EFFECT OF REACTION PHASE GROWTH ON THE IRRADIATION PERFORMANCE OF ROD-TYPE U-MO/AL DISPERSION FUEL

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ABSTRACT

Interdiffusional reaction phase between U-Mo fuel particles and an Al matrix is known to degrade thermal properties of a U-Mo/Al dispersion fuel. Coupled with high power ratings and geometry, reaction phase growth in a rod-type dispersion fuel has more influence than in a plate type fuel. The feed-back effect between reaction phase growth and degradation of thermal properties and in turn fuel temperature increase for rod-type dispersion fuels were investigated. Radial temperature distribution due to reaction phase growth during irradiation was calculated iteratively by varying fission density, reaction phase volume fraction, thermal conductivity, and oxide thickness with burnup. Microstructural evolution estimated based on temperature calculations showed a reasonable agreement with the PIE results for rod-type U-Mo/Al dispersion fuels irradiated in the HANARO reactor under various power histories. The fuel particle size was revealed to be a dominant factor that determined the fuel temperature. For an average fuel particle size smaller than 75 μm an undesirable temperature jump that can lead to melting of the Al matrix was predicted, whereas for a fuel particle size larger than 150 μm the fuel temperature gradually decreased toward the end of life without a break-away temperature increase.

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1. Introduction

Irradiation behavior of U-Mo/Al dispersion fuel has been investigated to develop high performance research reactor fuel[1]. Rod-type dispersion fuel has been used as driver fuel of the HANARO research reactor in KAERI[2]. Rod-type fuel has different heat dissipation routes compared to plate-type fuel. Temperature distribution at the beginning-of-life (BOL) can be

calculated based on the initial condition of irradiation and as-fabricated microstructure of fuel material. Centerline temperature of a dispersion fuel rod goes up to high temperature when thermal conductivity of fuel meat becomes low. Unfortunately, thermal conductivity of fuel meat decreases during the irradiation mainly because of interaction layer formation at the interface between the U-Mo fuel particle and Al matrix. The thermal conductivity of the interaction layer is not as high as the Al matrix. The growth of interaction layer is interactively affected by the temperature of fuel because it is associated with a diffusional reaction which is a thermally activated process. It is difficult to estimate the temperature profile during irradiation test due to the interdependency of fuel temperature and thermal conductivity changed by interaction layer growth. PLATE code is developed for the performance evaluation of plate-type fuel[3], but there is no counterpart for rod-type dispersion fuel. In this study, temperature histories of rod-type U-Mo/Al dispersion fuels during irradiation tests were estimated by considering the effect of interaction layer growth on the thermal conductivity of fuel meat.

2. Analysis Procedures

The temperature histories of the second irradiation tests (KOMO-2) in HANARO were analyzed by using reactor operation histories, fuel fabrication specifications and post-irradiation examination (PIE) results[4]. KOMO-2 tested a U-7Mo/Al dispersion fuel of 6.35 mm in diameter with 4.5 g-U/cc (494L2) and a U-7Mo/Al dispersion fuel of 5.49 mm in diameter with 4.5 gU/cc (494H2) in 2003 and KOMO-2 was irradiated up to 71.2 at% U-235 burn-up without the break-away swelling. PIE was carried out in the Irradiation Material Examination Facility (IMEF) facility in KAERI in 2004. Radial microstructure of the 494H2 fuel rod from clad to center of fuel rod at end-of-life (EOL) was shown in Fig. 1. Dispersion fuel near to center of fuel rod became fully reacted zone and the Al matrix is completely consumed by the reaction. End-of-life temperature was calculated by using the microstructural information obtained from the post irradiation examination. Fuel rod was simulated into four concentric rings with different thermal conductivity according to volume fraction of the interaction layer for the calculation by an axisymmetric finite element analysis as shown in Fig. 2.

