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ABSTRACT

In continuation of our effort to develop single buffer layer architectures for YBCO ($YBa_2Cu_3O_{7-y}$) coated tape conductors, we have studied RE_2O_3 (RE = Y, and rare earths) as candidate materials. Three types of crystal structures including the preferred cubic phase are known for the rare earth oxides. High quality simple cubic RE_2O_3 buffer layers were grown epitaxially on {100}<001> textured Ni substrates using both reactive evaporation and sol-gel processing. Detailed X-ray studies have shown that the Y_2O_3, Eu_2O_3, Gd_2O_3, and Yb_2O_3 were grown with a single epitaxial orientation. SEM micrographs indicated that both e-beam and sol-gel grown films were dense, continuous and crack free. High J_c YBCO films were grown on RE_2O_3-buffered Ni substrates with sputtered cap layers. Two new alternative buffer layer architectures were developed. A high J_c of 1.8 MA/cm^2 at 77 K and self-field was obtained on YBCO films with a layer sequence of YBCO (pulsed laser deposition)/Yb_2O_3 (sputtered)/Y_2O_3 (e-beam)/Ni. Also, a high J_c of over 1 MA/cm^2 at 77 K and self-field was obtained on YBCO films with a layer sequence of YBCO (ex-situ BaF_2 process)/CeO_2 (sputtered)/YSZ (sputtered)/RE_2O_3 (sol-gel or e-beam)/Ni. The performance of sol-gel grown buffers approached the quality of e-beam grown buffers.

INTRODUCTION

The development of second generation superconducting. wires based on YBCO ($YBa_2Cu_3O_{7-y}$) materials using the rolling-assisted biaxially textured substrates (RABiTS) approach has attracted several researchers all over the world.[1,2] In this approach, the {100}<001> cube texture in the starting Ni substrates was obtained by plastic deformation followed by recrystallization. High J_c YBCO films were reproducibly grown on textured-

Ni substrates with a standard architecture of YBCO/CeO_2/YSZ/CeO_2/Ni.[3,4] The starting CeO_2 was mostly obtained by either e-beam or pulsed laser deposition (PLD). Formation of cracks in CeO_2 films of dver 200 nm thick on Ni substrates has prevented us from utilizing CeO_2 as a single buffer layer so far.[2,5] Also, CeO_2 films go through a reduction to Ce_2O_3 during YSZ deposition. Even though this reduction can be either prevented or reversed, there is a clear need to develop alternative buffer layer architectures to simplify the wire development process. In addition, high quality buffer layers with small angle grain boundaries are necessary to produce high current YBCO coated conductors. Also, there is a need to develop scalable processing techniques for fabricating long lengths of coated conductors. In this regard, we have chosen e-beam, sputtering, and solution processing techniques to scale up the RABiTS process.

To develop single buffer layer architectures, we have studied RE_2O_3 (RE = Y and rare earths) as candidate materials.[6,7] Three types of crystal structures are known for the rare earth oxides.* They are hexagonal (type A), monoclinic (type B), and cubic (type C). Type A (hexagonal) has a space group of p3m1, and contains one molecule per unit cell. Each trivalent cation is bonded to seven oxygen atoms (four short bonds and three long bonds), while the two types of oxygen atoms are bonded to five and four metal atoms, respectively. Type B (monoclinic) has a space group of C2/m, and contains six molecules per unit cell, and also shows seven-fold cation coordination. The crystal lattice has three different cation sites and five different anion sites which bond to four, five or six metal atoms. Type C (cubic), has a space group of Ia3, with 16 molecules per unit cell. It is derived from the fluorite (CaF_2) structure by doubling the lattice parameter, and by removing one-fourth of the oxygen ions to maintain the charge neutrality between Y^{3+} and O^{2-}. The crystal lattice has two kinds of six fold coordinated yttrium ions. Stability relationships for these three structure types for RE_2O_3 are shown in Figure 1. The A $\leftrightarrow$ B phase boundary line is vertical, independent of temperature and lies between neodymium and samarium. The B $\leftrightarrow$ C phase boundary is fairly well understood and further extension of the line to higher temperatures is prevented by fusion of the rare earth oxides. Thus, Y_2O_3 and the five heaviest rare earth oxides namely Ho_2O_3, Er_2O_3, Tm_2O_3, Yb_2O_3, and Lu_2O_3 exist in only the cubic form at ambient pressures. Also, the B $\leftrightarrow$ C phase boundary is completely reversible for pure Dy_2O_3, Tb_2O_3, and Gd_2O_3, and for Eu_2O_3, and Sm_2O_3 in the presence of water as catalyst. The exact locations of these phase boundaries for RE_2O_3 films are under further investigation. The cubic lattice parameters for RE_2O_3 were reported earlier.[6] The lattice parameters can be tailored by choosing the appropriate RE_2O_3 (with one or more rare earth's) to match those of the substrate or the HTS film. Recently, we reported our initial demonstration of the growth of epitaxial RE_2O_3 buffer layers on textured Ni substrates using both reactive evaporation and sol-gel alkoxide routes.[6,7,9,10] In this paper, we report our successful demonstration of the growth of high current YBCO films on RE_2O_3-buffered Ni substrates with two new alternative buffer layer architectures.

