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

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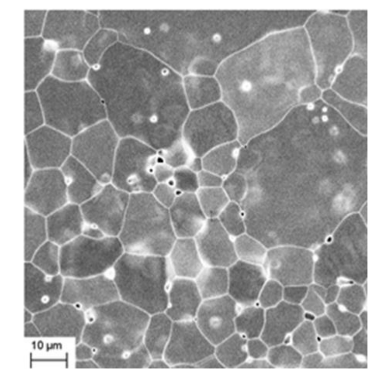
CMSNF Center for Materials Science of Nuclear Fuel

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



A typical microstructure of a porous ceramic.

- 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.

Sharp- and Diffuse-Interface Models of Grain Growth in Porous Ceramics

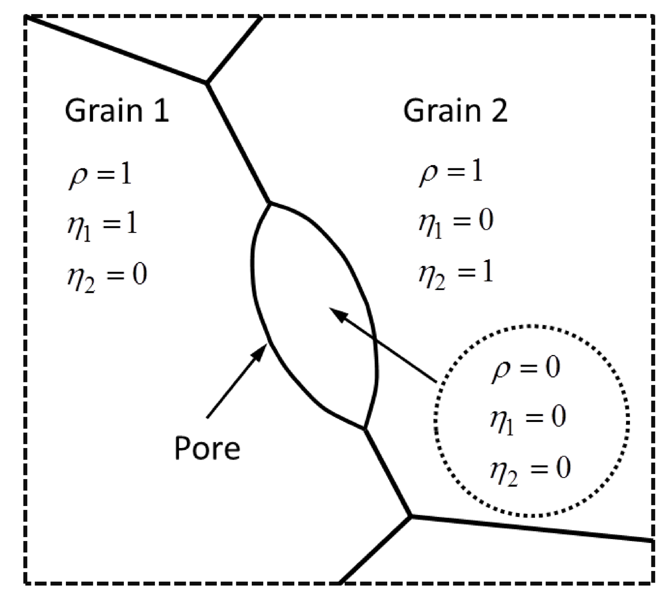
The Sharp-Interface Model

- 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.

$$v_b = -\gamma_b M_b \kappa_b, \qquad \text{(motion by mean curvature flow)}$$

$$v_p = \frac{\gamma_s D_s \delta\Omega}{K_B T}\nabla_s^2 \kappa_s. \qquad \text{(motion by surface diffusion)}$$

- 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.



A schematic illustration of the order parameters used in the diffuse-interface model. K. Ahmed et al., JNM, 2014

The Diffuse-Interface Model

- The conserved density field (η) and non-conserved orientation field (ρ) are used to fully represent the microstructure of a porous polycrystalline ceramic. The free energy of the non-uniform medium is given by,

$$F = \int_V [\, f(\rho,\eta_1,....,\eta_\alpha,....,\eta_p) + \frac{q\varepsilon^2}{2}|\nabla\rho|^2 + \frac{\varepsilon^2}{2}\sum_{\alpha=1}^{p}|\nabla\eta_\alpha|^2 \,]\, d^3r.$$

- From irreversible thermodynamics, the kinetic equations of the order parameters are derived as,

$$\frac{\partial\rho}{\partial t} = \nabla\cdot[M_s\rho^2(1-\rho)^2(\mathbf{I}-\hat{\mathbf{n}}_s\otimes\hat{\mathbf{n}}_s)]\nabla\frac{\delta F}{\delta\rho},$$

$$\frac{\partial\eta_\alpha}{\partial t} = -L\frac{\delta F}{\delta\eta_\alpha}; \qquad \alpha = 1, 2....p.$$

