Structural aspect of high-$J_c$ MOD-YBCO films prepared on large area CeO$_2$-buffered YSZ substrates

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Abstract. YBa$_2$Cu$_3$O$_7$ (YBCO) film was prepared by fluorine-free metalorganic deposition (MOD) on a 2-inch-diameter CeO$_2$-buffered yttria-stabilized zirconia (CbYSZ) substrate. The MOD-YBCO films on a flat CbYSZ prepared using a metal acetylacetonate-based coating solution demonstrated high critical current density ($J_c$), the average being higher than 4 MA/cm$^2$ at 77.3 K and in a self field using an inductive method. The $J_c$ distribution was uniform in the whole area of the MOD-YBCO films on the large size substrates. Cross sectional transmission electron microscopic images of the MOD-YBCO film exhibited a very smooth CeO$_2$/YSZ and CeO$_2$ buffer layer/YBCO film interfaces. Both the buffer and the YBCO layers were dense; however, a small amount of (100)-oriented YBCO occurred inside the matrix of (001)-oriented YBCO layers. Also, some distinct dislocations were observed at the CeO$_2$/YBCO interface. From around the stepped surface of the CeO$_2$ buffer layer there seemed to grow a slightly distorted (001)-oriented YBCO domain, however, the domain restored its (001)-oriented periodicity in the region at some distance above the steps. Such growth mode is considered to correlate the high $J_c$ of the MOD-YBCO film with very small average roughness of CeO$_2$ buffer layer.

1. Introduction

There have been various reports on a metalorganic deposition (MOD) method to prepare high-temperature superconducting (HTS) YBa$_2$Cu$_3$O$_7$ (YBCO) films, since MOD is one of the most attractive techniques for preparation of large-area HTS films at low cost [1]. The method has advantages of precise composition control and potential for manufacturing of large-area HTS films. Cerium oxide (CeO$_2$) is considered to be one of the most promising buffers for preparation of the epitaxial YBCO films. The CeO$_2$ buffer acts as a compensating layer of large lattice mismatch and as a diffusion barrier between substrate and YBCO. We have demonstrated that a complete (001) orientation and very smooth surfaces (observed by x-ray diffraction analysis and atomic force microscopic) are necessary to obtain high superconducting properties for our MOD-YBCO films [2]. We have also reported the epitaxial growth of MOD-YBCO films on CeO$_2$-buffered yttria-stabilized zirconia (CbYSZ), CeO$_2$-buffered sapphire-$r$, LaAlO$_3$ and SrTiO$_3$ substrates [1 and refs. therein]. However, we have never been successful in the direct preparation of YBCO on MgO substrates, though its preparation has been presented using physical deposition processes such as sputtering [3], pulsed laser deposition [4] and so on. It is because the epitaxial growth mechanism of YBCO in the

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chemical solution-related routes as MOD is considered to differ from that of the layer-by-layer type growth in the physical deposition processes.

In this paper, we report an epitaxial growth of the MOD-YBCO film on the CeO$_2$-buffer layer which shows uniform and high critical current density ($J_c$) over the whole area. Then we dedicate to a structural aspect of high-$J_c$ MOD-YBCO large-area film on CbYSZ substrate by high-resolution electron microscopy.

2. Experimental
The (001)-oriented CeO$_2$ buffer layer (40 nm) was deposited on a polished 2-inch-diameter YSZ substrate (K&R creation, Japan). The substrate was heated at 700°C and oxygen gas of a pressure $4 \times 10^{-2}$ Pa was introduced into the chamber to keep the radio frequency oxygen-plasma (13.56 MHz, 20 W) [2]. The fluorine-free MOD procedure for depositing YBCO films using a metal acetylacetonate-based solution has been employed as reported elsewhere [5]. The spin-coated sample was prefired in air at 500°C to remove most of the organic components. The prefired film was annealed for crystallization process of YBCO, in which temperature and oxygen partial pressure were controlled precisely. A conventional x-ray diffraction (XRD) apparatus (MAC Science MXP A with a pole-figure attachment mounted on a horizontal -2 goniometer or MFX-HP equipped with a large size sample holder) was employed to examine the crystalline quality and in-plane orientation for the films. Radiation of CuK$_x$ was used for the XRD measurements. Surface distribution of superconducting $J_c$ for the YBCO films was obtained by mapping analysis of inductive-$J_c$ data (THEVA Cryoscan; probe coil diameter: 5 mm). Thin foils for cross-sectional transmission electron microscopy (XTEM) observation were prepared by slicing, dimpling and ion-thinning. A high-resolution electron microscope (JEOL JEM-4000EX) was used and it was operated at 400 kV for the microstructure observations.

