The role of Pd over layer thickness on PLD YBCO coated conductor properties

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Abstract. Varying the thickness of the Pd layer, the role of the Pd films deposited on the cube-textured Ni-based substrates, was assessed in order to investigate the issues related to substrate oxidation phenomena and their impact on the quality of the buffer layer structures. Using a low temperature annealing, 600 °C, a threshold for the Pd film thickness, beyond which the Pd layer hinders the oxides formation on the substrate, can be defined. When the Pd-Ni interdiffusion is complete, oxide phases of Ni and of Ni-W coexist in the substrate. Besides, at temperatures as high as 800°C, the NiWO₄ formation is favoured as the thickness of the Pd layer increases. Despite of these phenomena, development of a buffer layer template suitable for YBCO growth are reported with a thickness for the Pd layer as low as 20 nm.

1. Introduction

For the development of superconducting tapes based on YBa₂Cu₃O₇₋ₓ (YBCO) films, the beneficial role of a Pd layer between the Ni- or Cu-based substrates and the buffer layer structures has been widely demonstrated [1-5]. On a cube-textured Ni-W substrates buffered with a 200 nm thick Pd layer and a standard buffer-layer architecture, CeO₂/YSZ/CeO₂ (YSZ is Y₂O₃ stabilized ZrO₂), YBCO films have been obtained with $J_C$ up to 2.2 MA/cm² in self-field. These results were obtained for YBCO films grown both by pulsed laser deposition (PLD) or by metal organic decomposition (MOD) techniques. The Pd has different fundamental properties. First of all, the lattice parameter and the coefficient of thermal expansion for the Pd have an intermediate value between the one of the substrate and those of some materials typically used as buffer layer, eg CeO₂ or MgO. This contribute to relieve the misfit stress and the thermal stress of the buffer layer structures grown on cube-textured metal tape. Therefore, the eteroepitaxial growth at relatively low temperatures, as low as 350 or 500°C, and a higher stability against the delamination are assured. The Pd layer shows an orientation distribution in the out-of-plane direction up to 60% sharper with respect to the substrate [5]. This characteristic can be related to the formation of a Ni-Pd solid solution at the interface that can alleviate the high lattice mismatch, about 10%, between the Pd layer and the Ni-W substrate [6]. Besides, it was observed that the quality of the CeO₂/substrate interface after an oxidation process is largely improved by the presence of the Pd layer [7].

In the present work, a study regarding the effects of the Pd layer thickness on the substrate oxidation process and on the properties of the CeO₂/YSZ/CeO₂ buffer layer structure is reported. The obtained results indicate that a Pd film thickness as low as 20 nm can promote the development of high quality buffer layer structures for the development of coated conductors based on YBCO film.
2. Experimental details

The Pd films were deposited by e-beam technique on the cube textured Ni$_{95}$W$_5$ (Ni-W) tapes. Details on the substrate preparation and on the Pd film growth are reported elsewhere [8, 5].

The buffer layer architecture, CeO$_2$/YSZ/CeO$_2$, and the YBCO film have been grown by Pulsed Laser Deposition (PLD) in two different vacuum chambers: one chamber is equipped with a Nd:YAG laser in the 4$^{th}$ harmonic configuration and used for the deposition of the buffer layer structures, and the other chamber equipped with a XeCl excimer laser and used for the YBCO film deposition. In the case of the buffer layer structure, the starting materials were high density sintered targets with purity of 99.99%. The seed layer of CeO$_2$ and the intermediate layer of YSZ were grown at 500°C in vacuum and in 10 mTorr of oxygen flowing atmosphere, respectively. Subsequently, the temperature was raised up to 750°C in vacuum at a rate of 10°C/minute, then a CeO$_2$ cap layer was deposited in a 10 mTorr atmosphere of flowing oxygen. The laser parameters were: fluence of about 2.2 J/cm$^2$ and frequency of 3 Hz. The deposition time is adjusted in order to obtain the following thickness for the deposited layers: 15 nm for the CeO$_2$ top layer, 110 nm for the intermediate YSZ layer and 45 nm for the CeO$_2$ seed layer. The growth rates are 2.5 Å/s and 0.7 Å/s for CeO$_2$ and YSZ, respectively. Details on YBCO film preparation are reported elsewhere [9].

Structural and morphological properties were analysed by means of X-ray diffraction (XRD) and Scanning Electron Microscopy (SEM). Standard X-ray $\theta$-2$\theta$ and $\omega$-scans were performed with the CuK$\alpha$ radiation using a Rigaku Geigerflex diffractometer equipped with a graphite monochromator on the diffracted beam, or with a 9 kW rotating anode Rigaku SmartLab diffractometer equipped with a Johansson monochromator on the incident beam. SEM investigations have been done using a LEO 1525 field emission high resolution scanning electron microscope. The zero resistance temperature transition, $T_C(R=0)$, was derived from the temperature dependence of the electrical resistance measured by a four-point method.

3. Results and discussion

3.1. Pd thickness and substrate oxidation mechanism

X-ray spectra of Pd/Ni-W samples subjected to annealing process at two different temperatures in 10 mTorr oxygen pressure for 30 minutes are reported in Figure 1. The thicknesses of Pd films is ranging from 0 to 100 nm. In the case of the uncoated substrate, the formation of a NiO layer for both the annealing temperatures, 600°C and 800°C, is observed.

At 600°C, Figure 1a, the complete interdiffusion between the Pd film and the substrate occurs for a Pd film thickness of 15 nm, and the formation of NiO as well as of NiWO$_4$ in the substrate is observed. In the case of thicker Pd films, a part of the Pd film resists against the interdiffusion and the NiWO$_4$ phase is not detectable.

