Mesoscale Response of Individual Grains of Polycrystalline $BaTiO_3$ to Electrical Loading

Mesut Varlioglu[1], Ulrich Lienert[2], Ersan Üstündag[1], Joel Bernier[2], Robert C. Rogan[1], Lawrence Margulies[3] and Henning Poulsen[3]

[1] *Iowa State University, Materials Science and Engineering, Ames, IA, U.S.A.*
[2] *Advanced Photon Source, Argonne National Laboratory, Argonne, IL, U.S.A.*
[3] *Risoe National Laboratory, Roskilde, Denmark*

Introduction

Ranging from sub-grain structures to grain clusters, the mesoscale is critical in understanding the constitutive behavior of ferroelectrics. The interaction of domains with grain boundaries and grain-to-grain constraints largely determine the overall response of a ferroelectric. Unfortunately, the most severe paucity of quantitative data also exists at the mesoscale. The newly developed 3-D XRD technique offers a unique opportunity to study the response of ferroelectrics to external stimuli at this scale by allowing the investigation of individual grains within a polycrystal. To demonstrate the feasibility of 3-D XRD in ferroelectrics research for the first time, a polycrystalline $BaTiO_3$ was subjected to quasi-statically cycled electric field.

Methods and Materials

The experiments were performed at beamlines 1-ID-C of the Advanced Photon Source (APS) and ID-11 of the European Synchrotron Radiation Facility (ESRF), Grenoble, France. In both cases, nominally the same polycrystalline $BaTiO_3$ samples (about 1x1x5 mm^3) were employed. The specimens were held upright in the geometry shown in Fig. 1 and run in transmission mode using ~80 keV X-rays. The direction of the electric field was perpendicular to the X-ray beam and it was stepped up to a maximum of ±20 kV/cm, well above the nominal coercive field of the material (~5 kV/cm). At each applied field value, the specimen was rotated perpendicular to the beam in 0.1° ω steps (Fig. 1) up to ±45° (at ESRF) and ±65° (at APS). The X-ray spot size was about 150x150 μm^2 at ESRF and 30x30 μm^2 at APS. Since the grain size of the $BaTiO_3$ samples was measured to be ~20-30 µm from SEM images, either sampling volume was large enough to capture a reasonable number of grains.

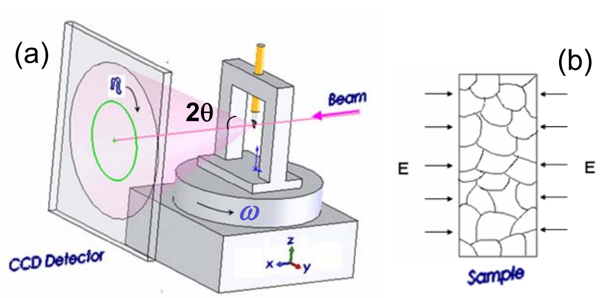

Fig. 1. (a) 3D-XRD setup. (b) Direction of the electric field perpendicular to the X-ray beam at ω = 0°.

The diffraction data were analyzed using the *GRAINDEX* software from Risoe. This analysis attempted to identify individual grains and track their response to electric field. In a parallel study, the macroscopic (polycrystalline) response of the material was obrained by integrating data within ±10° ω around the ω = 0° position (perpendicular to the beam) and parallel to the electric field using the *Fit2D* software from ESRF (Fig. 2).

Results and Discussion

The macroscopic behavior of the material (Fig. 2) was as expected from a polycrystalline ferroelectric: the electric field led to an increase of the 002 intensity due to domain alignment along the field direction. Fig. 2 also displays the MRD value (multiples of random distribution relative to an unpoled sample) obtained from this equation: $MRD = 3I_{002} / (I_{002}+2I_{200})$. The MRD is a better measure of texture evolution due to domain re-orientation and should be 1.0 for a random polycrystal. In Fig. 2, while the initial state of the specimen is not random (MRD ~ 0.7), the applied field leads to significant domain re-alignment (MRD ~ 1.5 at 2000 V).

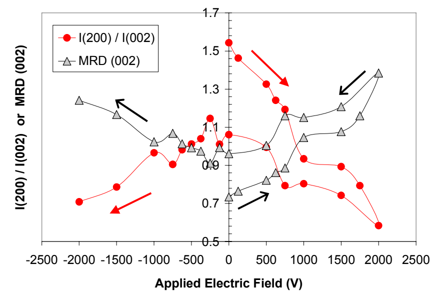

Fig. 2. Variation of the integrated intensity ratio (I_{200}/I_{002}) and MRD_{002} as a function of electrical loading. Diffraction images within ±10° of the ω = 0° position and along the electric field were summed to obtain these macroscopic data. The arrows indicate the progression of the electrical loading.

Unfortunately, the *GRAINDEX* analysis has not been successful so far in identifying individual grains. This is largely due to the presence of domain variants which results in closely spaced multiple Laue spots. However, a close inspection of the diffraction patterns does confirm the presence of significant variations in the response of grains to electrical loading (Fig. 3).

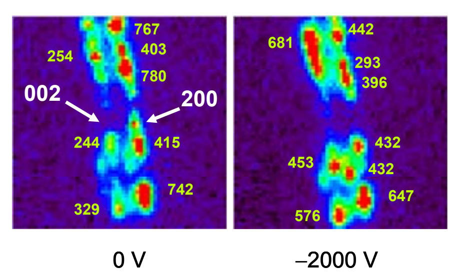

Fig. 3. A section of the 200/002 Debye rings as a function of applied electric field. Here, the data were summed within ±10° of the ω = 0° position and along the electric field. The numbers next to diffraction spots are peak intensities.

Acknowledgments

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