Analysis of measured thermophysical properties of U-Mo/Al dispersion fuels

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1. Introduction

The thermal conductivity of U-Mo/Al dispersion fuel is one of the important physical properties affecting the overall fuel performance by determining fuel temperature during operation [1]. However, its measured thermophysical data are scarce due to difficulty in sample fabrication and measurement. The thermophysical properties of U-Mo/Al dispersion fuel are dependent on the composition of the constituents, U loadings (or U density), interaction layer (IL) thickness, particle size, porosity, etc. The effects of them, however, are not clearly understood.

Some measured data are available in the literature [3] - [7]. ANL [2] reported the thermal conductivity data of U-Mo/Al samples fabricated with ground U-Mo powders, which had irregular particle shapes. Therefore, the particle-matrix bonding was less compact, and the porosity might be higher although the details were not reported. Therefore, this data could be used to provide an overall trend of thermal conductivity. Lee et al. [3] measured thermal properties of U-xMo/Al (x=6, 8, 10 wt.%) fabricated by using a hot-extrusion method in typically used ranges of U-Mo volume fraction and temperature. However, the dataset showed a large scatter and some unrealistic trends, which limits the use of the dataset. The possible cause was attributed to the poor homogeneity of the U-Mo particle distribution that produced a considerable uncertainty in U-Mo volume fraction in the samples. Huber [5] measured the thermal conductivities of several types of samples, cut from fuel plates. Huber included un-irradiated dispersion fuel samples that were different in powder types (ground or atomization), particle size, Mo content, Si content, U loading, porosity, and coating material. Huber also measured the thermal conductivity of irradiated fuel samples. From the dataset, an overall trend for each variable could be observed. However, U-loading and IL formation were not clearly characterized.

In this study, we measured the thermophysical properties of U-7Mo/Al and U-7Mo/Al-5Si fuels having U loadings of 5.0, 6.5, and 8.0 gU/cm^3 [6]. Some of the U-7Mo/Al fuel samples were heat-treated to measure the thermophysical properties of IL-formed U-Mo/Al [7].

2. Experimental methods

The typical method of the fabrication of a U-Mo/Al dispersion fuel plate is hot-rolling at ~500 °C. This method, however, produces 2 - 5 % porosity in the sample and ILs. The pore size is ~2 μm. The presence of pores and IL, however, are unfavorable for the measurement of thermal properties. In this study, therefore, an extrusion method at 400 °C was used to minimize porosity and IL formation in the samples. Another advantage of this method is to produce a good particle homogeneity in the samples.

Both atomized U-7Mo and Al(-5Si) powders were prepared at KAERI. The powder conditions followed the previous criteria applied to produce the HAMP-2 and HAMP-3 mini-plates such that particles are smaller than 150 μm. They were mixed in various U densities of 5.0, 6.5, and 8.0 g-/cm3 using a shake mixer. Each mixture was compacted into 8 mini-plates at room temperature. The compactions were hot-extruded at 400 °C in an Ar gas environment. This hot-extrusion step was repeated at least more than 3 times to increase particle homogeneity. A total of 6 different fuel types were prepared and subsequently machined for the sample sizes required as follows:

