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Pulsed Electron Deposition (PED) of Single Buffer Layer for “low-cost” YBCO Coated Conductors

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Abstract. The challenge for the commercialization of YBCO Coated Conductors (CC) is the development of a low cost manufacturing process to allow for a cheap, fast and continuous deposition of superconducting coatings with high electrical performance. We are currently investigating 2 ways to reduce the CC production costs: i) reducing the complexity of the CC architecture, by growing a single buffer layer based on doped CeO2, and ii) utilizing a new reel-to-reel apparatus for long length CC processing, equipped with a cheap and reliable deposition system (PED, Pulsed Electron Deposition).

In this work we report on the successful continuous deposition of very thick (up to 700 nm) doped-CeO2 single buffer layers on biaxially textured Ni–5at%W substrates by PED. XRD patterns display complete orientation and very good texture quality of our samples (FWHM out-of-plane values of ≈ 6°), over 20 cm length. Optical and electron microscopy show a dense and crack-free film surface and dielectric strength measurement confirms excellent insulating properties. Preliminary results indicate that the simplified single buffer layer structure could be a reliable solution for the reduction of HTS CC production costs.

1. Introduction

The technology for the production of 2nd generation HTS (YBCO-based) Coated Conductors (CC) is close to its maturity stage. Nowadays, high performance CC are available; recently, it has been reported a new record value of 102.935 A-m (Ic=173 A/cm over 595 m) [1]. However, their structural complexity and the production costs are crucial issues that prevent a wide market penetration.

In this work, we have studied possible solutions, focusing our activities on: 1) demonstrating the feasibility of a simple CC architecture, characterized by a single Buffer Layer (BL) structure (based on doped CeO2), and 2) using a simple and economic deposition process (PED, Pulsed Electron Deposition), suitable for long length processes.
There is an increasing interest towards CC with a single CeO$_2$ BL architecture, owing to the simplicity and cost effectiveness. The thickness of the BL should be high enough to ensure an efficient barrier effect, preventing the atomic diffusion between the metallic substrate and the conducting layer; by using textured RABiTS Ni-based tapes, having a lattice mismatch with CeO$_2$ of 8.9%, an estimation of the critical thickness ($t_c$, beyond which CeO$_2$ releases the elastic energy by creating defects and cracks) is $<150$ nm [2]. This is why it’s very difficult to deposit single CeO$_2$ BL, and why more complex and costly multi-buffer-layers architectures are usually grown.

We have recently reported [3, 4], the successful deposition of thick and crack-free single BL for CC fabrication, based on doped-CeO$_2$; by partially substituting Ce with different iso- or hetero-valent cations, having smaller ionic radius (Zr$^{4+}$, Sm$^{3+}$, Yb$^{3+}$), the lattice mismatch vs. Ni alloy substrate is reduced, leading to a significant increase of the BL critical thickness up to 280 nm. In addition, the doping increases the material fracture toughness, as demonstrated by dielectric strength measurements. RBS (Rutherford Back-Scattering) analysis shows the effectiveness of the single buffer layer as ionic diffusion barrier. XRD confirmed a complete doped-CeO$_2$ orientation and an excellent texture quality (FWHM in-plane and out-of-plane values of about 6°). YBCO films deposited by thermal co-evaporation on Yb-doped CeO$_2$ exhibit good transport properties: sharp $T_c$ transition ($T_{c0} = 89.3$ K) and $J_c = 0.9$ MA/cm$^2$ at 77 K (self field).

Concerning the choice of the deposition technique, several methods are currently under development, but it is not clear yet which one allows the best compromise between high performance CC and low production costs. Thick and crack-free single doped CeO$_2$ BL reported in ref. [3, 4] have been deposited by thermal and e-gun evaporation; however, these deposition techniques do not seem to be suitable for long length processes, due to problems in maintaining stable deposition conditions over long terms, contamination of the vacuum chamber, difficult control of the stoichiometry transferred from the target to the substrate, limited deposition rate, etc.

Pulsed Electron Deposition (PED) is a relatively new system that is attracting a great attention for its simplicity, reduced investment cost and flexibility. This technique, also known as Pseudo-Spark Discharge, Channel-Spark Discharge and Pulsed Plasma Deposition, is based on a pulsed high power electron beam, created in a low pressure gas discharge. The electron beam has typical values of current $>1$kA, energy $<15$keV, maximum discharge voltage ~25kV and power density exceeding $10^8$ W/cm$^2$; it impacts the target (penetration depth of 1-2 μm) producing a fast (~100 ns) ablation, leading to a non-equilibrium heating that preserves the stoichiometric material transfer from the target to the substrate, because of congruent evaporation.

A detailed review on the principles and applications of this technique are reported in reference [5].

