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Stable In-reactor Behaviors of Centrifugally Atomized U-10wt.%Mo Dispersion Fuel at Low Irradiation Temperature

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Abstract

In order to examine the in-reactor performance of very-high-density dispersion fuels, U-10wt.%Mo microplates with centrifugally atomized powder have been irradiated at low temperature. The U-10wt.%Mo fuels do not show breakaway swelling, but stable in-reactor irradiation behaviors, like U_3Si_2. *Moreover, centrifugally atomized U-10wt.%Mo microplates have finer and more uniform bubble size distribution than mechanically ground microplates. It seems to originate from the onset of gas bubble formation in the atomized powder at higher burnup, due to no deformation damage during the powder preparation process.*

1. Introduction

The conversion from high enriched uranium (HEU) to low enriched uranium (LEU) for use in research reactor fuel requires a large increase of uranium per unit volume to compensate for the reduction in enrichment. The relatively high- density compound U_3Si_2, with a uranium density of 11.6 g-U cm^{-3}, was found to possess a very stable irradiation behavior; however, fabricability limits do not allow fuel element loadings higher than 6 g-U cm^{-3} [1-5]. Therefore, very-high-density fuels having U-loadings up to 8~9 g-U cm^{-3} require both a very dense fuel dispersant (>15 g U/cm^3) and a very high volume loading in the dispersant (>50 vol.%). Uranium alloys including small amounts of alloying elements can be considered as fuel dispersants. 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 [6]. 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 element, molybdenum, had been shown to stabilize γ-U and at the same time yield a high uranium density [7].

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 shown for U_3Si_2 [4-5]. 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 shown for U_3Si [8-9]. If the volume loading of such fuel particles is high enough that a significant number of particles are touching, the fission gas bubbles can interlink across many particles and lead to unstable, rapid (breakaway) swelling.

Early irradiation experiments with uranium alloys showed the promise of acceptable irradiation behavior, if these alloys could be maintained in their cubic γ-U crystal structure [10]. If centrifugally atomized U-Mo powder can retain this gamma uranium phase during fuel element fabrication and irradiation, and if it is compatible with aluminum which forms the matrix of dispersion fuels, the uranium alloy would be a prime candidate for dispersion fuel for research reactors. It has recently been reported that very-high-density atomized U-10wt.%Mo powder prepared by rapid solidification retains the isotropic high temperature γ-U phase. Moreover, the gamma phase did not decompose into the equilibrium γ-U and U_2Mo two phase structure in U-10wt.%Mo alloy after annealing of up to 100 hours at 400°C [11]. In addition, the U-10wt.%Mo powder dispersed in aluminum did not show significant dimensional changes after annealing up to 2,000 hours at 400°C, and interdiffusion between U-10wt.%Mo and aluminum was found to be minimal [12]. In order to examine the in-reactor performance of very-high-density dispersion fuels, U-10wt.%Mo microplates with centrifugally atomized and mechanically comminuted powder have been irradiated to approximately 40at.% and 70at.% burnup at low temperature.

2. Experimental procedure

Low enriched uranium lumps (99.9 pct pure) and molybdenum buttons (99.7 % pct pure) were used for the preparation of the U-10wt.%Mo powders by mechanical comminution from as-cast ingot and rotating-disk centrifugal atomization [11, 13]. Dispersion fuel rods with a nominal volume fraction of 25% were prepared by blending the U-10wt.%Mo and aluminum powder and by extruding the blended powders at a working temperature of 400°C.

The microplate fuel samples were fabricated with external dimensions of 76 mm x 22 mm x 1.3 mm in aluminum cladding. 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. U-10wt.%Mo microplates, which were irradiated at an average fuel center temperature of 91°C, were discharged after 94 effective full-power days (EFPDs) of irradiation, and then discharged after 232 EFPDs of irradiation, achieving (calculated) ^{235}U burnups of 40at.% and 70at.%, respectively. PIE of the microplates is performed primarily using a scanning electron microscope.

