



Influence of grain boundary energy on the nucleation of complexion transitions

Stephanie A. Bojarski,^a Martin P. Harmer^b and Gregory S. Rohrer^{a,*}

^aDepartment of Materials Science and Engineering, Carnegie Mellon University, Pittsburgh, PA 15213, USA

^bDepartment of Materials Science and Engineering, Lehigh University, Bethlehem, PA 18015, USA

Received 19 May 2014; revised 15 June 2014; accepted 19 June 2014

Available online 2 July 2014

To test the effect of grain boundary energy on the nucleation of a transition from a low mobility to a high mobility grain boundary complexion, polycrystalline yttria-doped alumina was sandwiched between two single crystals of sapphire. Thermal groove measurements showed that the distribution of grain boundary energies at the single crystal–polycrystal interface is biased by the single crystal orientation. When annealed above the critical temperature for the complexion transition, the high mobility complexion preferentially nucleated at the higher energy interface.

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Keywords: Grain boundary; Complexion; Microstructure; Grain boundary energy; Alumina

It has recently been found that the structure and chemistry of a grain boundary can change abruptly as certain thermodynamic parameters are varied and this can lead to changes in the properties of the boundary, such as the energy and mobility [1]. For example, below a critical temperature, a monolayer of adsorbed impurities might be segregated to a grain boundary and above this temperature a bilayer of impurities might be stable [2]. These distinguishable states are now referred to as complexions [1–3]. While complexions have been characterized in a variety of materials, including metals [4], semiconductors [5] and ceramics [6], the most extensively studied complexions are found at the grain boundaries of doped aluminas [2]. In this case, the observations show that, when the temperature is changed, the transformation from one complexion to another is not immediate, or at least does not happen immediately at every grain boundary [7]. This suggests that the transformation from a metastable complexion to a stable complexion is limited by a nucleation energy barrier and/or that different types of grain boundaries have different transition temperatures. It has been hypothesized that higher energy grain boundaries have a lower nucleation energy barrier for the transformation and therefore transform first, or

that they have a lower critical temperature for the transformation [2,8,9]. Either of these two circumstances would lead to a mixture of transformed and untransformed boundaries at a constant temperature. The goal of this paper is to describe an experiment designed to test the hypothesis that grain boundaries with higher relative energies preferentially undergo complexion transitions.

Our experiment uses a unique sandwich-like sample, as shown schematically in Figure 1. The sample consists of a doped alumina polycrystal sandwiched between two single crystals of sapphire with known orientations. Grain boundary energies vary with the grain boundary plane orientation [10]. Therefore, the single crystals bias the distribution of grain boundary plane orientations at the single crystal–polycrystal interface and, with it, the distribution of grain boundary energies [11]. So, by using a sapphire crystal that has a surface orientation associated with high energy boundaries on one side and a crystal with an orientation associated with low energy boundaries on the other side, distinct grain boundary energy distributions will be created at the two single crystal–polycrystal interfaces. If the chemical potential of yttria, the temperature and the pressure are constant throughout the sample, the grain boundary energy is the only variable parameter. According to the hypothesis stated above, the transition to a higher mobility complexion should occur preferentially at the interface with a relatively higher energy.

* Corresponding author. Tel.: +1 412 268 2696; fax: +1 412 268 3113; e-mail: gr20@andrew.cmu.edu

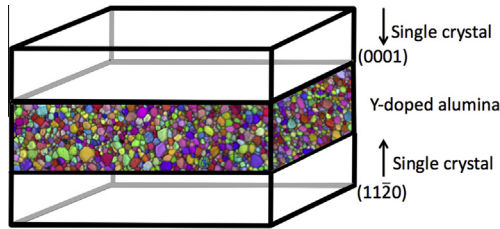


Figure 1. Schematic illustration of the sandwich sample. A polycrystalline Y-doped alumina polycrystal is sandwiched between two sapphire crystals of different orientation and diffusion bonded in a hot press or spark plasma sintering furnace.

For this experiment, we employ Y_2O_3 -doped Al_2O_3 . This pseudo-binary system is selected because there is already considerable documentation about the complexion transition in this system [12]. Based on a previous study of the grain boundary character distribution of yttria-doped alumina, it was concluded that grain boundaries with (0001) orientated planes have higher energies than those with (11 $\bar{2}$ 0) or ($\bar{1}$ 012) orientations [13]. Using this knowledge, we will test the idea that grain boundary energy influences the nucleation of the complexion transition by comparing the grain growth behavior at the higher energy (0001) plane with growth at the lower energy interfaces under the same conditions.

