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Addition of B_2O_3 precursors and their effect on texture and surface roughness of $La_2Zr_2O_7$ buffer layers

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***Abstract*-- $La_2Zr_2O_7$ (LZO) buffer layers were deposited on biaxially textured Ni-W substrates by chemical solution deposition method (CSD). In this study, the effect of B_2O_3 addition on texture and surface roughness of LZO films was investigated. The alkoxide-based precursor solution was employed to synthesize the precursor solution of LZO and the solution was coated on biaxially textured Ni-W substrates and subsequently annealed at 900°C for crystallization. The pure LZO film without B_2O_3 addition showed a (222) reflection in the X-ray diffraction (XRD) profile. The intensity of (222) reflection was enhanced and more rough surface was obtained after further repetition of coating. Contrary to this, the LZO film prepared by B_2O_3 added precursor solution shows well-developed (400) reflection peak in the XRD profile and excellent biaxial texture ($\Delta\theta$=4.3°, $\Delta\phi$=6.8°). The surface roughness of LZO films were also improved by addition of B_2O_3 even after multicoating (R_{rms} ~ 3.1nm). It was shown that the LZO film with smooth surface and biaxial texture was grown on the biaxially textured Ni-W substrates with addition of B_2O_3 in the precursor solution.**

1. INTRODUCTION

High temperature superconducting coated conductors have attracted much attention for their potential application to high power electric devices such as large scale motors, generators, transformers, etc. One of the most important prerequisites for coated conductor with high critical currents is smooth and crack-free buffer layers epitaxially grown on metallic substrates with excellent biaxial texture. In addition, low cost processing of high quality buffer layers is also important for commercialization. The chemical solution deposition (CSD) of buffer layer provides a cost-effective route to high quality buffer layer epitaxially grown on metal substrates [1-4]. At present, many potential buffer layer materials have been identified for YBCO-coated conductors on cube-textured Ni-based substrates using CSD such as CeO_2, $La_2Zr_2O_7$, $SrTiO_3$, etc.[3-5]. Among them, $La_2Zr_2O_7$ (LZO) with pyrochlore structure is one of the most important buffer materials for coated conductors, owing to its good lattice matching (a=3.81Å) with $YBa_2Cu_3O_{7-x}$ (a=3.83Å) [3].

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Chirayil *et al.* deposited biaxially textured LZO film through coating on Ni-W substrates and subsequent annealing at 1150°C with all alkoxide-based precursor solution based on La-isopropoxide and Zr-n-propoxide [3]. Recently, Knoth *et al.* report fabrication of biaxially textured LZO film on Ni-W substrate after annealing at 900°C [4]. Although they used very smooth substrate (R_{rms} ~ 0.5nm), the surface roughness was increased after LZO coating (R_{rms} ~ 2nm).

It is known that the addition of oxide with low melting point can decrease crystallization temperature and improve surface roughness, since the oxide with low melting point can act as a sintering additive [5]. In this paper, we present the preparation of the biaxially textured LZO film with smooth surface on relatively rough Ni-W substrates (R_{rms} ~ 5.5nm). In particular, B_2O_3 with low melting point (~450°C) was intentionally added to the precursor solution to improve biaxial texture and surface roughness of LZO films.

2. EXPERIMENTALS

2.1. Synthesis of precursor solution

A precursor solution for LZO film was synthesized by mixing La nitrate with Zr-n-propoxide which was stabilized by 2,4-pentanedione dissolved in methanol. The solution was refluxed for 1hr at 70°C and cooled to room temperature. Subsequently, ammonium tetraborate was dissolved in the precursor solution to provide boron oxide formation. The concentration of metal ions in the precursor solution was adjusted to 0.1M by evaporation and the amount of ammonium tetraborate addition was fixed to 2 mol%.

2.2. Deposition of LZO films

Ni-W substrates (supplied from KISWIRE) were applied to deposition of LZO. The Ni-W substrates show biaxial texture ($\Delta\theta$=4.8°, $\Delta\phi$=6.5°) and their surface roughness values were R_{rms} ~ 5.5nm. The surface of Ni-W substrates was degreased by sonication in acetone and LZO precursor solution was deposited by dip coating process. The coated precursor films were calcined at 300°C under ambient atmosphere and crystallization annealing was performed at 900°C under Ar/H2 (4%) mixed gas atmosphere for 1 hour. In this study, coating and calcination was repeated if it needed.

