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Demonstrating the Temperature Gradient Impact on Grain Growth in UO_2 Using the Phase Field Method

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Grain boundaries (GBs) are driven to migrate up a temperature gradient. In this work, we use a phase field model to investigate the impact of temperature gradients on isotropic grain growth. GB motion in 2D UO_2 polycrystals is predicted under increasing temperature gradients. We find that the temperature gradient does not significantly impact the average grain growth behavior because the curvature driving force is dominant. However, it does cause significant local migration of the individual grains. In addition, the temperature dependence of the GB mobility results in larger grains in the hot portion of the polycrystal.

Keywords: Grain Growth, Temperature Gradient, Phase Field Method, UO_2

1. Introduction Grain boundaries (GBs) play a critical role in microstructure evolution. For example, they act as defect sources and sinks,[1] they pin dislocation motion [2] and act as segregation sites for impurities and precipitates.[3,4] Thus, understanding and predicting GB behavior is of great importance.

One critical behavior is the migration of GBs to reduce the Gibbs free energy of the material. As summarized in Gottstein and Shvindlerman,[5] driving forces (DFs) for GB migration result from gradients in various properties of the material, including gradients of temperature, pressure, defect density and elastic energy. A temperature gradient drives a GB to migrate towards higher temperature, resulting in reduction in the energy. Yet, the temperature gradients that occur in most applications are small enough that their effects are typically negligible. However in certain applications, such as nuclear reactor fuel, gradients as large as $0.4 \text{ K}/\mu\text{m}$ can form.[6] Thus, in such applications, the temperature gradient DF could be significant.

The velocity of a migrating GB can be approximated by the simple model [5]

$$v = M_{\text{GB}}P, \quad (1)$$

where M_{GB} is the GB mobility and P the DF. The DF P takes the form of an energy per unit volume. The DF

to reduce the GB energy (typically called the curvature DF) is

$$P = \frac{\sigma_{\text{GB}}}{R}, \quad (2)$$

where σ_{GB} is the GB energy and R the GB curvature. The DF due to a temperature gradient ∇T can be expressed as [5]

$$P = \frac{\Delta S w_{\text{GB}}}{\Omega_a} \nabla T, \quad (3)$$

where ΔS is the entropy difference between the GB and the bulk, w_{GB} the GB width and Ω_a the molar volume of the material. While this model approximates the migration of a single GB, more complex models are necessary to predict the grain growth in a polycrystal. Many methods have been used to model grain growth, including front tracking,[7,8] Monte Carlo Potts models,[9,10] phase field (PF) methods,[11–13] cellular automation [14] and level set methods.[15] While each method was first developed for use with the curvature DF, many have been modified to include additional DFs.[16–20] While the Potts and PF models have been used to investigate the impact of a spatially varying GB mobility due to a temperature gradient,[21,22] none of the approaches have been used to model the temperature gradient DF.

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In this communication, we use the PF method to investigate the impact of the temperature gradient DF on normal grain growth, i.e. grain growth driven by the curvature DF with isotropic GB properties. The PF method is employed due to the relative simplicity of coupling it to the temperature gradient. We first implement a model of the temperature gradient DF using the PF method. We also compare the PF model predictions to an analytical model for a bicrystal. We then use the PF model to investigate the impact of various temperature gradients on grain growth in UO_2 polycrystals.

2. PF Model Development To model grain growth, we employ the quantitative PF model from Moelans et al.,[23] assuming isotropic GB energy and mobility. In this model, each grain is represented with an order parameter ϕ_i , where $\phi_i = 1$ within the grain, $\phi_i = 0$ in adjoining grains and $0 < \phi_i < 1$ within the GB. This grain growth model can be expanded to account for the temperature gradient (∇T) DF by adding a term to the Allen–Cahn evolution equation, resulting in

$$\frac{\partial \phi_i}{\partial t} = -L \left(\frac{\partial F}{\partial \phi_i} + A \nabla T \cdot \nabla \phi_i \right), \quad (4)$$

where L is the order parameter mobility, F the free energy functional and A a constant that defines the magnitude of the DF due to a temperature gradient. To ensure that the magnitude is consistent with the analytical model, we derive an expression for the value of A using a straight boundary that is perpendicular to the x -axis under a constant temperature gradient in the x -direction. The equation for the velocity of a GB in the PF model can be stated as [23]

$$v = \frac{\partial \phi_i / \partial t}{\partial \phi_i / \partial x}. \quad (5)$$

