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# Modified Lanthanum Zirconium Oxide buffer layers for low-cost, high performance YBCO coated conductors

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#### ABSTRACT

The pyrochlore Lanthanum Zirconium Oxide, La<sub>2</sub>Zr<sub>2</sub>O<sub>7</sub> (LZO), has been developed as a potential replacement barrier layer in the standard RABiTS three-layer architecture of physical vapor deposited CeO<sub>2</sub> cap/ YSZ barrier/Y<sub>2</sub>O<sub>3</sub> seed on Ni–5%W metal tape. The main focus of this research is to ascertain whether: (i) we can further improve the barrier properties of LZO; (ii) we can modify the LZO cation ratio and still achieve a high level of performance; and (iii) it is possible to reduce the number of buffer layers. We report a systematic investigation of the LZO film growth with varying compositions of La:Zr ratio in the La<sub>2</sub>O<sub>3</sub>-ZrO<sub>2</sub> system. Using a metal-organic deposition (MOD) process, we have grown smooth, crack-free, epitaxial thin films of  $La_xZr_{1-x}O_y$  (x = 0.2–0.6) on standard Y<sub>2</sub>O<sub>3</sub> buffered Ni–5W substrates in short lengths. Detailed XRD studies indicate that a single epitaxial LZO phase with only (001) texture can be achieved in a broad compositional range of x = 0.2 - 0.6 in La<sub>x</sub>Zr<sub>1-x</sub>O<sub>y</sub>. Both CeO<sub>2</sub> cap layers and MOD-YBCO films were grown epitaxially on these modified LZO barriers. High critical currents per unit width, *I<sub>c</sub>* of 274–292 A/cm at 77 K and self-field were achieved for MOD–YBCO films grown on La<sub>x</sub>Zr<sub>1-x</sub>O<sub>v</sub> (x = 0.4-0.6) films. These results indicate that LZO films can be grown with a broad compositional range and still support high performance YBCO coated conductors. In addition, epitaxial MOD La<sub>x</sub>Zr<sub>1-x</sub>O<sub>y</sub> (x = 0.25) films were grown directly on biaxially textured Ni–3W substrates. About 3  $\mu$ m thick YBCO films grown on a single MOD-LZO buffered Ni-3W substrates using pulsed laser deposition show a critical current density,  $J_c$ , of 0.55 MA/cm<sup>2</sup> ( $I_c$  of 169 A/cm) at 77 K and 0.01 T. This work holds promise for a route for producing simplified buffer architecture for RABiTS based YBCO coated conductors.

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#### 1. Introduction

The main objective of this research is to fabricate potentially lower-cost, high performance second generation YBCO (YBa<sub>2</sub>Cu<sub>3</sub>- $O_{7-\delta}$ ) coated conductors using the RABiTS (Rolling Assisted Biaxially Textured Substrate) template for electric-power applications [1–4]. Efforts are currently underway to replace the existing physical vapor deposited three buffer layer RABiTS architecture of Yttrium Oxide, Y<sub>2</sub>O<sub>3</sub> seed/Yttria Stabilized Zirconia, YSZ barrier/ Cerium Oxide, CeO<sub>2</sub> cap with buffers deposited by industrially scalable methods, such as slot-die coating of metal–organic deposition (MOD) precursors [1,2]. However, for optimizing the MOD film growth and process conditions of short samples, spin coating was used to deposit the films. MOD processes offer significant cost

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advantages, such as a non-vacuum process with 100% materials utilization compared to physical vapor deposition (PVD) processes [5–12]. MOD–CeO<sub>2</sub> has been developed as a potential cap layer and has enabled the growth of high performance MOD-YBCO films [13,14]. Sputtered Y<sub>2</sub>O<sub>3</sub> seed layers show an improved texture relative to the Ni–W substrate texture. Recently, MOD–La<sub>3</sub>TaO<sub>7</sub> (LTO) has been developed as a potential seed layer with improved texture on Ni-3W substrates [15,16]. La<sub>2</sub>Zr<sub>2</sub>O<sub>7</sub>, LZO, has been developed as a potential barrier layer to replace the sputtered YSZ layer [17,18]. Typically, a minimum of 75-nm thick LZO is required to act as a Ni-diffusion barrier [17-19]. In this work, we focused on: (i) further improving the barrier properties of LZO, (ii) studying the effect of varying compositions of La:Zr ratio with the compositional window of  $La_xZr_{1-x}O_y(x = 0.1-0.7)$  in the  $La_2O_3-ZrO_2$  system, and (iii) simplifying the buffer architecture. We report our successful growth of modified MOD-LZO films on standard sputtered Y<sub>2</sub>O<sub>3</sub> buffered Ni-5W substrates in short lengths and demonstrate the growth of high performance YBCO films. In addition, we have also demonstrated the growth of high I<sub>c</sub> YBCO films on a single LZO buffer laver.





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#### 2. Experimental procedure

