Fabrication of Long Lengths of YBCO Coated Conductors using a Continuous Reel-to-Reel Dip-Coating Unit

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Abstract—A low-cost, non-vacuum, solution precursor route has been developed to produce epitaxial Gd_2O_3 and Eu_2O_3 buffer layers and $YBa_2Cu_3O_{7-\delta}$ (YBCO) superconductors on biaxially textured metal substrates. On sol-gel Eu_2O_3 seed layers with sputtered YSZ and CeO_2 top layers, YBCO film with a J_c of over 1 MA/cm^2 at 77 K was obtained. On all solution buffer layers (CeO_2/Eu_2O_3/Ni), YBCO film with a J_c of 200,000 A/cm^2 at 77 K was grown using pulsed laser deposition (PLD). Meter lengths of epitaxial and crack-free Gd_2O_3 buffer layers were fabricated on cube textured Ni-W (3 at. %) substrates for the first time. High quality YBCO films were deposited on Rolling-Assisted Biaxially Textured Substrates (RABiTS) using a trifluoroacetate (TFA) precursor approach. The precursors were either spin-coated or dip-coated and decomposed in a newly developed fast 3-hour burn-out step followed by post-annealing. In a stationary burn-out route, we have produced 40 cm long crack-free YBCO TFA precursors on RABiTS. On short segments, YBCO films with a J_c of over 500,000 A/cm^2 at 77 K were grown on all PLD buffered-Ni substrates (CeO_2/YSZ/CeO_2/Ni).

Index Terms— Solution precursors, RE_2O_3 buffer layers, YBCO coated conductors, textured metal substrates

I. Introduction

For the promise of high-temperature superconductivity to be realized, practical processes are needed to manufacture useful lengths of high-temperature superconducting (HTS) wires. The goal is to produce robust, economical, and scaleable processes for making wire that can carry sufficient current at liquid nitrogen temperatures. The second-generation YBCO coated conductors are the potential candidates. Also, YBCO coated conductors have good superconducting properties in significant magnetic fields at the liquid nitrogen temperatures. Three methods have been developed for producing long lengths of YBCO coated conductors.

The first method is the "Ion Beam Assisted Deposition (IBAD), which was developed by Iijima et al. [1] and later significantly improved at Los Alamos [2]. In the IBAD technique, YSZ (Yttria-stabilized zirconia) or MgO buffer templates with a biaxial cube texture were produced on polycrystalline nickel alloy substrates using a well-collimated secondary gun directed towards the substrate at a certain angle to sputter off non-favorable orientations during the buffer layer deposition. This is followed by Pulsed Laser Deposition (PLD) to grow high quality YBCO layers. The second method is the Rolling-Assisted Biaxially Textured Substrates (RABiTS) process, developed at Oak Ridge [3,4]. In this process the biaxial texture templates were produced in the starting nickel or nickel alloy substrates using mechanical deformation of the metal rods over 95 % followed by annealing. The buffer layers and YBCO superconductors were then grown epitaxially on biaxially textured nickel tapes or sheets. The third method is the Inclined Substrate Deposition (ISD), which was developed in Japan [5], and further pursued in Europe and United States. In this approach, the biaxially textured YSZ or MgO buffer layer templates were produced by keeping the starting polycrystalline nickel alloy substrates at certain angle with respect to the on-axis flux of the buffers. The YBCO layers were then deposited by PLD.

Chemical solution epitaxy has emerged as a viable, non-vacuum process for fabricating epitaxial buffer layers and YBCO superconductors [6-12]. The roll-textured nickel substrates are the ideal starting templates for this process. The major advantages of solution processes are: The coating of metal-organic precursor solution is done at room temperature, followed by annealing at high temperatures to obtain highly crystalline phases. The control of precursor stoichiometry or doping is both easy and precise. This is completely a non-vacuum, high volume deposition process. The material utilization is almost 100 % and the substrates could be coated on both sides. In order to scale-up the solution process, a reel-to-reel dip-coating process was developed. The dip-coated precursors could be post-annealed in a batch or in a continuous process. We have recently developed an all alkoxide route to produce epitaxial oxide buffer layers on roll-textured nickel substrates [8-11].

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In this paper, we report our successful demonstration of the growth of epitaxial Gd_2O_3 seed layers on biaxially textured substrates with reduced magnetism in meter lengths. In addition, we will also report our recently developed fast burn-out step for YBCO TFA precursors.

