Effect of BaCeO₃ and BaSnO₃ Additives on Microstructural Development and Critical Current Density of Melt Textured YBa₂Cu₃O₇-x

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Jongprateep, Oratai; Dogan, Fatih; Strasik, Michael; and McCrary, Kevin E., "Effect of BaCeO₃ and BaSnO₃ Additives on Microstructural Development and Critical Current Density of Melt Textured YBa₂Cu₃O₇-x" (2005). Faculty Research & Creative Works. Paper 519.
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Effect of BaCeO₃ and BaSnO₃ Additives on Microstructural Development and Critical Current Density of Melt Textured YBa₂Cu₃O₇₋ₓ

Oratai Jongprateep, Fatih Dogan, Michael Strasik, and Kevin E. McCrary

Abstract—Improvement of superconducting properties, such as critical current density \( J_c \) and trapped magnetic field, of melt textured YBa₂Cu₃O₇₋ₓ (Y123) require introducing of effective pinning sites, e.g., nonsuperconducting inclusions, twin boundaries and other defects. It has been shown that addition of small quantities of BaCeO₃ (<0.5 wt%) into Y123 results in an increase of the \( J_c \). However, higher cerium concentrations affect the solidification process and inhibit the growth of melt textured Y123 single crystals. In this study, the effect of BaCeO₃ additions on the growth and microstructural development of melt textured Y123 single crystals were investigated. The relationship between the solidification kinetics, microstructural development and superconducting properties of Y123 melt textured single crystals with cerium additives is discussed.

Index Terms—Cerium oxide, critical current density, Y123.

I. INTRODUCTION

ONE OF THE critical requirements for practical applications of high \( T_c \) superconductor is the high critical current density \( J_c \). Improvement in the electrical and magnetic properties of Y123 can be achieved by enhancement of flux pinning. The interface between the superconducting matrix and second-phase particles can play a significant role in the enhancement of \( J_c \) [1], [2]. Doping of Y123 with cerium has been known to improve the critical current density in Y123 via refinement of Y211 particles and formation of sub-micron sized BaCeO₃ particles in Y123 [3]–[10]. However, it was found that addition of larger amounts of cerium oxide (>10 wt% BaCeO₃) inhibits growth of large melt textured single crystals. On the other hand, addition of BaSnO₃ was found to be effective for continuous crystal growth during the solidification process and led to refinement of Y211 particles in Y123 [11]–[16]. It was also shown that \( J_c \) of Y123 increases with small additions of BaSnO₃ (<1 wt%).

In this study, Y123 samples with large amounts of BaCeO₃ (5–15 wt%) and small amounts of BaSnO₃ (0.5–5 wt%) were processed. The experiments were aimed to determine the optimal amount of BaCeO₃ in Y123 that would result in faster solidification kinetics and higher \( J_c \). The role of BaCeO₃ and BaSnO₃ additives and their amounts on the microstructural development and superconducting properties of Y123 melt textured single crystals were studied.

II. EXPERIMENTAL

A. Sample Preparation

The Y123 pellets were prepared by the top-seeded melt-textured method. The initial powder was prepared by mixing of Y₂O₃, CuO, BaCO₃, SnO₂, and CeO₂. (99.9% Alfa Aesar, Ward Hill, MA) powders followed by calcinations at 920 °C for three times with intermediate grinding. The starting compositions were Y123 with 5 wt% BaCeO₃ and 0.5 wt% BaSnO₃, Y123 with 10 wt% BaCeO₃ and 2.5 wt% BaSnO₃, Y123 with 10 wt% BaCeO₃ and 5 wt% BaSnO₃, Y123 with 15 wt% BaCeO₃ and 2.5 wt% BaSnO₃, and Y123 with 15 wt% BaCeO₃ and 5 wt% BaSnO₃. The powders were uniaxially pressed to obtain pellets with 25 mm diameter under a pressure of 100 MPa. A Sm123 crystal was placed on top center of the pellet as a seed crystal for solidification of Y123. The powder compacts were heated from room temperature to 1050 °C at a heating rate of 47 °C/h and held at 1050 °C for 0.5 h. The samples were cooled rapidly to 1010 °C followed by slow cooling between 1010-990 °C at a rate of 0.3 °C/h. The solidified samples were then oxygenated at 600 °C for 7 days under flowing oxygen. For magnetometer measurements, samples were prepared in size of ~4 × 4 × 2 mm³.

B. Microstructural Analysis

The microstructure was characterized by scanning electron microscopy (SEM JEOL-T330A) techniques. The samples used in the characterization were cut from the edge of the pellets. The size distribution of Y211, BaCeO₃ and BaSnO₃ particles were analyzed using Scion Image Software. Due to the highly anisotropic shape of needle-like Y211 phase, the width of the particles was measured for the analysis. A minimum of 60 particles was counted to obtain the average size of the particles.

C. \( J_c \) Measurements

Critical current densities, \( J_c \), of the samples were measured with a SQUID magnetometer (Quantum Design PPMS). An external field from ~5 to 5 T was applied along the caxis of the samples. From the M–H loop and the Bean critical state...
model [17], [18], $J_e$ values were determined using the following relationship:

$$J_e(H) = \frac{20[M(H) - M(D)]}{A(1 - A/3B)}$$

(1)

where $M$ is the sample magnetization in the field $H$ in emu/cm$^3$; $A$ and $B$ ($A < B$) are the dimensions (in centimeters) of samples in (00l) plane.

