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http://www.inl.gov

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Development of a Model for Irradiation-Induced Grain Growth in UO_2 thin films

Md Ali Muntaha,* Michael R. Tonks,*, Larry Aagesen,†

*Department of Materials Science and Engineering, University of Florida, P.O. Box 116400, Gainesville, FL, md.muntaha@ufl.edu, michael.tonks@ufl.edu
†Idaho National Laboratory, Idaho Falls, larry.aagesen@inl.gov

INTRODUCTION

Grain boundary migration occurs in polycrystalline materials to reduce the total grain boundary energy of the material, resulting in the growth of larger grains and the disappearance of smaller grains. Various driving forces are possible for grain growth, including reduction in grain boundary energy, stored elastic energy, and stored defect energy. The kinetics of grain growth are typically defined by the grain boundary mobility, which is an exponential function of the temperature. Thus, grain growth typically only occurs at elevated temperatures, especially in ceramics such as UO_2.

Radiation can induce grain growth at much lower temperatures than for thermally-activated grain growth [1, 2]. For example, Kaoumi et al. [1] showed accelerated grain growth in ion irradiated metallic thin films from Zr, Cu, Pt, and Au, and supersaturated solid solutions of Zr–Fe and Cu–Fe. Later, Yu et al. [3] showed irradiation-accelerated grain growth for in situ ion irradiated UO_2 thin films. Kaoumi et al. [1] developed an analytical model that predicted the evolution of the average grain size in irradiated materials based on the direct impact of thermal spikes on grain boundaries, accelerating curvature-driven growth. Bufford et al. [2] developed a mesoscale model of grain growth in which they introduced randomly distributed regions of locally increased grain boundary mobility and compared the predictions with data from in situ ion irradiations of Au thin films.

In this work, we develop a model of irradiation-induced grain growth in UO_2 using the MARMOT mesoscale nuclear materials simulation tool. We couple the existing thermally activated grain growth model with a heat conduction model that includes random heat sources representing thermal spikes. We compare the results with the irradiation data on UO_2 thin films [3].

SIMULATION APPROACH

The MARMOT mesoscale nuclear materials simulation tool [4], based on the MOOSE framework [5], contains an implementation of a popular phase field grain growth model [6]. The model assumes isotropic grain boundary energy and mobility, and therefore can be used to model grain growth in UO_2 using average values for its grain boundary energy and mobility such as those presented by Tonks et al. [7]. We represent the impact of thermal spikes by having randomly distributed regions of short-lived heat generation that locally raise the temperature by very high amounts (even above the melting temperature) for short amounts of time. We assume that the grain boundary mobility M_{GB} follows the standard Arrhenius expression,

$$M_{GB} = M_{GB}^0 e^{-\frac{Q}{k_b T}}, \tag{1}$$

where M_{GB}^0 is the prefactor, Q is the activation energy, k_b is the Boltzmann constant, and T is the temperature, even when the local temperature raises above the melting temperature.

We modify the model to include the impact of thermal spikes resulting from irradiation by coupling it with a heat conduction model that determines the temperature over time t by solving the partial differential equation

$$\rho c_p \frac{\partial T}{\partial t} = \nabla \cdot k \nabla T + Q, \tag{2}$$

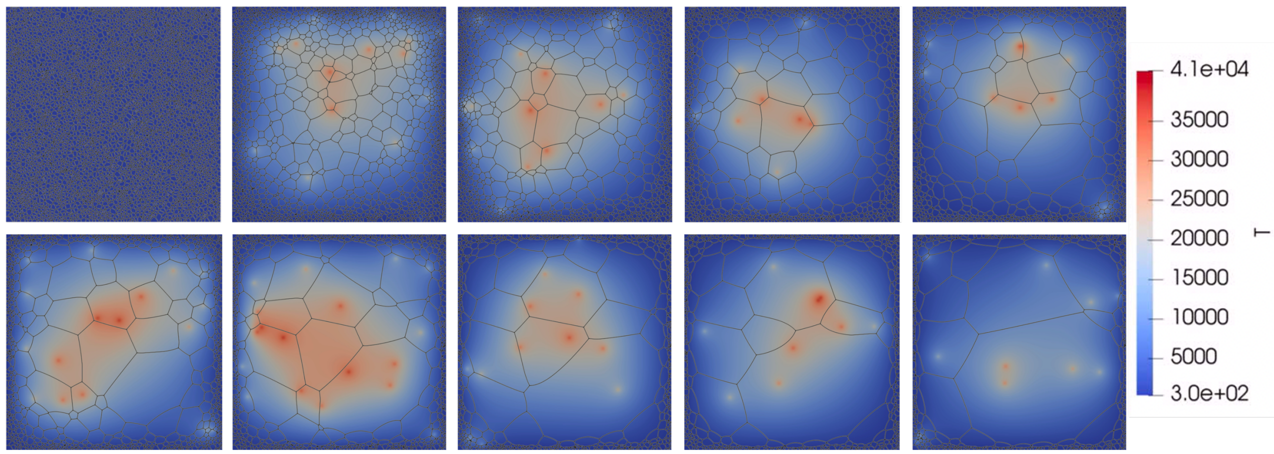
where ρ is the density, c_p is the specific heat, k is thermal conductivity, and Q is the heat generation due to thermal spikes. For the density, specific heat, and thermal conductivity, we use values from the literature for UO_2. The heat generation for the thermal spikes is a stochastic term for which its magnitude is a function of the irradiation flux. The thermal spikes result in short-lived regions of very high temperature that quickly disappear as the heat is conducted away.

