

# Effect of deposition temperature on the orientation and electrical properties of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ films prepared by laser CVD using liquid-source evaporation

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## Abstract

$\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO) films were prepared on a multilayer-coated Hastelloy C276 tape by laser chemical vapour deposition using liquid-source evaporation. The orientation of the YBCO films changed from *a*-axis to *c*-axis when the deposition temperature was increased from 1010 K to 1090 K. The *c*-axis-oriented YBCO film prepared at 1046 K exhibited critical temperature of 92 K and critical current density of  $1.7 \text{ MA cm}^{-2}$  at a deposition rate of  $13 \mu\text{m h}^{-1}$ .

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**Keywords:** A. Film; YBCO; Laser CVD; Liquid precursor

## 1. Introduction

Metalorganic chemical vapour deposition (MOCVD) has been widely studied in the preparation of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO)-coated conductors intended for practical applications because of its advantages, such as conformal deposition and suitability for use in large-scale continuous production systems. Watanabe et al. reported the rapid formation of a 200-m-long YBCO-coated conductor by using a reel-to-reel multistage CVD system [1]. Selvamanickam et al. reported the high-speed preparation of a Gd-doped YBCO-coated conductor at  $24 \mu\text{m h}^{-1}$  [2].

Laser CVD has stronger potential than MOCVD for the preparation of YBCO-coated conductors. The chemical reaction of the film can be enhanced by intense laser irradiation, which results in significant oriented growth and a high deposition rate [3–5]. The high-speed preparation of YBCO-coated conductors by laser CVD would be a promising technique for reducing production costs relative to those associated with current methods. In our previous studies, we prepared YBCO films at high deposition rates (55–100  $\mu\text{m h}^{-1}$ ) on single-crystal substrates [6] and on multilayer-coated Hastelloy C276 tape [7]. However, the critical current density ( $J_c$ ) of  $0.5 \text{ MA cm}^{-2}$  should be increased to satisfy industrial demands.

Yttrium, barium and copper metalorganic compounds have been used as solid precursors for the fabrication of YBCO films in previous studies, in which the composition of the resulting YBCO films was controlled by changing the vapour pressure of each precursor source material. For industrial mass production, the evaporation of multicomponent solid precursors would be difficult for the deposition of a long YBCO-coated conductor that exhibits a critical current density  $J_c$  greater than  $1 \text{ MA cm}^{-2}$ . Solid precursors often result in degraded surface states when long deposition times are used, and the evaporated flux of the precursor typically decreases with increased deposition time. We have developed a liquid-source evaporation system for the preparation of  $\text{CeO}_2$  and YBCO films via a laser CVD process [8].

In the present study, YBCO superconducting films were prepared by laser CVD using liquid-source evaporation, and the effects of the deposition temperature on the orientation, crystallinity, microstructure and electrical properties of the YBCO films were investigated.

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## 2. Experimental

A schematic of the laser CVD apparatus has been reported elsewhere [8]. The  $\text{Y}(\text{dpm})_3$ ,  $\text{Ba}(\text{dpm})_2/\text{Ba}(\text{tmod})_2$  and  $\text{Cu}(\text{dpm})_2$  (dpm, dipivaloylmethanate; tmod, 2,2,6,6-tetramethyl-3,5-octanedionate) precursors were dissolved in tetrahydrofuran ( $\text{C}_4\text{H}_8\text{O}$ ) as a source solution in a Y:Ba:Cu molar ratio of 1:2.4:2.8 with a Y ( $\text{dpm})_3$  concentration of  $0.01 \text{ mol L}^{-1}$ . The solution was delivered at a supply rate of  $0.11 \times 10^{-4} \text{ L s}^{-1}$  by a plunger pump.

