Irradiation-induced Interdiffusion between U-Mo vs. Al-Si

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1. Introduction

Current U-Mo/Al dispersion fuel development efforts are focusing on testing possible remedies for retardation of the interaction layer growth during irradiation. The addition of Si to an Al matrix has been found to be promising to reduce the interactions from experimental observations[1]. Out-of-pile annealing of U-Mo vs. Al-Si diffusion couples have resulted in a reduced interaction layer thickness when compared to U-Mo vs. Al diffusion couples. Because high Si concentrations in interaction layers have been observed, the reduced interaction layer thickness has been attributed to the high Si concentration in interaction layers, which might suppress the interdiffusion of U and Al. Furthermore, it is believed that the presence of Si in the interaction layer control the ratio of Al to U-Mo not to increase more than 3, while the ratio is generally ranging from 4 to 5 without Si. Therefore, it is deduced that Si would stabilize the interaction layer also during irradiation not to form gross fission gas voids. Irradiation tests, such as RERTR-6 and IRIS-3 irradiation tests, using Al-Si matrices instead of an Al matrix have shown remarkable reductions in the interaction layer growth [2].

However, the US AFIP-1 irradiation test at ATR and the European E-FUTURE1 test showed that interaction layers grow significantly due to their high fuel temperatures. It is proposed that the performance of Si on the interaction reduction would be decreased when the interaction layer grows since the Si content in the interaction layer becomes diluted. Also it is found that it is difficult to obtain high Si content in the interaction layer by interdiffusion between the U-Mo particle and Al-Si matrix during irradiation. Post-irradiation examination (PIE) of the KOMO-3 irradiation test using U-7wt%Mo/Al-2wt%Si has shown that the concentration of Si content in the interaction layer is less than 11 at% as shown in Fig. 1(a)[3].

One way to enrich Si in the interaction layer higher than 11 at% is pre-irradiation annealing of U-Mo/Al dispersion fuel. As shown in Fig. 1(b), more than 20 at% of Si could be enriched in the interaction layer by out-of-pile annealing at 580°C. The KOMO-4 and KOMO-5 irradiation tests at HANARO have irradiated pre-irradiation annealed U-Mo/Al-Si dispersion fuel in order to estimate the performance of the Si-rich layer as a diffusion barrier during irradiation.

In this study, the interdiffusion behavior of the pre-irradiation annealed U-Mo/Al-Si dispersion fuel and as-extruded U-Mo/Al-Si dispersion fuel during irradiation are analyzed.

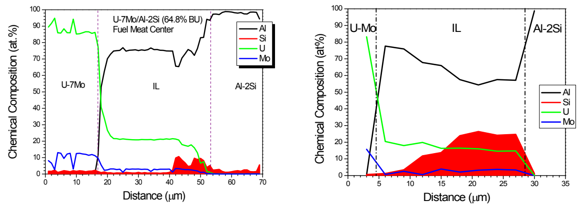


Fig. 1. Si concentration profiles in the interaction layers; (a) after the KOMO-3 irradiation test of U-7Mo/Al-2Si, and (b) after out-of-pile annealing of U-7Mo/Al-2Si at 580°C for 5 h.

2. Experimental Procedures

Centrifugally atomized U-7wt%Mo alloy powder having a diameter of 210~300 μm and gas atomized Al-5wt%Si alloy powder were used for the fabrication of U-Mo/Al-Si dispersion fuel samples. The powder mixture of U-Mo and Al-Si was hot-extruded into a rod-type dispersion fuel sample at 400°C. Pre-irradiation annealing of the U-Mo/Al-Si dispersion fuel sample was conducted at 580°C for 1 hour under vacuum. Microstructures of the specimens were observed by scanning electron microscopy (SEM) and elemental compositions of pre-formed interaction layers were measured by energy dispersive X-ray spectroscopy (EDS). Irradiated U-Mo/Al-Si dispersion fuel samples were observed and characterized by optical microscopy and shielded electron probe micro-analysis(EPMA) in the Irradiated Materials Examination Facility(IMEF) of KAERI.

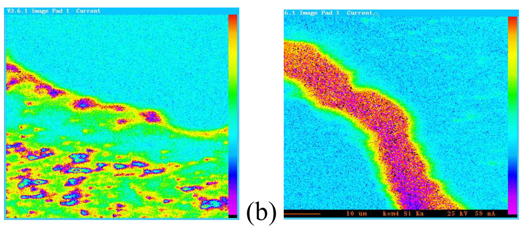
(a)  (b)

Fig. 2. EPMA X-ray elemental mapping of Si for (a) U-7Mo/Al-5Si and (b) pre-annealed U-7Mo/Al-5Si after irradiation.

