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Microstructure and Properties of Y-123/Y-211 Bulk Superconductors with BaCeO₃ and BaO₂ Addition

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YBa₂Cu₃O_{7- δ} (YBCO) bulk single-grain superconductors were prepared by a TSMG process. CeO₂, BaCeO₃ and BaO₂ powders were added to nominal composition Y_{1.5}Ba₂Cu₃O₇ with the aim to refine Y₂BaCuO₅ (Y-211) secondary particles. Added powders were refined by milling in a friction milling and characterized by X-ray powder diffractometry. Thermal properties of the systems were characterized by DTA. Microstructure of samples was studied by PLM. Successful refinement of Y-211 particles size was confirmed by IP measurements. Transition temperatures and field dependences of the magnetic moment for further determination of the critical current densities were obtained using a VSM.

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1. Introduction

The Top Seeded Melt Growth (TSMG) process is one of the most effective method to fabricate large singlegrain YBCO bulk superconductors. The important microstructural component of YBCO bulk single-grain superconductors are small Y-211 particles trapped in to YBCO matrix [1]. It is well known, that addition of Pt or Ce can effectively refine Y-211 particles [2]. For practical applications a cheaper Y-211 refinement by Ce should be preferred. It was shown that Ce in the form of $BaCeO_3$ is more effective than addition in the form of CeO_2 [3]. Non-superconducting Y-211 particles have a strong influence on the pinning of magnetic flux lines in the melt-grown YBCO bulks. Critical current density, J_c , increases with the $\mathrm{V}_{211}/\mathrm{d}_{211}$ ratio (V_{211} - volume fraction and d_{211} - size of Y-211 particles) [4]. It means that the local differences in the size and concentration of Y-211 particles could cause the inhomogeneity of J_c in the samples. In this contribution we studied the influence of BaO_2 addition on the refinement of Y-211 particles, which strongly affects on magnetic properties of YBCO superconductors.

2. Experimental

Five different YBCO compositions were fabricated by TSMG process. The composition of 1 mol $Y_{1.5}Ba_2Cu_3O_y$ + 0.25 mol Y_2O_3 with addition of CeO₂ or BaCeO₃ and BaO₂ powders were mixed with appropriate amounts (Table) in a mixer for 30 min, and then intensively milled for 15 min in a friction mill. Mixture of powders was uniaxially pressed into the cylindrical pellets of 20 mm in diameter. SmBa₂Cu₃O_x single crystals were placed in the middle of the top surface of pellets as nucleation seeds. The samples were grown in a chamber furnace with the time/temperature profile optimized for high Y-123 crystal quality [5]. Microstructure analyses were investigated by polarized light microscopy (PLM). The quantitative microstructural data of Y-211 particles were obtained using image processing software. TABLE

Appropriate amounts of powders CeO_2 , $BaCeO_3$ and BaO_2 in mol added to nominal composition of $Y_{1.5}Ba_2Cu_3O_y + 0.25 Y_2O_3$.

	S1	S2	S3	S4	S
CeO_2	0.042	0.042	-	-	0.042
$BaCeO_3$	-	-	0.22	0.22	-
BaO_2	0.142	0.284	0.142	0.284	-

Thermal properties of $Y_{1.5}Ba_2Cu_3O_y$ with additions of CeO₂, BaCeO₃ and BaO₂ powders were investigated by Differential Thermal Analysis (DTA) in the temperature range from 40 °C to 1085 °C in an artificial air atmosphere. Small samples for oxygenation process, performed at 400 °C for 150 hours in a flowing oxygen atmosphere, and magnetization measurements had a shape of a slab with the dimensions $2 \times 2 \times 1$ mm³. The smallest dimension was parallel to the *c*-axis of the crystal.

The magnetization measurements were performed in a vibrating-sample magnetometer (VSM) with magnetic fields up to 6 T applied parallel to the *c*-axis of the crystal, at a temperature of 77 K. The critical current densities, J_c , were calculated using the extended Bean model [6] for rectangular samples. The critical transition temperatures, T_c , were determined from the magnetic transition curves taken after zero-field cooling as the $T_c(50\%)$, of this curve in an applied external magnetic field of 2 mT.

3. Results and discussion

During the fabrication of the samples the most important role by the TSMG process is to find a temperature window for the epitaxial growth of YBCO single-grain bulk superconductors [5]. The window below the temperature of heterogeneous nucleation from the seed and beyond the temperature of self-nucleation of the Y-123 phase is very narrow. The quality of the grown samples depends on the position in this window. Prepared samples exhibited growth of small Y-123 crystal from the seed

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but there were also observed self- nucleated Y-123 crystals in the rest parts of the pellets. Only sample S was grown as single-grain crystal. From the results of DTA measurements (Fig. 1) it is clearly seen that the standard composition, S, exhibits two peaks which correspond to two peritectic reactions. Presence of BaO_2 in the system prevents formation of CuO. Therefore the first peritectic reaction below 950 °C does not appear, and the temperature of second peritectic reaction shifts from 1020 °C to 1040 °C in the compositions S1-S4.



Fig. 1. DTA curves of mixed nominal powders of standard composition (S) and compositions with BaO_2 additions (S1-S4).

The bimodal distribution of Y-211 particles is typical for the samples with the standard composition, S. The samples (S1-S4) also have bimodal distribution, but with higher amount of submicron Y-211 particles. The size of Y-211 particles is much smaller for the samples with BaO2 addition and the size depends also on the form of Ce addition. The mean particle size of Y-211 (d_{211}) measured by image processing was $d_{211} = 0.7 \ \mu m$ for the samples with BaCeO₃ addition and $d_{211} = 0.9 \ \mu m$ for the samples with CeO₂ addition. In the standard sample Y-211 particles have mean size of 1.8 μm .



Fig. 2. Critical temperatures, T_c , of prepared YBCO samples with BaO₂ addition.

Magnetization measurements of T_c with different amount of BaO₂ addition are shown in Fig. 2. We can see that the samples with lower amount of BaO₂ addition (S1 and S3) lead to higher T_c . The value of critical temperatures in the case when Ce is added as BaCeO₃ $(T_c(50\%)S1 = 90.84 \text{ K})$ or CeO₂ $(T_c(50\%)S3 = 90.78 \text{ K})$ are almost the same. The sharp transition to normal state occurred in all prepared samples (S1–S4) but the sharpest one was for the sample S3.



Fig. 3. Field dependences of critical current densities, $J_c(B)$ for prepared YBCO samples with BaO₂ addition.

The critical current densities, J_c , for all prepared samples at 77 K are shown in Fig. 3. We can see an increase of J_c value for sample S4 in comparison to J_c of the standard YBCO sample S. In the case of S1-S3 samples we have lower value of J_c , probably due to significant pushing effect of the Y-211 particles, and thus low concentration of Y-211 particles in measured pieces.

4. Conclusions

The influence of BaO_2 addition on microstructure and magnetic properties of YBCO bulk superconductors with Ce addition for Y-211 particle refinement was investigated. Changes in the peritectic temperature were observed when BaO_2 was added. The main positive contribution of BaO_2 is the significant refinement of Y-211 particles and thus effect of increasing critical current density, when Ce is added in the form of $BaCeO_3$. The lower amount of BaO_2 addition leads to higher value of critical temperature.

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