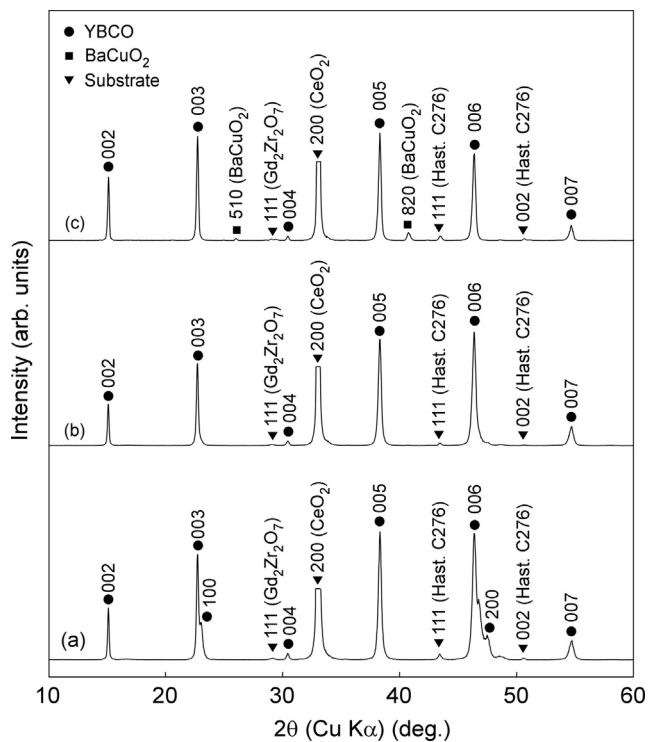


Fig. 1. XRD patterns of YBCO films prepared at various  $T_{\text{dep}}$ : 1010 K (a), 1046 K (b) and 1090 K (c).

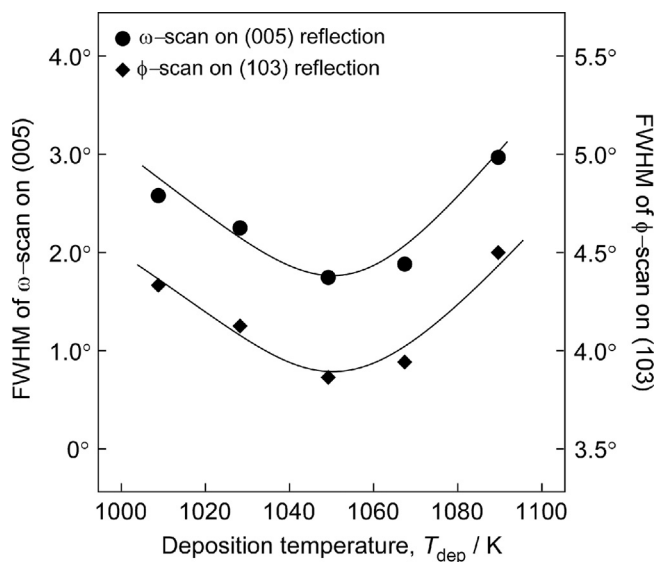


Fig. 2. The effect of the  $T_{\text{dep}}$  on the FWHM of  $\omega$ -scan on the (005) reflection and that of  $\phi$ -scan on the (103) reflection of the YBCO films.

The solution was sprayed into and evaporated in the evaporation chamber at 653 K. The flow rates of Ar and  $\text{O}_2$  gases were  $1.52 \times 10^{-6} \text{ m}^3 \text{ s}^{-1}$  and  $0.34 \times 10^{-6} \text{ m}^3 \text{ s}^{-1}$ , respectively.