Thermal conductivity of fuel meat was calculated based on the modified Hashin and Shtrikman relation developed by CEA and employed in the PLATE code as shown in Eq.(1).

$$k = \frac{-k_f + 3V_f + 2k_m - 3V_f k_m}{4} + \frac{\sqrt{8k_f k_m + (k_f - 3V_f k_f - 2k_m + 3V_f k_m)^2}}{4} \qquad (1)$$

Interaction layer thickness was calculated based on a well known correlation used in the PLATE code as shown in Eq. (2),

$$y^2 = 2.2442 \times 10^{-19} \cdot (1.625 - 6.25W_{Mo}) \cdot \dot{f} \cdot \Delta t \cdot \exp\left(\frac{-10000}{RT}\right) \qquad (2)$$

where y is the interaction layer thickness (cm), W_{Mo} is the weight fraction of Mo in the U-Mo fuel, $\dot{f}$ is the fission rate density (f/cm^3-sec), Δt is time (sec), R is the ideal gas constant (1.987 cal/mole-K), and T is absolute temperature (K). Thermal conductivity of U-Mo alloys used in the calculation is a function of temperature as shown in Eq.(3).

$$k_{U-Mo} = 0.034T - 0.56 \text{ (W/mK)} \qquad (3)$$

Figure 3 shows a flow chart for fuel temperature calculation procedures of this study. First, interaction layer thickness corresponding to each fission density was calculated according to the

PLATE correlation for interaction layer growth. After considering the swelling from fission gas bubble, fission product, and interaction layer, thermal conductivity of the dispersion fuel meat was calculated by considering volume fraction of interaction layer, U-Mo fuel and Al matrix at each fission density. Fuel temperature was obtained from a cylindrical heat transfer equation by using the estimated thermal conductivity of the dispersion fuel meat, Al clad, and oxide film. Oxide film thickness outside of the clad was obtained from the Griess model[5].

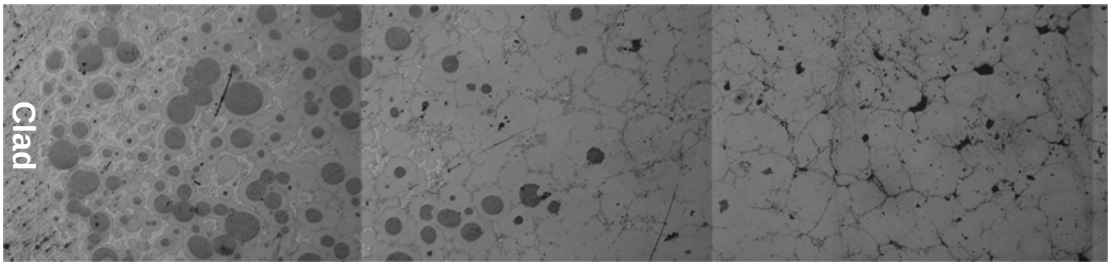


Fig. 1. End-of-life microstructure of 494H2 fuel. (Left side is clad and right side is center of fuel rod.)

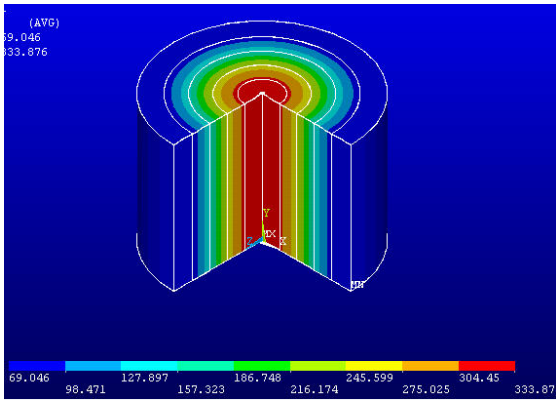


Fig. 2. Temperature calculation for 494H2 using microstructure-based axisymmetric finite element method.

