

Chemical Solution Deposition of Epitaxial $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}/\text{CeO}_2/\text{La}_2\text{Zr}_2\text{O}_7$ Multilayer Films on Biaxially-Textured Ni-W Tapes

Hongyan Li*, Xinni Tang, Yuanqing Chen

School of Materials Science and Technology, Xi'an University of Technology, Xi'an, China

Email address:

xblhy@xaut.edu.cn (Hongyan Li)

*Corresponding author

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Abstract: $\text{CeO}_2/\text{La}_2\text{Zr}_2\text{O}_7$ composite buffer layers were deposited on Ni-W tapes using chemical solution method. $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (YBCO) films were then deposited on the $\text{CeO}_2/\text{La}_2\text{Zr}_2\text{O}_7$ composite buffer layers using a low-fluorine solution route. Effect of the annealing temperature and annealing atmosphere on the texture of the CeO_2 layers was investigated. It was found that CeO_2 film annealed at 1000°C under the atmosphere of N_2 containing of 5vol. % H_2 shows a high degree of c-orientation on $\text{La}_2\text{Zr}_2\text{O}_7$ (LZO) film. Effect of oxygen partial pressure on the properties of YBCO films deposited on the CeO_2/LZO buffer layers was also investigated. The YBCO annealed under N_2 containing of 500ppm O_2 shows the optimal properties, with critical current density (J_c) of $1\text{MA}/\text{cm}^2$ at 77K , 0T .

Keywords: Superconductor, Film, Texture, Critical Current Density

1. Introduction

High temperature superconducting (HTS) coated conductors based on $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (YBCO) films are being developed due to its potential of power and energy applications [1]. The realization of textured YBCO coated conductors on flexible metallic substrates is the basic requirement for such engineering applications. In the past two decades, rolling assisted biaxially textured substrates (RABiTS) [2], ion beam assisted deposition (IBAD) [3] and inclined substrate deposition (ISD) have been developed as main techniques to achieve the required biaxially textured YBCO superconducting coated conductor [4]. Among them, the RABiTS technique was frequently used, where an oxide buffer layer architecture was epitaxially grown on the metallic tape followed by the YBCO superconducting layer.

CeO_2 exhibits a small lattice mismatch to YBCO and to the Ni-5W substrate as well as a good chemical stability and high solid density. Therefore, it has been considered as an ideal buffer material for coated conductor [5]. However, the major disadvantage arises from the appearance of micro-cracks as its thickness exceeds 50 nm [6]. Therefore,

CeO_2 has to be combined with other layers to form effective multi-buffer layer architectures. The best results have been reproducibly obtained using the established multi-layered buffer architecture $\text{CeO}_2/\text{YSZ}/\text{CeO}_2/\text{Ni}$ [7]. Currently, the pyrochlore $\text{La}_2\text{Zr}_2\text{O}_7$ (LZO) have attracted much attention as alternative buffer layers for simplifying the standard three-layer system of $\text{CeO}_2/\text{YSZ}/\text{CeO}_2$ by replacing at least the first two layers YSZ/CeO_2 . It was found that combination of CeO_2 with LZO, namely the CeO_2/LZO composite buffer layer, can induce high-performance YBCO coated conductors [8].

Chemical solution deposition (CSD) has emerged as a highly cost-effective method, and has become a competitive alternative to physical vapor deposition techniques for YBCO coated conductors [9–10]. In addition, the main advantages of this method include precise control of the metal oxide precursor stoichiometry, ease of compositional modification on a molecular level, high deposition rates, etc. At present, CSD preparation of buffer layers and YBCO superconducting layers based on RABiTS substrates has become a very

attractive approach for manufacturing high-temperature superconducting wire.

However, the chemical solution deposition of both the CeO_2/LZO buffer layer and YBCO superconducting layer on the Ni-W tape is still a challenge. Optimization of the processing so as to maintain high texture of both the buffer layer and the superconducting layer should be overcome. In this paper, we reported the CSD preparation of the CeO_2/LZO buffer layer and YBCO layer on the Ni-W tapes. Effect of the processing atmosphere on the properties of the CeO_2/LZO and YBCO layer has been investigated.

2. Experimental

Using stoichiometric 2, 4-pentadionates of La and Zr elements, the LZO precursor solution was prepared with methanol and propionic acid as solvents. The total cation concentration of LZO precursor solutions was fixed at 0.3mol/L. To prepare the solution for the CeO_2 film, cerium nitrate was dissolved in a mixture solution of methanol and monoethanolamine.

