Analyses on the U-Mo/Al Chemical Interaction and the Effects of Diffusion Barrier Coatings

Ho Jin Ryu*, Woo Jeong Kim, Woo Hyung Cho, Yong Jin Jeong, Yoon Sang Lee, Jong Man Park, Chang Kyu Kim
Korea Atomic Energy Research Institute: 1045 Daedeok-daero, Yuseong-gu, Daejeon 305-353, Republic of Korea
Corresponding author: hjryu@kaeri.re.kr

1. Introduction

While many HEU-fueled research reactors have been converted by adopting LEU U_3Si_2 fuel in harmony with the Reduced Enrichment for Research and Test Reactors (RERTR) program, some high performance research reactors still need the development of advanced fuels with higher uranium densities[1]. Currently, gamma-phase U-Mo alloys are considered promising candidates to be used as high uranium density fuel for the high performance reactors. For the production of U-Mo alloy powder, the centrifugal atomization technology developed by KAERI has been considered the most promising way because of high yield production and excellent powder quality when compared with other possible methods such as grinding, machining or hydriding-dehydriding [2].

However, severe pore formation associated with an extensive interaction between the U-Mo and Al matrix, although the irradiation performance of U-Mo itself showed most stable, delay the fuel qualification of U-Mo fuel for high performance research reactors[3]. Because the reaction products, i.e. uranium aluminides (UAl_x), is less dense than the mixed reactants, the volume of the fuel meat increases after formation of interaction layer(IL). In addition to the impact on the swelling performance, the reaction layers between the U-Mo and Al matrix induces a degradation of the thermal conductivities of the U-Mo/Al dispersion fuels[4].

The chemical interaction between the U-Mo and Al matrix are analyzed in this study to find remedies to reduce the growth of the interaction layers during irradiation. In addition, various coating technologies for the formation of diffusion barriers on U-Mo particles are proposed as a result of the analyses.

2. Irradiation Tests Results

A series of irradiation tests, named the KOMO test, has been continued in KAERI since 2000 in order to qualify rod type U-Mo fuel with U-densities of 5-6 gU/cc by utilizing atomized U-Mo powder,. The purposes of the KOMO test are multiple; 1) upgrading HANARO research reactor to get a more compact core and a longer cycle life, 2) solving the back-end option of spent fuel, and 3) scientific contribution to understanding of U-Mo fuel performance. Post-irradiation test in the Irradiated Materials Examination Facility (IMEF) showed the variation of fuel swelling, interaction layer thicknesses and oxide thicknesses with fuel design parameters and irradiation conditions. The KOMO irradiation tests at HANARO have contributed to understand the effects of U-Mo particle size, U-Mo volume fraction, irradiation temperatures, and Si content in the Al matrix.

According to the previous KOMO-1,2,3 tests, the formation of interaction layers in rod-type U-Mo/Al dispersion fuels with U-loadings up to 5~6 gU/cc is very severe and inevitable due to the high temperature irradiation conditions. Although a use of large-sized U-Mo particles in dispersion fuel showed better performance by reducing the interfacial areas between the U-Mo and Al, this could not be an ultimate remedy to the interaction problem. Instead, small addition of Si up to 2wt% into Al matrix and a use of ternary U-7wt%Mo-1wt%Zr showed positive results in accordance with previous out-of-pile annealing tests [4].

KOMO-4 irradiation test of rod-type U-Mo-X(X=Zr,Ti)/Al-xSi(x=2,5,8wt%) dispersion fuel with 5.0 g-U/cc U loadings was designed in order to investigate the optimum Si content in matrix and the effects of alloying element addition(Ti, Zr) on U-Mo with a combination of Al-Si matrix at a high temperature condition of ~200^{o}C. Several U-7Mo/Al dispersion fuels with different fuel particle sizes (105~210 μm, 210~300 μm, and 300~425 μm) were also included to compare the IL growth behavior for each particle size. The KOMO-4 irradiation test bundle was loaded at OR-3 hole of the HANARO reactor on 22 Dec. 2008 and was discharged on 03 Jan. 2010 after 132.1 EFPD (Effective Full Power Day) irradiation.

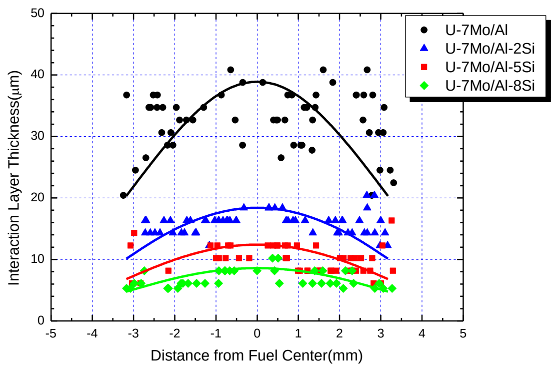


Fig.1. Interaction layer thickness distribution of irradiated U-Mo/Al-Si samples (~50% BU).

