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Effects of indium doping on the superconducting properties of $\text{YBa}_2\text{Cu}_3\text{O}_y$ sintered compounds and thin films

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Abstract

We investigated the effects of indium doping on the superconducting properties of YBCO sintered samples and thin films. In_2O_3 -doped YBCO and $\text{YBa}_2\text{Cu}_{3-x}\text{In}_x\text{O}_y$ sintered samples showed a gradual decrease in the critical temperature (T_c) with increasing indium content; however, a T_c value above 80 K was maintained even up to 30 vol% addition and $x = 0.4$, respectively. $\text{Ba}_3\text{Cu}_3\text{In}_4\text{O}_{12}$ was detected by X-ray diffractometry and energy-dispersive X-ray spectroscopy as a reaction product for both sintered samples. The normalized J_c under a magnetic field of 0.1 T showed a maximum at $x = 0.3$. Indium-doped YBCO films prepared by pulsed laser deposition showed a similar dependence of T_c on indium content as the sintered samples.

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1. Introduction

Enhancement of the critical current density (J_c) of $\text{REBa}_2\text{Cu}_3\text{O}_y$ (REBCO) superconductors is important for the application of RE123 superconducting melt-textured bulk and thin films to high-performance permanent magnets or high-current-carrying wires. Introduction of artificial pinning centers (APC) into RE123 superconducting matrices is a very effective technique for improving J_c under high magnetic fields. Significant improvements in J_c - B properties have been achieved for pulsed-laser deposited REBCO (PLD-REBCO) thin films using REBCO sintered targets with the addition of APC materials such as Y_2BaCuO_5 (Y211) [1], Y_2O_3 [2], BaZrO_3 [3], BaNb_2O_6 [4], BaSnO_3 (BSO) [5], or Au [6]). We reported a good correlation between the stability of APC materials in REBCO films and REBCO sintered compounds, and suggested that BaSnO_3 is a suitable APC material for REBCO films [7]. Actually, BSO-doped YBCO films showed a high global pinning force [5]. This suggests that a study of the doping of various materials into sintered samples would be extremely helpful in the search for new APC materials suitable for high- J_c thin films in high-magnetic fields.

In the present study, we investigated the effects of In_2O_3 addition on the

superconducting properties of YBCO by preparing two kinds of samples: In_2O_3 -containing YBCO and nominal $\text{YBa}_2\text{Cu}_{3-x}\text{In}_x\text{O}_y$ (YBCIO). To our knowledge, there are a few reports on In-doped REBCO superconductors [8-9], but none on the addition of In_2O_3 into the superconducting phase or on In-doped REBCO thin films. In addition to the experiments on sintered samples, we studied the effects of In_2O_3 addition to PLD-YBCO thin films.

2. Experimental

$\text{YBa}_2\text{Cu}_{3-x}\text{In}_x\text{O}_y$ (YBCIO) samples with a nominal composition x of 0-0.5 were prepared from Y_2O_3 (99.9%), BaCO_3 (99.95%), CuO (99.99%) and In_2O_3 (99.999%) using a standard solid-phase reaction technique. Appropriate amounts of the reagents were thoroughly ground and calcined at 1173 K for two periods of 12 h in air, with intermediate regrinding. The resultant YBCIO powder was pressed into pellets and sintered at 1233 K in air for two periods of 12 h, with intermediate regrinding. In the case of preparing In_2O_3 -doped YBCO samples, In_2O_3 powders were added to YBCO powders sintered at 1233 K in concentrations of 2–30 vol%. The resulting powders were thoroughly ground, pressed into pellets and then sintered at 1233 K in air for 12 h. All

