PROGRESS IN DEVELOPING VERY-HIGH-DENSITY LOW-ENRICHED-URANIUM FUELS

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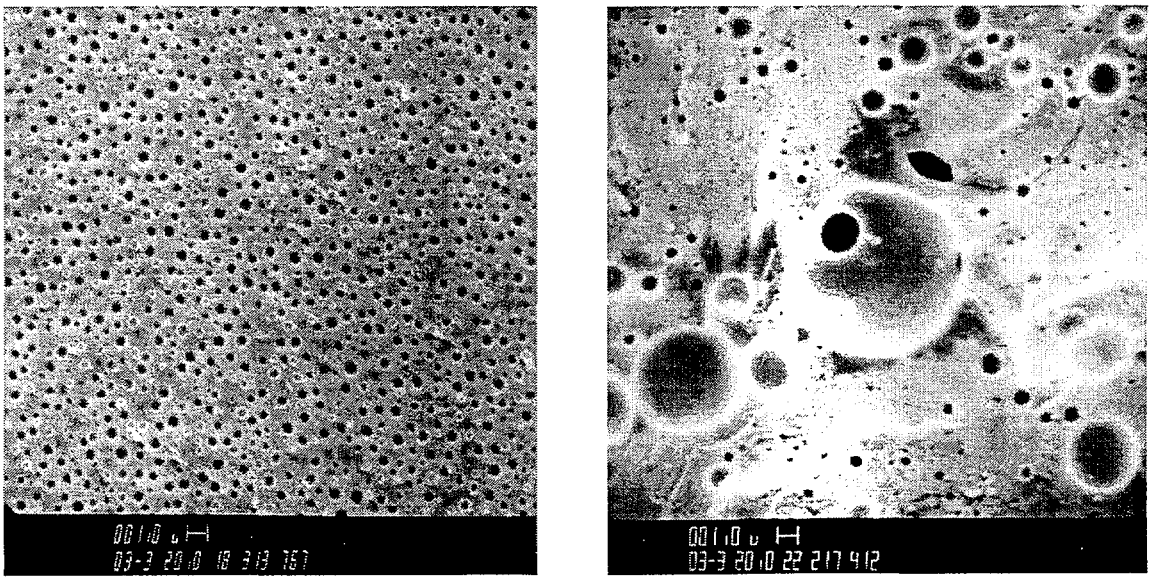
ABSTRACT

Preliminary results from the postirradiation examinations of microplates irradiated in the RERTR-1 and –2 experiments in the ATR have shown several binary and ternary U-Mo alloys to be promising candidates for use in aluminum-based dispersion fuels with uranium densities up to 8 to 9 g/cm^3. Ternary alloys of uranium, niobium, and zirconium performed poorly, however, both in terms of fuel/matrix reaction and fission-gas-bubble behavior, and have been dropped from further study. Since irradiation temperatures achieved in the present experiments (approximately 70°C) are considerably lower than might be experienced in a high-performance reactor, a new experiment is being planned with beginning-of-cycle temperatures greater than 200°C in 8-g U/cm^3 fuel.

1. Introduction

Two years ago, at RRFM'97, we reported the resumption of very-high-density fuel development by the U.S. Reduced Enrichment for Research and Test Reactors program at Argonne National Laboratory (ANL) [1]. Our goal is to achieve a uranium density in dispersion fuel meat of 8 to 9 g U/cm^3, which requires both a very dense fuel dispersant (>15 g U/cm^3) and a very high volume loading of the dispersant (>50 vol.%). The only uranium compounds having such densities are the U_6X compounds, such as U_6Fe and U_6Mn, which have been shown to perform poorly under irradiation [2]. However, alloys of uranium and some transition elements that maintain uranium in the metastable γ (cubic) phase have shown good irradiation performance in bulk form to intermediate burnup under fast reactor conditions. Sufficiently small amounts of one such alloying agent, molybdenum, had been shown to stabilize γ-U and yield at the same time a high uranium density [3]. Even smaller ternary additions of other elements to the right of molybdenum in the periodic table also had been found to provide a powerful stabilizing effect [4].