With a straight boundary, the equation defining the order parameter evolution becomes

$$\frac{\partial \phi_i}{\partial t} = -LA \frac{\partial T}{\partial x} \frac{\partial \phi_i}{\partial x}. \quad (6)$$

Combining the two expressions and expressing the PF mobility L in terms of the GB mobility M_{GB} and PF model interfacial width w_{int} (using the expression from Moelans et al. [23]) gives

$$v = -\frac{4}{3} \frac{M_{\text{GB}}}{w_{\text{int}}} A \frac{\partial T}{\partial x}, \quad (7)$$

where

$$M_{\text{GB}} = M_{\text{GB}}^0 e^{-Q/k_b T}, \quad (8)$$

with M_{GB}^0 the mobility pre factor, Q the GB activation energy and k_b the Boltzmann constant. By equating

Equations (7) and (1) using Equation (3) allows us to solve A to obtain

$$A = -\frac{3}{4} \frac{\Delta S w_{\text{int}} w_{\text{GB}}}{\Omega_a}. \quad (9)$$

Note that w_{int} is a modeling parameter that defines the width of the interface in the PF model, while w_{GB} is the actual physical width of the GB. Also note that while this expression was derived using the special case of a flat boundary, it is still accurate for curved boundaries.

The grain growth model including the temperature gradient DF has been implemented in the mesoscale simulation code MARMOT [24] under development at Idaho National Laboratory. We verify this model by comparing the GB velocity in a bicrystal under a constant temperature gradient predicted by the PF model to that predicted by the analytical model from Equations (1) and (3). We modeled a bicrystal of UO_2 , where the GB energy $\sigma = 1.58 \text{ J/m}^2$ [25] (for a $\Sigma 5$ tilt boundary, though our model assumes isotropic GB properties), the GB mobility $M_{\text{GB}} = 9.21 \times 10^{-9} e^{-2.77 \text{ eV}/k_b T} \text{ m}^4/\text{Js}$, [26] $\Delta S = 8.0 \text{ kJ}/(\text{K mol})$, [5] $\Omega_a = 24.62 \times 10^{-6} \text{ m}^3/\text{mol}$ (calculated from the density and the molar mass) and $w_{\text{GB}} = 0.5 \text{ nm}$. [5] We modeled a 2D bicrystal that is $20.0 \mu\text{m} \times 10 \mu\text{m}$, with an initial grain size of $10 \mu\text{m}$. The temperature gradient was $0.8 \text{ K}/\mu\text{m}$, increasing linearly to the right. In order to reduce the computational expense of the simulations in this work, we employ an interface width in the PF model of $w_{\text{int}} = 600 \text{ nm}$, which is three orders of magnitude larger than actual GB widths. However, as long as the GB width is small compared with the average grain size, our simulation predictions are not significantly impacted. To verify our model at various temperatures, three simulations were employed with left-hand temperatures of 1900, 2000 and 2100 K, respectively. An example of the bicrystal domain is shown in Figure 1(a).

The results from all three left-hand temperatures predicted a GB velocity that is somewhat slower than that from the analytical model, as shown in Figure 1(b). When directly comparing the average velocities from the analytical model and the PF simulations, the PF model velocity was slower by $9.5 \pm 0.5\%$ for all three temperatures. We repeated the simulations using an interfacial width $w_{\text{int}} = 300 \text{ nm}$, and obtained the same result. The method we used to introduce the temperature gradient DF (Equation (9)) assumed that there are no temperature gradient effects in the original GB migration model. [23] However, this reduction in velocity implies that the original model may predict an artificial GB migration down a temperature gradient. While we could modify Equation (9) with some fit factor, we choose to leave the equations as derived, and simply consider the discrepancy in all analysis in this work. We will investigate the discrepancy in more detail in future work.

