INTRODUCTION

\( \text{U}_3\text{Si}_2 \) has attracted much attention as a promising candidate for accident tolerant fuel (ATF) in light water reactors (LWRs) due to its high uranium density and thermal conductivity compared to traditional oxide fuel [1]. The advantages of thermophysical properties such as high thermal conductivity at room temperature support its use even as an accident tolerant fuel. As one of the most important thermophysical properties, thermal conductivity is crucial for accurate prediction of fuel performance under steady and transient states [2, 3]. The thermal conductivity of a material is a microstructure dependent property, although in most of the current fuel performance codes the thermal conductivity has no specific dependence on fuel microstructure [4, 5]. The thermal conductivity degradation of a nuclear fuel occurs after its exposure to neutron irradiation. During irradiation, the generation of soluble and insoluble fission products, point-defect clustering, growth of voids and gas bubbles, thermal-driven grain growth, and precipitation can significantly change the fuel microstructure, which can all decrease its thermal conductivity in both time and space. The degradation of thermal conductivity can eventually lead to the overheating of the central part of fuel elements, causing the failure of fuels. Therefore, it is very important to understand and predict the thermal conductivity changes with the microstructure damages during irradiation.

One of the most significant microstructure changes in the fuel is the formation of fission gas bubbles. There are two types of fission gas bubbles in irradiated \( \text{U}_3\text{Si}_2 \) fuels: intragranular bubbles – those inside fuel grains, usually of nanometer scale and evenly distributed; and intergranular bubbles – those on grain boundaries, usually have larger sizes than intragranular bubbles and unevenly distributed [6, 7]. Predicting the thermal conductivity of \( \text{U}_3\text{Si}_2 \) with respect to the volume fraction of gas bubbles is important to understanding the degradation of thermal conductivity. Plenty of simulation [8-11] and experimental [12-14] efforts on the thermal conductivity of \( \text{UO}_2 \) have been made. However, there are limited data on the thermal conductivity degradation due to the microstructure damage regarding to the \( \text{U}_3\text{Si}_2 \) fuel [15]. It is believed that the electronic mechanism is the main contribution to the thermal conductivity in \( \text{U}_3\text{Si}_2 \), which is very different from \( \text{UO}_2 \) where the phonon mechanism is responsible for thermal transport [16]. Data has been previously reported on the thermal conductivity of \( \text{U}_3\text{Si}_2 \), and, in a matrix with Al, the positive coefficient with temperature [15]. Recently, White et al. [3] reported the thermophysical properties of \( \text{U}_3\text{Si}_2 \) up to 1773K. The thermal conductivity was observed which increases linearly with temperature. This further confirms the difference from \( \text{UO}_2 \), where the thermal conductivity decreases with increasing temperature.

In this work, we study the effect of intragranular and intergranular porosity on the effective thermal conductivity of \( \text{U}_3\text{Si}_2 \). The simulations are performed using MOOSE software developed by Idaho National Laboratory [17].

METHODOLOGY

We study the effective thermal conductivity in the single crystal \( \text{U}_3\text{Si}_2 \) with intragranular gas bubbles, and bicrystal and polycrystalline \( \text{U}_3\text{Si}_2 \) with intergranular gas bubbles separately due to the large size difference between the bubbles. We use the conventional phase-field method to construct the gas bubbles and grain structures. In the phase-field model, the grain parameter \( \eta_i(r, t) \) (\( i = 1, 2, \ldots, p \)) is chosen to describe the polycrystalline structure, which is 1.0 inside \( i \)-th grain and zero outside \( i \)-th grain, and varies smoothly from 1.0 to zero across the grain boundaries, and \( p \) is the total number of grains. \( c_g \) is chosen to describe the fission gas Xe composition, which is 1.0 inside the bubble and zero in the matrix, and continuously changes across the bubble surface. The free energy of the system is given in terms of the phase parameters as

\[
F = \int \left( \mu f(c, \eta_i) + \frac{\kappa_c}{2} |\nabla c|^2 + \sum_{i=1}^{n} \frac{\kappa_i}{2} |\nabla \eta_i|^2 \right) \, dV
\]

