

US-RERTR Advanced Fuel Development Plans: 1999*

by

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ABSTRACT

Twelve fuel alloys were included in the very-high-density RERTR-1 and RERTR-2 microplate irradiation experiments. Experience gained during fabrication and results from the post-irradiation examination of these fuels has allowed us to narrow the focus of our fuel development efforts in preparation for the next set of irradiation experiments. Specific technical problems in both the areas of fuel fabrication and irradiation performance remain to be addressed. Examples of these are powder fabrication, fuel phase gamma stability versus density, and fuel-matrix interaction. In order to more efficiently address metal alloy fuel performance issues, work will continue on establishing a theoretical basis for alloy stability and metal alloy dispersion fuel irradiation performance. Plans to address these fuel development issues in the coming year will be presented.

INTRODUCTION

In the past year, several significant events have taken place in the US-RERTR advanced fuel development effort. The new fuel test vehicle described at last year's meeting [1] has been proven to be effective and efficient in the testing of novel fuel types.

Sixty four microplate fuel test plates in the RERTR-1 and RERTR-2 experiments have been irradiated to approximately 40% and 70% U^{235} burnup in the Advanced Test Reactor (ATR). The post irradiation examination (PIE) of these experiments is under way, and the first (to our knowledge) low-temperature, high burnup fuel performance data has been generated for U-Mo and U-Nb-Zr alloys.

A new fuel fabrication laboratory has been brought on line at ANL-W. This facility will allow experiments in fuel powder fabrication, fuel plate fabrication, and physical metallurgy of uranium alloys to be carried out in support of program goals.

The goal of the US-RERTR fuel development effort remains unchanged-the development of a dispersion fuel with a uranium density of up to 9 g U/cm^3 that will meet the operating and safety requirements of high power research reactors. Preliminary metal alloy dispersion fuel irradiation test results are promising; there are no indications of fundamental problems with this approach under the set of testing conditions used for these experiments. Our advanced fuel development effort will thus proceed along the

same path in 1999 as in the previous year. The emphasis will be on dealing with fabrication issues and on understanding fuel alloy behavior in- and ex-reactor. This effort will culminate in the irradiation of a refined fuel test matrix in 1999.

SUMMARY OF POST-IRRADIATION EXAMINATION RESULTS

The RERTR-1 and -2 tests were scoping experiments designed to separate fuel alloys that show promise from those that are clearly poor performers. This experiment was designed with a conservative fuel loading of 25 volume percent to ease fabrication and to minimize the chance of plate failure in the face of completely unknown irradiation performance. Fuel types included in this test are listed in Table 1. A more detailed account of the experiment and PIE is found in reference [2].

Table 1. Fuel alloys chosen for scoping irradiation tests
(alloying element in wt%)

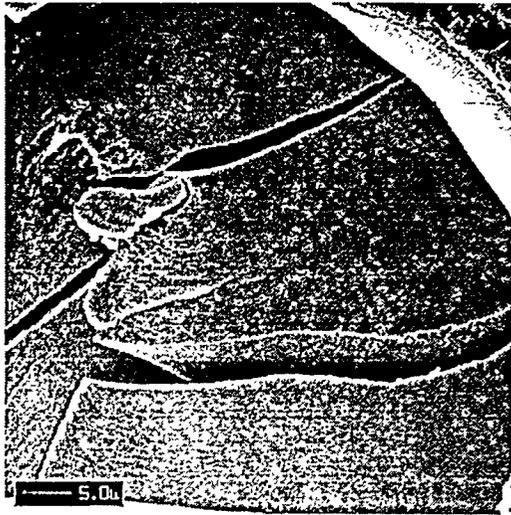
Most γ stable	Intermediate γ stability	Least γ stable ($\alpha + \gamma'$)	Intermetallics	Precipitate Dispersion within Fuel Particles
U-10Mo*	U-6Mo	U-5Nb-3Zr	U ₂ Mo	U-10Mo-0.05Sn
U-8Mo	U-6Mo-1Pt	U-4Mo	U ₃ Si ₂ #	
U-9Nb-3Zr	U-6Mo-0.6Ru			
	U-6Nb-4Zr			

