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The effect of susceptor inclination angle on the quality of superconducting YBCO thin films prepared by a photo-assisted MOCVD system

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ABSTRACT

Superconducting $YBa_2Cu_3O_{7-x}$ thin films were deposited on $LaAlO_3$ (100) substrates by a photo-assisted metal-organic chemical vapor deposition (MOCVD) technique. In our experiment, growth temperature, chamber pressure and growth time were all fixed; the only process variable was the susceptor inclination angle (Φ) between the axis of gas inlet and the susceptor surface. It was found that the growth rate of YBCO films increases monotonically as Φ increases, whereas the superconducting critical current density (J_c) has a maximum (1.2×10^6 A/cm^2) near $\Phi = 22.5°$. The experimental results were found to be well correlated with the three-dimensional simulations of gas flow inside the MOCVD reactor, with Φ being the only process variable.

1. Introduction

High temperature superconductor (high-T_c) $YBa_2Cu_3O_{7-x}$ (YBCO) thin films have significant potential for applications in mobile microwave telecommunication systems [1–3]. Various film preparation techniques, such as laser ablation [4], magnetron sputtering [5,6], and metal-organic chemical vapor deposition (MOCVD) [7–12], have been utilized for epitaxial growth of high-quality YBCO films. Among them, MOCVD technique has its own advantage in uniform growth over large areas. The technique of photo-assisted MOCVD [9,13–15] has some additional advantages, such as the capability to grow both thin and thick high-quality YBCO films, with high growth rate and having single-crystal-like qualities, e.g., no SEM visualizable grain boundaries [14,15]. Thus, the weak link effects caused by the grain boundaries of superconducting YBCO films are reduced [16,17].

In this paper, YBCO films are grown by a photo-assisted MOCVD technique. We examined the effect of various gas flow patterns on the superconducting critical current density (J_c). The variation of gas flow patterns in the quartz reactor of a photo-assisted MOCVD system was controlled by variation of the susceptor inclination angle (Φ), i.e., the angle between the axis of the gas inlet and the surface of the susceptor, as shown in Fig. 1. Obviously, as Φ changes, the gas flow pattern in the reactor chamber will be changed accordingly. This change of gas flow pattern should affect the growth mechanism of the growing YBCO films. Presumably, the film growth rate, crystal structure/orientation and surface/cross-section morphology of the resulting YBCO films will also be affected, as will the J_c of the films. In order to optimize the growth rate and the J_c value of superconducting YBCO films, the correlations between the film growth factors and the susceptor inclination angle (Φ) are investigated by both experiments and numerical simulations.

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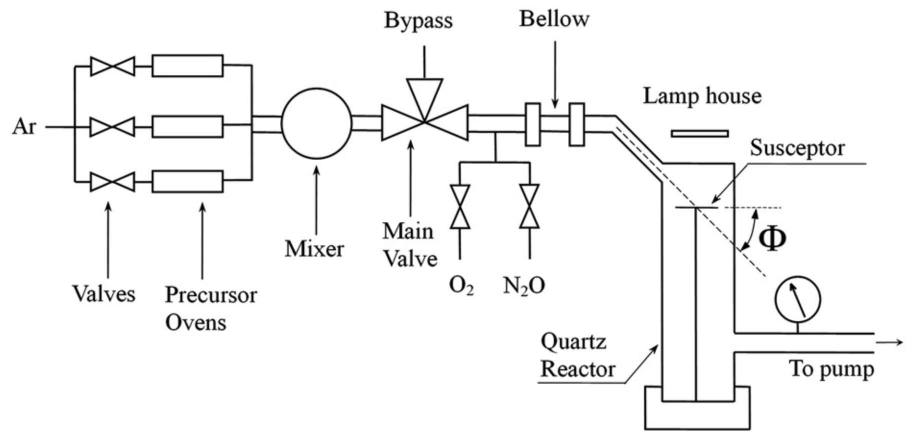

Fig. 1. Schematic diagram of the photo-assisted MOCVD reactor system.

2. Film preparation and characterization

The YBCO thin films (with thickness $\leqslant 1$ μm) were prepared on $LaAlO_3$ (1 0 0) (LAO(1 0 0)) substrates in a photo-assisted MOCVD system as shown in Fig. 1. Tungsten halogen lamps acted as the sole energy source. The major reason for utilizing this kind of energy source instead of traditional ones is based on the superior activation capability of tungsten halogen lamps. As compared with thermal activation, the effects of Boltzmann distribution can be overcome by directing the photo-activation [18,19]. This system is similar to one reported previously [14], but with the addition of a mixing chamber positioned right after the precursor ovens. The angle Φ was varied by rotating the surface of the susceptor with respect to the fixed center axis of the inlet tube. As Φ varied, the lamp housing would also be varied to maintain the illumination of lamps on the susceptor surface in the same way, independent of the change of Φ. The susceptor surface was not rotated horizontally. The substrate temperature, reactor pressure, and film growth time were kept at 825 °C, 7.5 torr, and 3 min, respectively. The values of Φ used for YBCO film growth were 0°, 22.5°, 45° and 67.5°.

