Effect of Gadolinium substitutions in $La_2Zr_2O_7$ films grown on textured Ni-5at.%W substrate by chemical solution deposition

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Abstract

In the $YBa_2Cu_3O_{7-x}$ coated conductor, the buffer layer is crucial for the texture transfer from the Ni-5%W substrate to the superconducting layer. We developed original $La_{2-x}Gd_xZr_2O_7$ buffer layers ($x \leq 1$) by chemical solution deposition to decrease the structure mismatch between the substrate and the buffer layer. They show a strong cube texture up to the surface, a crack free surface and a very low roughness. In conclusion, the $La_{2-x}Gd_xZr_2O_7$ layers with $0.4 < x < 0.9$ are promising and should be employed in the future as buffer layer in coated conductor to replace $La_2Zr_2O_7$ buffer layer.

Introduction

The second generation of high temperature superconductor tapes, called coated conductors, presents a considerable interest in the field of high power applications with low energy consumption [1]. Among the different architectures, the association of a rolling-assisted biaxially textured substrate (RABITS) and Chemical Solution Deposition methods (CSD) are considered as one of the most promising approach for the cost-effective processing of long lengths of coated conductors [2]. These coated conductors are built with a $YBa_2Cu_3O_{7-x}$ (YBCO) layer deposited on a Ni-5%W bi-axially textured substrate coated with intermediate buffer layers. In this architecture the superconducting layer is cube-textured along its principal axes, [001] and [100], to overcome weak superconducting coupling in high angle grain boundaries [3]. The buffer layer is an essential ingredient because it acts as crystallographic template for the growth of YBCO and chemically insulates the YBCO layer from the metal substrate. Therefore, the candidates for buffer materials must be selected based on: i) similar lattice parameters with both substrate and YBCO to support epitaxial growth; ii) the efficiency of the barrier against oxidation of the metallic substrate during YBCO film deposition and metal diffusion from the substrate to the YBCO. $La_2Zr_2O_7$ (LZO) has emerged as a promising buffer layer on Ni-5%W and has been obtained by CSD method by a four steps process: 1) synthesis of the precursor solution; 2) deposition of the precursor film on a substrate by dip-coating; 3) low temperature pyrolysis phase to remove the organic matter (typically 400°C); 4) high temperature heat-treatment for nucleation, crystallization and densification of the buffer layer (typically 1000°C under reducing atmosphere). We employed for the first time the CSD method to substitute Gd for La in $La_2Zr_2O_7$. The objectives were to modify the lattice parameter of the buffer layer in order to decrease the structural mismatch between the buffer layer and the substrate and then to improve the texture transfer. Indeed the structural mismatch $NiW/La_2Zr_2O_7$ is 7.5% and $NiW/Gd_2Zr_2O_7$ 5%. In the present work, we report the synthesis of $La_{2-x}Gd_xZr_2O_7$ buffer layers (LGZOx with $x \leq 1.2$). We have employed X-ray diffractions, and microscopies (Atomic Force Microscopy, Scanning Electron Microscopy, Transmission Electron Microscopy) to study the influence of the gadolinium substitution on the texture and the microstructure.

Experimental

The precursor $La_{2-x}Gd_xZr_2O_7$ solutions ($0 \leq x \leq 1$) are produced by mixing a stoichiometric amount of lanthanum (III) 2,4-pentanedionate hydrate, gadolinium (III) 2,4-pentanedionate

hydrate and zirconium (IV) 2,4-pentanedionate with propionic acid under a continuous stirring at 50°C for 15 minutes. The final solution concentration is 0.6 mol/L. Biaxially textured Ni-5at.%W tapes are used as substrates (Evico, Dresden). Before coating they are cleaned in an ultrasonic bath for 15 minutes in acetone and for 15 minutes in methanol and then are heated up to 800°C for 1 hour in a reducing atmosphere under a continuous flow of Ar-5%H_2 gas to restore the surface properties [02]. The substrates are dip-coated at 60 mm/min and then the precursors layers are dried for 2 minutes at 60°C using infrared lamps. Finally, the coatings are heated at 450°C for 1 hour under primary vaccum and then annealed at 1000°C for 30 minutes under Ar-5%H_2 gas flow. The heat treatment is the same for the different $La_{2-x}Gd_xZr_2O_7$ samples ($0 \leq x \leq 1.2$). X-ray data were collected using a Seifert four circles diffractometer equipped with a Cu K_α source at room temperature in air. Transmission electron microscopy (FEI Titan 50-80), scanning electron microscopy (FEG-SEM, Zeiss Ultra +) and atomic force microscopy (AFM, Veeco D3100) were used to determine the thickness, the morphology and the roughness of the buffer layers.