*Fabricated and irradiated using both atomized powder and filings

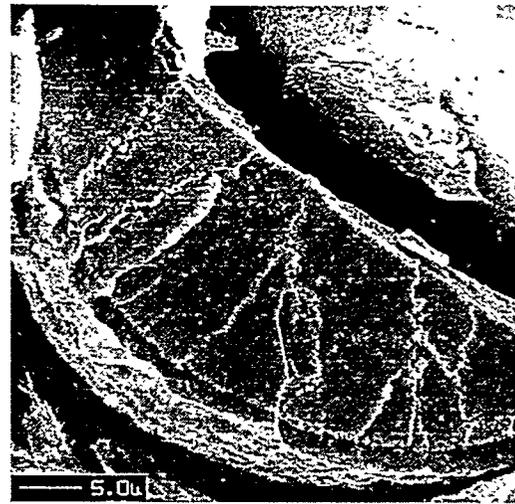
#Fabricated and irradiated using both atomized powder and comminuted powder

Although only partially complete, we consider these experiments successful in having achieved the stated goals. Examination of some of the fuels by SEM (scanning electron microscopy) has been completed, and several promising fuel candidates have been identified. Results from the 40% burnup RERTR-1 test indicate that U-Mo alloys have much better potential as fuel materials than U-Nb-Zr alloys. Two U-10Mo based fuels from RERTR-1 have been examined, and both look promising. The first U-10Mo fuel, made by the rotating disk atomization process at the Korea Atomic Energy Research Institute (KAERI) looks to be better in terms of fission gas bubble behavior. The U-10Mo fuel microstructure compares well to that of U₃Si₂ irradiated in the same experiment, and there is minimal fuel-matrix interaction. Figures 1(a) and (b) show micrographs of these two materials for comparison. Small fission gas bubbles are distributed on what appear to be grain boundaries in the atomized U-10Mo. Shown in Figure 1(c) is the microstructure of a U-10Mo specimen containing fuel produced by grinding a fuel pin with a rotary file. This fuel type showed a more varied microstructure from particle to particle than the atomized powder. Some areas of the fuel have a microstructure similar to the U-10Mo atomized powder (Figure 1(c) bottom) while other areas showed a higher density of bubbles distributed throughout the fuel (Figure 1(c) top). Much of the fuel was intermediate in behavior.

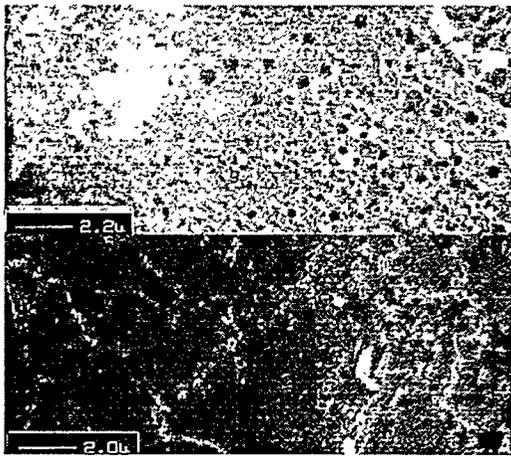
One possible explanation for the difference in fuel phase behavior is that fission gas bubbles in metallic fuel require defects such as sub-grain boundaries to nucleate. Fuel



(a)



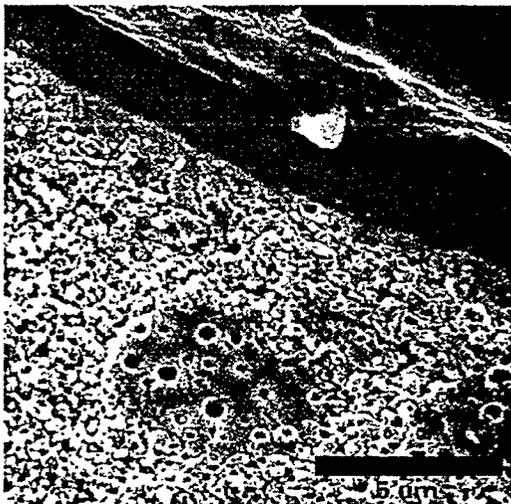
(b)



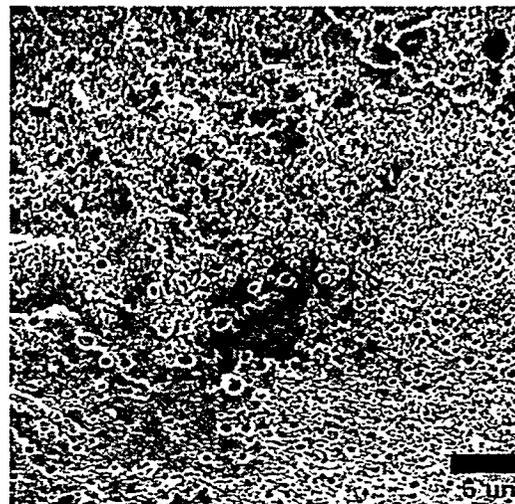
(c)



