High Functional GdB₂C₃O_{7-x} Thin Films Fabricated by Pulsed Laser Deposition

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Abstract -- REBCO coated conductors (RE: rare earth elements) have recently drawn great attention since they are known to possess stronger flux pinning centers in high magnetic fields compared with YBCO coated conductors. In this study, GdBa₂Cu₃O_{7-d}(GdBCO) was selected to investigate the influence of the distance between target and substrate and substrate temperature on the superconducting properties of GdBCO films on the SrTiO₃(100) substrate. Samples were fabricated by pulsed laser deposition (PLD) with a Nd:YAG laser (355nm). Under a given oxygen pressure of 800mTorr, we changed the distance between target and substrate from 5.5cm to 7.0cm and the substrate temperature from 750 $^{\circ}$ C to 850 $^{\circ}$ C. The crystallinity and texture of GdBCO films were analyzed by X-ray diffraction (XRD), and the surface morphology was observed by the scanning electron microscopy (SEM). Tc and Jc values were measured by the four point probe method. High quality GdBCO films with Tc of 89.7K and Jc over 1 MA/cm² at 77 K in self field were successfully fabricated by optimizing processing parameters. The detailed processing conditions, microstructure and superconducting properties will be presented for a discussion.

1. INTRODUCTION

RE_{1+x}Ba_{2-x}Cu₃O_{6+x}(REBCO, RE=Gd, Nd, Sm, and Eu) superconducting materials have higher critical temperature irreversible fields, compared YBa₂Cu₃O_{7-v}(YBCO) superconductor. It was reported that the separation of the superconducting phase and the non-superconducting in the Gd_{1+x}Ba_{2-x}Cu₃O_{6+δ} (GdBCO) was caused by the substitution, such as the formation of the solid solution by the spinodal decomposition [1-4]. Pulsed laser deposition (PLD) has become a widespread method for the growth of thin films of high temperature superconductors (HTS). The reason that PLD has been spreading is its reproducibility from pure single elements to multi-component materials complex stoichiometry and above all, its ability to deposit films easily from a single process [5-7]. Applications of HTS require a high quality films with high transition temperature, a narrow transition width, and high critical currents. We also obtained high quality GdBCO films with a maximum T_c value and a high J_c by optimizing processing parameters. Moreover, the transport properties of ceramic

superconducting films depend to a great extent on the deposition parameters (substrate temperature, laser fluence, gas pressure, etc) and on the particular set-up. Much effort has been done to understand the influence of each of the multiple parameters during the deposition to optimize the quality of the films and to overcome the intrinsic problems of the technique [8-10].

The oxygen pressure and the substrate temperature are known to play a major role in the properties of these films, not only in influencing the oxygen stoichiometry, but also the plasma expansion process, the number of particulates on the surface, the deposition rate, the kinetic energy of the deposition species, etc. [11]. In this research, we studied the detailed processing conditions, microstructures, and superconducting properties will be presented and their relation will be discussed.

2. EXPERIMENTAL PROCEDURE

The GdBCO target was sintered by conventional solid reaction method. The starting materials were Gd₂O₃, Ba₂O₃ and CuO. The mixed powder was reground, pressed and shaped into pellets. In order to improve the homogeneity of the targets, sintering was carried out three times after regrinding and repressing. The targets obtained in this way were found to have GdBCO single phase.

The Nd:YAG laser of the 355nm wavelength was used to fabricate GdBCO superconducting films on cleaved and polished (100) SrTiO3 substrates. Focused the laser beam with the energy density 2.5J/cm² was injected into the target to an elliptical spot using lens and a turning mirror outside vacuum chamber. The Nd:YAG laser was operated at a repetition rate of 5Hz. The laser beam had an incident angle of approximately 45° with respect to the target. To investigate the film properties, we performed with oxygen pressure held at 800mTorr in all cases, and we varied the pressure of the distance between target and substrate (d_{TS}) from 5.5 cm to 7 cm and the substrate temperature from 750 ℃-850 ℃. After the GdBCO films were deposited, substrates were cooled down to 500°C. Then the system was backfilled with O2 to 500Torr and finally cooled to room temperature. The crystallinity of GdBCO films were analyzed by X-ray diffraction (XRD) and the surface morphology was observed by the scanning electron microscopy (SEM). T_c and J_c values were measured by the four point probe method.

