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Thermal Property Simulation and Modeling for Irradiated U-Mo Dispersion Fuel: to predict thermal conductivity of interaction layer

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1. Introduction

To minimize nuclear proliferation; high-enriched uranium (HEU) has to be replaced with low-enriched uranium (LEU). However, this replacement deteriorates the levels of power and neutron flux. In order to use LEU, increasing uranium loading is required. U-Mo/Al dispersion fuel has been the most potential candidate since the U-Mo alloy can increase uranium loading significantly [1].

The problem of U-Mo/Al dispersion type fuel is the formation of interaction layer (IL) between U-Mo particles and aluminum matrix. When the dispersion fuel is irradiated, the fuel particles react with the matrix and form IL. The composition of the IL is complex. The microstructural analysis of the un-irradiated U-7Mo/Al-2Si showed that the chemical composition of the IL was dependent on the annealing temperature where a mixture of (U, Mo) Al_x-Si_x was formed [2]. After irradiation, the IL becomes amorphous and its composition is unknown [3]. Some properties including density and chemical composition that provided the best fitting for the measured fuel meat swelling were recently provided in Ref. 4. The thermal conductivity of IL is critical on the fuel performance during irradiation. The objective of this study is to predict the thermal conductivity of IL.

2. Simulation modeling

The simulation was performed using “COMSOL Multiphysics” version 5.1 finite element analysis (FEA) software [5]. Two segments were selected as basic models for the simulation; one was irradiated at a low burn-up of 5.195×10^{21} f/cm^3 (referred as TL) and the other was irradiated at a high burn-up of 6.49×10^{21} f/cm^3 (referred as TK) [8]. To determine dimensions of each model, the volume fraction of fuel particles, the IL, and the matrix (if applicable) were calculated using “ImageJ” software [6] from the images of TL and TK provided in Ref. 8.

Figure 1 showed the optical microscopy images of the TL segment [8]. The matrix was highlighted with a red color in the figure. The average area percentages of fuel particles, IL, and matrix calculated by ImageJ were approximately 60.88, 31.78, and 7.34.

The optical microscopy images of TK segment [8] were shown in Figure 2. The average calculated percentages of the fuel particles and the IL were equal to the reported data from Burkes et al. [8]. Table 1 showed the dimensions of the simulated models representing TL and TK segments.

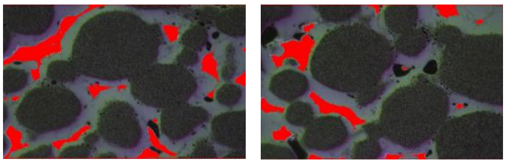

Figure 1. Optical metallographs of TL segment [8]

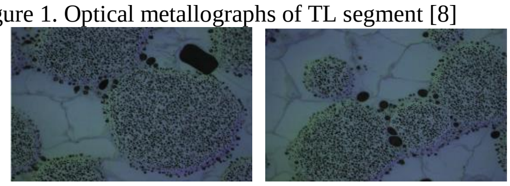

Figure 2. Optical metallographs of TK segment [8]

Table 1: The characteristics of irradiated fuel segments

	TL	TK
Fuel particle vol. %	60.88	49.1
IL vol. %	31.78	50.9
Matrix vol. %	7.34	0
Fuel particle fission density($\times 10^{21}$ f/cm^3)	5.195	6.49

2.1 Thermo-physical properties of irradiated materials

For the simulation, U-7wt.%Mo and Al-2wt.%Si were used as the fuel and matrix, respectively. Since IL was formed by reaction between the fuel particles and the matrix during the irradiation, this model assumed that IL consisted of UAl_3 and UAl_4. Their properties including density, heat capacity, and thermal conductivity were obtained as flows.

2.1.1 Density

According to Kim et al. [4], the density of U-Mo fuel particles and IL were 17.7 g/cm^3 and 5.71 g/cm^3, respectively. The value of pure aluminum density, 2.7 g/cm^3, was used for the matrix [7].

2.1.2 Heat Capacity

The heat capacities for the fuel particles and IL were temperature dependent. Therefore, functions of temperature had to be applied. Following equation was used for the fuel heat capacity [7]:

$$Cp_{fuel}\left[\frac{J}{kgK}\right] = (29.84 - (8.9\times10^{-3})\mathrm{T} + (4.32\times10^{-5})T^2 - (2.06\times10^{-8})T^3)\ /\ 0.2152, \quad (1)$$

where T was operating temperature in K.

