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High-J_c YBCO coatings on reel-to-reel dip-coated Gd_2O_3 seed buffer layers epitaxially fabricated on biaxially textured Ni and Ni-(3at%W-1.7at%Fe) alloy tapes

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ABSTRACT

A low-cost, non-vacuum reel-to reel dip-coating system has been used to continuously fabricate epitaxial Gd_2O_3 buffer layers on mechanically strengthened, biaxially textured Ni-(3at.%W-1.7at%Fe), defined as Ni-alloy, metal tapes. X-ray diffraction analysis of the seed Gd_2O_3 layers indicated that well textured films can be obtained at processing temperatures (T_p) between 1100 and 1175°C. Processing speed did not significantly affect the crystalline quality of the Gd_2O_3. Scanning electron microscopy revealed a continuous, dense and crack-free surface morphology for these dip-coated buffers. The Gd_2O_3 layer thickness led to remarkable differences in the growth characteristics of the subsequent YSZ and CeO_2 layers deposited by *rf*-magnetron sputtering. Epitaxial YBCO films grown by pulsed laser deposition on the short prototype CeO_2/YSZ/Gd_2O_3/Ni-(3at%W-1.7at%Fe) conductors yielded self-field critical current densities (J_c) as high as $1.2x10^6$ A/cm^2 at 77 K. Pure Ni tapes were used to asses the viability of dip-coated buffers for long length coated conductor fabrication. The YBCO films, grown on 80 cm long and 1 cm wide CeO_2/YSZ/Gd_2O_3 buffered Ni tapes by the industrially scalable *ex-situ* BaF_2 precursor process, exhibited end-to-end self-field J_c of $6.25x10^5$ A/cm^2 at 77 K.

INTRODUCTION

The recent demonstration of high superconducting performance in conductors based on continuous fabrication of high temperature superconducting (HTS) coatings with high degree of biaxial alignment onto rolling-assisted biaxially textured flexible metal substrates (RABiTS) has been realized as one of the most promising techniques for producing second-generation HTS wires (coated conductors) [1,2]. The RABiTS utilizes the biaxial texture of the Ni and/or Ni-alloy metal substrate as a template to deposit epitaxial buffer layers that provide a barrier to chemical interactions with the substrate, and yield chemically and structurally compatible surfaces for the epitaxial growth of HTS films. To date, various combinations of oxide buffer layers have been tried on RABiTS [1-5]. So far, the most successful and hence the most preferred buffer layer architecture has been the combination of CeO_2 and yttria stabilized zirconia (YSZ), yielding $YBa_2Cu_3O_{7-\delta}$ (YBCO) films with high critical current densities (J_c) exceeding 10^6 A/cm^2 (77 K, 0 Tesla) [1-4]. Fabrication of these buffer layers are generally carried out by vacuum deposition processes such as electron beam evaporation (e-beam), pulsed laser deposition (PLD) or magnetron sputtering [1-4]. On the other hand, for the commercial development of a practical superconducting wire technology, more adaptive and easily scalable processing techniques are highly desired. Non-vacuum chemical-solution based processes, such as sol-gel and metal-organic decomposition (MOD), offer many desirable aspects. These solution-based processes are attractive for their low-cost and mechanical simplicity, as well as the ease of scalability. In addition, since the precursors are mixed at atomic levels, the metal-oxide precursor stoichiometry and composition can be controlled precisely.