Our effort has focused on determining which, if any, of these alloys can be fabricated into aluminum-based dispersion fuel and will perform acceptably under irradiation. The two key issues to be addressed are the reaction of the fuel alloy with the aluminum matrix and the irradiation behavior of the dispersion. The former issue is important because excessive reaction will consume the matrix aluminum and, perhaps, a significant amount of the aluminum-alloy cladding. The latter issue relates principally to the behavior of the fission gas in the fuel. If the mobility of the fission gas is small enough, it will be contained in small bubbles that do not interlink, as, for example, in U_3Si_2, shown in Fig. 1(a). Such a fuel will exhibit a steady but stable increase in volume during irradiation. On the other hand, if the fission gas is very mobile, some bubbles grow preferentially, becoming large and interlinking with adjacent bubbles, as illustrated for U_3Si in Fig. 1(b). If the volume loading of such fuel particles is high enough that a significant number of particles are touching, as will be the case for the high densities to which we aspire, the fission gas bubbles can interlink across many particles and lead to unstable, rapid (breakaway) swelling.



(a) U_3Si_2 (b) U_3Si

Fig. 1. Examples of stable (a) and unstable (b) fission-gas-bubble morphologies.

Small dispersion fuel plates containing U-10Mo, U-8Mo, U-6Mo, U-4Mo, U-6Mo-1Pt, U-6Mo-0.6Ru, U-10Mo-0.05Sn, U-9Nb-4Zr, U-6Nb-4Zr, U-5Nb-3Zr, U_2Mo, and U_3Si_2 were irradiated in the Advanced Test Reactor (ATR) to screen their behavior. (The number preceding the alloying element signifies its content in weight percent.) The U_3Si_2 samples provided normalization to previous results; the U_2Mo samples were irradiated since γ-U decomposes into α-U plus U_2Mo. In addition to machined or ground powders of all alloys, powders of U-10Mo and U_3Si_2 produced by centrifugal atomization were provided by the Korea Atomic Energy Research Institute (KAERI). In this paper we summarize the results of these experiments obtained to date (early February 1999) and briefly describe a follow-on experiment.

2. Experiment Description

We have called the fuel plates fabricated for these experiments "microplates" owing to their small size. Their external dimensions are 76 mm x 22 mm x 1.3 mm. The fuel zone is elliptical in shape with major and minor axes of approximately 51 mm and 9.5 mm, respectively; the fuel zone thickness is nominally 0.5 mm. In spite of their small size, we have shown by mechanical analysis that the fuel zone of a microplate is essentially unrestrained in the thickness direction and, consequently, behaves in the same manner as a larger plate. Since the goal of these experiments was to determine as quickly as possible basic information of fuel/matrix interaction and fuel particle swelling, we chose to fabricate plates at about one-half of the goal density in order to ease fabrication problems. The plates were produced by standard powder metallurgy and hot roll-bonding techniques. Although we planned a 25-vol% loading, actual loadings (determined by X-ray density measurements) fell between 26 and 41 vol.%, with an average of approximately 30 vol.%. The enrichment ranged between 19.1 and 19.8%, and a microplate typically contained about 0.11 g of ^{235}U.

The irradiation vehicles, designated RERTR-1 and RERTR-2, each consisted of a flow-through "basket" holding eight vertically stacked flow-through capsules. Each capsule held four microplates in a miniature fuel element configuration. The irradiation vehicles occupied small I-hole positions (I-22 and I-23) in the control drum region of the ATR. Based on calculations, the neutron flux, microplate power, surface heat flux, and fuel centerline temperature were approximately 1.3×10^{18} n m^{-2} s^{-1}, 500 kW, 5.5×10^5 W m^{-2}, and 70°C, respectively, at the axial position of highest neutron flux at the start of the irradiation. Vehicle RERTR-1 was irradiated for 94 effective full-power days (EFPD) during the period August 23, 1997, through November 30, 1997, and RERTR-2 was irradiated for 232 EFPD during the period August 23, 1997 through July 6, 1998, achieving (calculated) ^{235}U burnups between 39 and 45% and between 65 and 71%, respectively.

