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COMPARISON BETWEEN CeO_2 BUFFER LAYER FILM FORMED BY CHEMICAL SOLUTION DEPOSITION IN THE PRESENCE OF DIFFERENT ORGANIC ADDITIVES

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ABSTRACT

Chemical solution deposition (CSD) derived cerium oxide (CeO_2) films were successfully produced on rolling assisted biaxially textured (100) <001> Ni. In here, the surface of the Ni was first treated with NaOH to see the possibility of improving adherence of the solution to the Ni. Furthermore, different organic additives added solution were experimented and the dip coated precursor films were heat treated at 1000 °C for 1 hour under a reducing atmosphere by flowing Ar-5%H_2 gas. Scanning electron microscopy (SEM) analysis showed surface oxide consisting of both solutions added with triton X-100 and polyvinyl alcohol (PVA) respectively were able to improve the quality of films. A denser film can be produced by adding glacial acetic acid in the triton-X 100 added solution. On the other hand, triethanolamine (TEA) did not help in producing a good coverage and smooth surface of CeO_2 film. Instead the films were cracked and parts of them delaminated from the substrate. X-ray diffraction (XRD) pattern indicated that both (111) and (200) CeO_2 were formed. Raman spectra analysis confirmed the formation of pure phase CeO_2.

INTRODUCTION

Significant research has been devoted to high temperature superconducting tapes based on the second-generation coated conductors. This is due to their high critical current density and intrinsically high irreversibility field displayed by the conductors. Coated conductors are consisted of $YBa_2Cu_3O_{7-x}$ (YBCO) superconducting film on a thin buffer layer deposited on a textured metallic substrate [1-4]. The biaxially textured substrates can be achieved by rolling of a Ni foil followed by recrystallisation. Such process is termed Rolling Assisted Biaxially Textured Substrate (RABiTS) process. RABiTS tapes have a (100) <001> surface; often onto this RABiTS tape, 70-100nm thick buffer layer made from oxide is deposited. CeO_2 is one of the most commonly used buffer layer for YBCO film. CeO_2 has a compatible thermal coefficient, matching lattice parameter with YBCO, as well as chemically compatible [3]. CeO_2 buffer layer is necessary as chemical barrier between the YBCO layer and the Ni substrate. It is also to reduce lattice mismatch between Ni and YBCO and more importantly it acts as a template for the epitaxial growth of YBCO. Morever, CeO_2 provides a corrosion resistance barrier for Ni. To date, CeO_2 has been grown by various physical and

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chemical methods. CSD has been considered as the most promising route to produce CeO_2 films since it is a low cost non-vacuum technique for commercial production of YBCO coated conductor. The work of perfecting the chemical precursor for CeO_2 film formation is challenging as the adherence of the solution onto the Ni substrate is not very good. This leads to shrinkage and subsequent cracking of the film. In this paper, we have studied on the effect of NaOH modified Ni surface and the effect of various organic additives in Ce-precursor solution to improve the wet ability of the solution on the Ni surface. Our aim is to a produce a process which produces film with good coverage, does not shrink, smooth and uniform throughout.

METHODOLOGY

High purity cold-rolled (100) <001> Ni based RABiTS tape with a rolling reduction of about 70 % were supplied by Karlsruhe Institute of Technology, Germany [5]. The tape was cut into dimension of 0.5 cm x 1 cm and was cleaned in an ultrasonic cleaner using acetone for 15 minutes before deposition. The substrate was treated with 0.3M NaOH which was heated to 80 ^{0}C. Cerium precursor solution was prepared by dissolving cerium (III) nitrate hexahydrate, $Ce(NO_3)_3.6H_2O$ (MERCK, 227371) with distilled water. The concentration of solution was adjusted to 0.25M. Then solution was stirred at room temperature for at least 30 minutes until it became fully dissolved and transparent solution. To further improve the adhesion and the wet ability of precursor film on Ni substrate, four main organic additives; triethanolamine (MERCK 822341), triton X-100 (FLUKA 93420), glacial acetic acid (MERCK 100083) and polyvinyl alcohol (MERCK S25443931) were experimented. Different volume percent and weight percent of additives were used. The CeO_2 films were dip coated on Ni substrate with withdrawing speed of 2cm/min whilst keeing it in the solution for 10s. CeO_2 precursor films were then dried at 100 ^{0}C for 15 min and annealed in the furnace under 5%H_2–Ar gas flow for 1H to inhibit the oxidation of Ni substrate. Microstructures and surface morphologies of the CeO_2 thin films were analyzed by using ZEISS SUPRA 35VP field emission scanning electron microscope (FESEM) equipped with an energy dispersive X-Ray (EDX) analyzer. The phase formed was investigated by Siemen D 5000 X-ray diffraction (XRD) using Cu-K_α radiation. Jobin Yvon HR 800 UV Raman spectroscopy was also used.