Results and discussion

X-ray diffraction θ-2θ scans for $La_{2-x}Gd_xZr_2O_7$ buffer layers are shown in Figure 1a (LGZOx with $0 \leq x \leq 1.0$). The crystal structure of LGZO buffer layers is described as the $La_2Zr_2O_7$ (LZO) pyrochlore structure. For the different samples only the (004) peak is observed showing a very strong c-axis preferred orientation of the buffer layer on the Ni-5%W substrate. Undesirable (222) peak was not detected (Figure 1a) while it was always observed for the $Gd_2Zr_2O_7$ buffer layers obtained by chemical solution deposition [4-5]. The lattice parameter decreases almost linearly with increasing gadolinium fraction x (Figure 1b), as expected from the smaller ionic radius of Gd^{3+} (1.00 Å) compared to La^{3+} (1.03 Å). Consequently, the structural lattice mismatch between the buffer layer and the substrate also decreases as a function of the Gd fraction, from 7.5% for LZO to 6.25% for LGZO0.9. However at gadolinium contents higher than 0.9, the intensity of the (004) peak decreases. We have increased the annealing temperature up to 1020°C nevertheless we observe no improvement. We concluded that the upper limit of the solid solution in the $La_{2-x}Gd_xZr_2O_7$ system seem to be close to x = 0.9. A higher Gd content seems disturb the crystal growth of the layer, a similar feature has been reported for cerium substitution in $La_2Zr_2O_7$ layer [02].
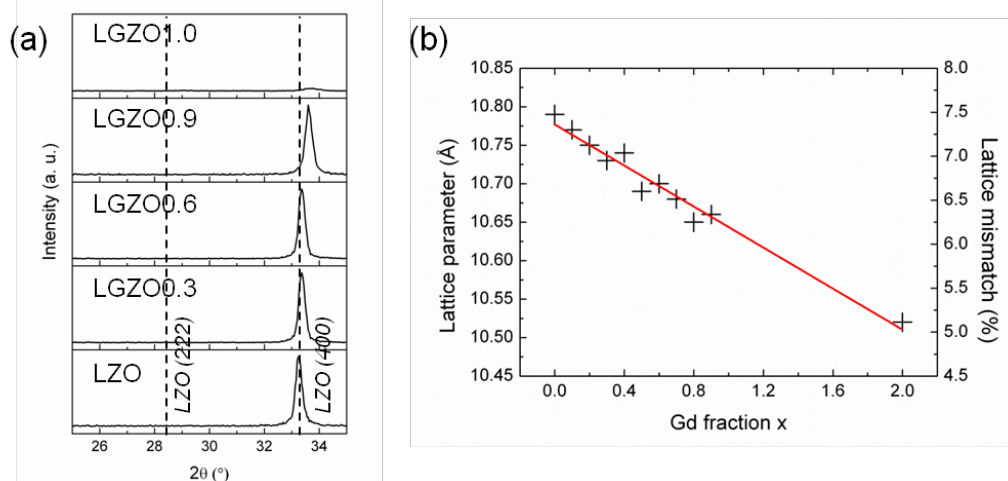

Figure 1 : (a) X-ray diffraction θ-2θ scans for $La_{2-x}Gd_xZr_2O_7$ buffer layers, (b) Variation of lattice parameters and structure lattice mismatch LGZO/Ni-5at.%W across the $La_{2-x}Gd_xZr_2O_7$ series as a function of x.

