Improvement of Silicide Coating Method as Diffusion Barrier for U-Mo Dispersion Fuel

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1. Introduction

Gamma phase U-Mo alloys are regarded as one of the promising candidates for advanced research reactor fuel when it comes to the irradiation performance [1]. However, it has been reported that interaction layer formation between the U-Mo alloys and Al matrix degrades the irradiation performance of U-Mo dispersion fuel. The excessive interaction between the U-Mo alloys and their surrounding Al matrix lead to excessive local swelling called 'pillowing'. For this reason, KAERI suggested several remedies such as alloying U-Mo with Ti, or Al matrix with Si. In addition, silicide or nitride coatings on the surface of U-Mo particles have also been proposed to hinder the growth of the interaction layer [2-5].

In this study, centrifugally atomized U-Mo-Ti alloy powders were coated with silicide layers. The coating process was improved when compared to the previous coating in terms of the ball milling and heat treatment conditions. Subsequently, silicide coated U-Mo-Ti powders and pure aluminum powders were mixed and made into a compact for the annealing test. The compacts were annealed at 550℃ for 2hr, and characterized using scanning electron microscopy (SEM) and energy dispersive x-ray spectroscopy (EDS).

2. Experimental Procedures

U-7wt%Mo-1wt%Ti alloy powders were produced by a centrifugal atomization method. Superheated molten uranium-alloy was fed through a small nozzle onto a graphite disk spinning at about 30,000 rpm, and liquid alloy droplets were then spread from the disk by centrifugal force and cooled in an argon atmosphere [6]. The sizes of the atomized U-7wt%Mo-1wt%Ti powders were varied from 45-150μm, and only powders from 105-150μm were used in this study.

The sizes of the Si powders used to mix with U-7wt%Mo-1wt%Ti powders were less than 5μm, and the mixing ratio between the Si powders and U-7wt%Mo-1wt%Ti powders were varied to optimize the mixing process for homogeneous coating. Si powders, which were 45μm in size originally, were grounded by a ball milling process for minimizing the size. Smaller Si powders could easily make contact with larger U-7wt%Mo-1wt%Ti powders owing to its larger surface area. For the same reason, the mixing ratio was also modified, and modified experimental conditions for the silicide coating are presented in Table. 1.

Conditions	Previous	Present
Mixing Ratio (U : Si)	1:2	5:1
Si powder size	≤20	≤5

Table. 1. Modified experimental conditions for silicide coating

Through the mixing process, smaller Si particles made mechanically good contact and surrounded bigger U-7wt%Mo-1wt%Ti particles. After the mixing process, the mixed powders were heat-treated in a vacuum furnace at T (T=800, 900, 1000℃) for 30mins to form silicide-coating layers on the surface of the U-7wt%Mo-1wt%Ti powders. Previously, mixed powders were heat-treated at 1000℃ for 1hr under a vacuum of about $3 \cdot 10^{-5}$torr. In this study, the duration for the heat-treatment was shortened to 30 min to reduce the thickness of the silicide coating layer. In addition, the heat-treatment temperature was also varied to 800℃, 900℃ and 1000℃ to analyze the relationship between the heat-treatment temperature and the thickness of the silicide layer.

After the heat-treatment process, the silicide coated U-7wt%Mo-1wt%Ti powders were mixed with pure aluminum powders and made into a compact. Subsequently, the compacts were annealed at 550℃ and 600℃ for 2hr, respectively, and the aspects of the interaction layer growth between the silicide coated U-7wt%Mo-1wt%Ti and the Al matrix were analyzed by SEM and EDS.