Yttrium-doped alumina powder was prepared by mixing pure alumina powder (Puratronic, Alpha Aesar 99.995% pure) with 500 ppm $Y(NO_3)_3 \cdot 6H_2O$ (Alpha Aesar, Ward Hill, MA). Sapphire single crystals (10 × 10 × 0.5 mm) were purchased (MTI Corp, Richmond, CA) and thoroughly cleaned to avoid contamination. The sandwich sample was organized in a graphite die such that 1.5 g of Y-doped alumina powder was interfaced with the graphite plunger and the (0001) oriented sapphire was then laid parallel to the die with the polished side of the crystal oriented upward. About 4 g of Y-doped alumina powder was then layered onto the (0001) oriented sapphire, a (11 $\bar{2}$ 0) orientated sapphire single crystal was placed on top of the powder, with the polished side oriented downward, and covered with another 1.5 g of Y-doped alumina powder before inserting the ram. After uniaxial pressing, the samples were spark plasma sintered (Thermal Technologies, LLC, Santa Clara, CA) using the following thermal treatment: a ramp of 100 °C min⁻¹ to 800 °C at a pressure of 10 MPa for an initial calcination dwell of 45 min, followed by an additional 100 °C min⁻¹ ramp to 1300 °C and sintering at 50 MPa for 30 min. For comparison, a second set of samples was produced in a hot press. For this experiment, the alumina powder was doped with 100 ppm yttria and a (0001) oriented sapphire crystal was paired with a ($\bar{1}$ 012) oriented crystal. The sample was constructed as described above and then sintered by hot isostatic pressing in a vacuum at less than 10 MPa (Astro 1000, Thermal Technology Inc., Santa Rosa, CA) for 60 min to achieve near theoretical density.

Orientation data was collected on a Quanta 200 field emission scanning electron microscope with OIM EDAX Delphi software configured with TSL analysis (EDAX Inc, Mahwah, NJ). The EBSD step size was roughly one-tenth the average grain size, the accelerating voltage was 15 kV and the working distance was

10 mm. Contact atomic force microscopy topographs were recorded using a SolverNext NT-MDT atomic force microscope (NT-MDT, Zelenograd, Moscow) and Pyrex-nitride tips (Contact mode, Nanoworld, Neuchâtel, Switzerland). The topographic images were taken with a step size of 10 nm and a field of view of 5–20 μ m.

The orientation maps in Figure 2a and b show the microstructure of the samples after densification. The regions of constant orientation on the left and right sides are the edges of the single crystals. The polycrystalline material between the single crystals is dense, about 2.5 mm thick, and shows a normal distribution of grain sizes, with an average size of 1.0 μ m. The single crystal surfaces do not remain perfectly flat because they sinter with the alumina powder. However, the interfaces did not advance significantly into the polycrystal, and it was assumed that the average orientation of the interface was maintained. The polycrystal along the (0001) and ($\bar{1}$ 012) orientated interfaces had a similar appearance.

To evaluate the grain boundary energy distributions in the as-sintered state, the sample was thermally grooved in air at 1350 °C for 15 h. At least 100 individual boundaries were analyzed along both the (0001) and (11 $\bar{2}$ 0) planes. Following the procedures described by Saylor and Rohrer [14] and Dillon et al. [15], which adapt the original Mullins [16] analysis for the half angle of the groove, the grain boundary to surface energy ratio was determined for all of the grooves. For each boundary, a mean dihedral angle determined from multiple measurements of the same groove. Cumulative distribution plots for the measured dihedral angles and the relative grain boundary energies at the (0001) and (11 $\bar{2}$ 0) interfaces are compared in Figure 3(a). As expected from an earlier study of the grain boundary character distribution [13], the interface with the (11 $\bar{2}$ 0) orientation has a lower mean energy (0.55) than the one with the (0001) orientation (1.15). Furthermore, the standard deviation of the higher energy interface (0.30) is larger than that of the lower energy interface (0.21). Random boundaries spatially separated from the single crystals by at least two grains have an intermediate average energy (0.71). This indicates that the single crystals bias the energy distribution so that grain boundaries at the (0001) interface have, on average, higher energies than random boundaries and that those at the (11 $\bar{2}$ 0) interface have lower energies than random boundaries.

The sample containing the (0001) and (11 $\bar{2}$ 0) oriented crystals was then heated in air for 8 h at 1500 °C, which is above the temperature at which a grain boundary complexion transition is expected [13]. The sample was then polished to a 0.05 μ m diamond finish and thermally grooved at 1400 °C for 15 h in air. Orientation maps of this sample are illustrated in Figure 2(c) and (d). After the high temperature anneal, there is a bimodal grain size distribution in the bulk of the polycrystal, where the average size of grains smaller than 10 μ m was 3.2 μ m and the average size of grains larger than 10 μ m is 19.8 μ m. However, there is also a distinct difference between the two interfaces. The microstructure along the (0001) interface contains nearly all larger grains, but the microstructure along the (11 $\bar{2}$ 0) interface contains mostly smaller grains. Both single crystals grew into the polycrystal, where

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