EXPERIMENTAL PROCEDURE

An e-beam evaporation technique was used to deposit RE_2O_3 films directly on Ni. Using thermodynamic considerations for the formation of metal oxides, we employed both reducing atmospheres (mixture of 4% H_2 and 96% Ar) and water vapor to oxidize the film *in situ* to form stoichiometric RE_2O_3. The details of film growth were reported earlier.[6] The Y_2O_3, Gd_2O_3, and Yb_2O_3 films were grown on textured Ni substrates at temperatures around 650 "C. Similarly, Gd_2O_3, Yb_2O_3, and Eu_2O_3 films were. grown on textured Ni substrates using sol-gel alkoxide precursors. The sol-gel chemistry and details of the solution preparations will be reported elsewhere.'." The coating solution was typically

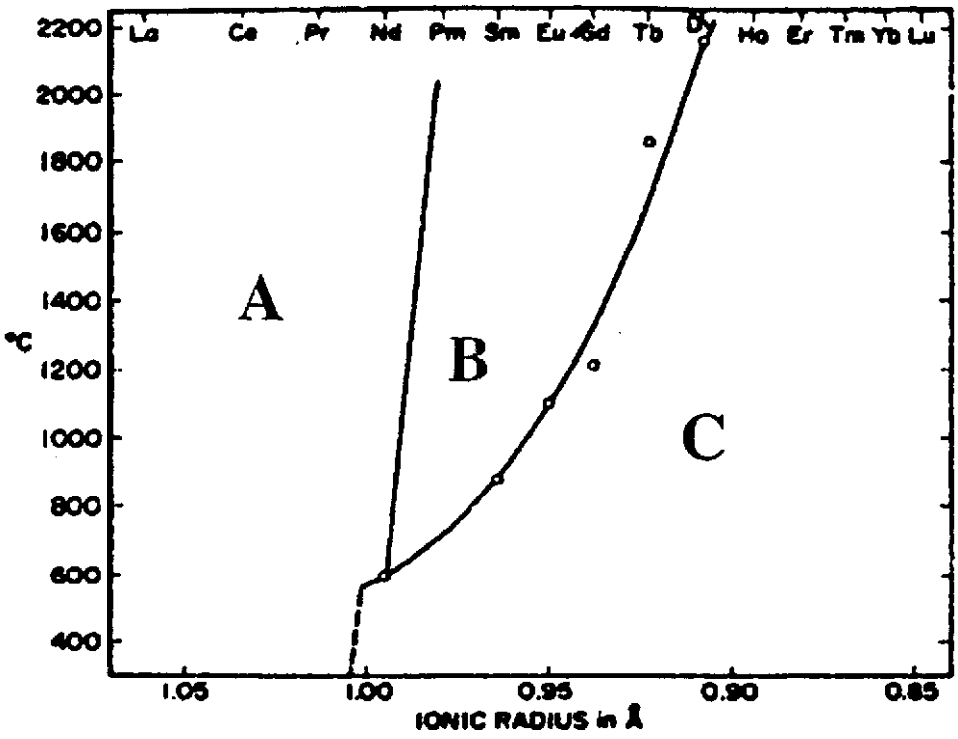

Figure 1. Structural stability relationships for RE_2O_3 (adapted from reference 8). The symbol A represents the hexagonal structure, B represents the monoclinic structure. and C represents the cubic structure.

