Effect of Substrate Temperature on the Critical Current Density in the YBa₂Cu₃O_{7-x} Superconducting Films

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Abstract: A comparative study of effect of substrate temperature (T_s) on the critical current density (J_c) in YBa₂Cu₃O_{7-x} (YBCO) superconducting films is reported. YBCO superconducting films were fabricated by using the pulsed laser deposition (PLD) technique. The substrate temperature was ranged from 720 to 800 °C in order to find its optimum value for depositing YBCO films. Results of structural measurements the YBCO films showed that the lattice parameter *c* were depended on the substrate temperature. The largest value of lattice parameter *c* calculated from Xray diffraction patterns was obtained for the YBCO film deposited at 780 °C. More interestingly, the critical current density of the YBCO films measured at 65 K also depended on the substrate temperature. The highest value of critical current density of ~ 5.7 MA/cm² was also found at the YBCO film deposited at 780 °C. Consequently, the optimum value of T_s of 780 °C might be concluded.

Keywords: YBCO, thin film, J_c.

1. Introduction

Significant efforts have been carried out on fabricating the second-generation high temperature superconductors (HTS), among them the YBa₂Cu₃O_{7- δ} (YBCO) is the first HTS showing superconducting state at liquid nitrogen temperature (77 K) [1]. The main purpose has been aimed to utilize the magnetic properties of YBCO at 77 K, those are highly demanded for power applications such as electric transmission lines, motors, generators... Fabrications of HTS tapes or films have been done by using a wide variety of deposition techniques consisting of pulsed laser deposition (PLD), metalorganic chemical vapor deposition (MOCVD), RF-magnetron sputtering... [2-5]. Among these techniques, the PLD has been compared to be one of the most promising ones due to its own advantages: a high precise control of film stoichiometry, reproducibility, uniformity and simplicity, so it has been applied to our research in fabricating YBCO films [2].

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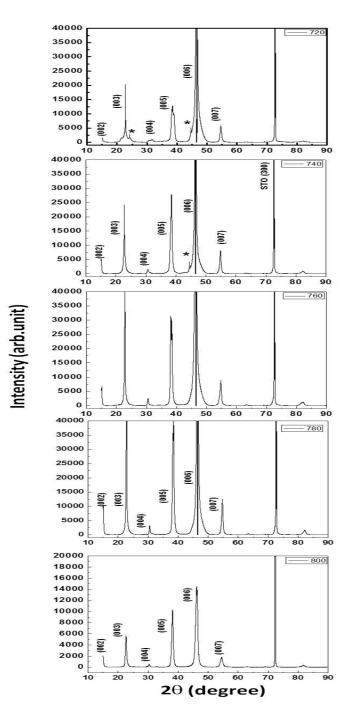


Fig. 1. XRD results of the YBCO films deposited at different substrate temperatures.

As for various applications of superconductors, J_c is the most important parameter for the material. The superconducting films with high critical current density J_c are obviously needed either for high current applications such as YBCO coated conductors [2] or for low current applications such as YBCO coated for microwave filters [6]. Technically, the YBCO films with high J_c are likely to show the *c*-axis oriented, well ab- plane aligned properties as well as less grain boundaries [4]. Consequently, for the effective usage of YBCO superconducting films for these two types of applications, the appropriate fabricating conditions consisting of substrate temperature (T_s) (might be called as growth temperature), ambient gas pressure... are required to be optimized. Among them, T_s is the most sensitive parameter since it is directly related to the layer-by-layer condensation process of YBCO plasma on the substrate. In this paper, the effect of T_s , then, is reported to find the optimum T_s to deposite YBCO films with high J_c.

2. Experiment

The YBCO target used in our experiment was prepared by using the solid state reaction method. The appropriate amounts of starting materials of Y_2O_3 , BaCO₃ and CuO were throughly mixed by using the wet-ball milling. The mixed powder was sintered at 880°C in air for 24 hours then ground by using the mortar and pestle. The sintering/grinding process was repeated several times. The obtained powder was pressed into a pellet, and then sintered at 920 °C in air for 24 hours. The oxygen annealing process for the pellet was followed at 500 °C for 15 hours, then cooled to room temperature.