Recently, it has been reported that various RE_2O_3 (rare-earth; RE=Gd, Yb, and Eu) and $RE_2Zr_2O_7$ (RE=La and Nd) oxide films can grow epitaxially on textured Ni substrates by using solution-based methods [5,6]. While short segments of sol-gel seeded Ni substrates with sputtered CeO_2/YSZ over layers have yielded YBCO with J_c in the range of 0.4 - $1.1x10^6$ A/cm^2 at 77 K, long lengths of high-J_c YBCO coatings on solution seed buffer layers have not yet been produced. Oxide buffer layers have also been grown directly on Ni-alloy tapes by non-vacuum solution-based methods. However, despite a good crystalline structure, to date there have been no reports on the growth of high-J_c YBCO films on these sol-gel buffered alloy tapes. On the other hand, it is highly desirable to work with textured Ni-alloy substrates, which are much stronger and have reduced magnetism. Indeed, the low mechanical strength and ferromagnetism of pure Ni present significant challenges to long-length manufacturing and hinders its usage in applications where AC losses are an issue.

In the RABiTS process, it is well understood that integrity of a seed buffer layer is extremely important for the epitaxial growth of subsequent oxide buffer and HTS layers [4,7]. In this research, we aimed to develop reproducible and high-quality seed buffer layers on Ni and Ni-alloy tapes via non-vacuum solution-based techniques. This study also provides a baseline for the ultimate development of an all-solution route for development of coated conductors. Here, we first report the epitaxial growth and property characterizations of sol-gel processed, reel-to-reel dip-coated Gd_2O_3 seed buffer layers on mechanically strenghtened textured Ni-alloy substrates. Second, we demonstrate the continuous fabrication of Gd_2O_3 seed layers in long lengths on both Ni-alloy and pure Ni tapes. Third, for the first time, the electrical transport properties, measured over lengths of YBCO films grown using the *ex-situ* BaF_2 precursor process on pure Ni-tapes, are also reported.

EXPERIMENTAL PROCEDURE

Preparation of biaxially textured Ni/Ni-alloy tapes and coating procedure

Biaxially textured Ni (99.99%) and Ni-(3at%W-1.7at%Fe) substrates were obtained by progressive cold rolling of polycrystalline, randomly oriented Ni and Ni-alloy bars to about a 95%-98% deformation. After mechanical deformation, 50 μm thick and 1 cm wide substrates were first cleaned with isoproponal in a reel-to-reel ultrasonic cleaning unit, and then annealed at 1100 ^oC (for pure Ni) and 1250^{o}C (for Ni-alloy) for 1 h in a reducing, forming gas (96%Ar + 4%H_2) atmosphere. The procedure yields the desired {100}<100> cube texture to typical levels of 6 – 8 degrees FWHM.

A 2-methoxyethanol solution of gadolinium methoxyethoxide/acetate, prepared by an alkoxide sol-gel synthesis route, was used to coat the Ni and Ni-alloy tapes. The details of the solution preparations were reported earlier [7,8]. In a reel-to-reel dip-coating unit tapes with dimensions of 10 cm in length and 1cm in width were coated to determine the optimum processing parameters of Gd_2O_3 seed layers on Ni-alloy. These double-sided, dip-coated tapes were then transported through a pre-heated furnace at various temperatures (T_{an}) and speeds (S_{an}) ranging from 1050 to 1250 ^oC and 5 to 30 cm/h in flowing forming gas atmosphere, respectively. The same optimum growth conditions were also used for the growth of Gd_2O_3 films on Ni tapes.

Preparation of YBCO test structures and characterization techniques

Sputtering by the *rf*-magnetron technique was used to deposit both YSZ and CeO_2 overlayers on short segments of the dip-coated substrates, yielding the buffer-layer sequence, CeO_2/YSZ/Gd_2O_3/Ni-(3at%W-1.7at%Fe). Sputtering conditions for both oxides consist of a mixture of 10 mTorr forming gas and 2×10^{-6} Torr H_2O, and a substrate temperature of 780 ^oC. Typical film thicknesses for these YSZ and CeO_2 layers were around 200 nm and 20 nm, respectively. To assess the quality of the CeO_2/YSZ/Gd_2O_3/Ni-alloy multilayer structure, YBCO films were deposited by pulsed laser deposition (PLD), using a XeCl excimer laser system, operated with energy density of $\approx$4 J/cm^2. Typical YBCO film thicknesses were 200 nm.