0.25–0.5 M rare earth methoxyethoxide in 2-methoxyethanol. The coating was done either by spin coating at 2000 rpm for 45 sec or by linear dip-coating in which the Ni strip was immersed in the precursor solution and then withdrawn at a rate of 3 cm/min. After coating, the Ni substrates were annealed in a mixture of 4% H_2 and 96% Ar (Forming gas) at temperatures ranging from 1050-I 150 °C for 1 h and quenched to room temperature.

The rf magnetron sputtering technique was used to grow Yb_2O_3 on e-beam grown Y_2O_3-buffered Ni substrates at 780 °C.[7] Similarly YSZ and CeO_2 cap layers were grown on RE_2O_3-buffered Ni substrates at 780 °C using rf magnetron sputtering. The plasma power was 7.5 W at 13.56 MHz. The resulting Yb_2O_3, YSZ, and CeO_2 films were smooth and dense. Pulsed laser deposition was used to grow YBCO at 780 °C and 200 mTorr O_2 on sputtered Yb_2O_3 layers. Precursor YBCO films were grown on CeO_2-buffered YSZ/RE_2O_3 (sol-gel/e-beam)/Ni substrates by electron beam co-evaporation of Y, BaF_2, and Cu at a combined deposition rate of ~ 6 Å/sec. Details of the experimental conditions were reported earlier.[4,11]

The films were analyzed by X-ray diffraction. A Philips model XRG3100 diffractometer with Cu K_α radiation was used to record powder diffraction patterns. A Picker four-circle diffractometer was used to determine the texture of the films. The out-of-plane alignment (omega scans) was measured by scanning the (100) planes of the film. The in-plane alignment (phi scans) was determined by measuring the (Ill) planes of the film. The RE_2O_3 (222) pole figure was also collected to determine whether the film had a single cube texture. SEM micrographs were taken using a Hitachi S-4100 field emission scanning electron microscope. The beam voltage used was 10 kV. The thickness of the films was determined by both Rutherford backscattering spectroscopy (RBS) and Alpha Step profilometer scans. Electron Backscatter Kikuchi Diffraction (EBKD) patterns were obtained on RE_2O_3 layers and the experimental details were published earlier.[6,12] The resistivity and transport critical current density, J_c was measured using a standard four-probe technique. Values of J_c were calculated using a 1-μV/cm criterion.

RESULTS AND DISCUSSION

Using reactive evaporation, high quality Y_2O_3, Gd_2O_3, and Yb_2O_3 films were grown with a single cube texture directly on {100}<001> textured-Ni substrates.[6] Similarly, using a non-vacuum process, high quality Gd_2O_3, Yb_2O_3, and Eu_2O_3 films were grown with a single cube texture directly on {100}<001> textured-Ni substrates.',[10] Detailed microstructure studies indicate that these RE_2O_3 buffers were dense, continuous and crack-free. A superconducting YBCO film 300 nm thick deposited onto the Yb_2O_3 (sputtered)/Y_2O_3 (e-beam)/Ni by PLD had a J_c of 1.8 MA/cm at 77 K and self field.[6,7] This demonstration of high-current YBCO films on these RE_2O_3-buffered Ni substrates indicates that RE_2O_3 buffers are an excellent diffusion barrier for Ni.