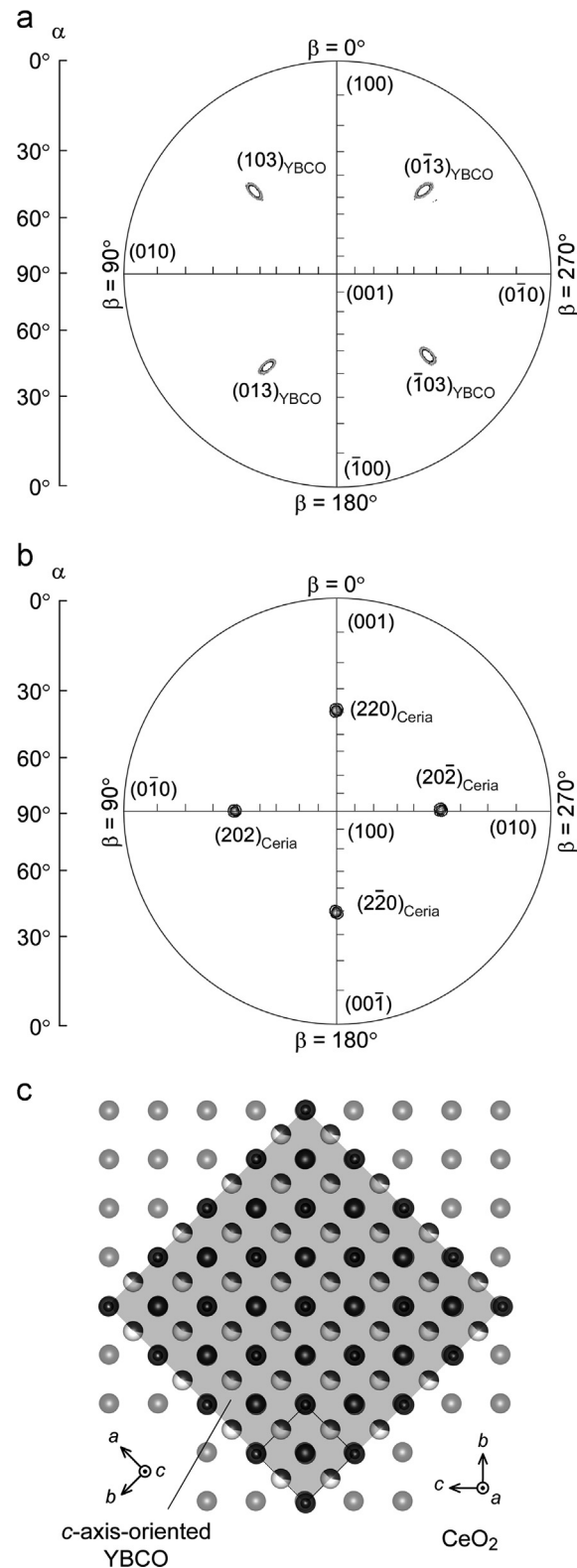


Fig. 3. X-ray pole figures of the (103) reflection of the YBCO films prepared at  $T_{\text{dep}} = 1046 \text{ K}$  (a) and the (220) reflection of the  $\text{CeO}_2$  layer (b). Image (c) illustrates the in-plane orientation relationship of the  $c$ -axis-oriented YBCO film on  $\text{CeO}_2$  (100) plane.

Hastelloy C276 tape with  $\text{CeO}_2/\text{LaMnO}_3/\text{MgO}/\text{Gd}_2\text{Zr}_2\text{O}_7$  buffer layers was cut into pieces with dimensions of  $10\text{ mm} \times 5\text{ mm} \times 0.1\text{ mm}$ , which were used as substrates. The buffer layers were prepared by ion-beam-assisted deposition and pulsed-laser deposition [9]. The Hastelloy C276 tape was preheated on a hot stage at 923 K. The total pressure was maintained at 0.5 kPa. The as-deposited YBCO films were heat treated in the sequence 573 K for 14.4 ks, 673 K for 14.4 ks and 773 K for 14.4 ks under a pure  $\text{O}_2$  atmosphere (100 kPa) to compensate oxygen vacancies and to transform the YBCO from a tetragonal to an orthorhombic structure. After heat treatment, the  $\delta$  decreased from 0.96 to 0.18 [10], indicating the transition to orthorhombic structure ( $\delta < 0.65$ ) [11].