(d)



(e)



(f)

Figure 1. Fracture surfaces of irradiated fuel particles. All at ~40% burnup from RERTR-1 experiment except (d), at 70% burnup from RERTR-2. (a) U_3Si_2 atomized, (b) U-10Mo atomized, (c) U-10Mo high (top) and low (bottom) gas bubble density regions, (d) U-10 Mo RERTR-2, (e) U-6Mo-0.6Ru, (f) U-8Mo.

powder filings will have high dislocation densities due to the large amount of cold deformation introduced during grinding. The times and temperatures during fabrication combined with the effects of irradiation may be sufficient to allow these dislocations to form sub-grains, which might act as nucleation sites for fission gas bubbles [3]. Seeing this difference in behavior at low burnup, we have expedited the microscopic analysis of a high burnup U-10Mo fuel plate from the RERTR-2 experiment. A micrograph of this fuel plate is shown as Figure 1(d). The key point of interest is that breakaway swelling did not occur at higher burnup, and that fuel-matrix interaction occurs in a slow and predictable manner.

Two higher density fuel alloys, U-6Mo-1Pt and U-6Mo-0.6Ru were also examined using SEM. The microstructure of a typical fracture surface from U-6Mo-0.6Ru is shown in Figure 1(e). The microstructure of U6Mo-1Pt is nearly identical. Comparison of the U-6Mo-0.6Ru microstructure with those in Figures 1(a)-(c) indicates that a higher density of bubbles has been generated in these fuel alloys during irradiation. X-ray diffraction indicates that the fuel in the U-6Mo-1Pt plate has undergone considerable decomposition from the gamma phase during fuel plate fabrication. This observation is consistent with our initial hypothesis that gamma-stabilized uranium fuels should perform better than alpha-uranium under irradiation.

The microstructure of a U-8Mo fuel plate from RERTR-1 is shown in Figure 1(f). It appears to be similar or slightly better in behavior to the U-6Mo-X based alloys, with a fairly uniform distribution of fission gas bubbles. The bubble distribution looks to be similar to that of the U-10Mo at high burnup.

The alloy from the U-Nb-Zr system that had the best potential to meet fuel density criterion, U-5Nb-3Zr, was identified as a poor performer. In contrast to the U-Mo fuels, the fuel phase and aluminum matrix show extensive reaction, and fission gas bubbles have inter-linked to form large cavities at 40% burnup. U-5Nb-3Zr is the only U-Nb-Zr alloy that has been examined to date.

ISSUES IN FUEL FABRICATION AND IRRADIATION PERFORMANCE

Although U-Mo alloys appear to be promising fuel phase materials, several issues remain to be addressed before a metal alloy dispersion fuel becomes a practical RERTR fuel solution. Among these are powder production, understanding the phase stability and transformation behavior of fuel alloy(s) and consequent effects on fuel performance, determining the effect of fuel phase microstructure on fuel performance, and determining fuel behavior at higher loading. The issues above will be addressed in work leading up to the next set of US-RERTR irradiation experiments and in the irradiation experiments themselves.

Powder Production

Alloys being investigated for use as fuel phases are ductile and cannot be converted to powder form by the processes routinely used for oxides or intermetallics. The atomized U-10Mo and U₃Si₂ powder used for some RERTR-1 and -2 fuel plates was manufactured

by KAERI using the method of rotating disk atomization. While rotating disk atomization is a viable powder production option, three other methods [4] of powder production have been or are being investigated within the US-RERTR program. These processes are grinding, cryogenic milling, and hydride-dehydride processing. In addition, a gas atomization process was investigated using gold as a surrogate for uranium.

Gamma Stability and Fuel Performance

The initial supposition made in this work is that gamma phase uranium will perform better under irradiation than alpha-phase uranium, regardless of the type and amount of additions necessary to keep the alloy in the metastable state. In other words, if a U-Mo and a U-Nb-Zr alloy have similar TTT (Time-Temperature-Transformation) behavior, they should perform equally under irradiation. The minimum alloy content required to keep the fuel phase predominantly gamma through the fabrication process and during irradiation should be sufficient. This necessarily simple hypothesis was made in the absence of any meaningful data. As we gain knowledge about the irradiation behavior of the RERTR-1 and RERTR-2 fuels, it appears that the behavior of metal fuel alloys is, as expected, more complex. For example, several published TTT curves for U-8Mo and U-10Mo predict minimal transformation of the gamma phase during fabrication, yet there is clearly a difference in fuel behavior. In order to select a fuel that best meets the tradeoff between density and irradiation performance, the relationship between gamma-phase stability, specific alloying elements, and irradiation performance must be more clearly mapped.