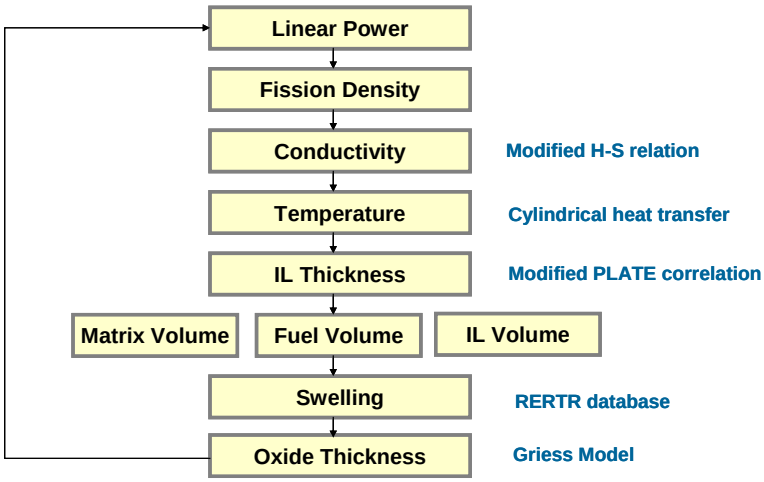


Fig. 3. Flow chart of fuel temperature calculation procedures.

2. Results and Discussion

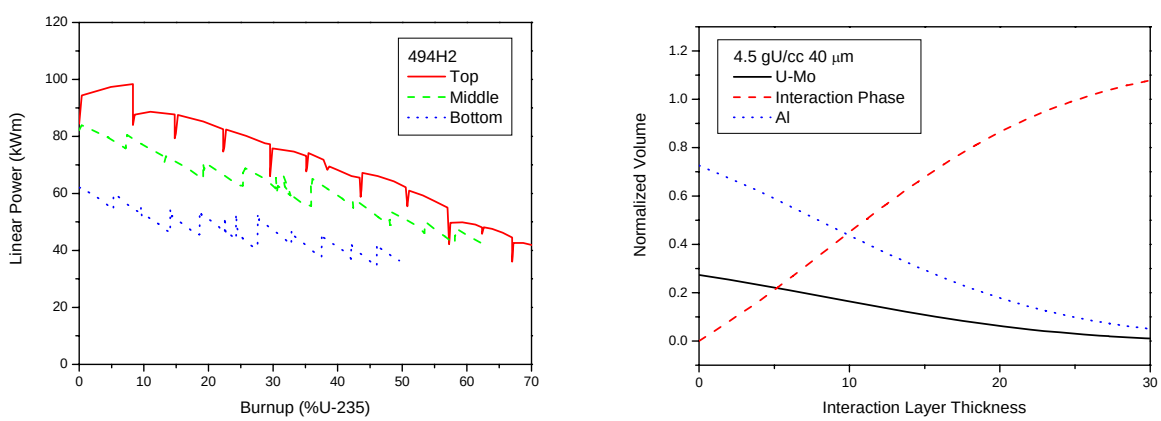
HANARO irradiation test condition for rod-type U-Mo/Al dispersion fuel was quite different from RERTR irradiation test. Fission density (~1.36x10^{21} f/cm^3) and fission rate (~1.26x10^{14} f/cm^3-s) for HANARO irradiation were lower than RERTR irradiation test. Fuel temperature of rod-type dispersion fuel was higher than plate-type fuel, and then the volume fraction of interaction layer can be increased up to 100%. Therefore the PLATE correlation for interaction layer growth developed from the results of RERTR irradiation test needs a modification. A constant was introduced to modify the correlation by comparing the end-of-life fuel temperature estimated from the calculation presented in this study and the microstructure-based finite element analysis. Factor of 2 was selected as a multiplying constant as shown in Eq.(4) by considering the end-of-life temperatures of top section and middle section of the 494H2 fuel rod as listed in Table 1. This constant can be corrected after in-detail verification study by using interaction layer thickness, swelling, and oxide thickness of irradiated dispersion fuel.

$$y^2 = 2 \times 2.2442 \times 10^{-19} \cdot (1.625 - 6.25 W_{Mo}) \cdot \dot{f} \cdot \Delta t \cdot \exp\left(\frac{-10000}{RT}\right) \qquad (4)$$

Fuel temperature histories with increasing burnup were calculated for the 494H2 fuel rod by using the modified correlation. Linear power histories for each fuel and each section in a fuel rod with burnup were plotted in Fig. 4(a). Fig. 4(b) represents the variation of normalized volume of U-Mo, interaction layer and Al matrix with increasing interaction layer thickness.