In addition, high J,, 300 nm thick YBCO films were also grown onto the CeO_2 (sputtered)/YSZ (sputtered)/Yb_2O_3 (e-beam) or Eu_2O_3 (dip-coated)/Ni by the ex-situ BaF_2 precursor process. The θ-2θ scans of these YBCO films indicated the presence of a c-axis aligned film. A small impurity peak at ~ 28.7". which may be indexed as belonging to $BaCeO_3$, indicates that there is some interaction between the CeO_2 layer and YBCO. Typically for high J_c YBCO films, this $BaCeO_3$ impurity is almost suppressed. A small amount of NiO impurity was also observed in the film. As shown in Figure 2, the XRD results from ω and ϕ scans on YBCO/CeO_2/YSZ/Yb_2O_3 (e-beam)/Ni revealed good epitaxial texturing. The full width at half-maximum (FWHM) values for Ni (002), YSZ (002), CeO_2 (002) and YBCO (006) are 7.8°, 6.8". 8.0° and 6.7°, and those of Ni (11 1), YSZ (111), CeO_2 (111) and YBCO (103) are 8.6°, 8.5°, 8.9" and 9.7°, respectively. Similarly, ω and ϕ scans on YBCO/CeO_2/YSZ/Eu_2O_3 (dip-coated)/Ni are shown in Figure 3. The FWHM values for Ni (002). Eu_2O_3 (004), YSZ (002) and YBCO (006) are 7.8°, 8.0°, 7.9° and 6.9°, and those of Ni (11 1), Eu_2O_3 (222), YSZ (111) and YBCO (103) are 10.3°, 10.8°, 10.9' and 10.5". respectively. Detailed pole figure analysis indicated the presence of a single cube texture for Yb_2O_3 (e-beam) and Eu_2O_3 (sol-gel) layers. The BBS spectra for 300 nm thick YBCO films on CeO_2/YSZ/Eu_2O_3 (dip-coated)/Ni are shown in Figure 4. The simulation indicates that the film thickness for Eu_2O_3, YSZ, CeO_2 and YBCO are 60 nm, 295 nm, 10 nm and 300 nm, respectively.

The room-temperature resistivity of the 300 nm thick YBCO film on CeO_2/YSZ/Yb_2O_3 (e-beam)/Ni was around 260 microOhm·cm and the T_c measured was about 90 K. Extrapolating values of the resistivity (between 300 K and above T_c) to below zero indicates the presence of predominantly cl YBCO film. The field dependence of J_c for the same film is shown in Figure 5. As per Figure 5, the zero field J_c measured was around 1 MA/cm^2 at 77 K. The J_c at 0.5 T was about 25 % of the zero field J_c. This indicates the presence of a strongly linked YBCO film. The irreversibility field is also above 7 T. Similarly, the room temperature resistivity of the 300 nm thick YBCO film on CeO_2/YSZ/Eu_2O_3 (sol-gel)/Ni was low and the T_c measured was about 90 K. The field dependence of J_c for the same film is shown in Figure 6. The zero field I_c measured was 16 A which translates to a J_c of I.1 MA/cm^2. The J_c at 0.5 T is about 20 % of the zero field J_c. Also, a reproducible J_c of over 0.4 MA/cm^2 at 77 K and self field was obtained on 300 nm thick YBCO films on CeO_2 (sputtered)/YSZ (sputtered)/Gd_2O_3 or Yb_2O_3 (sol-gel)/Ni.[9,10] From Figures 5 and 6, we can conclude that RE_2O_3 buffers with CeO_2 and YSZ sputtered caps provide a good template for further growth of high J_c YBCO films through the ex-situ BaF_2 precursor process. Furthermore, we note that the performance of the RE_2O_3 sol-gel buffers approached that of the e-beam RE_2O_3 buffers. Efforts are being made to grow YBCO films directly on RE_2O_3-buffered Ni substrates. SEM micrograph for 300 nm thick YBCO film on CeO_2 (sputtered)/YSZ (sputtered)/Eu_2O_3 (dip-coated)/Ni substrate is shown

in Figure 7. The microstructure of the YBCO film looks porous but with plate-like morphology similar to those observed previously for epitaxial Tl-1223 films.

