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In-situ electron beam growth of $YBa_2Cu_3O_{7-x}$ coated conductors on metal substrates

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Abstract

High temperature superconductor $YBa_2Cu_3O_{7-x}$ (YBCO) films have been grown by *in-situ* electron beam evaporation on artificial metal tapes such as ion-beam assisted deposition (IBAD) and rolling assisted biaxially textured substrates (RABiTS). Deposition rate of the YBCO films is 10 ~ 100 Å/sec. X-ray diffraction shows that the films are grown epitaxially but have inter-diffusion phases, like as $BaZrO_3$ or $BaCeO_3$, at their interfaces between YBCO and yttrium-stabilized zirconia (YSZ) or CeO_2, respectively. Secondary ion mass spectroscopy depth profile of the films confirms diffused region between YBCO and the buffer layers, indicating that the growth temperature (850 ~ 900 °C) is high enough to cause diffusion of Zr and Ba. The films on both the substrates show four-fold symmetry of in-plane alignment but their width in the -scan is around 12 ~ 15°. Transmission electron microscopy shows an interesting interface layer of epitaxial CuO between YBCO and YSZ, of which growth origin may be related to liquid fluxes of Ba-Cu-O. Resistivity vs temperature curves of the films on both substrates were measured. Resistivity at room temperature is between 300 and 500 cm, the extrapolated value of resistivity at 0 K is nearly zero, and superconducting transition temperature is 85 ~ 90 K. However, critical current density of the films is very low, ~ 10^3 A/cm^2. Cracking of the grains and high-growth-temperature induced reaction between YBCO and buffer layers are possible reasons for this low critical current density.

Keywords : $YBa_2Cu_3O_{7-x}$, coated conductor, RABiTS, IBAD, *in-situ* e-beam

1. Introduction

The high temperature superconductor (HTS) $YBa_2Cu_3O_{7-x}$ (YBCO) is one of the materials being considered for coated conductors for electrical power applications [1,2]. Deposition of a high temperature superconductor film epitaxially onto a textured buffer layer or textured substrate so as to achieve biaxial texture is the process used to make superconductors of YBCO, requiring of both good c-axis grain orientation and also good in-plane orientation. The classic experiment by Mannhart *et al.* showed that critical current density drops by an order of magnitude for current transfer between two grains with parallel c-axes, but with the a-axis misoriented by only 14° [3,4]. The best Jc values for YBCO are obtained for thin films grown epitaxially on single crystal substrates, such as $SrTiO_3$ or $LaAlO_3$, which are impractical for conductors due to high cost, small size, and poor mechanical properties. Instead, Ni, Ni alloys, and Ag are used as substrate materials in one of two processes. In the first process, a buffer layer, of yttrium-stabilized zirconia (YSZ) for example, is deposited on polycrystalline substrate by ion beam

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assisted deposition (IBAD) [5,6]. The YBCO is then deposited directly on this template or on top of one or more intermediate buffer layers that are grown epitaxially on the YSZ. In the second process, the substrate itself is biaxially textured by thermomechanical processing, so called and rolling assisted biaxially textured substrates (RABiTS) [7], typically a very large rolling reduction followed by heating to obtain the preferred grain orientation needed to form a template for the HTS film and any intermediate buffer layers. A protective coating may be made over the HTS film to protect it from the environment and to facilitate attachment of electrical contacts.

The requirements are challenging: *c*-axis epitaxial with the in-plane alignment grain to grain better than 2 degrees, onto a strong metallic tape kilometers long, and with critical current densities as good as found under ideal conditions on single crystal oxide substrates. We are researching a process called *in-situ* electron beam evaporation. The economical constraints require deposition rates greater than 100 Å/sec and thickness of microns or more, over large areas. These require that the overall system pressure be low to provide large mean free path and thus high rates and permit the use of *in-situ* process control using electrons to probe the surfaces. Thus the use of molecular oxygen at the values needed for thermodynamic stability (10 ~ 30 mTorr) at reasonable growth temperatures is not advisable. The higher activity of atomic oxygen and ozone are being explored along with molecular oxygen at reduced pressure (5 x 10^{-5} Torr). Preliminary observations discussed here indicate that YBCO is stable within a band of atomic oxygen flux and temperature, similar to that published for YBCO in molecular oxygen by Lindemer *et al.* at oxygen pressure many orders of magnitude higher in flux than our attempts with atomic oxygen [8,9]. In this paper, we report growth behaviors of YBCO coated conductor films on IBAD and RABiTS by *in-situ* e-beam.

2. Experiments

Details of e-beam evaporation process were published elsewhere [10]. Several techniques to control composition during deposition are being used [11,12]. Artificial metal substrates have been supplied from 3M (RABiTS), Oak Ridge Natioanl Lab (RABiTS) and Los Alamos National Lab (IBAD). Two different types of IBAD YSZ/Ni tapes are used: with and without CeO_2 top buffer layers Substrate temperature during deposition is 850 ~ 900 °C.