State-of-the-art Y<sub>2</sub>O<sub>3</sub> buffered, biaxially textured, Ni-5W (5 at. wt.%) substrates were used for this study [2]. The substrates typically were cut to 1-cm wide and 5-cm long from 4-cm wide and 75-µm thick tapes. Modified LZO precursor solutions with various compositions of  $La_x Zr_{1-x}O_y$  (x = 0.1–0.7) were prepared from Lanthanum isopropoxide (Alfa, La 40% assay), Zirconium n-propoxide in *n*-propanol (Alfa, 70%), and 2-methoxyethanol (Alfa, spectrophotometric grade). The details of the solution preparation were similar to that of the standard LZO (where x = 0.5) preparation conditions [10,12]. About 0.75 M total cation concentration was used to grow 100-nm thick LZO films in a single coat on Y<sub>2</sub>O<sub>3</sub> buffered Ni-5W substrates. Spin coating was used to deposit modified LZO films at a spin rate of 2000 rpm for 30 s in a controlled humidity of 40-60%. The spin coated films were then crystallized at 900 °C for 15 min in a flowing gas mixture of Ar/H<sub>2</sub> 4%, and subsequently quenched to room temperature. About 75 nm thick sputtered CeO<sub>2</sub> cap and 0.8 µm thick MOD-YBCO were deposited on modified MOD-LZO using American Superconductor's proprietary TFA process [9]. For simplified buffer work, Ni-3W (3 at.%) substrates were used. On single MOD buffers, YBCO films containing 1 vol.% BaZrO<sub>3</sub> (BZO) were deposited by pulsed laser deposition (PLD) at 790 °C in 230 mTorr oxygen, using an average laser energy density of 2 I/cm<sup>2</sup>. Following deposition, the films were annealed under 500 Torr oxygen for 30 min. The thickness of YBCO determined from the TEM cross-sectional images was found to be 3 µm. YBCO films were then prepared for current density measurements by depositing silver for current and voltage leads, followed by oxygen annealing at 500 °C for 1 h. The films were characterized for phase purity and texture using X-ray Diffraction (XRD). Secondary Ion Mass Spectroscopy (SIMS) depth profile analyses were carried out to study the metal and oxygen diffusion properties of buffers. The resistivity and transport critical current density, J<sub>c</sub>, were measured using a standard four-point probe technique. Microstructural analysis was done by cross-section transmission electron microscopy (TEM) and scanning electron microscopy (SEM).

#### 3. Results and discussion

ntensity (a.u.)

26

La20Zr80 La30Zr70

La40Zr60

a 50 Zr 50

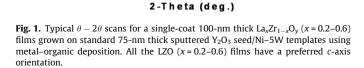
28

Typical  $\theta$ -2 $\theta$  XRD patterns for a single-coat 100-nm thick La<sub>x</sub>Zr<sub>1-x</sub>O<sub>y</sub>(x = 0.2-0.6) films grown on standard 75-nm thick sputtered Y<sub>2</sub>O<sub>3</sub> seed/Ni-5W templates are shown in Fig. 1. All of the cubic fluorite-type LZO (La 20–60%) films have a preferred *c*-axis

Zr<sub>v</sub>O<sub>7</sub> (004)

34

3 6

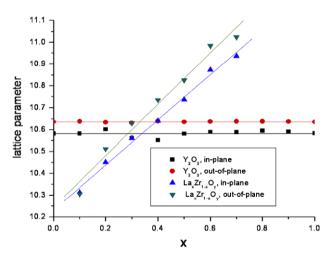


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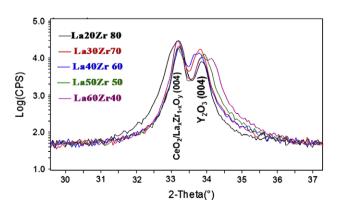
30

orientation. No (1 1 1) texture was observed for any of these films. The position of the LZO (004) peak shifts towards lower  $2\theta$  values (from the right side of  $Y_2O_3$  (0 0 4) peak for lower La content (20%) to the left side of  $Y_2O_3$  (004) for higher La content (60%) films) due to an increase in the lattice parameter with increase in La content. Detailed XRD results from  $\omega$  and  $\phi$  scans revealed good epitaxial texturing. The full widths at half maximum (FWHM) values for all the modified LZO films are comparable to that of the underlying Y<sub>2</sub>O<sub>3</sub> seeds. Highly textured modified LZO films with a wider composition window were achieved. Both in-plane and out-of-plane lattice parameters for single-coat 100-nm thick  $La_xZr_{1-x}O_y$ (x = 0.1-0.7) films are reported in Fig. 2. All the LZO (La 10-70%) films have been indexed as a pseudo-cubic structure. A single phase was observed using XRD for films with a composition of La 20-60%, with more than 10% impurity phases observed in the other compositions.

In order to evaluate the performance of the modified MOD–LZO barrier layers, both sputtered CeO<sub>2</sub> cap and MOD–YBCO films were deposited. Typical  $\theta - 2\theta$  XRD patterns for a sputtered CeO<sub>2</sub> cap layer on a single-coat 100-nm thick La<sub>x</sub>Zr<sub>1–x</sub>O<sub>y</sub> (x = 0.2-0.6) barrier/75-nm thick sputtered Y<sub>2</sub>O<sub>3</sub> seed/Ni–5W templates are shown in Fig. 3. All the CeO<sub>2</sub> films have a preferred *c*-axis orientation, with



**Fig. 2.** Both in-plane and out-of-plane lattice parameters for a single-coat 100-nm thick La<sub>x</sub>Zr<sub>1-x</sub>O<sub>y</sub> (x = 0.1-0.7) films grown on standard 75-nm thick sputtered Y<sub>2</sub>O<sub>3</sub> seed/Ni-5W templates using metal-organic deposition. All the L2O (x = 0.1-0.7) films have been indexed as a pseudo-cubic structure. A single peak was observed for films started with a composition of x = 0.2-0.6. However, for the other compositions, mixed pyrochlore and fluorite phases were observed.



**Fig. 3.** Typical  $\theta - 2\theta$  scans for a 75-nm thick sputtered CeO<sub>2</sub> cap layer on a singlecoat 100-nm thick La<sub>x</sub>Zr<sub>1-x</sub>O<sub>y</sub> (x = 0.2–0.6) barrier/75-nm thick sputtered Y<sub>2</sub>O<sub>3</sub> seed/Ni–5W templates. All of the CeO<sub>2</sub> films have a preferred *c*-axis orientation.

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