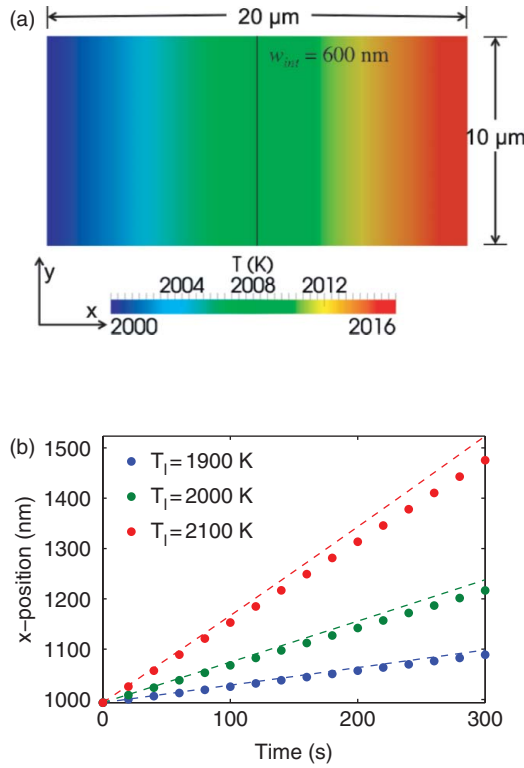


Figure 1. Results from the bicrystal simulations using the temperature gradient DF with a linear gradient of $0.8 \text{ K}/\mu\text{m}$, where (a) shows the bicrystal domain with a left-hand temperature $T_l = 2000 \text{ K}$ shaded by temperature, with the black line the GB, and (b) shows the GB position vs. time at the three left-hand temperatures, with the dashed line being the analytical model. Note that GB velocity predicted by the PF model is about $9.5 \pm 0.5\%$ slower than that from the analytical model.

3. Polycrystal Results Now, we investigate the importance of the temperature gradient DF under conditions similar to those found in a fast reactor with UO_2 fuel. We model a UO_2 rectangular 2D domain that is $250 \mu\text{m} \times 80 \mu\text{m}$ with 300 initial grains. The grains were initialized using a Voronoi tessellation and have an initial average grain size of $8.2 \mu\text{m}$, with symmetric boundary conditions (BCs) for the PF model on the left and right and periodic BCs on the top and bottom. The grain growth is modeled for 2000 min. We apply five linear temperature gradients, all with an average temperature of 2050 K . The area of each grain is recorded with time, such that we can calculate the distribution and mean of the grain size. The simulations employed the same parameter values as the bicrystal simulations from the previous section. Mesh adaptivity was employed to decrease the computational expense. See Figure 2(a) for the initial polycrystal domain with the adapted mesh.

All the simulations, irrespective of the temperature gradient, reduced from the initial 300 grains to 67 ± 2 grains after 2000 min. The final microstructures with no temperature gradient and with $\nabla T = 0.8 \text{ K}/\mu\text{m}$ are

