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

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Abstract

In order to examine the in-reactor performance of very-high-density dispersion fuels for high flux performance research reactors, U-10wt.%Mo microplates containing centrifugally atomized powder were irradiated at low temperature. The U-10wt.%Mo dispersion fuels show stable in-reactor irradiation behaviors even at high burn-up, similar to U_3Si_2 dispersion fuels. The atomized U-10wt.%Mo fuel particles have a fine and a relatively uniform fission gas bubble size distribution. Moreover, only one of third of the area of the atomized fuel cross-sections at 70at.% burn-up shows fission gas bubble-free zones. This appears to be the result of segregation into high Mo and low Mo.

Key Words : in-reactor performance, very-high-density dispersion fuels, research reactors, U-10wt.%Mo, centrifugally atomized powder

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 content 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 very stable irradiation characteristic; 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 [6-7]. 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. 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.

The fuel dispersants for research and test reactors have been prepared by the comminution of as-cast or heat-treated uranium alloy [8-9]. In order to simplify the preparation process and improve the properties, a rotating-disk centrifugal atomization method has been used [10]. In addition, centrifugal atomization as rapid solidification processing (RSP) of nuclear fuel materials can provide beneficial features which could not be achieved using the conventional methods. It is known that this process, in which the powder is prepared by a centrifugal force, has the advantages that the powder has a rapidly solidified microstructure, a relatively narrow particle size distribution, and a spherical shape [11]. It has been reported that very-high-density atomized U-10wt.%Mo powder prepared by rapid solidification retains the isotropic γ-U phase [12]. Moreover, the gamma phase did not decompose into the equilibrium γ-U and U_2Mo two phase

