Analyses of Interaction Phases of U-Mo Dispersion Fuel by Synchrotron X-ray Diffraction

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1. Introduction

Gamma phase U-Mo alloys are one of the promising candidates to be used as advanced high uranium density fuel for high power research reactors due to their excellent irradiation performance. However, formation of interaction layers between the U-Mo particles and Al matrix degrades the irradiation performance of U-Mo dispersion fuel. One of the remedies to the interaction problem is a Si addition to the Al matrix. Recent irradiation tests have shown that the use of Al-(2~5wt%)Si matrices retarded the growth of interaction layers effectively during irradiation. Recently, KAERI has proposed silicide or nitride-coated U-Mo fuel for the minimization of the interaction layer growth [1]. The silicide or nitride coatings are expected to act as interdiffusion barriers and their out-of-pile tests showed the improved diffusion barrier performances of the silicide and nitride layers.

In order to characterize constituent phases in the coated layers on U-Mo particles and the interaction layers of coated U-Mo particle dispersed fuel, synchrotron X-ray diffraction experiments have been performed at the ESRF (European Synchrotron Radiation Facility), France as a KAERI-CEA cooperation program.

2. Experimental Procedures

Centrifugally atomized U-7wt%Mo and U-7wt%Mo-1wt%Ti particles with 90~150㎛ in diameter were used for nitride or silicide coating. A vacuum rotator heat treatment furnace was used for nitride coating on the surface of U-Mo particles. The operation conditions were listed in Table 1.

Table 1. The operation conditions of nitride coating.

Parameters		Conditions
Back ground pressure		1×10^{-6} torr
Working pressure		3×10^{-5} torr
N_2-gas flow		80 sccm
Revolution speed		30 rpm
Temperature		1000 ℃
Holding Time	U-7Mo	6 hours
	U-7Mo-1Ti	1~4 hours

For silicide coating on U-Mo particles by vacuum annealing, U-Mo particles and pure Si powder were mixed in a ball mill with a rotation speed of 30 rpm for 8 hours. The mixed powder was annealed at 1000℃ for 1 hour under a vacuum of about 7×10^{-5} torr.

U-Mo/Al dispersion rods were extruded after mixing Al powder and coated U-Mo particles. In order to form the interaction layers, dispersion samples were annealed at 580 ℃ for 1hour in a vacuum furnace.

Cross-sectional microstructures of the specimens were observed by using SEM. To refine the compounds in the coating layers and interaction layers, synchrotron X-ray diffraction patterns from the coated particles and the annealed dispersion samples were analyzed by using the Fullprof program [2].

3. Results and Discussion

Silicide coating layers were formed on the surface of U-7wt%Mo particles with a thickness of about 7~8 ㎛, as in Fig. 1(a). After annealing at 580℃ for 1h, interaction layers were formed as in Fig. 1(b). Fig. 2 shows the silicide coating layers of about 10-15 ㎛ and interaction layers formed on the surface of U-7wt%Mo-1Ti particles. While silicide coating layers were able to inhibit formation of interaction layers, interaction layers formed on uncoated surfaces of the particles.

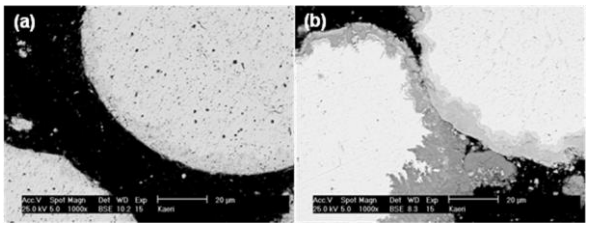

Fig. 1. SEM images of silicide coated U-7wt%Mo particles. (a) as extrude; (b) as annealed

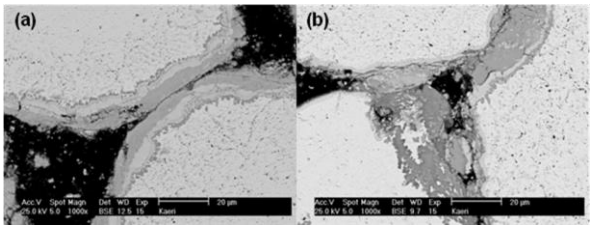

Fig. 2. SEM images of silicide coated U-7wt%Mo-1wt%Ti particles. (a) as extrude; (b) as annealed

Nitride coating layers were formed on the surface of U-7wt%Mo particles with a thickness of about 1~2 ㎛ and the layers were formed on the surface of U-7wt%Mo-1wt%Ti particles with a thickness of 10~15 ㎛, as shown in Fig. 3 and Fig. 4. Although they are thinner than the silicide coating layers, annealing test result showed that interaction layer formation was suppressed.