3. Experimental results

Fig. 1 shows scanning electron micrographs for two U-10wt.%Mo microplates at 40at.% burnup. Fig. 1-(a) is from a U-10wt.%Mo microplate prepared using centrifugally atomized fuel powder, whereas Fig. 1-(b) is for a U-10wt.%Mo microplate prepared using mechanically fuel powder. The cell structure of as-atomized particles showing Mo micro-segregation changed into a homogenized grain structure during the irradiation process. In addition, the grains of the atomized particles after irradiation become coarser in the range of 5~10 μm in grain size rather than in the range of 2~3 μm in cell size of as-atomized particles [11]. The fuel-matrix interaction layer of the atomized spherical particles is very uniform and in the range of 2~3 μm microns in thickness at 40at.% burnup; however, the interaction layer on the mechanically ground fuel powder is more irregular and appears to be as much as twice the thickness. The fission gas bubble morphologies appear quite different for these two fuel powder types. The maximum bubble diameter of atomized and ground U-10wt.%Mo microplates is approximately 0.4 μm and 0.8 μm, individually. The average bubble diameter of atomized and ground U-10wt.%Mo microplates is approximately 0.2 μm and 0.1 μm, respectively. The mechanically ground powder has a higher density of gas bubbles, having in general bubbles of larger diameter that are distributed uniformly within fuel particles as well as on grain or subgrain boundaries. Some prominent coarse bubbles can be observed in grains. In contrast, the atomized fuel powder has fewer bubbles of generally smaller diameter that appear to be concentrated on grain boundaries almost exclusively.

Scanning electron micrographs of atomized and ground U-10wt.%Mo microplates at 70at.% burnup are shown in Fig. 2. The bubble size and the population density increase greatly, as the burnup of the microplates increase. The maximum bubble diameter of atomized and ground U-10wt.%Mo microplates is approximately 0.8 μm and 1.2 μm, respectively. The average bubble diameter of atomized and ground U-10wt.%Mo microplates is approximately 0.2 μm and 0.4 μm, respectively. There are fission gas bubble-free zones of ~35% in area fraction and 5~10 μm in zone size in the atomized particles; however, less than 10% in area fraction and smaller than 3 μm in zone in the ground particles. Gas bubble-free zones, not entering the stage of bubble formation, are located primarily around the perimeter rather than toward the center of the atomized particle. Small bubble-free zones are sometimes formed in bubble-rich zones in places. However, the ground particles don't show small and uniform bubbles in grains. This comparison shows that the onset of gas bubble formation in the atomized powder occurs at a higher burnup vis-à-vis ground powder. The atomized particles have a more uniform interaction layer than the ground particles and almost the same thickness of 2~3 μm as the ground particles. The interaction layer between U-10wt.%Mo particle and Al matrix has very few bubbles.

Back-scattered scanning electron images of an atomized U-10wt.%Mo microplate at

70at.% burnup are shown in Fig. 4. The microplate at 70at.% burnup does not have the micro-segregation shown Mo-depleted zones in the cell boundaries of as-atomized powder. And no traces of aluminum penetration are confirmed within the atomized particle. The average size of bubble-free islands of 3~10 μm is almost the same as the homogenized grain size of 5~10 μm. Fission gas bubbles are nucleated along grain boundaries and then grow continuously toward grains, primarily leaving many bubble-free zones within grain.

4. Discussion

The U-10wt.%Mo fuels, irrespective of powder kind, do not show breakaway swelling at all, indicating the maintenance of metastable cubic γ-U phase; however, pure uranium and U-Zr-Nb alloy, etc. that exist in the orthorhombic α-U phase are poor performers under irradiation due to anisotropic growth that induces grain-boundaries tearing and resultant breakaway swelling. The average bubble diameter of atomized U-10wt.%Mo is almost the same as that of atomized U_3Si_2 [1-5]. 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.