II. Experimental

A. Preparation of Ni or Ni-alloy tapes

Both Ni substrates and Ni-W (3 at. %) (stronger and substrates with reduced magnetism) were chosen for this study. Biaxially textured Ni (100) or Ni-W (3 at. %) substrates were obtained by mechanically deforming the metal rods over 95 % followed by annealing at 1000-1200 °C for 1 h in the presence of Ar/H_2 (4%) gas. Meter lengths of 50 μm thick and 1 cm wide as-rolled metal substrates were cleaned by sonification in isopropanol in a reel-to-reel substrate cleaning unit. The cleaned substrates were then annealed in Ar/H_2 (4%) to get the required cube texture in a reel-to-reel annealing furnace at the optimum temperatures and time.

B. Coating and heat-treatment

A 2-methoxyethanol solution of europium methoxyethoxide/acetate or gadolinium methoxyethoxide/acetate precursors was used for the spin-coating/dip-coating process of buffer layers. The solution preparations were reported earlier [8-11]. The typical concentrations of the solutions were 0.25-0.5 M. In the reel-to-reel dip-coating unit, both ends of the annealed Ni or Ni alloy tapes were electrically spot-welded to Ni leaders mounted on two reels. The take-up reel was driven continuously by a stepper motor and the pay-out reel was tensioned by a variable torque motor. The textured tapes from the pay-out reel were withdrawn from the coating bath at a constant speed of 3-30 m/h. The tapes were coated on both sides. The coated tapes were then annealed in a pre-heated furnace. The furnace was kept at 1050-1150 °C with the constant purging of Ar/H_2 (4%) gas. The tapes were annealed for a residence time of 30-60 minutes. For all solution buffers, cerium methoxyethoxide precursors were used for the CeO_2 cap layer.

For the YBCO growth, Y, Ba, and Cu trifluoroacetate (TFA) precursor solutions were prepared. The concentration of the solution was typically 2 M total cations. The YBCO precursor solutions were spin-coated at 2000 rpm for 120 sec or dip-coated at a constant speed of 20-30 m/h. The coated tapes were heat-treated in two stages. The first stage is the burn-out step. It is very important to obtain crack-free, feature-less brown colored precursors at this stage. This step takes nearly 10 hours as reported in the literature [7,13]. We have developed a fast 3-hour burn-out step to obtain crack-free YBCO precursors. The heat-treatment step for the first burn-out step is shown in Figure 1. The first burn-out stage resulted in amorphous Y_2O_3, Cu_2O/CuO and crystalline BaF_2 phases. The second stage is the post-annealing process where the YBCO precursors are annealed in a controlled atmosphere furnace equipped with $wet/O_2/Ar$ mixtures. The YBCO conversion is typically carried out at 735 °C with a pO_2 of 150-180 ppm and H_2O dew point of 40 °C. The linear gas flow conditions were used. The process time varies from 1- 3 hours depending on the film thickness and width of the substrate.

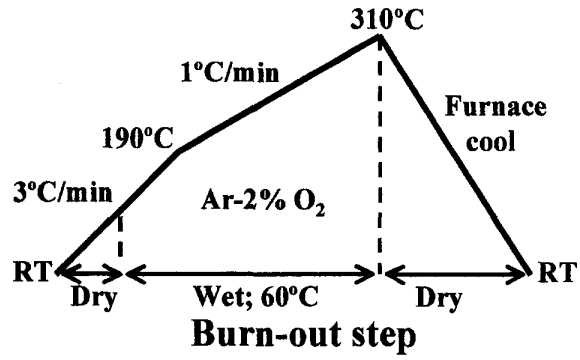

Fig.1. The heat-treatment step for the first burn-out step of YBCO TFA precursors

C. Characterization

The buffer layers and YBCO superconductors were characterized by x-ray diffraction (XRD), scanning electron microscopy (SEM), and electron backscatter Kikuchi diffraction (EBKD). The thickness of both buffers and YBCO films were determined by both Rutherford backscattering spectroscopy (RBS) and Alpha Step profilometer scans. The carbon concentrations in solution buffers have been determined using proton resonance (E_H = 1.73 MeV) from RBS. The resistivity and transport critical current density, J_c was measured using a standard four-probe technique. Ag contacts were made by sputtering. After Ag contacts were made, YBCO films were annealed in O_2 at 500 °C for 30 min and slow cooled to 350 °C and further furnace cooled to room temperature in pure O_2. The voltage contact spacing was 0.4 cm. Values of J_c were calculated using a 1-μV/cm criterion.

D. Preparation of YBCO test structures

On short samples of dip-coated Eu_2O_3, YSZ and CeO_2 top layers were deposited by rf magnetron sputtering at 780 °C in 10 mTorr of Ar/H_2 (4%) gas. The plasma power was 75 W at 13.56 MHz. Ex-situ YBCO BaF_2 precursors were deposited on CeO_2-buffered YSZ (sputtered)/Eu_2O_3 (dip-coated)/Ni substrates using electron beam co-evaporation of Y, BaF_2, and Cu. Details of the experimental conditions were reported earlier [14]. The CeO_2-buffered YSZ (sputtered)/Eu_2O_3 (dip-coated)/Ni substrates were annealed under N_2 at 650 °C for 1 h prior to the YBCO precursor deposition. The YBCO films on all solution buffers (CeO_2 (1-coat)/Eu_2O_3 (4-coats)/Ni) were grown using pulsed laser deposition at 780 °C and pO_2 of 185 mTorr.