Table 1. Comparison of end-of-life temperature calculated by microstructure-based finite element method and temperature calculation developed in this study.

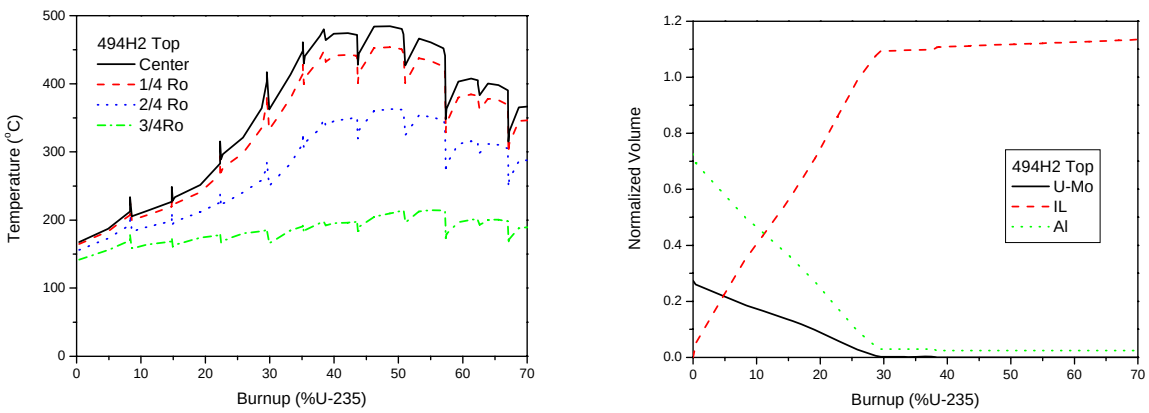
Calculation Methods	494H2 Top	494H2 Middle
FEM based on Microstructural Information of End of Life	348°C	290 ^{o}C
This Study	360 ^{o}C	265 ^{o}C



(a) (b)

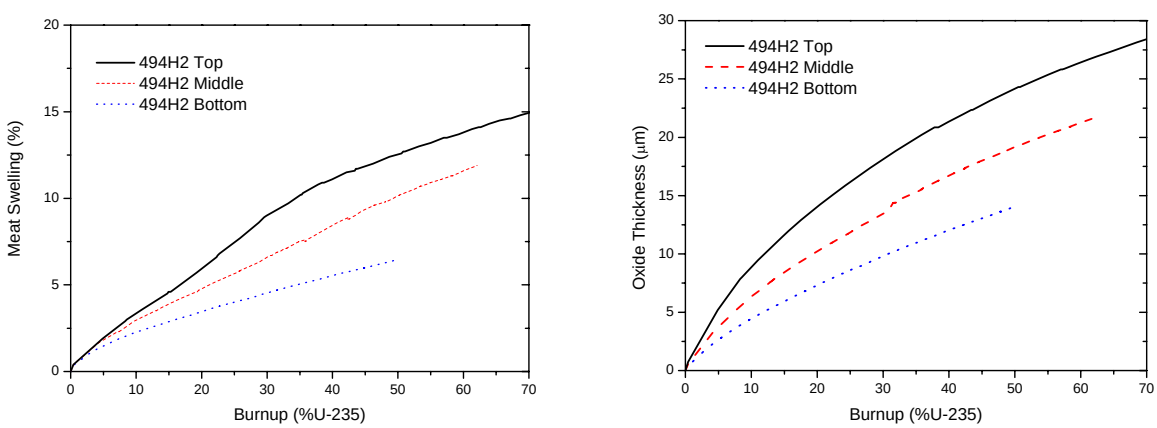
Fig. 4. (a) Linear power histories of top, middle and bottom section of 494H2 fuel rods in the KOMO-2 irradiation test.(b) The variation of volume fraction of interaction layer, U-Mo fuel, and Al matrix.