Centrifugally atomized U-10wt.%Mo microplates have finer and more uniform bubble size distribution than mechanically ground microplates. The possible reasons are supposed as follows. The U-10wt.%Mo dispersion fuel has a granular appearance, suggesting grain refinement. The atomized particles do not have prominent fabrication damage except for excessive vacancy due to raped solidification during centrifugal atomization. Whereas, the ground particles have severe deformation damage with high-density dislocation formed during mechanical comminution process. A portion of metastable γ-U phase is decomposed into α-U phase and centering grain boundaries during hot rolling for the preparation of the microplates, performed after isothermal annealing at 500°C for 2 hours. It has been reported recently that the communited U-10wt.%Mo powder contained a significant amount of γ-U, which must have come from partial decomposition of the γ-U phase during fabrication, unlike the U-10Mo atomized powder [14]. In addition, heavily deformed areas having high stored strain energy are liable to be nucleated and grown into subdivided grains around grain boundaries during hot rolling or irradiation test at elevated temperature. As recrystallized subgrain boundaries around grain boundaries have very high fission gas mobility, the onset of gas bubble formation in the ground powder occurs at a lower burnup vis-à-vis ground powder. The fission gas bubbles appear to be forming at the boundaries of the small grains. 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. Thence, U-10wt.%Mo microplates with ground fuel particles have coarser and more uneven bubble size distribution than U-10wt.%Mo microplates with atomized fuel particles.

5. Conclusions

The U-10wt.%Mo fuels, irrespective of powder kind, do not show breakaway swelling at all, indicating the maintenance of metastable cubic γ-U phase, but very stable in-reactor irradiation behaviors. Centrifugally atomized U-10wt.%Mo microplates show finer and more uniform bubble size distribution than mechanically ground microplates. The possible reasons are supposed as follows. The atomized particles do not have prominent fabrication damage; however, the ground particles have severe deformation damage with high-density dislocation formed during mechanical comminution process, induced to recrystallized subgrain around grain boundaries with high fission gas mobility during hot rolling or irradiation test at elevated temperature. The grain refinement and onset of gas bubble formation in the ground powder occurs at a lower burnup than in the atomized powder. It causes the ground microplates to have coarser and more uneven bubble size distribution compared with the atomized microplates.

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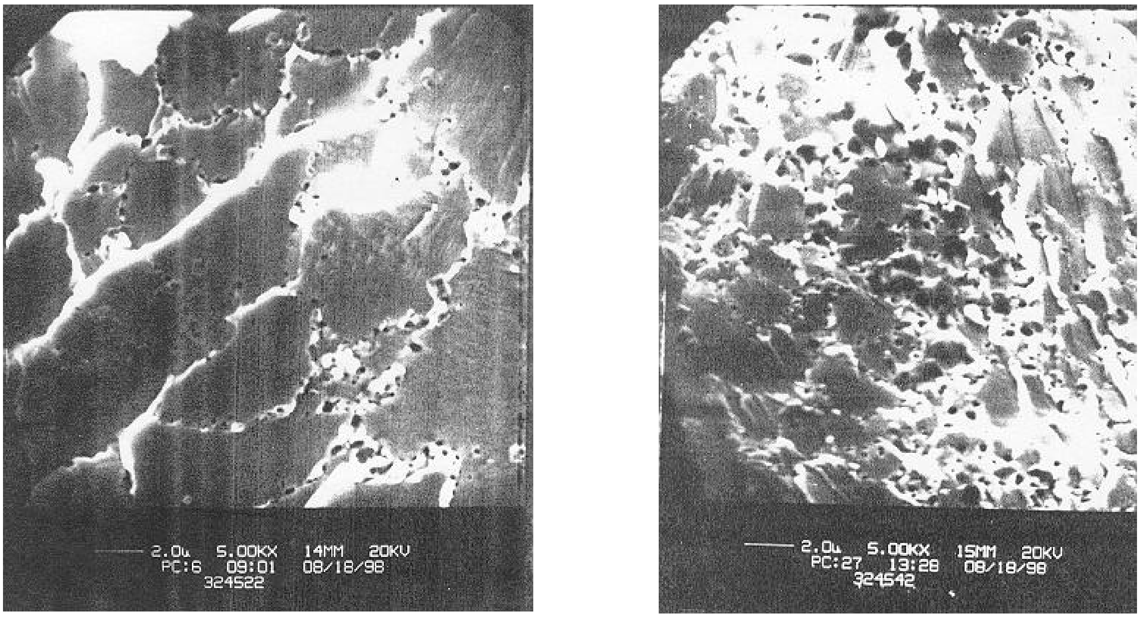

Fig. 1. Scanning electron micrographs for U-10wt.%Mo microplates at 40at.% burnup;

(a) Atomized fuel powder, (b) Ground fuel powder.