The YBCO film phase was investigated by X-ray diffraction (XRD; Rigaku RAD-2C). The out-of-plane texture of the YBCO films was evaluated on the basis of the full width at half maximum (FWHM) of the peaks generated by  $\omega$ -scans (rocking curves) of (200) and (005) reflections. The in-plane orientation of the YBCO film was evaluated on the basis of the FWHM of the peak generated by a  $\varphi$ -scan of the (103) reflection, which was measured by a pole-figure X-ray diffractometer (XRD; Rigaku Ultima IV). The microstructure was observed using a field-emission scanning electron microscope (FESEM; JEOL JSM-7500F). A schematic of the epitaxial relationship was drawn using the VESTA software package [12]. The electrical conductivity was measured from 30 K to 300 K using a dc four-probe method. The critical current ( $I_c$ ) was measured in liquid  $\text{N}_2$  (77 K) using a dc four-probe method with the criterion of  $1\text{ }\mu\text{V cm}^{-1}$ . Au electrodes were deposited onto the sample surface by using an ion-coater and were connected to thin copper wires (0.1 mm in diameter) with Ag paste. For the  $I_c$  measurements, Ag electrodes that were approximately 6  $\mu\text{m}$  thick

were deposited onto the sample surface via dc sputtering. The four electrodes were spaced at 1 mm interval.

### 3. Results and discussion

The deposition temperature ( $T_{\text{dep}}$ ) was increased from 1010 to 1090 K as the laser power ( $P_L$ ) increased from 97 to 160 W. The relationship between the  $T_{\text{dep}}$  and the  $P_L$  was almost linear. Fig. 1 shows the XRD patterns of YBCO films prepared at  $T_{\text{dep}} = 1010$ –1090 K. YBCO films co-oriented along the  $a$ - and  $c$ -axes were prepared at  $T_{\text{dep}} = 1010$ –1027 K (Fig. 1(a)). YBCO films oriented along the  $c$ -axis were prepared at  $T_{\text{dep}} = 1046$ –1090 K (Fig. 1(b)). A small amount of  $\text{BaCuO}_2$  was codeposited in the  $c$ -axis-oriented YBCO film prepared at  $T_{\text{dep}} = 1090$  K. As the  $T_{\text{dep}}$  was increased,  $a$ -axis preferred orientation was suppressed. This trend related to the preferred orientation of the YBCO films was the same as that observed for YBCO films prepared by laser CVD using solid precursors [13,14].

Fig. 2 shows the FWHM of the out-of-plane ( $\omega$ -scan) and in-plane crystallinities ( $\varphi$ -scan) of YBCO films prepared at  $T_{\text{dep}} = 1010$ –1090 K. The FWHM of the peak generated by the  $\omega$ -scan showed a minimum of  $1.8^\circ$  at  $T_{\text{dep}} = 1046$  K. The FWHM of the peaks generated by the  $\varphi$ -scan exhibited a trend similar to that of the peaks generated by the  $\omega$ -scan. The FWHM of the  $\varphi$ -scan showed a minimum of  $3.8^\circ$  at  $T_{\text{dep}} = 1046$  K. The FWHM value of  $3.8^\circ$  for the  $\varphi$ -scan in the present study was comparable to that of films prepared by MOCVD ( $3.6$ – $3.7^\circ$ ) [1,15] and by laser CVD using solid precursors ( $3.8^\circ$ ) [7].

Fig. 3 shows the XRD pole-figure pattern generated from the (103) reflection of the YBCO films prepared at  $T_{\text{dep}} = 1046$  K and that generated from the (220) reflection of the  $\text{CeO}_2$  film deposited