samples were subsequently annealed in flowing oxygen at 1183 K for 12 h, cooled to 773 K with a 12 h stay, and then cooled to room temperature in a furnace. In_2O_3 -doped YBCO and YBCIO films were prepared by a pulsed laser deposition technique using sintered In_2O_3 -doped YBCO and YBCIO targets, respectively. The detailed deposition conditions were described in a previous paper [5]. The electrical resistivity of the samples was measured by a standard four-probe technique to determine their critical temperatures (T_c). J_c of the samples at 77 K was calculated from B - M curves measured by a superconducting quantum interference device at magnetic fields of 0-1.0 T. X-ray diffractometry (XRD) was employed to identify the phases present in the samples and determine lattice constants. The sample surfaces were characterized using scanning electron microscopy (SEM). The chemical compositions of precipitates on the sample surfaces were determined by energy-dispersive X-ray spectroscopy (EDX).

3. Results and discussion

Fig. 1(a) shows the T_c of the In_2O_3 -doped Y123 sintered samples as a function of the doping level. Samples with In_2O_3 added showed a gradual decrease in T_c with increasing In_2O_3 content. This T_c behavior is comparable to that found for BaZrO_3 ,

which is a typical APC material for REBCO thin films, and exhibits high stability in the superconducting phase [7, 10]. The In_2O_3 -doped samples maintained a T_c above 80 K even for a doping level of 30 vol%. YBCIO samples showed results similar to those of the In_2O_3 -doped samples, as seen in Fig. 1(b). This suggests that indium doping does not have such a serious effect on the superconducting properties as that reported for the synthesis of $(\text{Y}, \text{In})\text{Ba}_2\text{Cu}_3\text{O}_y$ superconductors [8].

XRD patterns for pure YBCO, 7.8 vol% In_2O_3 -doped YBCO and YBCIO ($x = 0.4$) samples are shown in Fig. 2. A $\text{Ba}_3\text{Cu}_3\text{In}_4\text{O}_{12}$ compound was detected for both the In_2O_3 -doped YBCO and YBCIO samples. The (222) main peak intensity of $\text{Ba}_3\text{Cu}_3\text{In}_4\text{O}_{12}$ ($2\theta \sim 29.55^\circ$) increased with increasing In_2O_3 content for both indium-containing samples. $\text{In}_2\text{Cu}_2\text{O}_5$, which was reported as an impurity phase in the preparation of $(\text{Y}, \text{In})\text{Ba}_2\text{Cu}_3\text{O}_y$ superconductors, was not identified in the indium-containing samples by XRD analysis. In addition, no In_2O_3 were found in the In_2O_3 -doped YBCO samples. These results indicate that $\text{Ba}_3\text{Cu}_3\text{In}_4\text{O}_{12}$ was formed by a reaction of In_2O_3 and YBCO. Y211 was also identified in both indium-containing samples, whereas no BaCuO_2 was detected in either. Y211 was probably produced during the formation of the Ba and Cu-rich $\text{Ba}_3\text{Cu}_3\text{In}_4\text{O}_{12}$ phases. No clear change in the lattice constants calculated XRD peaks was observed for both In_2O_3 -containing YBCO

and YBCIO samples.

SEM images of In_2O_3 -containing YBCO and YBCIO samples are shown in Fig. 3. Fine precipitates indicated by arrows were observed on the surfaces of the 2.1 and 9.5 vol% In_2O_3 -doped YBCO (Figs. 3(a) and (b)). In particular, precipitates with a characteristic square shape were clearly observed for the 9.5 vol% In_2O_3 -doped YBCO samples. There was no appreciable change in the grain size due to the addition of In_2O_3 . The chemical composition of the square precipitates was determined by EDX to be $\text{Ba}:\text{Cu}:\text{In}:\text{Y} = 3.0:2.9:3.3:0.15$. This atomic ratio roughly corresponds to $\text{Ba}_3\text{Cu}_3\text{In}_4\text{O}_{12}$, although a small amount of yttrium was contained in the precipitates. In contrast, such square precipitates were not observed for the YBCIO samples (Figs. 3(c) and (d)). This is possibly due to that most of $\text{Ba}_3\text{Cu}_3\text{In}_4\text{O}_{12}$ particles were formed inside of the YBCO crystal grains. The grain size of the YBCIO samples was found to decrease with increasing x . The J_c - B properties at 77 K were measured for the YBCIO sintered samples.