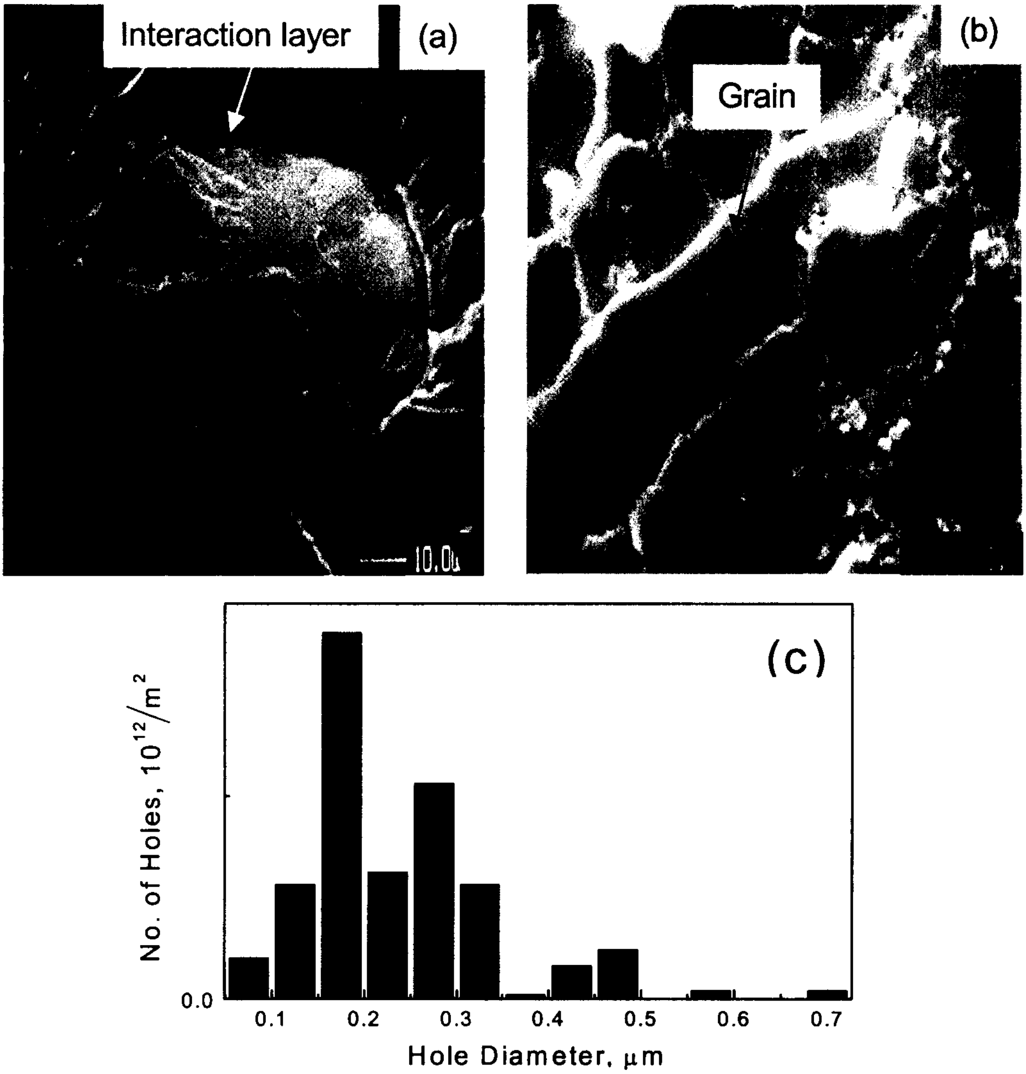

Fig. 1. Scanning Electron Micrographs (a-b) and Apparent Fission Gas Bubble Size Distribution (c) on the Fracture Surface in the Fuel Meat of Atomized U-10wt.%Mo Microplates at 40at.% Burn-up

structure in U-10wt.%Mo alloy after annealing for up to 100 hrs at 400℃. In addition, the U-10wt.%Mo particles dispersed in aluminum did not show significant dimensional changes after annealing up to 2,000 hrs at 400℃, and interdiffusion between U-10wt.%Mo and aluminum was found to be minimal [13].

The two key issues for the in-reactor performances of atomized U-10wt.%Mo dispersion fuel 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 possibly a significant amount of the aluminum-alloy cladding. The latter issue relates principally to the irradiation behavior of the fission gas in the fuel. If the diffusivity of fission gas atoms is small enough, the atoms will be contained in small bubbles that do not interlink, as shown for U_3Si_2 [4-5]. Such a fuel will exhibit a slow 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 [14-15]. 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 rapid swelling. In this study, the U-10wt.%Mo microplate meats with a nominal volume fraction of 25% were irradiated to characterize the in-reactor behavior of atomized U-Mo dispersion fuels at approximately 40at.% and 70at.% burn-up at low temperature. Thereafter, the in-reactor performance of atomized U-10wt.%Mo dispersion fuels was examined primarily using a scanning electron microscope (SEM), compared with that of ground fuels.

2. Experimental

Low enriched uranium lumps (99.9% pure) and molybdenum buttons (99.7% pure) were used for preparation of the U-10wt.%Mo powders by rotating-disk centrifugal atomization [12]. Dispersion fuel meats with a nominal fraction of 25vol.% fuel particles were prepared by blending the U-10wt.%Mo and aluminum powder and by rolling the blended powders at a working temperature of about 485℃. The microplate fuel samples were fabricated with external dimensions of 76 mm × 22 mm × 1.3 mm in aluminum cladding by the hot roll-bonding method. The fuel

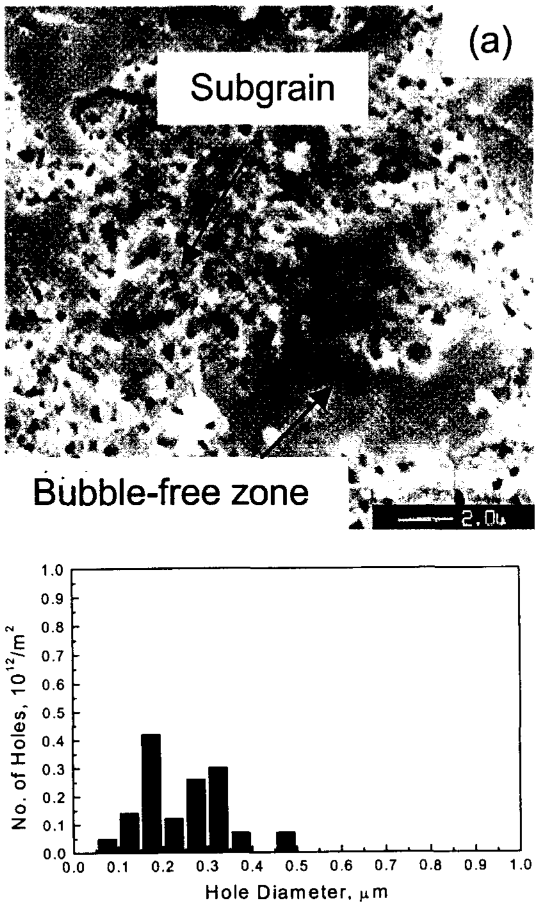

Fig. 2. Scanning Electron Micrograph (a) and Apparent Fission Gas Bubble Size Distribution (b) on the Fracture Surface in the Fuel Meat of Atomized U-10wt.%Mo Microplates at 70at.% Burn-up