Microstructure and Fuel Performance

It is important to have an understanding of the relationship between fuel powder microstructure and irradiation performance of the particles in the fuel dispersion. For example, although U-10Mo filings performed well under irradiation, it appears that there is a lower density of fission gas bubbles present in the atomized U-10Mo powder, at least at lower burnup. Alloy microstructure-performance relations will be established as results continue to be generated from the PIE. This will include better characterization of fuels both in the as-fabricated and irradiated conditions. Work will proceed along the lines of establishing the defining microstructural characteristics of metal alloy fuels that perform well under irradiation.

Fuel Performance at High Loading

The average measured fuel zone uranium densities of the RERTR-1 and RERTR-2 alloy microplates range from 4.3 to 4.9 g U/cm³. In order to be considered for fuel in high power research reactors, the uranium density in the fuel zone will have to be increased to roughly 8.5 g/cm³. This represents a 73-97% increase over the loadings used for the first experiments. In past experiments, high fuel loadings have, in some cases, proven to be detrimental to fuel performance at high burnup.

FURTHER CHARACTERIZATION OF ALLOYS AND IRRADIATED FUEL

The PIE of RERTR-1 and RERTR-2 will continue. An emphasis has been placed on microscopic examination of the fuels, in order to gain a better understanding of microstructural changes and interactions.

An attempt will also be made to investigate the fine scale bubble structure of some alloy phases using TEM (Transmission Electron Microscopy). This technique will allow the association between fission gas bubbles and dislocations to be determined. It will also allow the resolution of gas bubbles on a scale finer than is possible in the SEM, provide microstructural information, and provide some degree of crystal structure information. The post-irradiation crystal structure of both representative 'good' and 'bad' fuels must be determined in order to more clearly establish the link between the initial crystal structure and the phase stability during irradiation with fuel performance. Of particular interest will be a comparison of the microstructures of ground and atomized powder after irradiation. Characterization of these microstructures and their different fission gas behaviors will no doubt provide insight into the mechanism of fission gas bubble nucleation.

Proceeding with the theory that the degree of gamma stability defines fuel performance to a large measure, work will continue on developing an understanding of the reasons for extension of gamma stability due to ternary additions to U-Mo alloys [5]. Realizing the inherent trade-off between gamma stability and density, a predictive model is being developed for approximating the gamma stability of uranium alloyed with refractory and noble metals. An effective model will allow the efficient optimization of fuel materials to meet both density and fuel performance criteria

The transformation behavior of U-Mo-Pt, U-Mo-Ru, and U-Mo-Os alloys will be determined using a combination of dilation and resistance measurements combined with metallography. Experimental measurements will be used to refine the alloy stability model. This work is being carried out jointly with KAERI, who have already generated transformation data on U-Mo-Pd and U-Mo-Ir alloys.

RERTR-3 AND RERTR-4 EXPERIMENTS

Based on the post-irradiation examination results to date, a general test plan has been developed for the RERTR-3 and RERTR-4 experiments. These experiments are planned for reactor insertion into the ATR during the first half of 1999. The experiments will be similar in many aspects to RERTR-1 and RERTR-2 in that microplate size specimens will be irradiated under moderate temperature and flux conditions. Both experiments will be essentially identical except for discharge burnup. RERTR-3 will be irradiated to approximately 40% burnup and RERTR-4 to near 80%. The RERTR-3 PIE should begin late in 1999, with PIE of RERTR-4 beginning mid-year 2000.

This next set of irradiation experiments will focus primarily on uranium-molybdenum-based alloy fuels. Emphasis will again be placed on attempting to stabilize these alloys in the gamma phase. Binary uranium-molybdenum alloys such as U-10Mo and U-8Mo will be included in the matrix. Although these alloys look very promising, the optimum combination of high alloy density and gamma stability almost certainly lies among ternary and perhaps quaternary alloys. Candidate ternary fuels are U-Mo-Pt, U-Mo-Ru, and U-Mo-Os alloys; the exact compositions are yet to be determined. Fuels will be chosen based on a figure-of-merit that takes into account neutronic efficiency and calculated gamma stability [5]. The gamma stability will then be experimentally determined. If the candidate alloy has the predicted high gamma stability, then it is likely that it will be included in the irradiation test. Approximately a half-dozen of the best candidates will be selected for irradiation. This differs from the approach of RERTR-1 and -2 in that the alloys will be from a relatively narrow composition range.