Since UAl_3 and UAl_4 for IL had different heat capacity equations [9], a new correlation was derived from the average value from these two equations within 50-300℃. The new equation was described as follows:

$$Cp_{IL}\left[\frac{J}{kgK}\right] = 0.225\times T + 401, \quad (2)$$

where T was operating temperature in ℃.

Heat capacity of the aluminum matrix, however, was independent from the operating temperature. Constant value of 900 J/kgK was used for the matrix heat capacity [7].

2.1.3 Thermal Conductivity

The model for thermal conductivity of U-Mo particles was derived by Burkes et al. [10] throughout Equations 3-10. The first step was to calculate thermal conductivity of un-irradiated U-Mo alloy using Equation 3 to 6.

$$k^0_{U-Mo} = \left(1-\sqrt{1-x_{Mo}}\right)k_{Mo} + \sqrt{1-}\left((1-x_{Mo})k_U + (x_{Mo})k_{c,\ Mo}\right), \quad (3)$$

$$k_U(T) = 21.73 + (1.591\times10^{-2})T + (5.907\times10^{-6})T^2, \quad (4)$$

$$k_{Mo}(T) = 150.0 - (4\times10^{-2})T\ , \quad (5)$$

$$k_{c,Mo}(T) = -274.4 + 985.2x_{Mo} - (1.941\times10^3){x_{Mo}}^2 + (3.64\times10^{-2})T + (7.365\times10^{-5})T^2 + (5.793\times10^{-2})x_{Mo}T, \quad (6)$$

where k^0_{U-Mo} was the thermal conductivity of the un-irradiated fuel, k_{Mo} and k_U were the thermal conductivities of Mo and U as functions of temperature. x_{Mo} was Mo concentration in weight percent and T was the operating temperature in K.

Then, Equations 7-10 determined thermal conductivity of irradiated U-Mo alloy (k_{U-Mo}).

$$k_{U-Mo} = .25\left(A + \sqrt{A^2 + 8k^0_{U-Mo}k_g}\right), \quad (7)$$

$$\mathrm{A} = (2-3P)k^0_{U-Mo} + (3P-1)k_g, \quad (8)$$

$$k_g = 0.1\ (8.247\times10^{-5}T^{0.8363}) + .9(4.351\times10^{-5}T^{0.8616}), \quad (9)$$

$$\mathrm{P} = \left(\frac{\Delta V}{V}\right)_G + \left(\frac{\Delta V}{V}\right)_s, \quad (10)$$

where k_g was the thermal conductivity of fission gases including Xe and Kr and P was the percent volume change.

To calculate Equation 10, following equations derived by Kim et al. [4] were used:

$$\left(\frac{\Delta V}{V}\right)_G = \frac{\left(\frac{\Delta V}{V_0}\right)_G}{1+\left(\frac{\Delta V}{V_0}\right)_G}, \quad (11)$$

$$\left(\frac{\Delta V}{V_0}\right)_G = 0.02 + 0.027(f_d - 2) + 0.0058(f_d - 2)^2 - 0.04f_d\ ,$$

$$\text{For } 2\times10^{21}\,\frac{\text{fission}}{cm^3} < f_d \quad (12)$$

$$\left(\frac{\Delta V}{V_0}\right)_s = 0.04f_d,$$

$$\text{For } 2\times10^{21}\,\frac{\text{fission}}{cm^3} < f_d \quad (13)$$

The thermal conductivity of Al-2wt.%Si matrix was measured by Cho et al. [12]. At room temperature, it was 191.4 W/mK and was 197.6 W/mK at 200℃. For the simulation, this modeling used the value of 191.4 W/mK for the matrix thermal conductivity within the range of 50-200℃. And 197.6 W/mK was used for the operating temperature higher than 200℃.

2.2 Model Geometry

Simulated models for TL segment consisted of three phases; fuel particles, Al matrix, and IL. The model for TK segment consisted of just two phases, fuel particles and IL. This modeling assumed that there would be no thermal expansion nor swelling due to irradiation; IL thickness would not grow throughout the operation. The fuel particles had constant and uniform radius of 33.5μm which was the average fuel radius reported by Keiser Jr et al. [11].

This model assumed that U-Mo particles and IL were distributed in FCC array in the matrix. ILs were overlapped together in the model representing TL segment. To determine IL thickness for the model, equations provided in Ref. 4 were used. Table 2 and Figure 3 showed the final dimensions for the geometry of TL and TK model.