where \( \kappa_c \) and \( \kappa_i \) are the gradient coefficients for concentration and grain parameters, respectively, \( \mu \) is the bulk energy parameter. The bulk energy term is defined as

\[
f = \sum_{i} \left( \frac{\eta_i^4}{4} - \frac{\eta_i^2}{2} \right) + \left( \frac{c^4}{4} - \frac{c^2}{2} \right) + \gamma_{gb} \sum_{i=1}^{n} \sum_{j>i}^{n} \eta_i^2 \eta_j^2 + \gamma_{gb} \sum_{i=1}^{n} \eta_i^2 \]

where \( \gamma_{gb} \) is set to 1.5 to ensure a symmetric diffuse interface, and \( \sigma_{sur} \) and \( \sigma_{gb} \) are the surface energy and grain boundary energy, respectively.

The evolution of grain parameters and Xe composition are controlled by the Allen-Cahn and Cahn-Hilliard equations as [8, 9, 11]

\[
\frac{\partial \sigma}{\partial t} = \nabla \cdot \nabla \sigma \frac{\delta F}{\delta \sigma}
\]

\[
\frac{\partial \eta_i}{\partial t} = -L \frac{\delta F}{\delta \eta_i}
\]

where \( L \) is the kinetic coefficient, \( M \) is the concentration mobility, and \( t \) is the time.
Based on the above phase-field model, the grain structure with gas bubbles of the U$_3$Si$_2$ fuel can be constructed. To estimate the effective thermal conductivity of such a microstructure, the distribution of temperature in the phase-field constructed microstructures can be obtained by solving the steady-state heat conduction equation given by [10]

$$\nabla \cdot (\kappa(\eta)\nabla T) = 0,$$

where $\kappa$ is the local thermal conductivity depending on the microstructural information such as the Xe composition and grain parameters.

For each microstructure we studied, Eq. (4) is solved with a constant temperature $T$ of 800 K on the left side and a heat flux $j_0$ of 100 MW/m$^2$ for single crystals and a heat flux of 29.4 MW/m$^2$ for bicrystals and polycrystals on the right side. The rest of the boundaries are periodic. After Eq. (4) is solved, the effective thermal conductivity $\kappa_{\text{eff}}$ can be estimated by [10]

$$\kappa_{\text{eff}} = -\frac{j_0 L_i}{T_L - T_R},$$

where $T_L$ and $T_R$ are the temperatures at the left and right sides, respectively, $L_i$ is the length of the domain along the temperature gradient direction.

The effective thermal conductivity of the entire system was calculated using the asymptotic expansion homogenization method implemented in MARMOT by Hales at al. [11, 18].

RESULTS

Determination of the bulk thermal conductivity

The thermal conductivity of the polycrystalline U$_3$Si$_2$ with an average grain size of 35.0µm at different temperatures was reported in recent experiments [2, 3]. However, the bulk thermal conductivity of U$_3$Si$_2$ has not been reported, which is required in order to calculate the effective thermal conductivity of U$_3$Si$_2$ with both intragranular and intergranular gas bubbles. As reported by Millett et. al. [9] the effective thermal conductivity of a dense polycrystalline with an average grain size $d$ is related to the bulk thermal conductivity and Kapitza conductance as [9, 19]

$$\kappa_{\text{eff}} = \frac{\kappa_0}{1 + \frac{G_i}{G_k} d},$$

where $\kappa_0$ is the bulk thermal conductivity, $G_i$ is the Kapitza conductance, and $d$ is the average grain diameter. We can estimate the bulk thermal conductivity and Kapitza conductance by fitting the experimental thermal conductivity.

In the simulation, a simple bicrystal system was first created using the phase-field method. Grain 1 is placed at the center of the simulation cell while grain 2 is placed at both ends of the simulation cell to satisfy the periodic boundary conditions. The two grains have the same size. The average grain size varies from 5.0 µm to 35.0 µm. We choose three different bulk thermal conductivities, 8.55W/mK, 8.60W/mK, and 8.65W/mK to get the best fitting of the experimental data at 35.0 µm. Figure 1 shows the grain size dependence of the calculated effective thermal conductivity of a bicrystal with different bulk thermal conductivities. The calculated effective thermal conductivity data is fitted to Eq. (6) to obtain the bulk thermal conductivities. The calculated effective thermal conductivity gives the best fitting results. Thus, we use this value as the bulk thermal conductivity of U$_3$Si$_2$ in the following simulations.

