

EPITAXIAL GROWTH OF Yb_2O_3 BUFFER LAYERS ON BIAXIALLY TEXTURED-Ni (100) SUBSTRATES BY SOL-GEL PROCESS

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ABSTRACT

In order to develop an alternative buffer layer architecture using the sol-gel process to produce YBCO ($\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$) coated conductors, Yb_2O_3 has been chosen as the candidate material. Buffer layers of Yb_2O_3 were epitaxially grown on biaxially textured-Ni (100) substrates by the sol gel process for the first time. The Yb_2O_3 precursor solution was prepared from an alkoxide sol-gel route in 2-methoxyethanol and was deposited on textured-Ni (100) substrates by either spin coating or dip coating methods. The amorphous film was then processed at 1160°C under flowing (96%)Ar/ H_2 (4%) gas mixture for one hour. The Yb_2O_3 film exhibited a strong c-axis orientation on the Ni (100) substrates. The phi and omega scans indicated good in plane and out of plane orientations. The X-ray (222) pole figure showed a cube-on-cube epitaxy. High current YBCO films were grown on the Yb_2O_3 sol-gel buffered-Ni substrates.

INTRODUCTION

The RABiTS (Rolling-Assisted Biaxially Textured Substrates) approach developed at the Oak Ridge National Laboratory (ORNL) has emerged as one of the leading techniques for the fabrication of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO) coated conductors [1,2] in the development of superconducting wires for high temperature, high-field applications. The process involves obtaining cube textured Ni (100) substrates by cold-rolling Ni rods followed by recrystallization. The buffer layers are then epitaxially grown on the textured metal substrates followed by the deposition of in-plane oriented superconducting films. Critical current densities of YBCO films over 1 MA/cm² at 77 K have been demonstrated on rare-earth oxide (RE_2O_3) buffer layers with various architectures via vacuum process [3,4]. The goal of this research is to develop the RABiTS process using a non-vacuum approach such as the sol-gel or the MOD processes.

The non-vacuum approach has many advantages. The techniques are very cost-effective and easily scalable. The solution process gives better homogeneity and composition control because the precursors are mixed at atomic levels. Substrates with various shapes and forms can be coated with ease by spin coating or dip coating methods.

Previously, buffer layers prepared by the sol-gel technique showed epitaxial growth on single crystal substrates [5-9], but resulted in multiple orientations on textured-Ni (100) substrates [10-12]. Beach et al. at ORNL recently demonstrated the epitaxial growth of a rare-earth oxide, Gd_2O_3 , buffer layer on textured-Ni (100) substrate by sol-gel process for the first time [13]. The Gd_2O_3 precursor solution was prepared via an all alkoxide sol-gel route. The textured film had a continuous and a dense microstructure without any cracks.

In this paper, we report the preparation of the Yb_2O_3 precursor solution, film deposition and characterization of the sol-gel buffer layer on textured-Ni (100) substrate for the first time.

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We also demonstrate the deposition of a high J_c YBCO film on a sol-gel grown buffer layer with sputtered cap layers, YSZ and CeO_2 .

EXPERIMENTAL

The solution preparation was carried out under Ar atmosphere using a Schlenk-type apparatus. The isopropanol was dried by distillation from aluminum isopropoxide. The ytterbium ingot and 2-methoxy ethanol (Alfa) were used without further purification.

The flow chart of the ytterbium precursor preparation is shown in Figure 1. The first step involved the preparation of ytterbium isopropoxide from ytterbium metal. This was achieved by reacting the ytterbium metal filings with dry isopropanol in the presence of mercuric catalyst. The ytterbium isopropoxide was extracted using a Soxhlet extractor with isopropanol and recrystallized to obtain a highly pure final product.