The studied YBCO superconducting films were fabricated on (100) $SrTiO_3$ (STO) by using the PLD technique. The 248 nm wavelength KrF excimer laser operated at an energy of 250 mJ was applied to ablate the target surface. The target – substrate distance was kept at 4.5 cm and the oxygen gas pressure was maintained at 200 mTorr. The substrate temperatue was varied from 720 to 800°C in order to find the optimum value.

For characterizing the YBCO films, The crystalline structures were examined by using the X-ray diffraction (XRD), the surface morphologies were compared by using the scanning electron microscopy (SEM). The magnetization data were measured by using the MPMS XL-5 system with the field applied parallel to the *c*-axis of the films.

3. Results and discussion

In order to find the phase formation, the YBCO films fabricated at different substrate temperature were initially examined by XRD with the results are given in Fig. 1.

Overall, the XRD patterns of all YBCO films contain (00*l*) peaks (where *l* ranged from 2 to 7) of the YBCO phase without the impurity indicates that the YBCO films are c-axis oriented. The interdistance between the (00*l*) peaks is compared to be almost the same which is corresponding to the inter-layers of YBCO crystal structure. Besides strong peaks found at $2\theta \sim 72.5^{\circ}$ identified to be STO, the appearance of the XRD peaks are observed to be differrent with increasing T_s. For the YBCO film deposited at T_s = 720°C, the XRD pattern shows the presence of relatively broad peaks and their intensity is recorded to be small. The exposure of the tiny peaks (marked by (*) symbols) is attributed to the (*h00*) orientations suggesting the formation of *a*-axis phases. The growth orientations of the YBCO films deposited at T_s = 740°C and 760°C are found to be almost similar to that of the YBCO film deposited at T_s = 720°C. Interestingly, the peak intensity and broaden are observed to increase reduced, respectively, those indicate the crystallinity improvement. The highest crystallinity improvement is obtained for the YBCO film deposited at T_s = 780°C, which is evidenced by no formation of *a*-axis phases. Over 780°C, the appearance of (001) peaks is inversed: broaden and small intensity which reveals the degradation of the film crystallinity. The lattice parameter c was calculated by taking the (001) peaks of YBCO phase with different values of T_s as listed in Table 1. Table 1. Lattice parameter c and surface roughness of the YBCO films

T_{s} (°C)	c (Å)	R _{r.m.s}
720	11.6701	17,5
740	11.6892	14.3
760	11.7001	10.1
780	11.7102	7.8
800	11.7008	8.2

720 1μm 740° C 1μm 760°C 0 1μm 780°C 1μm 800°C <u>1μm</u>

Fig. 2. SEM images of the YBCO films deposited at different substrate temperatures. The formation of the a-axis grains is found to reduce as increasing temperature and the smooth film surface was observed at 780°C.

The formation of the a-axis and c-axis phases is examined by using the surface morphology analyses. The typical SEM images of the YBCO films are shown in Fig. 2. For the YBCO film deposited at $T_s = 720^{\circ}$ C, the surface could be seen to consist of two part. The lower part (black background) is relatively smooth, and the upper part is covered by large rectangular shapes those are horizontally developed. The rectangular shapes are then called as the a-axis grains. This surface formation is likely to be caused by the low surface temperature effect [7]. Based on the fact that YBCO is a material whose small thermal conductivity, which induce a high temperature gradient following the fim thickness, the surface temperature might be lower than T_s . As a results, the a-axis grains are naturally generated. Meanwhile, there is no big changes in the surface morphology of the YBCO films deposited at $T_s = 740$ and 760°C. The formation of the a-axis grains is still observed but the average size and density are found to reduce. Interestingly, the YBCO film deposited at $T_s =$ 780°C shows a smooth surface with a disappearance of the a-axis grains. The a-axis orientation is completely replaced by the c-axis one. As T_s is further increased, however, the film surface is noncontinuous, consisting of obvious cracks (indicated by arrows) as displayed in Fig. 2.