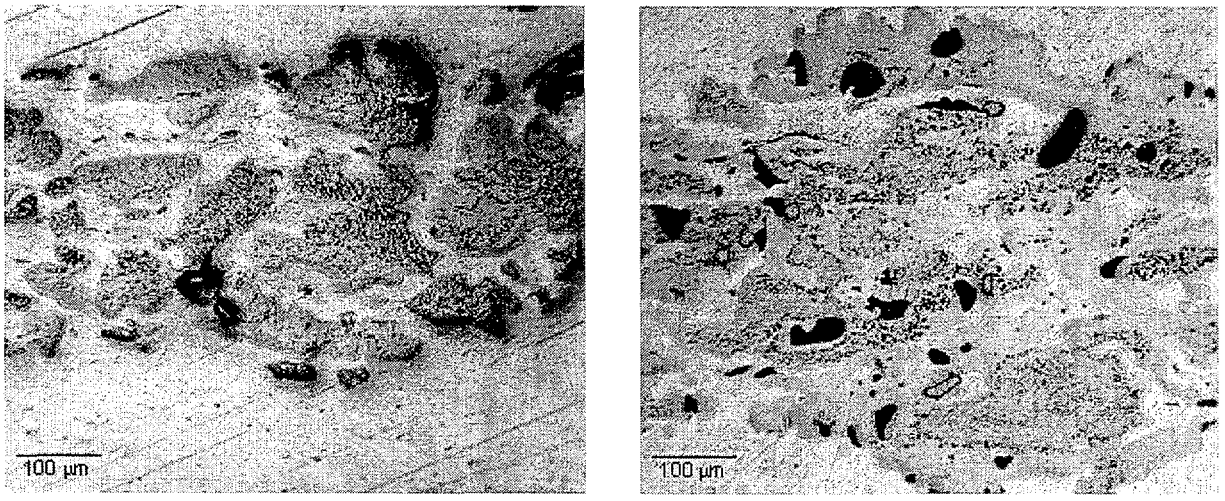
3. Experiment Results

Considerable information about the extent of the fuel/matrix reaction was derived during fabrication. One of the fabrication steps is a one-hour blister anneal at 485°C. Plate thickness measurements taken before and after the anneal revealed significant thickness increases in all of the U-Nb-Zr plates and in the U-4Mo plates, indicating reaction to form less-dense compounds. As the alloying addition and, hence, the gamma stability decreased, the reaction became more pronounced. The binary U-Mo alloys with at least 6 wt.% Mo and the ternary U-Mo alloys showed little or no thickness increase. More recently, X-ray diffraction was used to study the phase composition of fuel particles removed from fabricated fuel plates. Except for the U-10Mo atomized powder, all of the powders examined contained a significant amount of α-U, which must have come from partial decomposition of the γ-U phase during fabrication. This will be addressed further when the results of the metallography are discussed.

After irradiation and cooling, the capsules were removed from the baskets and shipped to the Alpha-Gamma Hot Cell Facility (AGHCF) at ANL's site in Illinois. Examinations have included visual inspection, thickness measurements, gamma scanning, and metallography. All examinations but the metallography have been completed. Visual inspection revealed significant corrosion of the cladding surface, especially over the fuel zone. Cladding breaches, subsequently shown to be due to pitting corrosion, were identified in five microplates. This is a common problem in the ATR environment and is not related to fuel performance. Thickness measurements on the RERTR-1 plates showed, in general, decreased thickness over the fuel zone, consistent with erosion or spallation of a corrosion product. The plates from RERTR-2, on the other hand, increased modestly in thickness over the fuel zone, consistent with swelling observed during metallographic examination. Samples from several microplates have been submitted for burnup analysis. We also plan to perform a standard postirradiation blister anneal test on selected microplates.

The most-important information has come from optical metallography of ground and polished sections and, especially, from scanning electron microscopy (SEM) of fracture surfaces of fuel particles. Since we are in the midst of our examinations, however, the following must be considered as a preliminary assessment.

Optical metallography revealed that the U-10Mo, U-8Mo, U-6Mo, U-6Mo-1Pt, and U-6Mo-0.6Ru alloys behaved very well. The fuel particles are surrounded by thin layers of reaction product, and fission gas bubbles are small and relatively uniformly distributed, as shown for U-6Mo at approximately 70% burnup in Fig. 2(a). In contrast, U-5Nb-3Zr at approximately 40% burnup exhibited extensive reaction with the matrix (little matrix remains) and cladding (significant



(a) U-6Mo at ~70% burnup (b) U-5Nb-3Zr at ~40% burnup

Fig. 2. Optical micrographs illustrating the good irradiation behavior of U-Mo alloys and the poor irradiation behavior of U-Nb-Zr alloys.

penetration) as well as an unstable fission-gas-bubble behavior (interlinkage has begun), as shown in Fig. 2(b). The large voids seen in Fig. 2(b) are sintered remnants of voids formed during the fuel/matrix reaction. The amount of reaction alone precludes use of this fuel at the high volume loading needed to achieve our density goal in an aluminum-matrix dispersion. In addition, the unstable fission-gas-bubble behavior in the unreacted fuel would preclude its use even if fuel/matrix reaction were not a problem. Therefore, we are abandoning further study of U-Nb-Zr alloys.

