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Single grain (LRE)-Ba-Cu-O superconductors fabricated by top seeded melt growth in air

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Abstract. We have recently reported a practical processing method for the fabrication in air of large, single grain (LRE)-Ba-Cu-O [where LRE = Nd, Sm, Eu and Gd] bulk superconductors that exhibit high T_c and high J_c. The process is based initially on the development of a new type of generic seed crystal that can promote effectively the epitaxial nucleation of any (RE)-Ba-Cu-O system and, secondly, by suppressing the formation of (LRE)/Ba solid solution in a controlled manner within large LRE-Ba-Cu-O grains processed in air. In this paper we investigate the degree of homogeneity of large grain Sm-Ba-Cu-O superconductors fabricated by this novel process. The technique offers a significant degree of freedom in terms of processing parameters and reproducibility in the growth of oriented single grains in air and yields bulk samples with significantly improved superconducting and field-trapping properties compared to those processed by conventional top seeded melt growth (TSMG).

1. Introduction

(RE)-Ba-Cu-O (where RE = Y, Yb, Tm, Er, Ho and Dy) bulk superconductors can be processed relatively easily in air in the form of large, single grains by the so-called top seeded melt growth (TSMG) technique [1] with typical superconducting transition temperatures, T_c's, of up to 92 K. Importantly from an applications perspective, these materials have been shown to trap large magnetic fields compared to those generated by conventional iron-based and rare earth permanent magnets [2-4]. LRE-Ba-Cu-O bulk superconductors based on light rare earth (LRE) elements such as LRE= Nd, Sm, Eu and Gd, on the other hand, need to be processed in a controlled atmosphere in order to suppress the formation of LRE/Ba solid solution [5,6], although these materials do generally exhibit higher T_c's of up to 96 K. Significantly, the critical current density, J_c, and irreversibility fields of bulk (LRE)BCO superconductors at a given temperature are generally higher than in YBCO [7-9]. However, the successful fabrication of single grain (LRE)BCO bulk materials to date has been achieved only by a very few, mainly academic, research groups [10-12] due primarily to the difficulty of controlling solid solution formation and the lack of availability of a suitable seed crystal, which are key factors in the processing of these materials. Recently a practical processing method for the fabrication of (LRE)BCO single grains has been developed by Hari Babu *et al* at the IRC in Superconductivity [13]. The process is based initially on the development of a new type of generic seed crystal that can promote effectively the epitaxial nucleation of any (RE)-Ba-Cu-O system [14] and, secondly, the suppression of the formation of (LRE)/Ba solid solution in a controlled manner within large LRE-Ba-Cu-O grains processed in air. This paper describes the spatial variation of superconducting properties of single grain (LRE)BCO grains fabricated using this recently developed technique.

2. Experimental

$SmBa_2Cu_3O_{7-\delta}$ (Sm-123) and Sm_2BaCuO_5 (Sm-211) single phase powders were synthesised via a solid state reaction process. Sm-123 + 30 wt% Sm-211 + x wt% BaO_2, where x = 0, 1, 2, 4, and 10 precursor powders were mixed thoroughly using a mortar and pestle and pressed uniaxially into cylindrical pellets. Excess BaO_2 was added to the precursor composition to suppress Sm/Ba solid solution [15,16]. A small Mg-doped NdBCO melt processed grain with dimensions of approximately 1.5 x 1.5 x 1 mm^3 was placed on top of each pellet with the *ab*-plane of the seed in direct contact with the surface of the sample. Differential thermal analysis (DTA) of a seed crystal of starting composition Nd-123 + 12 mol% Nd-422 + 1w% MgO, obtained from its parent melt processed bulk, is shown in Fig. 1. The peritectic decomposition temperatures of various RE-Ba-Cu-O systems are also indicated in this figure. It can be seen that the melting point of the Mg-doped seed is at least 15 ^{O}C higher than that of any other REBCO system. The preparation and properties of Mg-doped seed crystals are reported in detail in [14]. The pellet and seed crystal arrangement was placed in a box furnace, heated rapidly to 1080 ^{O}C, held for 1 hour to ensure complete peritectic decomposition of the Sm-123 phase, cooled rapidly to 1050 ^{O}C, cooled slowly to 1010 ^{O}C at the rate of 1 ^{O}C /h and furnace cooled to room temperature. Finally, the pellets were annealed at 350 ^{O}C in a flowing O_2 atmosphere for 100 hours prior to characterisation. The annealed samples were polished to investigate their microstructural features in detail.