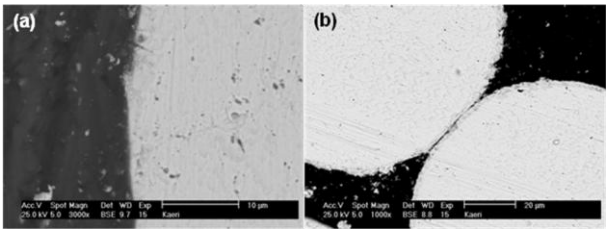

Fig. 3. SEM images of nitride coated U-7wt%Mo particles. (a) as extruded; (b) as annealed

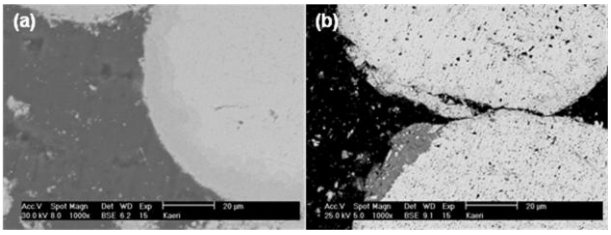

Fig. 4. SEM images of nitride coated U-7wt%Mo-1wt%Ti particles. (a) as extruded; (b) as annealed

In order to refine the crystallographic information of the compounds in the coating layers and interaction layers, specimens were experimented by high energy XRD at the ESRF. The whole pattern analyses result by the Fullprof program showed that the interaction layers were composed of UAl_3, U_6MoAl_{43}, UMo_2Al_{20} and UAl_4[3,4] as shown in Fig. 5 and Fig. 6. It is also found that the peak intensity of UAl_3 and UAl_4 were changed by the addition of 1wt% Ti.

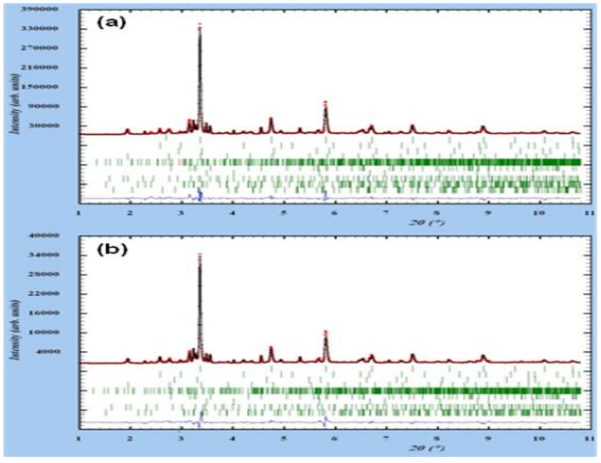

Fig. 5. The Fullprof analysis results of extruded dispersion fuel rods made of (a) silicide coated U-7wt%Mo powder, and (b) U-7wt%Mo-1wt%Ti powder.

The crystallographic refinement by the Fullprof program showed that the compounds of silicide coating layers were identified as U_3Si_5, U_3MoSi_2 and U_3Si and nitride coating layers were composed of UN, UN_2 and U_4N_7. The intensities of Bragg positions corresponding to interaction layers were very weak in the samples with nitride coated U-Mo than in those with silicide coated U-Mo. The results of microstructural observation and phase analysis by the Fullprof program shows that nitride coating is more effective in inhibiting the formation of interaction layers.

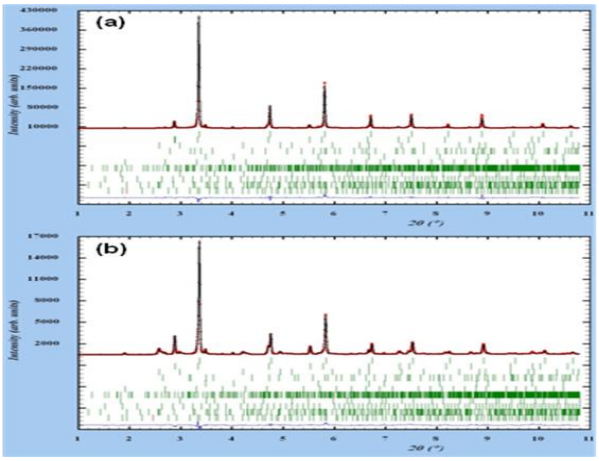

Fig. 6. The Fullprof analysis results of extruded dispersion fuel rods made of (a) nitride coated U-7wt%Mo powder, and (b) U-7wt%Mo-1wt%Ti powder.

4. Conclusions

1. The crystallographic refinement by the Fullprof program showed that interaction layers were composed of UAl_3, U_6MoAl_{43}, UMo_2Al_{20} and UAl_4.
2. Silicide coating layers have been identified to consist of U_3Si_5, U_3MoSi_2, U_3Si and nitride coating layers were composed of UN, UN_2, U_4N_7.
3. Nitride coated samples showed less interaction formation than silicide coated samples.

ACKNOWLEDGMENTS

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