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Studying alumina boundary migration using combined microscopy techniques

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Abstract. Thermal grooving and migration of grain boundaries in alumina have been investigated using a variety of microscopy techniques. Using two different methods, polycrystalline alumina was used to investigate wet, (implying the presence of a glassy phase), and dry grain boundaries. In the first, single-crystal Al_2O_3 was hot-pressed via liquid phase sintering (LPS) to polycrystalline alumina with an anorthite glass film at the interface. Pulsed laser deposition was used to deposit approximately 100-nm thick glass films. Specimens were annealed in air at 1650°C for 20 h to induce boundary migration. Boundary characterization was carried out using visible light (VLM) and scanning electron (SEM) microscopies. Effects on migration due to surface orientation of grains were investigated using electron backscatter diffraction (EBSD). The second method dealt with heat treating dry boundaries in polycrystalline alumina to monitor boundary migration behavior via remnant thermal grooves. Heat treatments were conducted at 1650°C for 30 min. The same region of the sample was mapped using VLM and atomic force microscopy (AFM) and followed over a series of 30 min heat treatments. Boundary migration through a pore trapped inside the grain matrix was of particular interest.

1. Introduction

Ceramics are often anisotropic materials where second phases and a broad grain-size distribution are often found, making transmission electron microscopy (TEM) and X-ray diffraction (XRD) for orientation studies difficult.[1] The introduction of electron backscatter diffraction (EBSD) to the microscopy community has helped to alleviate some of these difficulties. In EBSD, the surface orientation and phase of each grain may be determined by indexing Kikuchi bands formed in the scanning electron microscope (SEM) by backscattered electrons and, subsequently, produce an orientation map, or inverse pole figure (IPF) map, of the sample surface. A large, bulk sample may be used in the SEM allowing easier sample preparation and a larger sampling volume, in contrast to TEM

and XRD. The reader is referred elsewhere for an extensive description on the use of EBSD as applied to ceramics.[2]

Liquid-phase sintering (LPS) of alumina substrates to a polycrystalline slab with an amorphous thin film at the boundary allows investigation of the driving force for GBM with an emphasis on orientation effects.[3] The use of LPS allows tight control over grain size, pore size, interface orientation and composition. Since LPS occurs at high temperatures, this experimental set-up models typical processing conditions.[1, 3]

Processing ceramics at high temperatures leads to thermal grooving on the surface of the grain boundaries.[4, 5] Thermal grooves may be used in understanding stationary and migrating grain boundaries.[3, 5] As GBM proceeds, the grooves follow the boundary, but leave a remnant groove behind. Atomic force microscopy (AFM) enables the monitoring of grooves and GBM.[5, 6] In AFM, topographic information is determined quickly and easily by scanning a cantilevered tip across the sample surface enabling hills and valleys on the surface to be mapped by monitoring the degree of tip deflection.

2. Monitoring boundary migration using EBSD

2.1. Experimental

Pulsed-laser deposition (PLD) was used to deposit approximately 100 nm of anorthite ($CaAl_2Si_2O_8$) glass on single-crystal alumina of known orientation, (either *c*(0001) or *m*$\{10\bar{1}0\}$).[7] A Lucalox™ tube, high-purity polycrystalline alumina stabilized with up to 0.5 wt% magnesia, was cut into 2 mm square pieces, polished flat and then hot-pressed in air to the film/substrate assemblage inside a box furnace.[8] Use of a single crystal substrate allowed investigation of specific boundary orientations, while polycrystalline alumina allowed the study of several boundary types in a single sample. The manufactured boundary was constructed with an initially flat interface so free-energy differences across the boundary due to curvature effects were not present.

After sample fabrication, a surface was prepared perpendicular to the interface by polishing with successive grits of diamond lapping films and followed with a diamond suspension polish on a felt pad to a 0.05 μm finish. The assemblage was then heat treated in air at 1650˚C for 20 h to induce GBM. The extent and direction of migration was monitored with visible light microscopy (VLM) (Olympus BH2 UMA) and SEM (Hitachi S-900). The grain surface orientation of the boundary was determined using EBSD. Specimens were coated with 1–2 nm Pt to avoid charging under the 20 kV electron beam on a FEI XL30 FESEM.

