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INTERDIFFUSION IN THE Si-RICH LAYER OF U-Mo/Al-Si FUEL

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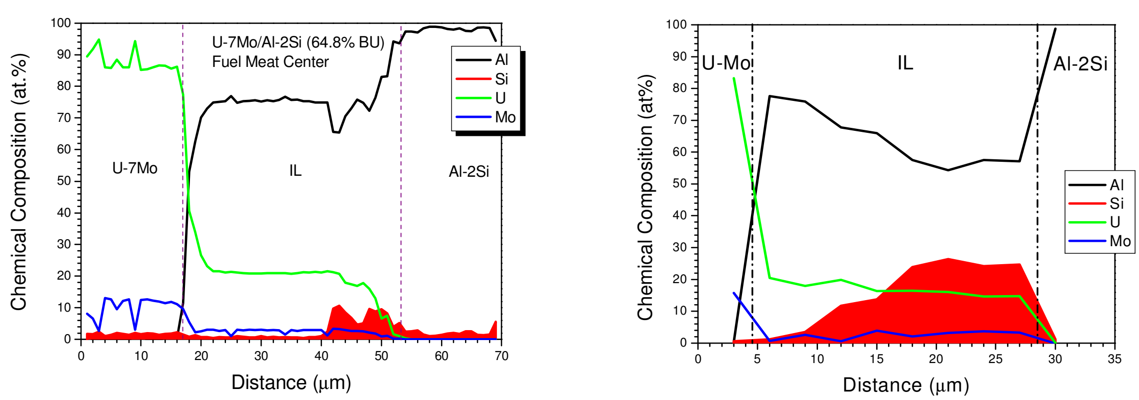
ABSTRACT

Pre-irradiation-annealed U-7wt%Mo/Al-5wt%Si dispersion fuel has been irradiated at HANARO in order to investigate the effects of a pre-formed Si-rich layer on the interaction layer growth of U-Mo/Al-Si dispersion fuel during irradiation. Post-irradiation examination results showed that Al diffuses more actively than Si during irradiation. Composition profiles in the interaction layer of the pre-annealed dispersion fuel before and after the irradiation test were analyzed. Initial 30~50 at% of Si in the pre-formed layer in the pre-annealed U-7wt%Mo/Al-5wt%Si sample was decreased to less than 15 at% Si as the interaction layer grew additionally during irradiation. While (Al,Si)/(U,Mo) atomic ratio in the interaction layer was near to 3 for non-annealed U-7Mo/Al-5Si, a higher x value, up to 4.5, was measured in the interaction layer of the pre-annealed U-7Mo/Al-5Si after irradiation. It became clear that the high Si content in the interaction layer as in pre-annealed fuel did not necessarily yield a lower x value close to 3. However, a high Si/Al ratio was retained at the U-Mo fuel/interaction layer interface for the pre-annealed sample after irradiation.

1. Introduction

The growth of interaction layers during irradiation has been considered to be one of the most challenging issues in the development of U-Mo/Al dispersion fuel[1]. Since the Al matrix is consumed by the interaction, the thermal conductivity of U-Mo/Al dispersion fuel decreases and the fuel temperature increases. In addition, the accumulation of radiation damage during irradiation induces amorphization of the interaction layers[2,3] and significant pillowing of the fuel plate may occur by the growth of fission gas voids primarily at the interaction layer/Al matrix interfaces. The addition of Si to the Al matrix has been found to be a promising remedy to reduce the interactions[4]. Irradiation tests using Al-Si matrices instead of Al matrix have shown reductions in the interaction layer growth[5]. However, the performance of Si on the interaction reduction might be decreased when the interaction layer grows since the Si content in the interaction layer becomes diluted. Although the dilution of Si in the U-Mo/Al-Si dispersion fuel can be delayed by increasing the Si content in the Al-Si matrix, minimizing the total Si content is recommended for improving the reprocessing efficiency of the spent fuel[6].

However, 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 HANARO 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)[7]. However, out-of-pile annealing of U-Mo/Al-Si has resulted in a high Si concentration in interaction layers as shown in Fig. 1(b)[8].The PIE of the BR1 reactor fuel has shown that a thin USi layer limited interaction between the uranium and aluminum cladding effectively during irradiation[9]. This result suggests that a Si-rich layer is very efficient as a diffusion barrier to the interaction between uranium and aluminum. Therefore, it is necessary to estimate the performance of the Si-rich layer as a diffusion barrier. In this study, the microstructures, compositions and the diffusion barrier performance of a pre-formed Si-rich-layer on U-Mo particles have been examined by irradiation tests at HANARO.