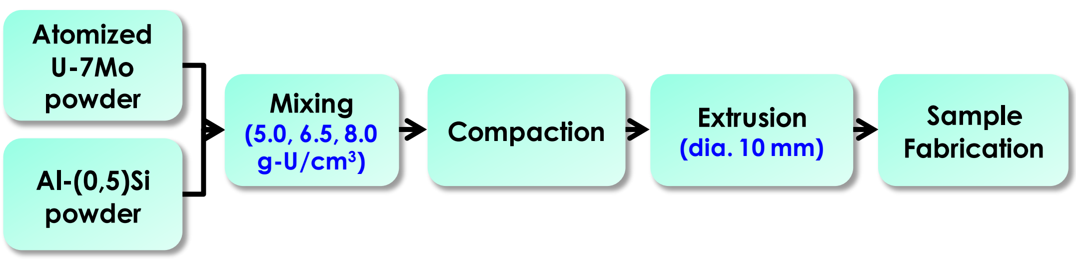


Fig. 1. A schematic showing sample preparation

1) Laser Flash Measurement
   - Diameter: ~ 10 mm, Thickness: 1.8 - 2.2 mm

2) DSC Measurement
   - Diameter: ~ 6 mm, Thickness: 0.5 – 1.0 mm

Microstructure images of all samples were obtained before the measurements. Fig. 2 shows SEM images of the as-fabricated U-7Mo/Al and U-7Mo/Al-5Si dispersion samples of 5.0, 6.5, 8.0 gU/cm^3 U-loadings, respectively. It is seen that the U-Mo particles were distributed with high homogeneity without IL and pores. Fig. 3 shows SEM images of heat-treated U-7Mo/Al dispersion samples at 530 °C for 8 hours. The grey areas are the ILs, which formed uniformly around U-Mo particles without pores due to Al matrix depletion.

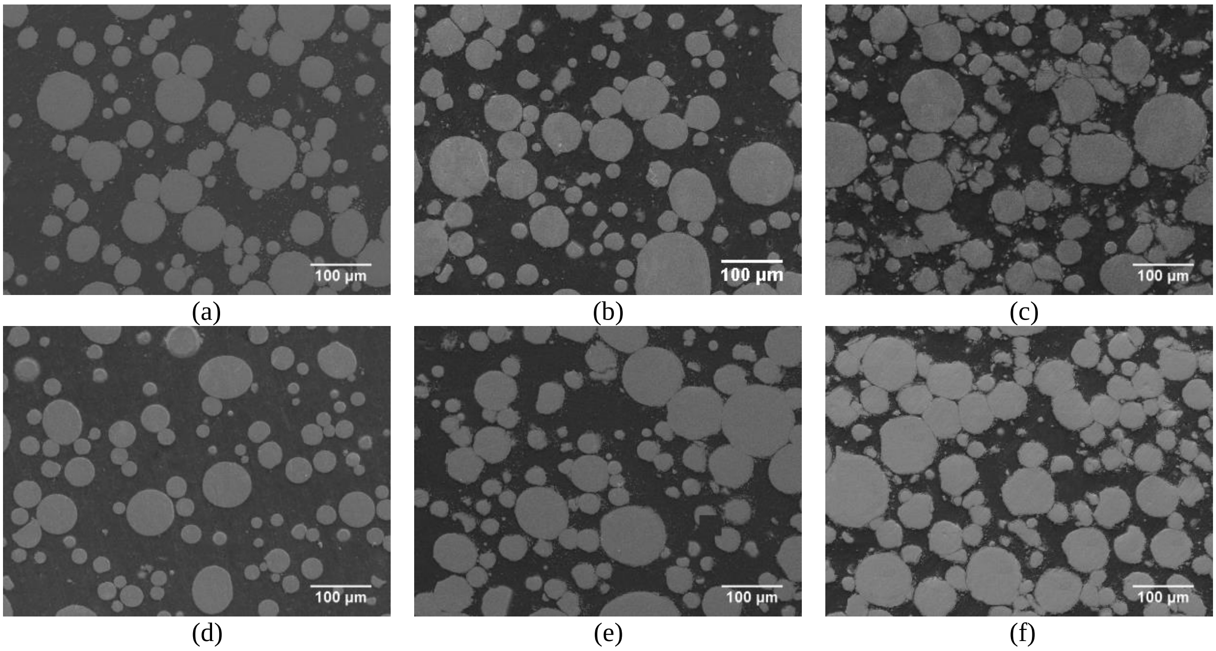


Fig. 2. SEM images of as-fabricated U-7Mo/Al fuels: (a) 5.0 g-U/cm^3, (b) 6.5 g-U/cm^3, and (c) 8.0 g-U/cm^3 and U-7Mo/Al-5Si fuels: (d) 5.0 g-U/cm^3, (e) 6.5 g-U/cm^3, and (f) 8.0 g-U/cm^3

