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EFFECT OF ALLOY ADDITIONS TO AL MATRIX AND U-MO FUEL ON THE INTERACTION LAYER GROWTH DURING IRRADIATION - KOMO-4 IRRADIATION TEST RESULTS

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ABSTRACT

In accordance with the high power irradiation test program of rod-type U-Mo dispersion fuels (KOMO-4), twelve different U-Mo-X(X=Zr,Ti)/Al-Si fuel rods have been irradiated in HANARO. The objectives of this irradiation test are to find a suitable third alloying element for U-Mo fuel, a minimum content of Si in matrix Al, the optimal combination between them, and the maximum allowable power. The test fuel rods were irradiated for 132.1 EFPD and discharged at average burnup of 47.1at%U-235. The maximum peak linear power for test fuel rods was 105.7 kW/m, and the beginning-of-life temperature was ranged from 171 to 200 °C, depending on location. In this study, we investigate microstructure evolution in the irradiated U-7wt%Mo/Al-Si fuel rods by the additions of 5wt% Si in Al and the third element (1wt% of Zr or Ti) in U-Mo. We propose a mechanism to explain the role of Ti and Zr addition to U-Mo when Si is added to the matrix.

1. Introduction

Uranium-molybdenum alloy particle dispersion fuel in an aluminum matrix with a high uranium density has been developed for a high performance research reactor in the RERTR program [1]. An unacceptable volume expansion caused by interaction layer and subsequent high porosity formations between the UMo fuel particles and Al matrix has been an obstacle

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for further development of this fuel.

Several relevant studies to understand the detailed inter-diffusion mechanism in the interaction layer (IL) and to mitigate its growth are under progress [2-16]. The group at Argonne National Laboratory proposed to modify the matrix by adding Si and/or add a third transition metal element to the U-Mo fuel. Various elements such as Al, Si, Ti, Zr, Sn, Y, Cu, Cr and Pt were evaluated in out-of-pile [8-13] and in-pile studies [14-16]. Promising results were obtained for Ti [9-10, 15] and Zr [11-12, 14]. Diffusion couple tests have demonstrated that an addition of a small amount of Ti or Zr into γ(U-Mo) can not only reduce the interaction layer growth with Al or Al-Si alloy, but also induces a high accumulation of Si in the interaction layer (IL) developed with Al-Si alloys, especially in the case of Ti [9,10].

A series of KOMO irradiation tests to develop the rod-type high uranium density U-Mo fuel have been carried out at KAERI [17-20]. The KOMO-4 test is the latest one. The previous results have shown that extensive formation of interaction phase in U-7Mo/Al dispersion fuel could be considerably reduced by an addition of Si in Al matrix, an addition of Ti or Zr in U-Mo, and a use of large-sized fuel particles that reduces the interface area between the fuel and matrix. However, the latest test (KOMO-3) also revealed that 2% of Si in Al was still insufficient to properly suppress the interaction layer growth for rod-type fuel geometry due to higher temperatures. It also indicated that the combined effect of Si and a third allying element on in-reactor performance of dispersion fuel is worth pursuing further investigation.

In KOMO-4, U-Mo-X(X=Zr,Ti)/Al-Si fuel rods have been irradiated in HANARO. The objectives of this irradiation test are to find a suitable third alloying element for U-Mo fuel, a minimum content of Si in matrix Al, the optimal combination between them, and the maximum allowable power. In this study, we investigate microstructure evolution in the irradiated U-7wt%Mo/Al-5wt% Si fuel rods by the additions of the third alloying element (1wt% of Zr or Ti) in U-Mo. We further examine correlations between interaction layer growth and test parameters. A mechanism to explain the role of Ti and Zr added to U-Mo is also a part of the present work.

2. KOMO-4 irradiation experiments

Centrifugally atomized U-7Mo, U-7Mo-1Ti, and U-7Mo-1Zr fuel powders were used for the fabrication of the rod-type dispersion fuel meats. Twelve different test rods were irradiated in the HANARO: Among them, four (4) test rods were post-irradiation examined and used for the analysis in this paper. Table 1 shows the irradiation parameters for test fuel rods.