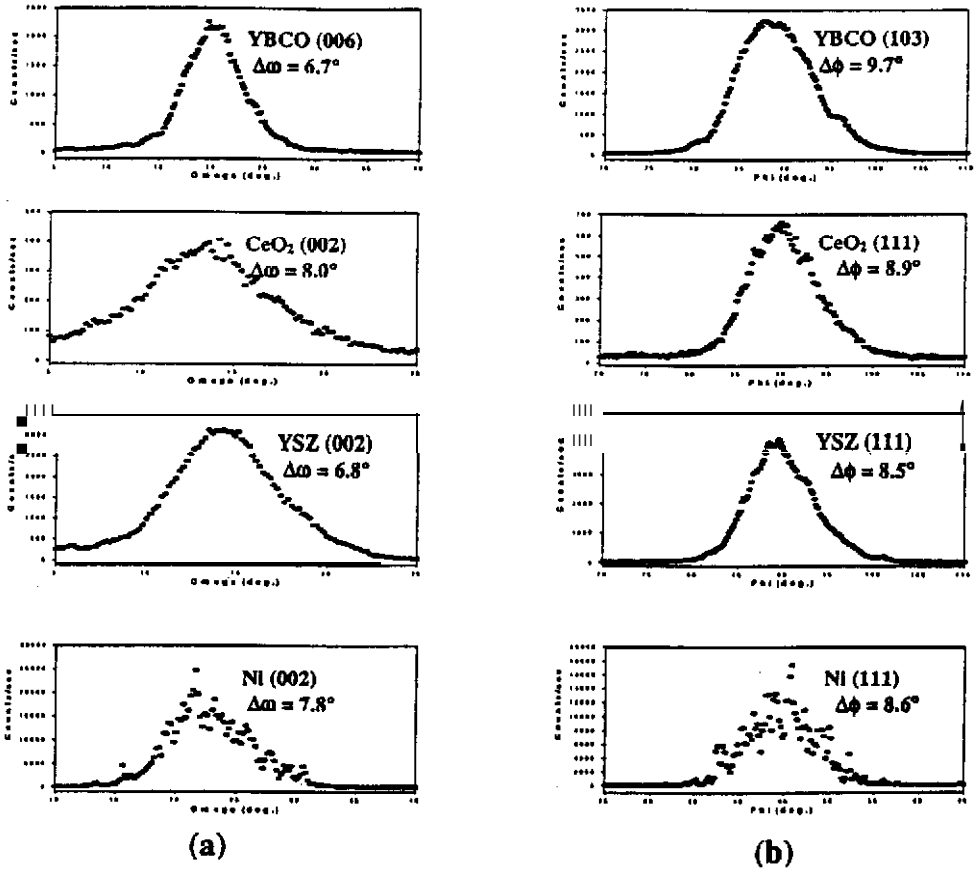

Figure 2. **The** ω and φ **scans** for a 300 **nm thick** YBCO **film** on **CeO_2 (sputtered)/YSZ (sputtered)/Yb_2O_3 (e-beam)/Ni** substrates.

