Microstructure and superconducting properties of melt-processed RE-Ba-Cu-O with dispersed barium cerate particles

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Abstract. We studied microstructure and superconducting properties of melt-processed RE123 (RE=Y and Dy) bulk superconductors with a large amount of BaCeO3 addition. The melt-textured bulk samples with compositions of Y123 + xBaCeO3 (x=0.05–0.4) and 0.5 wt% of Pt were fabricated in air. The BaCeO3 particles with the diameter of 1-2 μm were trapped in the melt-textured samples. The critical current density (Jc) in a low field region increased with the amount of BaCeO3 inclusions. This suggests the BaCeO3 particles act as effective flux pinning centers, as well as Y211 particles in conventional melt-textured Y-Ba-Cu-O bulk materials. We found that the size of BaCeO3 particles could be reduced by substituting Zr on Ce site. The Y123 bulk sample with 30 mol% of Ba(Ce0.75Zr0.25)O3 and 0.5 wt% Pt showed a high Jc value of 110 kA/cm² at 77 K. The Dy123 bulk samples dispersed BaCeO3 and Ba(Ce0.75Zr0.25)O3 particles also exhibited excellent Jc–B properties.

1. Introduction

Improvement in trapped fields of melt-processed RE-Ba-Cu-O bulk superconductors leads to the developments of several engineering applications such as magnetic separation systems, superconducting motors and magnetron sputtering devices. The field trapping capability of bulk superconductors is related with the critical current density (Jc) and grain size of REBa2Cu3Oy (RE123). Therefore, many efforts have been performed on the improvement in Jc of melt-processed RE-Ba-Cu-O materials. It well-known that the non-superconducting RE2BaCuO5 (RE211) particles dispersed in the superconducting RE123 phase act as flux pinning centers. The Jc increases with decreasing the size of RE211 inclusions [1-3], since the interface between RE211/RE123 essentially contributes to flux pinning. Recently, some complex perovskite compounds such as (A,B1-x)REBa2O5 (A= U, W and Mo, B=Pt and Zr) [4], RE2Ba4CuMOy (M=Zr, Hf, Nb, Ta, W and Mo) [5] have also been reported to be new artificial pinning centers.

It is known that a small addition of CeO2 is effective in reducing the size of RE211 secondary particles in RE-Ba-Cu-O [6, 7]. The added CeO2 reacts with RE211 to form BaCeO3 particles during the melt-process. However, it is still unclear whether BaCeO3 particles can act as a pinning center. In this work, we have been studied the superconducting properties of melt-textured RE123 (RE=Y and Dy) materials with a large amount of BaCeO3, and found that the BaCeO3 is one of effective flux pinning centers in RE123 bulk superconductors.
2. Experimental

BaCeO$_3$ powder was synthesized by a solid state reaction of BaO and CeO$_2$ at 1273 K for 4 h. The particle size of BaCeO$_3$ powder was determined to be 0.8 μm by a scanning electron microscopic (SEM) observation. The powders in molar ratios of Y$_{123}$:BaCeO$_3$ = 100:x (x = 5-40) were thoroughly mixed. 0.5 wt% of Pt was added to some raw materials. The mixtures were uniaxially pressed into pellets 20 mm in diameter and 12 mm in thickness. The cold-seeding method was applied in air for melt-processing of bulk samples. A melt-textured Nd$_{123}$ (001) bulk crystal about 2 mm in diameter was used as a seed and placed on the center of the pellet. The pellets were heated to 1313 K and held for 2 h, cooled to 1293 K in 1 h, then cooled at a rate of 0.3-0.5 K h$^{-1}$ to 1253 K and finally cooled at a rate of 100 K h$^{-1}$ to room temperature.

The melt-textured bulk samples were annealed in flowing oxygen at 723 K for 100 h. For the measurements of the superconducting properties, rectangular pieces with dimensions of about 2 × 2 × 1 mm$^3$ were cut from the bulk samples. Critical temperature ($T_c$) was measured with a Quantum Design SQUID magnetometer in the zero-field-cooled (ZFC) mode applying magnetic field of 1 mT. For the determination of $J_c$, magnetization loops were measured for $H$//c at 77 K [8].

3. Results and discussion

In each sample with BaCeO$_3$ addition, a single grain growth occurred from a seed crystal. Figure 1 shows the SEM photograph of the sample with the composition of Y$_{123}$ + 30 mol% BaCeO$_3$ and 0.5 wt% Pt. White particles of 1-2 μm diameters were observed in the whole area of the sample and identified to be BaCeO$_3$ by energy dispersion X-ray spectrometry (EDS).

