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Phase Stability Studies of Unirradiated Al - U-10wt.%Mo Fuel at Elevated Temperature

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Abstract

The phase stability of atomized U-10wt. %Mo powder and the thermal compatibility of dispersed fuel meats at 400°C and 500°C have been characterized. Atomized U-10Mo powder has a good γ-U phase stability, and excellent thermal compatibility with aluminum matrix in a dispersion fuel. It is thought that the good phase stability is related to the large supersaturation of Mo atoms in the atomized particles. The reasons for the excellent thermal compatibility have been considered to be as follows. Before thermal decomposition of γ-U in particle, supersaturated Mo atoms at γ-U grain boundaries inhibit the diffusion of Al atoms. After thermal decomposition of γ-U into α-U and U_2Mo, the intermetallic compound of U_2Mo seems to retard the penetration of Al atoms. The penetration mechanisms of aluminum atoms in the atomized particles are assumed to be classified as (a) diffusion through the reacted layer between fuel particles and Al matrix leaving a kernel-like unreacted island and (b) diffusion along grain boundaries showing several unreacted islands and more reacted regions.

1. Introduction

The conversion from high enriched uranium (HEU) to low enriched uranium (LEU) for use in research reactor fuel requires a large increase in uranium per unit volume to compensate for the reduction in enrichment. The relatively high density compound with a uranium density of 11.6 g-U/cm^3, U_3Si_2 was found to possess very stable irradiation behavior. However, fabrication limits allow loading higher than 6 g-U cm^{-3} [1-5]. Hence, in the renewed fuel development for research and test reactors, attention has shifted to high density uranium alloys. 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 [6]. It has been reported that high density atomized U-Mo powder prepared by rapid solidification has the metastable isotropic γ-U phase supersaturated with substitutional molybdenum, especially in U-10wt.%Mo alloy [7]. If indeed the centrifugally atomized U-Mo powder can retain this gamma phase during fuel element fabrication and irradiation, and if it is compatible with aluminum matrix, the uranium alloy would be a prime candidate for dispersion fuel for research reactors.

In this study U-10wt.%Mo alloy powder which has high density above 15 g-U cm^{-3} was prepared by rotating-disk centrifugal atomization. The fuel rods were made by extruding a blend of atomized U-Mo powder and aluminum powder. The γ-U phase stability of atomized U-10wt.%Mo powder and the thermal compatibility of atomized U-10wt.%Mo-Al dispersion fuel during annealing at elevated temperatures have been examined, compared with the thermal compatibility of U_3Si_2 dispersion fuel.

2. Experimental Procedure

The molten U-10wt.%Mo alloy after induction-melting in graphite crucible coated with a high-temperature-resistant ceramic was fed through an orifice onto a rotating graphite disk in an argon atmosphere, respectively. In order to obtain the desired size distribution and shape, the atomization parameters were adjusted [8]. The atomized powder was collected in a container at the bottom of the funnel-shaped chamber. Dispersion fuel rods were prepared by extruding the blended powders of U-10wt.%Mo and aluminum at a working temperature of 400°C. Atomized powder and compatibility specimens were annealed for incremental times at 400°C and 500°C. After each annealing interval, the dimensional changes of the specimens were measured.

The samples were polished to 0.3 μm diamond paste, and examined by a scanning electron microscope (SEM) to characterize the microstructure of the atomized particles and the fuel meats. Electron-probe micro-analysis (EPMA), energy dispersive spectrometry X-ray analysis (EDX), and X-ray diffraction analysis (XRD) using Cu K_α radiation were also used to determine the chemical composition and the phase of the samples.

3. Experimental Results

The γ-U phase of U-10Mo powder annealed at 400°C upto 100 hours was retained [7]. However, the micrograph of the U-10Mo powder after 350 hours showed fine γ-U cell structure with decomposed α-U and γ'-U_2Mo phases around the grain boundary (Fig. 1-(a)). The X-ray diffraction pattern showed that the major phase of U-Mo powder after 350 hours was γ-U phase. Some γ-U phases were decomposed as coarse α-U and γ'-U_2Mo phase after annealing. The micrograph of U-10Mo powder annealed at 500°C after