The microstructure of dispersion fuel rods before and after irradiation was characterized by optical microscopy (OM) and scanning electron microscopy (SEM). Concentration profiles of reaction layers were obtained by point-to-point counting techniques using an EPMA.

3. Results and discussion

Microstructure evolution in the irradiated U-7wt%Mo-1wt% X(X=Ti, Zr)/Al-5wt% Si fuel rods was examined by using OM and SEM. Fig. 1 shows typical OM images of irradiated fuel cross sections. We have measured the IL thickness on OM images of fuel cross sections and averaged the measured values. Fig. 2 shows a comparison of the averaged IL thicknesses. The IL thickness decreases as the Si content in the matrix increases. This result is consistent with previous in-pile and out-of-pile test data. Addition of Ti or Zr in U-Mo showed further reduction of IL growth. The US RERTR-8 test showed that the effect of Ti of Zr addition in U-Mo was minimal because of low irradiation temperatures. At low temperatures, the Si effect is so dominant that the additional effect of Zr or Ti addition was not clearly conclusive. However, because the fuel temperatures in the KOMO-4 test are higher, the effect of Zr or Ti is magnified and noticeable. In addition, we could notice that Zr addition to U-Mo was more effective than Ti. This is somewhat contradictory from out-of-pile test results, in which Ti was better than Zr [11]. The main reason is because at high temperatures, where the out-of-pile tests were performed, the transformation of the meta-stable γ phase to α+γ' inadvertently affected the results. However, at the still lower temperatures of the present case than the out-of-pile tests, this does not occur. Hence, Zr having a higher affinity to Si than Ti shows a stronger effect. The averaged IL thicknesses of both U-Mo-1Ti/Al-5Si and U-Mo-1Zr/Al-5Si are generally similar to that of the U-Mo/Al-8Si sample. Therefore, either Ti or Zr addition in U-Mo is a promising method to help reduce the amount of Si addition to the Al matrix, particularly for high temperature applications like the KOMO-4 test.

Table 1. Irradiation condition for test rods and .

Sample I.D.	Fuel meat composition	Max. Burn-up (at%U-235)	Max. linear power (kW/m)	Central BOL Temp. (^{O}C)
676-5S1	U-7Mo/Al-5Si	49.0	100.2	189.2
676-8S1	U-7Mo/Al-8Si	48.0	98.48	188.4
677-5S1	U-7Mo-1Ti/Al-5Si	51.1	105.7	199.5
588-5S1	U-7Mo-1Zr/Al-5Si	48.7	100.9	189.9

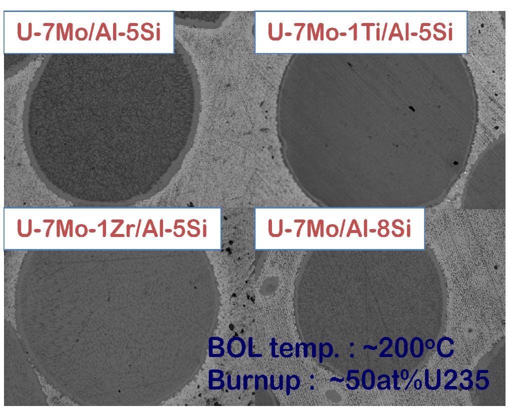


Fig. 1. OM images of fuel particle cross sections.