(a) (b)

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^{o}C for 5 h.

2. Experimental Procedures

Centrifugally atomized U-7wt%Mo powder having a diameter of 210~300 μm and gas atomized Al-5wt%Si 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 rod-type dispersion fuel meat at 400^{o}C. Pre-irradiation annealing of U-Mo/Al-Si dispersion fuel samples was conducted at 580^{o}C for 1 h 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). Table 1 lists details of the KOMO-4 irradiation test conditions[10]. Irradiated U-Mo/Al-Si dispersion fuel samples were observed and characterized by optical microscopy and shielded EPMA in the Irradiated Materials Examination Facility(IMEF) in KAERI.

Table 1. Details of the Fuel Sample and Irradiation Test Conditions of KOMO-4

Compositions of fuel meat	U-7wt%Mo/Al-5wt%Si
Uranium density	5.0 gU/cm^3 (LEU)
U-Mo particle size	210-300 μm
Fuel meat diameter	6.35 mm
Fuel meat length	50 cm
Max. Linear power	96 kW/m
BOL temperature	195^{o}C
Irradiation hole	OR3 (HANARO)
EFPD	132 days (Dec.22 2008 – Jan. 03, 2010)
U-235at% Burnup	55% LEU

3. Results and Discussion

Fig. 2(a) represents that no remarkable interaction layer was formed on U-Mo particles after hot extrusion of dispersion fuel meat at 400^{o}C. However, the pre-irradiation-annealed U-7Mo/Al-5Si sample has an interaction layer with an average thickness of 10 μm as shown in Fig. 2(b). The thickness of the interaction layer is not uniform. The composition profiles in the pre-formed interaction layer, measured by EDS, are shown in Fig. 3(a). High concentrations of Si ranging from 30 to 50 at% are measured and large variation of Si content is observed in the interaction layer. An x-ray elemental map for Si, which can be seen in Fig. 3(b), reveals that the distribution of Si was not uniform. The irregular interaction during pre-irradiation-annealing might be attributed to the phase transformation of the meta-stable γ to $\alpha+\gamma$ or $\alpha+\gamma$'at 580^{o}C[8]. Due to the high Si-content in the interaction layer, a thickness of Si depletion zone from the interface is estimated to be approximately 50 μm by considering the mass balance of Si.

The microstructures of irradiated U-7Mo/Al-5Si dispersion fuel samples with and without a pre-formed Si-rich layer are compared in Fig. 4. The interaction layer thicknesses are similar to each other as shown in Fig. 5. It is difficult to say that the pre-annealed U-7Mo/Al-5Si has a superior performance, because the pre-formed Si-rich layer does not work as an effective diffusion barrier to Al as expected. When compared to U-7Mo/Al-5Si without pre-irradiation-annealing, it seems that no benefit was obtained by the pre-irradiation annealing. However, considering the Si-depleted zone around the interaction layer as shown in Fig. 4(b), the pre-formed Si-rich layer resulted in a lower interaction thickness against nearly pure Al. When compared to the U-7Mo/Al dispersion fuel with interaction layer thickness greater than 30 μm as shown in Fig. 5, it seems that the interaction was retarded by the presence of the Si-rich layer.