In order to circumvent the pressure-temperature limitations of molecular oxygen, activated species of oxygen, such as atomic oxygen or ozone has been used [10,13]. The active oxygen sources have an activity higher than molecular oxygen and therefore will more readily oxidize the metal atoms. Naturally, these activated species cannot be in thermodynamic equilibrium (otherwise O_2 would form), so it is not clear how relevant a thermodynamic comparison is. It is difficult to establish where the corresponding stability is located for activated oxygen, because it is hard to measure the flux (or pressure) accurately. We are attempting to quantify this for activated oxygen and YBCO.

The process is not yet known. It is perhaps via the secondary electrons (~ 100 eV, where the cross-section for electron impact dissociation of O_2 is largest), or perhaps due to dissociation of the molecule at the hot molten surface of Ba and/or Y, by the electron stimulated dissociation of the metal oxides (DIET, or dissociation induced by electron transition).

Crystal structure and phases are investigated by x-ray diffraction (XRD). In-plane alignment of the films is very important so that -scan of some samples have been conducted. Composition of the films is characterized by inductively coupled plasma emission spectroscopy (ICP). Sticking coefficients of Y, Ba, and Cu are dependent on substrate kinds. Depth profiles of the samples are analyzed by secondary ion mass spectroscopy (SIMS). Cross-sectional transmission electron microscopy (TEM) is

carried out to investigate microstructure of the interfaces. Resistivity vs. temperature measurement was performed using a four-point probe method. Critical current density of the films is obtained using Keithley current source (228A). Wires or pogo pins are used now for contact methods, which should be checked from a point of view that the tape is sufficient to tolerate the mechanical pressure. It is found that this has resulted in cracking in the RABiTS buffer layers.

3. Results and discussion

3.1 X-ray diffraction

Fig. 1(a) shows XRD patterns of a YBCO film on RABiTS with CeO_2-YSZ-CeO_2 texturing layers.

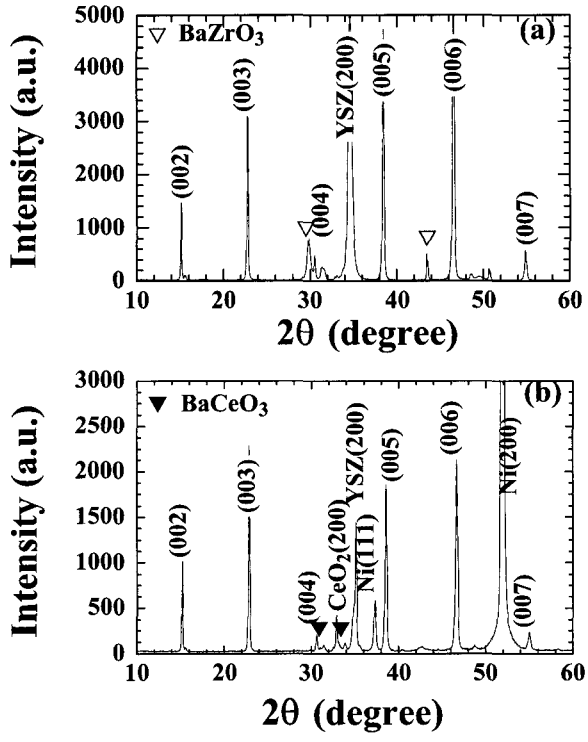

Fig. 1. (a) XRD of a YBCO film on IBAD-YSZ tapes without CeO_2 texturing layers and (b) XRD patterns of YBCO films on RABiTS with CeO_2-YSZ-CeO_2 texturing layers.

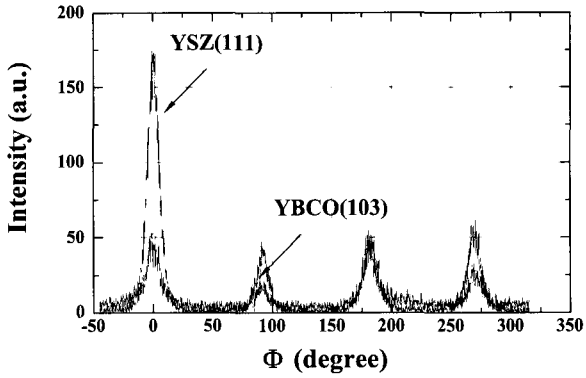

Fig. 2. Φ-scan of a YBCO film on IBAD-YSZ.