Figure 5(a) shows the temperature histories for top section of the 494H2 rod. Centerline temperature increased sharply after 20% U-235 burnup and reached a peak temperature around 480°C after 40% U-235 burnup. The variation of normalized volume of U-Mo fuel, interaction layer and Al matrix was plotted in Fig. 5(b). Circumferential zone ($3/4r_o$) adjacent to clad did not showed an abrupt temperature jump similar to plate-type dispersion fuel because the lower temperature in the region keeps the interaction layer thinner and the thermal conductivity lower. U-Mo fuel and Al was completely consumed after 30% U-235 burnup as shown in Fig. 5(b). Then thermal conductivity did not change much after whole area of dispersion fuel meat is converted to interaction layer and fuel temperature simply follow the trend of linear power history. Swelling of fuel meat calculated according to the RERTR irradiation test results [3], and the oxide thickness of clad calculated by the Griess model increased as a function of linear power and burnup as shown in Fig. 6.



(a) (b)

Fig. 5. Variation of (a) fuel temperature and (b) normalized volume of U-Mo, interaction layer and Al matrix bat top section of a 494H2 fuel rod in the KOMO-2 irradiation test



(a) (b)

Fig. 6. Variation of (a) fuel meat swelling and (b) oxide thickness at clad surface in top, middle and bottom sections of a 494H2 fuel rod in the KOMO-2 irradiation test.

Figure 7 shows the effect of fuel particle size on the centerline temperature and meat swelling when the average diameter of U-Mo fuel particles in 4.5 gU/cc dispersion fuel varies from 35 μm to 150 μm. Centerline temperature and meat swelling was decreased by the increase in the average diameter of fuel particles because the specific interfacial area for interaction decreases and then volume fraction of less conducting interaction layer increases slower. The effect of fuel particle size was already experimentally confirmed in the KOMO-2 irradiation test as presented in Fig. 8. When the end-of-life microstructure of U-Mo/Al dispersion fuel with small-sized fuel particles (33 μm) and large-sized fuel particles (74 μm) was compared, dispersion fuel with large-sized particles exhibited less volume fraction of interaction layer. Completely reacted zone was expanded to 3/4 r_o for dispersion fuel with small-sized particle, whereas only center to 1/4 r_o area was converted in the dispersion fuel with large-sized particle.