Table 2: Dimensions for the models representing TL and TK segments. Following values were in the unit of micron

Model	**Radius of fuel**	**Length of unit cell**
TL	33.5	101.1431
TK	33.5	108.6593

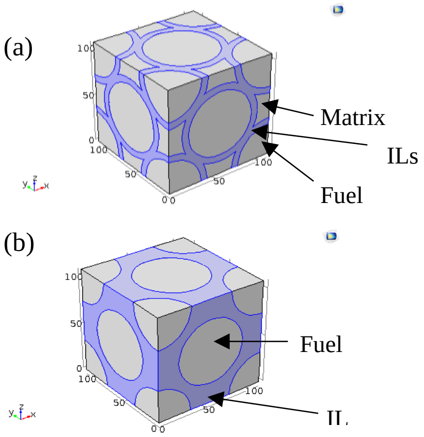

Figure 3. Simulated models generated for (a) TL model with IL thickness of 6.008 micron and (b) TK segment in COMSOL

2.3 Boundary conditions

For this modeling, there was no heat generation from the fuel particles. A constant heat load was applied to evaluate the effective thermal conductivity of the models. Both TL and TK models were under the same boundary conditions. For each model, all side surfaces of the unit cell were insulated; heat could not escape from these sides. The top surface held at a constant temperature, equal to the initial operating temperature of the unit cell, while a constant heat flux of 250 W/cm^2 was applied to the opposite surface.

3. Experiment

To evaluate the effective thermal conductivity of the composites, the following equation was used:

$$k_{eff} = \frac{q" \times L}{dT} \quad (14)$$

where q" was the applied heat flux (=250 W/cm^2), L was the height of the model geometry, and dT was the temperature distribution of the top and bottom surfaces. The average surface temperatures were calculated by COMSOL Multiphysics program.

To evaluate thermal conductivity of IL, the data-fitting method was applied by using a random numerical value to the IL thermal conductivity for making the effective thermal conductivity of the simulated model to match with the experimental data reproduced from Ref. 8.

4. Result and Discussion

4.1 TL model

By applying data-fitting method on the IL thermal conductivity, the effective thermal conductivity from the modeling was matched with the experimental data. Figure 4 showed the comparison between the results from the simulation and the experimentally measured data [8]. According to the experimental data, the fuel meat thermal conductivity for TL segment was 14.6-25 W/mK within the range of the operating temperature.

In Figure 5, "COMSOL with ImageJ" showed obtained thermal conductivity of IL using volume fractions provided in Table 1. The figure also provided the values assumed by Burkes et al. [8] and the values generated by analytical modellings [13].

Burkes et al assumed that the thermal conductivity of IL for TL segment would be 1.13 times to un-irradiated thermal conductivity of fuel particles [8]. For the thermal conductivity of un-irradiated fuel particle, the following equation derived from the correlation of Burkes et al. [8] was used:

$$k_{fuel} = -(1.294 \times 10^{-5}){T_m}^2 + (4.11 \times 10^{-2})T_m + 10.8, \quad (15)$$

where T_m was the operating temperature in ℃.

As could be seen from Figure 5, results from the simulation and the prediction by the analytical models had different characteristics compared with the Burkes' assumption. For TL segment, the value for IL thermal conductivity was strongly related to the remaining matrix since thermal conductivity of the matrix was much greater than that of the fuel particles. Burkes used the sample with 13.2 percent of the matrix [8]. The values estimated by the analytical models [13] using the same matrix volume fraction with Burkes were matched mostly with the reported experimental data except the range of 75-150℃. At this range, the value was greater than expected.

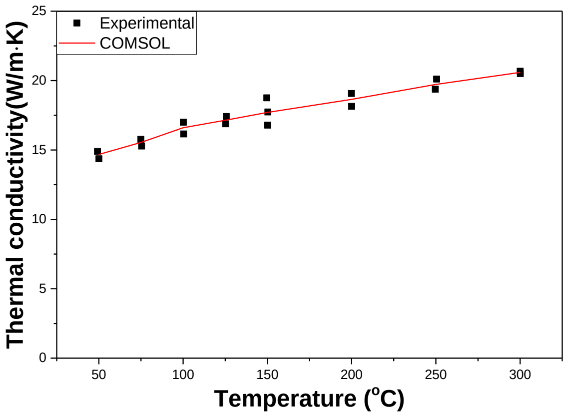

Figure 4. Comparison between the experimentally measured data [8] and the COMSOL predicted thermal conductivity for TL model