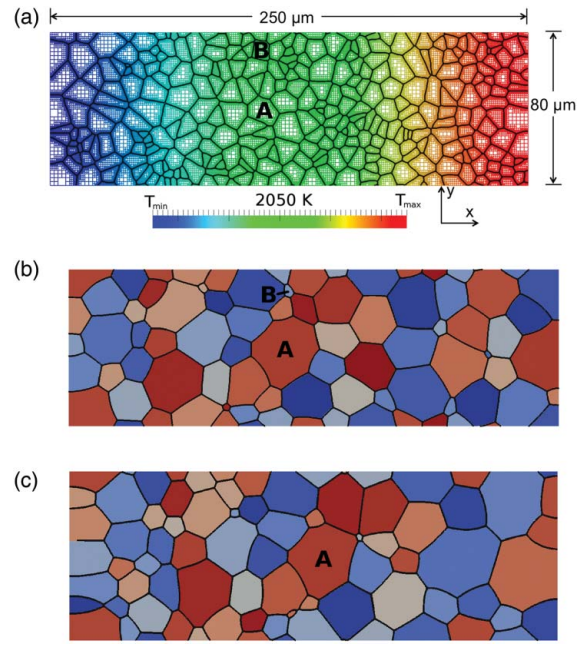


Figure 2. Polycrystal simulation domain, where the initial grain configuration showing the adapted mesh is shown in (a). The constant temperature gradient is also shown. Note that the values for $T_{\min}$ and $T_{\max}$ vary for the different gradients, ranging from $T_{\min} = T_{\max} = 2050 \text{ K}$ for no gradient to $T_{\min} = 1950 \text{ K}$ and $T_{\max} = 2150 \text{ K}$ for $\nabla T = 0.8 \text{ K}/\mu\text{m}$. The centroid position with time is recorded for the grains labeled A and B. The final grain configuration after 2000 min is shown in (b) with no temperature gradient and in (c) with a gradient of $0.8 \text{ K}/\mu\text{m}$, where grain B has disappeared. Note that with the temperature gradient, the grains are smaller on the cold side and larger on the hot side.

shown in Figure 2(b) and 2(c), respectively. By comparing the position of grain A (shown in Figure 2(a) and 2(b)), it is clear that there is significant grain migration towards the hot end of the polycrystal in the presence of a temperature gradient. To obtain a quantitative measure of the grain migration velocity, we fit a line to the x -position of the centroid of grains A and B (Figure 2(b)) at four different times for the five different temperature gradients, showing a linear migration towards the hot side of the fuel, demonstrated for grain A in Figure 3(a).

To investigate these grain velocities, we compare them to the GB velocity calculated from Equations (1) and (3) at $T = 2050 \text{ K}$, which represents the maximum possible grain velocity at that temperature. The actual grain velocity at the same average temperature will always be less than this ideal GB velocity, as it depends on the orientation of its GBs with respect to the temperature gradient. As shown in Figure 3(b), the grain velocity increases with increasing temperature gradient, but not linearly as with the ideal velocity. In addition, the grain velocity appears to be independent of grain size, as grains A and B show similar behavior even though grain A is significantly larger than grain B.

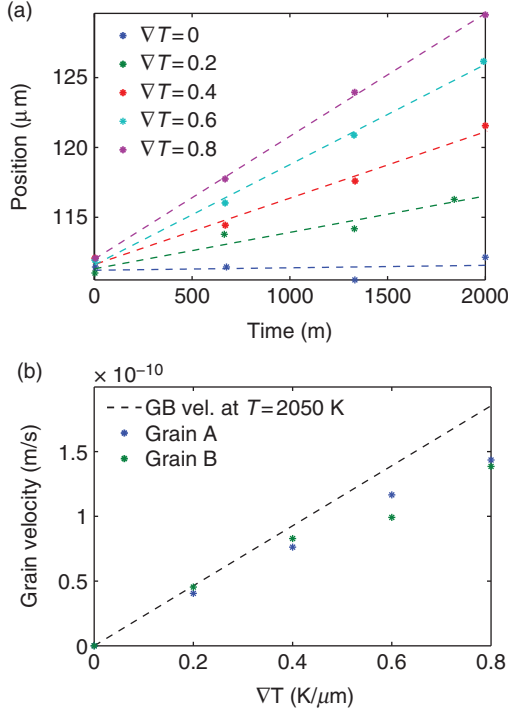


Figure 3. Velocity of grains A and B, where (a) shows how the grain velocity is calculated (shown with values from grain A) from the slope of linear fits to the x-position of the grain centroid for each temperature gradient, where the fits are shown as dashed lines. (b) Shows the grain velocity of A and B as a function of the temperature gradient. The GB velocity predicted by Equations (1) and (3) at $T = 2050$ K, which represents the maximum possible GB velocity at that temperature, is shown as a black dashed line.