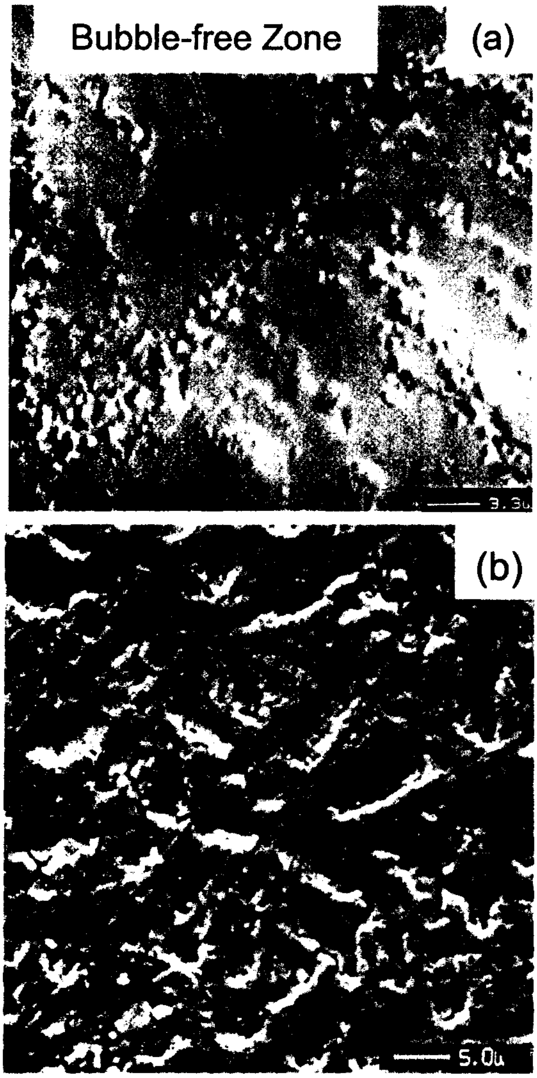

Fig. 3. Back-Scattered Scanning Electron Image of a Fractured Surface (a) and Scanning Electron Image of a Polished Surface (b) of Atomized U-10wt.%Mo Microplate at 70at.% Burn-up

zone was elliptical in shape with major and minor axes of approximately 51 mm and 9.5 mm, respectively; the thickness of fuel zone is nominally 0.5 mm.

Atomized U-Mo dispersion fuels have been developed to use the nuclear fuels for high performance reactors. The average burn-up, maximum thermal neutron flux, and peak fuel centerline temperature of the fuels for high performance reactors are generally up to 50at.%, 1×10^{19} $n/m^2\cdot s$ (unperturbed) and 200℃, respectively. In this experiment, U-10wt.%Mo microplate meats with a nominal fraction of 25vol.% fuel particles were irradiated in higher burn-up at lower temperature in the Advanced Test Reactor (ATR), one of the high performance