3. RESULTS AND DISCUSSION

In order to find the high-quality GdBCO films by optimizing the deposition parameters, we have grown a series of samples on $SrTiO_3$ (100) substrates by using the pulsed laser deposition (PLD) with a Nd:YAG laser(355nm). First of all, we investigated that there exist an optimum target-to-substrate distance(d_{TS}) to achieve highest T_c in a given PO_2 of 800mTorr since it is well-known that T_c values of YBCO and REBCO samples are function of oxygen pressure for various d_{TS} . It is known that there is a certain optimum target-to-substrate distance for each oxygen partial pressure in order to produce the excellent superconducting properties [12].

Fig. 1 shows XRD patterns of the GdBCO film with various d_{TS} . Except for the d_{TS} of 5 cm with the mixture of c-axis and random orientation, all films have a good c-axis orientation. The resistance vs. temperature measurements of GdBCO films measured by the standard four probe method are shown in Fig. 2 $T_{c,zero}$ and $T_{c,oneset}$ values of GdBCO films show $T_{c,zero}$ ranging between 63.8K and 89.7K and a transition width($\triangle T = T_{c,onset} - T_{c,zero}$) from 17.7K to 1.3K. High T_c value of 89.7K and narrow transition value of 1.3K were obtained from d_{TS} of 6 cm.

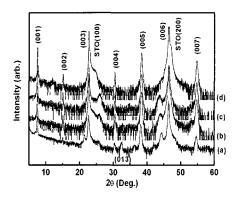


Fig. 1. XRD-patterns of GdBCO films on STO with difference of target-to-substrate.

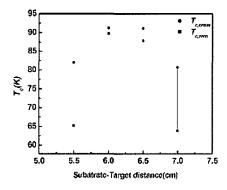
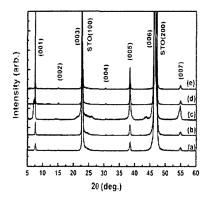


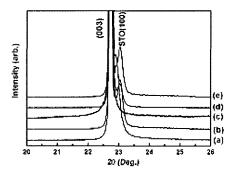
Fig. 2. $T_{c, zero}$ value and $\triangle T_c$ value as a function of target-to-substrate.

The results of I-V measurements also are shown the highest value of $1.76MA/\text{cm}^2$ at d_{TS} of 6 cm. We found that the optimum target-to-substrate is 6 cm for PO_2 of 800mTorr.

Secondly, under the oxygen pressure of 800mTorr and with the target-to-substrate distance of 6 cm, we deposited GdBCO films at various substrate temperature values.

XRD analysis results for these samples are represented in Fig. 3 The typical X-ray diffraction pattern of the films shows the films has grown to be highly c-axis-oriented and no other second phase are observed at substrate temperature $800\,^{\circ}\mathrm{C}$. The film deposited at $800\,^{\circ}\mathrm{C}$ also shows exclusively (00L) peaks with stronger intensity than any other peaks of the different temperature. But a-axis oriented peak of GdBCO (010) peak in vicinity of 23.1° and GdBCO (020) peak in vicinity of 47.3° are observed.





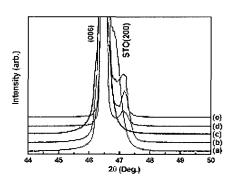


Fig. 3. X-ray diffraction patterns of each GdBCO film deposited at (a) $750\,^{\circ}$ C, (b) $775\,^{\circ}$ C, (c) $800\,^{\circ}$ C, (d) $825\,^{\circ}$ C, and (e) $850\,^{\circ}$ C.

In Fig. 4, SEM micrographs of laser-deposition GdBCO thin films on $SrTiO_3(100)$ substrates show the presence of typically submicron particulates, circle shaped outgrowths. All in the range of substrate temperature, needle-shaped crystals, which are thought to be crystals with a-axis orientation, are hardly observed. In the range of $770\,^{\circ}\text{C}$ -850 $^{\circ}\text{C}$ the surface was smooth, and for $750\,^{\circ}\text{C}$ it became rough. All films have outgrowth particles of similar size and density with different shapes.