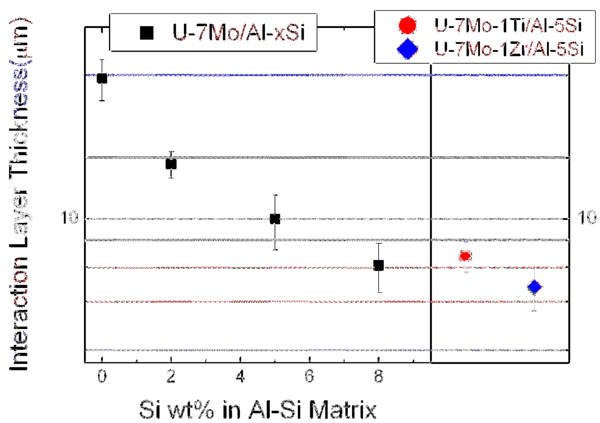
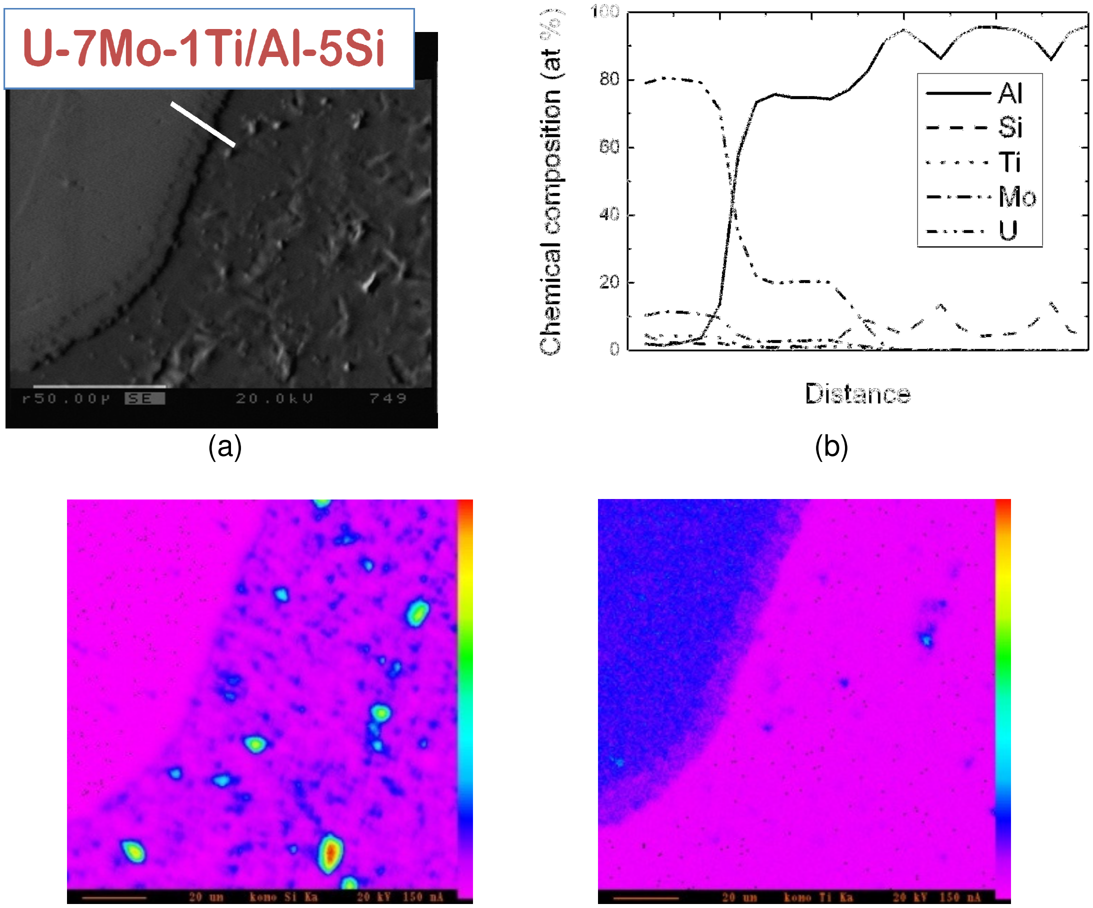


Fig. 2. Averaged IL thickness of irradiated fuels.

Chemical composition profiles in and around the IL in the irradiated fuels were measured by EPMA. Fig.3 shows Si and Ti X-ray mapping images and typical composition profiles along the IL thickness direction for the irradiated U-7Mo-1Ti/Al-5Si fuel. Si X-ray mapping image and compositional profile show that Si was accumulated at the IL-matrix interface so that a thin Si-rich layer was formed on the matrix side of IL. The Si accumulation at the IL-matrix interface was also reported in irradiated U-Mo/Al-Si fuels (Leenaers [21],Ryu [22]). Ti X-ray mapping also showed the presence of Ti in IL, although not prominent.