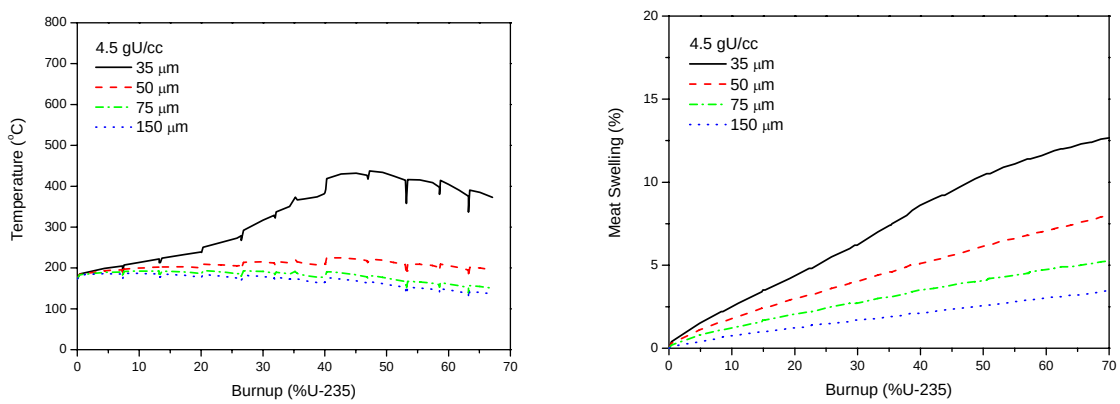
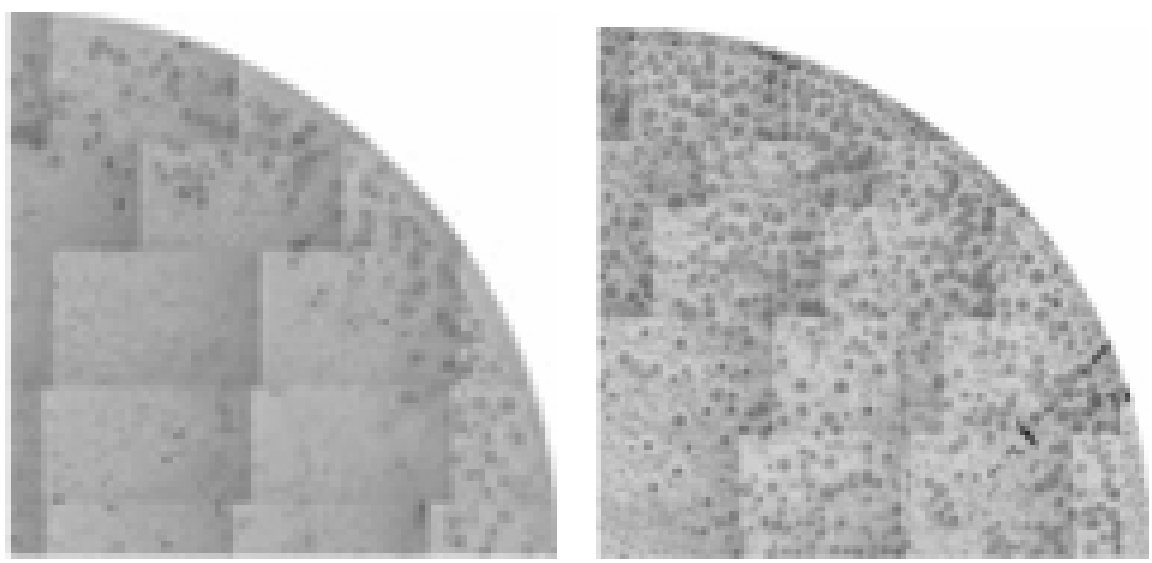


Fig. 7. Variation of (a) fuel temperature and (b) fuel meat swelling of a 4.5 gU/cc dispersion fuel as a function of fuel particle size.



(a) (b)

Fig. 8. End-of-life microstructure of dispersion fuel with (a) small fuel particles (33 μm) and (b) large fuel particles(74 μm) in the KOMO-2 irradiation test.

Effect of uranium loading on the thermal conductivity and centerline temperature of U-Mo/Al dispersion fuel with a 494L2 irradiation condition was calculated as plotted in Fig. 9. A dispersion fuel rod with higher uranium loading density keeps lower thermal conductivity as burnup proceeds. The maximum centerline temperature increased with increasing uranium loading density. When the uranium loading density is higher than 6 gU/cc, fuel temperature was estimated to exceed the melting temperature of the Al matrix.

