HIGH-URANIUM-LOADED U_3O_8-Al FUEL ELEMENT DEVELOPMENT PROGRAM

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ABSTRACT

The High-Uranium-Loaded U_3O_8-Al Fuel Element Development Program supports Argonne National Laboratory efforts to develop high-uranium-density research and test reactor fuel to accommodate use of low-uranium enrichment. The goal is to fuel most research and test reactors with uranium of less than 20% enrichment for the purpose of lowering the potential for diversion of highly-enriched material for nonpeaceful usages.

The specific objective of the program is to develop the technological and engineering data base for U_3O_8-Al plate-type fuel elements of maximal uranium content to the point of vendor qualification for full scale fabrication on a production basis. A program and management plan that details the organization, supporting objectives, schedule, and budget is in place and preparation for fuel and irradiation studies is under way. The current programming envisions a program of about four years duration for an estimated cost of about two million dollars.

During the decades of the fifties and sixties, developments at Oak Ridge National Laboratory led to the use of U_3O_8-Al plate-type fuel elements in the High Flux Isotope Reactor, Oak Ridge Research Reactor, Puerto Rico Nuclear Center Reactor, and the High Flux Beam Reactor. Most of the developmental information however applies only up to a uranium concentration of about 55 wt % (about 35 vol % U_3O_8). The technical issues that must be addressed to further increase the uranium loading beyond 55 wt % U involve plate fabrication phenomena of voids and dogboning, fuel behavior under long irradiation, and potential for the thermite reaction between U_3O_8 and aluminum.

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INTRODUCTION

Dispersions of uranium oxide (U_3O_8) in aluminum, and clad in aluminum, are used in the High Flux Isotope Reactor (HFIR) fuel at Oak Ridge National Laboratory. Since 1966 and through October 1978, 168 fuel elements (90,720 fuel plates) have been used successfully. Maximal fuel loading is about 35 wt % U (about 17 vol % U_3O_8) of 93% enrichment in the uranium-bearing-portion (meat) of a HFIR fuel plates; thickness of meat within a HFIR fuel plate varies from about 250 to 760 μm (about 0.010 to 0.030 in.). Also, Puerto Rico Nuclear Center Reactor (PRNC), which has been shut down, used a U_3O_8-Al dispersion, plate-type fuel elements developed by Oak Ridge National Laboratory. The fuel loading in PRNC was 55 wt % U (35 vol % U_3O_8) of 20% enrichment in the meat, which was about 630 μm (about 0.025 in.) in thickness. Presently, the Oak Ridge Research Reactor (ORR) and the High Flux Beam Reactor are also using U_3O_8-Al dispersion-type fuel elements.

As part of the U_3O_8 dispersion fuel development for HFIR and PRNC, test samples were made, irradiated, evaluated, and deemed satisfactory.[1,2] Thus, 55 wt % U represents the maximal fuel loading of qualified U_3O_8 dispersion-type aluminum plate fuel.

The potential for development of higher uranium loadings for U_3O_8 dispersion-type aluminum plate fuel does exist. For example, again as part of the U_3O_8 dispersion fuel development for HFIR, test samples were made that contained up to 74 vol % U_3O_8 in the meat.[3,4] As expected, for uranium loadings greater than about 60 wt % U (about 40 vol % U_3O_8), depending on void content, the continuous phase of the dispersions changed from aluminum to U_3O_8. However, all cold-pressed U_3O_8-Al dispersions exhibited sufficient green strength after compacting to permit some handling without breaking. All also appeared to roll-clad satisfactorily, as was evident from the absence of internal cracks.

This paper summarizes the program and management plan and pertinent technical issues of the High-Uranium-Loaded U_3O_8-Al Fuel Element Development (HFED) program at ORNL. The technical issues that must be addressed to further increase the uranium loading beyond 55 wt % in U_3O_8 dispersion-type aluminum plate fuel involve plate fabrication (i.e., such as adequate control of cladding thickness, dogboning, a continuous aluminum phase in the meat, and uranium homogeneity), fuel behavior under long irradiation (i.e., unacceptable swelling and/or shrinking), and the potential for the thermite reaction between U_3O_8 and aluminum.

OBJECTIVE

The specific objective of this program is to develop the technological and engineering data base that is needed to permit fabrication and qualification of U_3O_8-Al plate-type fuel element of maximal uranium content for research and test reactors.