We have determined the in-plane and out-of-plane texture of the LGZO layers by φ-scans of the LGZO(222) reflection and ω-scans of the LGZO(400) reflection in the rolling direction (RD) and transverse direction (TD) of the Ni-5%W tapes. Figure 2 represents the φ-scan and

the ω-scans of LGZO0.5 and summarizes the average full width at half maximum (FWHM) measured for the La$_{2-x}$Gd$_x$Zr$_2$O$_7$ series as a function of x. In the in-plane direction, the FWHM values are constant as a function of Gd content and remain close to 6.2° which is very close to the values reported for LZO and GZO buffer layers [05]. These values indicate a successful transfer of the substrate biaxial texture to the buffer layer. In the out-plane direction the FWHM values increase almost linearly with increasing gadolinium percentage, from 8° to 9.5° in the RD and from 5.1° to 6.0° in the TD directions. However, they are below the FWHM values reported for Gd$_2$Zr$_2$O$_7$ buffer layers obtained by chemical solution deposition, 10.2° in the RD and 6.9° in the TD direction [5], and are in the appropriate range for the deposition of YBCO layers with high Jc [6].
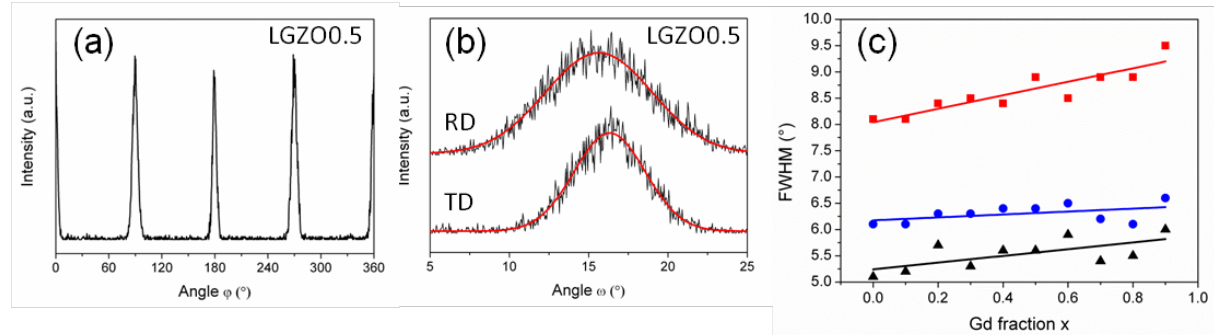

Figure 2 : (a) φ-scan and (b) ω-scans of the LGZO0.5(222) and LGZO0.5(400) reflections in the rolling direction (RD) and transverse direction (TD) of the Ni-5at.%W tapes. (c) Variation of the FWHM value across the La$_{2-x}$Gd$_x$Zr$_2$O$_7$ series as a function of x. (●) = φ-scan (■) = ω-scans TD, (▲) = ω-scans RD.

For the different buffer layers, homogenous and crack free surfaces are observed by SEM analysis, as shown in Figure 3 for LGZO0.8. No modification of the grain size is observed as a function of Gd content. The grain diameter is close to 30 nm and is in agreement with the values reported in the literature for LZO and GZO thin layers [3-4]. The thickness of the buffer layers has been estimated by TEM cross-section analysis to be between 60 and 80 nm (Figure 3). Dark regions with a rectangular shape are observed on the TEM images of the different LGZO layers. They are homogenously distributed throughout the bulk of the materials and their diameter is between 10 and 20 nm. In the literature they are described as nanovoids and are observed for LZO and GZO layers [7]. The origin of the nanovoid formation mechanism is not clearly established and different explanations are proposed [7]: i) the escaping of carbon-rich gases creates the nanovoids and they are intrinsic to chemical solution deposition method; ii) the nanovoid appearance is intrinsic to pyrochlore structure and has no relation with the deposition technique.
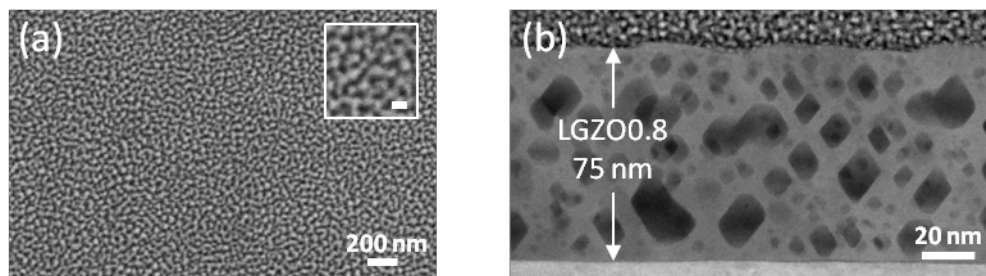