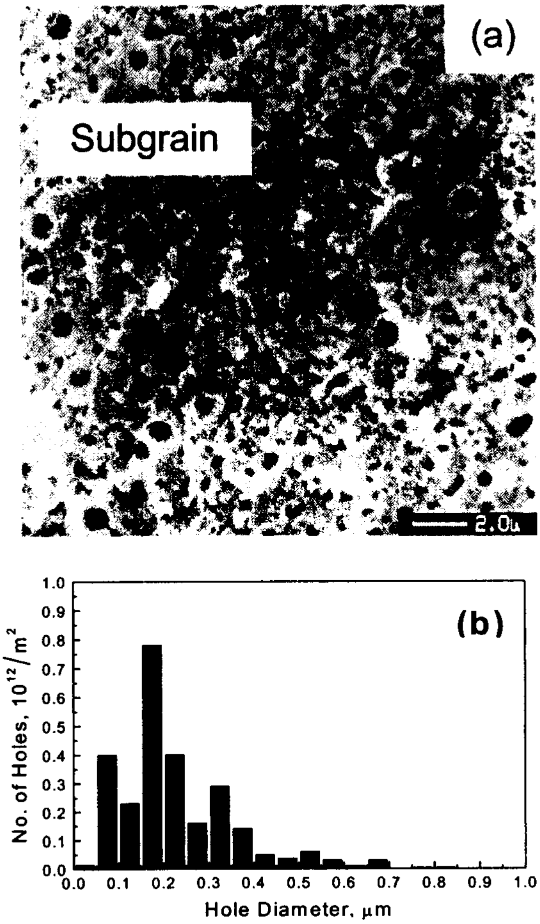

Fig. 4. Scanning Electron Micrograph (a) and Apparent Fission Gas Bubble Size Distribution (b) on the Fracture Surface in the Fuel Meat of Ground U-10wt.%Mo Microplates at 70at.% Burn-up

reactors, than in the high performance reactors, in order to investigate the feasibility of the U-Mo dispersion fuel materials as the very-high-density fuels. Based on calculations, the burn-up, neutron flux, and fuel centerline temperature were approximately 70at.%, 1.3×10^{18} n/m$^2\cdot$s, and 65℃, respectively, at the axial position of highest neutron flux at the start of the irradiation [16].

The irradiation vehicles, designated RERTR-1 and RERTR-2, each consisted of a flow-through "basket" holding vertically stacked flow-through capsules. Each capsule held several 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 the calculations, the neutron flux, microplate power, surface heat flux, and fuel centerline temperature were approximately 1.3×10^{18} n/m$^2\cdot$s, 500 W, 5.5×10^5W/m^2, and 65℃, respectively, at the axial position of the highest neutron flux at the start of irradiation. Vehicle RERTR-1 was irradiated for 94 effective full-power days (EFPD), and RERTR-2 was irradiated for 232 EFPD, achieving (calculated) ^{235}U burnups of about 40at.% and 70at.%, respectively. Thereafter, post-irradiation examinations of the microplates were performed, primarily using a scanning electron microscope.

3. Results and Discussions

Fig. 1 shows scanning electron micrographs of fracture surfaces and apparent fission gas bubble size distribution in the fuel meat of the atomized U-10wt.%Mo microplate at 40at.% burn-up. The fuel-matrix interaction layer of the atomized spherical particles is uniform and in the range of ~2 μm in thickness at 40at.% burn-up. Fission gas bubbles generally appear to be concentrated on the primary grain boundaries almost exclusively. The maximum and the average bubble diameter of atomized U-10wt.%Mo microplates are approximately 0.8 μm and 0.2 μm, individually. The atomized powder has a lower bubble population density of about 2.6×10^{11}/m^2. Scanning electron micrograph and apparent fission gas bubble size distribution of fracture surfaces in the fuel meats of the atomized U-10wt.%Mo microplates at 70at.% burn-up are shown in Fig. 2. There are fission gas bubble-free zones of ~35% in area fraction and 5~10 μm in zone size in the atomized particles. Gas bubble-free zones, not entering the stage of bubble formation, are located primarily around the perimeter rather than around the center of the atomized particle. Small bubble-free zones sometimes formed in bubble-rich zones in places. The bubble size and the population density increase greatly, as the burn-up of the microplates increases from 40at.% to 70at.%. The maximum bubble diameter and the average bubble diameter of atomized fuel particles are approximately 0.7 μm and 0.2 μm, respectively. The bubble population density of the atomized fuel powder is about 1.4×10^{12}/m^2. A back-scattered scanning electron image of an atomized U-10wt.%Mo microplate at 70at.% burn-up is also shown in Fig. 3-(a). The average size of bubble-free islands of 3~10 μm is almost similar to the homogenized grain size of 5~10 μm in the fuel meat at 40at.% burn-up. Fission gas bubbles are associated with a granular microstructure that appears to have nucleated along grain boundaries and then grow continuously toward the center of the grains, leaving many bubble-free zones within the grain. A somewhat different picture is shown in Fig. 3-(b), where the scanning electron microscopy samples are polished. The granular appearance of the fracture surfaces, which indicated grain refinement, is no longer visible and a more representative gas bubble morphology can now be described. There are gas bubbles, as well as the grain refined microstructure, with the lower Mo segregated γ-U phase that formed during solidification in the atomization process [12]. The bubble-free areas, on the other hand, are the high Mo γ-U phase fraction that apparently does not recrystallize at 70at.% burnup and does not develop visible gas bubbles.