The supporting objectives for this program are as follows:

1. To conduct a screening study to determine the effects of high-uranium-loaded U_3O_8-Al dispersions on the fabricability of miniature-type, irradiation-test fuel plates;

2. to utilize and extend the above screening study to full-sized ORR reactor-type fuel plates, and to assemble full-sized ORR-type fuel elements;
3. To conduct irradiation tests to validate the performance of the product produced by the foregoing technological and engineering development; and
4. to provide for joint industry-government cooperation, and to provide technical data to industry.

RELATIONSHIP OF PROGRAM TO NATIONAL PROGRAM

The program is in direct support of Argonne National Laboratory (ANL) efforts to develop high-uranium-density research and test reactor fuel to accomodate use of low-uranium enrichment. The use of low-uranium enrichment (less than 20% enrichment) in place of highly-enriched fuel for these reactors would give the potential benefit of reduced possibility of ^{235}U diversion. Argonne National Laboratory coordinates and manages the national program for the Department of Energy (DOE). Argonne National Laboratory through DOE field offices at Chicago, Illinois, and Oak Ridge, Tennessee, has delegated the responsibility for conducting the High-Uranium-Loaded U_3O_8-Al Fuel Element Development Program to the Metals and Ceramics Division at ORNL.

ORGANIZATION

The program organization chart, present in Fig. 1, shows the first level of program work breakdown structure and the program management hierarchy. Key personnel are M. M. Martin, Program Coordinator; Fuel Fabrication Task Leader, G. L. Copeland; and General Support (Fuel Irradiation Testing and Examination) Task Leader, E. L. Long, Jr.

The purpose of the fuel fabrication subtask is to develop the fabrication technology for U_3O_8-Al plate-type fuel elements of maximal uranium content to the point of vendor qualification for full-scale fabrication on a production basis. The work includes the development of fabrication procedures, material specifications, and the fabrication of irradiation-test samples and irradiation-test full-size elements.

The purpose of the general support subtask is to verify the dimensional stability and metallurgical integrity of the U_3O_8-Al plate-type fuel elements produced in the fuel fabrication subtask. The fuel irradiation testing and evaluation work includes design and construction of an apparatus in the ORR reactor for irradiating miniature U_3O_8-Al bearing fuel plates and all postirradiation examination. It requires irradiation of miniature fuel plates and also the irradiation of a full-size element in the ORR reactor. During the conceptual design phase, full consideration will be given to incorporation of the irradiation testing and evaluation of other developmental aluminum-base plate-type fuels of the national program.