Textured Ni-5%W alloy tapes were used as substrates. The Ni-5%W tapes were annealed in a flowing 5vol.% H_2/N_2 gas at 850°C for 20 min. After that, LZO gel films were coated on the Ni-W tapes using a dip-coating method. After being dried at 180°C in air for 10 minutes, the coated LZO gel films were crystallized at 1050°C for 60 min in a flowing 5 vol.% H_2/Ar gas. The LZO films were coated and heat treated twice so as to form a film with thickness of 170 nm. Then, the CeO_2 films were coated on the LZO films and heat treated at 950-1050°C under the atmosphere of H_2/N_2 gas. Thus, $\text{CeO}_2/\text{LZO}/\text{NiW}$ composite films were obtained.

The YBCO films were prepared using a solution with molar ratio of Y:Ba:Cu:F=1:2:3:2. The solution preparation process of the YBCO solution has been reported elsewhere [11]. The YBCO gel films were coated on the CeO_2/LZO buffered NiW tapes. The gel films were firstly pyrolyzed at 400°C in oxygen. After that, the films were annealed at 775°C under humidified N_2/O_2 gas with oxygen partial pressure of 50-1000ppm. Finally, the films were post annealed at 450°C in oxygen for 2h.

7000S-type X-ray diffractometer (XRD) was applied to detect the orientation and the phase structure of the films. Atomic force microscopy (AFM, Bruker Dimension Icon) was used to examine the morphology and surface roughness of CeO_2/LZO buffer layers. Surface morphologies of the YBCO films were observed under JEM-6700F scanning electron microscopy (SEM). The J_c of the films was investigated by a J_c -cryoscan device.

3. Results and Discussion

Firstly, the effect of the annealing atmosphere on the crystallinity of CeO_2 film was investigated. The films were annealed at 950°C. The content of H_2 was changed by the controlling the volume ratio of H_2/N_2 . Figure 1 shows the XRD of the CeO_2 films annealed at different H_2/N_2 ratio. All

the CeO_2 films were deposited on the Ni-W tapes. It is found that the when the content of H_2 is 4vol.%. The $\text{CeO}_2(111)$ peak was observed. However, when the H_2 was increased to 6vol.%, only the $\text{CeO}_2(200)$ peak was observed, indicating that the film was of pure c-orientation. The CeO_2 films were then annealed at 950°C, 1000°C and 1050°C. It was found that the degree of the c-orientation was basically improved with the annealing temperature. When the film was annealed at 1000°C, the degree of the c-orientation reaches a high value of 98%. Further increase the annealing temperature has almost on effect on the c-orientation. Therefore, we select the 1000°C as the annealing temperature for CeO_2 film.

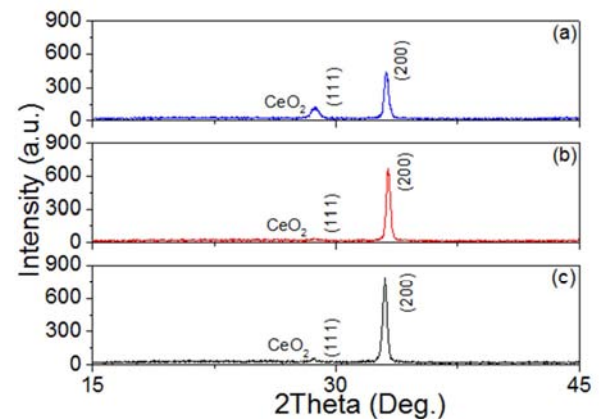


Figure 1. XRD patterns of the CeO_2 film annealed under different atmosphere: (a) 4% H_2/N_2 ; (b) 6% H_2/N_2 ; (c) 8% H_2/N_2 .

To prepare the CeO_2/LZO composite films, the LZO film was firstly prepared on Ni-W tapes. The LZO film was annealed at 1050°C under N_2 with 5vol. % H_2 . After that the CeO_2 film was coated on the LZO and annealed. Figure 3 shows the XRD pattern of the CeO_2/LZO composite film. It can be seen that the only $\text{CeO}_2(200)/\text{LZO}(400)$ peak was observed, indicating that the film is of high c-orientation. The film morphology and the roughness were observed by AFM, as shown in Figure 4. As can be seen that the film shows a flat surface with a roughness of 3-4nm within $10\mu\text{m} \times 10\mu\text{m}$. The flat surface is beneficial for growth of high-quality YBCO films.