2. Deposition of Single BL by PED

In order to test the efficiency of the PED system, we repeated some of the successful depositions obtained by using the e-gun technique (i.e., Yb and Zr doped CeO$_2$). In addition, we selected Ta as a new dopant, for its small ionic radius (to reduce the CeO$_2$ lattice mismatch with the Ni-based substrate) and to study the effect of a higher valence dopant (Ta$^{5+}$) [6]. As already pointed out, the limited reduction in the lattice mismatch alone, cannot account for the large increase in the doped CeO$_2$ $t_c$, suggesting that the type and amount of dopant certainly play a role in making the BL structure more resistant towards crack formation.

The targets are prepared by mixing the oxides in the stoichiometric ratios; the powders, finely ground in a ball mill, are pressed in pellets with 50–60 bars (5 minutes at ambient temperature) and sintered at 1300 °C for 8 hours. Biaxially textured Ni-5at.%W alloy (RABiTS, from evico GmbH) are used as substrates, cut in 2 cm long pieces and annealed in 10 mbar of forming gas (Ar –5% H$_2$) at 700 °C for 60 minutes. Subsequently, doped layers are deposited using a commercial PED source (Organic Spintronics, Bologna, Italy).

Depending on the different targets, the experiments are performed with a source voltage = 14-18 kV, and pulse frequency = 4 Hz. PED operates with O$_2$ internal pressure as discharge ignition gas.
Deposition conditions are: $O_2$ Pressure = $1 \times 10^{-4}$ mbar, Temperature = 650–750 °C. The post-growth treatment is carried out for 10 minutes, under the same thermodynamic conditions (closed shutter). The calculated deposition rate is $5.5 \AA$/sec (1.4 \AA/pulse), similar for the different target compositions. X-ray diffraction (θ-2θ geometry) shows that all the doped CeO$_2$ samples are highly oriented and with no secondary phases (figure 1); the degree of (200) out-of-plane orientation, $C_{(200)}$, is given by the following formula:

$$C_{(200)} = 100 \frac{I_{200} / W_{200}}{(I_{200} / W_{200} + I_{111} / W_{111})}$$

(1)

Where $I_{hkl}$ is the measured intensity of the (hkl) reflection in the 0-2θ pattern of CeO$_2$ films, and $W_{hkl}$ represents the tabulated intensity for the (hkl) reflection of ceria powders [7]. The $C_{(200)}$ is $> 99\%$ in all the studied samples and a good texture quality is confirmed by the rocking curves (FWHM values = 5.2 – 6.6°).

![Figure 1. X-ray diffraction pattern of Zr (15%) doped CeO$_2$/Ni-5at.%W (left) and corresponding (200) reflection rocking curve (right).](image)

The film thickness has been measured by cross section SEM analysis (figure 2). The measured values agree with the calculated ones, that can be derived from the thin film colour, by using the interference condition; this simple method allow to determine, with the accuracy of ±20nm, the thickness of the growing film simply by optical inspection.

![Figure 2. Cross section SEM image of Zr (15%) doped CeO$_2$/Ni-5at.%W. Film thickness is 500 nm.](image)

It is not possible to perform a precise thickness measurement by step-profiler, because of significant film surface roughness; SEM (Philips 515) analysis shows that the film surface is covered of sub-micro inclusions (figure 3). The presence of particulates on the surface of film grown by PED is well known [8], and the possible solutions to prevent/minimize their formation is currently under study.
A remarkable result from SEM analysis is that all the samples are crack-free, in spite of the film thickness (> 700 nm). This implies that the mechanical properties of the doped-CeO$_2$ are dramatically different compared to the undoped-CeO$_2$. Further studies need to be done, in order to understand the physical/chemical properties of the doped CeO$_2$ that make the material so robust; the presence of some kind of defects (inclusions?) might play a role in relaxing the stress in other ways instead of cracking.

It is worthwhile to note that doped-CeO$_2$ indeed is very tough; the measured dielectric strength at 77 K of thick (Yb, Zr and Ta) doped CeO$_2$ is found to be for all samples > $4 \times 10^6$ V/cm, more than one order of magnitude higher than the value measured under the same conditions on crack-free undoped-CeO$_2$ (100 nm thick).