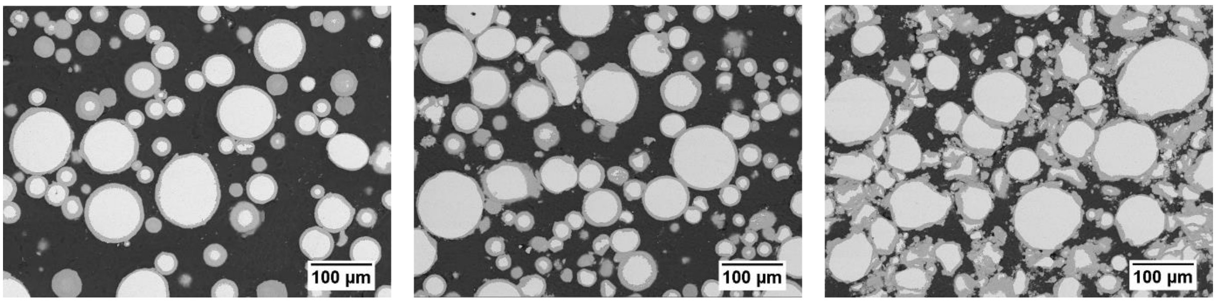
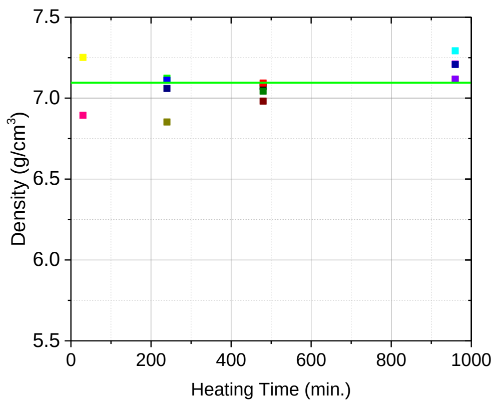


Fig. 3. SEM images of heat-treated U-7Mo/Al dispersion fuels at 530 °C for 8 hours: (a) 5.0 g-U/cm^3, (b) 6.5 g-U/cm^3, and (c) 8.0 g-U/cm^3 [7].

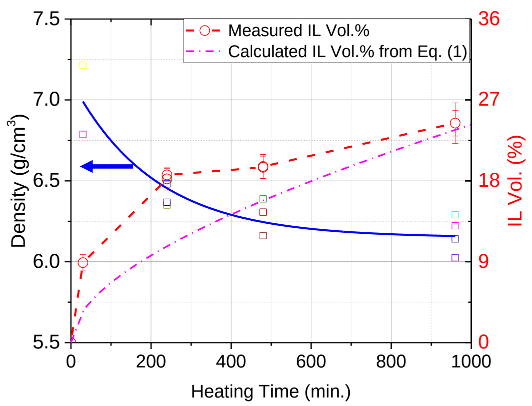
Fig. 4 shows the changes of density and IL volume fraction as a function of heating time when heated at 500 °C. The densities of as-fabricated samples are shown in Fig. 4 (a) in which the averaged value is ~7.0 gU/cm^3. The densities and IL volume fractions of heat-treated samples are shown in Fig. 4 (b). As expected, the density decreased exponentially, while the IL volume fraction increased exponentially. The IL volume fraction was compared with the theoretical models. The IL growth model in our-of-pile was suggested as follows [8]:

$$Y_{Al}^2 = 1.5\times10^{18}\exp\left(-\frac{38000}{T}\right)t \tag{1}$$

where Y_{Al} is the IL thickness (μm) in an Al matrix, T is the temperature in K and t is the time in s. The calculated IL thickenss can be converted to IL volume fraction by using the FCC model described in [8]. As seen in Fig. 4, the theoretical model estimated slightly lower IL volume fractions.