Figure 3 : (a) SEM images of LGZO0.8 layer (50 nm scale bar), (b) TEM dark-field image of LGZO0.8 in cross-section.

AFM investigations of the surface structure of the LGZO samples were carried out and are illustrated in Figure 4. The different pictures confirm the homogenous grain size distribution

observed by the SEM analysis. Nevertheless the root mean square (RMS) roughness of the LGZO buffer layers decreases as a function of Gd ratio, from 2.4 to 1 nm measured on $4\mu m^2$ square samples (Figure 4c). Indeed, we observe also a decrease of the grains size as a function of Gd substitution, from 45 nm for LGZO0.1 to 25 nm for LGZO0.9. The RMS roughness of LGZO layers is lower than the values reported for LZO layer, 2.81 nm [03]. A smooth surface is absolutely necessary for the successful epitaxial growth of YBCO layer on the top of the buffer layer.
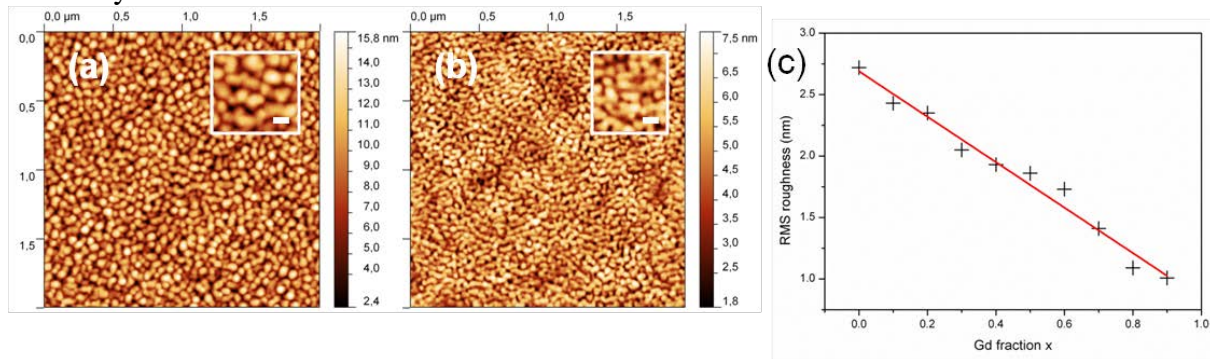

Figure 4 : AFM images of (a) LGZO0.1 and (b) LGZO0.9 on Ni-5%W (scale bar = 50 nm). (c) Variation of the RMS roughness across the $La_{2-x}Gd_xZr_2O_7$ series as a function of x.

Conclusions

This work presents the synthesis and characterization of $La_{2-x}Gd_xZr_2O_7$ layers $(0 < x < 1)$ on Ni-5%W. The structural mismatch between the buffer layer and the substrate decreases linearly as a function of the Gd fraction. Nevertheless, we observe no improvement in the texture of the buffer layers. The SEM and AFM analysis revealed a dense morphology with no reduction of porosity as a function of Gd content. The roughness of the $La_{2-x}Gd_xZr_2O_7$ layers decreases linearly as a function of the Gd percentage. Their surfaces are very smooth which is very important for the epitaxial growth of the YBCO layer. The $La_{2-x}Gd_xZr_2O_7$ layers with x between 0.4 and 0.9 have all the characteristics to be promising buffer layers and their integration in coated conductor will be described in a future article focused on superconducting properties.

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