3. Results and Discussion

From the previous research, it was noted that the previous coating conditions as presented in Table 1, resulted in poor homogeneity and almost 20μm thick silicide coating layers, as shown in Fig. 1. The poor homogeneity was induced by non-contact between Si powders and U-7wt%Mo-1wt%Ti powders due to the non-optimized mixing ratio and sizes of the Si powders. Furthermore, too high a heat-treatment temperature (1000℃) was suspected to be the reason for the thick silicide coating layers.
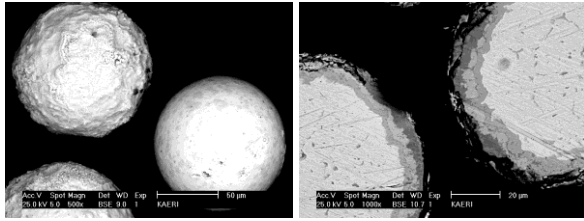
Fig. 1. Micrographs of (a) the surface, and (b) the cross-section of the silicide-coated U-7wt%Mo-1wt%Ti powders from the previous coating method.

In this sense, the sizes of the Si powders, the mixing ratio and the temperature for the heat-treatment were modified. The results from the modified coating process are shown in Fig. 2, and showed the improved homogeneity and uniformity of the silicide coating layers. In addition, it was shown that the thinner silicide coating layers could grow at a lower heat-treatment temperature.

In the case of the 800℃ heat-treated sample, the thickness of the silicide coating layers was less than 1μ m. However, few U-7wt%Mo-1wt%Ti particles seemed to be uncoated, or the silicide coated layers were too thin to analyze by SEM. As the heat-treatment temperature increased to 900℃, the silicide coating layers were uniformly and homogeneously formed on the surface of U-7wt%Mo-1wt%Ti powders, and its thickness was around 4-5μm. For the 1000℃ heat-treated sample, the thickness of the silicide layers was around 5-20μm thick and its surface was too rough although the silicide coating layer was uniformly grown.
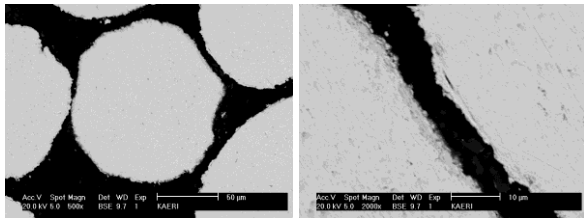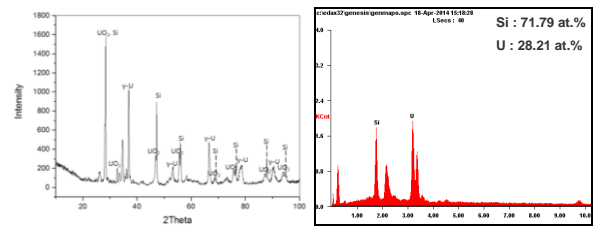

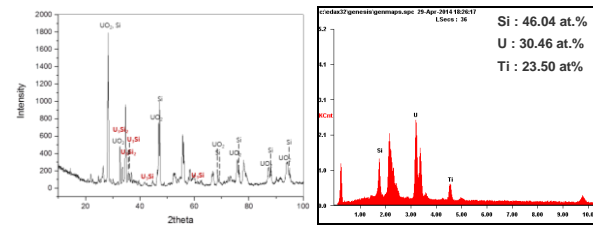(a) Heat-treated at 800℃ for 30min.
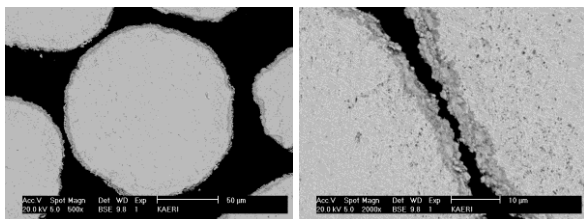
(b) Heat-treated 900℃ for 30min.
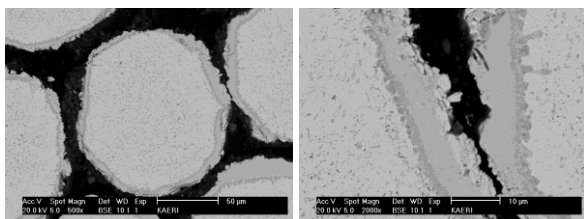
(c) Heat-treated 1000℃ for 30min.