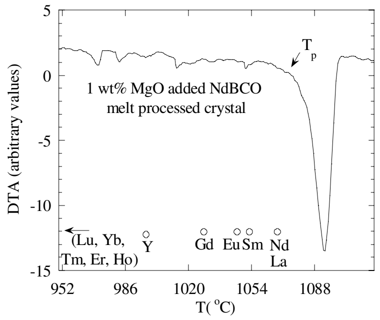

Figure 1. DTA signal of a melt processed single grain of composition Nd-123 + 12mol% Nd-422 + 1wt% MgO. The open circles indicate the peritectic temperatures of other undoped (RE)BCO systems.

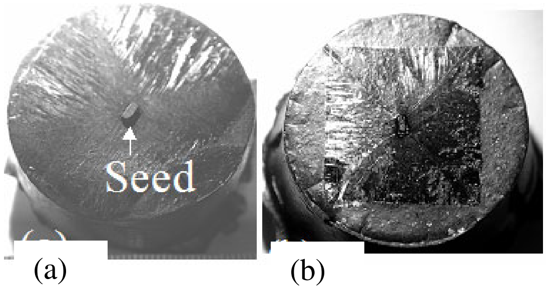

Figure 2. Top view of SmBCO single grains melt processed using a Mg-doped Nd-Ba-Cu-O seed in air for SmBCO containing (a) 1 wt% and (b) 10 wt% BaO_2. Sample diameter is 20 mm.

3. Results and discussion

Figure 2 shows the top view of two SmBCO single grains fabricated by TSMG and containing an excess of (a) 1 wt% and (b) 10 wt% BaO_2. Both samples exhibit four growth facet lines, which is characteristic of the TSMG technique. The square-shaped geometry of each grain indicates that the orientation of the crystal is similar to that of the seed. Pole figure analysis confirmed further that the *c*-axis of each SmBCO grain is perpendicular to the top surface of the pellet. The grain size in Fig. 2(b) is smaller than that in Fig. 2(a), suggesting further that the growth rate of Sm-123 is affected by the presence of excess BaO_2 in the precursor composition. In addition, the average size of Sm-211 phase inclusions in the bulk Sm-123 matrix is reduced from ~ 3 to 1.5 μm by increasing the BaO_2 in the precursor pellet from 0 to 4 wt%BaO_2, which is potentially important for increased flux pinning.

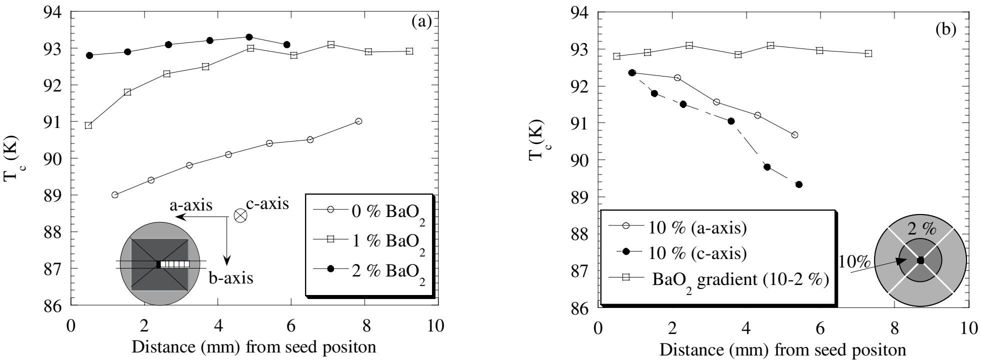

Figure 3. Spatial variation of T_c for single SmBCO grains processed in air for (a) SmBCO enriched with 0, 1, and 2 wt% BaO_2 and (b) SmBCO enriched with 10 wt% BaO_2. Data for a single grain containing an internal variation in the concentration BaO_2 oxide from 10 to 2 wt% is also shown in (b).