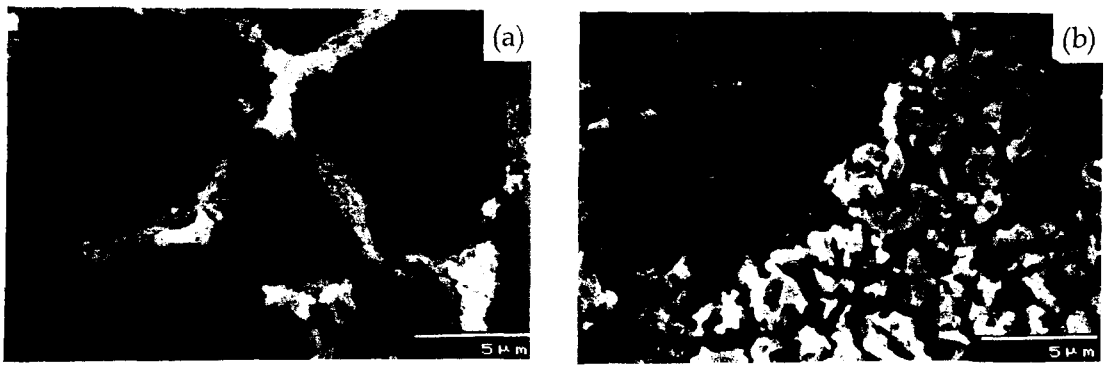

Fig. 1. Back-scattered scanning electron images of the atomized U-10Mo powder after annealing; (a) at 400°C after 350 hours, (b) at 500°C after 500 hours.

24 hours showed fine γ-U cell structure with the fibric structure of decomposed phases. Most γ-U grains of atomized U-10Mo powder after annealing for 500 hours were already decomposed as coarse α-U and γ'-U_2Mo phase (Fig. 1-(b)). The X-ray diffraction pattern of the atomized U-10Mo powder showed that half of the γ-U phase of U-10Mo powder after 100 hours remained as it were; however the greater part of the γ-U phases of U-10Mo powder after 500 hours was decomposed as the α-U phase and the γ'-U_2Mo phase, including some γ-U phase. Fig. 2 shows the dimensional changes of the Al - 45vol.% U-10Mo fuel samples annealed at 500°C for various times. Half of the swelling in the fuel samples at 500°C occurred within 10 hours, so the swelling appeared to reach a plateau gradually with annealing time. The intermediate phase layer formed around the interface between U-10Mo fuel and aluminum matrix increased in proportion to square root with annealing time. The volume change of the dispersion fuels specimens was less than that of similar Al - U_3Si_2 dispersion fuel specimens for the same time, independent of annealing time [9]. Even after annealing for 500 hours the Al - U-10Mo dispersion fuel samples did not show a large volume increase, up to 34%. Back-scattered scanning electron images of the fuel samples after annealing at 500°C for 200 hours are shown in Fig. 3. The region of the fuel samples imaged in Fig. 2 may be divided into two general areas: (a) white islands, (b) dark-grey regions. Metallographic examinations of the samples showed that most particles exhibited an irregular interface and a rim of an intermediate phase. Fine particles had several unreacted islands (white), and reacted intermetallic compounds (dark-grey) as a matrix in the U-10Mo particles. The fuel meats showed two aspects of the penetration with aluminum atoms. The particles were composed of a considerable amount of reacted areas around the circumferential part and generally had a "kernel-like" structure with an unreacted island, and several unreacted islands. The penetration degree of most atomized particles with a kernel-like structure did not reach half of particle cross-section despite long annealing at elevated temperatures. The area scan analyses of the U-10Mo samples annealed for 200 hours,

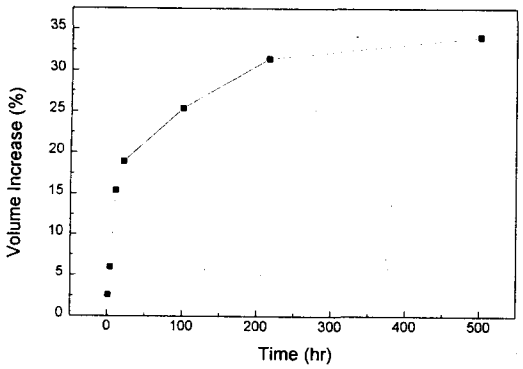

Fig. 2. The dimensional increase of the Al - 45vol.% U-10Mo fuel samples at at 500°C for various times.

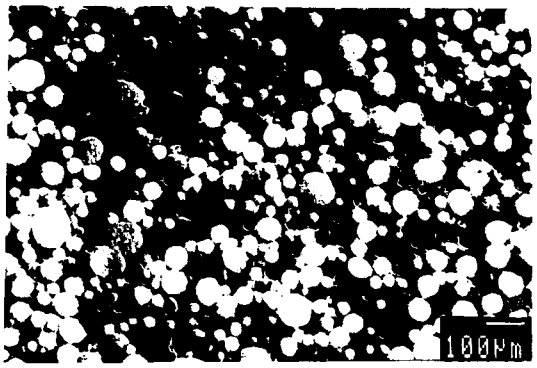

Fig. 3. Back-scattered scanning electron images of the fuel samples after annealing at 500°C for 200 hours.

by using energy dispersive X-ray spectroscopy (EDX), indicated that white regions were composed of 81 at.%U, 17 at.%Mo and 2 at.% Al, whereas dark-grey regions consisted of 20at.%U, 4 at.%Mo and 75 at.%Al, that is, (U,Mo)Al_3. Uranium-aluminide with a small amount molybdenum, mainly UAl_3 was formed in the U-Mo particles due to the diffusion of Al atoms. Electron probe micro-analysis (EPMA) traces of the fuel sample also confirmed that there was some formation of intermediate phase regions between U-Mo particle and Al matrix. The atomized U-10Mo particles had more unreacted regions, compared with the atomized U_3Si_2 particle. This led to a volume change of 31% which was less by 8% than that of the atomized U_3Si_2 fuel samples (Fig. 1) [9].