The X-ray diffraction was employed to analyze phase, structure and texture of the buffer layers as well as the YBCO coatings deposited on short samples. Pole figures were collected by a 4-circle diffractometer. Microstructural investigations were conducted using a Hitachi S-4100 high-resolution field emission type scanning electron microscope (HRSEM). A standard four-point contact technique was applied to measure the superconducting critical temperature (T_c) and J_c of the YBCO films. Values of J_c were assigned at a 1 μV/cm criterion.

RESULTS AND DISCUSSION

Growth of Gd_2O_3 seed layers on biaxially textured Ni-alloy substrates

A series of experiments at various processing temperatures and speeds ranging from 1050 to 1250 ^oC and 5 to 30 cm/h in flowing forming gas atmosphere, respectively, were made to determine the optimum processing conditions of Gd_2O_3 seed layers on Ni-alloy tapes. High quality Gd_2O_3 layers were achieved at temperatures between 1100-1150^oC and speeds < 20 cm/hr. A typical XRD θ-2θ scan for a dip-coated Gd_2O_3 seed layer processed at 1150^oC and S_{an} of 10 cm/hr on a Ni-alloy substrate is shown in Fig. 1. The pattern indicates excellent c-axis orientation of the films. The epitaxial orientation of the Gd_2O_3 films on textured Ni-alloy substrates was characterized by XRD pole figure analysis. Inset shows the logarithmic-scale (222) pole figure of the same Gd_2O_3 film, exhibiting a single domain epitaxy with the Gd_2O_3[001]//substrate[001] and Gd_2O_3[110]//substrate[100].

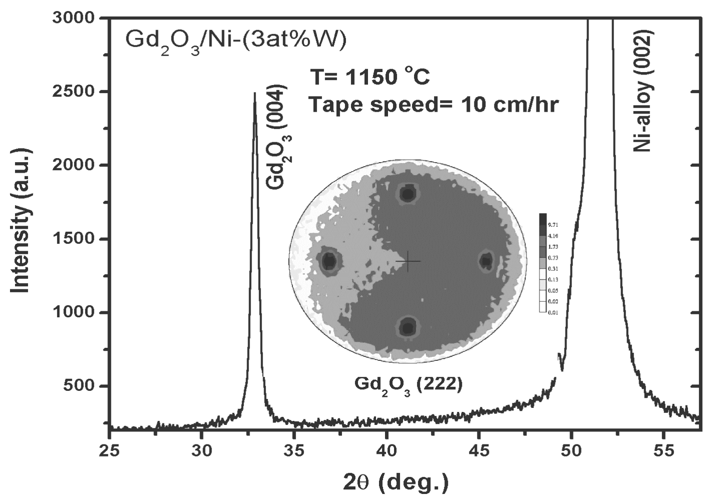

Figure 1. XRD θ-2θ pattern of a 25 nm thick dip-coated Gd_2O_3 film grown on biaxially textured Ni-(3at%W) substrates. The inset shows (222) logarithmic pole figure of the same film.

For implementation of low-cost, non-vacuum dip-coated Gd_2O_3 seed layers into long-length wire manufacturing, it is important grow dense and crack-free layers. To assess this, plan-view SEM micrographs of the surface morphology for 25 nm and 75 nm thick Gd_2O_3 films fabricated on textured Ni-alloy substrates are presented in Figs. 2a, and 2b, respectively. While each sample exhibits a continuous, crack-free and dense surface morphology, significant surface roughening develops for the thicker coatings (Fig. 2b). On the other hand, thinner coatings (25 nm) exhibit extremely smooth and featureless surface microstructure (Fig. 2b), with an rms roughness of ~3 nm as determined by atomic force microscopy measurements, on a 2.5X2.5 μm area, shown in Fig. 2c. Such surface morphology should be realized to achieve the chemical and structural integrity of the subsequent buffer and HTS layers.