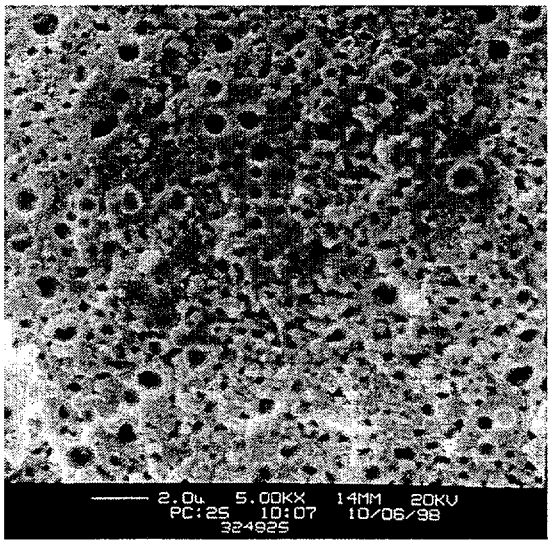
Scanning electron micrographs of fracture surfaces of several of the 70%-burnup fuel samples are shown in Figs. 3 and 4. The appearance of each of the four fuels shown in Fig. 3 is quite similar. The fuel has a granular appearance, suggesting grain refinement. The fission gas bubbles appear to be forming at the boundaries of the small grains. There is no obvious evidence of a two-phase microstructure in any of the irradiated samples. Therefore the decomposition of the γ-U phase which occurred during fabrication was probably reversed early during irradiation. Such an effect has been reported in the literature as owing to fission-spike mixing at high fission rates. There is no evidence of interlinking of the relatively uniformly distributed fission gas bubbles. Though not evident in the micrographs shown here, we have seen small areas in many of the fuel particles that have a different appearance. We think that this might be due to compositional inhomogeneities; however, we have not yet been able to perform electron-beam microchemical analysis owing to the high gamma field of the samples.

Fig. 4 compares the behavior of an atomized powder to that of ground powder. The thin interaction layer is evident in the lower-magnification views of both fuels. The higher magnification views are similar to each other and to those shown in Fig. 3. Grain refinement has occurred in all of the alloys, resulting in an average grain size of <0.5 μm. However, the presence of significant areas of the atomized fuel with no visible bubbles indicate that grain refinement started earlier (at lower burnup) in the ground powder than in the atomized powder. We believe this to be the result of a high degree of deformation (that is, initial dislocation density) in the former introduced during grinding.

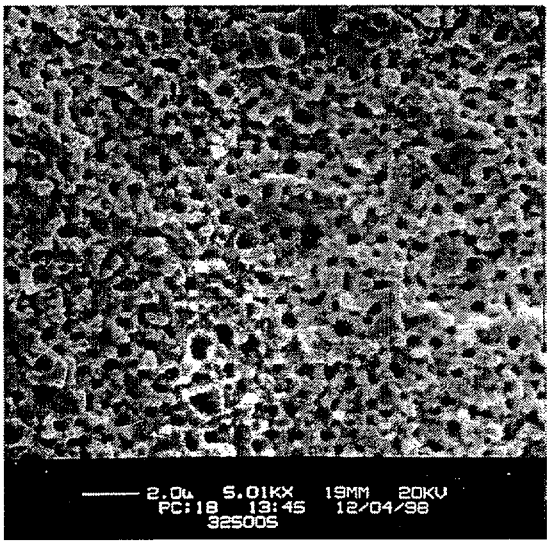
4. Next Experiment

While we are very encouraged by the results from the binary and ternary U-Mo alloys in the RERTR-1 and –2 experiments, a number of issues still must be addressed, including the effects of higher loading, higher fuel temperature, higher burnup, and scale-up to full-sized fuel plates. Based on our understanding of dispersion fuels in general and the observed fission-gas-bubble behavior in the U-Mo alloys, we feel confident that loading, burnup, and scale-up will present no problems from an irradiation point of view. Therefore, we have decided to concentrate on the effects of higher temperature in our next experiment. This is particularly important since a reactor requiring the highest uranium density most likely will operate at high power density and, hence, high fuel temperature.