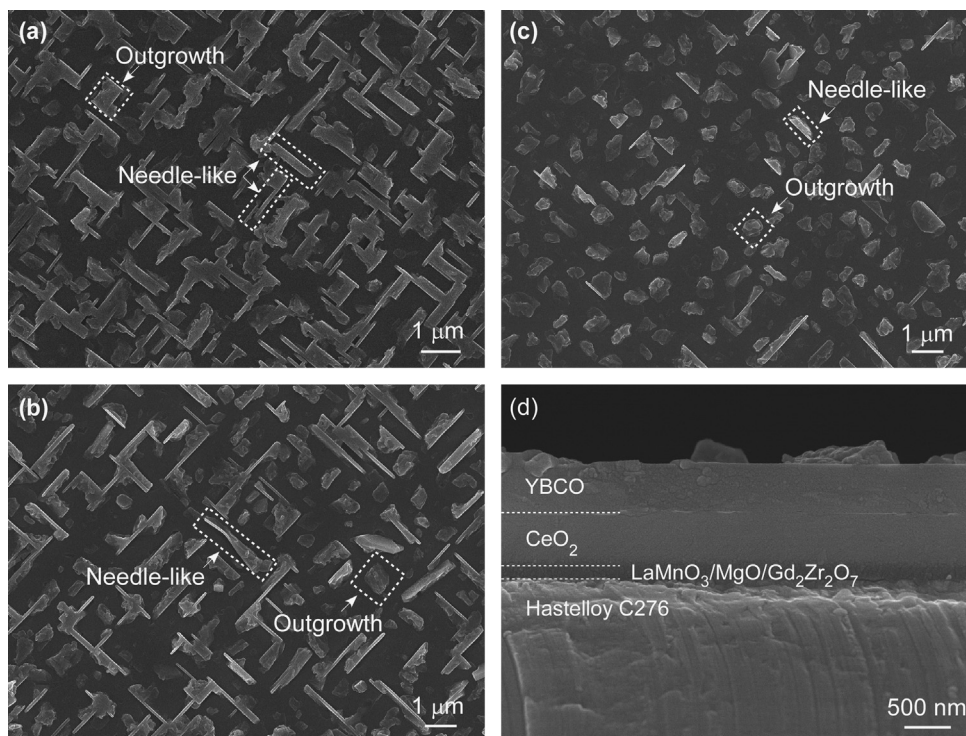


Fig. 4. Surface (a)–(c) and cross-sectional (d) SEM images of YBCO films prepared at various  $T_{\text{dep}}$ : 1010 K (a), 1027 K (b) and 1046 K (c) and (d).

onto the Hastelloy C276 tape. The X-ray pole-figure pattern of the YBCO (103) reflection showed a four-fold pattern at approximately  $\alpha=45^\circ$  (Fig. 3(a)), which was attributed to the YBCO {103} planes with an angle of  $46^\circ$  between the YBCO (001) and (103) planes. The X-ray pole-figure pattern of the (220) reflection of the  $\text{CeO}_2$  film showed a four-fold pattern at approximately  $\alpha=45^\circ$  (Fig. 4(b)), which was attributed to the  $\text{CeO}_2$  {220} planes with an angle of  $45^\circ$  between the  $\text{CeO}_2$  (100) and (110) planes. The azimuth angles ( $\beta$ ) of the pole peaks from the YBCO (103) reflection were rotated  $45^\circ$  with respect to those of the  $\text{CeO}_2$  (220) reflection, which indicates an in-plane epitaxial growth relationship of YBCO [100]// $\text{CeO}_2$  [011] for the  $c$ -axis-oriented YBCO film (Fig. 3(c)). The YBCO films prepared at  $T_{\text{dep}}=1010$ – $1090$  K showed the same in-plane epitaxial growth relationship as that prepared at  $T_{\text{dep}}=1046$  K.

Fig. 4 shows the surface and cross-sectional images of the YBCO films prepared at  $T_{\text{dep}}=1010$ – $1090$  K. The YBCO film prepared at  $T_{\text{dep}}=1010$  K exhibited a flat surface with needle-like and outgrowth grains (Fig. 4(a)). The flat areas on the surface of the YBCO film were attributed to the (001) plane of the