RESULTS AND DISCUSSION

In Figure 1(a), we compared the XRD patterns of CeO_2 films grown on Ni RABiTS treated with and without NaOH. As seen, when the Ni was not treated with NaOH, the diffraction pattern has no peak. On the other hand, for Ni RABiTS that was treated with NaOH prior to dip coating, it was found that both (111) and (200) CeO_2 peaks were detected. NaOH helps with the formation of CeO_2.

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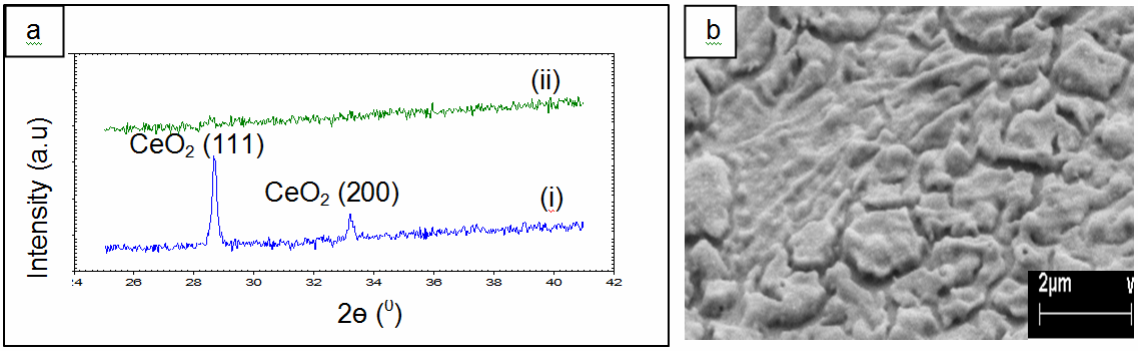

Figure 1: (a) XRD patterns for coating on Ni RABiTS treated (i)with and (ii)without NaOH. (annealed at 1000℃ for 1H in Ar-5%H_2), (b) SEM micrograph of coating as in (i).

Different types of organic additives were then added in the solution to see if the surface of film could be improved. The first additive used was a non-ionic TEA which should in theory reduce the surface tension of the liquid hence improving the wet ability. However as seen in Figure 2(a), the morphology of the coating has became worse by the addition of TEA. There is the possibility that the amine group in the TEA may have reacted with the precursor solution, hindering polymerization of the metal ions leading to severe cracking of the coating. Figure 2(b) and (c) are representative images of coatings made using solution with the addition of triton X-100 without (b) and with (c) addition of glacial acetic acid. As seen in both figures, the coatings are more uniform with better coverage with glacial acetic acid added (Figure 2(c)). Acetic acid is a chelating agent which can control metal ions in aqueous system and to form stable water soluble complexes with multivalent metal ions. The chelation process helps in the formation of good adherence of film and thus denser surface of film can be achieved. Pores were due to the decomposition of organic additives after pyrolysis during annealing process. The fourth solution was prepared with PVA addition. PVA also showed a relatively good surface quality as showed in Figure 2(d). This is because PVA has excellent film forming and adhesive properties which help in the coating of film. The overall surface of CeO_2 film was smooth and crack free from solution added with PVA, only some thicker part of film was cracking. However, the difficulty in using PVA is that it is very difficult to dissolve the PVA.

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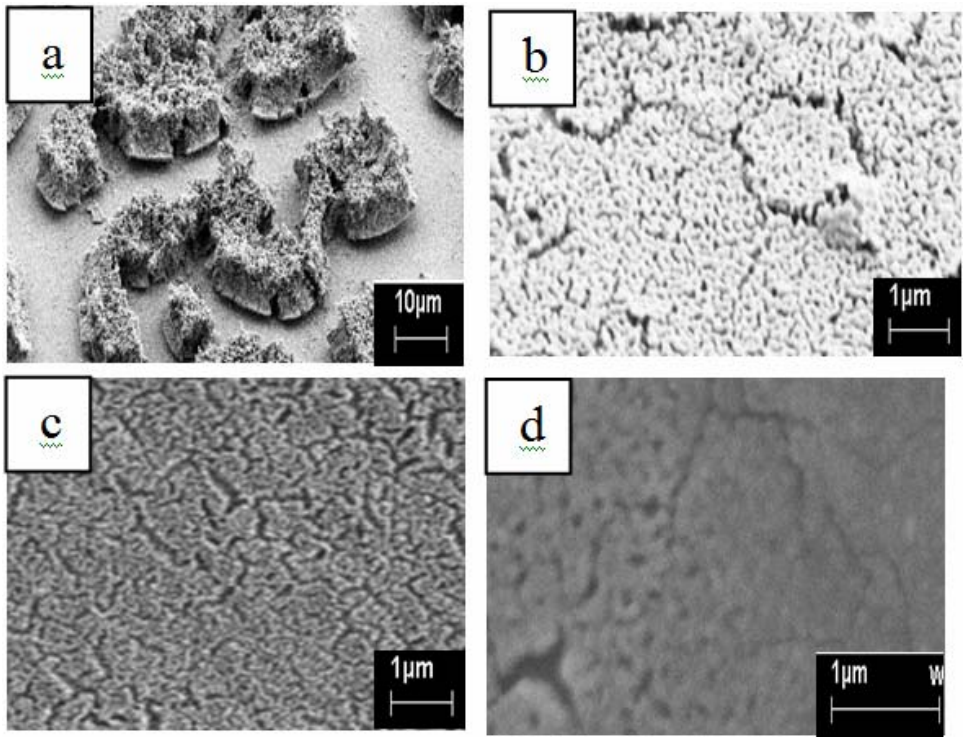