Fig. 2. The cross-sectional micrographs of silicide coated U-7wt%Mo-1Ti powders at various temperatures for 30min, respectively.

As shown in figure 3, the phases and chemical compositions of the silicide layers were analyzed by XRD and EDS, respectively. Unfortunately, we could not find any peak of the silicide layers from the XRD patterns in the case of the 800℃ heat-treated sample, while in the cases of 900℃ and 1000℃, heat-treated samples showed U_3Si and U_3Si_2 peaks. It was noted that the compositions of Ti in the silicide layers was also very low for the 800℃ heat-treated sample while other samples showed relatively higher Ti compositions in the silicide layers.
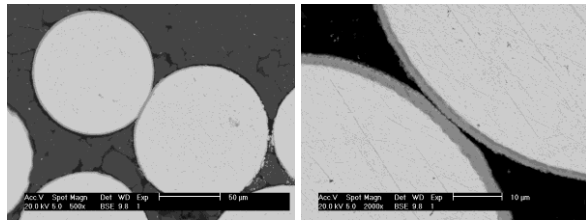

(a) Heat-treated at 800℃ for 30min.
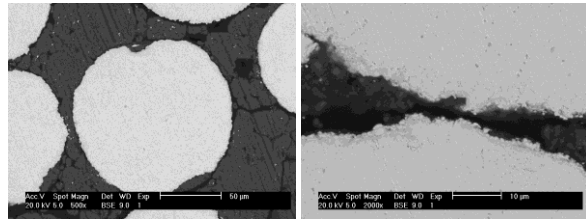

(b) Heat-treated at 900℃ for 30min.
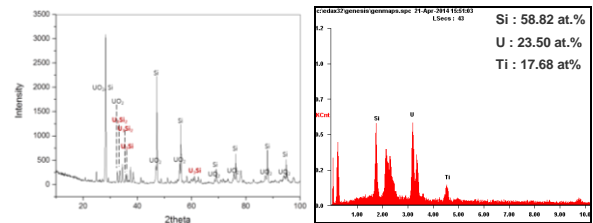
(c) Heat-treated at 900℃ for 30min.

Fig. 3. X-ray diffraction patterns and EDS spectra of the silicide coated U-7wtTMo-1wt%Ti powders heat-treated at various temperatures for 30 min, respectively.

Cross-sectional micrographs for the results of the annealing test are presented shown in Fig. 4, including the results of uncoated U-7wt%Mo-1wt%Ti/Al compacts for comparing the effects of silicide coating. In the case of uncoated U-7wt%Mo-1wt%Ti/Al compact, it showed that 5μm thick interaction layers formed at the interface of the uncoated U-7wt%Mo-1wt%Ti/Al matrix. For the silicide coated U-7wt%Mo-1wt%Ti/Al compacts, on the other hand, the growth of the interaction layer was effectively suppressed regardless of the heat-treated temperatures.
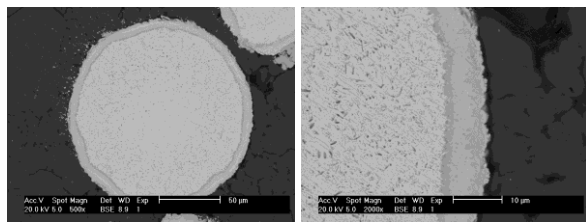
(a) Uncoated U-7wt%Mo-1wt%Ti/Al matrix compact.