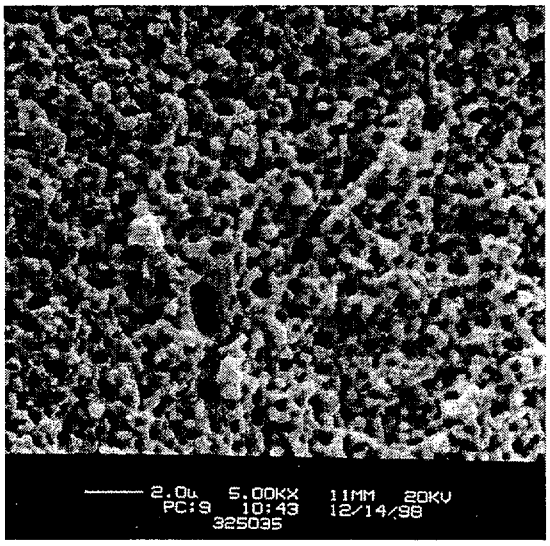
The simplest method to perform a high-temperature irradiation and to produce data that are the simplest to analyze is to use a high-temperature irradiation loop. Unfortunately, that is also the most expensive method. However, preliminary design calculations indicate that fuel centerline temperatures as high as 250°C can be achieved in small plates irradiated in one of the outboard A-holes of the ATR. Much-higher fluxes are available in the A-holes than in the I-holes used for the RERTR-1 and –2 experiments, but the A-hole diameter is only 40% that of the I-hole. Consequently, the fuel plates must be even smaller than our microplates. The current concept envisions a plate having dimensions approximately 41 mm x 10 mm x 1.3 mm with an elliptical-shaped fuel zone having dimensions 22 mm x 4.8 mm x 0.5 mm. Even though the fuel zone is only half as wide, mechanical analysis again has shown that the dimensions are large enough to provide plate-like behavior. Two columns of four plates each would be contained in a single capsule and as many as six capsules could be stacked vertically. The uranium density in the plates will be 8.0 g U/cm^3, nominally. In order to obtain the desired temperatures, enrichment greater than 20% is planned for many of the fuel plates. In addition to allowing the desired temperatures to be reached, the higher enrichment will allow, in a single ATR cycle, fission densities equivalent to LEU fission densities achieved at up to 70% burnup. Many of the U-Mo alloys tested in RERTR-1 and –2 will be tested; both ground and atomized powders will be used. We plan to insert this test, designated RERTR-3, in the ATR during the summer of 1999.