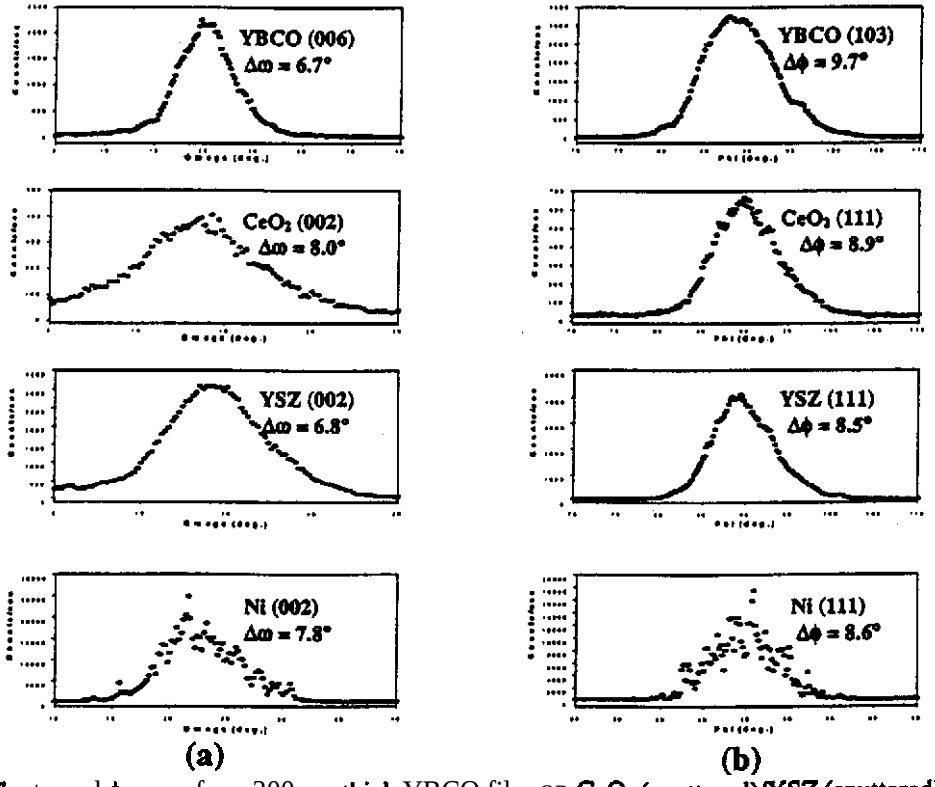

Figure 3. **The** to and φ scans for **a** 300 **nm thick** YBCO film OR **CeO_2 (sputtered)/YSZ (sputtered)/Eu_2O_3 (dip-coated)/Ni** substrates.

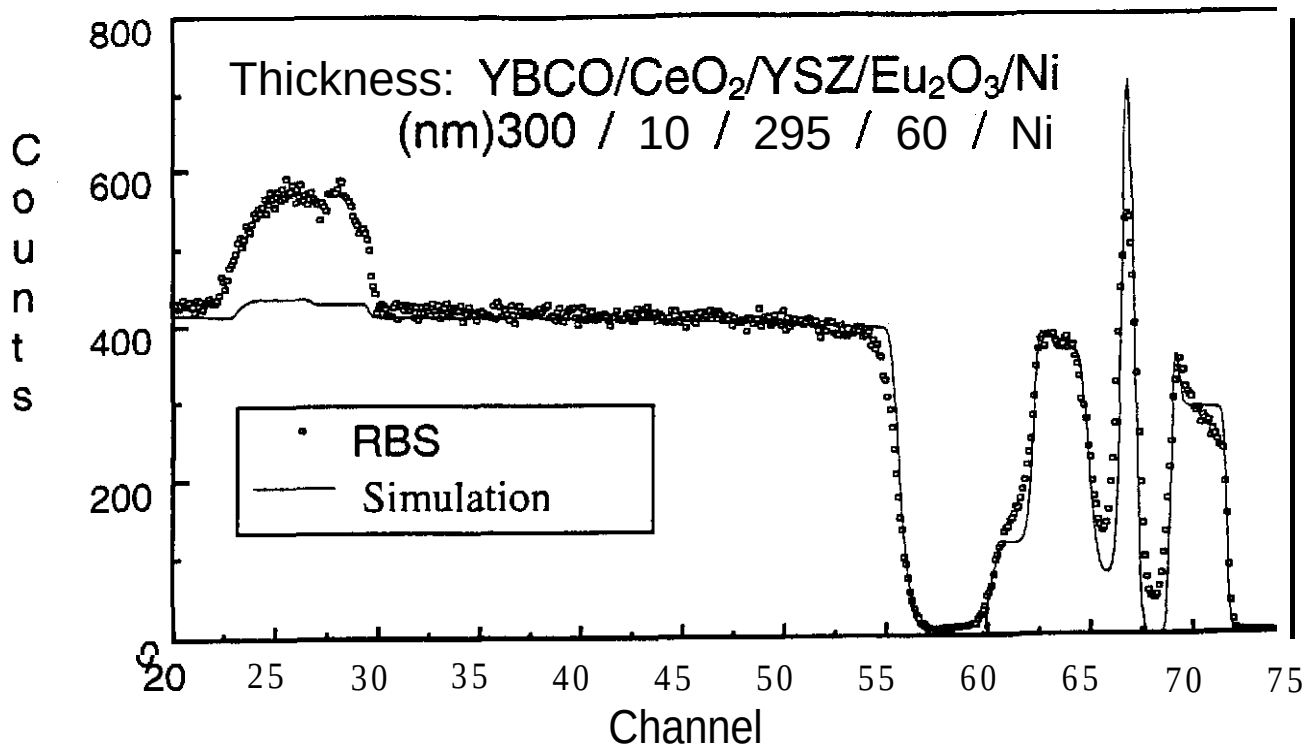

Figure 4. Rutherford backscattering spectra of 300 nm thick YBCO film on CeO_2 (sputtered)/YSZ (sputtered)/Eu_2O_3 (dip-coated)/Ni substrates. The spectra were measured with 5.0 MeV He^+ ions at near-normal incidence, detected at 160" scattering angle.