The second step involved the exchange of the isopropoxide ligand for a methoxyethoxide ligand. About 1.66g (6.25 mmoles) of ytterbium isopropoxide was weighed out in a 200 ml round bottom flask and dissolved in 50 ml of 2-methoxyethanol. After the solution was refluxed for one hour, approximately 30 ml of the isopropanol and 2-methoxy ethanol solvent mixture was removed by distillation. The solution was rediluted with 2-methoxyethanol and distilled repeatedly to ensure complete ligand exchange. Finally, the volume of the solution was adjusted with 2-methoxy ethanol to 25 ml to make a 0.25 M Yb_2O_3 precursor solution.

The precursor solution was partially hydrolyzed by adding approximately one part of 1.0 M water in 2-methoxyethanol to four parts of the stock solution. The solution was allowed to age overnight at room temperature prior to coating. Part of the solution was fully hydrolyzed to obtain gels. The gels were dried on a hot plate and then fired at 900°C in air for one hour to obtain the powder for phase identification.

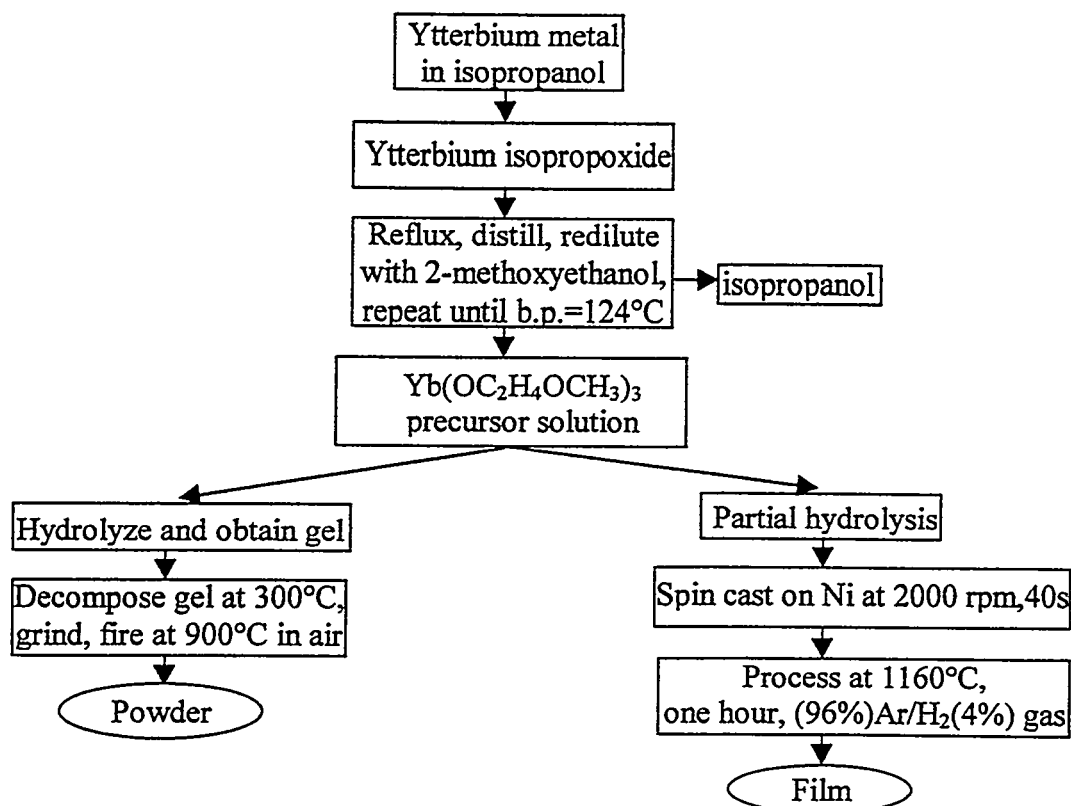


Figure 1. Flow chart of the Yb_2O_3 precursor solution preparation.