The value for Q varies stochastically in space across the domain and in time. A given probability P_c of a cascade occurring is a function of the irradiation flux and has units of cascades per unit volume per unit time. When a cascade event is randomly determined to occur during a certain time step, the center location of the cascade is randomly determined from a uniform distribution. The value of Q is then nonzero in a spherical region of radius r_c around the center point for a specific spike duration t_s. The magnitude of the heat generation rate is

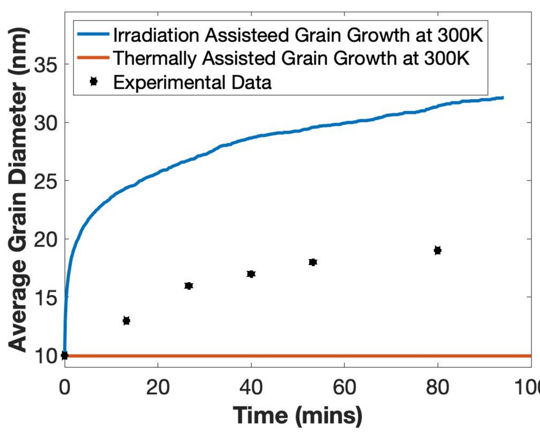
$$Q = \frac{q_s}{V_s t_s}, \tag{3}$$

where q_s is the average thermal spike energy in eV per spike and $V_s = 4/3\,\pi r_c^3$ is the spike volume. We reduce the required simulation time by assuming that no grain growth appears between thermal spikes and that there is no interaction between thermal spikes. Thus, we can increase the probability and scale the time by a similar amount to the point where only one or two thermal spikes occur every time step.

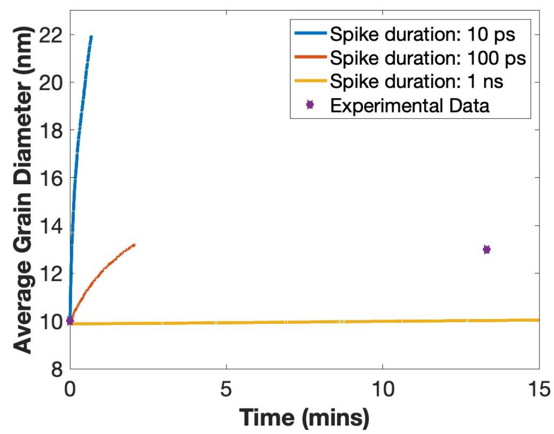
We model the room temperature (T = 300 K) ion irradiations of UO_2 thin films carried out by Yu et al. [3], with an ion flux of 6.25×10^{12} ions/cm^{2}s. We assume no heat transport from the top and bottom of the thin films and that the grains are columnar, such that they can be represented in 2D without significant error. Heat is removed from the system at the sample edges by the holder. Thus, we assume that the temperature at the outer edge of the domain is fixed at 300 K. We also assume zero flux boundary conditions for the phase field variables representing the grains. We assume $r_c = 4.84$



(a)



(b)



(c)

Fig. 1: Results from 2D simulations of grain growth in a 710×710 nm UO_2 thin film with 5000 initial grains undergoing room temperature ion irradiation. (a) Snapshots of the polycrystal domain over time assuming a spike duration of 10 ps, shaded by the temperature in K and the grain boundaries in black. The corresponding times in minutes are: 0, 1, 2.7, 5.9, 8.5 (top row); 10.6, 19.7, 29.8, 40.5, 52.5 (bottom row). (b) Plot of the grain diameter versus time with a spike duration of 10 ps, a simulation with just thermal grain growth, and the experimental data [3]. (c) Plot of the grain diameter versus a much shorter time using various spike durations.

nm and q_c = 25.65 keV. The duration of a thermal spike is unknown but potentially ranges from 1 to 100 ps. We model a 710×710 nm domain with 5000 initial grains. The time step is equal to the thermal spike duration.

SIMULATION RESULTS

The results from the room temperature simulation of grain growth during ion irradiation are shown in Fig. 1a. The grain growth is modeled for 60 minutes, assuming a thermal spike duration of 10 ps. No grain growth would occur at room temperature without ion irradiation. Due to the thermal spikes, a large gradient in temperature occurs. The temperature is very high at the center of a spike (as high as 40,000 K), but drops off quickly away from a spike. These maximum temperatures are much higher than the melting temperature for a few ps but then quickly drop back down. The temperatures are higher for thermal spikes near the center of the domain than near the boundary due to the 300 K fixed boundary conditions on the temperature. These high local temperatures result in local bursts of grain growth in the center regions of the domain. The grains at the boundaries stay small, as they do not experience high temperatures.

A plot of the average grain diameter from the simulation assuming a spike duration of 10 ps is shown in Fig. 1b. The plot also shows the grain size over time for a simulation without ion irradiation. Without ion irradiation, no change in grain size occurs. However, with ion irradiation, the grain size quickly increases and then levels out. However, with a spike duration of 10 ps, the predicted grain growth is much more than was observed in the experiments by Yu et al. [3]. The grain growth behavior is highly sensitive to the assumed spike duration, as shown in Fig. 1c for much shorter amounts of growth. The grain growth rate decreases with increasing spike duration. This occurs because the magnitude of the heat generation goes down with increasing spike duration as shown in Eq. (3). From these results, it is clear that a longer spike duration can be determined that will compare much better with the experiments.

CONCLUSIONS

We have developed a mesoscale model of irradiation induced grain growth using the MARMOT tool. We represent thermal spikes by adding a stochastic heat generation term to the heat equation and using the evolving temperature to determine the local grain boundary mobility. We apply the model to simulate the room temperature ion irradiations of UO_2 thin films from Yu et al. [3]. Assuming a spike duration of 10 ps results in more grain growth than shown by the experiments. However, longer spike durations decrease the growth rate, such that a longer duration can be determined that will compare better with the experimental data.

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