2.2. Results and Discussion

In all cases, the initial boundary migrated into the polycrystalline region of the sample during annealing. Representative portions of the interfaces of *c*-Al_2O_3 pressed to polycrystalline Al_2O_3, (referred to as *c*/poly), and *m*-Al_2O_3 pressed to polycrystalline Al_2O_3, (referred to as *m*/poly), are shown in figures 1 and 2. The surfaces shown were tilted to 70° in order to view the remnant grooves and optimize EBSD conditions.[2] The initial boundary position was marked by the remnant grooves. While similar behavior was seen in both sample geometries, the *c*/poly migrated in a manner that kept the boundary nearly parallel to the initial boundary. The boundaries in *m*/poly samples became jagged and curved around the grains. After polishing the as-annealed surface for EBSD analysis, these behaviors were confirmed in the bulk using the IPF maps in figure 3. The average GBM distance at the surface was measured to be 6.1 and 5.3 μm for the *c*/poly and *m*/poly samples, respectively; however, any orientation dependence on GBM has not been fully explained.

The *c*-plane of alumina is considered energetically stable and will reconstruct into a terrace-and-step morphology when exposed to high temperature. The *m*-plane is unstable at high temperatures and will facet into the $s\{1\bar{1}01\}$ and $r\{\bar{1}012\}$ planes forming a hill-and-valley configuration.[9] Reconstruction increases the total surface area, but lowers the total surface energy. The single crystal at the surface of the boundary was under an energetically unfavorable condition with the neighboring

grains in the *m*/poly samples. The unfavorable misorientation caused the boundary to migrate in the curved manner observed rather than in the parallel motion exhibited in the *c*/poly samples.

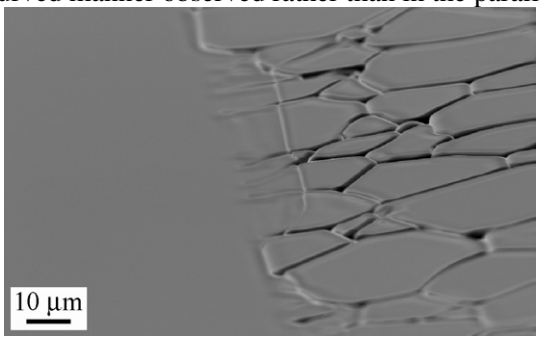

Figure 1. SEM image of the as-annealed surface of a *c*/poly specimen. The surface was tilted to 70° and shows migration of the initial boundary into the polycrystalline region. Remnant grooves mark the initial boundary position.

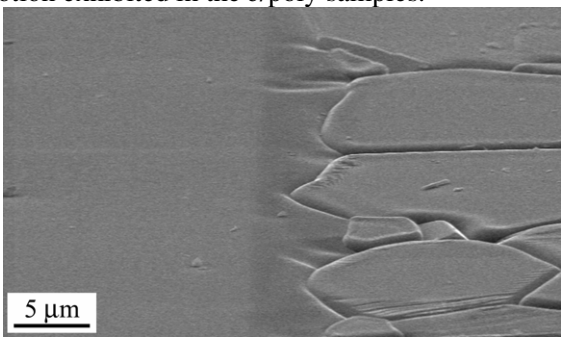

Figure 2. SEM image of the as-annealed surface, tilted to 70°, of an *m*/poly specimen. The remnant groove again marks the initially straight boundary position, but the boundary has now become curved.

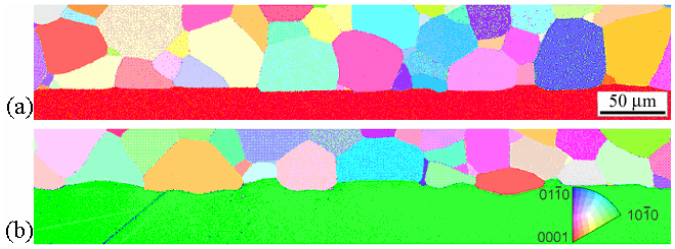

Figure 3. IPF maps of a *c*/poly (a) and an *m*/poly sample (b). Both samples were polished after annealing. The orientations mapped are with respect to the surface orientation.