The Yb_2O_3 precursor solution was spin coated onto textured-Ni substrates. The (100) cube textured-Ni substrates were prepared by annealing the cold rolled Ni substrates to recrystallize at 900°C for 2 hours under vacuum. The Ni substrates were cleaned ultrasonically in isopropanol for 1 hour prior to coating. The coatings were applied with a spin coater operated at 2000 rpm for 40 seconds. The samples were annealed in a preheated oven at 1160°C for one hour under flowing (96%) $\text{Ar}/\text{H}_2(4\%)$ gas and then quenched to room temperature.

The Yb_2O_3 films were characterized in detail by X-ray diffraction. Powder diffraction patterns were obtained with a Philips XRG3100 diffractometer with $\text{Cu K}\alpha$ radiation. A Picker four-circle diffractometer was used to determine the texture of the film. The out-of-plane alignment (omega scans) was measured by scanning the (400) planes of the film. The in-plane alignment (phi scans) was determined by measuring the Yb_2O_3 (222) planes. The Yb_2O_3 (222) pole figure was also collected to determine whether the film had a single cube epitaxy. The microstructure of the Yb_2O_3 films was analyzed using a Hitachi S-4100 field emission scanning electron microscope. The beam voltage used was 5 kV. The thickness of the film was determined with an Alpha-Step 500 (Tencor Instruments) profilometer.

RESULTS AND DISCUSSION

Yb_2O_3 Buffer Layer Characterization

The X-ray diffraction pattern of the ytterbium precursor powder fired at 900°C revealed a pure Yb_2O_3 phase. The pseudocubic Yb_2O_3 phase can be indexed with a lattice parameter of 3.69 Å. The lattice mismatch of Yb_2O_3 with YBCO is -3.50% .

The Yb_2O_3 thin film after annealing displayed a golden color. The film thickness of a single coat was determined to be approximately 1500Å . The theta/2-theta scan (Figure 2) of the Yb_2O_3 film showed a strong peak at $34.5^\circ 2\theta$ from the Yb_2O_3 (400) reflection and a very

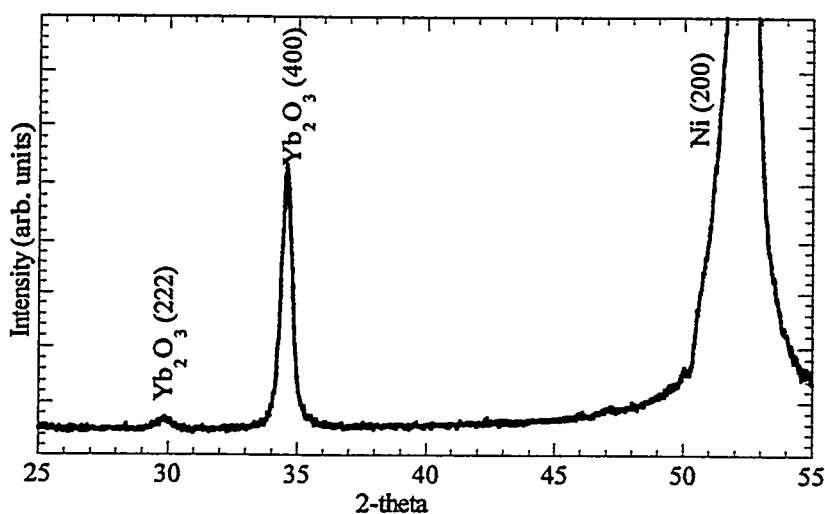


Figure 2. The theta/2-theta scan of the Yb_2O_3 film on Ni (100) substrate shows a strong c-axis orientation.

weak peak at 2θ of 29.5° from the Yb_2O_3 (222) reflection. This indicated a good out-of-plane alignment of the Yb_2O_3 film on the Ni(100) substrate. Figure 3 shows the phi and omega scans of Yb_2O_3 film on textured-Ni (100) substrate. The full-width-at-half-maximum (FWHM) values of Yb_2O_3 (400) and Ni (200) omega scans were 9.89° and 7.06° , respectively.