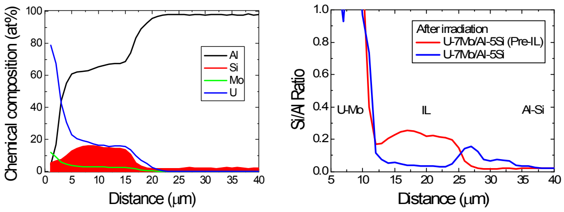


Fig. 3. (a) Composition profiles of the pre-annealed U-7Mo/Al-5Si after irradiation, and (b) Si/Al ratio comparison between irradiated U-7Mo/Al-5Si samples with and without pre-irradiation annealing.

3. Results and Discussion

Fig. 2 shows x-ray elemental maps for Si of the non-annealed U-7Mo/Al-5Si and the pre-irradiation-annealed U-7Mo/Al-5Si after irradiation. EPMA analyses of irradiated U-7Mo/Al-5Si dispersion fuel showed that initial 30-50 at% of Si concentration in the pre-formed layer was reduced uniformly to less than 15 at% Si as interaction layers grew additionally during irradiation as shown in Fig. 3(a). It means that diffusion of Al into the interaction layer was active and dominant. When Si/Al ratios of irradiated U-7Mo/Al-5Si dispersion fuel samples with/without pre-annealing were compared as shown in Fig. 3(b)[4], the pre-annealed sample shows the higher Si/Al ratio at the inside of the interaction layer while the non-annealed sample shows only a small bump in the ratio at the interface of interaction layer and the matrix. In the interaction layer of the non-annealed sample, the Si/Al ratio is very low.

The changes of diffusion paths before and after irradiation are compared by using a pseudo-ternary composition diagram of (U,Mo)-Al-Si as shown in Fig. 4(a). Differences in diffusion paths of the pre-annealed and the non-annealed U-7Mo/Al-5Si samples after irradiation are compared in Fig. 4(b). Al-rich phase was formed more in the non-annealed samples while its Si-content is not so high compared to the pre-annealed sample. (Al+Si)/(U+Mo) atomic ratio was nearly equal to 4.5, in the interaction layer of the pre-annealed U-7Mo/Al-5Si after irradiation as shown in Fig. 5(a). The composition is similar to that of the interaction layer of U_3Si_2/Al reported by Leenaers et al.[5]. It means that high Si in interaction layers does not always stabilize $(U,Mo)(Al,Si)_3$ under irradiation conditions.

When uranium and silicon exist together, aluminum diffuses in the interaction layers and no change in U/Si ratio before and after irradiation. It means U and Si maintain strong bonds each other. No aluminum can pass the interaction layers to form UAl_x. When uranium and silicon do not exist together, only small Si react with U to form a Si rich layer. At the same time Al diffuse to U-Mo to form UAl_x because Si-rich layer may not be tight enough. There are many bare U-Mo without enough Si-rich layer so Al can diffuse to react with U-Mo. Once the Si-rich layer formed, it can slow down the diffusion of Al and the (Al+Si)/(U+Mo) atomic ratio was maintained to be nearly 3 as shown in Fig. 5(b).

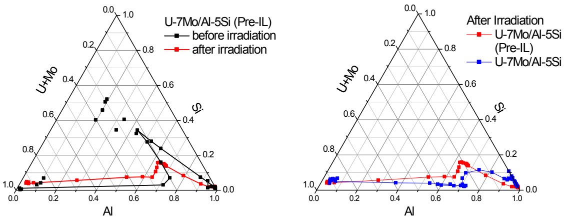


Fig. 4. Diffusion path comparisons between irradiated U-7Mo/Al-5Si samples with and without pre-irradiation annealing , before and after irradiation.

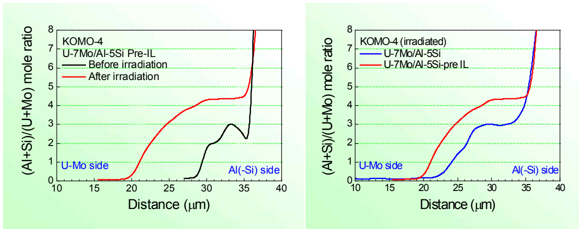


Fig. 5. (Al+Si)/(U+Mo) atomic ratio comparisons between irradiated U-7Mo/Al-5Si samples with and without pre-irradiation annealing, before and after irradiation.

4. Conclusions

By pre-irradiation forming of Si-rich layers, in-pile interdiffusion mechanisms depending on the Si distribution can be understood for U-7wt%Mo/Al-5wt%Si. The preferential pile up of Si at the periphery of interaction layers reduces the interaction during irradiation. The presence of Si in the interaction layers does not stabilize the composition as close to $(U,Mo)(Al,Si)_3$, which is different from out-of-pile diffusion test results.

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