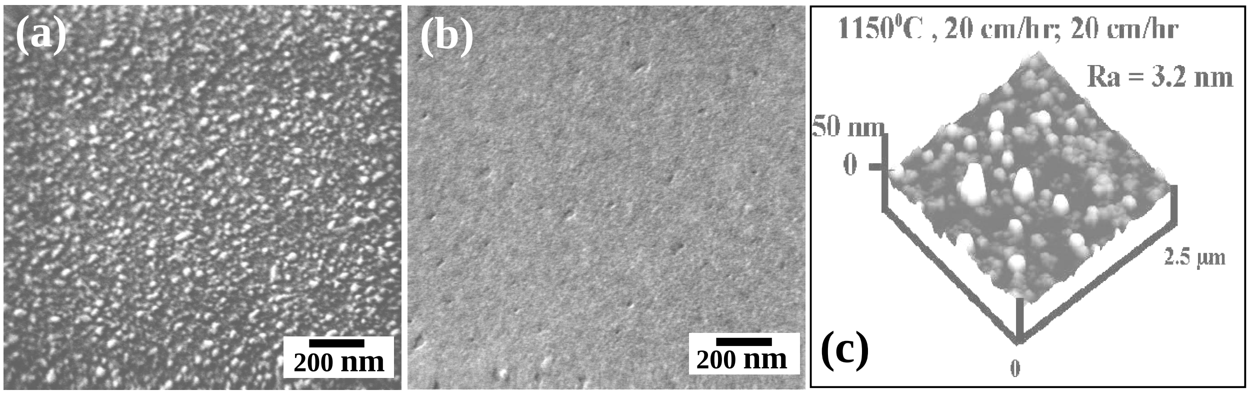

Figure 2. High-resolution SEM micrographs for (a) 73 nm, and (b) 25 nm thick sol-gel dip-coated Gd_2O_3 seed layers on (100) textured Ni-(3at%W-1.7at%Fe) substrates. (c) Displays the AFM image of the same film shown in part b.

Structural and superconducting properties of YBCO films on CeO_2/YSZ/Gd_2O_3/Ni-alloy

To demonstrate dip-coated Gd_2O_3 on Ni-alloy substrates could be used as a template to grow high quality YBCO films, barrier YSZ and cap CeO_2 layers were deposited on Gd_2O_3 by *rf*-sputtering. The results of XRD θ-2θ scans (not shown here) showed that CeO_2/YSZ bilayers grown on thicker Gd_2O_3 layers (Fig. 2a) have a polycrystalline nature, whereas films deposited on thinner seed layers(Fig. 2b,c) exhibit excellent c-axis oriented growth. This growth behavior seems overwhelmingly governed by the surface morphology of the Gd_2O_3 films. It is known that surface roughness exacerbates adverse interfacial reaction between the film and the substrate. Therefore, the relatively coarse surface morphology of thicker Gd_2O_3 films may contribute to polycrystalline growth behavior of the CeO_2/YSZ overlayers. Following deposition of CeO_2/YSZ overlayers, suitability of the CeO_2/YSZ/Gd_2O_3/Ni-alloy structure was tested with the growth of YBCO films. Figure 3 compares the magnetic field (H//c-axis) dependence of J_c at 77 K with those of YBCO films grown on solution derived CeO_2/YSZ/Gd_2O_3(dip-coated)/Ni and on standard, all vacuum deposited, buffer layer RABiTS architecture of CeO_2/YSZ/CeO_2/Ni. The inset of this figure shows the superconducting transition region of the temperature-dependent resistivity, yielding a zero-resistance T_c = 90.5 K. The zero-field transport J_c of the YBCO/CeO_2/YSZ/Gd_2O_3/Ni-alloy sample is $1.2x10^6$ A/cm^2 and it exhibits a high irreversibility field (H_{irr}) of 7 T. This J_c performance is comparable to that of epitaxial YBCO films on CeO_2/YSZ/Gd_2O_3(dip-coated) and CeO_2/YSZ/CeO_2 buffered biaxially textured Ni substrates.