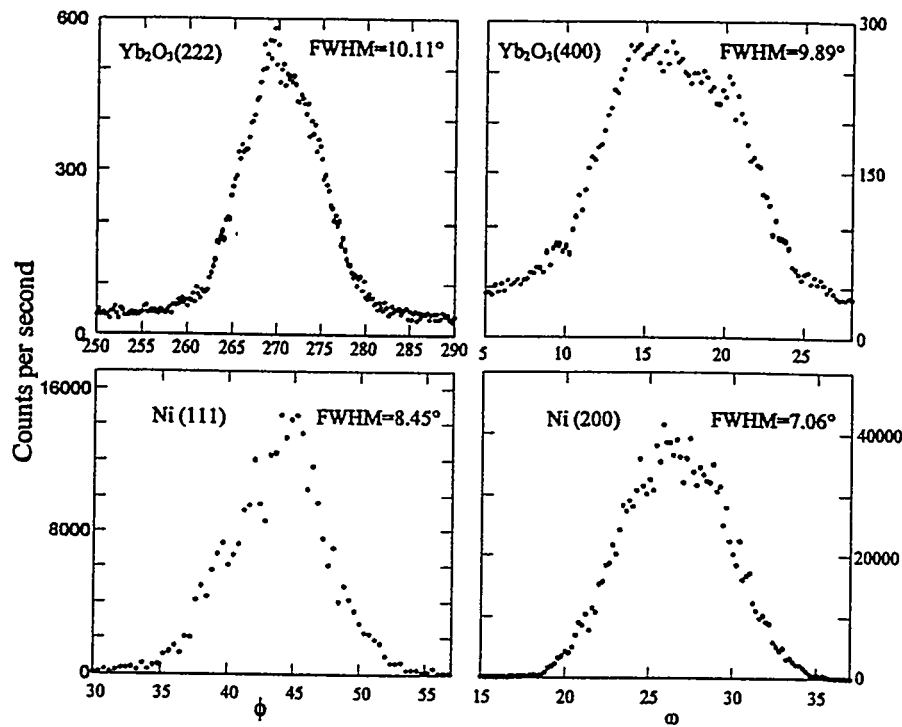


Figure 3. The phi and omega scans of Yb_2O_3 film and Ni (100) substrate.

The FWHM values of the phi scans were 10.11° for Yb_2O_3 (222) plane and 8.45° for Ni (111) plane. The pole figure (Figure 4) of the Yb_2O_3 sol-gel buffer layer on textured-Ni (100) substrate had predominantly a single cube epitaxy. From the phi scans and the pole figure, we can conclude that the Yb_2O_3 film exhibits an excellent in-plane alignment on the Ni (100) substrate.

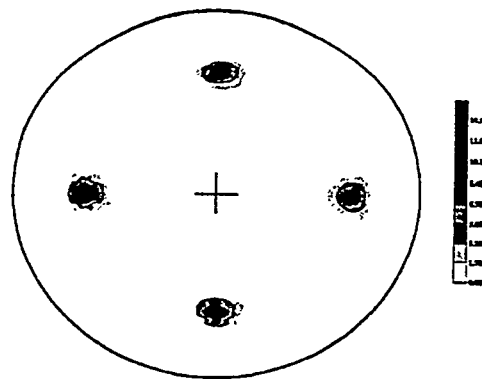


Figure 4. Pole figure of the sol-gel grown Yb_2O_3 film shows a single cube epitaxy on textured-Ni (100) substrate.

The SEM micrograph of the Yb_2O_3 film, shown in Figure 5, was taken at a magnification of 20 kX. The microstructure showed a dense, continuous and a crack free Yb_2O_3 film on the Ni substrate.