(a) Density before heat-treatment



(b) Density and IL volume percent after heat-treatment.

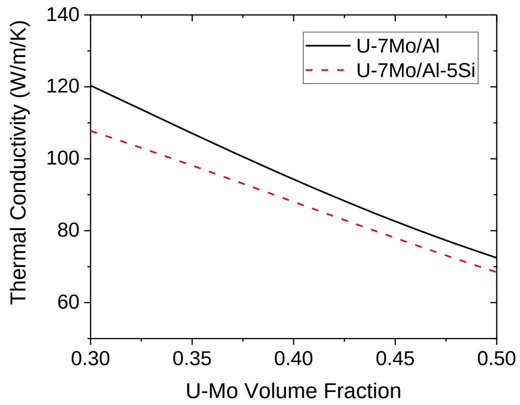
Fig. 4. Density and IL volume fraction of 5.0 g-U/cm³ U-7Mo/Al as a function of heating time at 500 °C

Thermal diffusivity, heat capacity, and density of samples the were measured. The thermal diffusivity (α) was measured at 30, 50, 100, 150, 200, 250, 300, 350, and 400 °C using a laser flash method (LFA427 by Netzsch). The heat capacity was meausred over a temperature range of 30 to 400 °C using DSC404C (Netzsch). The measurements were carried out at a heating rate of 5K/min in an argon atmosphere with a flow rate of 50 ml/min. The density was meausred at room temperature by adopting the Archimedes' method. The density at temperatures higher than room temperature was calculated by using the thermal expansion coefficients of fuel components [6], [7].

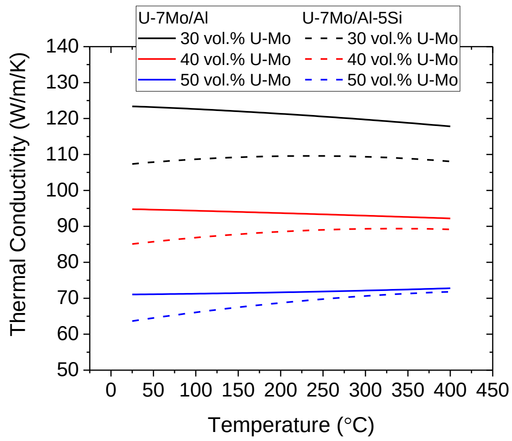
3. Thermal Conductivity

Using the measured data, the thermal conductivity of U-Mo/Al dispersion fuel was calculated as a function of temperature and U-Mo volume fraction. Fig. 5 shows the thermal conductivity of U-7Mo/Al and U-7Mo/Al-5Si dispersion fuels. The effects of U loading, Si addition to Al matrix, and temperature are observed as follows:

- The thermal conductivity decreases with U-Mo volume fraction linearly in the volume fraction range of 0.30 – 0.50.
- The addition of Si reduces the thermal conductivity of the fuels. However, the effect is weakened as the U loadings increases and temperature increases.
- The thermal conductivity increases or decreases with temperature depending on the amount of U-Mo and Al(-Si) volume fraction. For lower U loadings (i.e. less than 5.0 gU/cm3), the thermal conductivity generally decreases with temperature. For higher U loadings, the thermal conductivity increases with temperature.



(a)



(b)

Fig. 5. Thermal conductivity of U-7Mo/Al and U-7Mo/Al-5Si dispersion fuels (a) as a function of U-Mo volume fraction at 150 °C, and (b) as a function of temperature.

The effects of IL were evaluated using the measured data as well. Fig. 6 shows the thermal conductivity of U-7Mo/Al as a function of IL volume fraction assuming the consumptions of U-Mo and Al. The thermal conductivity decreases linearly with IL volume fraction regardless of U-Mo volume fractions.

