A Study on Silicide Coatings as Diffusion barrier for U-7Mo Dispersion Fuel

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1. Introduction

Fuel development has been the US Reduced Enrichment for Research and Test Reactors (RERTR) program since 1978. For the LEU conversion of HEU using research and test reactors, uranium density in the fuel meat has to be increased to compensate the reduced U-235 enrichment. 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 [1,2]. 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 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 [3-6].

In this study, centrifugally atomized U-7Mo alloy powders were coated with silicide layers at 900 $^{\circ}$ C for 1hr. U-Mo alloy powder was mixed with MoSi₂, Si and ZrSi₂ powders and subsequently heat-treated to form uranium-silicide coating layers on the surface of U-Mo alloy particles.

Silicide coated U-Mo powders and characterized using scanning electron microscopy (SEM), energy dispersive x-ray spectroscopy (EDS) and X-ray diffractometer (XRD).

2. Experimental Procedures

The silicide coating method is presented in Fig. 1. U-7Mo alloy powders were produced by a centrifugal atomization method in KAERI. 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 [7]. The sizes of the atomized U-7Mo powders were varied from 45-150 μ m, and only powders from 65-150 μ m were used in this study.

Three kinds of powders were $MoSi_2$, Si and $ZrSi_2$. The sizes of the $MoSi_2$, Si and $ZrSi_2$ powders used to mix with U-7Mo powders were less than 5µm. $MoSi_2$, Si and $ZrSi_2$ powders, which were 45µm in size originally, were grounded by a ball milling with zirconia balls for minimizing the size. Smaller powders could easily make contact with larger U-7Mo powders owing to its larger surface area. U-7Mo alloys were mixed with the $MoSi_2$, Si and $ZrSi_2$ powders by using a v-mixer with zirconia balls for 18 hours. After the mixing process, mixed powders were annealed at 900°C for 1 hour under a vacuum of about 6·10⁻⁵ torr to form silicide-coating layers on the surface of the U-7Mo powders.

The microstructures of silicide-coated powders were observed by SEM, and silicide coating layers were characterized by EDS and XRD.

Sample	Heat-treated conditions		
	Temperature	Duration	Mixing Ratio(vol.)
(a) MoSi ₂	900 ℃	1hr	1:5
(b) Si	900 ℃	1hr	1:5
(c) ZrSi ₂	900 ℃	1hr	1:5

Table. 1. Experimental conditions for the silicide coating.

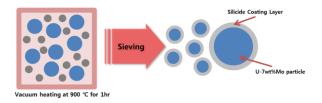


Fig. 1. A schematic diagram of silicide coating method.

3. Results and Discussion

The silicide-coated U-7Mo particles by mixing condition in Table .1. U-7Mo powders were mixed with $MoSi_2$, Si and $ZrSi_2$ powders in the volume ratio of 1:5 and annealed at 900 °C for 1hr.

General silicide coating layers were formed on the surface of U-7Mo particles to a thickness of about $2\sim3\mu$ m, and various U-Si alloy phases were formed as shown in Fig. 2. MoSi₂ coating layers were formed on the surface of U-7Mo particles to a thickness of about $2\sim3\mu$ m, as shown in Fig. 2. (a). Si coating layers were formed on the surface of U-7Mo particles to a thickness of about $2\sim3\mu$ m, as shown in Fig. 2. (b). ZrSi₂ coating layers were formed on the surface of U-7Mo particles to a thickness of about $2\sim3\mu$ m, as shown in Fig. 2. (b). ZrSi₂ coating layers were formed on the surface of U-7Mo particles to a thickness of about $2\sim3\mu$ m, as shown in Fig. 2. (c).

The surface of a silicide coated particle was very rough and Silicide powder attached to the surface of the coating layer.

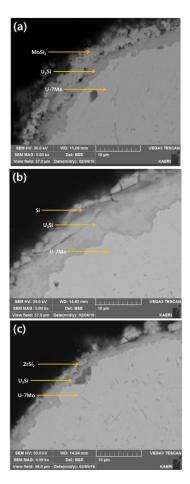


Fig. 2. Cross-sectional SEM microstructures of (a) MoSi₂, (b)Si and (c)ZrSi₂ coated on a U-7Mo particle

The XRD analysis of the coating layers showed that, they consisted of compounds such as U_3Si_2 , USi_2 , as shown in Fig. 3. After the heat-treatment, unreacted MoSi₂, Si and U_3Si mixed-layer formed on the surface of U-7Mo particles, and thickness of the layer was less than 3µm in the case of (a) MoSi₂, (b) Si. On the other hand, in the case of (c) ZrSi₂ formed multi-phase. Those coating layers were composed of U_3Si_2 , USi₂, from the outside to the inside of U-7Mo particles, respectively.

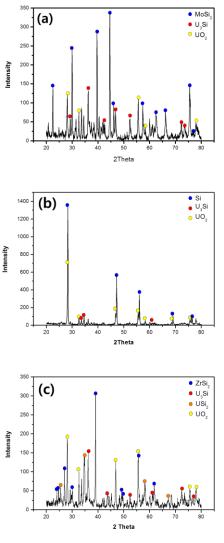


Fig. 3. X-ray diffraction patterns of (a) $MoSi_2$, (b) Si and (c) $ZrSi_2$ coated on a U-7Mo particle

3. Conclusions

- The MoSi₂ coating layers were formed on the surface of U-7Mo particles to a thickness of about 2~3μm. The Si coating layers has a thickness of about 2~3μm. The ZrSi₂ coating layers has a thickness of about 1~2μm.
- 2. The surface of a silicide coated particle was very rough and silicide powder attached to the surface of the coating layer.
- 3. The XRD analysis of the coating layers showed that, they consisted of compounds such as U_3Si_2 , USi_2 .

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