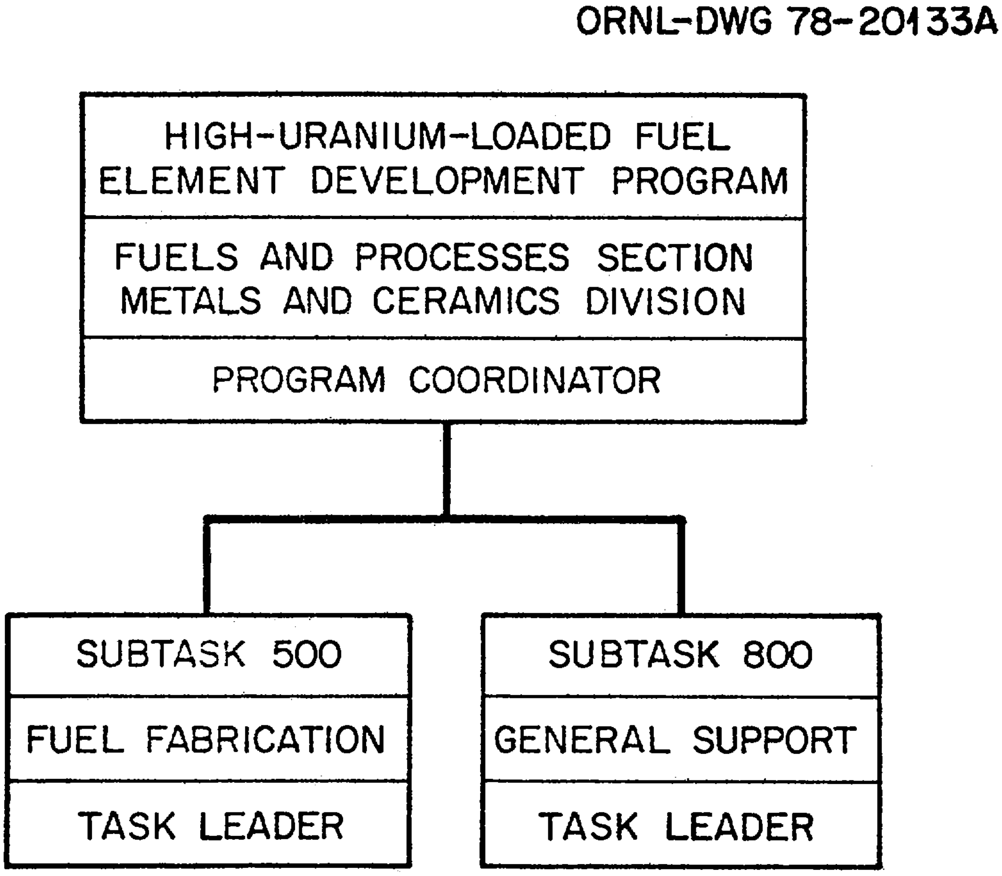

Fig. 1. Technical Organization Chart of the Program.

SCHEDULE

We anticipate that a program of about four years duration will be pursued. Continuation of the studies, however, will be based on ANL annual selection of the fuel candidates suitable for further study. Therefore, the program for development of the high-uranium content U_3O_8-Al plate-type fuel elements has been broken down into four approximately one-year phases as follows: Phase I - Inital screening-fabrication and conceptual-irradiation-design studies; Phase II - Continuation of any remaining fabrication and metallurgical studies and preparation for irradiation testing; Phase III - Prototype fuel element fabrication and irradiation testing; and Phase IV - Development of procedures, specifications and aid to fuel fabricators.

BUDGET

The completion of this program will involve the use of both operating and capital equipment funds. Operating funds shown graphically in Fig. 2, reach maximal commitment in FY 1980. Capital equipment items for FY 1979 include powder compacting press, vacuum annealing furnace, fluoroscope, and a channel-spacing metrology device for interim examination of miniature-fuel-plate assembly in the pool of the ORR.

GENERAL FUEL PLATE FABRICATION PROCEDURES

Plate type fuel elements of U_3O_8 dispersed in aluminum are fabricated by powder metallurgy techniques in which fuel and aluminum powders are blended, compacted, and then encased in an aluminum envelope. Metallurgical bonding and the primary sizing operations are accomplished by hot rolling the assembled envelope which is sometimes termed a "picture-frame" billet.

The general processing steps used are as follows:

1. weighing and blending the U_3O_8 powder (typically the principal particle size is 44 to 88 µm with about 25% <44 µm) with the aluminum powder (principal particle size is 0 to 44 µm);
2. forming compacts by cold pressing at 410 MPa (30 ton/in^2);
3. degassing the compacts at 500°C for 1 hr in vacuum to remove pressing lubricants and moisture;
4. cleaning the frames and cover plates by chemical etching (this procedure allows direct bonding of 6061 aluminum alloy to itself and elminates the need for alclad frames and cover plates);
5. assembling the degassed compacts, frame, and cover plates to form a billet which is then welded partially around the perimeter;
6. hot rolling the billet to 84% reduction at 490°C;
7. annealing at 490°C for 1 hr;
8. cold rolling to 20% reduction;
9. if desired, annealing to the "0" temper for alloy 6061; and
10. finishing the plates by determining core location, shearing, forming if required, and inspecting.