![Fig. 1. Effective thermal conductivities as a function of grain size for different bulk thermal conductivities.](image)

Effective thermal conductivity of U$_3$Si$_2$ with intragranular gas bubbles

The size of the intragranular gas bubbles are generally in several nanometers. Single crystals with the size of 1.0µm×1.0µm in 2D and of 0.5µm×0.5µm×0.5µm in 3D are used to study the effective thermal conductivity of U$_3$Si$_2$. Identical gas bubbles are randomly seeded in the grain interior of U$_3$Si$_2$. The volume fraction of gas bubbles can be

![Fig. 2. Effective thermal conductivity of U$_3$Si$_2$ as a function of porosity for different sizes of intragranular gas bubbles in 2D (a) and 3D (b).](image)
controlled by adjusting the number of bubbles. A minimum bubble distance of $0.2 \text{nm}$ is set to ensure there is no overlap between bubbles. Using the fuel microstructures generated by phase field simulations, we can simulate the effective thermal conductivity of $\text{U}_3\text{Si}_2$ by solving Eqs. (3) and (4). We investigated the effects of gas bubble size, size distribution, and the bubble volume fraction on the effective thermal conductivity of $\text{U}_3\text{Si}_2$ [3]. Figure 2 shows the predicted porosity dependence of the thermal conductivity of $\text{U}_3\text{Si}_2$ at 300 K both in 2D and 3D. Simulations show that the effective thermal conductivity of $\text{U}_3\text{Si}_2$ decreases as porosity increases. The simulated thermal conductivities show a notable difference between the 2D and 3D simulations. The 2D simulation data are clearly lower than those of the 3D data, which suggests that the 2D simulations have a more rapid decrease in conductivity than the 3D simulations as porosity increases, which agrees with previous studies [13, 20].

According to Hastings et al. [21], the porosity dependence of the effective thermal conductivity can be described by the following expression,

$$\kappa^\text{eff} = \kappa_0 \left(1 - P \beta \right)^{-1},$$  \hspace{1cm} (7)

where $P$ is the porosity and $\beta$ is a factor depending on the pore conductivity, shape, and distribution. To determine the parameter $\beta$, the simulated thermal conductivities in 2D and 3D are used to fit the model separately. The thermal conductivities of $\text{U}_3\text{Si}_2$ with different bubble sizes are used to fit the model due to the minor effect of bubble size. The fitting results are shown in Fig. 2(a) and the determined parameters $\beta = 2.04, 2.15, 2.14$, and 2.06 for bubble radius $r = 2$, 5, 8, and 10 nm in 2D simulations, respectively. The fitting results are shown in Fig. 2(b) and the determined parameters $\beta = 2.04, 2.15, 2.14$, and 2.06 for bubble radius $r = 2$, 5, 8, 10, and 15 nm in 3D simulations, respectively. For the selected gas bubble sizes the gas bubble size and size distribution has a minor impact on the thermal conductivity for three chosen classical intergranular gas bubble sizes. The differences increase with the increase of porosity, which may come from the diffuse interface effect of the gas bubble area/volume calculations.

Next, we will derive a correlation to convert the 2D thermal conductivities to 3D based on Bakker’s derivations [22], in which the derived relationship between 2D and 3D thermal conductivities with gas bubble phase having a circular/spherical shape is related to the conductivities of gas bubble and matrix phases, and the composition of gas bubble phase. However, Bakker’s formula was applied to the polycrystalline materials where the grain boundary conductivity plays an important role in determining the effective thermal conductivity, giving different conductivities when the composition of gas bubble phase is zero for 2D and 3D simulations. This is different from the thermal conductivities of single crystals in this study. Thus, we modified the Bakker’s derivation (Eq. (1) in Ref. [22]) to ensure that the thermal conductivity has the same value for 2D and 3D when the composition of gas bubble phase is zero as

