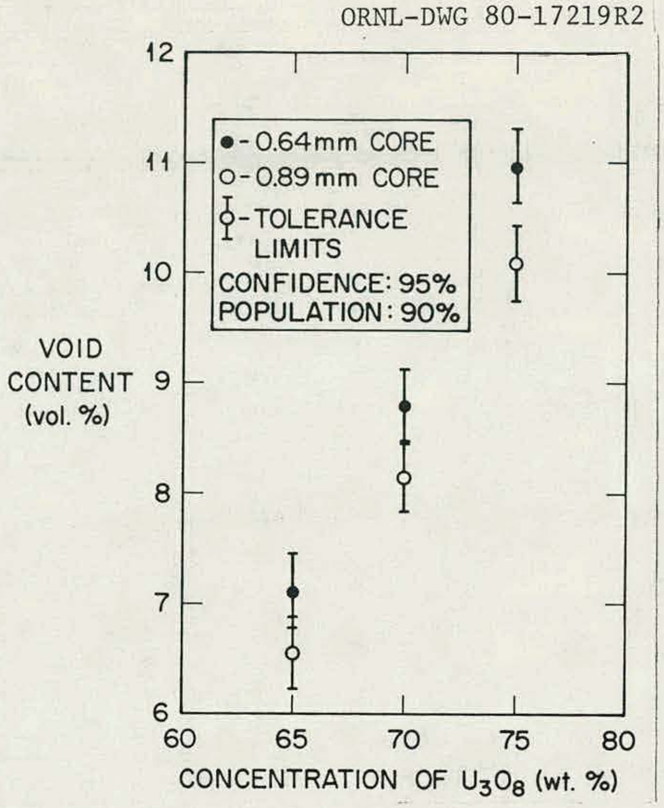

Fig. 2. The Void Content of U_3O_8-Al Cores Depends on the Concentration of U_3O_8 and to a Lesser Extent on the Core Thickness.

FORMING

Many fuel element designs for research and test reactors require that the plates be curved to a radius of curvature of about 140 mm (5.5 in.). The ability to form the plates to this curvature without core cracking is of concern as the loading or the core thickness increases. To determine if this forming is a problem, we fabricated full-sized plates that contained 75 wt % U_3O_8-Al (the highest concentration proposed for the first irradiation test) and core thicknesses of 0.51, 0.76, and 0.89 mm (20, 30, and 35 mils). These plates were rolled, fully annealed, and then successively formed to smaller radii of curvature with interim visual and radiographic examinations for core cracks. No cracking was observed after forming to the smallest available radius die. This die produced a radius of 84 mm (3.3 in.) in the plates. Subsequent metallographic examination of the plates confirmed the absence of cracks. Figure 3 shows the cross sections of the three plates. We do not anticipate any problems in forming within the limits investigated.

THICKER CORES

A potential method of increasing the uranium loading is to increase the core thickness of the plates. To determine the feasibility of this approach we fabricated miniature fuel plates containing 75 wt % U_3O_8-Al dispersions with a core thickness of 1.52 mm (60 mils) and overall plate thickness of 2.29 mm (90 mils). No problems were encountered in the fabrication of these plates. The dogboning was less than in corresponding thinner cores — both in absolute magnitude and percentage. These plates exhibited only 4% core thickening as measured metallographically. No attempts were made to form a curvature in the plates since they were not full width. Plates of this thickness may possess sufficient stiffness so that forming will not be necessary.