$c$ -axis-oriented YBCO grains, whereas the needle-like grains were attributed to the  $a$ -axis-oriented YBCO grains. Because the growth rate along the  $\langle 100 \rangle$  and  $\langle 010 \rangle$  directions is faster than that along the  $\langle 001 \rangle$  direction, these  $a$ -axis-oriented YBCO grains often grow outward from the surface of  $c$ -axis-oriented YBCO films [16]. The outgrowth particles were a Cu-rich phase, as observed by TEM-EDX analysis [7], and were distributed along the boundaries of the  $a$ -axis-oriented and  $c$ -axis-oriented YBCO grains. When the  $T_{\text{dep}}$  was increased to 1027 K, the number of needle-like  $a$ -axis-oriented grains decreased and the shape of the outgrowth particles tended to be granular (Fig. 4(b)). When the  $T_{\text{dep}}$  was increased to 1046 K, the formation of the needle-like grains were suppressed (Fig. 4(c)). All the films exhibited a dense cross section (Fig. 4(d)). Their thickness decreased from 740 nm to 590 nm when the  $T_{\text{dep}}$  increased from 1010 to 1090 K, which indicates the deposition rates of  $15$ – $12 \mu\text{m h}^{-1}$ . In the films prepared by laser CVD, the number of needle-like  $a$ -axis-oriented grains effectively decreased when the  $T_{\text{dep}}$  ( $P_L$ ) increased.

Fig. 5 shows the temperature dependence of the electrical resistivity of the YBCO film prepared at  $T_{\text{dep}}=1046$  K and the effect of the  $T_{\text{dep}}$  on the  $T_c$  of the YBCO films. The electrical resistivity of the YBCO film prepared at  $T_{\text{dep}}=1046$  K linearly

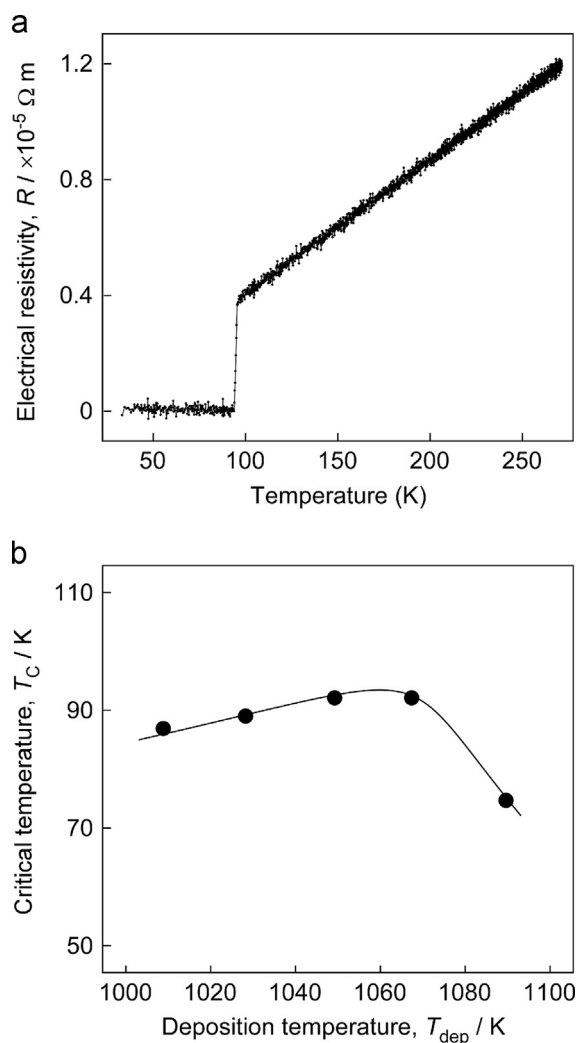


Fig. 5. Temperature dependence of electrical resistivity of the YBCO film prepared at  $T_{\text{dep}}=1046$  K (a) and the effect of the  $T_{\text{dep}}$  on the  $T_c$  of the YBCO film (b).