For grain growth due to the curvature DF, the average grain size changes linearly with time, where the slope is a function of the DF, Equation (2). However, this behavior is only obtained once the grain size distribution has reached a steady-state form. In our simulation with a zero temperature gradient, the grain area reaches this equilibrium behavior after 1000 min, and the slope $\dot{A} = \sigma_{GB}M$, as shown in Figure 4(a). The behavior is consistent with the curvature DF. In addition, all the simulations, irrespective of the temperature gradient, exhibit a similar linear increase with the same slope.

We also plotted the size of the largest grain with time and found some impact of the magnitude of the temperature gradient on its behavior. The change in the size of the largest grain is accelerated after 1000 h for the 0.6 and 0.8 K/μm gradients (see Figure 4(b)), with the acceleration increasing with temperature gradient. The final grain size distribution (Figure 4(c)) shows that this acceleration in the change of the largest grain size with time is due to the formation of a slightly bimodal grain size distribution, with a slight peak at the largest grain sizes.

To elucidate the cause of this change in the grain size distribution, we compare approximate magnitudes of

the curvature and temperature gradient DFs. We approximate the curvature DF using Equation (2), with the radius of curvature calculated from the average grain area A_{av} according to $R = \sqrt{A_{av}}/2$. The temperature gradient DF is approximated using Equation (3). However, note that this approximation of the temperature gradient DF represents a maximum value (only experienced by GBs perpendicular to the temperature gradient), while our approximation of the curvature DF represents an average value. As shown in Figure 4(d), the average curvature DF decreases with time, due to the increase in average grain area, while the temperature gradient DF is constant. For all grain sizes in our simulations, the average curvature DF is greater than the maximum temperature gradient DF, even if the temperature gradient DF was increased by 9.5% to account for the error observed in our bicrystal simulations, indicating why the temperature gradient did not significantly impact the change in the average grain area (Figure 4(a)). However, independent of the DF, the GB mobility will always be higher in hotter regions of a polycrystal (Equation (8)). Thus, grains on the hot end grow larger than those on the cold end, causing the observed change in the grain size distribution. This gradient in grain size is clearly observable in the final microstructure from the 0.8 K/μm gradient shown in Figure 2(b). Note that if we normalized each grain size by the average grain size at the corresponding temperature, the grain size distribution would be unchanged by the temperature gradient, as shown in [21].

4. Discussion The average temperature gradient experienced by the fuel in light water reactors (LWRs) is about 0.2 K/μm, while it is about 0.4 K/μm in oxide fuel fast reactors. Thus, from our simulations it appears that the temperature gradient DF will have no impact on the average grain size within the fuel. In fact, the maximum temperature gradient DF will be smaller than the average curvature DF for all fuels with an average grain size less than 97.0 μm for LWR conditions and less than 48.5 μm for fast reactor conditions (with the DFs calculated as before). However, the temperature dependence of the GB mobility will have an effect, resulting in faster moving GBs and thus larger grains in the hotter portion of the fuel.

While the temperature gradient has little impact on the average GB behavior, it will have an impact on the local behavior. Our simulations showed a distinct motion of grain A for all temperature gradients greater than zero (Figure 3(b)). This grain motion within the fuel caused by the temperature gradient could result in additional fission gas pick-up, increased interaction between fission gas bubbles and GBs, and local modifications of the stress fields due to grain anisotropy.

The temperature gradient DF could also play an important role in non-nuclear applications. Local grain