Fig. 4 shows the dependence of the normalized J_c on x for the YBCIO samples. J_c was determined using Bean's critical state model from the DC magnetization hysteresis loops. $J_c(0.1 \text{ T})$ and $J_c(0)$ indicate J_c values at the self field and 0.1 T, respectively. $J_c(0.1 \text{ T})/J_c(0)$ increases with x and shows a maximum at $x = 0.3$, which indicates that

the $\text{Ba}_3\text{Cu}_3\text{In}_4\text{O}_{12}$ reaction product possibly acts as a pinning center. The decrease in the normalized J_c for the YBCIO samples with x over 0.3 is probably due to suppression of crystal growth of the YBCO phase due to the indium doping, as shown in Fig. 3(d).

Fig. 5 shows the dependence of T_c on the In_2O_3 content of In_2O_3 -doped targets and the indium content x of YBCIO targets for indium-doped YBCO thin films. The T_c values of the indium-doped targets are plotted again for comparison. Both films show a similar dependence of T_c on indium content. The correlation between the T_c of the thin films and that of the sintered targets agrees with the previous studies on barium oxide or RE_2O_3 addition to bulk REBCO [11]. For the thin films prepared using In_2O_3 -doped YBCO targets, a sudden decrease in T_c is observed for 6 vol% In_2O_3 addition. The decrease in T_c with APC amount observed for the In-doped films using the In_2O_3 -doped targets is almost comparable to that for the BSO-doped YBCO films with high pinning potential [5, 7].

4. Conclusions

The effects of indium doping on the superconducting properties of YBCO sintered samples and thin films were investigated. In_2O_3 -doped YBCO and YBCIO sintered

samples maintained a T_c above 80 K even for large amounts of indium addition. It was found that $\text{Ba}_3\text{Cu}_3\text{In}_4\text{O}_{12}$ was formed as a reaction product in the indium-doped sintered samples. $J_c(0.1 \text{ T})/J_c(0)$ was observed to increase with increasing indium doping up to $x = 0.3$. A similar dependence of T_c on the indium doping level was found for both the thin films and the sintered samples. Such studies on the addition of various materials into sintered samples are useful for searching for new APC materials.

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Figure captions

Fig. 1 Dependence of T_c on (a) In_2O_3 content and (b) indium content x for sintered samples.

Fig. 2 XRD θ - 2θ spectra for sintered samples. (a) 7.8 vol% In_2O_3 -doped YBCO (b) YBCIO ($x = 0.3$) and (c) pure YBCO. Closed triangles, open triangles and open circles indicate $\text{Ba}_3\text{Cu}_3\text{In}_4\text{O}_{12}$, Y211 and YBCO phases, respectively.

Fig. 3 SEM surface images of indium-containing sintered samples. (a) 2.1 vol% In_2O_3 -doped YBCO, (b) 9.5 vol% In_2O_3 -doped YBCO, (c) YBCIO ($x = 0.1$) and (d) YBCIO ($x = 0.3$). Square precipitates indicated by arrows were analyzed by EDX.

Fig. 4 Dependence of normalized J_c at 77 K on content x . $J_c(0.1 \text{ T})$ and $J_c(0)$ indicate J_c values at 0.1 T and self field, respectively.

Fig. 5 T_c vs. (a) In_2O_3 content of In_2O_3 -doped targets and (b) indium content x of YBCIO targets for In-doped YBCO thin films. The T_c values of the targets are plotted

again for comparison.

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Fig.2 BLP19/ISS2009

Fig.3 BLP19/ISS2009

Fig.4 BLP19/ISS2009

Fig.5 BLP19/ISS2009