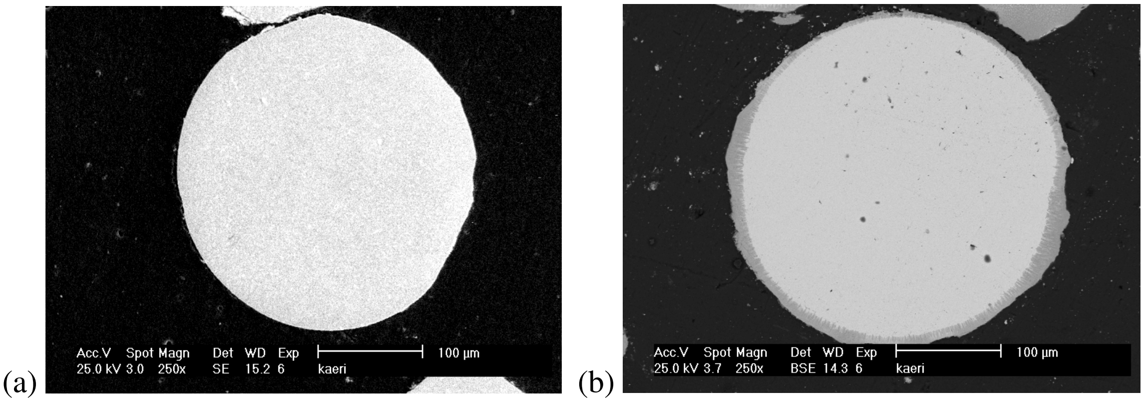


Fig. 2. SEM micrographs of cross-sections of U-7Mo/Al-5Si dispersion fuel meat; (a) after extrusion at 400°C, and (b) after out-of-pile annealing at 580°C for 1 h.

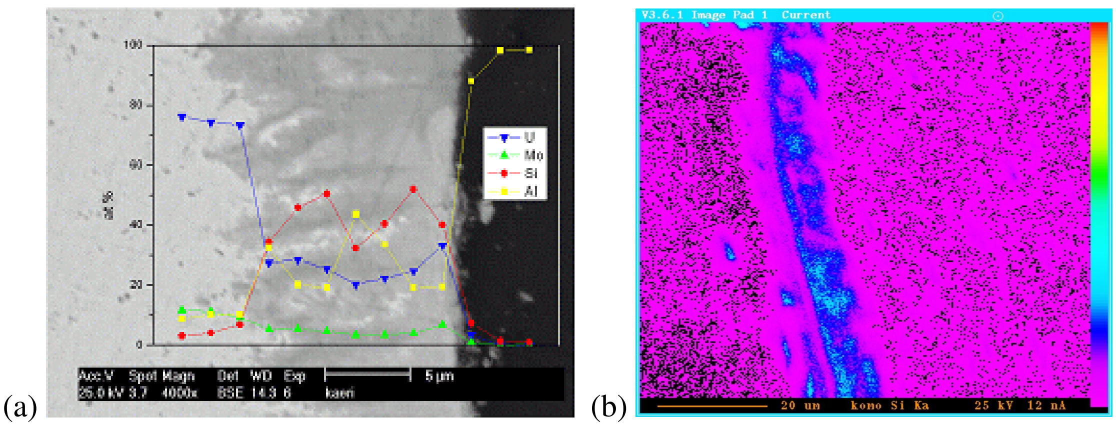


Fig. 3. (a) An SEM micrograph superimposed with composition profiles measured by EDS and (b) an x-ray elemental map for Si of the pre-irradiation-annealed U-7Mo/Al-5Si sample.

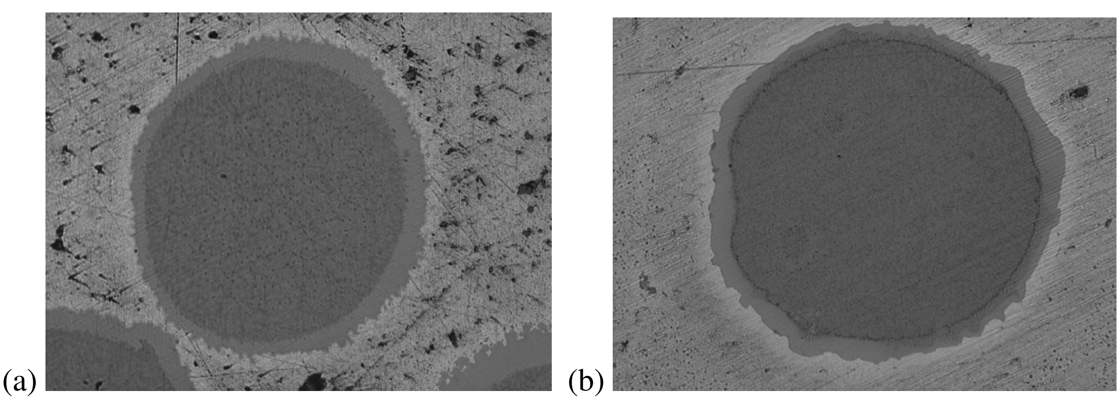


Fig. 4. Optical micrographs of irradiated U-7Mo/Al-5Si dispersion fuel samples; (a) without pre-irradiation annealing, (b) with pre-irradiation annealing.