In Fig. 5, the GdBCO films had $T_{c.zero}$ values of 78.2K-89.7K. The circles, the triangles, the rhombus, the squares and the polygons represent the resistance curves for the films deposited at 750 °C, 775 °C, 800 °C, 825 °C, 850 °C, respectively. All GdBCO films exhibited a narrow width ($\triangle T_c$) except the film deposited at 750 °C.

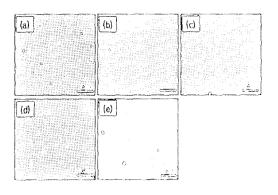


Fig. 4. FE-SEM images of GdBCO films deposited at different substrate temperatures: (a) 750° C, (b) 775° C, (c) 300° C, (d) 825° C, and (e) 850° C.

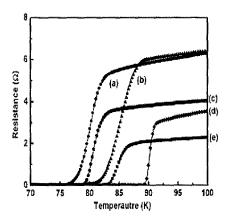


Fig. 5. R-T curves of GdBCO films deposited at $PO_2=800mT$ orr : (a) $750\,^{\circ}$ C, (b) $775\,^{\circ}$ C, (c) $800\,^{\circ}$ C, (d) $825\,^{\circ}$ C, (e) $850\,^{\circ}$ C.

Fig. 6 shows the substrate temperature dependence of Tc of GdBCO films deposited at PO₂ =800mTorr.

Transition temperature versus substrate temperature relations are somewhat scattered, but $T_{c,zero}$ (zero T_c) values of around 90K were successfully attained at the substrate temperature $800\,^{\circ}\mathrm{C}$. The films deposited at higher temperature (800 $^{\circ}\mathrm{C}$) showed better quality but the films of more higher temperature (upward of $800\,^{\circ}\mathrm{C}$) or of more lower temperature (downward of $800\,^{\circ}\mathrm{C}$) drop the quality. The reason is that all the samples except for the sample of substrate temperature $800\,^{\circ}\mathrm{C}$ exhibited a-axis orientation caused the decrease of the $T_{c,zero}$ from the XRD analysis.

The I-V curve for this sample is shown in Fig. 7 he highest Jc value of 1.76MA/cm at 77K was achieved from the sample prepared with substrate temperature $800\,^{\circ}\mathrm{C}$, oxygen pressure of $800\,\mathrm{mTorr}$, and the target-to-substrate distance of $6\,\mathrm{cm}$.

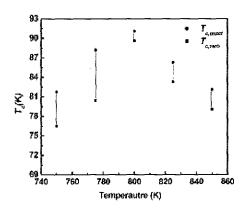


Fig. 6. The substrate temperature dependence of the transition temperature GdBCO films deposited at PO₂=800mTorr.

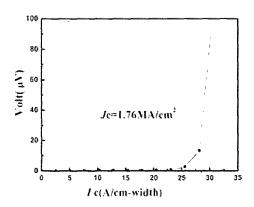


Fig. 7. I-V curve of the GdBCO film deposited on STO.

4. CONCLUSION

We have shown the target-to-substrate distance effect of the $5.5~\mathrm{cm}$ - $7.0~\mathrm{cm}$ range and the substrate temperature effect of the $750~\mathrm{C}$ - $850~\mathrm{C}$ range on the preparation of the superconducting $\mathrm{GdBa_2Cu_3O_{7-x}}$ thin films on the $\mathrm{SrTiO_3}$. For all films prepared with the Nd:YAG laser (355nm) their superconducting properties were quit different according to the target-to-substrate distance and substrate temperature. The highest T_c value of $89.7\mathrm{K}$ and J_c value of $1.76~\mathrm{MA/cm}$ was obtained from the sample prepared with substrate temperature $800~\mathrm{C}$, oxygen pressure of $800~\mathrm{mTorr}$, and the target-to-substrate distance of $6~\mathrm{cm}$. The origin for suppressed T_c values of above and under $800~\mathrm{C}$ is that all the samples except for the sample of substrate temperature $800~\mathrm{C}$ exhibited a-axis.

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