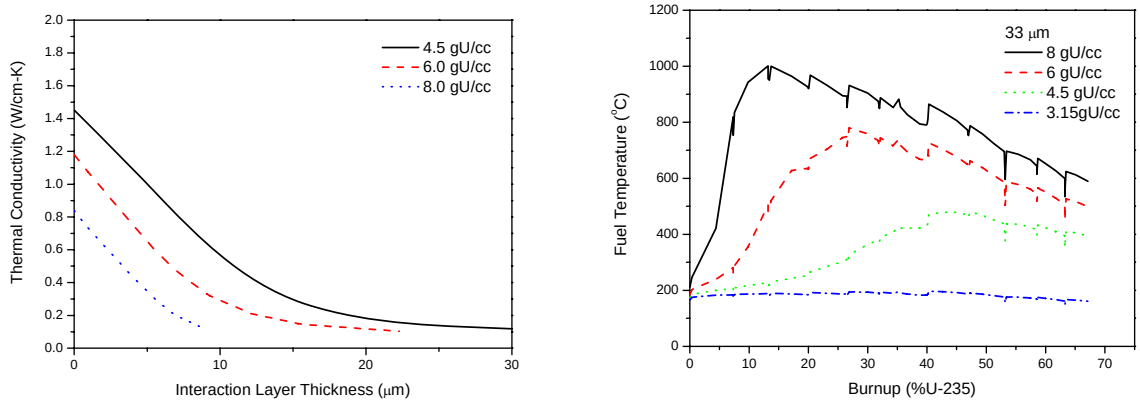
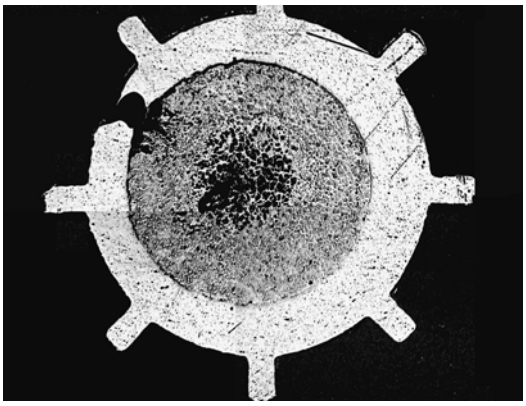
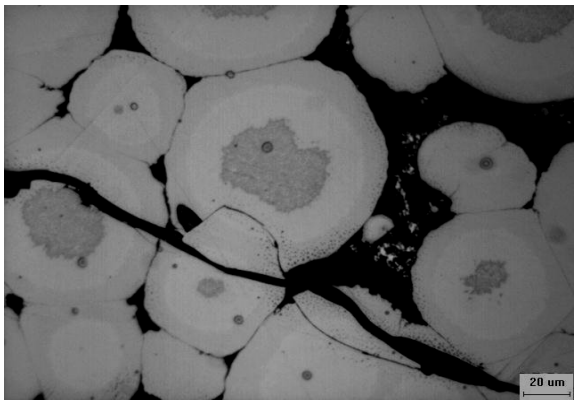


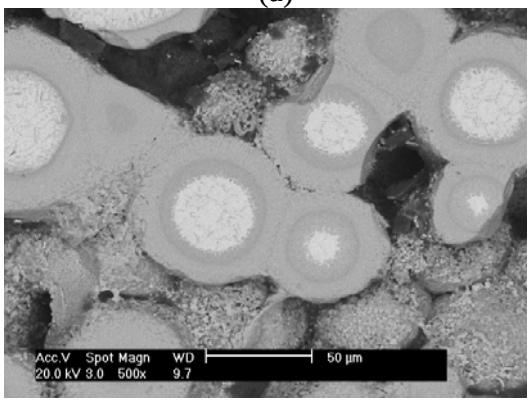
Fig. 9. Effect of uranium loading on (a) the thermal conductivity of fuel meat with increasing interaction layer thickness and (b) the variation of fuel temperature with increasing burnup.



(a)



(b)