Fig. 6 compares the cross-sectional optical micrographs of U-7Mo/Al-5Si samples before and after irradiation. Swelling measured from diameter changes before and after irradiation was 5 % ($\Delta V/V$) for the pre-annealed U-7Mo/Al-5Si. Volume fractions of the constituent phases were measured by using image analysis techniques as listed in Table 2. The volume fractions of irradiated sample were corrected by considering of swelling of the fuel meat. The volume fraction of the interaction layer after irradiation is 8.7 %, which is not much greater than 8.6% of the as-annealed sample. It might be associated with the non-uniform distribution of interaction layer of the as-annealed sample, which is not resolvable by optical microscopy. Only 3.3 vol.% of the initial Al-Si matrix was consumed by interaction during irradiation. By considering the ratios of the molar volumes of Al to UAl_3 or UAl_4, it was estimated that the volume fraction of the interaction layer was increased by 4~5 % during irradiation.

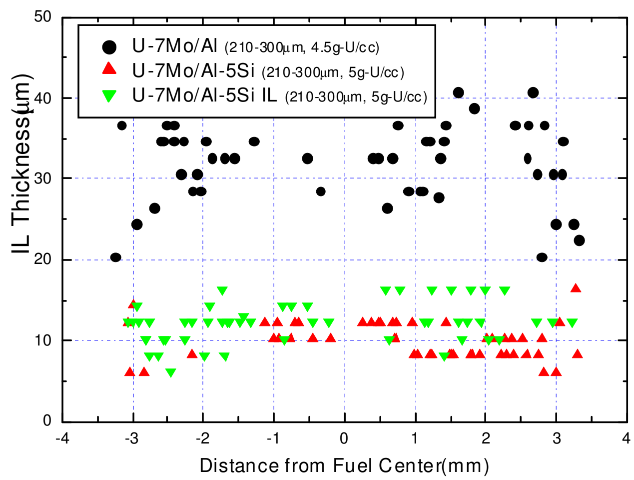
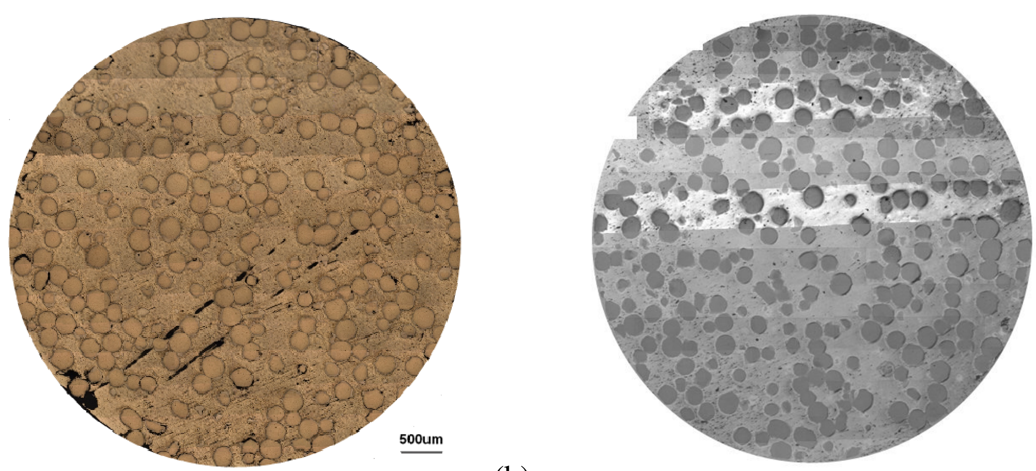


Fig. 5. Distribution of interaction layer thicknesses after irradiation of U-7Mo/Al and U-7Mo/Al-5Si along the radial distance from the fuel center.



(a) (b)

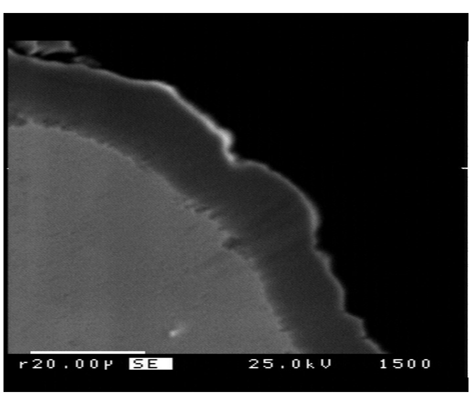
Fig. 6. Optical micrographs of U-7Mo/Al-5Si samples; (a) after pre-irradiation annealing, (b) after irradiation test.