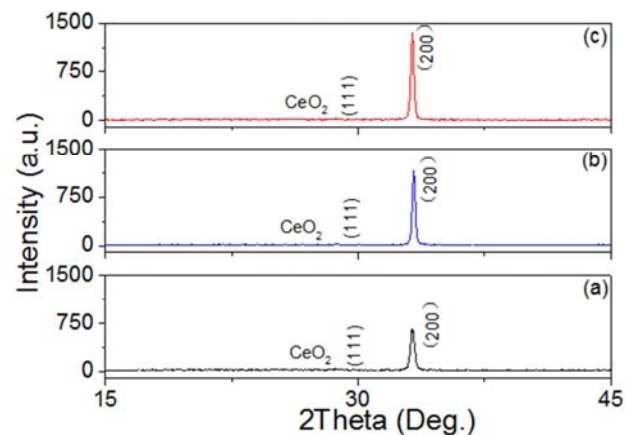


Figure 2. XRD patterns of CeO_2 films annealed at: (a) 950°C; (b) 1000°C; (c) 1050°C.

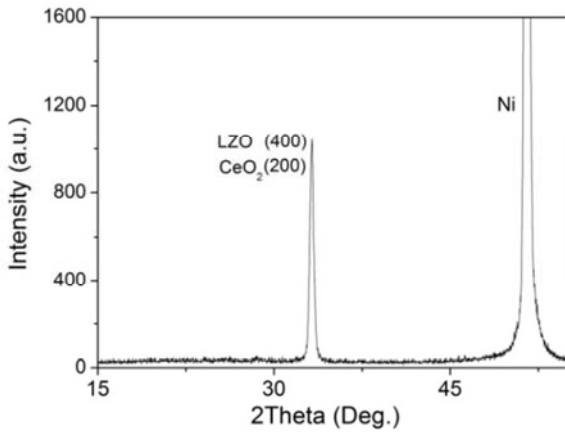


Figure 3. The XRD pattern of the CeO₂/LZO composite film.

The YBCO films were prepared on the CeO₂/LZO buffer layers using a low-fluorine solution and annealed under different atmosphere. The oxygen partial pressure was controlled at 50ppm, 200ppm, 500ppm, and 1000ppm. The XRD patterns of the YBCO films on CeO₂/LZO buffer layers were shown in Figure 5. It can be seen that all the films are of c-orientation. BaCeO₃ phase was detected due to the reaction between the Ba²⁺ with CeO₂ film. At a low oxygen partial pressure, no (200) peak of YBCO phase was detected. However, when the oxygen partial pressure was higher than 500 ppm, the (200) peak was observed, indicating that a-oriented YBCO grains were formed. The room temperature resistance of the YBCO films was shown in Table 1. It can be found that the resistance of the YBCO film annealed with the oxygen partial pressure of 200ppm is lower than that of any other films. For the film annealed under oxygen partial pressure of 50ppm, the film is almost insulating.

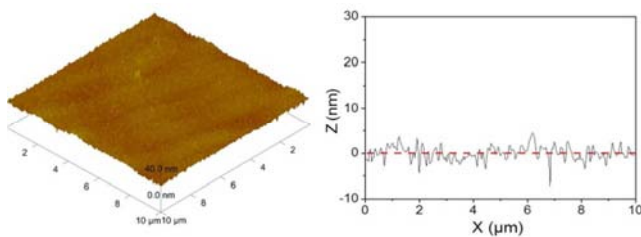


Figure 4. Surface morphology of the CeO₂/LZO composite film and its roughness.

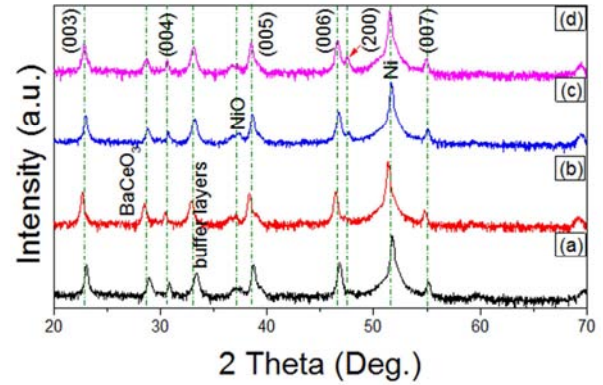


Figure 5. XRD of YBCO/CeO₂/LZO/Ni films with different oxygen partial pressures: (a) 50ppm; (b) 200ppm; (c) 500ppm; (d) 1000ppm.

Table 1. Room temperature resistances of YBCO films annealed under different oxygen partial pressures.