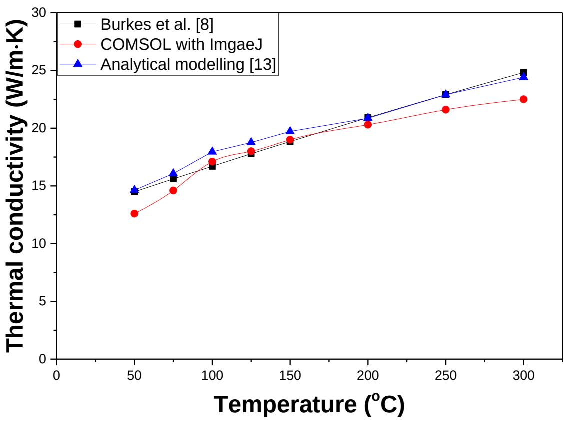

Figure 5. Data-fitted IL thermal conductivities for TL model simulated in COMSOL, values assumed by Burkes et al. [8], and results predicted by analytical models [13]

The simulation using the matrix volume fraction measured by ImageJ [6] tended to have IL thermal conductivity lower than the Burkes' assumption. However, the value was matched with the assumption where the result from the analytical models did not matched.

4.2 TK model

Figure 6 showed the measured data collected from Burkes et al. [8] and the results from the simulation applied data-fitting method on the IL thermal conductivity for TK segment. Those values applied to IL were provided in Figure 7. In the same figure, the data assumed by Burkes et al. [8] and the values generated by analytical models [13] were also included. Since Burkes et al assumed that the thermal conductivity of IL should be equal to that of un-irradiated fuel particles, the result was derived from Equation 15.

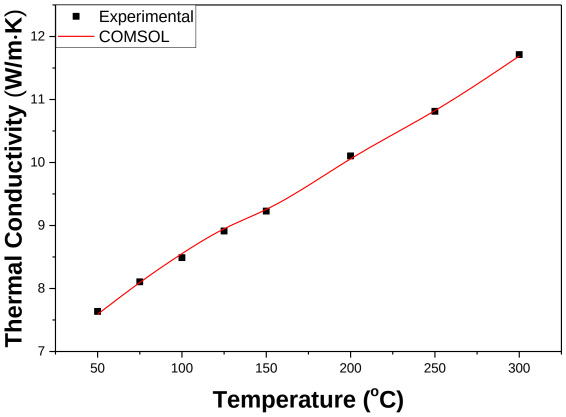

Figure 6. Comparison between experimental data and the predicted effective thermal conductivity for TK model by COMSOL.

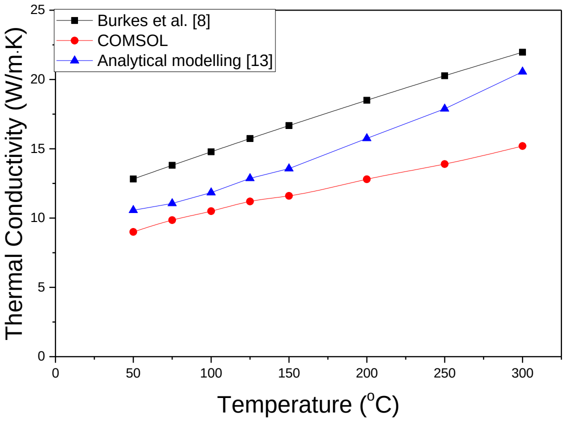

Figure 7. Data-fitted thermal conductivity of IL for TK segment, values assumed by Burkes et al. [8], and results derived by analytical models [13]

Figure 7 showed that the overall IL value from the simulation had lower than that from both the analytical models and the Burkes' assumption. Since TK segment did not contain the matrix in its geometry, thermal conductivity of fuel particles was critical on IL thermal conductivity. The value calculated from Equation 3-13 was higher than thermal conductivity of U-Mo particles estimated by Burkes et al. [8]. Therefore, the result for IL from COMSOL had the thermal conductivity lower than the assumption.

5. Conclusion

The modeling from COMSOL Multiphysics estimated the thermal conductivity of IL for both TL and TK segments. The overall results of the simulation for the models had IL thermal conductivity lower than the assumption from Burkes et al. [8]. According to the simulation for TL model, the thermal conductivity of IL was predicted to be 12.56-21 W/mK. The value for simulated TK model, however, was estimated to be 9-15 W/mK.

Acknowledgments

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