Figure 2: SEM micrographs of CeO_2 layer produced from 0.25M $Ce(NO_3)_3.6H_2O$ added with (a) 5 vol% TEA; (b) 5 vol% triton X-100; (c) 5 vol%triton X-100 plus 5 vol% glacial acetic acid; (d) 36.85 wt % PVA (All foils were annealed at 1000℃ for 1H in 5%H_2–Ar).

XRD patterns of all four samples are shown in Figure 3(a). All coatings showed a preferential (111) and (200) CeO_2 orientation at 2ɵ ~ 28° and 33° respectively. Further study in annealing treatment is however ongoing to seek the possibility of (200) CeO_2 formation. The fact that epitaxial growth cannot be achieved in this work could be due to the NaOH addition. The increase in the thickness may destroy the epitaxial relationship. Figure 3(b) presented a Raman spectrum with sharp peak at frequency 465cm^{-1}.This representative spectrum confirmed the formation of pure CeO_2 phase for annealed film. The Raman-active mode in CeO_2 thin film corresponds to the frequency of ω_R = 466 cm^{-1}, which is attributed to a symmetrical stretching mode of the $Ce–O_8$ vibrational unit [9]. Furthermore, there are no other peaks visible from the whole frequency range which indicated that NaOH layer does not react with the additives to form undesired phases besides CeO_2 phase.

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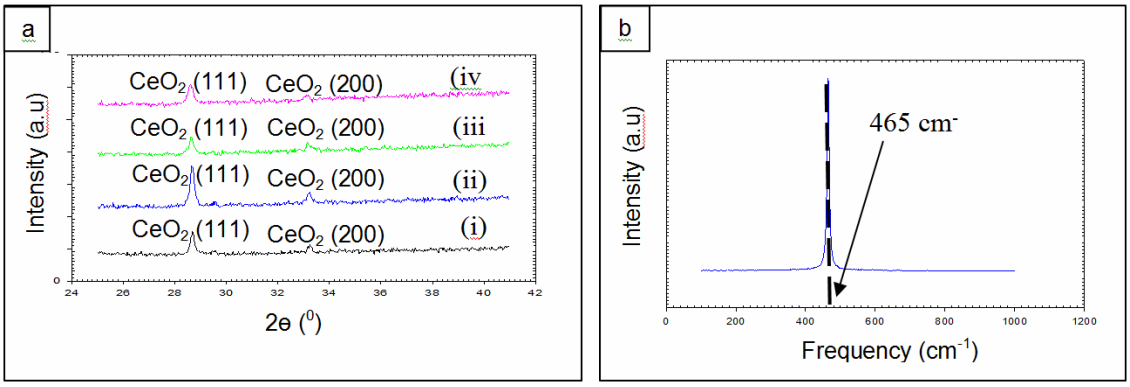

Figure 3: (a) XRD spectrum for annealed films produced from solution added with (i) triton X-100; (ii)triton X-100 plus glacial acetic acid; (iii) TEA; and (iv)PVA respectively. (b) Raman spectra of coating as in (i)(annealed at 1000℃ for 1H in 5%H_2–Ar)

CONCLUSION

This study has analyzed the effect of NaOH treated Ni surface as well as various organic additives on the quality of coating produced in the Ce-based nitrate precursor. CSD through dip coating was chosen for the deposition of CeO_2. NaOH treated surface has helped in the formation of CeO_2 phase. However NaOH layer increased the thickness of annealed CeO_2 film and disturbing the epitaxial growth of the film. A denser film with no cracking can be produced by adding 5 vol% triton X-100 and acetic acid in the Ce-precursor. PVA was also a good additive while TEA added solution formed film with severe cracking.

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