Fig. 4 shows the (Al+Si)/(U+Mo+Ti) and Si/(Al+U+Mo+Ti) ratio plots in and around IL of the irradiated U-7Mo-1Ti/Al-5Si. For a comparison, atomic ratios obtained from the IL of irradiated

U-7Mo/Al-5Si fuel were also shown. The (Al+Si)/(U+Mo+Ti) ratio in the IL was about 3, which is consistent with that of the sample without Ti. The Si/(Al+U+Mo+Ti) ratio in Fig. 4 shows again the accumulation of Si at the IL-matrix interface and shows a minimum in the IL. On the fuel side of the interaction layer, a slight increase in the Si ratio is seen. These results are also consistent with those observed in the U-7Mo/Al-5Si. However, when we see the Si atomic ratio variation from the IL-matrix interface to fuel side, we can find that Si ratio was considerably decreased when Ti was added in U-7Mo.



(a) (b)

(c) (d)

Fig. 3. Compositional profile in and around the IL in the irradiated U-7Mo-1Ti/Al-5Si fuels.

(a) SEM image, (b) Compositional profile along the IL,

(c) Si X-ray mapping image, (d) Ti X-ray mapping image

Fig.5 shows Si mapping images and composition profiles along the IL for the irradiated U-7Mo-1Zr/Al-5Si fuel. This irradiated fuel also shows the accumulation of Si at the IL-matrix interface. The enlarged compositional profile of Fig. 5(d) revealed the presence of Zr in IL.

Fig. 6 shows the (Al+Si)/(U+Mo+Zr) and Si/(Al+U+Mo+Zr) ratios in and around ILs for irradiated U-7Mo-1Zr/Al-5Si. The (Al+Si)/(U+Mo+Zr) ratio in the IL is in the range of 2 - 3. This ratio was slightly smaller than that in the Ti addition sample. The trend of the Si atomic ratio shown in Fig. 6(b) is similar to that of Fig. 4(b). This fact reveals that Zr and Ti effect on Si behavior in and around IL is almost identical.

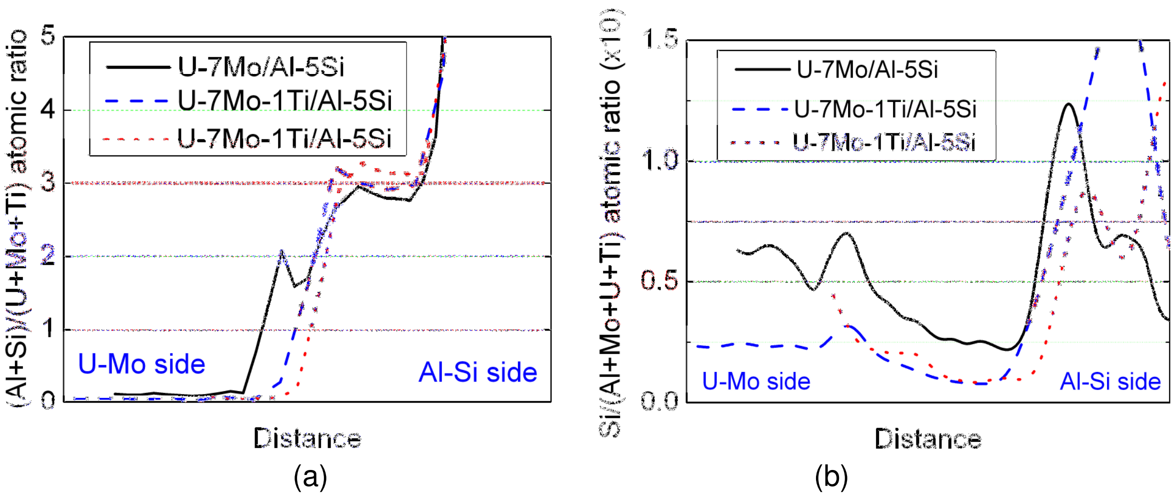


Fig. 4. Atomic ratio in and around ILs for irradiated U-7Mo-1Ti/Al-5Si fuels.
(a) (Al+Si)/(U+Mo+Ti) (b) (Al+Si)/(U+Mo+Ti)