Solid metal-organic β-diketonates precursors, such as 2,2,6,6-tetramethyl-3,5-heptanedionate (TMHD) of Y, Ba, and Cu, in the respective forms of $Y(TMHD)_3$, $Ba(TMHD)_2$, and $Cu(TMHD)_2$, were used for the growth of YBCO films. They were separately contained in three different ovens, with their heating temperatures individually controlled. Argon was used as the carrier gas. The oxidizing reagent was a mixture of O_2 and N_2O, with an optimized volume ratio of 3:2, as reported previously [9]. The growth temperatures of these samples were measured with K-type thermal couples.

In order to minimize the effects of thermal profile and mass distribution on YBCO film, only small pieces of LAO(1 0 0) substrates (about 5 mm × 10 mm) were placed at the center of the 2″ susceptor. So uniformity is essentially not considered in this study. Other factors, such as possible variation of composition ratio of the inlet gases, can also be overlooked with this simplified approach.

The YBCO thin films were characterized by XRD (2θ-scan, Φ-scan, and rocking curve), scanning electron microscopy (SEM), and energy dispersive spectroscopy (EDS). Superconducting properties of these samples, such as the superconducting critical temperature (T_c) and superconducting critical current density (J_c), were determined from magnetization measurements made using a Quantum Design MPMSXL-7 system [20]. In particular, values of J_c were indirectly derived from the field dependent magnetization measurements using the Bean Model [21–26]. Dimensions of the rectangular YBCO samples were $a \times b \times c$, where a, b and c are the width, length and thickness of the films, respectively. Then J_c can be calculated from the following equations [26]:

$$J_c = 20(M^+ - M^-)/a(1 - a/3b) = 20\Delta M/a(1 - a/3b), \quad (1)$$

where $\Delta M = M^+ - M^-$ (in emu/cm^3) is the difference between the upper and lower branches of $M(H)$ curve. Also, $b > a$ is required for Eq. (1), and the applied magnetic field H is applied perpendicular to the surface of the YBCO film.

3. Computer simulation of gas flow patterns

To correlate gas flow patterns with growth rate, crystal structure, morphology, and superconducting properties, a three-dimensional simulation for gas flow patterns was conducted with a commercial software program (FLUENT 6.0). Related processing parameters for this simulation were inlet gas velocity = 5 m/s; top wall temperature of the quartz reactor = 300 °C; side wall temperature of this reactor = 200 °C; inlet gas temperature = 300 °C; susceptor temperature = 825 °C; total reactor pressure = 7.5 torr (with partial pressures of O_2, $P(O_2)$ = 2.25 torr and N_2O, $P(N_2O)$ = 1.5 torr). The values of the angle Φ were 0°, 22.5°, 45°, 67.5° and 90°. However, YBCO film growth was not done at Φ = 90°, because the reactor arrangement with Φ = 90° is not attainable in the present reactor. The simulated gas flow pattern for Φ = 90° is therefore presented for reference only.

4. Results and discussion

The three-dimensionally simulated gas flow patterns inside the quartz reactor of the photo-assisted MOCVD system are presented in Fig. 2, where the susceptor inclination angle, Φ, is the only process variable. The values of Φ were 0°, 22.5°, 45°, 67.5° and 90°. Fig. 2(a-1), (b-1), (c-1), (d-1) and (e-1) are simulated gas flow patterns for cross-sections through the reactor symmetry plane. Fig. 2(a-2), (b-2), (c-2), (d-2) and (e-2) are the corresponding simulated gas flow patterns at layers 1 mm above the susceptor surface.

Fig. 3 shows the growth rate of the YBCO thin films as a function of the susceptor inclination angle Φ. The superconducting critical current density (J_c) were calculated according to Eq. (1), where M^+ and M^- were measured at T = 77 K and H = 0 Oe. Fig. 4 shows J_c versus Φ with the transition temperature T_c determined by the $M(T)$ magnetization measurement. The results indicate that a maximum for J_c exists at $\Phi \approx 22.5°$. Surface and cross-section morphologies of the YBCO films grown at different Φ values are revealed by the SEM micrographs shown in Figs. 5 and 6, respectively. XRD 2θ