

# A compositional gradient $\text{Ce}_{1-x}\text{Zr}_x\text{O}_2$ buffer architecture for producing high-performance YBCO film

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## ABSTRACT

In this work, YBCO films were epitaxially grown on  $\text{Ce}_{1-x}\text{Zr}_x\text{O}_2$  (CZO) buffered yttria stabilized zirconia (YSZ) single crystal substrates through a fluorine-free sol-gel process. The influence of two different CZO buffer architectures on the microstructure and superconducting properties of YBCO films was investigated. According to XRD results in combination with (S)TEM analysis, it was confirmed that YBCO film grown on the gradient CZO buffer layer (CZO-G) exhibits better in-plane and out-of-plane texture than YBCO film deposited on the uniform CZO buffer layer (CZO-20). And besides, the superconducting performances especially the current-carrying capacity under magnetic field of the former was much higher than that of the latter, which means that the enhanced flux pinning force will be presented in YBCO film on CZO-G buffer layer. The maximum pinning force of the YBCO film grown on CZO-G buffer architecture is up to  $17.67 \text{ GN/m}^3$  @ 65 K. HR-STEM analysis shows that the stacking faults within YBCO matrix gave rise to the considerable nanoscale lattice-strains which may suppress the formation of Copper pairs and produce nanoscale non-superconducting regions served as pinning centers to significantly improve the flux pinning performance of YBCO film. Given the above, it has been implied that high-performance YBCO coated conductor can be developed by using a compositional gradient  $\text{Ce}_{1-x}\text{Zr}_x\text{O}_2$  films as buffer architecture.

## 1. Introduction

The second-generation high temperature superconductors, also called as REBCO coated conductors (CCs), have been or will be widely used in many kinds of fields such as Power Source field (e.g. Superconducting Cables, FCL and Energy storage), Transportation field (Superconducting Maglev) and Medical Treatment field (e.g. MRI, MCG and NMR). It has been well known that, in comparison with the conventional conductors, REBCO CCs have many unique physical effects such as Zero-resistance and Meissner effects which allows them to be used in electric power and high magnetic fields, and besides, REBCO CCs have extremely large current-carrying capability which is as several hundred times as that of the conventional conductors. Therefore, the development of REBCO CCs has been receiving more extensive attention than ever before [1–6]. Comparing with the first-generation

superconducting tapes ( $\text{BiSrCaCuO}$ , BSCCO) with silver sheathed, the REBCO CCs are fabricated on the rolling assisted biaxially textured (RABiTS) nickel tapes [7] or ion beam assisted deposition (IBAD)  $\text{MgO}$ /Hastelloy substrates using various film deposition techniques to overcome the main drawbacks of the BSCCO superconducting tapes such as the high amount of expensive silver and the low irreversibility fields at high temperatures [8]. Among these techniques, chemical solution deposition (CSD) is considered as a promising approach for developing CCs in a large scale due to its low cost and easy control of stoichiometry. However, it is quite difficult to fabricate the typical architecture of REBCO CCs with sandwich-like buffer layers through all CSD approach because of the complex multilayer stacks [9, 10]. Hence, in order to achieve the so called all CSD fabrication of CCs, many efforts have been focusing on the issue of simplifying the architecture of buffer layer for CCs [11–14].  $\text{La}_2\text{Zr}_2\text{O}_7$ ,  $\text{CeO}_2$  and their derivatives with the

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doping of rare earth elements have been receiving a wide range of attention and they are considered as the most suitable materials for developing simplified buffer layer for CCs [15–19]. Our group has developed a novel buffer layer of zirconium-doped ceria with highly (001)-preferred orientation on RABiTS, which is expected to make it possible that Zr-doped  $\text{CeO}_2$  can be employed to fabricate the novel CCs with a simplified buffer architecture [20]. Although, up to now, high performance CCs with multilayer buffer architecture have been fabricated by CSD in combination with several physical deposition techniques such as pulsed laser deposition or ion beam assisted deposition [21, 22], there are few reports on the preparation of CCs by all-CSD approaches. It has been known that the superconducting properties of YBCO films derived from the traditional TFA-MOD process can be greatly degraded due to the release of hydrofluoric acid gas which has strong corrosion to the buffer layer and YBCO films [23].

In this study, therefore, we attempt to fabricate epitaxial YBCO/ $\text{Ce}_{1-x}\text{Zr}_x\text{O}_2$  bilayer films on (001)-oriented yttria stabilized zirconia (YSZ) single crystal substrates via all chemical solution deposition approaches to verify the feasibility for developing a novel CCs with a simplified buffer architecture.  $\text{Ce}_{1-x}\text{Zr}_x\text{O}_2$  (CZO) thin films were prepared by an inorganic-salts-based CSD method, and YBCO films were prepared by a fluorine-free CSD route which would promote to produce high performance YBCO films. In addition, the influence of the zirconium-doped concentration gradient in CZO buffer layers on the microstructure and superconducting properties of YBCO films was investigated.

