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ABSTRACT

A phase-field model for irradiation-enhanced densification is currently under development in the NEAMS code MARMOT. To begin, a simplified model of sintering without irradiation effects is developed. This model requires a method to generate sintering initial conditions, and the accepted phase-field rigid-body motion sintering model must either be fully implemented in MARMOT or shown to be unnecessary. It is shown to be unnecessary. Sintering simulation results without rigid body motion are shown. Shortcomings in the results are discussed and plans on how to address them are presented.

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1. Introduction

Traditionally, fuel performance codes have relied on empirical data fits. These empirical models are advantageous in several ways. They are simple models that are computationally efficient and they are accurate as long as the model stays within the available data range. However, empirical fits also have drawbacks. They are limited by the available range of data, the necessary data may be expensive or otherwise difficult to obtain, and the models themselves tell us nothing about physics controlling the processes.

In order to improve the range of fuel performance models, the Nuclear Energy Advanced Modeling and Simulation (NEAMS) program is working to develop a new set of fuel performance codes with minimal dependence on empirical models. The mechanistic models utilized by these codes will improve the range of accurate predictions beyond the limits of current empirical models[1].

One way in which the NEAMS program seeks to limit the use of empirical fits is through multiscale modeling and simulation. Results of nanometer-scale first principle calculations are fed into molecular dynamic simulations, which in turn feed into mesoscale simulations, which inform engineering scale fuel-performance models. This limits the need for empirical data to inform the engineering scale model about what is happening on the mesoscale.

One of the processes that must be modeled by fuel performance codes is irradiation-enhanced densification. During reactor operation, pores existing within the fuel close, causing the fuel pellets to decrease in volume[2]. The volume change in turn effects the heat conduction, temperature profile, and performance of the fuel. Densification is similar to the ceramic process

of sintering, but it is sped up by fission products. This project seeks to develop a mesoscale phase-field model for densification in the NEAMS code MARMOT. The results of this model will be used to help inform a fuel performance model on the engineering scale. Finally, the engineering scale model will be implemented in the NEAMS fuel performance code BISON[3].

2. Phase-Field Modeling of Sintering

To begin development of a densification model, the physics should be simplified by not including the effects of irradiation. This simplified model is then a sintering model. Sintering can be modeled in MARMOT via the phase-field method. Currently, the standard phase-field sintering model is the Wang Rigid Body Motion (RBM) model[4–6]. This model allows the individual particles to move as rigid bodies within the simulation, and is believed to improve the accuracy of simulations[7]. It does this by adding a rigid body velocity term to the standard phase-field equations. The rigid body velocity is applied to all points within a grain such that all of the points within the grain move together.

Previously, only small sintering problems have been modeled in MARMOT. In order to model large problems there are several changes that must be made to the code. First of all, the RBM model as currently implemented does not scale well. But before improving the scalability of the model we wanted to check how much effect the model really has. Second, we need a way to create initial conditions for simulations with particles following a given size distribution all in contact one with another, both in 2D and 3D simulations.

2.1 Checking the Accuracy of the RBM Model

In studying the RBM model, we believe its use is unjustified. The reported justification for the rigid body model is to capture rigid body motion of the particles caused by rapid diffusion along particle surfaces. However, the phase field model is very adept at modeling diffusion and

should be able to capture this effect without special treatment. Furthermore, the results showing the model improves accuracy are incorrect. Kazaryan et al.[5] developed a toy phase-field model of a single pore on an infinite grain boundary with an analytical solution. The solution has the pore closing as the gas diffused on the grain boundary and into the bulk. They then simulated this system with and without an early version of the RBM model[6] and found that only the RBM model predicted pore closure. The simulations, however, did not model an infinite grain boundary but were subject to conservation of mass within the simulated region. In a finite system, the pore should not close completely because the pore gas will reach its solubility limit within the bulk and be unable to further diffuse out of the pore. It is possible that the reported results are caused by an error in their early version of the model causing it to violate conservation of mass.