At the higher annealing temperature, 800°C, Figure 1b, even the thicker Pd layer interdiffuses almost entirely in the substrate, and oxide phases of Ni and of Ni-W coexist. In Table 1, the integrated

Figure 1. XRD spectra acquired on Ni-W tapes coated with a Pd layer of thickness 1) 100 nm, 2) 50 nm, 3) 15 nm and 4) without the Pd layer. Samples were annealed in a low oxygen pressure (10 mTorr), at temperature of: (a) 600°C and (b) 800°C. Symbols * and § indicate features related to the interdiffusion process and to the substrate, respectively [5]. Dashed lines mark NiWO$_4$ stronger peaks, continuous lines mark NiO peaks.
intensities of the diffraction peaks of NiO and NiWO$_4$ are listed. The greater the thickness of the Pd layer, the easier the formation of NiWO$_4$. This reveals that an increasing amount of W is involved in the oxidation process. It is possible to speculate that the Pd presence in the Ni-W matrix, promotes the W migration in a region where the oxidation is easier, e.g. on the surface, or even favors the oxygen diffusion inside the substrate, for example due to an increase of the lattice disorder.

Thermo-Gravimetric analyses, TGA, performed in air on the bare substrate and on the Pd coated substrate are reported in Figure 2. The sample with the Pd coating shows an increase mass higher if compared with the uncoated one. Moreover, the cross sections, analyzed by SEM, reveal different thicknesses for the oxide penetration in the bare and in the covered substrate, of about 5 and 10 $\mu$m, respectively. This remarkable thickness difference together with TGA results confirm the greater ease to absorb oxygen for the coated sample.

### 3.2. Pd thickness and coated conductors properties

In Figure 3a, the XRD spectra of CeO$_2$/YSZ/CeO$_2$ buffer layer structures deposited on Pd/Ni-W substrates, with different thickness of the Pd layer, are reported. The intense (00$\ell$) peaks reveal a good epitaxial growth for each layer, with the [00$\ell$] direction perpendicular to the substrate surface. As shown in Figure 4 panel a) for the sample grown on a 20 nm thick Pd layer, despite the oxidation and the interdiffusion processes, the buffer layer structures surfaces are smooth and continuous.

In the Figure 3b the spectrum for the YBCO sample deposited on the buffer layer structure with a 110 nm Pd over layer. The YBCO film is completely c-axis oriented and a full width at half maximum (FWHM) for the (006) YBCO peaks $\omega$-scan of about 2.4° in the rolling direction was

| sample | Pd (nm) | NiO   | NiWO$_4$ |
|--------|--------|-------|----------|
| 1      | 100    | 40.2  | 96.2     |
| 2      | 50     | 165.4 | 52.1     |
| 3      | 15     | 312.7 | 37.4     |
| 4      | 0      | 103.7 | 0        |

Table 1 Integrated intensity of NiO and NiWO$_4$ XRD peaks for Pd/Ni-W samples annealed at 800°C in 10mTorr of O$_2$ atmosphere.

Figure 2 Mass gain profile and temperature, from in air TGA test on an uncoated Ni-W substrate and on a 200 nm thick Pd buffered substrate.

Figure 3. a): XRD spectra for CeO$_2$/YSZ/CeO$_2$ buffer layer structure deposited on Ni-W tape coated with a Pd layer of thickness (1) 165 nm, (2) 110 nm and (3) 20 nm. Marks are the same as figure 1. b): XRD spectrum for the YBCO film deposited on CeO$_2$/YSZ/CeO$_2$ buffer layer structure with a 110 nm Pd over layer.
detected, indicating a sharp out-of-plane orientation. The morphological characterization of the YBCO surface, Figure 4b, reveals that the film is compact and dense. Some features typical of the YBCO film grown by PLD, like droplets and out-growth, are present. Similar characteristics are obtained in the YBCO films grown exploiting Pd layers of about 20 and 165 nm thick.

Regardless of the thickness of the Pd film, the dependence of the electrical resistance with the temperature shows similar behavior, as reported in Figure 5. YBCO films exhibit a narrow superconducting transition and zero resistance critical temperature values, $T_C$, of about 89 K. As a result, Pd/Ni-W structures with a Pd layer 20 nm thick, have been validate as suitable for the development of the YBCO based coated conductor.

4. Conclusions

The role of the Pd layer thickness on the substrate oxidation process in the case of the Pd/Ni-W structure has been investigated. Results show that if a complete Pd-Ni interdiffusion occurs an easier NiWO$_4$ formation is observed. Besides, the oxidation process is progressively enhanced as the thickness of the Pd layer increases. However, at temperature as low as those used for the deposition of the first layers of the buffer structure, 500°C, a part of the Pd film is not involved by the interdiffusion. As a result, the epitaxial growth of a high quality buffer structure and YBCO film is allowed even exploiting a Pd layer with thickness as low as 20 nm.

References

[1] He Q et al. 1997 Physica C 275 155–61.
[2] Celentano G et al. 2005 IEEE Trans. Appl. Supercond. 15 2691–4.
[3] Ciontea L et al. 2008 Journal of Physics: Conference Series 97 012302.
[4] Vannozi A et al. 2011 IEEE Trans. Appl. Supercond. 21 2908-11.
[5] Mancini A et al. 2008 Supercond. Sci. Technol. 21 015003.
[6] Je J H et al. 2002 Physica C 383 241–6.
[7] Mancini A et al. 2011 IEEE Trans. Appl. Supercond. 21 2891-11.
[8] Varesi E et al. 2002 Physica C 372-376 763.
[9] Varesi E et al. 2003 Supercond. Sci. Technol. 16 498-505.