Oxygen partial pressure (ppm)	50	200	500	1000
Sheet Resistance (Ω)	>10 ⁴	17	20	25

The surface morphology of the YBCO films was shown in Figure 6. As can be seen that when the oxygen pressure reaches 1000ppm, there are some a-axis grains observed. In contrast, the film prepared under a low oxygen pressure of 200ppm shows no a-axis grains, which is consistent to the results of the XRD patterns. It should be noticed that the supercurrent transports along the Cu-O planes in the YBCO lattice. That is to say that the supercurrent transports along the a-b planes, other than the a-c, or b-c plane. Therefore, the a-axis YBCO grains will block the supercurrent transportation. Only the c-oriented YBCO grains are beneficial to transport the supercurrent. That is to say, the YBCO film annealed under oxygen partial pressure of 200ppm would be of high superconductivity.

The film superconductivity was measured using inductive method. The J_c distribution was shown in Figure 7. The average J_c value of the YBCO films annealed under oxygen partial pressure of 200ppm, 500ppm, and 1000ppm is 0.93MA/cm², 1.04 MA/cm², and 0.78 MA/cm² respectively. Therefore, the oxygen pressure of 200ppm is the optimal parameter to control the film quality. Under this low oxygen partial pressure, not only the growth of a-axis grains is prevented, but also the oxidation of the Ni tape is prohibited.

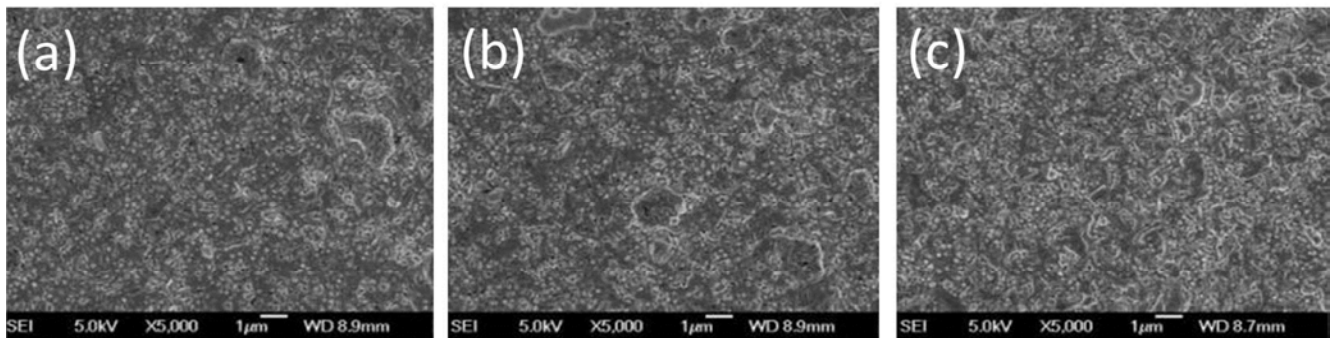


Figure 6. Surface morphology of the YBCO films deposited on CeO₂/LZO buffer layer under different oxygen partial pressure: (a) 200ppm; (b) 500ppm; (c) 1000ppm.

1.23	1.58	1.32	1.29	1.16
0.98	1.05	0.99	0.88	0.90
1.03	1.56	0.62	0.87	0.95
0.97	1.00	0.76	0.66	0.86
0.73	0.80	0.90	0.74	1.53

(a)

1.95	1.72	1.54	1.34	1.83
0.80	1.00	1.41	1.08	1.00
0.83	1.03	1.01	0.93	0.97
0.83	0.98	1.08	0.86	0.96
0.71	0.94	1.05	0.89	0.87

(b)

0.66	1.02	1.06	0.98	1.05
0.82	1.01	0.78	0.76	/
1.11	1.00	0.55	0.66	0.77
1.13	0.79	0.64	0.85	0.84
0.82	0.94	0.76	0.79	0.65

(c)

Figure 7. J_c distribution of the YBCO films annealed under different oxygen partial pressure: (a)200ppm; (b)500ppm; (c)1000ppm.

4. Conclusion

Using chemical solution deposition method, CeO_2/LZO buffer layers were deposited on the NiW tapes. After that, YBCO films were prepared on the CeO_2/LZO buffer layers. Influence of the annealing temperature and atmosphere on the texture of the CeO_2 films was investigated. High quality

CeO_2/LZO buffer layers was produced when the CeO_2 films were annealed at 1000°C under 6vol.% $\text{H}_2\text{-N}_2$ atmosphere. Effect of the oxygen partial pressure on the properties of YBCO films was investigated. Annealed at 775°C with oxygen partial pressure of 200 ppm. YBCO films with J_c larger than $1\text{MA}/\text{cm}^2$ was obtained on the solution-derived CeO_2/LZO buffer layers.

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