Grain Growth in Porous Oxides: Diffuse-Interface Modeling and Experiments

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Introduction

- All physical properties of ceramics depend strongly on the grain size. Furthermore, the grain size affects their behavior under extreme conditions such as irradiation, high temperatures, and stress.
- Modeling the process of grain growth in porous ceramics is complicated by the interaction between the pore and the grain boundary.
- The classical models by Nichols, Brook, and Carpay give only a qualitative description of the problem since they assume homogeneous microstructures and rigid body motion of the pore.
- Therefore, the phase field model parameters are directly related to the regular thermodynamic and kinetic ($T\alpha\alpha$, $\sum_kT\gamma_k\alpha\alpha\gamma\delta\alpha\gamma\beta\delta\omega$) behavior.
- More advanced model have been introduced to take into account the details of the pore and boundary geometries and motions. Evans, Riedel and Svoboda proposed the sharp-interface description of the problem. We have recently introduced the phase field (diffuse-interface) description of the problem. The phase field model alleviates all the unrealistic assumptions of the classical models and obviates all the numerical difficulties of the sharp-interface model.
- We present here for the first time 3D simulations of the grain growth process in porous ceramics.

The Sharp-Interface Model

- In the sharp-interface description by Evans, Riedel and Svoboda, the grain boundary migrates under the influence of its mean curvature, while the pore moves via surface diffusion.
  \[ v_p = -g \frac{\alpha}{\kappa}, \quad (\text{motion by mean curvature flow}) \]
  \[ v_p = \frac{\beta}{\kappa^2}, \quad (\text{motion by surface diffusion}) \]
- At triple-junctions, the balance of forces, fluxes, and continuity of chemical potential must hold. Solving this problem for general geometries is a cumbersome task, and only 2D simulations of this model have been performed.

The Diffuse-Interface Model

- The conserved density field ($\rho$) and non-conserved orientation field ($\eta$) are used to fully represent the microstructure of a porous polycrystalline ceramic. The free energy of the non-uniform medium is given by.
  \[ F = \frac{1}{2} \left[ \rho \eta_1 \eta_2 \eta_3 \right] + \frac{\gamma}{2} \left[ \sum_{i=1}^3 \left( \rho \eta_i \right)^2 \right] \]
- From irreversible thermodynamics, the kinetic equations of the order parameters are derived as.
  \[ \frac{\partial \rho \eta_i}{\partial t} = \nabla \cdot \left( \rho \eta_i \mathbf{v} \right) \]
- The kinetic equations are solved using a standard explicit finite difference scheme. In order to solve the problem efficiently in 3D, we have utilized parallel computing.

Asymptotic Matching and Model Implementation

- We carried out a formal asymptotic analysis of the phase field model in the limit where the interface thickness vanishes. The equations of motions of the grain boundary and the pore (free) surface in the diffuse-interface description are given by.
  \[ v_p = -\frac{\alpha}{\kappa^2}, \quad v_p = \frac{\beta}{\kappa^2} \]
  \[ v_p = \frac{\beta}{\kappa^2}, \quad v_p = (1+q)\frac{\alpha}{\kappa^2} \]
- Therefore, the phase field model parameters are directly related to the regular thermodynamic and kinetic (sharp-interface) parameters as follows.
  \[ \frac{\partial^2 \rho \eta_i}{\partial t^2} = \frac{\partial}{\partial \eta_i} \left( \frac{\partial F}{\partial (\rho \eta_i)} \right) \]
- The kinetic equations are solved using a standard explicit finite difference scheme. In order to solve the problem efficiently in 3D, we have utilized parallel computing.

Results

Effect of porosity on the kinetics of grain growth in UO₂ at 2000K. The grain growth process slows down as the amount of porosity increases. K. Ahmed et al., JNM, 2014

Effect of porosity on the kinetics of grain growth in CeO₂ at 1700K. The grain growth process slows down as the amount of porosity increases.

Comparison of the extrapolated model predictions and the experimental data of grain growth in 4% porous UO₂ at 2000K. K. Ahmed et al., JNM, 2014

3D snapshots and 2D cross-sectional views of the microstructure evolution of CeO₂ at 1700K. Pore breakaway is evident. In a heterogeneous microstructure, some pores detach from the migrating grain boundaries and some move along with them. Pores on two-grain junctions (edge pores) separate easily from a migrating boundary, while pores on two- and three-grain junctions tend to move along with it. Moreover, an isolated pore inside a grain could get picked up by a migrating boundary. A pore could go through a series of attachments and detachments. It is worthy noting that grains that experienced pore breakaway are larger than their neighbors, which demonstrates that pore breakaway initiates abnormal grain growth.