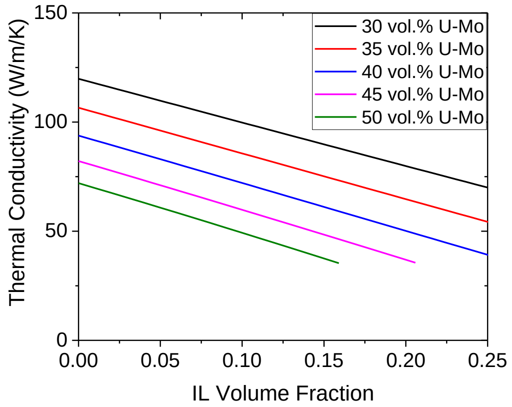
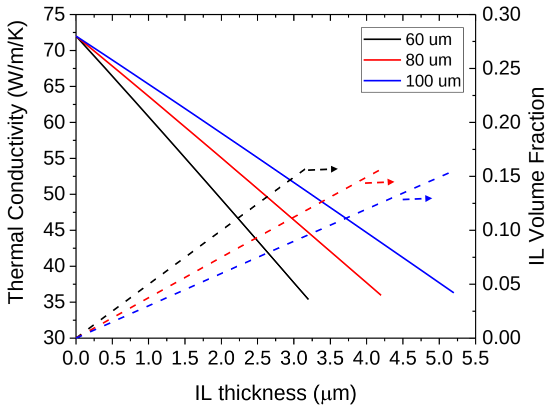
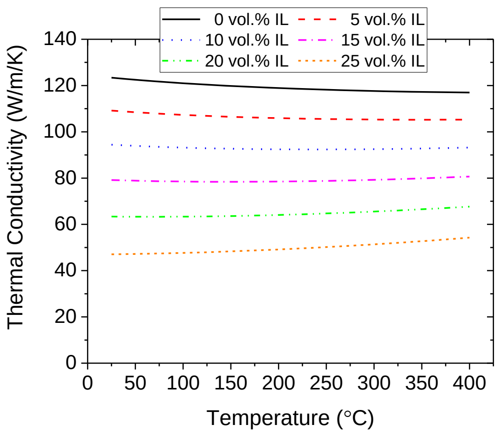


Fig. 6. Thermal conductivity of U-Mo/Al dispersion fuels as a function of IL volume fraction.



(a)



(b)

Fig. 7. Thermal conductivity of U-Mo/Al dispersion fuels: (a) as a function of IL thickness when U-Mo volume fraction is 0.50 with several sizes of U-Mo particle (60, 80, 100 μm), and (b) as a function of temperature with different IL volume fractions when U-Mo volume fraction is 0.30.

Fig. 7 (a) shows the effect of particle size fixing the U-Mo volume fraction at 0.50. The smaller the particle size, the IL volume fraction becomes larger at the same IL thickness. Consequently, the sample with smaller U-Mo particles has lower thermal conductivity when IL grows. Fig. 7 (b) shows the effect of temperature with different IL volume fractions fixing the U-Mo volume fraction at 0.30. When IL does not exist, the thermal conductivity decreases with temperature. However, as the IL volume fraction increases, the thermal conductivity itself decreases, but it's temperature trend changes to increase. This is mainly due to the consumption of the Al matrix.

3. Conclusions

The thermal conductivity of U-7Mo/Al(-5Si) dispersion fuels were evaluated by using newly measured data. The addition of 5wt.% Si to Al matrix decreases the thermal conductivity of fuels while its effect decreases as the uranium loadings and temperature increase. The volume fractions of fuel constituents are the major factors determining the thermal conductivity. The thermal conductivity decreases linearly with U-Mo and IL volume fractions in the measured volume fraction range (U-Mo volume fraction of 0.30 – 0.50 and IL volume fraction of 0 – 0.25). The particle size effect on thermal conductivity was found by a modeling. As the particle size decreases, the IL volume fraction becomes larger at the same IL thickness. These differences of IL volume fraction cause a considerable difference in thermal conductivity. Thermal conductivity changes to an increasing function of temperature as U-Mo and IL volume fractions increase.

ACKNOWLEDGMENT

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