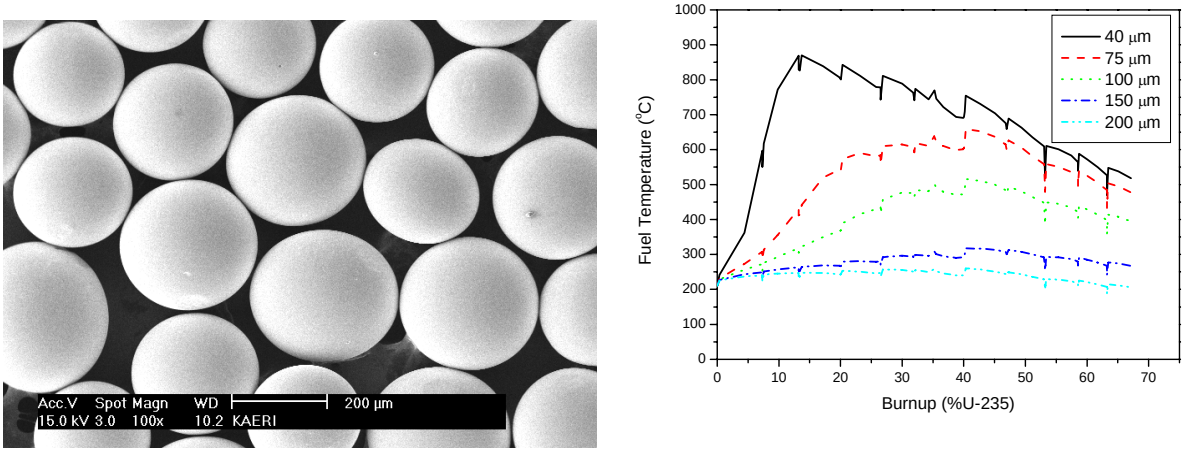
(c)

Fig. 10. (a) A cross-section image of a failed fuel rod of KOMO-1 test and (b) a scanning electron micrograph of a failed fuel rod of KOMO-1 test. (c) A scanning electron micrograph of out-of-file annealing test at 600°C for 24 hrs.

Figure 10(a) shows a cross-section image of a dispersion fuel rod with 6 gU/cc failed at 8% burnup in the KOMO-1 irradiation test[6]. When a scanning electron micrograph for that fuel meat was compared with an out-of-pile annealing tested dispersion fuel meat at 600°C for 24 hrs, microstructural similarity such as multiple ring and absence of Al matrix was found as shown in Fig. 10(b) and Fig. 10(c). This result supports that the fuel temperature of a dispersion fuel with 6 gU/cc increases abruptly as calculated in this study and the failure of the fuel rod in the KOMO-1 irradiation test is attributed to temperature jump related volume expansion of fuel meat due to the abrupt formation of interaction layer.

To overcome the limitation of uranium loading density in U-Mo/Al dispersion fuel, very large U-Mo particle with 100-500 μm in diameter were fabricated by centrifugal atomization process as shown in Fig. 11(a)[7]. Centerline temperature of U-Mo/Al dispersion fuel with 8 gU/cc was calculated with varying average fuel particle size from 40 μm to 200 μm as plotted in Fig. 11(b). When the average fuel particle size is ranging from 40 to 100 mm, undesirable temperature rises were predicted due to the active interaction layer growth, while stable fuel temperature history was calculated when the average fuel particle size is larger than 150 μm. Dispersion of U-Mo fuel particle larger than 150 μm is expected to be a remedy to release temperature-related problems in rod-type dispersion fuel with a high uranium loading density.

Verification and validation of the fuel temperature calculation program developed in this study will be carried out further by using various post-irradiation examination results obtained from the HANARO irradiation tests. This performance analysis program for rod-type dispersion fuel will be used for a safety analysis of the KOMO-3 irradiation test focusing on the dispersion fuel with very large fuel particles up to 500 μm.



(a) (b)

Fig. 11. (a) Morphology of atomized U-7Mo particles larger than 150 μm. (b) Calculated fuel temperature of 8 gU/cc U-Mo/Al dispersion fuel with varying fuel particle size.

4. Conclusions

Fuel temperature history of rod-type U-Mo/Al dispersion fuel was calculated by using a modified correlation of interaction layer growth. Fuel temperature of rod-type dispersion fuel showed strong feedback behavior due to the lower thermal conductivity of the interaction layer. As the uranium loading density increases up to 8 gU/cc, unacceptable temperature increase above 600°C was predicted. It was found that dispersion of large fuel particle (>150 μm) was effective in mitigating thermal degradation associated with the interaction layer.

Acknowledgements

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