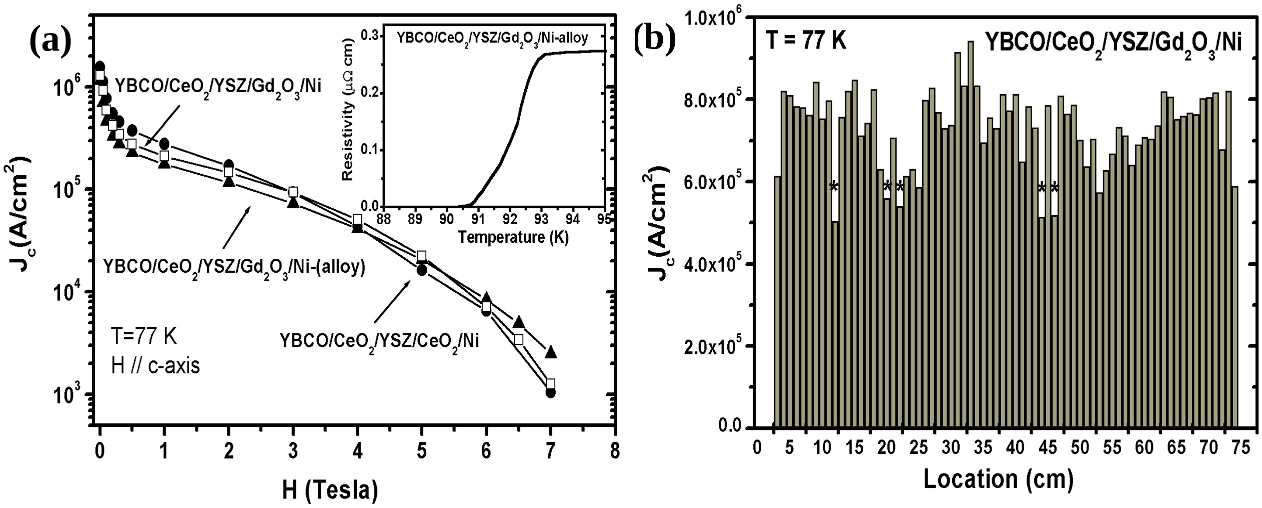

Figure 3. (a) Magnetic field dependence of J_c (H=0 T), measured at 77 K, for a YBCO film on the $CeO_2/YSZ/Gd_2O_3$/Ni-(3at%W-1.7at%Fe) multilayer structure. Also shown for comparison are J_c vs H for the $YBCO/CeO_2/YSZ/Gd_2O_3/Ni$ and $YBCO/CeO_2/YSZ/CeO_2/Ni$. The inset shows the resistive superconducting transition region of the present sample. (b) Sectional J_c values for the YBCO film obtained on a 1 m sol-gel seeded Ni tape. Visible line features marked by (✳).

<u>Long-length fabrication of RABiTS architecture based on dip-coated Gd_2O_3/Ni tapes</u>

Having realized excellent structure and superconducting properties of YBCO films on short segments of CeO_2/YSZ capped, sol-gel synthesized Gd_2O_3 seed layers; we implemented the same architecture on pure Ni tapes. Figure 3b shows the sectional J_c(77 K, self-field) results of a 80 cm long $YBCO/CeO_2/YSZ/Gd_2O_3/Ni$ tape, where the YBCO film was deposited using the *ex-situ* BaF_2 precursor process in a reel-to-reel system. The sample exhibits an average J_c value of $7.5x10^5$ A/cm^2 and it supports an end-to-end J_c of $6.25x10^5$ A/cm^2, with some regions reaching to J_c values of $1x10^6$ A/cm^2. The low J_c sections marked with, "✳", are associated with visible line features along the YBCO thickness, resulting from the variations in the composition of YBCO(BaF_2) precursor films. Despite the existence of these flaws, this result establishes a new benchmark for performance that was attained on a RABiTS structure having a non-vacuum solution processed seed layers.