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,

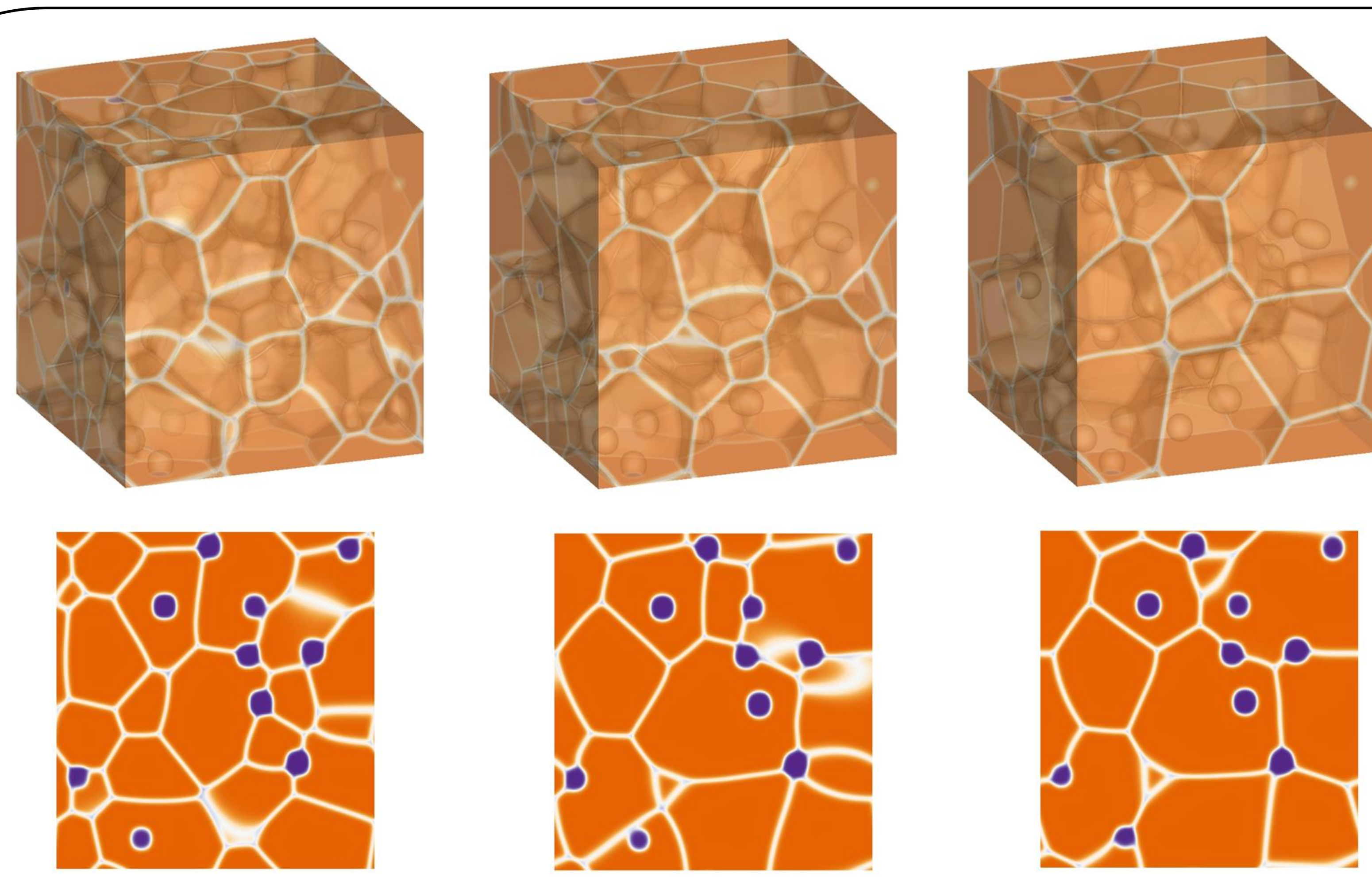
$$v_b = -L\varepsilon^2\kappa_b, \quad v_p = (1+q)M_s\varepsilon^2\nabla_s^2\kappa_s.$$

- Therefore, the phase field model parameters are directly related to the regular thermodynamic and kinetic (sharp-interface) parameters as follows,

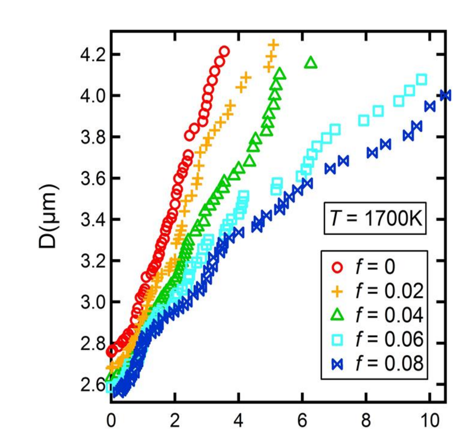
$$L\varepsilon^2 = \gamma_b M_b, \quad (1+q)M_s\varepsilon^2 = \frac{\gamma_s D_s \delta\Omega}{K_B T}.$$

- 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.