When the interaction layer thicknesses with the Si content in the Al matrix were compared, higher Si content in the matrix reduces interaction layer growth further as shown in Fig. 1. The average fuel temperature of the KOMO-4 test is around 200^{o}C, which is much higher than 120^{o}C of the RERTR-6 irradiation test conditions. The higher temperatures of the KOMO-4 test than those of the RERTR-6 test produced thicker ILs as compared in Fig. 2. Therefore it became clear that higher Si contents were needed to suppress the interaction layer growth in high temperature irradiation conditions.

Si was accumulated preferentially at the interaction layer/matrix interface and more Si was observed in the higher Si content added dispersion fuel rod. The Al/U ratios were less than 3 for all Si added dispersion fuel rods, while the ratios were more than 4 for the dispersion fuel rods without Si. Reduction of the population of Si precipitates in the recoil zone about 13 μm around the U-Mo particle is observed. Ti and Zr additions to U-Mo also reduce the interaction layer growth additionally, consistent with the lower temperature RERTR-8 test results.

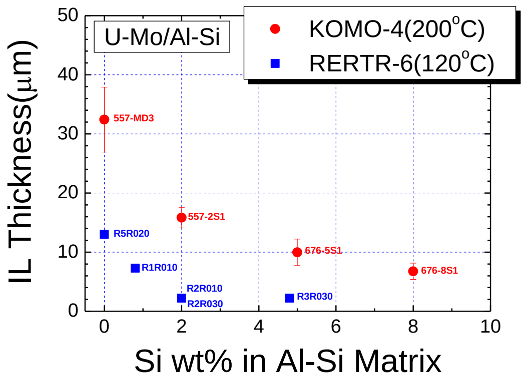
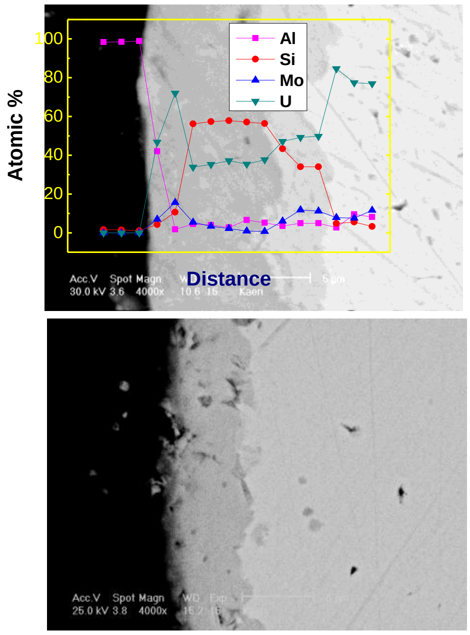


Fig. 2. Comparison of interaction layer(IL) thickness between the KOMO-4 and RERTR-6 tests.

3. Diffusion Barrier Coatings

Silicide or nitride coating technologies have been developed in KAERI in order to reduce the interaction between the U-Mo and Al matrix during irradiation[5]. The silicide or nitride coatings are expected to act as diffusion barrier layers on the U-Mo particle. Out-of-pile diffusion tests showed improved diffusion barrier performances of the silicide and nitride layers. Fig. 3 shows the microstructure of a silicide coated U-Mo particle and a nitride coated U-Mo particle.

In order to test the performance of diffusion barrier coated U-Mo particles, the KOMO-5 irradiation test is underway since May 23, 2011. Intermediate visual inspections on the KOMO-5 irradiated fuels revealed a sound fuel surface without any break-away swelling, as shown in Fig. 10[11]. It is expected that the irradiation test will be finished after attaining 60 at% U-235 burnup in May-June 2012 and the first PIE results of the KOMO-5 will be obtained in September-October 2012.



(a)

(b)

Fig. 3. SEM micrographs of (a) a silicide coated U-Mo particle and (b) a nitride coated U-Mo particle.

4. Conclusions

It has been proposed that diffusion barrier coatings should be applied on U-Mo particles in order to reduce the interaction layer growth between the U-Mo and Al matrix of U-Mo/Al dispersion fuel. Silicide or nitride layers formed on U-Mo particles have presented an excellent performance preventing chemical interaction in out-of-pile annealing tests. Currently, the KOMO-5 irradiation test for full size U-Mo/Al dispersion fuel rods is underway to investigate the irradiation behaviors of silicide or nitride coated U-7Mo/Al(-Si) dispersion fuels.

ACKNOWLEDGMENTS

This study was supported by the National Nuclear R&D Program of the Ministry of Education, Science and Technology (MEST) of Korea.

REFERENCES

[1] J.L. Snelgrove, G.L. Hofman, M.K. Meyer, C.L. Trybus and T.C. Wiencek, *Nucl. Eng. Des*., **178** (1997) 119.
[2] C.K. Kim, J.M. Park, and H.J. Ryu, *Nucl. Eng. Technol*., **39** (2007) 617.
[3] H.J. Ryu, Y.S. Kim, J.M Park, H.T. Chae, C.K. Kim, *Nucl. Eng. Technol.*, **40** (2008) 409.
[4] J.M. Park, H.J. Ryu, S.J. Oh, D.B. Lee, C.K. Kim, Y.S. Kim, G. L. Hofman, *J. Nucl. Mater*., 374 (2008) 422.
[5] H.J.Ryu, J.S.Park, J.M.Park, C.K.Kim, *Nucl. Eng. Technol*., **43** (2011) 159.