III. RESULTS AND DISCUSSION

A. Microstructural Development

Fig. 1 shows formation of needle or pillar shaped Y211 particles in Y123. BaCeO$_3$ and BaSnO$_3$ are revealed as highly faceted and fine particles with the size in sub-micrometer range. In general, BaCeO$_3$ and BaSnO$_3$ particles were well-dispersed at lower concentrations of additives. However, for the sample with higher amount of the additives, it was found that the particles agglomerated locally as shown in Fig. 2. It is known that small inclusions such as BaZrO$_3$ (<100 nm) in semi-liquid phase of Y123 above the peritectic decomposition temperature segregate during the solidification process and form large clusters of particles [19].

B. Effect of Additives on Crystal Growth

The size of the single crystals, initiated at the seed crystal of the sample, varied with the composition of the additives. The higher ratio of BaCeO$_3$ to the total amount of additives (BaCeO$_3$ and BaSnO$_3$) resulted in earlier termination of the crystal growth and smaller crystal sizes (Fig. 3). This may be attributed to the particle pushing phenomenon at the solidification front during the crystal growth process.

The effect of BaCeO$_3$ concentration on the size of Y211 particles in Y123 was studied. The size of Y211 particles was within the range of 1.1–3.1 μm. It appeared that increasing amounts of BaCeO$_3$ addition resulted in coarsening of Y211 particles as shown in Fig. 4. The slower dissolution rate of coarser Y211 inclusions reduces the solidification rate of Y123 because of slow dissolution rate of larger particles in the melt. This also could give rise to particle pushing at the growth interface resulting in earlier termination of crystal growth. Cerium addition can alter the kinetics of the Y211 coarsening according to the following equation [7], [20]:

$$R = (D/\Gamma t)^{1/3}$$

(2)

where $R$ is the mean radius for Y211 particles, $\Gamma$ is the Gibbs–Thompson coefficient, $t$ is the holding time of Y211 particles in the melt, and $D$ is a diffusion constant.
TABLE I

<table>
<thead>
<tr>
<th>Sample Composition</th>
<th>Critical Current Density (10^4 A/cm^2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>5 wt% BaCeO_3+ 0.5 wt% BaSnO_3</td>
<td>3.2</td>
</tr>
<tr>
<td>10 wt% BaCeO_3+ 2.5 wt% BaSnO_3</td>
<td>10.9</td>
</tr>
<tr>
<td>10 wt% BaCeO_3+ 5 wt% BaSnO_3</td>
<td>16.0</td>
</tr>
<tr>
<td>15 wt% BaCeO_3+ 2.5 wt% BaSnO_3</td>
<td>6.6</td>
</tr>
<tr>
<td>15 wt% BaCeO_3+ 5 wt% BaSnO_3</td>
<td>12.2</td>
</tr>
</tbody>
</table>

Fig. 5. Effect of ratio of BaCeO_3 to total amount of additives (BaCeO_3 and BaSnO_3) in Y123 on critical current density at zero magnetic field.

It is believed that the addition of small amounts of cerium—based compounds can increase the viscosity of the semi—solid melt. This may lead to a decrease in the diffusion constant. Thus, the coarsening of the Y211 particles is hindered as a result of BaCeO_3 addition at small amounts [7]. However, as the amount of BaCeO_3 addition increases, the size of Y112 increases as shown in Fig. 4. The coarsening mechanism of Y211 inclusions at higher amounts of BaCeO_3 additions is not understood at this time and requires further systematic studies.

C. Critical Current Density

The critical current density measurements were conducted using a SQUID magnetometer at 77 K. The magnetic field (−5T<H<5 T) was applied and the corresponding magnetization (M) was recorded. The Jc values were calculated from the M–H loop using the extended Bean’s critical state model. Jc values of samples with different amounts of additives are shown in Table I with increasing BaCeO_3 concentration. The Jc of the samples was in the range of 32–160 kA/cm^2 measured at 77 K in zero magnetic field. It appears that increasing amount of BaSnO_3 results in higher Jc. Fig. 5 reveals the relationship between the critical current density and the ratio of BaCeO_3 to total amount of additives. It is shown that the critical current density is reduced with increasing ratio of BaCeO_3/BaCeO_3+BaSnO_3 concentration.

Coarsened Y211 particles and local agglomeration of BaCeO_3 particles may also result in the reduction of the critical current density of the samples. Since the interface between the superconducting matrix and second phase particles acts as flux pinning sites, large agglomerated particles result in reduced interfacial area within the superconducting matrix. This could lead to a decrease of available flux pinning sites, which in turn results in lowering of Jc.

IV. Conclusion

The effect of BaCeO_3 and BaSnO_3 additions on the growth kinetics, microstructural development and superconducting properties of melt textured Y123 was studied. It was shown that an excessive amount of BaCeO_3 led to the agglomeration of these particles within the Y123 matrix and a coarsening of Y211 particles. Increasing amounts of BaCeO_3 concentration in Y123 resulted in lowering of Jc while higher amounts of BaSnO_3 give rise to an increase of Jc.

ACKNOWLEDGMENT

The authors also would like to acknowledge Dr. J. Switzer and H. Kothari for the use of SQUID magnetometer.

REFERENCES


Fatih Dogan received the Ph.D. degree in materials science and engineering from the Technical University of Berlin, Berlin, Germany, in 1989. Since 2002, he has been a Professor at the University of Missouri-Rolla. His current research interests include multifunctional electronic ceramics and composites; nanoscale science and engineering; solidification and crystal growth; high-temperature superconducting materials, dielectrics, piezoelectrics, thermophotovoltaic emitters, mixed ionic electronic conductors, and solid oxide fuel cells.

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Oratiai Jongprateep was born in Bangkok, Thailand, in 1977. She received the B.S. and the M.S. degree in materials science and engineering from Columbia University, New York, in 2000 and 2002, respectively. She is now working toward the Ph.D. degree at the University of Missouri-Rolla. Her current research interests concern high-temperature superconducting materials and rapid prototyping.