4. Discussion

By rapid solidification of U-Mo alloy from the melt, the centrifugally atomized powder retained a gamma phase as metastable state. However, the γ-U phase of U-10Mo alloy annealed below the eutectoid temperature (560°C) had a tendency to be decomposed into a thermodynamically stable lamellar structure including α-U and γ'-U_2Mo phases [10]. A scanning electron micrograph carried out on the U-10Mo powder annealed at 400°C upto 100 hours, illustrated that U-10Mo particle have a fine grain structure below 3 ㎛ in size with microsegregation of molybdenum atoms [7]. These results originated from the supersaturation of Mo in the metastable γ-U solid solution of U-10Mo alloy. Large content of substitutional Mo atoms with low diffusivity caused the migration of U atoms difficulty, and inhibited the decomposition and the coarsening of γ-U. This confirmed that the γ-U phase of atomized U-10Mo powder could be retained at 400℃ for an extended time, presumably because the diffusion-controlled transformation is retarded at

increased Mo content. Fine laminae were nucleated primarily at γ-U grain boundaries at 400°C (Fig. 1). γ-U in the grains is decomposed as α-U and γ-U with higher Mo content than γ'-U_2Mo phase as would be expected for normal eutectoid reaction [11-12]. Then transformation occurred continuously involving the formation of ordered intermediate phase (γ'). However, the decomposition of γ-U at 500°C took place primarily by the cellular mechanism [13].

As the aluminum reacts with the fuel, the fuel's volume increases due to the difference between densities of the original particle and reaction product, and due to the pores produced by the Kirkendall effect [14]. The volume of the U-10Mo dispersion fuel sample annealed at 400℃, remained the same even after 2000 hours anneal, without formation of intermediate phase layer [15]. However, the swelling of this U-10Mo fuel sample after annealing at 500℃ for 200 hours was considerable up to 31%, with formation of (U,Mo)Al_3 of 8 μm in thickness. The increase in annealing temperature accelerated the penetration rate of aluminum atoms in the fuel particles. The U-10Mo dispersion fuel samples did not show a large volume increase, compared with that of U_3Si_2 dispersion fuel specimens. The possible reasons for the excellent thermal compatibility could be supposed as follows. Such results originated from larger atomic radius and lower diffusivity of supersaturated substitutional molybdenum atoms in the metastable γ -U solid solution, relative to those of silicon atoms. Before phase decomposition of γ-U, substitutional Mo atoms with large content, supersaturated especially at γ-U grain boundaries, inhibited the diffusion of Al atoms along grain boundaries. After phase decomposition of γ-U into α-U and U_2Mo, as separated U-10Mo particles were mainly composed of U_2Mo, the intermetallic compound retarded the penetration of Al atoms. These factors resulted in excellent thermal compatibility of U-10Mo with aluminum at elevated temperature. In the initial stage of annealing at 500℃, atomized U-10Mo particles showed a thin kernel-like intermediate phase layer around the perimeter. However, after the middle stage of annealing at 500℃, atomized U-10Mo particles sometimes began to have several unreacted islands, showing more reacted regions.

5. Conclusions

The phase stability of atomized U-10wt.%Mo powder and the thermal compatibility of dispersed fuel meats at elevated temperatures were characterized.

1) After annealing at 400°C for 350 hours atomized U-10wt.%Mo powder showed fine γ-U grain structure with decomposed α-U and γ'-U_2Mo phases only around the grain boundaries. However, the greater part of γ-U phases of atomized U-10wt.%Mo powder annealed at 500°C for 500 hours was already decomposed as α-U and γ'-U_2Mo phases, with some retained γ-U phases.
2) Atomized U-10Mo powder had an excellent thermal compatibility with aluminum matrix in a dispersion fuel.
3) The reasons for the excellent thermal compatibility were considered to be as follows. Before thermal

decomposition of γ-U in particle, supersaturated Mo atoms at γ-U grain boundaries inhibited the diffusion of Al atoms. After thermal decomposition of γ-U into α-U and U_2Mo, the intermetallic compound of U_2Mo seemed to retard the penetration of Al atoms.

4) The penetration mechanisms of aluminum atoms in the atomized particles were assumed to be classified as (a) diffusion through the reacted layer between fuel particles and Al matrix leaving a kernel-like unreacted island and (b) diffusion along grain boundaries showing several unreacted islands and more reacted regions.

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