Fig. 7 shows an SEM micrograph and an x-ray elemental map for Si of the pre-irradiation-annealed U-7Mo/Al-5Si sample 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. 8(a). 20-40 at% of Al concentration was increased to 60-70 at% after irradiation. 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. 8(b)[11], 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. Si-depleted interaction layers might have a low resistance to pore formation as in U-Mo/Al dispersion fuel with the pure Al matrix[12]. Si-rich interaction layers will be beneficial to suppress the pore growth at the U-Mo/interaction layer interface at a higher burnup.

The changes of diffusion paths are compared by using a pseudo-ternary composition diagram of (U,Mo)-Al-Si as shown in Fig. 9(a). Differences in diffusion paths of the pre-annealed and the non-annealed U-7Mo/Al-5Si samples are compared. 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, x, was nearly equal to 4.5, in the interaction layer of the pre-annealed U-7Mo/Al-5Si after irradiation as shown in Fig. 9(b). The composition is similar to that of the interaction layer of U_3Si_2/Al reported by Leenaers et al.[13]. It means that high Si in interaction layers does not always stabilize (U,Mo)$(Al,Si)_3$ under the irradiation conditions.

Table 2. Volume fractions of phases in pre-annealed U-7Mo/Al-5Si dispersion fuel before and after irradiation. (* obtained from a reverse calculation by using the matrix consumption)

Phase	Pre-irradiation Annealed (vol.%)	Irradiated	
		Measured (vol.%)	Swelling Corrected (vol.%)
U-Mo	24.8	31.5	33.1
Interaction Layer	8.6 (~4*)	8.3	8.7
Matrix	66.6	60.2	63.3

(a) 

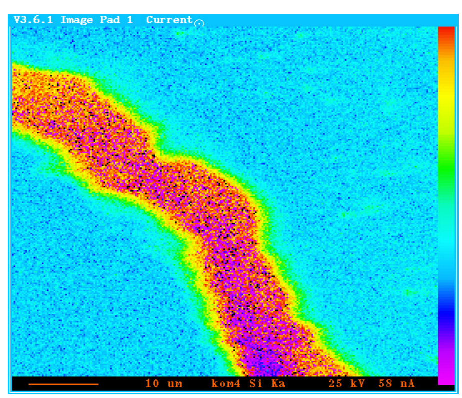
(b) 

Fig. 7. (a) An SEM micrograph and (b) an x-ray elemental map for Si of the pre-irradiation-annealed U-7Mo/Al-5Si sample after irradiation.

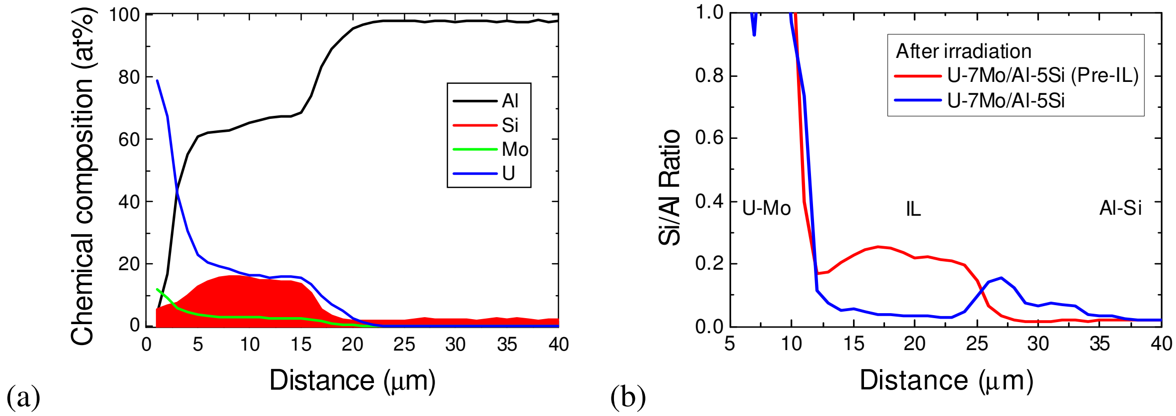


Fig. 8. (a) Composition profiles of the pre-annealed U-7Mo/Al-5Si measured by EPMA after irradiation, and (b) Si/Al ratio comparison between irradiated U-7Mo/Al-5Si samples with and without pre-irradiation annealing.