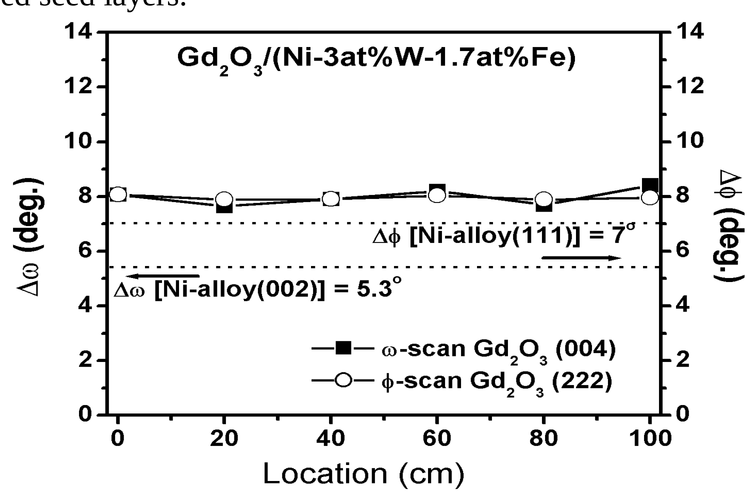

Figure 4. The FWHM values of ω- and ϕ-scans for sol-gel Gd_2O_3 seed layer deposited on biaxially textured Ni-(3at%W-1.7at%Fe) substrate as a function of position along the tape length.

In the light of the above results, we have just started long length implementation of sol-gel Gd_2O_3 seed layers on strengthened Ni-alloy tapes. Figure 4 shows the ω-rocking curve and ϕ-scan FWHM values of the Gd_2O_3 layer as a function of position along the 1 m length of Ni-alloy tape. The dashed lines serve as visual guide for the in- and out-of-plane texture of the metal substrate. As evident from the figure, Gd_2O_3 film exhibits very uniform texture with similar in- and out-of-plane FWHM of about 8^o, indicating epitaxial growth along the entire length of the Ni-alloy substrate. Hence, these results, together with those obtained from microstructural investigations implies that the production of long length YBCO-based coated conductors on strengthened Ni-alloy tapes is possible by the combination of both non-vacuum and vacuum processes. Currently, efforts are underway to complete the buffer structure with CeO_2/YSZ over layers for the further growth of high-quality YBCO films in lengths on Gd_2O_3 buffered Ni-alloy substrates. In addition, we are also investigating an all-solution route to produce both buffers and the YBCO superconductors for the practical development of coated conductors.

SUMMARY AND CONLUSION

We have demonstrated for the first time the successful fabrication of high-J_c YBCO coatings on sol-gel processed, reel-to-reel dip-coated Gd_2O_3 seed buffer layers on textured, strengthened Ni-alloy substrates, with sputtered CeO_2/YSZ over layers. The crystalline structure and the surface quality of the Gd_2O_3 were excellent. On short prototype Ni-alloy tapes, having solution processed seed layers, high-quality epitaxial YBCO films with J_c(77 K, self-field) values exceeding $1x10^6$ A/cm^2 were grown for the first time. After establishing this proof of principal on short prototype conductors, we have presented our results on the long length demonstration of dip-coated Gd_2O_3 seed layers on biaxially textured Ni as well as on Ni-alloy tapes. On both tapes, Gd_2O_3 showed a very uniform texture along tape length (100 cm) and the YBCO films grown by using *ex-situ* BaF_2 precursor approach on the non-vacuum and vacuum derived 80self-field J_c value of $7.5x10^5$ A/cm^2 at 77K with some regions reaching to $1x10^6$ A/cm^2.

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