In order to check this, we recreated the original simulation with and without the current form of the RBM model. Then we ran additional simulations of a similar system, but with a free external surface to act as a sink to the pore gas. These systems are shown in Figure 1. We ran

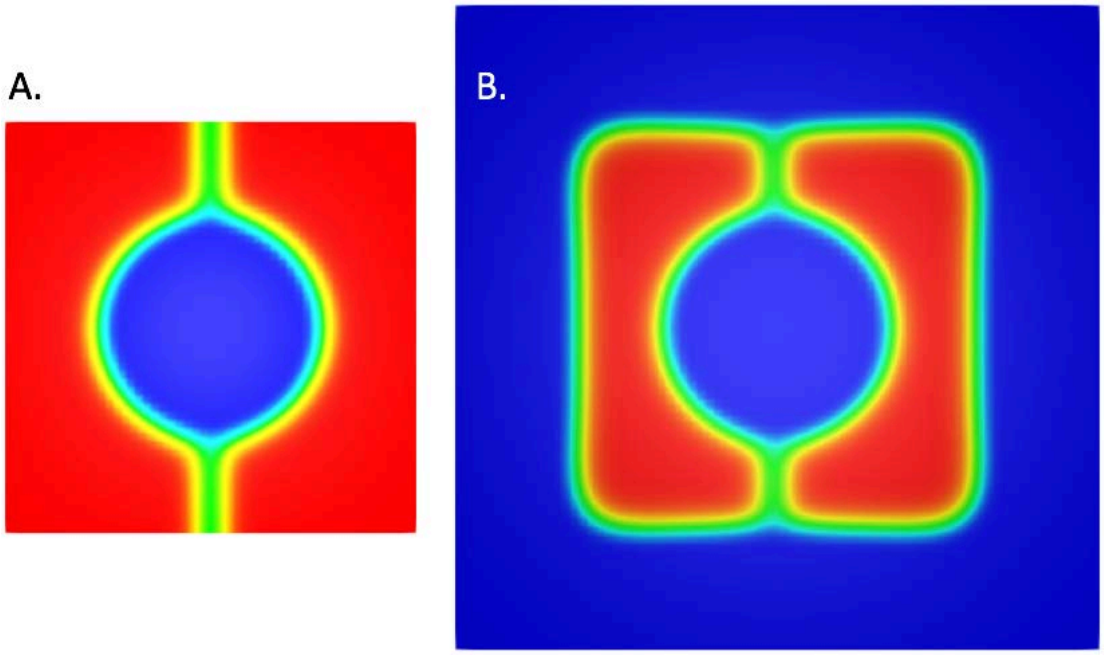

Figure 1: *A. Initial condition of grain boundary pore simulation. B. Similar simulation with an external pore gas sink.*