Figure 2 shows the temperature dependence of dc susceptibility for the samples with the composition of Y$_{123}$:BaCeO$_3$ = 100:30. In addition, figure 3 shows the $J_c$-$B$ curves for the Pt-free Y$_{123}$ sample with 30 mol% of BaCeO$_3$ and the Pt-added Y$_{123}$ samples with various BaCeO$_3$ contents. As shown in figure 2, onset-$T_c$ for Pt-free sample is 89.5 K, which is lower than that for conventional Y$_{123}$ bulk material (90-91 K). Such a low value of $T_c$ led to lowering $J_c$ properties as shown in figure 3. We found however that a small addition of Pt improved superconducting properties and the onset-
$T_c$ reached 90.5 K with a sharper transition. The $J_c$ and irreversibility field were also enhanced with an improvement in $T_c$. Pt addition generally leads to the size reduction of Y211 particles formed by the peritectic decomposition of Y123 phase during the partial melting stage. Dispersion of fine Y211 particles prevents the loss of Ba-Cu-O liquid component and thus maintains the initial composition, which will keep the quality of sample [9]. In the $J_c$-$B$ curves for the Pt-added samples, the $J_c$ in a low field region increased with increasing BaCeO$_3$ content. These results indicate that the BaCeO$_3$ inclusions act as effective flux pinning centers, as well as Y211 particles in conventional melt-textured Y-Ba-Cu-O bulk materials.

If the size of BaCeO$_3$ particles decreases, it is possible to increase $J_c$ in the Y123 samples. To reduce the particle size of BaCeO$_3$, we employed finer ball-milled BaCeO$_3$ powder as a starting material. Figure 4 shows the $J_c$-$B$ curve of the sample fabricated from the precursor of Y123 with 30 mol% of the finer BaCeO$_3$ powder and 0.5 wt% Pt. The $J_c$ increased with the employment of finer BaCeO$_3$ powder, since the size of BaCeO$_3$ inclusions was decreased. The partial substitution of Ce$^{4+}$ ions in BaCeO$_3$ by Zr$^{4+}$ ions was also effective in reducing the particle size of BaCeO$_3$. Figure 5 shows the SEM photograph of the sample with the composition of Y123 + 30mol% Ba(Ce$_{0.75}$Zr$_{0.25}$)O$_3$ and 0.5 wt% Pt, where Ba(Ce$_{0.75}$Zr$_{0.25}$)O$_3$ was prepared by a solid state reaction of BaO, CeO$_2$ and ZrO$_2$. Here it is notable that the size of Ba(Ce, Zr)O$_3$ particles was smaller than that of BaCeO$_3$ presented in figure 1. As displayed in figure 4, the Y123 bulk sample with 30 mol% of Ba(Ce$_{0.75}$Zr$_{0.25}$)O$_3$ exhibited a high $J_c$ value of $1.1 \times 10^5$ A cm$^{-2}$ at 77 K in self field.

The preparation and properties of Dy123 with BaCeO$_3$ and Ba(Ce, Zr)O$_3$ were also investigated. Figure 6 shows the $J_c$-$B$ curves at 77 K for the melt-textured Dy123 samples with (a) 5 mol% of BaCeO$_3$ (b) 30 mol% of BaCeO$_3$ and (c) 30 mol% of Ba(Ce$_{0.75}$Zr$_{0.25}$)O$_3$. From the comparison between (a) and (b), $J_c$ in a low field region increased with increasing BaCeO$_3$ content as in the case of Y123 system. In addition, the Ba(Ce$_{0.75}$Zr$_{0.25}$)O$_3$ added Dy123 bulk sample exhibited excellent $J_c$–$B$ properties of $1.15 \times 10^5$ A cm$^{-2}$ and $5.0 \times 10^4$ A cm$^{-2}$ at 0 T and 3 T, respectively.

4. Summary
We investigated the superconducting properties of melt-textured Y123 bulk with different BaCeO$_3$

![Figure 3. $J_c$-$B$ curves at 77 K for the melt-textured Y123 samples with various BaCeO$_3$ contents.](image1)

![Figure 4. $J_c$-$B$ curves at 77 K for the melt-textured Y123 samples with 30 mol% of BaCeO$_3$ (a) without ball-milling and (b) with ball-milling treatment, and (c) the Y123 sample with 30 mol% of Ba(Ce$_{0.75}$Zr$_{0.25}$)O$_3$.](image2)
additions. The BaCeO$_3$ particles with the diameter of 1-2 μm were dispersed in the melt-textured samples. The Y123 bulk samples with 5-40 mol% of BaCeO$_3$ exhibited onset-$T_c$ of 90.5 K, when a small amount of Pt was added. The $J_c$ in a low field region increased with BaCeO$_3$ content. The size of BaCeO$_3$ particles could be reduced by the employment of fine BaCeO$_3$ starting powder or by substituting Zr on Ce site, leading to an enhancement in $J_c$. The Dy123 bulk samples dispersed BaCeO$_3$ and Ba(Ce, Zr)O$_3$ particles also exhibited good $J_c$–$B$ properties. The BaCeO$_3$ can be employed as novel artificial pinning centers in melt-textured RE123 bulk superconductors.

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