3. RESULTS AND DISCUSSIONS

3.1. Effect boron oxide addition on the texture development of LZO films

As described in the previous section, LZO films were deposited on the biaxially textured Ni-W tapes by CSD process. Fig. 1 shows an X-ray diffraction profile of LZO film deposited using precursor solution without boron oxide addition. No trace of NiWO4 and other secondary phase was detected with X-ray diffraction. Both of (400) and (222) reflection peaks of LZO are observed in the diffractogram. The integrated intensity ratio of LZO (222) to LZO (400) was estimated to be about 0.72 for 1 layer coated film and 0.57 for 3 layer coated film, respectively. The intensity of both (400) and (222) reflection peaks of LZO are increased with repetition of coating. This indicated that large amount of randomly oriented grains are existed in the LZO films. Since the roughness of underlying layer or substrate can increase volumetric portion of randomly oriented grains in the coated film, the origin of randomly oriented grains in LZO films can be attributed to relatively large surface roughness of metal substrates (~7nm) [6].

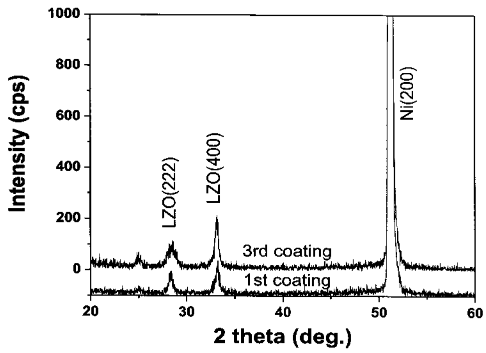

Fig. 1. X-ray diffraction profiles of LZO films.

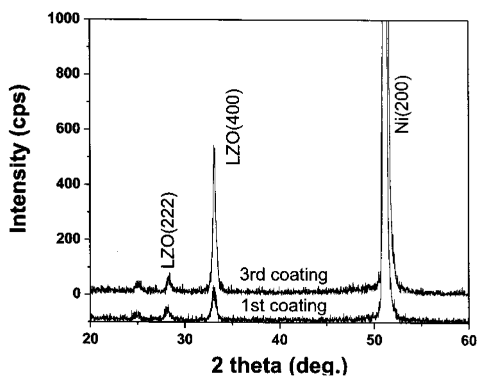

Fig. 2. X-ray diffraction profiles of LZO films with B_2O_3 addition. Peaks near 25° were originated from the Ni-W substrates.

Fig. 2 shows X-ray diffraction profile of LZO film deposited using precursor solution with boron oxide addition. A further analysis of the XRD data indicated that, regardless of the amount of boron addition, the cubic lattice parameter remained essentially constant. This excludes the possibility of the dissolution of boron into the perovskite lattice. Although (222) reflection peak was detected in the diffractogram and did not show noticeable increase with repetition of coating, the intensity of (400) peak was almost tripled by 3 layer coating. Then the integrated intensity ratio of LZO (222) to LZO (400) was 0.29 for the 1 layer coated film and 0.09 for the 1 layer coated film, respectively. Compared with undoped LZO films, LZO films doped with B_2O_3 showed much improved crystallinity and preferred orientation to (100) direction. This means that the addition of B_2O_3 with low melting point enhanced the crystallization of LZO films by accelerating material transport with presence of liquid phase, as reported in the glass-ceramics [7,8].