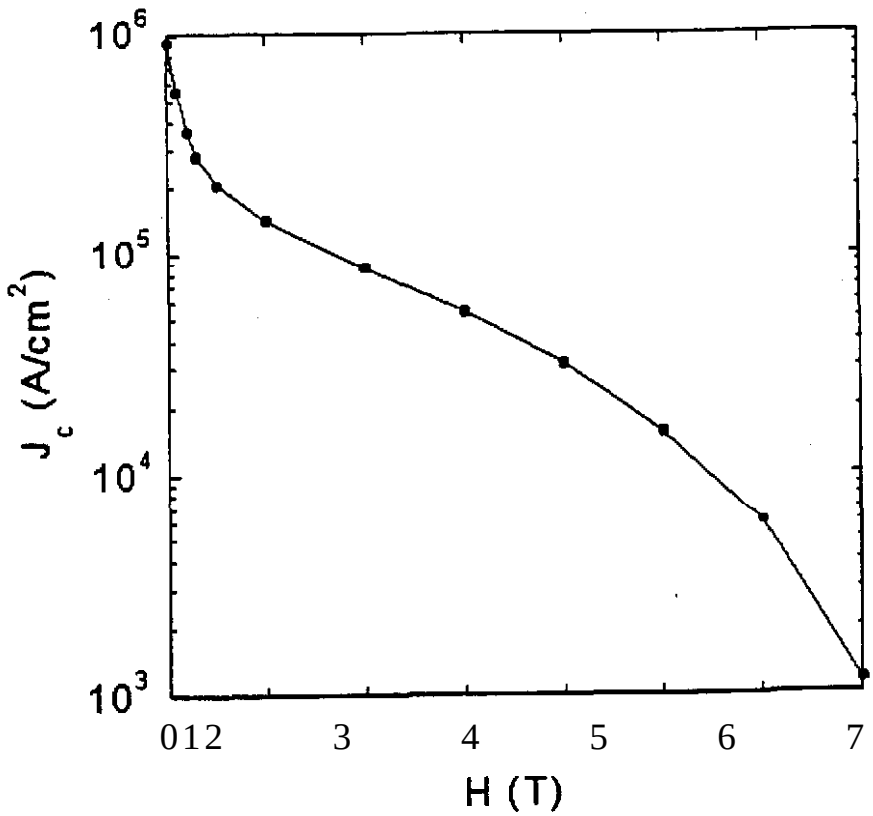

Figure 5. The field dependence of critical current density, J_c for 300 am thick YBCO film on CeO_2 (sputtered)/YSZ(sputtered)/Yb_2O_3(e-beam)/Ni substrates.