Scanning electron micrograph and apparent fission gas bubble size distribution of fracture surfaces in the fuel meats of the ground U-10wt.%Mo microplates at 70at.% burn-up are shown in Fig. 4. The ground particles do not show small and uniform bubbles in grains. There are fission gas bubble-free zones much less than 10% and smaller than 3 μm in zone size in the ground particles. The maximum bubble diameter and the average bubble diameter of ground fuel particles are approximately 1.0 μm and 0.3 μm, respectively. The bubble population density of the ground fuel powder is about $2.6 \times 10^{12}/m^2$.

The in-reactor performance of U-10wt.%Mo microplates, irrespective of powder kind, shows stable irradiation behavior even at high burn-up, similar to that of U_3Si_2 [1-5]. The U-10wt.%Mo dispersion fuels show no indication of breakaway swelling [16]. A U-10wt.%Mo alloy can exist in two states: the heterogeneous state appears to be stable at room temperature; a homogeneous solution of Mo in γ-U appears to be stable above 570℃. It can crystallize either in the form of a supersaturated α solid solution or in the form of $\acute{\gamma}$-U_2Mo phase. An as-cast U-10wt.%Mo ingot was heat-treated in a vacuum for 100 hrs at 900℃ to ensure compositional homogeneity, and then quenched to form a metastable γ-U phase [9]. The mechanically ground powder was prepared from the as-cast ingot contained some αU around the grain boundaries, which must have come from the partial decomposition of the metastable γ-U phase during hot rolling, performed at 500℃ for 2 hrs, in contrast to atomized U-10wt.%Mo powder. There is no evidence of interlinking of the relatively uniformly distributed fission gas bubbles, even in the ground powder. This indicates that there is no obvious evidence of a two-phase microstructure in any of the irradiated microplates, irrespective of powder kind. Therefore it seems that the partial decomposition of the γU phase in the ground powder which occurs during fabrication is probably reversed early during irradiation. It has been well established that a transition (αU+U_2Mo $\rightarrow$ γ solid solution) is caused by irradiation, even when the mean temperature of the specimen does not exceed 100~150℃. Such an effect has been reported in the literature as owing to fission-spike mixing at high fission rates. The removal of heat from the neighborhood of such a thermal spike is accomplished in a very short period of time. As a result, the formation of a spontaneous crystallization centre in the thermal spike area seems improbable and the crystallizing volume acquires the structure of one of its neighboring lattices. Hence, as the diffusion of the molybdenum atoms and the alignment to the average molybdenum composition in the U-10wt.%Mo particles develop further in spite of an average fuel center temperature of 65℃, a transition into the γU phase takes place.

The atomized U-10wt.%Mo dispersion fuel has a finer and more uniform size distribution of fission gas bubbles than the mechanically ground dispersion fuel. The possible reasons are supposed as follows. The atomized particles do not have prominent deformation damage formed during the powdering process. Whereas, the ground particles have severe deformation damage with a high dislocation density formed during the mechanical comminution process. During rolling or irradiation of the ground U-10wt.%Mo dispersion fuels, the dislocations interact and tend to cluster into arrangements of high dislocation density that are separated by regions with a relatively low dislocation density [17-19]. The clustering of dislocations is a general observation in deformed polycrystalline alloys [19-21]; the typical dislocation configurations are dislocation tangles, two-dimensional dislocation boundaries (or walls), and three-dimensional dislocation cell structures. The different dislocation configurations are derived from energy minimization, where glide-dislocation configurations increasingly approach the minimum energy per unit length of dislocation line as the dislocation density - and hence interaction between dislocations - increases. Strain-free blocks within a crystal are formed by the spatial rearrangement of dislocation into lower-energy arrays by polygonization. Recrystallization generally occurs in all of the alloys, resulting in an average grain size of submicron. It is promoted by increasing the amount of cold work, as the activation energy for recrystallization is a function of the amount of deformation. The heavily deformed areas of the ground U-10wt.%Mo have a highly stored strain energy with a high degree of deformation, that is, initial dislocation density. They tend to nucleate and grow into subgrains around the grain boundaries during hot rolling or irradiation. Hence, the onset of grain subdivision in the ground U-10wt.%Mo occurs at a lower burn-up, compared with the atomized powder. The ground U-10wt.%Mo at 70at.% burn-up has a granular appearance all over the particles, suggesting a grain refinement of about 0.8 μm in subgrain size similar to the coarse fission gas bubble size. However, the atomized powder at 70at.% burn-up shows a partial granular appearance and bubble-free zones. It indicates that a severe grain subdivision by recrystallization did not yet occur in some regions of the atomized particles.