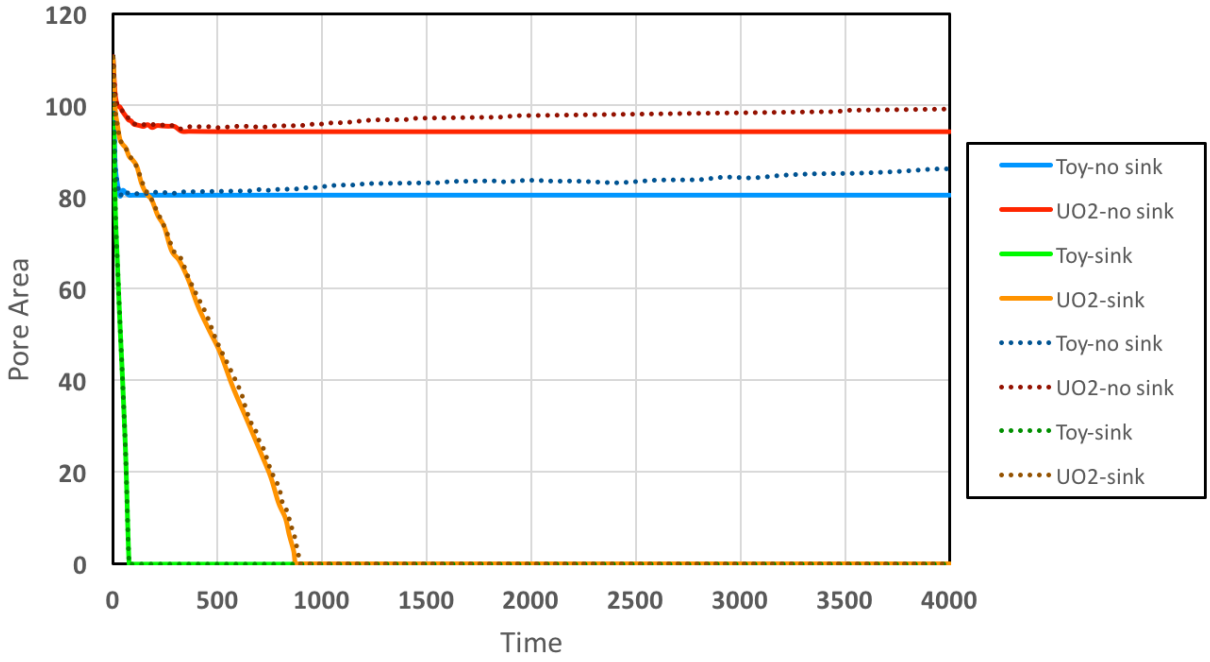

Figure 2: *Results of grain boundary pore simulations. Solid lines represent simulations without RBM while dashed lines include RBM.*

simulations both using toy problems similar to that proposed by Kazaryan et al. and with UO_2 properties already included in MARMOT. Our results are shown in Figure 2. The simulations without a sink show the pore size initially decreasing as some gas diffuses into the bulk, but ultimately reaching a constant size after the pore gas reaches its solubility limit. Simulations with sinks, however, shored the pore closing as predicted. The presence of the RBM model did not affect these results.

Furthermore, Figure 2 shows that the RBM model did violate conservation of mass by marginally increasing the total amount of pore gas in the system over time. However, it is not clear if this is a problem with the model itself or with its implementation in MARMOT.

2.2. Initial Condition Generation

Large sintering simulations require a random collection of particles that are all in contact one with another. We accomplished this by writing a MATLAB script which randomly generates circles in 2D or spheres in 3D, moving them to follow a free energy function, and outputting their positions and radii in a text file.

Next, an initial condition was added to MARMOT that could read the text file and assign variables to the circles without the same variable being used in two adjacent circles. This facilitates the interaction with a tool in MARMOT called GrainTracker that allows the simulations to increase the number of particles without increasing the computational cost. A sample initial condition generated in this manner which follows a size distribution given in Burk et al.[8] is shown in Figure 3.

3. Sintering Simulations in MARMOT

The initial condition in Figure 3 was simulated using only the standard phase-field equations in MARMOT using real UO_2 properties and designed to replicate the experiment done by Burk et al.[8]. The simulation started at room temperature, increased in temperature at a rate of 4°C per minute, held steady at 1,650°C for two hours, then cooled at 5°C per minute back to room temperature. Density was calculated using a flood algorithm. It finds all of the connected points on the mesh where the gas concentration is above a certain threshold and

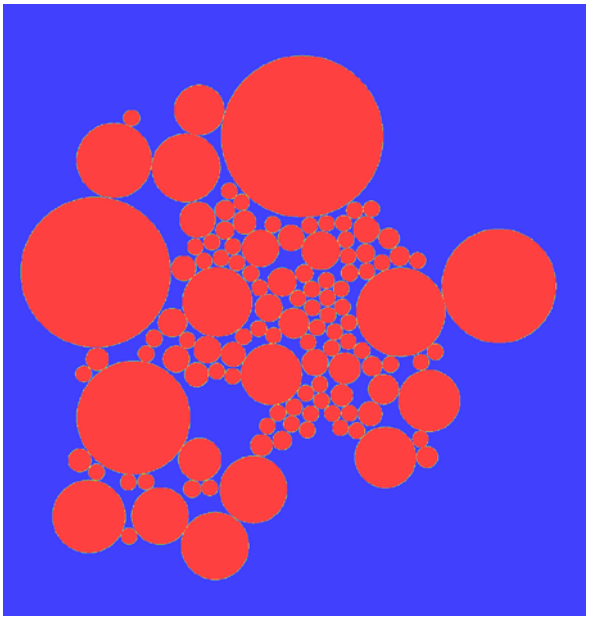

Figure 3: *Initial condition for MARMOT sintering simulation following particle size distribution from Burk et al.*

adds the volumes of those points to determine the volume of each pore. The results of this simulation are shown in Figure 4.