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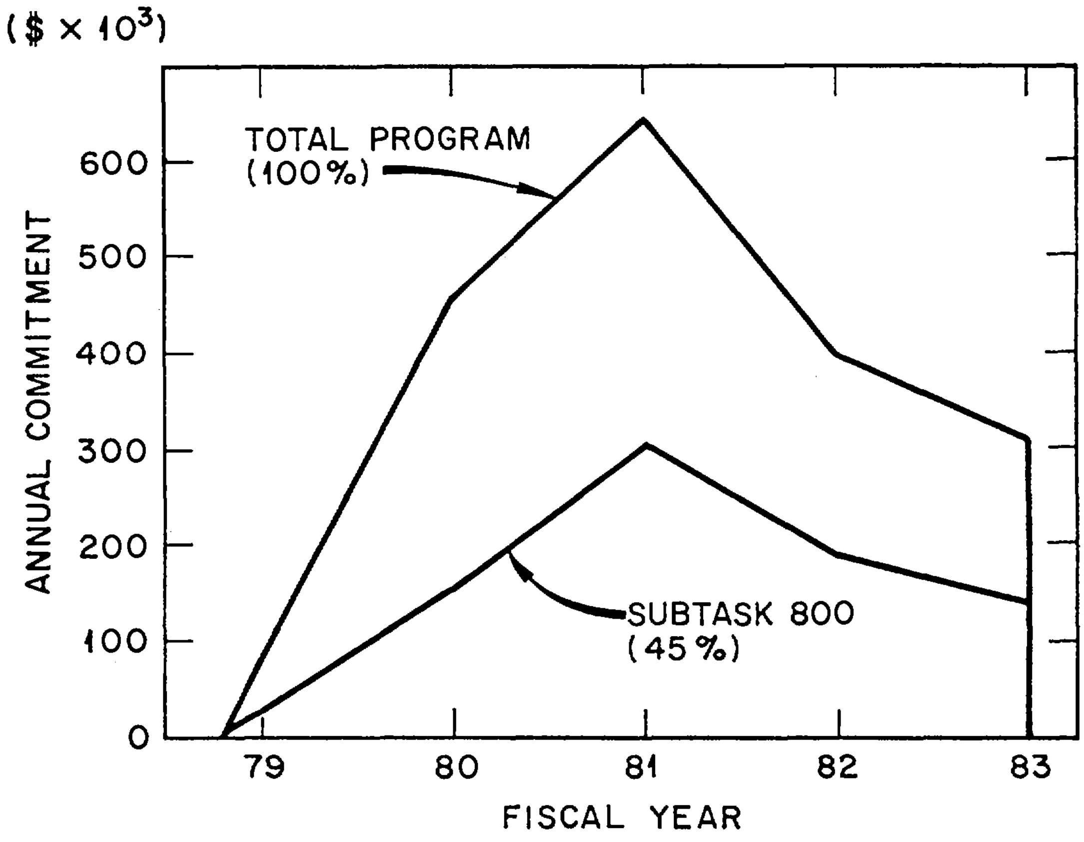

Fig. 2. Estimate of Operating Funds Required for Completion of Program.

TECHNICAL ISSUES

The technical issues of fabrication voids, dogboning, and thermite reaction are discussed below.

1. Fabrication Voids

Fabrication Voids explain variations in irradiation performance of many low-uranium-content fuel dispersions for nuclear reactors. For example, core swellings predicted from a rate of 6.3 vol % per 10^{21} fissions/cm^3 and from the assumption that the initial void volume is 100% effective in accommodating this growth agreed relatively well with the measured values.[1] The technical issue is the effectiveness and stability of fabrication voids that can be introduced into the fuel dispersions of high-uranium content.

The void content of roll-clad aluminum-base dispersions depends on (1) the type and concentration of the fuel compound, (2) the aluminum cladding alloy, and (3) the amount of cold-rolling deformation.[5] For a particular material combination, the first roll-bonding reduction of as little as 15% in thickness establishes a constant void concentration for all subsequent hot-rolling passes. The final void content of the completed fuel plate shows only a secondary dependency on hot deformation and heat treatment. Fig. 3 presents the void content of experimental fuel plates as a function of U_3O_8 concentration in both the hot and cold rolled conditions.[4] Clearly, increasing U_3O_8 concentration and cold reduction for roll-clad U_3O_8-bearing plates increases the fabrication void content. The in-reactor performance of plates with greater than about 7 vol % fabrication voids remains to be demonstrated.

2. Dogboning

Dogboning in fuel plates manufactured by roll bonding and using the picture-frame technique, is a problem that has been with us for years. And, it tends to be more severe with higher fuel loadings. The relative strength of core to frame and cover plates, ratio of thickness of cover plates to core, and rolling conditions such as temperature, reduction per pass, and number of passes, all affect the degree of dogboning. Fig. 4 presents the compressive strength of various materials to aid in showing the effect of relative strength of dogboning.[6] For example, identically fabricated fuel plates which contained either U_3O_8 or UAl_x ($x \sim 3$) dispersoids at concentrations up to 20 vol %, exhibit a similar degree (about 20% core thickening) of dogboning because the compressive yield strengths are similar. Above 30 vol %, the dogboning in the UAl_x-bearing plates is more severe because UAl_x cores are much stronger than those containing U_3O_8.