Irradiation-induced recrystallization and enhanced bubble growth on the newly formed grain boundaries were also observed for UO_2 power reactor fuels. Recrystallization and intergranular bubble growth have been definitively confirmed for UO_2 fuels. "Subdivision" of the original grains has been observed in high burn-up uranium dioxide. The peripheral region of LWR fuel pellets reveals an increasingly porous microstructure with burn-up [22-26]. Observation of this "rim effect" showed that an extremely fine-grained structure formed by subdivision of the original fuel grain was associated with the porous microstructure. The large fission gas bubbles generally appear to be forming at the subgrain boundaries, as the recrystallized boundaries around the grain boundaries have very high fission gas mobility. Upon grain subdivision in U-10wt.%Mo fuel particles, fission gas atoms diffuse from the bulk to the grain faces, where they accumulate in gas bubbles that grow until grain-face saturation occurs. The coarse bubbles shown in Fig. 2 and Fig. 4 are associated with grain-corner bubbles formed by the intersection of grain edges within the subgrain boundary structure. Fission gas that collects along the grain edges vents upon intersection with these "dead-end" nodes as they continue to collect additional gas. The large bubbles in the grain subdivision region of U-10wt.%Mo fuel particles are due to the accumulated gas in the "dead-end" nodes. Moreover, in the case where fabrication-induced damage leads to an earlier onset irradiation-induced recrystallization, the higher gas bubble population density in the ground U-10wt.%Mo microplates will occur at a larger bubble size. This is because of the shorter time that bubbles have to accumulate and grow on the subgrain boundaries. Subsequent to irradiation-induced recrystallization, the growth rate of gas bubbles on the recrystallized grain surface increases. Fission-gas bubbles nucleate at the newly formed boundaries and then grow at an accelerated rate relative to that of fission-gas bubbles in the bulk material. They accelerate the growth rate of gas bubbles of the ground U-10wt.%Mo particles at a lower burn-up against the atomized particles. The fine bubbles shown in Fig. 2 and Fig. 4 are associated with grain boundary bubbles. About one-third of the area of the bubble-free zones in the atomized fuel cross-sections at 70at.% burn-up, which appears to be associated with the higher segregated Mo fraction of the γ-U phase, indicates that a considerable amount of fuel particles do not start grain refinement. The bubble sizes are very fine below size resolvable experimentally by scanning electron microscopy (SEM) [27]. In addition, a slower progress of the grain refinement on the atomized U-10wt.%Mo microplates induces lower gas bubble mobility and growth rate, which eventually result in finer size distribution than the ground microplates. Hence, the atomized U-10wt.%Mo microplates have lower population density of gas bubbles than the ground microplates having coarser size distribution, as the fine gas bubbles generally have higher density of gas atoms and higher internal pressure than the coarse gas bubbles [2].

4. Conclusions

In order to examine the in-reactor performance of very-high-density dispersion fuels for high flux performance research reactors, U-10wt.%Mo dispersion fuels containing centrifugally atomized and mechanically ground powder were irradiated at low temperature.

(1) The U-10wt.%Mo dispersion fuels show stable in-reactor irradiation behavior, indicating maintenance of the metastable cubic γ-U phase.

(2) The atomized U-10wt.%Mo fuel particles have an overall finer and more uniform fission gas bubble size distribution of gas bubbles relative to the mechanically ground fuel particles.

(3) The atomized U-10wt.%Mo fuel particles at 70at.% burn-up show a considerable amount of fission gas bubble-free zones, in contrast to the ground fuel cross-sections containing very small fission gas bubble-free zone.

(4) The possible reasons for different in-reactor behavior of the atomized powder relative to ground powder are supposed as follows.

i) The atomized particles do not have severe deformation damage with a high dislocation density formed during the mechanical comminution process.

ii) The grain refinement and the gas bubble formation in the ground particles occur at a lower burn-up than those in the atomized particles.

iii) The atomized particles contain 2 sub γ phase fractions as a result of segregation (coring).

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