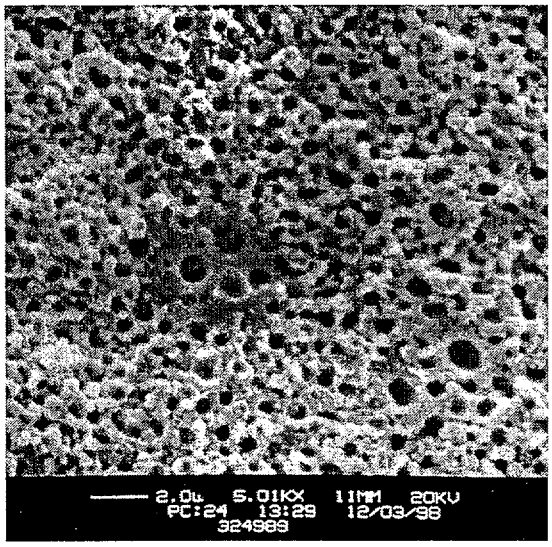
(a) U-10Mo



(b) U-8Mo



(c) U-6Mo



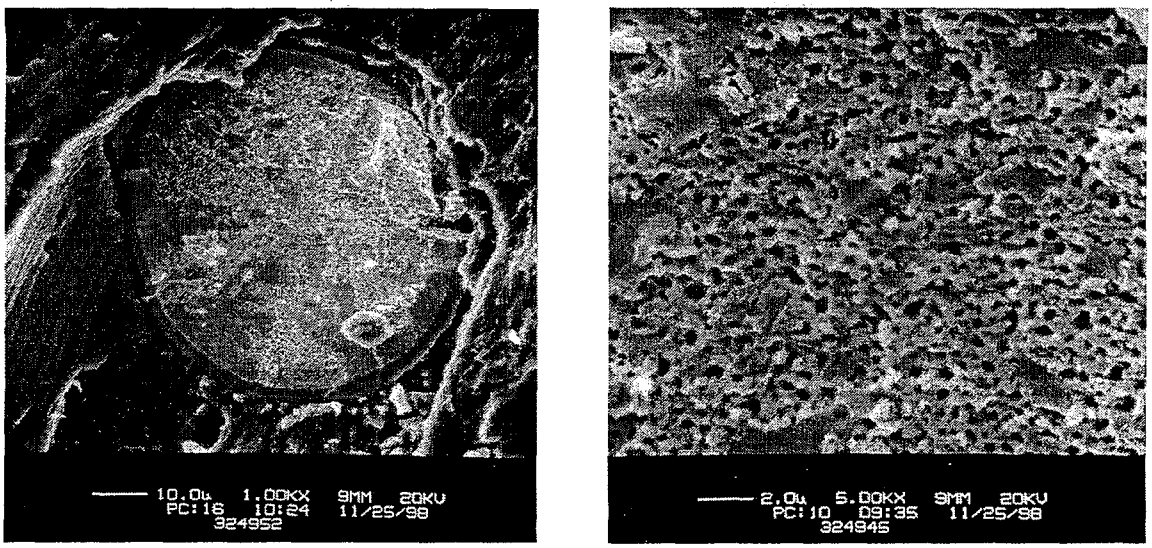
(d) U-6Mo-1Pt

Fig. 3. Scanning electron micrographs of typical portions of fuel particles from various U-Mo alloys at approximately 70% burnup illustrating similar behavior.

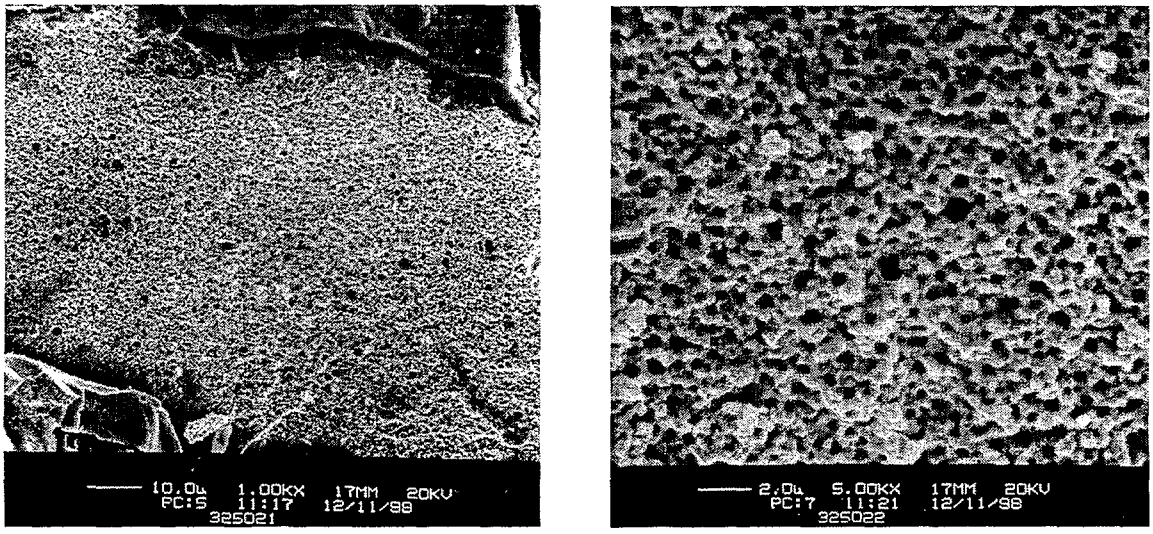
We are also discussing the irradiation of full-sized plates with the fuel fabricators BWX Technologies, Inc., and CERCA and with the CEA in France, SCK/CEN in Belgium, and JRC in the Netherlands. At this time, however, no plans have been formulated.

5. Conclusions

Preliminary results from the postirradiation examinations of microplates irradiated in the RERTR-1 and –2 experiments in the ATR have shown several binary and ternary U-Mo alloys to be promising candidates for use in aluminum-based dispersion fuels with uranium densities up to 8 to 9 g/cm^3. Ternary alloys of uranium, niobium, and zirconium performed poorly, however, both in terms of fuel/matrix reaction and fission-gas-bubble behavior. Therefore, these alloys have been dropped from further study. The fuel temperatures achieved in the RERTR-1 and –2 experiments, although prototypic of temperatures which might be expected in reactors requiring 4 to 5 g U/cm^3, are significantly lower than might be experienced in high-performance reactors. Therefore, a new experiment is being planned for irradiation beginning during the summer of 1999 with beginning-of-cycle temperatures greater than 200°C in 8-g U/cm^3 fuel.



(a) Atomized U-10Mo



(b) Ground U-6Mo-0.6Ru

Fig. 4. Scanning electron micrographs of portions of fuel particles comparing the behavior of atomized and ground U-Mo alloy particles at ~70% burnup.

6. References

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