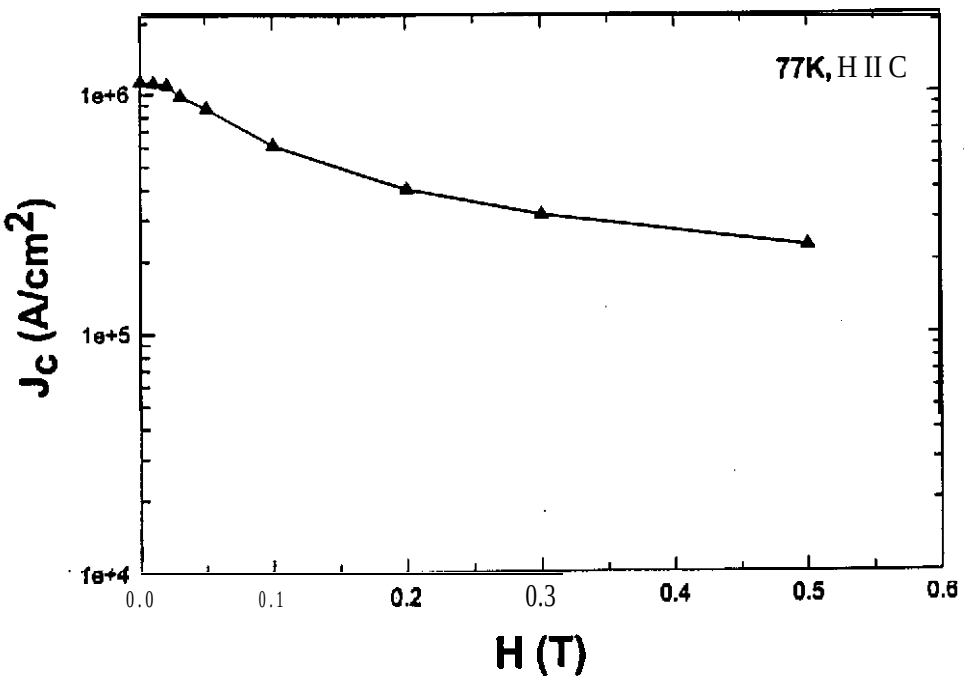

Figure 6. The field dependence of critical current density, J_c for 300 nm thick YBCO film On CeO_2 (sputtered)/YSZ(sputtered)/Eu_2O_3(dip-coated)/Ni substrates.

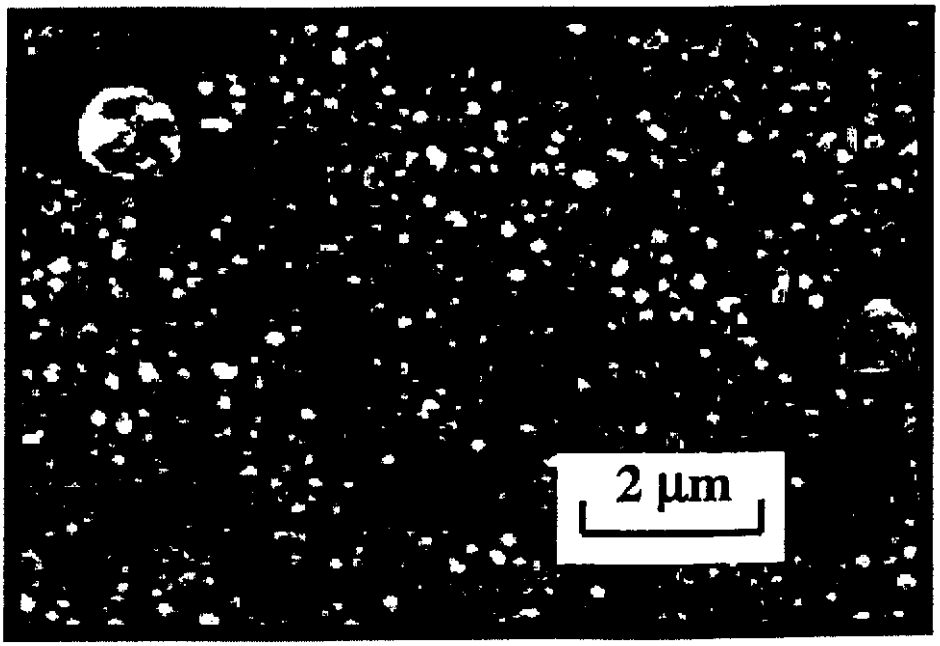

Figure 7. SEM micrograph for 300 nm thick YBCO film on CeO_2 (sputtered)/YSZ (sputtered)/Eu_2O_3 (dip-coated)/Ni substrates.

CONCLUSIONS

We have demonstrated that several high quality RE_2O_3 films can be grown epitaxially with a single cube-on-cube orientation on {100}<001> textured Ni substrates using both reactive evaporation and sol-gel processing. The microstructures of RE_2O_3 buffers grown by both techniques were dense, continuous and crack-free. The performance of sol-gel grown buffers approached the quality of e-beam grown buffers. High J_c YBCO films were grown on RE_2O_3 templates with two new buffer layer architectures. A high J_c of 1.8 MA/cm^2 at 77 K and self-field was obtained on this $YBCO/Yb_2O_3/Y_2O_3/Ni$ architecture. Also, a high J_c of over 1 MA/cm^2 at 77 K and self-field was obtained on this $YBCO/CeO_2/YSZ/RE_2O_3/Ni$ architecture.

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