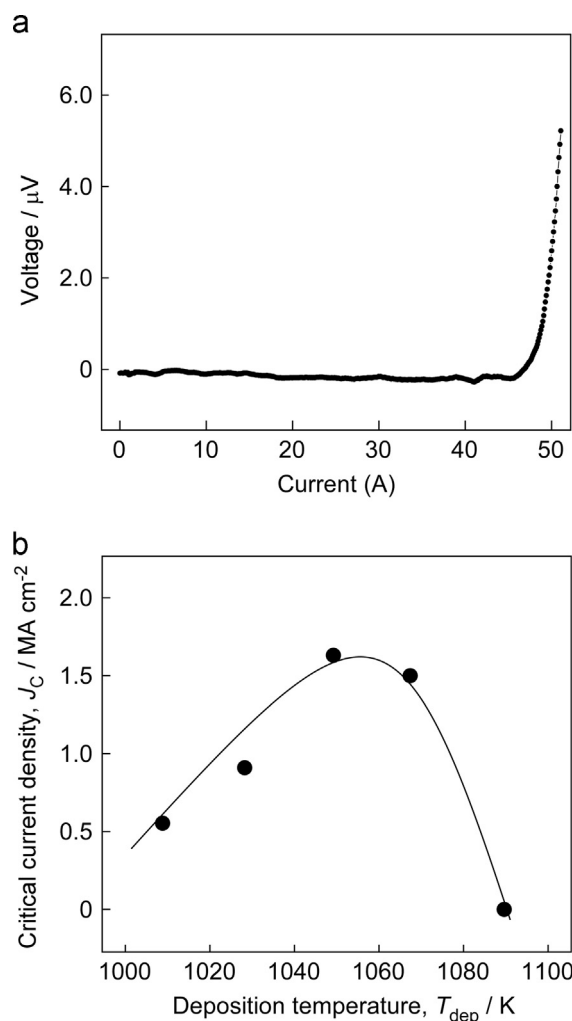


Fig. 6. The  $I$ - $V$  curve of the YBCO film prepared at  $T_{\text{dep}}=1046$  K (a) and the effect of the  $T_{\text{dep}}$  on the  $J_c$  of the YBCO films (b).



decreased with temperature and showed a sharp zero-resistance transition at 92 K (Fig. 5(a)). The electrical resistivity of other YBCO films prepared at  $T_{\text{dep}}=1010\text{--}1090\text{ K}$  showed temperature dependences similar to that of the film prepared at  $T_{\text{dep}}=1046\text{ K}$ , and the zero-resistance transitions of these films occurred at 74–89 K (Fig. 5(b)). When the  $T_{\text{dep}}$  was increased from 1010 to 1090 K, the  $T_c$  of the YBCO films first increased from 88 to 92 K and then decreased to 74 K.

Fig. 6 shows the current–voltage ( $I$ – $V$ ) curve for the YBCO film prepared at  $T_{\text{dep}}=1046\text{ K}$  and the effect of  $T_{\text{dep}}$  on the  $J_c$  of the YBCO films. In the case of the YBCO film prepared at  $T_{\text{dep}}=1046\text{ K}$ , the voltage remained zero as the current increased while it remained under 49 A (Fig. 6(a)), which indicates a  $J_c$  of  $1.7\text{ MA cm}^{-2}$ . The  $J_c$  of the YBCO film increased from 0.5 to  $1.7\text{ MA cm}^{-2}$  and decreased to  $1.6\text{ MA cm}^{-2}$  as the  $T_{\text{dep}}$  increased from 1010 to 1067 K (Fig. 6(b)). The YBCO film prepared at  $T_{\text{dep}}=1090\text{ K}$  showed no  $I_c$  at 77 K because this film contained a  $\text{BaCuO}_2$  phase. The  $J_c$  of  $1.7\text{ MA cm}^{-2}$  of the YBCO film in the present study was three times greater than that previously reported for films prepared by laser CVD using solid precursors ( $0.5\text{ MA cm}^{-2}$ ) [7].

#### 4. Conclusions

$c$ -axis-oriented YBCO superconducting films were prepared on multilayer-coated Hastelloy C276 by laser CVD using liquid-source evaporation. When the  $T_{\text{dep}}$  ( $P_L$ ) was increased, the orientation changed from  $a$ -axis to  $c$ -axis. The in-plane epitaxial growth relationship was YBCO [100]// $\text{CeO}_2$  [011] for the  $c$ -axis-oriented YBCO films. The YBCO films showed a high  $T_c$  of 88–92 K and a high  $J_c$  of  $0.5\text{--}1.7\text{ MA cm}^{-2}$ . The deposition rate of the YBCO films was  $12\text{--}15\text{ }\mu\text{m h}^{-1}$ .

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