YBCO(00*l*) peaks are shown in both the samples. In Fig. 1(b), it is shown that XRD patterns of YBCO Films on IBAD-YSZ tapes without CeO_2 texturing layers. All the YBCO films were grown with c-axis normal to the substrate. There are several secondary phases like as Y_2O_3 and $BaZrO_3$. $BaZrO_3$ phase is identified in IBAD samples that indicate interaction between YBCO and YSZ buffer layer. Figures show typical XRD patterns of a YBCO sample grown at 895 °C with a growth rate of 100 Å/sec. YBCO(00*l*) reflections and secondary phase peaks of $BaZrO_3$ and $BaCeO_3$ are identified in the patterns.

In Fig. 2, -scan of YBCO on IBAD-YSZ is shown. FWHM of IBAD-YSZ films shows 17 ~ 19.7° (Φ_o = 18.65°). The best FWHM of IBAD-YSZ -scan is attained around 10° at 1.2- m thickness of YSZ [14]. FWHM of YBCO films shows 16.6 ~ 17.4° (Φ_o = 16.95°). Even though YBCO shows smaller FWHM values than YSZ, there are some nonzero signals between YBCO peaks. It indicates that most of YBCO grains are well aligned biaxially but there are some randomly oriented grains.

3.2 Depth profile of Secondary Ion Mass Spectroscopy

In Fig. 3, it is shown that SIMS depth profiles of YBCO/RABiTS (run 109) and YBCO/IBAD-YSZ (run 130, run 136) samples grown at high temperature. Cr, Al, Mn, Si, Ti, Nb, and Fe are diffused into the interface and piled up as oxygen from YSZ comes into the metal side. Cr is

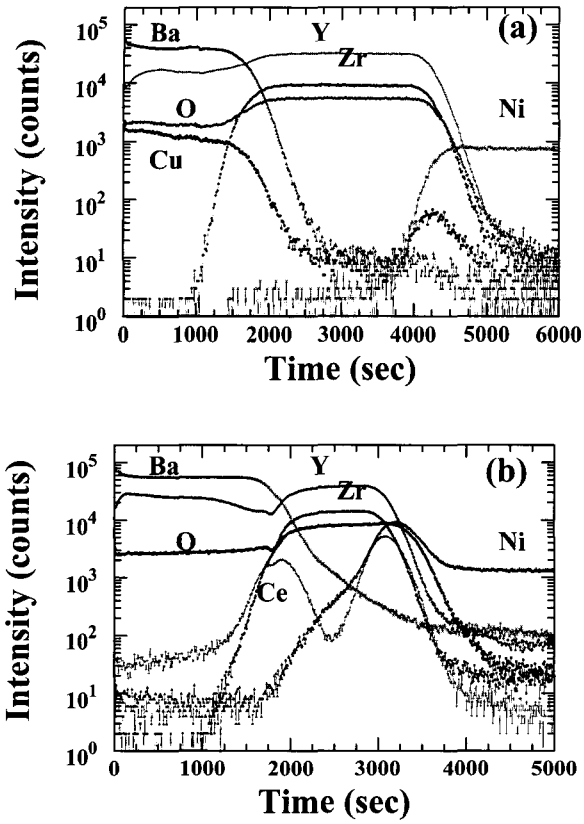

Fig. 3. (a) SIMS depth profiles of YBCO/IBAD and (b) depth profile of YBCO/RABiTS samples grown at high temperature.

dominantly accumulated at the interface, curiously the origin might be oxygen from the YSZ layer. Cr level of SIMS profile increased to $3x10^5$ from $2x10^3$. Note that Ni is extremely stable even though some oxygen diffused into the metal alloy layer. As increasing the baking temperature to 810 °C, an interesting behavior of migration is observed. Cr and Al starts to migrate back to the metal alloy layer whereas oxygen is not much diffused into the metal layer. Thus, some atoms of Cr and Al migrate as a metallic form. At the highest temperature, this migration behavior is revealed clearly for Cr, Al and even Ti. From this heat treatment experiments, it is clear that the atomic distribution near the interface is strongly dependent on process temperature. However, it is not concluded yet that this interface reaction would deteriorate the grown YBCO layers. The result shows that some elements diffuse into the YSZ buffer layers during deposition. CeO_2 layer is very widely diffused throughout the whole samples. From XRD results of the samples, $BaCeO_3$ peaks are found due to reaction of YBCO and CeO_2. Note that run 136 has "perfect" YBCO: this might result in sharper interface.