3. Results and discussions
3.1. The epitaxial CeO$_2$ buffer layer deposited on the YSZ substrate showed complete (001) orientation and very smooth and uniform surface morphology. The average roughness of the buffer layer was as small as 0.4 nm. The XRD -2 pattern of the MOD-YBCO (~200 nm) on CbYSZ is demonstrated in Fig.1 in a logarithmic scale. The (001)-oriented YBCO phase is dominant. A small amount of impurity components, i.e., the YBa$_2$Cu$_4$O$_8$ (Y124) and BaCeO$_3$ phases, was observed in the figure. XRD -scanning showed an orientation relationship: YBCO (001) || CeO$_2$ (100) || YSZ (100) and YBCO <110> || CeO$_2$ <100> || YSZ <100>, which was consistent with fast Fourier transform images calculated from XTEM images for the film (not shown here).
3.2. The MOD-YBCO films on the flat CbYSZ exhibited high-$J_c$ values, the average being higher than 4 MA/cm$^2$ at 77.3 K using an inductive method. The $J_c$ values are distributed uniformly in the whole area of the MOD-YBCO film on the large size substrate as shown in Fig. 2, in which the histogram was made from $J_c$ mapping data (49 points: at intervals of 5 mm for x-y directions) except edge area. The small fluctuation in $J_c$ should be attributed to the excellent matching of the crystal structure, lattice parameter and thermal expansion coefficient between the YBCO film, CeO$_2$ buffer layer and YSZ substrate as well as from the smooth and uniform surface morphology of the CeO$_2$ buffer layer as reported in our previous study [6].

3.3. A low-magnification image of the XTEM image for the (001)-oriented MOD-YBCO/ CeO$_2$ buffer/ YSZ substrate is shown in Fig. 3. The image exhibits a smooth YBCO/ CeO$_2$ and CeO$_2$/YSZ interfaces. Both the layers were dense, however, a small amount of the (100)-oriented YBCO, which was not observed in the XRD pattern in Fig. 1, occurred inside the matrix of (001)-oriented YBCO layers. Figure 4(a) shows a high-resolution XTEM image of the CeO$_2$/ YSZ interface. Changes in the contrast at the interface region indicate small distribution of local crystal orientation which accommodates misfit dislocation at the interface. Some distinct steps were observed at the YBCO/ CeO$_2$ interface as shown in a high-resolution XTEM image (surrounded by a dotted rectangle in Fig. 4(b)). From around the stepped surface of the CeO$_2$ buffer layer there seemed to grow a slightly distorted (001)-oriented YBCO domain, however, the domain restored its (001)-oriented periodicity in the region at some distance above the steps. Clear stacking faults are observed in the XTEM image (Fig. 4(b)). The planer stacking faults indicated in the figure may be attributed to an insertion of additional Cu-O plane (remarkable points are indicated by black arrows in the figure),
which results in the Y124 phase as observed in Fig. 1 [7]. Such a growth mode is considered to correlate the high $J_c$ of the MOD-YBCO film with very small average roughness of CeO$_2$-buffer layer. The characteristics in the MOD-YBCO observed using the high-resolution XTEM seem to have no significant difference compared with the physical- and chemical-vapor-deposited ones [7-9]. The preliminary TEM result of our prefired film consists of small randomly-oriented crystallites spread over an amorphous matrix. The small crystallites are considered to recrystallize to form the YBCO phase. At this moment it is impossible to judge whether the epitaxial growth starts just above the CeO$_2$ buffer layer or in the upper region. We should examine the detailed progress of YBCO crystallization to reveal the growth mechanism.

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Figure 4. High-resolution XTEM images of (a) the CeO$_2$ buffer/YSZ substrate and (b) the MOD-YBCO/ CeO$_2$ buffer interfaces. The interfaces are indicated by white arrows.