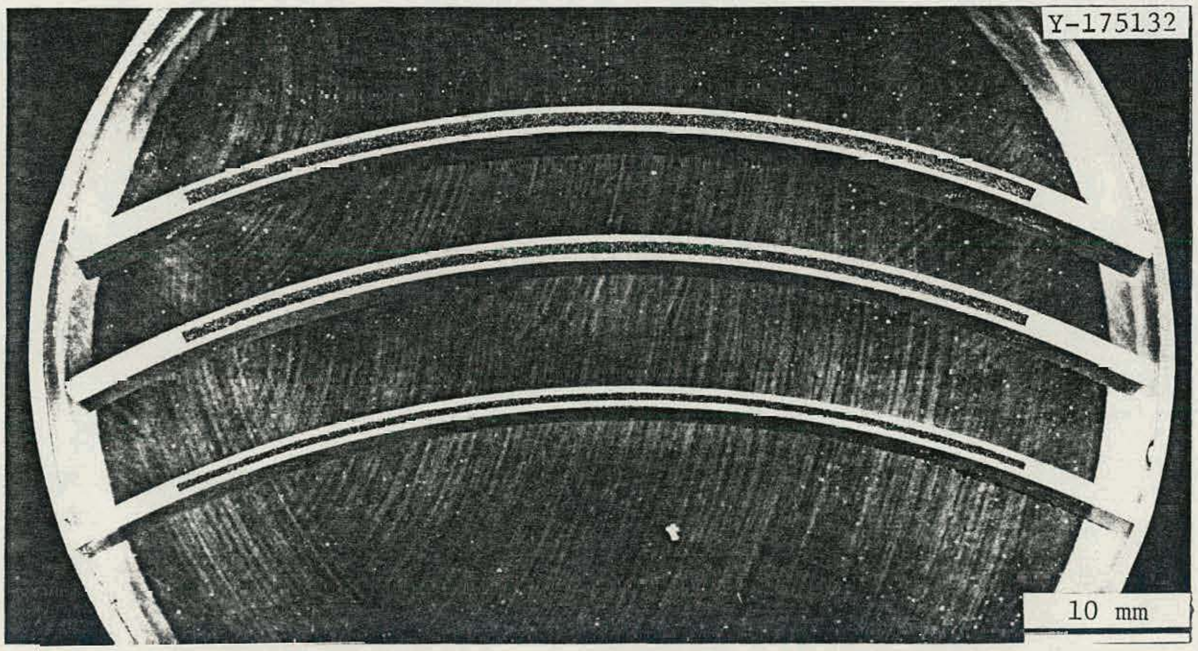

Fig. 3. Cross Sections of 75 wt % U_3O_8-Al Cored Plates Reveal no Cracking After Being Formed to a Radius of 84 mm. Core thicknesses are 0.89, 0.76, and 0.51 mm from top to bottom.

THERMAL CONDUCTIVITY

Thermal conductivity is important in determining the operating temperature of the fuel core and depends strongly on oxide concentration. The thermal conductivity in the thickness direction was measured for miniature fuel plates that contained from 60 to 82.5 wt % U_3O_8-Al.[10] Disk samples 6.35 mm (0.25 in.) in diameter were cut from areas of the fuel plates that had been selected for uniformity by eddy-current measurements and radiography. The thermal conductivity measurements were made in a comparative heat flow apparatus, which has been described previously.[11] The thermal conductivity of the fuel core correlates well with the total oxide plus void volume fraction in the core, as can be seen from the data presented in Fig. 4. The thermal conductivity values obtained indicate that centerline operating temperatures will not limit the fuel loading for most research reactor applications.

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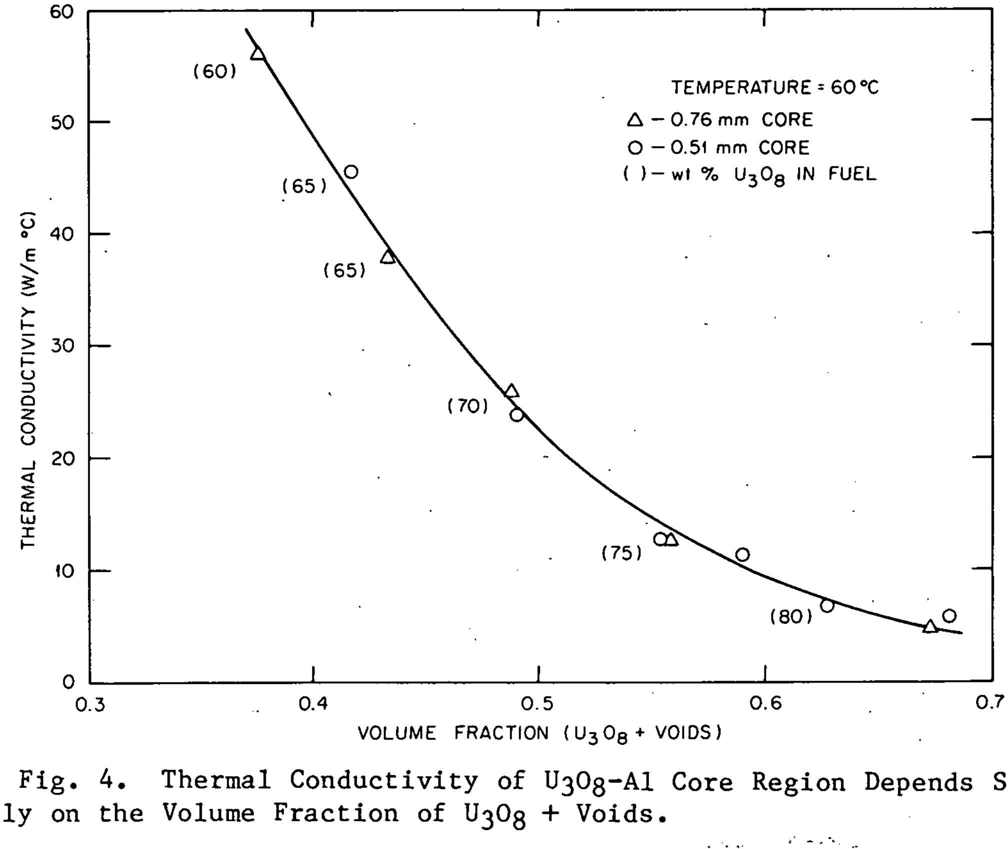

Fig. 4. Thermal Conductivity of U_3O_8-Al Core Region Depends Significantly on the Volume Fraction of U_3O_8 + Voids.