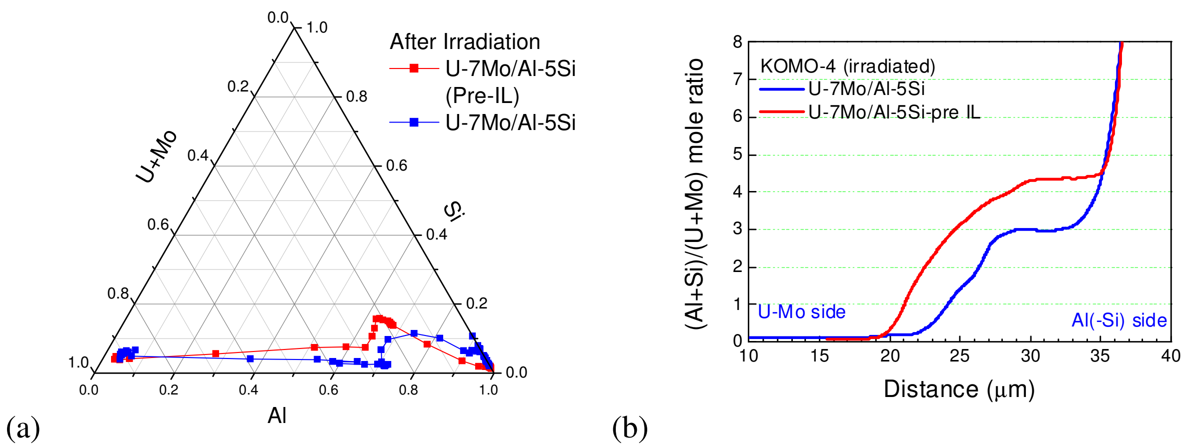


Fig. 9. (a) Diffusion path comparisons, and (b) (Al+Si)/(U+Mo) atomic ratio comparisons between irradiated U-7Mo/Al-5Si samples with and without pre-irradiation annealing.

Some benefits of the presence of pre-formed Si-rich layers might be found as follows. First, it resulted in the less interaction increment considering the initial interaction layer thickness and the Si-depleted zone around the interaction layers. Second, it retained the higher Si content in the interaction layer and no Si-depleted U-Mo-Al interdiffusion zone was developed as in the U-Mo/Al-Si sample without pre-annealing. Third, fewer fission gas pores at a higher burnup are expected due to the high Si/Al ratio at the U-Mo/interaction layer interface.

The PIE results of pre-annealed U-7Mo/Al-5Si will be reflected in the test design of the KOMO-5 irradiation test to be started in mid 2011. Thinner Si-rich layers less than 5 μm in thickness will be formed by controlling pre-annealing temperature and time in order to investigate the effect of the initial thickness of a pre-formed Si-rich layer on the dilution of Si during irradiation. Lower Si content less than 5 wt% will be used because 5 wt% of Si in the Al-Si matrix was sufficient to suppress the interaction layer growth under the HANARO irradiation conditions. Higher discharge burnup will be pursued in order to investigate the effect of burnup on the diffusion barrier performance of the pre-formed Si-rich layers.

4. Conclusions

Pre-irradiation annealed U-7wt%Mo/Al-5wt%Si with a 10 μm Si-rich layer has been irradiated at HANARO up to 55 at% burnup (KOMO-4). Its performance was similar to the non-annealed U-7Mo/Al-5Si, and interaction layer growth during irradiation was lower than the non-annealed sample, when considering the initial thickness and the surrounding Si-depletion zone after pre-annealing. The compositions of interaction layer was close to $(U,Mo)(Al,Si)_{4.5}$ not $(U,Mo)(Al,Si)_3$. High Si content approximately 15 at% was retained in the interaction layer without the UMo-Al interdiffusion zone as formed in the non-pre-annealed U-7Mo/Al-5Si after irradiation. Another irradiation test of pre-irradiation annealed U-7Mo/Al-2Si with a thinner Si-rich layer up to ~70 at% burnup has been planned as one of the irradiation test samples for the KOMO-5 test.

Acknowledgment

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