3. Thermite Reaction

A safety concern in the unlikely event of a U_3O_8-bearing reactor core meltdown accident involves the heat contribution of an exothermic reaction between the U_3O_8 and aluminum. An exothermic reaction involving U_3O_8 and Al to produce UO_2, U-Al compounds and Al_2O_3 is thermodynamically possible. This reaction has been studied by a number of investigators.[7] It was found that no significant reaction occurs at temperatures below about 930°C. Energy releases as high as 1.7 kJ/g of U_3O_8 occurred when the U_3O_8 loading was on the

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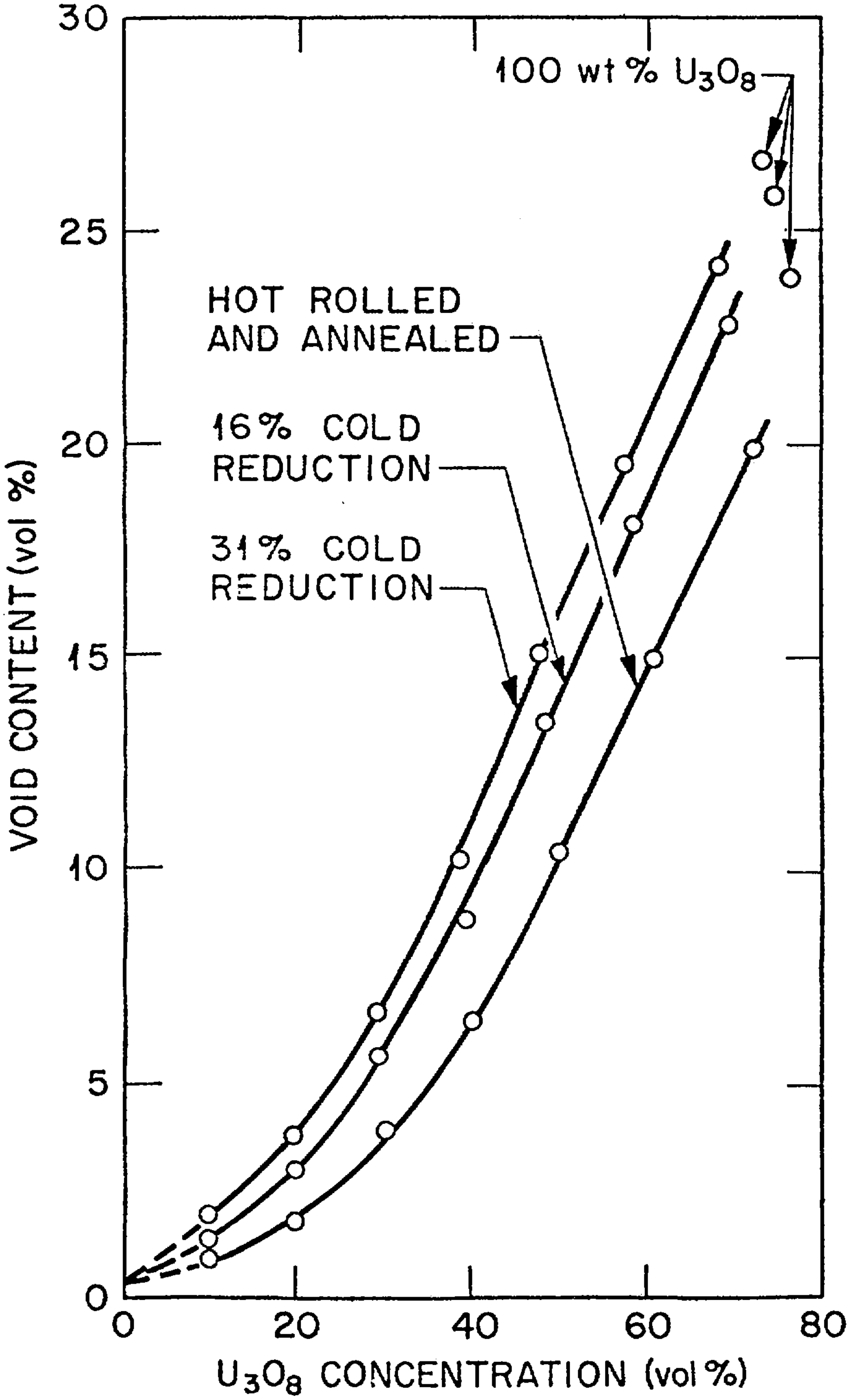

Fig. 3. Effect of U_3O_8 Concentration on the Void Content of High-Fired U_3O_8-Al Dispersion.