3.4 Transmission Electron Microscopy

Microstructure of YBCO films is very important to understand their transport properties. Cross-sectional view of transmission electron micrography is very useful to investigate grain growth and defect structures. As mentioned in the previous report on YBCO films on single crystal STO substrates [10], it was found that the films have 'faulted' and 'perfect' grains as well as some Y_2O_3 and CuO secondary phase grains. The YBCO film grown without initial liquid layer at 45 Å/sec shows Jc = 0.47 MA/cm^2 at 77 K and 0 T. Bottom part of the film shows layer-by-layer grown YBCO, *i.e.* 'perfect' grains. Top thin layers are highly faulted YBCO, which turns out to carry most of the supercurrents. It is interesting to note that if we estimate Jc only for the faulted region, it is about 3 MA/cm^2. So it would be good if we find conditions to increase the faulted YBCO thickness. On the other hand, there are two microstructurally distinct defect structures such as Y_2O_3 spheroids and CuO plates in the interface. Oxygen may be transported through Y_2O_3 balls into the layer-by-layer grown YBCO from highly faulted YBCO region. Exact phase of the CuO may be Cu_2O since CuO is usually grown as island form and unstable and Cu_2O is well lattice-matched with YBCO. Amorphous BaCuO is also found on the surface. Fig. 4 shows TEM micrograph of YBCO film on IBAD-YSZ. It is interesting to compare the microstructure of this film with the microstructure of the film on single crystals. The film shows a relatively large portion with a laminar structure and thick CuO structure below that layer. The thickness of CuO layer is about 2000 Å, four times thicker than STO case. However, the YBCO film exhibits only 504 A/cm^2 at 77 K and 0 T.

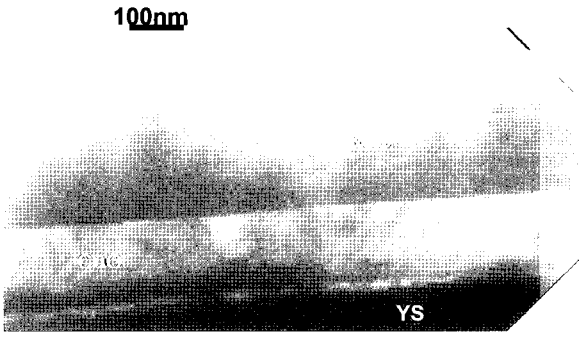

Fig. 4. TEM micrograph of a YBCO film on IBAD.

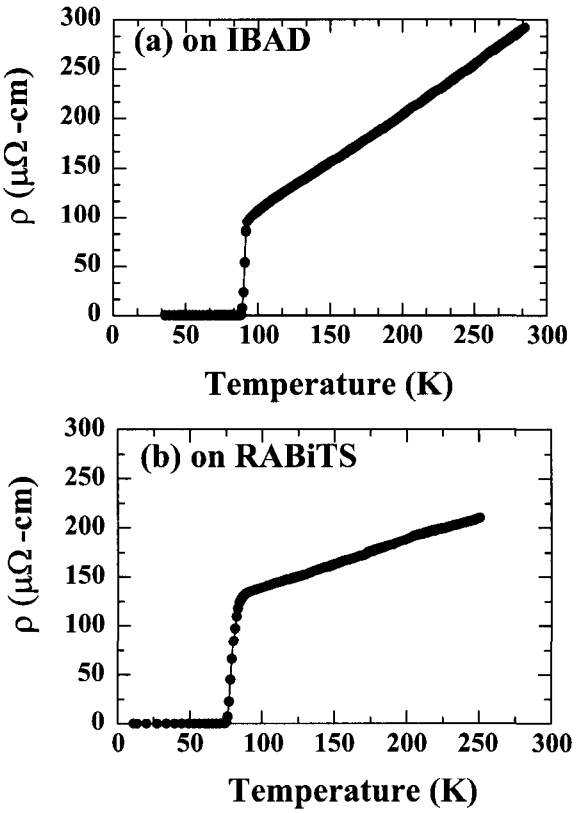

Fig. 5. Resistivity vs.Temperature and XRD results of YBCO films (a) on IBAD and (b) on RABiTS.

3.5 Resistivity vs. Temperature

Resistivity vs temperature curves of the films on both substrates were measured. Fig. 5 shows resistivity of the YBCO films on IBAD and RABiTS. Resistivity at room temperature is between 300 and 250 cm, the extrapolated value of resistivity at 0 K is non-zero, and superconducting transition temperature is 85 ~ 90 K. However, critical current density of the films is very low, ~ 10^3 A/cm^2. Cracking of the grains and high-growth-temperature induced reaction between YBCO and buffer layers are possible reasons for this low critical current density. It is very desirable to find out a robust buffer layer and/or to develop a low temperature processing condition.

4. Summary

YBCO films have been grown by *in-situ* electron beam evaporation on IBAD and RABiTS. The films are grown epitaxially but have $BaZrO_3$ or $BaCeO_3$ at their interface between YBCO and YSZ or CeO_2, respectively. The diffusion is attributed to high growth temperature. The films with four-fold in-plane alignment show broadness in the -scan around 12 ~ 15°. Resistivity vs temperature curves of the films were measured. However, critical current density of the films is quite low due to cracking of the grains and high growth-temperature induced reaction. It is very desirable to find out a robust buffer layer and/or to develop a low temperature processing condition.

Acknowledgements

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