Figure 4 shows two important problems. The first problem is that the density is initially much too high. Experiments start with powder densities between 5 and 6 g/cm^3[8–10]. This simulation had an initial density closer to 9.5 g/cm^3. The reason for this is that real UO_2 powder forms agglomerates with much larger pores[11]. Therefore, the MATLAB script needs be updated to include agglomerates and larger interior pores.

The second major problem is that the flood algorithm is finding small channels between particles that connect the external void to the interior pores, incorrectly counting those interior pores as part of the external void. Once sintering begins, the channels close and the measured density becomes more accurate. In 2D simulations this can be mitigated by adjusting the threshold value used by the flood algorithm, but in 3D simulations it will necessitate a new

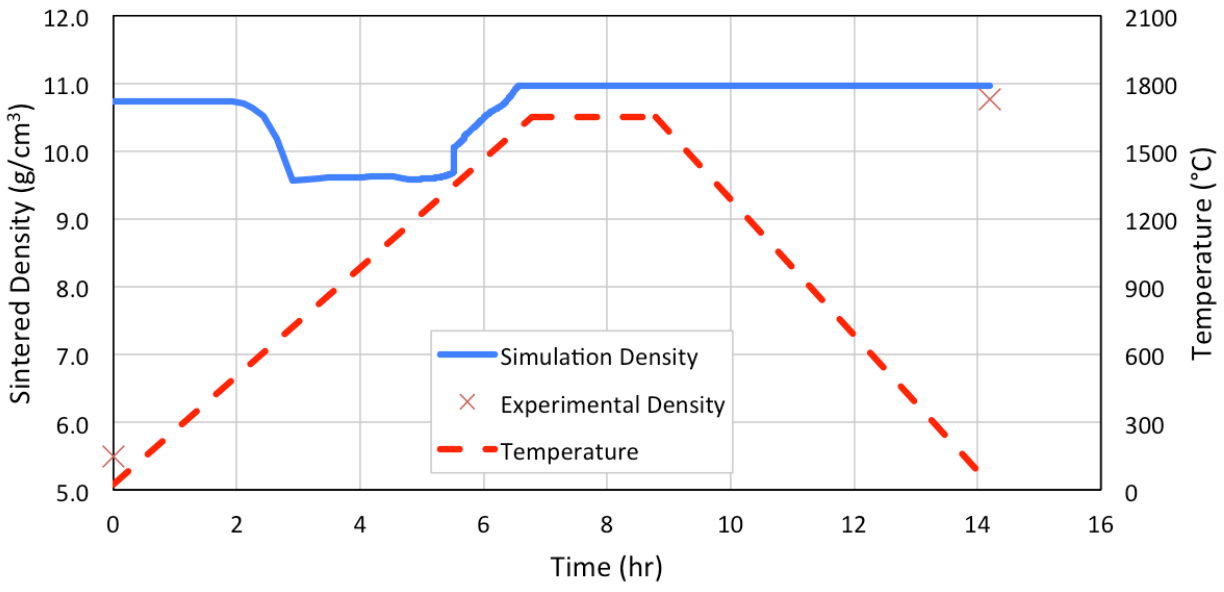

Figure 4: *Results of simulation recreating the experiment by Burk et al. along with the experimental results. The initial density is much too high on its own, but is also being artificially inflated by the method used to calculate density in the simulation.*

method to calculate density in the simulations.

4. Summary

Before irradiation effects can be included in the densification model, the sintering model must be validated. But before it can be validated it must be scaled to a level where it can be used to replicate experimental results. To do this we must generate accurate initial conditions, improve the implementation of the RBM model in MARMOT, and develop a method to better calculate the simulation density.

To generate initial conditions we use a MATLAB script. Random particles following a given size distribution and placed such that they are randomly distributed and in contact. The results are output to a text file which is read by MARMOT. MARMOT then assigns variables to the particles. However, more work is still needed to generate initial conditions with lower densities such that they match the initial conditions in experimental data.

We have determined that we can simulate sintering without using the RBM model. Rather, the standard phase-field equations should be sufficient to capture sintering. The RBM model does not improve the accuracy of sintering simulations, but it may violate conservation laws, which gave false results that were incorrectly interpreted as more accurate in previous works.

Finally, we will shortly begin investigating new ways to accurately determine simulation densities, both in 2D and 3D. 2D simulations are not as accurate as 3D simulations and so will not be as useful for validating the sintering model.

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