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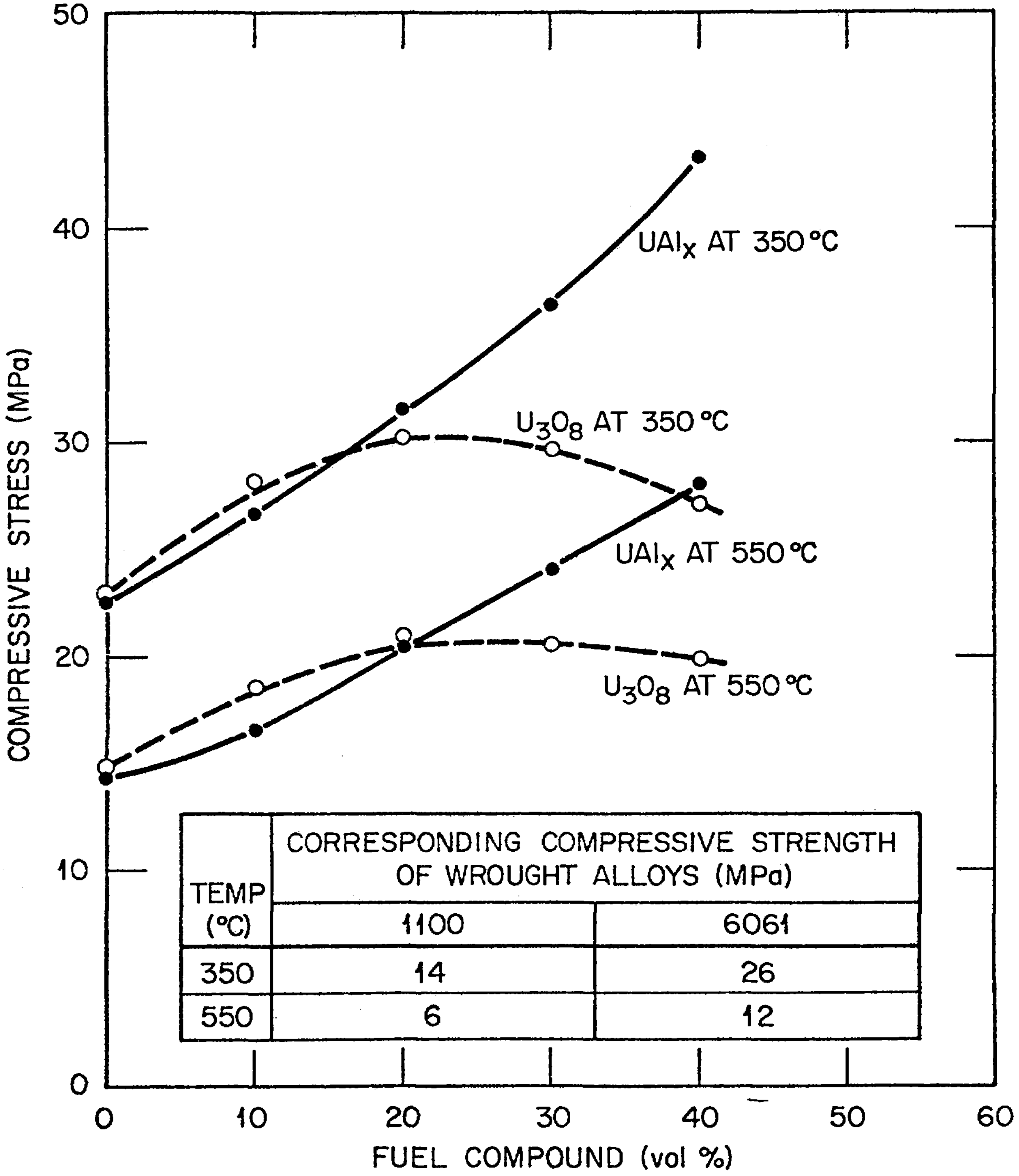

TEMP (°C)	CORRESPONDING COMPRESSIVE STRENGTH OF WROUGHT ALLOYS (MPa)	
	1100	6061
350	14	26
550	6	12

Fig. 4. Eleveated Temperature Compressive Strength of Fuel Dispersion Required to Produce 5% Strain.

