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Stability of grain boundary texture during isothermal grain growth in UO_2 considering anisotropic grain boundary properties

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ABSTRACT

In the present study, mesoscale simulations of grain growth in UO_2 are performed using a 2D level set representation of the polycrystal grain boundary network, employed in a finite element setting. Anisotropic grain boundary properties are considered by evaluating how grain boundary energy and mobility varies with local grain boundary character. This is achieved by considering different formulations of the anisotropy of grain boundary properties, for example in terms of coincidence site lattice (CSL) correspondence. Such modeling approaches allow tracing of the stability of a number of characteristic low-Σ boundaries in the material during grain growth. The present simulations indicate that anisotropic grain boundary properties have negligible influence on the grain growth rate. However, considering the evolution of grain boundary character distribution and the grain size distribution, it is found that neglecting anisotropic boundary properties will strongly bias predictions obtained from numerical simulations.

1. Introduction

The microstructure of uranium dioxide (UO_2) decisively determines the macroscopic thermo-mechanical properties of the material and also controls the processes whereby its microstructure evolves and eventually degrades. Grain growth occurs in the material under high-temperature conditions through migration of mobile, generally high-angle, grain boundaries. Grain boundary properties such as boundary energy and mobility tend to vary with the local grain boundary configuration, giving rise to anisotropic grain growth kinetics. In addition, grain boundary migration will be impeded by any presence of pores or particles. Further, as grain boundaries frequently constitute the origin of material damage and processes of creep, segregation of impurities and embrittlement it is evident that knowledge of the grain boundary structure is pivotal for tracing the state of the material. The present study focuses on the stability of grain boundary character distribution during grain growth in UO_2 under sintering conditions when anisotropic grain boundary properties are considered.

A full characterization of an individual grain boundary requires knowledge of three parameters in order to describe the crystallographic misorientation across the interface and two additional parameters to define the inclination of the boundary plane [1–3]. Although both grain boundary energy and mobility are usually observed to vary with the full set of five parameters, the dependence of the parameters on grain boundary character is complex and no general theory exists to describe it. Experimental observations on grain boundary energy and mobility are usually made on limited ranges of misorientation and for chosen subsets of boundary configurations, such as symmetrical tilt and twist boundaries with well-defined boundary planes. Recognizing the complexity of these relations, the concept of coincidence site lattice (CSL) boundaries provides a simplifying approach. The CSL concept is based on the identification of some degree of geometrical agreement between the two adjacent crystal lattices, on opposite sides of the grain boundary. Although based on geometrical considerations, rather than on experimental observations, the CSL concept remains influential when it comes to describe grain boundary character and related grain boundary properties [3]. For example, CSL boundaries are often observed to have low energy and high mobility character [4]. The CSL concept also remains important in the field of grain boundary engineering [5]. Regarding UO_2, it can also be noted that an increased diffusion rate for some CSL boundaries has been reported [6,7]. The CSL description of grain boundary character is adopted in the present work.

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When it comes to mesoscale modeling of grain growth, different approaches can be taken as discussed in Ref. [8]. As examples, grain growth in UO_2 is modeled by empirically-based closed-form expressions in Refs. [9–11]. Turning to numerical models, grain growth and pore migration in UO_2 is studied using a 2D Monte Carlo Potts (MCP) model in Ref. [12] while a 2D phase field (PF) model is employed in Ref. [13] to study grain growth in porous UO_2, assuming isotropic grain boundary properties. Zener-like pinning of grain boundaries by pores was found in Ref. [14] by using MCP simulations of grain growth in UO_2 and grain growth in a porous material was also simulated by a 3D MCP model in Ref. [15]. The inhibition of grain boundary migration due to pores as a type of Zener pinning is also discussed in Ref. [16]. Further, 2D MCP models are used to trace the evolution of CSL boundaries during grain growth – although not in UO_2 – in Refs. [17,18].

In passing it can also be noted that some model-based studies on UO_2 focus on void growth while keeping the grain boundaries fixed. This is done analytically in Ref. [19] and by phase field modeling in Refs. [20,21].

The level set method was introduced in Ref. [22] and provides a method to trace the evolution of interfaces in different physical settings. Level sets have been used for mesoscale modeling of polycrystals in several studies. Recrystallization in isotropic systems is modeled by level sets in Refs. [23–26]. Grain growth with isotropic grain boundary properties is approached by the same method in Refs. [27,28]. Grain growth with a Read–Shockley description of grain boundary energy is considered in the level set model employed in Ref. [29]. Particle pinning of grain boundaries, modeled by level sets, is considered in Ref. [30].

The present level set formulation is adopted from Refs. [31], where the focus lies on dynamic recrystallization. A similar model was also adopted in Ref. [32] to trace the evolution of grain boundary character distribution during grain growth in polycrystals with cubic structure.