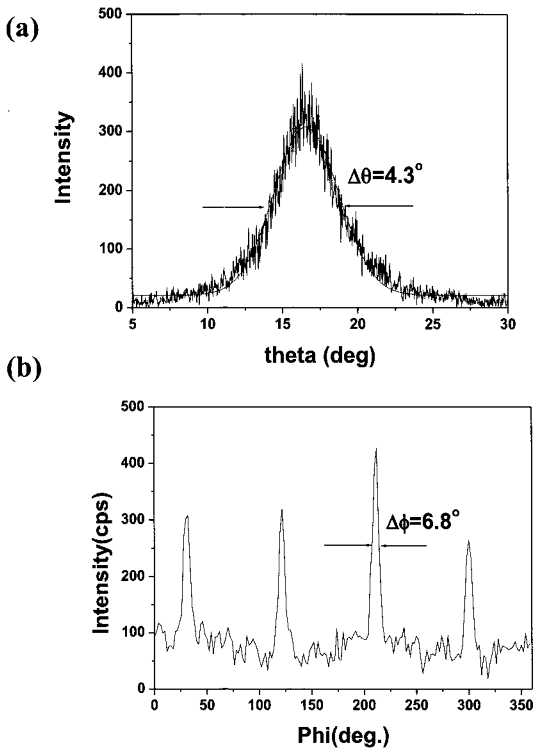

Fig. 3. X-ray diffraction of the LZO film doped with B_2O_3 (after 3 layer coating): (a) theta rocking of LZO (400) peak, (b) phi scan of LZO (111).

The texture analysis of the LZO film was performed with a theta rocking of LZO (400) peak and a phi scan of LZO (111) as shown in Fig. 3. The FWHM (full-width at half maximum) of theta rocking of LZO (400) peak and a phi scan of LZO (111) were estimated to be 4.3° and 6.8°, respectively. The degree of biaxial texture of the B_2O_3 -doped LZO film was nearly similar to that of the Ni-W substrate. This indicates that the LZO film was biaxially grown on the biaxially textured Ni-W substrate.

3.2. Effect boron oxide addition on the surface roughness of LZO films

As described in the previous section, B_2O_3 addition can improve the texture development of LZO films. This can presumably be attributed to improvement of diffusion process for texture development by B_2O_3 doping. Then, it can be inferred that the B_2O_3 addition can also enhanced the surface diffusion and planarize the surface.For the estimate of the surface morphology, each AFM image was obtained using an area of 5×5 μm^2, as shown in Fig. 4. The surface morphology of the undoped LZO film showed much rough surface. Compared with surface roughness of the Ni-W substrate (R_{rms} ~ 7nm), the surface roughness was deteriorated by undoped LZO coating. In particular, large grains were observed and no surface planarization was detected in the 3 layer coated LZO film without B_2O_3 addition (Fig. 4(c)).

Contrary to this, the AFM images of LZO films with B_2O_3 addition showed much smoother surfaces. The RMS rougness of the single coated LZO film was estimated to be 4.6nm which is much lower than that of Ni-W substrates. In addition to this, further repetition of LZO (doped with B_2O_3) coating provides smoother surface (R_{rms} ~ 3.1nm) which is comparable to that of LZO film prepared on Ni-W substrate with highly smooth surface (R_{rms} ~ 0.5nm) [4].

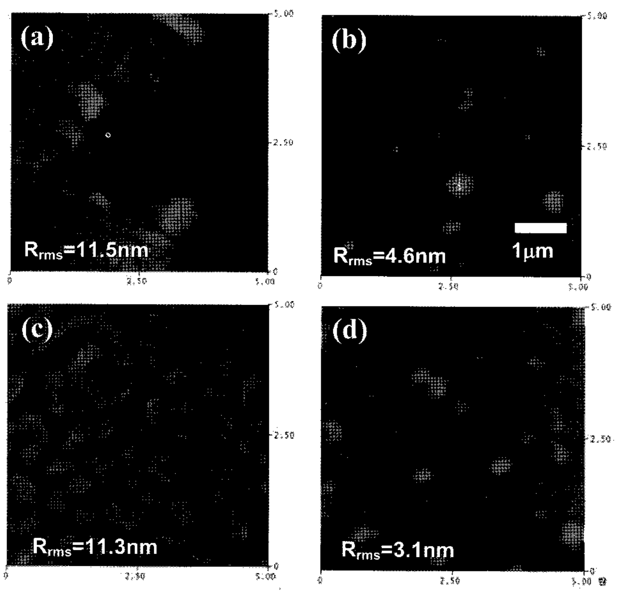

Fig. 4. AFM images of LZO films: (a) undoped (1 layer coating), (b) with B_2O_3 addition (1 layer coating), (c) undoped (3 layer coating), (d) with B_2O_3 addition (3 layer coating).

3.3. Sputtering of CeO_2 coating on LZO film with smooth surface

A further CeO_2 layer was grown on the LZO film by sputtering. Fig. 5 shows a XRD profile for CeO_2-coated LZO(CSD, 3 layer coated)/Ni-W.