order of 33 to 45 vol %. At 13 vol % the self heating from the reaction was found to be negligible. An ANL experiment[8] in the Transient Reactor Test (TREAT) facility using 17 vol % U_3O_8 plates exposed to reactor accident conditions shows that the exothermic reaction was not an important energy contribution. To provide additional data for the higher loaded plates, we plan to perform a series of experiments using plates from our development program to determine the exothermic reaction energy release as a function of U_3O_8 loading. We will also fabricate plates for samples if needed by ANL in TREAT experiments to simulate reactor accident conditions.

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DISCUSSION

FERADAY (Chalk River): In the kind of reactors that will be using this U_3O_8 fuel, have you done any studies of what happens on a loss of coolant, and on what fuel temperature you expect to reach? There is a possibility that thermite reaction could occur under certain loss-of-coolant conditions. Is that right?

MARTIN: I have not done those types of studies, but I would assume that if you get the temperature to about 900-950°C, you have a thermite reaction. I would assume that. I don't know what would happen after long burnup. I would think the thermite propensity would be a lot less.

LEWIS (U.S. Dept. of State): You've got a certain amount of volume in the fuel meat of a plate. Let's say that you're going to stick with an existing fuel plate design. You have a given amount of fuel meat volume to work with. That fuel volume can go into U_3O_8, aluminum and void. What I'm looking for is to increase the average uranium density in that fuel meat volume in order to permit me to go to lower enrichments. What I'm saying is if as you increase the U_3O_8 density weight percent in there, if you get extremely high void fractions, you may be really losing out in the average uranium density. What I'm asking is whether any trade-off studies have been done. For example, does it pay to go to 75 wt % because of the very high wt fractions you get.

TRAVELLI(ANL): Maybe I can put the question in different words. As you increase the volume fraction of the U_3O_8, is it possible that the wt fraction may not increase but actually decrease?

MARTIN: No. Not according to the last curve that I put on the board for you.

TRAVELLI: That is the point that I would like to put in words. because I think that Martin showed that curve and it looked like the weight fraction was not increasing as fast as the volume fraction. Because of the appearance of the voids, the voids were increasing as it was approaching the 100% U_3O_8.

MARTIN: Voids increase as the weight fraction increases. We can reduce these voids by using a higher strength cladding, for example.

MOSS (AI): In terms of void volume, are you sure it's not just a phase change of a reaction between the U_3O_8 and the aluminum going to UAl_x, if you will, which has a lower density. It's hard for me to picture you hot rolling something and ending up with something with a 20% void.

MARTIN: One can make the calculations with reasonable assumptions for densities and show that there is no volume change from the reactions between U_3O_8 and aluminum. I have not run an electron probe across to actually determine the extent and amount of reaction of the U_3O_8-Al dispersion that is consumed during fabrication. It's a very small amount; and I really can't detect it on a microscope. In a special effort after it's been reacted, I can detect it.

MOSS: How do you determine the void volume?

MARTIN: Void volume is determined on the basis of toluene-determined densitities of the particles that we start with and the densitites of the fuel plates from immersion. It appears that the calculated void content is associated with the U_3O_8 particles. When we hit 40 vol. % we find that for our particle size distribution that the continuous matrix phase changes from aluminum to U_3O_8.

LEWIS (Babcock & Wilcock): I think I heard you say that you were not going to irradiate your fuel plates, either the mini-plate or the full size plate, in a test loop. Is that right?

MARTIN: That is our plan.

LEWIS (Babcock & Wilcock): Were you also going to allow them to fail? I was wondering if you could elaborate on that. What kind of problems do you have in the reactor?

MARTIN: I would have to have special permission from ORR operators to plan failure on the fuel test.

LEWIS (Babcock & Wilcock): Is that what you were considering?

MARTIN: I don't plan for the plates to fail.

WOODRUFF (ANL): Is there any bonding problem with the clad on the 100% U_3O_8?

MARTIN: I don't think there would be.

WOODRUFF: You had said that you fabricated 100 wt % U_3O_8 plates.

MARTIN: Yes. Plates were previously evaluated in 1969. They survived heat treatments twice at 500°C for one hour without blistering. This is the last work that was done before the start of our program. That's why I made it the theme of my talk. We do have an opportunity to continue where we stopped.