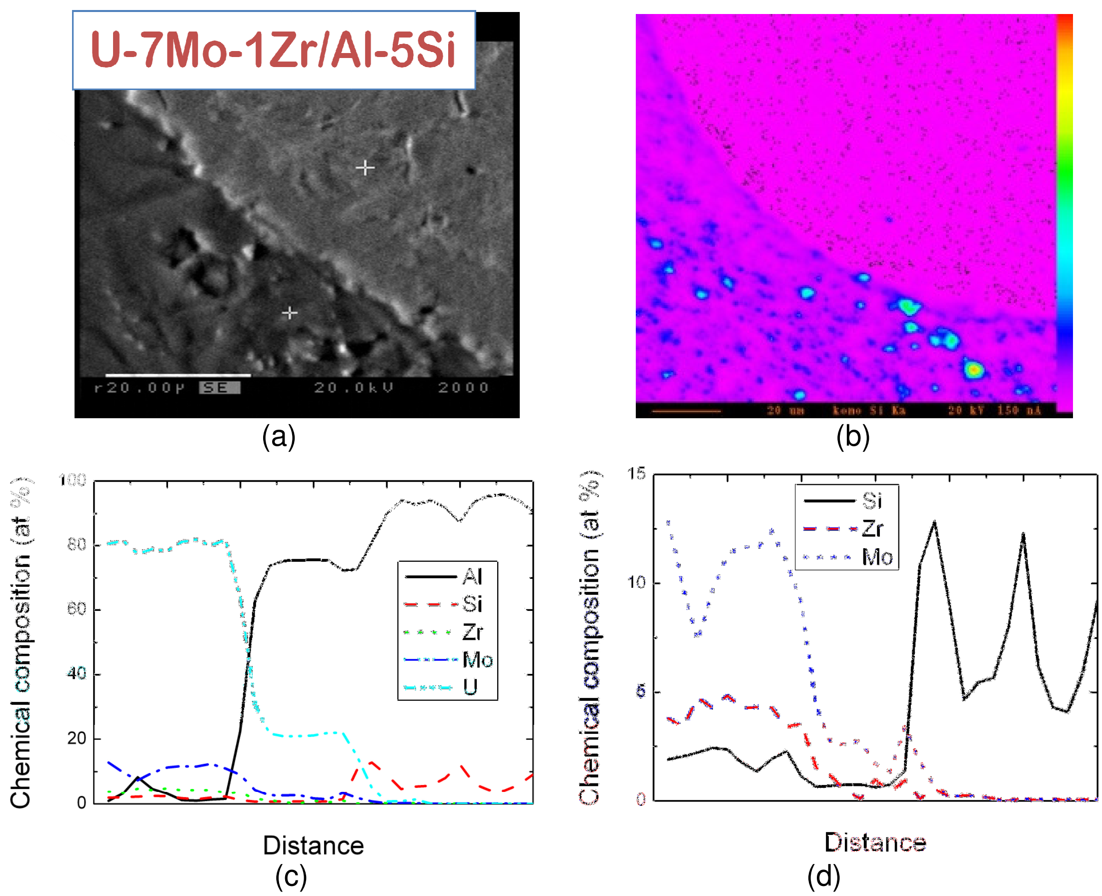
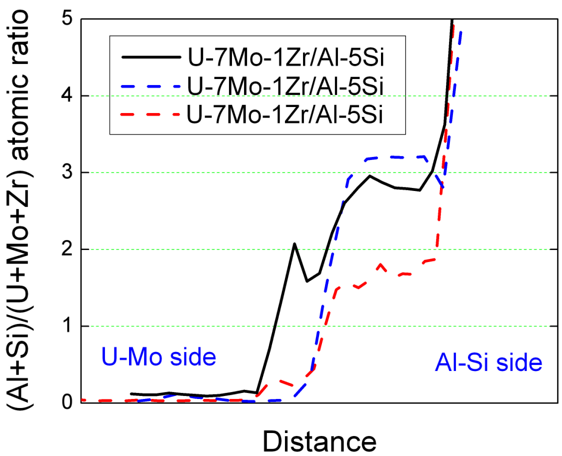
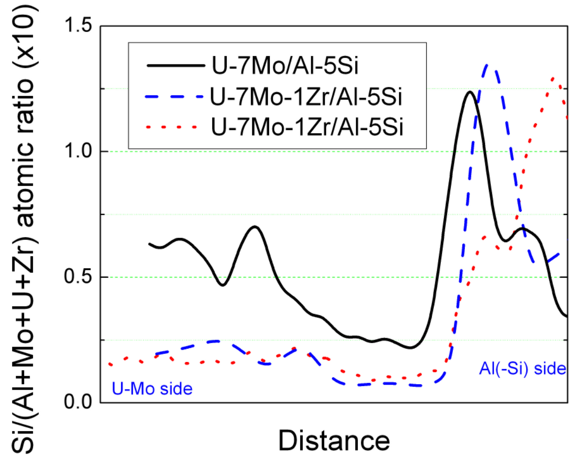