$$\frac{1 - (1 - \frac{\kappa_0}{\kappa_g})p - \kappa^\text{eff}}{1 - (1 - \frac{\kappa_0}{\kappa_g})p - \kappa^\text{eff}_{2D}} = 1 + p \left(C_1 + \frac{1}{\kappa_0 + C_2}\right), \hspace{1cm} \text{(7)}$$

where $\kappa^\text{eff}_{2D}$ and $\kappa^\text{eff}_{3D}$ are the effective thermal conductivities of $\text{U}_3\text{Si}_2$ in 2D and 3D, respectively, and $C_1$ and $C_2$ are the fitting constants. Using the predicted effective thermal conductivities of single crystal $\text{U}_3\text{Si}_2$ with intragranular gas bubbles from both 2D and 3D simulations, the fitted constants are determined as $C_1 = -1.213$ and $C_2 = 1.674$.

Note that the value of $\kappa^\text{eff}_{2D}$ is obtained by fitting the overall 2D data, which gives the fitting parameter $\beta$ of 2.10. The simulated and fitted thermal conductivities of $\text{U}_3\text{Si}_2$ in 3D using Eq. (7) are shown in Fig. 3. This correlation can be used to estimate the 3D effective thermal conductivity containing the intragranular gas bubbles based on its 2D data.

![Diagram showing comparison between simulated and fitted thermal conductivities of $\text{U}_3\text{Si}_2$ in 3D.](image)

**Effective thermal conductivity of $\text{U}_3\text{Si}_2$ with intergranular gas bubbles**

Since the fabricated $\text{U}_3\text{Si}_2$ fuels for LWRs are polycrystalline, it is important to study the effect of intergranular gas bubbles on the thermal conductivity of $\text{U}_3\text{Si}_2$. All simulations were performed in a 2D square domain with a size of $17 \mu \text{m} \times 17 \mu \text{m}$, with a total number of grains of 10. The average grain size is $5.37 \mu \text{m}$ if we assume the grain shape is square. The thickness of grain boundary is fixed at $0.17 \mu \text{m}$. The grain boundary conductivity is assumed to be the same as the bulk due to the metallic behavior of $\text{U}_3\text{Si}_2$ fuel. The phase field generated fuel microstructures with intergranular gas bubbles are shown in Fig. 4. The grains were initialized using a Voronoi tessellation [23]. The phase-field simulations were conducted to relax the microstructures to the equilibrium state for each thermal conductivity calculation. Three initial different circular intergranular gas bubble sizes were used in the microstructures.
The thermal conductivity was calculated as a function of grain boundary coverage for three different initial intergranular gas bubble radius at temperature of 300 K. The calculated thermal conductivity of U$_3$Si$_2$ with intergranular bubbles are shown in Fig. 5. The effective thermal conductivities of U$_3$Si$_2$ decreases as the grain boundary coverage increases. The grain boundary coverage is defined in two-dimensional domain as $X_G^e = l_b N_b / l_{GB}$, where $l_b$ is the length of an individual intergranular bubble, $N_b$ is the total number of intergranular bubbles, $l_{GB}$ is the total length of the GB network. For polycrystalline, the effective conductivity is described by Eq. (6).

When a grain boundary is laden with fission gas bubbles, the Kapitza conductance of the grain boundary decreases. A correlation of transferring 2D to 3D can be obtained by using Bakker’s derivations [13] with $k_{eff}^{2D}$ for intergranular bubbles.

In conclusion, mesoscale heat conduction simulations were performed to investigate the effect of fission gas bubbles, including intragranular and intergranular gas bubbles, on the effective thermal conductivities for both single crystal and polycrystalline U$_3$Si$_2$ fuels. Based on the thermal conductivities of single crystals of U$_3$Si$_2$ simulated in 2D and 3D, a correlation of transferring 2D to 3D conductivities is derived for the U$_3$Si$_2$ fuel. The correlation of the thermal conductivities of the polycrystalline U$_3$Si$_2$ fuel as a function of grain boundary coverage in 2D was also derived. We hope the current study can provide a better understanding of the effect of microstructure change on the thermal conductivity of U$_3$Si$_2$.

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References