Most mesoscale models of grain growth adopt the simplifying assumption of isotropic grain boundary properties in terms of boundary energy and mobility. Mesoscale models of grain growth where anisotropic grain boundary properties are taken into account are significantly more scarce, and particularly so related to UO_2. If anisotropic energy is considered, it is usually done by adopting a Read–Shockley model for low-angle boundaries where the interface energy depends solely on the scalar misorientation across the interface. The common approach in these models is to assume a constant energy for all high-angle grain boundaries. Anisotropic grain boundary energy and mobility is an important aspect of the kinetics of microstructure evolution [3,33]. The grain boundary character will also influence the diffusion of fission products in the material [34]. It can be noted that grain structures in UO_2 with a significant fraction of special boundaries – in terms of CSL boundaries – have been reported [35]. Such microstructure variations suggest anisotropic grain boundary properties to be an influential factor on the microstructure evolution in this material.

This paper is structured into five sections. Beginning with a note on modeling of grain growth kinetics in Section 2, subsections provide details on the formulation of anisotropic grain boundary properties based on CSL correspondence. Section 3 contains the level set formulation of grain growth. Mesoscale simulations of grain growth in UO_2 are performed in Section 4 where the results are also shown and discussed. Finally, some concluding remarks closes the paper in Section 5.

2. Modeling grain growth kinetics

The local migration velocity of a grain boundary can be written as

$$v = mp \tag{1}$$

where m and p is the grain boundary mobility and the driving pressure, acting on the grain boundary, respectively.

Focusing the study on grain growth, the driving pressure due to a purely curvature-driven grain boundary motion can be formulated as

$$p = -\kappa\gamma \tag{2}$$

where the local grain boundary curvature κ and the grain boundary energy γ were introduced.

In the present study, anisotropic grain boundary properties will be introduced in terms of the grain boundary energy γ and the grain boundary mobility m. This is detailed in the following subsections.

2.1. CSL classification of grain boundary character

In a polycrystalline aggregate it can be assumed that for some boundaries a certain degree of geometrical correspondence can be found between the atomic arrangements on opposite sides of the boundary. The notion of a *Coincidence Site Lattice* (CSL) is based on identification of the coinciding lattice positions in the two neighboring crystals. Boundaries where such geometrical agreement are found, have in several studies been discovered to posses "special" properties, such as low energy and high mobility. The degree of CSL correspondence is usually indicated by an integer number Σ, where $1/\Sigma$ is the fraction of coinciding lattice positions.

Crystal orientations need to be defined by three parameters which in the present study are taken as the three Euler–Bunge angles $(\varphi_1, \Phi, \varphi_2)$. Due to the many possible configurations of these parameter sets in adjacent crystals, it is unlikely to encounter ideal CSL boundaries. To address this, an acceptance criterion was introduced by Brandon in Refs. [36], defining a range of misorientation angles $\Delta\theta_\Sigma$ related to an individual CSL configuration. The Brandon criterion is given by,

$$\Delta\theta_\Sigma = \frac{\theta_\Sigma}{\Sigma^{1/2}} \tag{3}$$

where θ_Σ is a parameter that determines the range of misorientation angles that are assumed to belong to each CSL. In the present work, the common choice of $\theta_\Sigma = 15^\circ$ is made.

Based on the Euler angles, the orientation of each crystal can be defined by the orthogonal rotation matrix $\boldsymbol{g}(\varphi_1, \Phi, \varphi_2)$ that rotates the sample reference frame into the crystal reference frame. The misorientation between two crystals with rotations $\boldsymbol{g}_i$ and $\boldsymbol{g}_j$ can subsequently be evaluated as $\Delta\boldsymbol{g}_{ij} = \boldsymbol{g}_j\boldsymbol{g}_i^{\mathrm{T}}$.

In a cubic system, there are 24 symmetrically equivalent variants of each orientation $\boldsymbol{g}$. In evaluation of the misorientation, the common approach is to consider the set of variants that provide the minimum misorientation. By this approach, the scalar misorientation is found by performing the minimization.

$$\theta = \min_{\boldsymbol{O}_s \in \mathscr{G}_c} \left| \operatorname{acos}\left[\frac{1}{2}\{\operatorname{tr}(\boldsymbol{O}_s\Delta\boldsymbol{g}) - 1\}\right]\right| \tag{4}$$

where indices i and j are skipped for convenience and where $\boldsymbol{O}_s$ are the operators in the cubic symmetry group $\mathscr{G}_c$. The trace of a tensorial quantity is denoted by $\operatorname{tr}(\cdot)$ and the absolute value is taken of the argument in Eq. (4) since a negative sign is merely an indication of the rotation axis pointing in the opposite direction. Due to the minimization in Eq. (4), the maximum misorientation that will be encountered in a cubic polycrystal is 62.8°, as shown in Ref. [37].