3. Monitoring boundary migration using remnant grooves

3.1. Experimental

Lucalox™ tubes were again cut into 2 mm square pieces and polished using diamond lapping film until a 0.25 µm finish was achieved.[8] A box furnace operated in air was used to heat samples in an alumina crucible at a temperature increase of 20°C/min to 1650°C. The temperature was held at 1650°C for 30 min intervals and cooled at approximately 150°C/min to 1300°C. Further cooling to room temperature was at a rate of approximately 40°C/min. The series of heat treatments inducing migration was monitored via VLM and AFM. A detailed account of the sample preparation method and monitoring is described elsewhere.[5]

3.2. Results and Discussion

Using a VLM map and the remnant grooves, the same region of the polycrystalline compact was returned to with AFM (Digital Instruments Nanoscope III) after a series of four heat treatments. A pore was observed to be trapped inside a grain after the initial 30 min heat treatment, as seen in figure 4. With additional heat treatments, the grain to the right of the pore was observed to grow by means of Ostwald ripening. However, the migrating boundary did not absorb the pore, even as the boundary traveled completely through the pore. No change in pore size was observed.

Sintering theory suggests that pores should be absorbed during heat treatment and cause the polycrystalline alumina to densify during grain growth.[1] Migrating grain boundaries are natural sinks for the vacancies comprising a pore. However, the fact that Ostwald ripening was obviously occurring indicates that the sample was undergoing secondary recrystallization.[10] The pore in question was not absorbed by the grain boundary, suggesting that the grain-boundary vacancy concentration was at saturation and further densification was not possible.

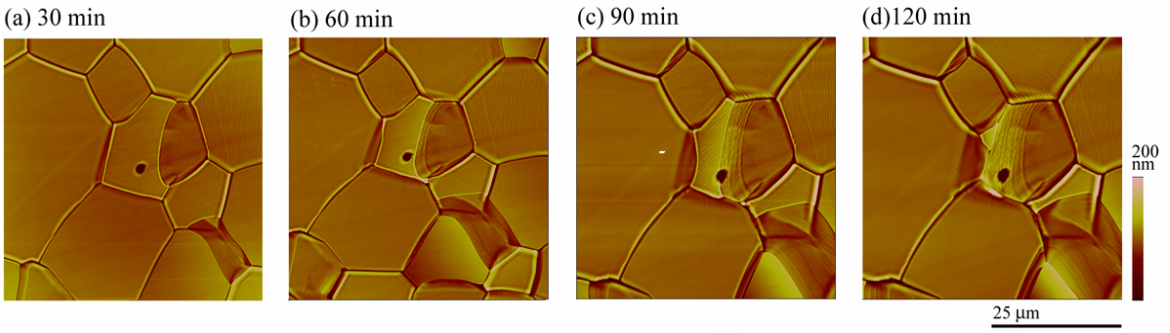

Figure 4. AFM images of the same set of grains after a series of heat treatments at 1650°C. The grain in the center of (a) has a pore initially trapped within. The interface between this grain and the adjacent grain can be seen to have migrated closer to the pore in (b), cut the pore in (c) and, finally in (d), moved entirely through the pore.

4. Summary and Conclusions

Polycrystalline alumina was hot-pressed to single crystal substrates, with two different crystallographic orientations at the boundary plane, to observe orientation effects on GBM. The *c*-plane was found to migrate nearly parallel to the initial boundary, while the *m*-plane migrated in a jagged manner. The difference in motion is attributed to the *m*-plane being an energetically unstable orientation at high temperatures. Orientation dependence on migration rate is not fully understood. Future work should include using EBSD to determine if particular orientation combinations migrate uniquely. Migration in the bulk needs to be compared to surface migration to see if similar behavior exists, particularly if the interface is faceted.

A pore in polycrystalline alumina was observed via AFM to not absorb into a grain boundary which migrated through it during heat treatment due to the sample being in a state of secondary recrystallization. The surface orientation of grains containing pores that are not observed to densify should be monitored via EBSD and cross-section TEM analysis to determine if a special relationship exists with the migrating boundary.

Acknowledgments

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