## 2. Experimental

CZO and YBCO films were coated successively on YSZ single crystal substrates through an all CSD routes. Firstly, CZO precursor solutions were synthesized by dissolving zirconium oxychloride ( $\text{ZrOCl}_2 \cdot 8\text{H}_2\text{O}$ ) and cerium nitrate ( $\text{Ce}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ ) into methanol under ambient conditions, followed by stirring continuously to yield homogeneous solutions with a total cation concentration of 0.3 mol/L. A series of CZO solutions with different stoichiometric ratio of Ce/Zr can be obtained through adjusting the additive amount of cerium and zirconium in the precursor solutions. Secondly, the YSZ single crystal substrates with the dimension of 10 mm × 10 mm × 0.5 mm were clean in acetone and alcohol using an ultrasonic bath, and then the CZO precursor solutions were coated on the YSZ substrates with a dip-coating method. Subsequently, the CZO gel films were subjected to pyrolysis at 230 °C for 30 min and further annealed at 850 °C–950 °C for 60 min in atmosphere. Finally, YBCO films were deposited on the CZO buffered YSZ substrates by a fluorine-free sol-gel method. The detailed preparation processes of YBCO precursor solution and the corresponding heat-treatment can be learned in our previous studies [24, 25]. In order to obtain thicker CZO films, the above dip-coating and heat-treatment processes can be repeated for several times. In this investigation, a stack of CZO films with a specified stoichiometric ratio of Ce:Zr = 0.8:0.2 (called as Zr-doped uniform composition architecture) were deposited on YSZ substrate layer by layer through repeating the preparation process for three times. Another stack of CZO films with various stoichiometric ratios of Ce:Zr = 0.7:0.3, 0.8:0.2 and 0.9:0.1 (called as Zr-doped concentration gradient buffer architecture) were deposited on YSZ substrate layer by layer. For convenience, the two CZO stacks with different architectures mentioned above are named as CZO-20 (CZO-20/CZO-20/CZO-20) and CZO-G (CZO-30/CZO-20/CZO-10), respectively, in what follows. As for the compositional gradient CZO buffer architecture, the epitaxial growth relationship between tiers is quite close to homoepitaxy, which would make it possible to avoid reaction and diffusion problems between heterogeneous interfaces in the traditional heterogeneous multilayer buffer architecture.

The phase composition and growth orientation of the as-prepared CZO and YBCO films were characterized by using 7000S-type X-ray diffractometer (XRD) and Rigaku SmartLab diffractometer operated

with  $\text{Cu } K_\alpha$  sources. The in-plane and out-of-plane texture were evaluated through the corresponding full width at half maximum (FWHM) values, which can be commonly measured from the  $\phi$ -scan and  $\omega$ -scan (also called rocking curve) curves of one of the preferred orientation reflections, respectively. The cross-sectional morphology and the interface microstructure of YBCO/CZO/YSZ multilayer structure were analyzed by a high-resolution transmission electron microscope (HRTEM, JEOL JEM-3010) and a spherical aberration corrected transmission electron microscope (TEM, JEOL JEM-ARM200F). The surface roughness of CZO films were measured by a Laser scanning confocal microscope (LSCM, Olympus OLS-4000). The superconducting transition temperature ( $T_c$ ) based on the resistance versus temperature curve were measured with pressed-on indium electrodes, using a standard four-probe method. Magnetization critical current density measurement ( $J_c$ ) was carried out on a multi-function vibrating sample magnetometer (VSM, Quantum Design VersaLab) to analyze the flux pinning properties of the YBCO/CZO/YSZ multilayer films. The size of the samples used in magnetic  $J_c$  measurement is around 2.5 mm × 2.5 mm. The applied magnetic field with a strength of 0–3 T was perpendicular to the film surface during the whole magnetization measurement.

## 3. Results and Discussion

The temperature for crystallization and epitaxial growth of metal oxide buffers, such as  $\text{La}_2\text{Zr}_2\text{O}_7$  and  $\text{CeO}_2$ , on NiW metal substrates is usually up to around 1000 °C, which has been reported in the previous references [26, 27]. However, the temperature for the crystallization and epitaxial growth of the oxide buffer layers on YSZ single crystal substrates is much different with that on the NiW substrates. As shown in Fig. 1, two diffraction peaks corresponding to (200) and (400) reflections of CZO-20 film can be seen when the crystallization temperature is set at 850 °C and the intensity of these two peaks becomes much stronger with the increase of the temperature, which indicates that higher temperature is conducive to promoting the crystallization and epitaxial growth of CZO-20 film. Although there is a relative large lattice mismatch between  $\text{CeO}_2$  film and YSZ substrates, this mismatch can be alleviated through introducing specific metal ions into the crystal lattice to slightly adjust the lattice constant of  $\text{CeO}_2$ . Based on this idea, a Zr-doped concentration gradient buffer architecture (CZO-G) referred in the Experimental section was prepared on YSZ single substrate. For comparison, a Zr-doped uniform composition architecture (CZO-20) was also deposited on YSZ substrate under the almost same preparation condition (950 °C for 60 min in atmosphere).

Fig. 2 shows the XRD results of the above two specimens. It can be seen from Fig. 2(a), (b) that the two buffer architectures exhibit highly (100)-preferred orientation. However, a broadened peak like humps

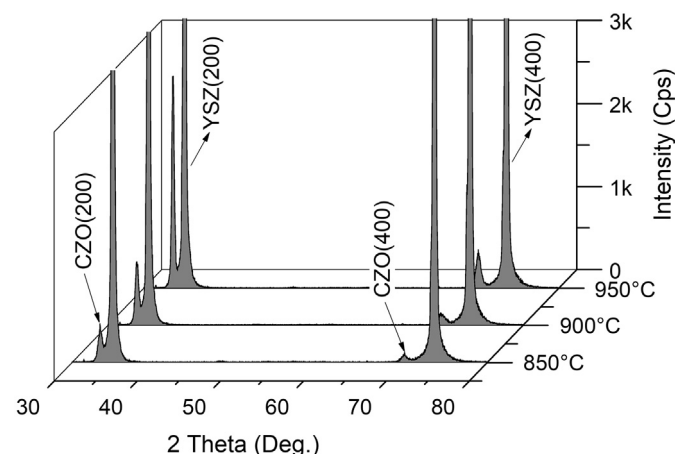


Fig. 1. XRD patterns of  $\theta$ -2 $\theta$  scan for CZO-20 films on YSZ single crystal substrates heat treated at different crystallization temperature.

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