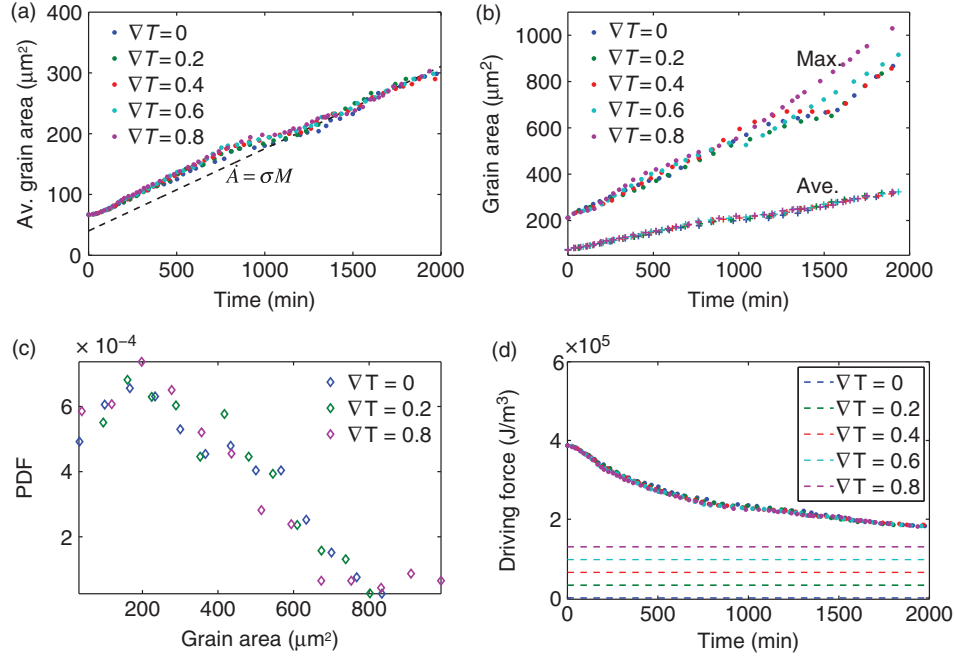


Figure 4. Quantified measures of the polycrystal grain growth, where all temperature gradient units are $\text{K}/\mu\text{m}$. In (a), the average grain area is plotted with time, with a linear slope $\dot{A} = \sigma M$ after enough time for the grain structure to adjust from the initial Voronoi tessellation. Note that the average grain area does not change with temperature gradient. However, the behavior of the maximum grain size with time does change for the larger two gradients, as shown in (b), where the dots indicate the maximum grain area and the plus signs the average. A numerical approximation of the probability density function of the final grain size distribution for three of the temperature gradients is shown in (c), where it is evident that for the largest gradient, a slight bimodal distribution has formed, with an increase in grains of large size. In (d), the curvature DF (dots) and the temperature gradient DF (dashed lines) are compared, clearly showing that the temperature gradient DF is smaller than the curvature DF for all gradients used in our simulations.

migration will take place in any application with a significant temperature gradient and could impact the local microstructure behavior. In applications with a significant temperature gradient and a large grain size, the curvature DF will be reduced and the temperature gradient DF may play a more dominant role than observed in our simulations. By equating the DFs from Equations (2) and (3), we can solve for the critical grain size

$$d_{\text{crit}} = \frac{\Delta S_{\text{WGB}}}{2\sigma_{\text{GB}}\Omega_a} \nabla T, \quad (10)$$

where the average grain size $d = 2R$. Thus, for given polycrystal parameters and temperature gradient, the temperature gradient DF may play a dominant role if the average grain size is greater than d_{crit} . Again note that this approximation of the temperature gradient DF represents a maximum value while the approximation of the curvature DF represents an average value.

5. Conclusions GBs are driven to migrate up temperature gradients, adding an additional GB migration DF. Here, we have implemented the temperature gradient DF into a PF model of grain growth. We verified the model by comparing with the analytical equation (3) and found that the PF model predicts GB velocities that are

$9.5 \pm 0.5\%$ slower than those calculated by the analytical model. The PF model was then used to predict the behavior in a UO_2 polycrystal with a range of temperature gradients. We found that while the temperature gradient does cause local GB motion up the gradient, the evolution of the average grain area with time is not impacted. This is because the temperature gradient DF is smaller than the curvature DF for all of the gradients that we investigated. The behavior of the largest grain size with time does change with the gradient, due to changes in the grain size distribution. However, these changes are not caused by the temperature gradient DF, but rather by the dependence of the GB mobility on temperature. Therefore, the temperature gradient DF may not need to be considered in macroscale models of the average grain size in LWR and fast reactor fuel, though the temperature gradient will modify the local grain behavior.

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