YBCO Deposition

With Yb_2O_3 sol-gel buffer layer as the initial layer, YSZ and CeO_2 cap layers were epitaxially grown by sputtering. The YBCO precursor was then deposited on CeO_2 by

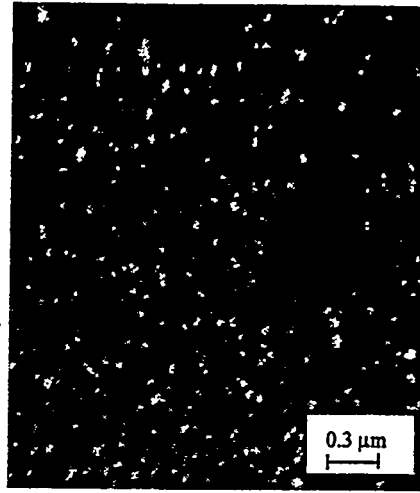


Figure 5. SEM micrograph shows a dense, continuous and a crack free Yb_2O_3 film.

e-beam deposition of BaF_2 , Yttrium metal, Copper metal and subsequently annealed in $\text{H}_2\text{O}/\text{O}_2/\text{Ar}$ atmosphere [14,15]. The RABiTS architecture and the temperature dependent resistivity measurement are shown in Figure 6. The YBCO film had a room temperature resistivity of $380\mu\Omega\text{-cm}$ and zero resistance at 90 K. The transport J_c of the YBCO film was $4.2 \times 10^5 \text{ A/cm}^2$ at 77 K and zero magnetic field. The J_c value decreased to $0.7 \times 10^5 \text{ A/cm}^2$ when a perpendicular magnetic field of 0.5 tesla was applied.

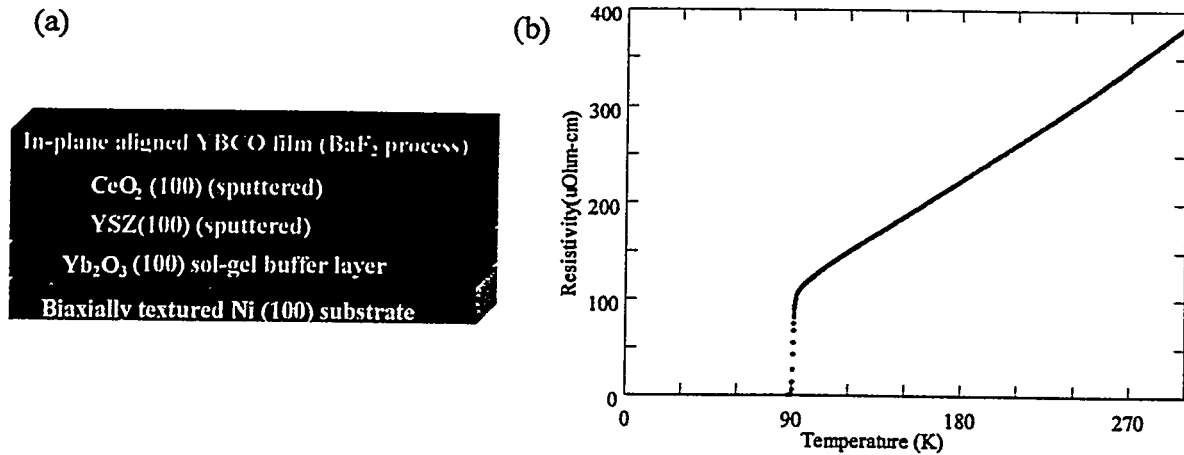


Figure 6. (a) RABiTS architecture with Yb_2O_3 sol-gel initial layer and (b) the resistivity curve.

CONCLUSIONS

We have demonstrated the epitaxial growth of Yb_2O_3 buffer layer on textured-Ni (100) substrates by the sol-gel process for the first time. The X-ray analysis of the buffer layer indicated good in-plane and out-of-plane alignments. The pole figure of the Yb_2O_3 film showed predominantly a single cube texture on Ni. The microstructure of the film showed a dense, continuous and crack free film. Reasonably high J_c of $4.2 \times 10^5 \text{ A/cm}^2$ at 77K and zero field was demonstrated on the RABiTS architecture: YBCO(BaF_2 process)/ CeO_2 (sputtered)/ YSZ (sputtered)/ Yb_2O_3 (sol-gel)/Ni. Future research is directed towards depositing epitaxial sol-gel buffer layers and in-plane superconducting films using only non-vacuum approach.

