Preparation of *c*-axis-oriented Y₂Ba₄Cu₇O_{15-δ} Films by Laser CVD with Ultrasonically Nebulized Precursor

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Abstract. *c*-axis-oriented Y₂Ba₄Cu₇O_{15- δ} (Y247) films were prepared on multilayer-coated Hasterolly tape substrate by laser chemical vapor deposition with ultrasonically nebulized liquid precursor. At a low precursor concentration of 0.01 mol l⁻¹ and deposition temperature of 933 K, single-phase Y247 film with significant *c*-axis orientation was obtained. At a precursor concentration of 0.1 mol l⁻¹ and deposition temperature 983 K, *a*-axis-oriented YBa₂Cu₃O_{7- δ} (Y123) was codeposited with *c*-axis oriented Y247 film.

Introduction

High-temperature superconducting (HTS) oxides are candidate materials for power cables, motors and sensing devices. Among HTS oxides, yttrium-barium-cupper complex oxides including YBa₂Cu₃O_{7- δ} (Y123) [1], YBa₂Cu₄O₈ (Y124) [2] and Y₂Ba₄Cu₇O_{15- δ} (Y247) [3] have been extensively studied for their potential applications. These complex oxides had a layered structure consisting of Cu₂O plane and CuO chain blocks, exhibiting superconduction parallel to *c*-plane.

A long-length HTS wire up to several kilometers is required to meet practical applications. Superconducting layer has often been prepared by pulsed laser deposition [4] and chemical vapor deposition (CVD) [5]. However, their low deposition rate and low yield have been drawbacks for practical production process. In addition, chemical composition and orientation of the superconducting layer should be controlled to acquire a high superconducting performance. We have developed laser CVD [6] and prepared *a*-axis-oriented CeO₂ film for buffer layer [7] and *c*-axis-oriented YBCO film for superconducting layer [8] on multilayer-coated Hastelloy tape substrate at a high deposition rate. Solid precursors have widely been used in most CVD process; however, a liquid precursor is more suitable for industrial production process because of its long-time stability and controllability of composition [9].

In the present study, we have developed a new laser CVD with ultrasonically nebulized liquid precursor and prepared single-phase c-axis-oriented Y247 film on multilayer-coated Hastelloy tape substrate.

Experimental procedure

Figure 1 shows a schematic of laser CVD apparatus. $Y(dpm)_3$, $Ba(dpm)_2/(tmod)_2$, and $Cu(dpm)_2$ metalorganic compounds mixed with tetrahydrofuran solvent at molar concentrations of 0.1 and 0.01 mol 1^{-1} were used as a precursor. The liquid precursor was nebulized by ultrasonic transducers and the precursor mist was carried into the chamber with Ar gas. The total pressure in the chamber was held at 20 kPa. (100) CeO₂-buffered multilayer-coated Hastelloy C273 tape (5 mm × 10 mm × 0.5 mm) was used as a substrate. The substrate was heated on a hot stage at a preheating temperature of 873 K, and a thermocouple was inserted backside of the substrate to measure the deposition temperature. A continuous-wave mode Nd:YAG laser beam (wavelength: 1064 nm; diameter: 20 mm; output 260 W) was introduced through a quartz window to irradiate the whole substrate. Deposition was conducted for 900 s.

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The crystal phase was analyzed by X-ray diffraction (XRD, Rigaku Ultima VI) using Cu K α X-ray radiation. The surface morphology was observed by a field-emission scanning electron microscope (FESEM, JEOL JSM-7500F). A schematic of crystal structure was illustrated by VESTA, a three-dimensional visualization system [10].

Results and discussion

Figure 3 shows the XRD pattern of the Y247 films. At precursor concentration of 0.01 mol l^{-1} and deposition temperature of 933 K, single-phase Y247 film with *c*-axis orientation was obtained (JCPDS #49-1220) (Fig. 2(b)). At a precursor concentration of 0.1 mol l^{-1} and deposition temperature of 983 K, *a*-axis-oriented YBa₂Cu₃O_{7- δ} (Y123) was codeposited with *c*-axis oriented Y247 film (Fig. 2(c)).



Fig. 1 A schematic of laser CVD apparatus with ultrasonic neblizer of liquid precursor.



Fig. 2 XRD patterns of the multilayer-coated Hastelloy substrate (a) and the Y247 films (b, c).



Fig. 3 A schematic of crystal structures of Y123, Y124 and Y247.

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Y247 has a similar structure to Y123 and Y124 as shown in Fig. 3. Y123 has an orthorhombic structure (space group: *Pmmm*; a = 0.383 nm, b = 0.389 nm and c = 1.170 nm; ICSD #62928) (Fig. 3(a)) [1]. Y124 and Y247 have an orthorhombic structure with a space group *Cmmm* (Y124: a = 0.384 nm, b = 0.387 nm and c = 2.723 nm, ICSD #69698 (Fig. 3(b)) [2]; Y247: a = 0.383 nm, b = 0.387 nm and c = 5.041 nm, ICSD #82676 (Fig. 3(c)) [3]). These compounds are characterized by a layered structure of CuO₂ plane and CuO chain blocks. CuO₂ locates at the boundary of each layer and Y atom locates between CuO₂ planes. Ba atoms locate at the position to form a defect perovskite structure of Ba, Cu and O atoms. Y247 has a long-length *c*-axis structure consisting of alternating blocks of Y123 and Y124.

Figure 3 shows the surface morphology of the *c*-axis-oriented Y247 films prepared at precursor concentration of 0.01 mol l^{-1} and deposition temperature of 933 K. Although thin cracks along (010)-and (001)-CeO₂ direction was observed, the *c*-axis-oriented Y247 film had a smooth surface.



Fig. 4 Surface morphology of the single-phase *c*-axis-oriented Y247 films.

Summary

Single-phase *c*-axis-oriented Y247 film was obtained on (100) CeO₂-buffered multilayer-coated Hastelloy C273 tape substrate at precursor concentration of 0.01 mol 1^{-1} and deposition temperature of 933 K. At precursor concentration of 0.1 mol 1^{-1} and deposition temperature 983 K, *a*-axis-oriented Y123 was codeposited with *c*-axis oriented Y247 film. *c*-axis-oriented Y247 film had a smooth surface.

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