In table 1 the main characteristics of the studied samples are summarized and compared to undoped-CeO$_2$:

| Material        | Critical thickness $t_c$ (nm) | Out-of-plane (200) orientation degree, $C_{(200)}$ (%) | Out-of-plane (200) $\Delta \omega$ FWHM (°) | Dielectric strength (MV/cm) | Mismatch with Ni-W substrate (%) |
|-----------------|------------------------------|------------------------------------------------------|-----------------------------------------------|------------------------------|---------------------------------|
| (Yb-45%)-CeO$_2$ | > 700                        | 99.8                                                 | 5.2                                           | 5.0                          | 7.8                             |
| (Zr-15%)-CeO$_2$ | > 500                        | 99.7                                                 | 6.1                                           | >5.9                         | 8.0                             |
| (Ta-10%)-CeO$_2$ | > 550                        | 99.6                                                 | 6.6                                           | 4.1                          | 8.1                             |
| Undoped-CeO$_2$ | \(\pm 100\)                 | \(\pm 100\)                                          | 6.0                                           | 0.3                          | 8.9                             |

3. Deposition of long-length Single BL by PED

To scale up the fabrication of longer length tapes, a PED system has been recently installed in a reel to reel apparatus, available at IMEM. Details of the apparatus have been reported in Ref. [3]. Although it was designed for thermal evaporation, the PED system has been easily allocated in the central (deposition) chamber (see figure 4).
The continuous (dynamic) depositions have been performed in the same way as the static ones. Instead of small pieces of Ni-5% RABiTS tapes placed on the heater, a continuous tape has been mounted in a reel-to-reel apparatus, and moved back and forth over the deposition zone at a velocity of 10 mm/min. Preliminary results on dynamic depositions are very encouraging; doped CeO$_2$ (Zr 15%) buffered Ni-5%W (RABiTS) substrates tapes have been easily produced (figure 5), with a high degree of reproducibility and uniformity in term of thickness, surface roughness and orientation.

![Figure 5. Zr (15%) doped CeO$_2$ buffered Ni-5%W RABiTS tape, grown by PED](image)

To check the tape homogeneity, short samples have been cut and characterized every 5 cm; XRD analysis shows that the (200) orientation degree is 99±0.5% along the 20 cm-long tape, and the texture quality (tested by rocking curves) gives a strong out-of-plane orientation (FWHM values = 6.5 ±0.5°), with values similar to those obtained for short length, static depositions (figure 6).

![Figure 6. CeO$_2$(Zr 15%)/Ni-5at.%W orientation and texture (out-of-plane) along 20 cm long tape (XRD data)](image)

Interestingly, pole figures of dynamic Zr(15%) doped-CeO$_2$ processed samples are better than those of static ones grown under similar conditions. While these latter exhibit a triple in-plane orientation (along the [110], [470] and [740] directions), the dynamic samples show a good (001)-[110] biaxial texture, resulting in FWHM in-plane and out-of-plane values of 9.8° and 7.0° respectively (figure 7).

![Figure 7. Texture analysis of static (left) and dynamic (right) CeO$_2$(Zr 15%)/Ni-5at.%W by pole figure.](image)

Furthermore, the longer length depositions seem to improve the surface quality of the film; this might be explained as a “dilution” over long length of particles from the target (in our system the target is placed above the moving tape).
It's also remarkable to note that several depositions have been made using the same target (CeO$_2$ – ZrO$_2$ 15%); its size was 18 mm (diameter) x 2 mm (thickness). The target has been used during 4 different processes, for more than 20 hours of deposition time (corresponding to about $3 \times 10^5$ pulses), without exhibiting significant damages on the surface. It confirms that the PED could be a suitable technique for long length (long time) depositions; moreover, the deposition process has been constant and regular (pulse frequency, shape of the ablated plume), leading to a stable deposition rate during long time. Finally, the high material transfer rate between target and substrate and the strongly focused geometry, make the PED a “clean” system; no contaminations, depositions or dust are visible in the vacuum chamber after several tents of operation hours, being the quartz/glass tube (to accelerate the electron beam) the only part that requires a periodical cleaning.

The optimization of the conditions for the YBCO deposition by PED is in progress. YBCO has been one of the first materials to be deposited using a PED system [9, 10] with very good properties, comparable to film grown by PLD [11, 12].

In conclusion, having in mind a necessary reduction of the HTS CC production costs, the preliminary results here reported demonstrate the possibility to fabricate a simple architecture (single BL) with a cheap technique (PED). Very thick (> 700 nm), and crack-free doped CeO$_2$ (dopant: Yb, Ta, Zr) have been grown with good structural and dielectric properties onto 2 cm-long RABiTS Ni-5%W tapes. The deposition of longer tapes (20 cm) has been easily performed by using a reel-to-reel system, confirming the stability and the reproducibility of PED during long time deposition processes; the tape movement, while keeping constant the film thickness and orientation, seems to improve other properties like texture and surface roughness.

Therefore, owing to the flexibility of the deposition technique, a very low cost CC fabrication process, entirely based on PED system, could be realizable; the scale-up to longer length or higher deposition rate could be done by installing in the vacuum chamber a linear array of pulsed electron guns, as reported in Ref. [5].

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