Microstructure and phase evolution of BaHfO₃ pinning centers in YBCO thin films fabricated with the TFA-MOD process

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Exploiting the electrical and magnetic properties of superconducting $YBa_2Cu_3O_{7-\delta}$ (YBCO) coated conductors for the commercial market requires both an economical deposition process as well as a means of immobilizing magnetic flux lines to increase performance of these materials in applied magnetic fields. A promising economical deposition process is the chemical batch process known as TFA-MOD [1]. The immobilization of magnetic flux lines has been demonstrated by the introduction of non-superconducting nanoparticles known as Artificial Pinning Centers (APCs) such as the perovskite phase $BaZrO_3$ (BZO) that can be effectively incorporated into the YBCO microstructure [2]. Finally, combinations of both these techniques have been shown [3,4].

To gain full control over this deposition process, it is desirable to influence the orientation and distribution of these APCs through the variation of external processing parameters. Thus, a comprehensive microstructural analysis of the phase evolution of these perovskite APCs and their dependence on external parameters is needed.

This contribution takes a closer look at the microstructure of films containing the similar perovskite phase $BaHfO_3$ (BHO) at various stages during the reaction process on single crystal $SrTiO_3$ (STO) substrates. BHO was chosen over BZO because the presence of the heavy element Hf in the BHO phase provides a better mass contrast in STEM than BZO as well as exhibiting a clear EDX signal for use in chemical mapping. Moreover, the crystallographic similarity between BZO and BHO means that many of the lessons learned here can be transferred to the BZO system.

Films with varying concentrations of BHO were deposited on STO substrates and quenched at different temperatures during the conversion process. Using the results from x-ray diffraction as a guide, TEM lamellae from samples at critical stages of the nucleation process were prepared using the targeted focused ion beam in-situ lift-out technique.

Results from a fully reacted sample are shown in this abstract. In figure 1, it can be seen that BaHfO₃ particles are distributed evenly throughout the film, including within CuO precipitates that form at the surface. Figure 2 shows a STEM Z-contrast image and numerous EDX maps acquired on an FEI Tecnai 20 TEM (200 kV, LaB₆). Hf-rich regions can be correlated with the perovskite BaHfO₃ phase using HRTEM and this phase grows without a preferred orientation within the YBCO matrix (figure 3). However, figure 3 also reveals that BaHfO₃ particles located at the STO / YBCO interface grow cube-on-cube with YBCO and STO. This is in contrast to APCs introduced using physical deposition processes - in which the secondary phase always grows cube-on-cube with YBCO - and has further consequences on the directional pinning properties of these films.

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- 2. T. Haugan et al., Nature **430** (2004) p. 867-870
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Figure 1. FIB cross section of a YBCO thin film doped with BaHfO₃. The cut was prepared with a Carl Zeiss 1540XB and the image was acquired using the attached in-lens secondary electron detector. Subsequent TEM images indicate that the white particles correspond to the BaHfO₃ perovskite phase. The larger, dark particle on the surface of the YBCO was shown to be CuO. BaHfO₃ particles appear embedded within these particles as well as the YBCO.



Figure 2. STEM Z-contrast image of the YBCO matrix with BHO nano inclusions and EDX maps of the same region. Regions of high Hf content also appear Cu-poor suggesting the BHO phase. BHO particle size appears to decrease near the substrate / YBCO interface.



Figure 3. HRTEM image of the STO / YBCO interface in the YBCO [100] zone axis near the Scherzer focus. Two BHO particles are visible. The BHO particle on the substrate surface grows cube-on-cube with YBCO and STO whereas the particle embedded entirely within the YBCO matrix is rotated through a random angle both in- and out-of-plane.