The target loadings for these experiments will be 8 g U/cm^3 , in the range required for high power research reactors. We have recently demonstrated that microplates containing tungsten as a fuel surrogate can be fabricated at loadings up to 60 volume percent without excess clad thinning. This is equivalent to more than 9 g U/cm^3 for U-10Mo-adequate for testing purposes. Although the fabrication of such test coupons is not representative of commercial fuel fabrication processes, it will allow fuel performance to be evaluated at very high loadings. If the fuel performs well at high loadings on the size scale of the microplates, attention will be given to fabricating larger fuel plates at high loadings.

In order to more clearly identify and separate the effects of fuel phase microstructure on irradiation performance, fuel powder derived from different techniques will be used. Atomized powder appears to have an advantage in fission gas bubble behavior, although its behavior at high burnup is not yet known. If the gas bubble behavior continues to be superior at high burnup, it will be desirable to weight the test matrix in favor of atomized powder. If homogeneous alloy powder can be successfully made by the hydride-dehydride process, it will be included in the test matrix. By including ground powder in both the annealed and as-ground state, a comparison can be made between the two fuel types that may yield insight into microstructure-performance relationships.

Fuel-aluminum interaction during irradiation was minimal at 40% burnup in the higher alloy U-Mo fuels examined to date, and does not appear to be a limiting factor in the use of these alloys as the fuel phase. Due to extensive interaction between aluminum and fuels with low alloy content, such as U-4Mo, it is also desirable to investigate magnesium matrix fuels [6]. Irradiation of these specimens will allow the performance of the low alloy fuels to be gauged independently of its interaction with aluminum and will serve to provide data for this alternate technology, should it become necessary to move away from an aluminum matrix for certain applications.

SUMMARY

The irradiation phase of the RERTR-1 and RERTR-2 scoping experiments has been completed. Both experiments are now undergoing post irradiation examination. Based on results to date, it appears that U-Mo and U-Mo-X based fuels are the best candidates for future irradiation experiments. RERTR-3 and RERTR-4 experiments will focus on these alloys and will be fabricated at higher fuel loading, approaching high power research reactor fuel requirements. It has become evident that the method of fuel powder fabrication and resultant microstructure play an important role in fuel behavior during irradiation, at least during the early phases.

The US-RERTR program will be working in the coming year to address issues related to the production and performance of metal alloy dispersion fuels. These issues include alloy powder production, understanding the phase stability and transformation behavior of fuel alloy(s) and consequent effects on fuel performance, determining the effect of fuel phase microstructure on fuel performance, and determining fuel behavior at loadings in the neighborhood of 8 g U/cm³.

As the field of possible fuels is narrowed further, the behavior of only a few of the best performing fuels will be determined at high temperatures, higher fission rates, and higher burnup in experiments subsequent to RERTR-3 and -4.

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REFERENCES

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- [1] C.L. Trybus, T.C. Wiencek, M.K. Meyer, D.J. McGann, C.R. Clark, "Design and Fabrication of High Density Uranium Dispersion Fuels," *Proc 20th Annual Int. Conf. Research and Test Reactors*, ANL Report (in press 1998).
 - [2] S. L. Hayes, M. K. Meyer, G. L. Hofman, and R. V. Strain, "Post Irradiation Examination of High-Density Uranium Alloy Dispersion Fuels," these proceedings.
 - [3] J. Rest, G.L. Hofman, I. Konovalov, A. Maslov, "Experimental and Calculated Swelling Behavior of U-10Mo Under Low Irradiation Temperatures," these proceedings.
 - [4] C.R. Clark, M.K. Meyer, J.T. Strauss, "Fuel Powder Production from Ductile Uranium Alloys," these proceedings.
 - [5] G.L. Hofman, M.K. Meyer, A. Ray, "Design of Gamma-Phase High Density Uranium Alloys for LEU Dispersion Fuel Applications," these proceedings.
 - [6] T.C. Wiencek, I.G. Prokofiev, D.J. McGann, "Development and Compatibility of Magnesium Matrix Fuel Plates Clad with 6061 Aluminum Alloy," these proceedings.