THERMITE REACTION

The phase assemblage Al-U_3O_8 is a nonequilibrium mixture and tends to react with heat evolution, a process known as a thermite reaction. However, this reaction has been of little concern since at fabrication temperatures up to 500°C (932°F) very little reaction occurs. Operating temperatures of the fuel plates in-reactor are much less than this, often less than 100°C (212°F), so that the potential thermite reaction was inconsequential except in the event of a loss-of-coolant accident involving fuel meltdown. In view of the possibility of a meltdown, studies were undertaken of the ignition temperature and magnitude of heat evolution for use in safety analyses.[12] These early studies showed that the peak energy release occurs in the 55 to 75 wt % U_3O_8 range, which is the region of interest for the current developmental high-uranium-loaded fuel. Therefore, we repeated the investigation of ignition temperature and maximal heat release for the compositions of interest using differential thermal analysis and heating of miniature fuel plates.[13] The observed ignition temperature of around 900°C (1650°F) agrees well with previous studies. The energy releases, however, were much lower than predicted and in general are low compared with the energy required to initiate them. The heating of miniature fuel plates to as high as 1400°C (2550°F) showed no violent thermal effects, explosions, or observable gas release. We conclude from these tests that increasing the U_3O_8 loading does not add a significant chemical reaction hazard to the other considerations of safe reactor operation.

CONCLUSIONS

We have recommended 75 wt % U_3O_8-Al (3.1 Mg U/m^3) as the highest loading to be tested in the initial irradiation test. This limit is based on a qualitative assessment of the loss of mechanical integrity as the aluminum matrix becomes discontinuous at higher loadings. We offer the following conclusions and comments with regard to fabricability, testing, and use of 75 wt % U_3O_8-Al cored fuel plates:

1. Fabrication is easily accomplished with current technology and materials.

2. Dogboning is a potential problem for some applications; however, this problem can be easily solved by using tapered compact ends.

3. Current radiography and x-ray scanning techniques can be used.

4. Ultrasonic testing for nonbonds is marginal because of the abrupt change in sound conductance at the cladding-core interface.

5. Core thicknesses up to 1.52 mm (60 mils) can be fabricated with good results.

6. Full-sized plates with core thicknesses up to 0.89 mm (35 mils) can be successfully formed to a radius of curvature of 84 mm (3.3 in.); this is a sharper radius than is required for most research reactor elements.

7. Void contents agree well with earlier data and should accommodate fission products.

8. Thermal conductivity measurements indicate that operating temperatures will be within acceptable limits.

9. Measurements of the energy releases from the thermite reaction show that the higher U_3O_8 loading does not add a significant chemical reaction hazard to other considerations of safe reactor operation.

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REFERENCES

1. A. E. Richt, C. F. Leitten, Jr., and R. J. Beaver, "Radiation Performance and Induced Transformations in Aluminum-Base Fuels," pp. 469–88 in *Research Reactor Fuel Element Conference, September 17–19, 1962, Gatlinburg, Tennessee*, TID-7642, Book 2 (1963).
2. M. M. Martin, "Effect of Dispersoid Concentration on Void Content of Composite Plates," *Fuels and Materials Program Quart. Prog. Rep. Dec. 31, 1969*, ORNL-4520, pp. 282–86.
3. *Ibid. March 31, 1970*, ORNL-4560, pp. 225–30.
4. M. M. Martin, W. J. Werner, and C. F. Leitten, Jr., *Fabrication of Aluminum-Base Irradiation Test Plates*, ORNL-TM-1377 (February 1966).
5. M. M. Martin, A. E. Richt, and W. R. Martin, *Irradiation Behavior of Aluminum-Base Fuel Dispersions*, ORNL-4856 (May 1973).
6. R. W. Knight, J. Binns, and G. M. Adamson, Jr., *Fabrication Procedures for Manufacturing High Flux Isotope Reactor Fuel Elements*, ORNL-4242 (June 1968).
7. G. W. Gibson, EG&G, Idaho Falls, Idaho, personal communication. February 1979.
8. D. O. Hobson and C. F. Leitten, Jr., *Characterization of U_3O_8 Dispersions in Aluminum*, ORNL-TM-1692 (February 1967).
9. M. M. Martin and W. R. Martin, *Fabrication Voids in Aluminum-Base Fuel Dispersions*, ORNL-4611 (October 1970).
10. R. K. Williams and R. S. Graves, Oak Ridge National Laboratory internal correspondence, January 1980.
11. R. K. Williams, R. S. Graves, and J. P. Moore, *A Study of the Effects of Several Variables on the Thermal Conductivity of 2 1/2 Cr-1 Mo Steel*, ORNL-5313 (April 1978).
12. J. D. Fleming and J. W. Johnson, "Exothermic Reactions in $Al-U_3O_8$ Composites," pp. 649–66 in *Research Reactor Fuel Element Conference, September 17–19, 1962, Gatlinburg, Tennessee*, TID-7642, Book 2 (1963).
13. A. E. Pasto, G. L. Copeland, and M. M. Martin, *A Quantitative Differential Thermal Analysis Study of the U_3O_8-Al Thermite Reaction*, ORNL-5659 (June 1980).