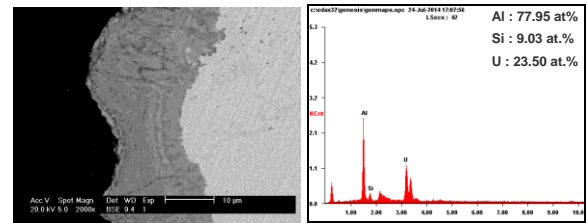(b) Silicide coated (heat-treated at 800℃ for 30min) U-7wt%Mo-1wt%Ti/Al matrix compact.
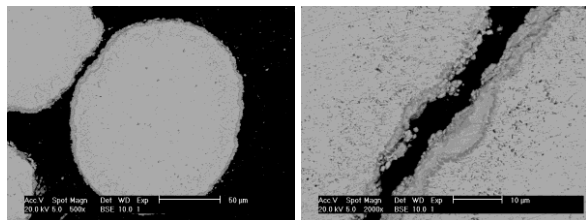
(c) Silicide coated (heat-treated at 900℃ for 30min) U-7wt%Mo-1wt%Ti/Al matrix compact..
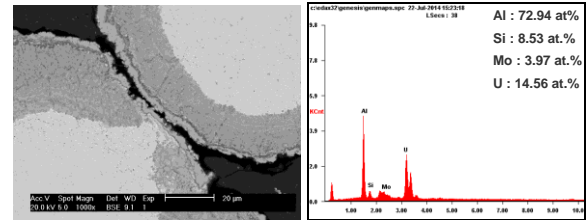

(d) Silicide coated (heat-treated at 1000℃ for 30min) U-7wt%Mo-1wt%Ti/Al matrix compact.

Fig. 4. The cross-sectional micrographs of the silicide coated U-7wt%Mo-1wt%Ti/Al compacts annealed at 550℃ for 2 hours, respectively.

However, as the annealing temperature increased to 600℃, silicide coated layers were not able to inhibit the formation of interaction between U-7wt%Mo-1wt%Ti and the Al matrix, as shown in Fig. 5. It was interesting that for the silicide coated (heat-treated 900℃ and 1000℃) U-7wt%Mo-1wt%Ti/Al matrix compacts, the compositions of the interaction layers were ternary or quaternary phases of U, Mo, Si and Al, as shown in Figs. 5(b) and (c). It seemed that, during the annealing test, Al penetrate d the silicide layers, which had a thermal induced crack on the surface, and formed a complex interaction phases, as shown in Fig. 5(a). However, an in-depth study should be followed to characterize the interaction phases, and to analyze the phase transformation mechanism.
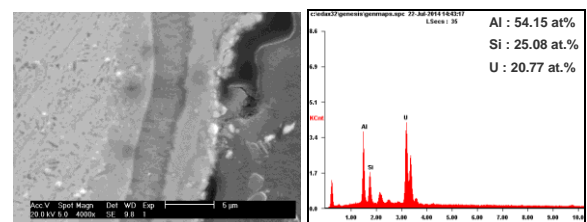

(a) Silicide coated (heat-treated at 800℃ for 30min) U-7wt%Mo-1wt%Ti/Al matrix compact.
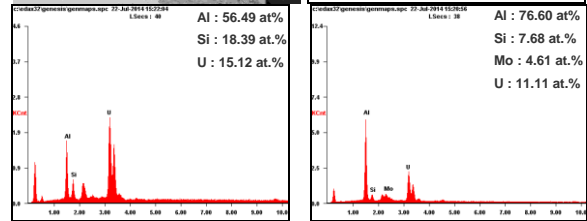
(b) Silicide coated (heat-treated at 900℃ for 30min) U-7wt%Mo-1wt%Ti/Al matrix compact.

(c) Silicide coated (heat-treated at 1000℃ for 30min) U-7wt%Mo-1wt%Ti/Al matrix compact..

Fig. 5. EDS spectra and cross-sectional images of annealed silicide-coated U-7wt%Mo-1wt%Ti/Al compacts at 600℃ for 2 hours.

4. Conclusions

1. Uniform, homogeneous, thickness controllable silicide layers were successfully coated on the surface of U-7wt%Mo-1wt%Ti powders.
2. U_3Si, U_3Si_2 silicide layers formed on the surface of U-7wt%Mo-1wt%Ti powders, and were identified by XRD and EDS analyses.
3. The silicide coating layer effectively suppressed the growth of interaction layers at 550℃ for 2 hours, while Al diffused into the U-7wt%Mo-1wt%Ti particles at 600℃ for 2 hours.

ACKNOWLEDGMENTS

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