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III. RESULTS AND DISCUSSION

A. Eu_2O_3 buffer layers

XRD results indicated the presence of epitaxial and crack-free 50 nm thick Eu_2O_3 buffer layers on {100}<001> Ni substrates. Longer annealing times during processing eliminated the presence of carbon in dip-coated buffers. To reproducibly grow YSZ epitaxially on sol-gel seed layers, the carbon has to be removed completely from the solution layers. On Eu_2O_3 seed layers with sputtered YSZ and CeO_2 top layers, YBCO film with a J_c of over 1 MA/cm^2 was obtained. The field dependence of J_c for the same film is shown in Figure 2. The J_c at 0.5 T is 0.23 MA/cm^2. This proves that Eu_2O_3 seed layers provide a good template to fabricate high current density YBCO films. On all solution buffer layers (CeO_2; 1-coat/Eu_2O_3; 4-coats), YBCO film with a J_c of 200,000 A/cm^2 was obtained. The field dependence of J_c for the same film is shown in Figure 3. The total buffer layer thickness is 220 nm. The YBCO films on all solution buffer layers had a porous microstructure. In addition, NiO impurities were also present. To further improve the current density, it is necessary to grow dense and thicker buffers.

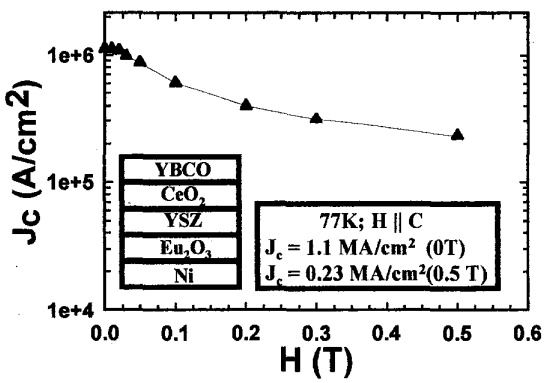

Fig.2. The field dependence of critical current density, J_c for 300 nm thick YBCO film (Ex-situ) BaF_2 process) on CeO_2 (sputtered) (10 nm)/ YSZ (sputtered) (295 nm)/Eu_2O_3 (dip-coated) (60 nm)/Ni substrates

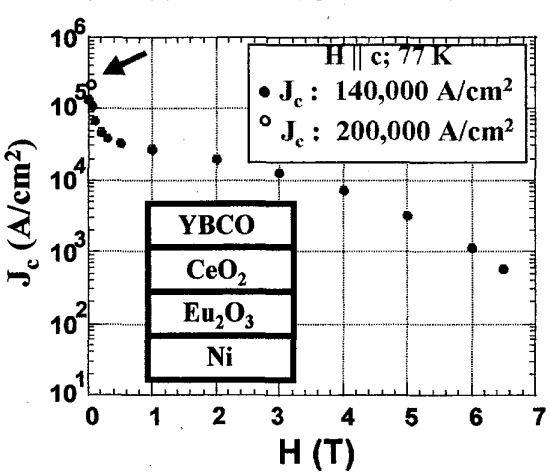

Fig.3. The field dependence of critical current density, J_c for 200 nm thick PLD grown YBCO film on CeO_2 (solution)(20 nm)/Eu_2O_3 (solution) (200 nm)/Ni substrates

B. Gd_2O_3 buffer layers on Ni-W (3 at. %) substrates

The annealed Ni-W (3 at. %) substrates are much stronger and have reduced magnetism than pure Ni substrates. The yield strength (YS) at 0.2 % is 150-200 MPa. This is comparable to 164 MPa YS for Ni-Cr (13 at. %) substrates. Detailed XRD studies indicate that Gd_2O_3 layers can be grown directly on Ni-W (3 at. %) substrates without any surface modifications. The full width at half maximum (FWHM) values for Ni (002) and Gd_2O_3 (004) are 5.0°, and 8.9°, and those of Ni (111) and Gd_2O_3 (222) are 7.3° and 9.0°, respectively. The four-fold symmetry in Gd_2O_3 (222) pole figure indicated the presence of a single cube textured buffer. Efforts are being made to grow superconductors on these layers. On all vacuum buffered-Ni-W (3 at. %) substrates (YBCO/CeO_2/YSZ/Y_2O_3/Ni-W (3 at. %)), YBCO films with a J_c of 1.9 MA/cm^2 at 77 K were obtained. Meter lengths of Gd_2O_3 seed layers on roll-textured Ni-W (3 at. %) substrates were produced. The texture along the one-meter tape is shown in Figure 4. The texture is uniform on most of the lengths. As can be seen from Figure 5, SEM picture indicates the presence of a continuous and crack-free Gd_2O_3 buffer layer.