The root mean square (r.m.s) surface roughness values of the YBCO films were evaluated by using AFM (not shown here) and also listed in Table. 1. The smallest surface roughness was obtained for the YBCO film deposited at $T_s = 780^{\circ}$ C that was consistent with the smooth surface observed in the SEM image.

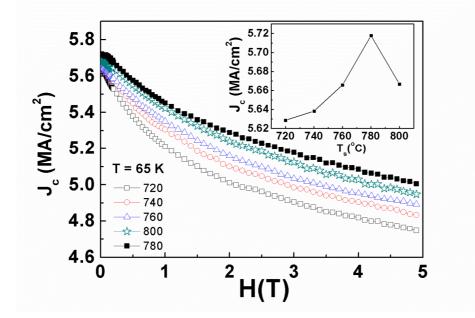


Fig. 3. J_c behaviors of the YBCO films deposited at different substrate temperatures (T_s). The highest J_c is obtained for the YBCO films deposited at $T_s = 780^{\circ}$ C.

Fig. 3 shows the field dependence of J_c for the YBCO films while the fields were applied parallel to the *c*-axis. J_c values were calculated from the simplified Bean model, $J_c = 20\Delta M/[b(1-b/3a)]$ where ΔM is the magnetization difference per unit volume, a and b (a > b) are the sizes of rectangular samples [8]. There was a dependence of self-field J_c (J_c^{sf}) on the substrate temperature T_s : J_c^{sf}

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increases from 5.62 MA/cm² to 5.71 MA/cm² as increasing T_s from 720 to 780, but then decreases back to 5.66 MA/cm² at T_s = 800 °C as shown in the inset of Fig. 3. The highest value of J_c^{sf} , hence, was obtained for the YBCO film at $T_s = 780$ °C. The dependence of in-field J_c on T_s is also studied. The overall decreases in J_c with increasing the applied field are observed with the YBCO films deposited at different T_s . The results are analyzed to be consistent with the intrinsic pinning theory [9]. Each kind of HTS is owning a limited number of intrinsic pinning centers (IPCs) those are going to pin vortex (in forms of quantized magnetic fields called fluxons). As the applied field is increased, the number of vortex is also increased. If the number of vortex is higher than that of IPCs, the remaining ones are going to move leading to the degradation of superconductivity and the decrease of J_c. As increasing T_s , the behavior of in-field J_c is investigated to be similar with that of self-field J_c . As T_s was increased from 720 °C to 780 °C, the J_c was found to increase. The investigation was compared to be in agreement with the changes in surface morphology as given in Fig. 2. The formation of a-axis grains has been reported to obstacle for the current flowing along the (ab) plane of the films [10]. Consequently, by reducing these formation, the current flowing ability would be enhanced. At $T_s =$ 780°C, the film surface was relatively smooth with no a-axis grains on top leading to the highest J_c. Unexpectedly, an opposited behavior occurred at $T_s = 800^{\circ}$ C, the J_c was decreased. The formation of crack-like factor (indicated by arrows) dividing the film in separated areas was likely to be a reason. The current was probably flowing in these separated areas rather than throughly the films, which induced a smaller J_c . These results might indicate that $T_s = 780$ °C was the optimum temperature for fabrication of the YBCO films showing high J_c.

4. Conclusions

The YBCO films were deposited at different substrate temperature. The temperature was ranged from 720 to 800°C while keeping other deposition conditions. The structural examinations showed the formation of the a-axis grains at low temperature. By increasing temperature, the formation was observed to reduce leading to a smooth surface of the YBCO film deposited at 780°C. The overheat effect was found as temperature was increased to 800°C which was evidenced by the occurrence of the crack-like factor. The J_c behavior estimated from the magnetization curves measured at 65 K was compared to be in agreement with the structural results. The formation of the a-axis grains and the crack-like factor was analyzed to obstacle the current flowing. The highest J_c was obtained for the YBCO films deposited at 780°C, which might suggest the optimum substrate temperature.

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