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REFERENCES

1. A. Goyal, D. P. Norton, J. Budai, M. Paranthaman, E. D. Specht, D. M. Kroeger, D. K. Christen, Q. He, B. Saffin, F. A. List, D. F. Lee, P. M. Martin, C. E. Klaubunde, E. Hatfield, and V. K. Sikka, *Appl. Phys. Lett.*, **69**, p. 1795 (1996).
2. D. P. Norton, A. Goyal, J. D. Budai, D. K. Christen, D. M. Kroeger, E. D. Specht, Q. He, B. Saffin, M. Paranthaman, C. E. Klaubunde, D. F. Lee, B. C. Sales, and F. A. List, *Science*, **274**, p. 755 (1996).
3. M. Paranthaman, D. F. Lee, A. Goyal, E. D. Specht, P. M. Martin, X. Cui, J. E. Mathis, R. Feenstra, D. K. Christen, and D. M. Kroeger, *Supercond. Sci. Technol.*, **12**, (1999) in press.
4. D. F. Lee, M. Paranthaman, J. E. Mathis, A. Goyal, D. M. Kroeger, E. D. Specht, R. K. Williams, F. A. List, P. M. Martin, C. Park, D. P. Norton, and D. K. Christen, *Jpn. J. Appl. Phys.*, **38**, p. L178 (1999).
5. S. S. Shoup, M. Paranthaman, B. B. Beach, E. D. Specht, and R. K. Williams, *J. Mater. Res.*, **12**, p. 1017 (1997).
6. M. Paranthaman, S. S. Shoup, D. B. Beach, R. K. Williams, and E. D. Specht, *Mat. Res. Bull.*, **32** (12), p. 1697 (1997).
7. D. B. Beach, J. S. Morrell, Z. B. Xue, and E. D. Specht, *Mat. Res. Soc. Symp. Proc.*, Fall (1998), in press.
8. J. S. Morrell, Z. B. Xue, E. D. Specht, and D. B. Beach, *Mat. Res. Soc. Symp. Proc.*, Fall (1998) in press.
9. D. B. Beach, C. E. Vallet, M. Paranthaman, E. D. Specht, J. S. Morrell, and Z. B. Xue, *Mat. Res. Soc. Symp. Proc.*, **495**, p. 263 (1998).
10. S. S. Shoup, M. Paranthaman, A. Goyal, E. D. Specht, D. F. Lee, and D. M. Kroeger, *J. Am. Ceram. Soc.*, **81**, p. 3019 (1998).
11. M. Paranthaman, S. S. Shoup, D. B. Beach, J. S. Morrell, A. Goyal, E. D. Specht, J. E. Mathis, D. T. Verebelyi, and D. K. Christen, *Proc. of the 9th CIMTEC-World Ceramics Congress and Forum on New Materials* (1998) in press.
12. A. Singhal, S. S. Shoup, M. Paranthaman, D. B. Beach, E. D. Specht, A. Goyal, D. F. Lee, and J. S. Lin, submitted to *J. Am Ceram. Soc.* (1999).
13. D. B. Beach, J. S. Morrell, M. Paranthaman, T. G. Chirayil, A. Goyal, E. D. Specht, Z. B. Xue, C. E. Vallet, and D. F. Lee, in preparation.
14. R. Feenstra, A. Goyal, D. K. Christen, M. Paranthaman, D. F. Lee, D. T. Verebelyi, E. D. Specht, J. D. Budai, D. P. Norton, and D. M. Kroeger, in preparation.
15. M. Paranthaman, D. F. Lee, R. Feenstra, A. Goyal, D. T. Verebelyi, D. K. Christen, E. D. Specht, F. A. List, P. M. Martin, D. M. Kroeger, Z. F. Ren, W. Li, D. Z. Wang, J. Y. Lao, and J. H. Wang, *IEEE Trans. On Appl. Supercond.* (1999) in press.