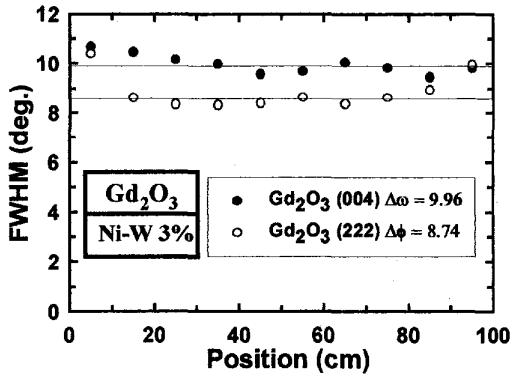

Fig.4. The FWHM values of ω and ϕ scans for sol-gel Gd_2O_3 seed layers deposited on textured Ni-W 3% substrates are plotted along the length of the dip-coated substrate

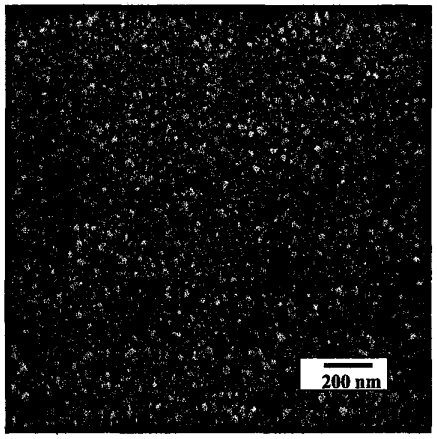

Fig.5. SEM micrograph for a 20 nm thick dip-coated Gd_2O_3 seed layer on biaxially textured Ni-W 3% (100) substrates

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C. YBCO TFA precursors

In a stationary burn-out step, 40 cm long crack-free YBCO TFA precursors on RABiTS were produced. In a reel-to-reel continuous burn-out step, 10 cm long crack-free YBCO precursors on RABiTS were produced. On short samples, YBCO films on $LaAlO_3$ (100) single crystal substrates had a J_c of over 2 MA/cm^2 at 77 K. However, the YBCO films on PLD RABiTS had a J_c of 500,000 A/cm^2 at 77 K. These results were obtained using fast burn-out precursor step. The film thickness is 0.3 μm. The field dependence J_c for these films are shown in Figure 6. In the burn-out step, the heat-treatment of YBCO precursors is rate limiting due to the high volatility of Cu TFA precursors. In order to carry out much faster burn-out step, other Cu containing precursor chemistry has to be developed.

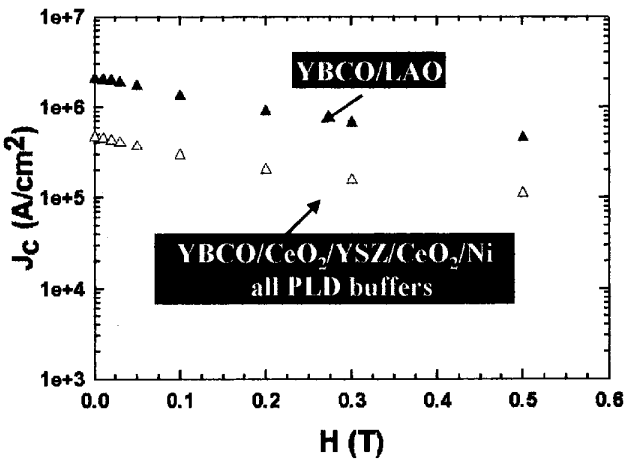

Fig.6. The field dependence of critical current density, J_c at 77 K and self-field for 0.3 μm thick YBCO film on both $LaAlO_3$ (100) single crystals and all PLD-buffered CeO_2/YSZ/CeO_2/Ni substrates

IV. CONCLUSIONS

The performance of sol-gel templates is approaching as that of vacuum templates. On Eu_2O_3 seed layers with YSZ and CeO_2 sputtered top layers, YBCO film with a J_c of 1.1 MA/cm^2 at 77 K was obtained. On all solution buffer layers, YBCO film with a J_c of 200,000 A/cm^2 at 77 K was obtained. Meter lengths of epitaxial Gd_2O_3 buffer layers were fabricated on roll-textured Ni-W (3 at. %) substrates. Faster burn-out step was developed for YBCO TFA precursors. YBCO films with a J_c of over 2 MA/cm^2 on $LaAlO_3$ substrates and over 500,000 A/cm^2 were obtained on PLD grown RABiTS.

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