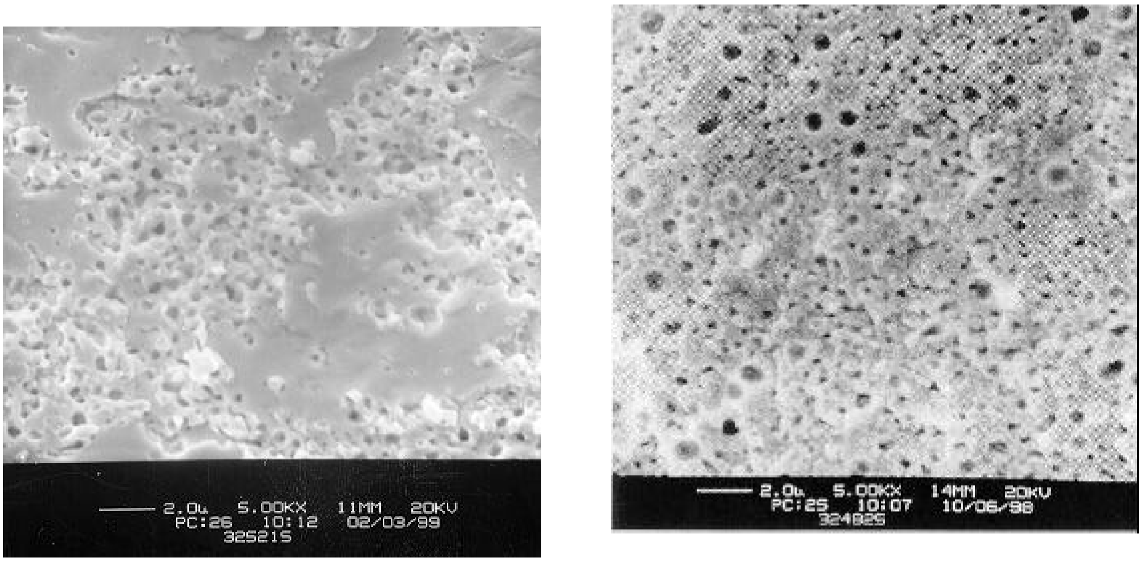

Fig. 2. Scanning electron micrographs for U-10wt.%Mo microplates at 70at.% burnup;
(a) Atomized fuel powder, (b) Ground fuel powder.

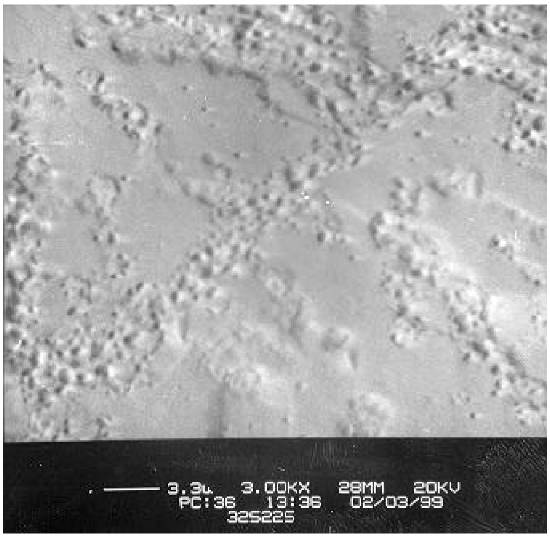

Fig. 3. Back-scattered scanning electron image of atomized U-10wt.%Mo microplate at 70at.% burnup.