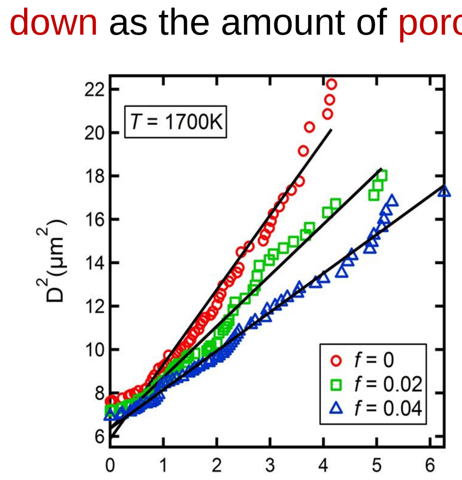
Results



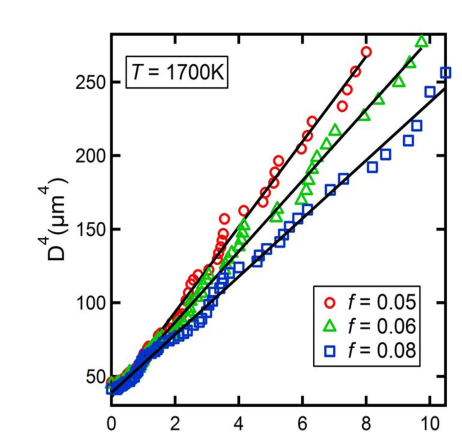
3D snapshots and 2D cross-sectional views of the microstructure evolution of CeO_2 at 1700K. Pore breakaway is evident. In a heterogeneous microstructure, some pores detach from the migrating grain boundaries and some move along with them. Pores on two-grain junctions (edge pores) separate easily from 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.



Effect of porosity on the kinetics of grain growth in CeO_2 at 1700K. The grain growth process slows down as the amount of porosity increases.

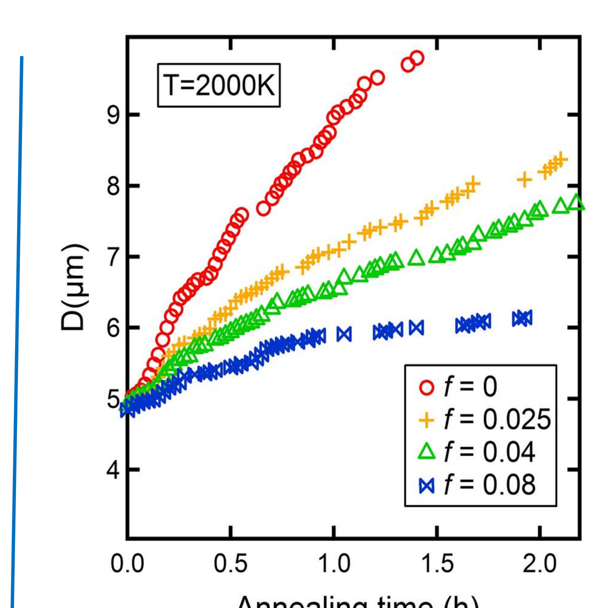


(a) Boundary-controlled kinetics

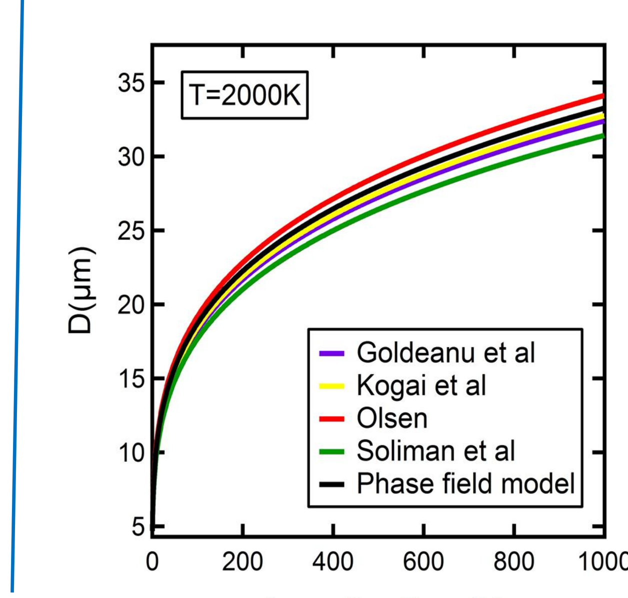


(b) Pore-controlled kinetics

The kinetics of grain growth in CeO_2 at 1700K. The grain growth kinetics changes from boundary-controlled to pore-controlled as the level of porosity increases. Furthermore, in each growth mode, the rate constant (or activation energy) is sensitive to the precise amount of porosity.



Effect of porosity on the kinetics of grain growth in UO_2 at 2000K. The grain growth process hinders as the amount of porosity increases. K. Ahmed et al., JNM, 2014



Comparison of the extrapolated model predictions and the experimental data of grain growth in 4% porous UO_2 at 2000K. K. Ahmed et al., JNM, 2014