Since the (400) reflection of LZO and (200) reflection of CeO_2 have similar diffraction angle (LZO (400): 33.13°, CeO_2 (200): 33.08° for Cu Kα line), only one (h00) reflection peak is observed in the diffractogram. Compared with XRD profile shown in Fig. 2, the intensity of (h00) reflection is doubled by further CeO_2 deposition. However, there still exist randomly oriented grain as shown in the reflection peak in 28.5° which is the position of (222) reflection of LZO or (111) reflection of CeO_2. The degree of biaxial texture estimated from the FWHM (full-width at half maximum) of theta rocking of CeO_2 (200) peak and a phi scan of CeO_2 (111) were estimated to be 5.2° and 6.2°, respectively as shown in Fig. 6. This implies that the biaxial texture of LZO buffer layer was successfully transferred to sputtered CeO_2 layer.

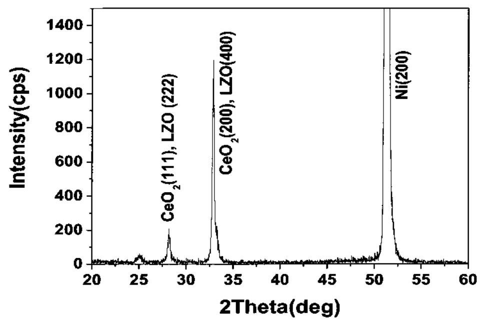

Fig. 5. X-ray diffraction profiles of CeO_2 (sputtered) /LZO(CSD, 3 layer coated)/Ni-W.

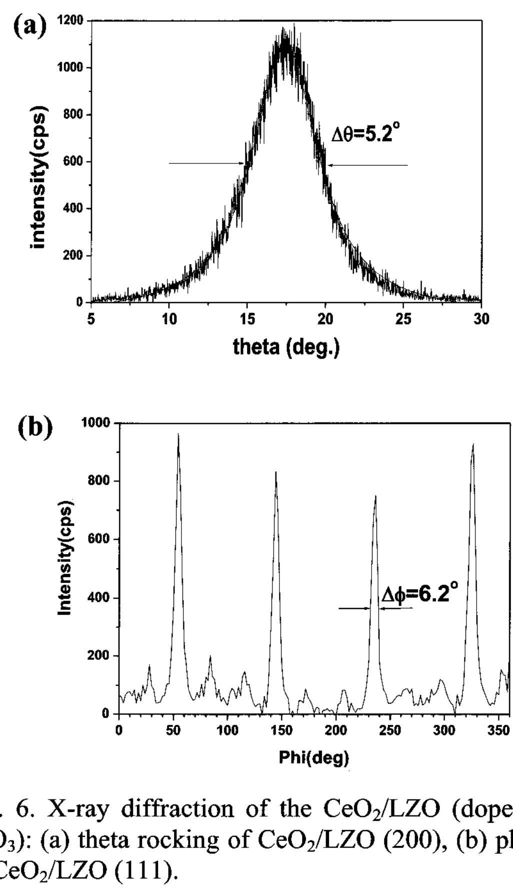

Fig. 6. X-ray diffraction of the CeO_2/LZO (doped with B_2O_3): (a) theta rocking of CeO_2/LZO (200), (b) phi scan of CeO_2/LZO (111).

4. SUMMARY

Biaxially textured $La_2Zr_2O_7$ films with smooth surface were deposited on relatively rough Ni-W substrates by chemical solution deposition. B_2O_3 with low melting point was intentionally added to the precursor solution to improve biaxial texture and surface roughness of LZO films. Both texture development and surface morphology of LZO films were improved by B_2O_3 addition. The LZO film prepared by B_2O_3 added precursor solution shows preferred (400) reflection peak in the XRD profile and good biaxial texture ($\Delta\theta$=4.3°, $\Delta\phi$=6.8°). The surface roughness of LZO films were also improved by addition of B_2O_3 even after multicoating (R_{rms} ~ 3.1nm). It was shown that the LZO film with smooth surface and biaxial texture was grown on the biaxially textured Ni-W substrates with addition of B_2O_3 in the precursor solution.

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