The spatial variation of transition temperature, T_c, in SmBCO grains containing BaO_2 was measured using a SQUID magnetometer. Small samples of dimensions (1 ± 0.2) x (1 ± 0.2) x (0.5 ± 0.2) mm^3 were cut from large grains parallel to the *a* and *c*-axes of the crystal. The onset T_c of each specimen, determined as the temperature at which the magnetic moment becomes measurably diamagnetic in the presence of a magnetic field of 2 mT, is plotted in Fig. 3(a) as a function of distance from the seed crystal for samples containing 0, 1, and 2 wt% BaO_2. The inset to this figure illustrates the position of each specimen within the original grain. A lower T_c with a broad transition width up to 15 K (not shown in the figure) is observed generally for the SmBCO grains grown without added BaO_2 due to the formation of a $Sm_{1+x}Ba_{2-x}Cu_3O_{7-\delta}$ solid solution phase. T_c is observed to increase to 91 K with a relatively sharp transition at the position of the seed, however, for the sample containing 1 wt% BaO_2 and to increase further and continuously to 93 K with increasing distance from the seed. Increased T_c and reduced transition width suggests that the formation of a Sm/Ba solid solution phase is suppressed by the addition of BaO_2. A similar, continuous increase in T_c with distance from the seed (i.e. grain nucleation site) and associated suppression of solid solution formation has been observed previously in NdBCO processed under reduced oxygen partial pressure (PO_2) [11,12]. It is thought that this is due to a continuous change in the composition of the liquid during the (LRE)-123 solidification process as the solid solution phase forms [15]. An effectively constant, high T_c (~ 93 K) with a very sharp transition width (ΔT_c < 1.4 K) is observed throughout the SmBCO sample by increasing the BaO_2 content to 2 wt%, as shown in Fig. 3(a). Similarly sharp transitions with high, constant T_c's were also observed along the *c*-axis for this grain [not shown in Fig. 3(a)]. However, a spatial variation of T_c was observed in single grain samples with higher concentrations of BaO_2 of up to 10 wt %. In contrast to the samples processed with lower BaO_2 concentration, the T_c within single grain containing 10 wt % BaO_2 is observed to decrease progressively from 92.4 K to 90.6 K with increasing distance from the seed along both the *a* and *c*-axes, as shown in Fig. 3(b). The origin of the observed decrease in T_c as the crystal growth proceeds is unclear and needs be studied further.

A precursor pellet with a concentration gradient of BaO_2 was melt processed into a single grain in order to investigate the variation of growth rate with BaO_2 content (growth rate is observed to decrease significantly for higher concentrations of BaO_2), whilst retaining a constant T_c within the grain. For this purpose, Sm-123 + 30 wt% Sm-211 precursor powder containing 10 wt% BaO_2 was pressed initially into a pellet of diameter 3 mm. The sample was then placed at the center of a 20 mm diameter die, surrounded with precursor powder of composition Sm-123 + 30 wt% Sm-211 + 2 wt% BaO_2, and re-pressed, as illustrated in the inset to Figure 3(b). This composite green body was melt processed in air as described in the experimental section. In this case, a single grain could be grown within the entire pellet due to the increased growth rate in the outer region of the sample, compared to that at the sample center. The spatial variation in T_c for this grain following annealing in O_2 is shown in Fig. 3 (b). The measured magnetic hysteresis loops at 77 K were observed to exhibit the peak effect that is seen commonly in samples processed under reduced PO_2 atmosphere, suggesting qualitatively that the flux pinning behavior in these samples is similar to that of single grains processed under reduced PO_2. The critical current, J_c, of the SmBCO grains is measured to be 40 - 60 kA/cm^2 at 77 K depending on the concentration of Sm-211 second phase particles, which is comparable to that observed for samples processed under reduced PO_2 [5,6]. In addition, the irreversibility field of the SmBCO grains was measured to be up to 7 T at 77 K. Finally, trapped field measurements were performed on single grains prior to cutting into specimens for T_c characterisation. Initially, each single grain was cooled to liquid nitrogen temperature (77 K) in a magnetic field of ~ 0.6 T provided by a stack of large NdFeB magnets. A Hall probe was placed on the sample surface, following removal of the field, and scanned manually over the Sm-123 grain. A maximum trapped field of 0.085 T was observed for the sample processed in air without added BaO_2. This increases significantly to 0.26 T, 0.43 T, 0.5 T and 0.33 T for the samples containing 1, 2, 4 and 10 wt% BaO_2, respectively, indicating that the addition of an optimum BaO_2 content is important if improved trapped magnetic fields are to be achieved in large grain bulk SmBCO processed under air.

4. Conclusion

Large, SmBCO grains melt processed in air using a practical, processing technique have been fabricated with high, homogeneous transition temperatures, indicative of suppressed $Sm_{1+x}Ba_{2-x}Cu_3O_{7-\delta}$ solid solution phase formation. Significantly improved trapped fields are observed in these bulk superconductors. The processing technique enables the fabrication in air of large grain bulk superconductors, and particularly LRE(BCO) materials, that exhibit good superconducting properties and offers both a significant degree of freedom in terms of processing parameters and reproducibility in the growth of oriented, single or multi-grain samples.

Acknowledgements

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