Fig. 5. Compositional profile in and around the IL in the irradiated U-7Mo-1Zr/Al-5Si fuels.
(a) SEM image, (b) Si X-ray mapping image, (c) Compositional profile along the IL, (d) enlarged profile



(a)



(b)

Fig. 6. Atomic ratio in and around ILs for irradiated U-7Mo-1Zr/Al-5Si fuels.
(a) (Al+Si)/(U+Mo+Zr) (b) (Al+Si)/(U+Mo+Zr)

The test results clearly show that addition of Zr or Ti in U-Mo further reduces the IL growth of U-7Mo/Al-5Si fuel during irradiation. EPMA results show that the concentration profiles of component elements were almost identical, regardless of Ti or Zr addition, except the Si atomic ratio in the IL and fuel particle periphery: The Si atomic ratio in IL and fuel was decreased when Zr or Ti were added in U-Mo. Therefore, it seems that lowering of Si atomic ratio in IL and fuel is closely correlated with the reduction of IL growth.

The mechanism of the role of Ti or Zr in U-Mo concocted with Si addition in the matrix can be considered as follows: According to the thermodynamic calculation conducted by Kim[5], Zr or Ti more preferentially reacts with Si at the IL-matrix interface and form a strong compound in which the Si is immobilized. The role of the presence of the Zr or Ti at the IL-matrix interface is that it behaves like a protecting barrier against Si diffusion from the IL-matrix interface to the inside of the IL. Because the Si is immobilized at the IL-matrix interface, the Si accumulates at the IL-matrix interface. In other words, Si depletion from the so-called fission fragment recoil zone can be sustained longer by reducing the Si loss from the IL-matrix interface. Therefore, Zr or Ti addition has an equivalent effect of increasing Si addition in Al matrix.

It is also plausible that when Si in the IL diffuses to the IL-fuel interface, it forms a Si-rich protective phase on the fuel particle as observed in out-of pile tests. In this case, two barrier layers on both interfaces synergistically reduce IL growth. However, based on the observation of relatively low Si concentration in the inside IL away from the IL-matrix interface in the present work, this possibility is rather slim, particularly at low temperatures.

4. Conclusions

From the KOMO-4 test, microstructure evolution in the irradiated U-7wt%Mo/Al-5wt%Si fuel rods by the additions of the third element (1wt% of Zr or Ti) in U-Mo has been investigated to verify the their effects on the interaction layer growth. Addition of Zr or Ti in the U-Mo fuel showed substantial positive effect on reducing the IL growth. Therefore, Ti or Zr addition in U-Mo is an effective method to reduce the necessary amount of Si in Al matrix. Preferential reaction of Zr or Ti with Si that reduces the Si diffusion to the inside of the IL, and thereby increases Si concentration at the IL-matrix interface seems to be the main reason for the retardation of IL growth in U-7Mo-(Ti,Zr)/Al-Si dispersion fuel.

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