Physica C 471 (2011) 876–879

Contents lists available at ScienceDirect

Physica C

journal homepage: www.elsevier.com/locate/physc

Effects of Y_2O_3 additions on the oxygen diffusion in top-seeded melt growth processed $YBa_2Cu_3O_{7-y}$ superconductors

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ARTICLE INFO

Article history:
Available online 12 May 2011

Keywords:
Single grain YBCO superconductor
Y_2O_3 addition
Oxygen diffusion
Superconducting property

ABSTRACT

To understand the effect of Y_2BaCuO_5 (Y211)/$YBa_2Cu_3O_{7-y}$ (Y123) interfaces on the oxygen diffusion in single grain $YBa_2Cu_3O_{7-y}$ superconductors, single grain Y123 superconductors with 0.05 and 0.3 moles of Y_2O_3 additions were fabricated by a top-seeded melt growth (TSMG) process. Y123 compacts with Y_2O_3 additions were subjected to melt growth heating cycles with a cooling rate of 1 °C/h through a peritectic temperature (1015 °C) and then annealed at 450 °C for 200 h in flowing oxygen. The superconducting temperature (T_c) and critical current density (J_c) were estimated for the three different regions (top surface (*s*), intermediate (*i*) and center (*c*)) of samples. The amount of Y211/Y123 interface area in single grain Y123 superconductors was successfully controlled by Y_2O_3 additions. The T_c values of *s* regions were higher than those of *i* and *c* regions, which indicates the presence of more oxygen at the sample surfaces. In addition, the T_c values of *i* and *c* regions of the Y123 sample with 0.3 mole Y_2O_3 addition were higher than those of the same regions of the Y123 sample with 0.05 mole Y_2O_3 addition due to the promoted oxygen diffusion through Y211/Y123 interfaces and other related defects. In spite of the promoted oxygen diffusion by Y_2O_3 addition, the large T_c difference among the regions still existed, which suggests sluggish oxygen diffusion into single Y123 grains.

1. Introduction

The top-seeded melt growth (TSMG) process, which is a modified melt growth process combined with top seeding, has been widely adopted to fabricate large single grain $YBa_2Cu_3O_{7-y}$ (Y123) superconductors [1–4]. The levitation forces of the single grain Y123 superconductors fabricated by a TSMG process were large due to the high critical current density (J_c) and large grain size. The TSMG processed Y123 superconductors can be used as a bearing part of the levitation applications such as a flywheel energy storage system [5]. The J_c at magnetic fields is dependent on the defect density inside Y123 grains. The possible flux pinning sites are oxygen deficiency [6], dislocations [7], stacking faults [8], chemical doping [9] and second particle phases [10,11]. The size of defects should be reduced to a nanometer scale in order to insure strong pinning capability at external magnetic fields [6–9].

One of important factors which decide a superconducting transition temperature (T_c) and J_c of a Y123 superconductor was oxygen deficiency ($7-y$ in $YBa_2Cu_3O_{7-y}$) [12]. The flux pinning induced by the oxygen deficiency appeared as a peak effect at intermediate magnetic fields [6]. Due to the peak effect, the J_c of samples with oxygen deficiency was higher than that of the fully oxygenated Y123 sample. The oxygen annealing condition was dependent on the sample size and the interior microstructure regarding oxygen diffusion [13–15]. Oxygen diffusion into the poly-grain samples is fast due to the presence of high angle grain boundaries (grain size of a few μm [16]), whereas oxygen diffusion into the large sized Y123 grains is much more difficult due to a lack of oxygen diffusion paths [15]. Due to the above reasons, the oxygenation time for sintered Y123 samples was several tens of hours [17], whereas the time for the large single grain Y123 samples of a few cm was over several hundred hours [14]. Various defects such as a dislocation, twin, platelet, crack and Y_2BaCuO_5 (Y211)/Y123 interface were present in the melt processed Y123 superconductors [15,18], but it was not clear which defects acted effectively as diffusion paths. The transmission electron microscopy study showed the twin formation around the Y211 particles trapped within Y123 grains, which indicated that the Y211/Y123 interfaces and platelets acted as oxygen diffusion paths [18]. The microstructure analysis suggested the possible way of facilitating the oxygen diffusion in large sized single grain Y123 superconductors.

The purpose of this study is to macroscopically understand the effect of Y211/Y123 interfaces on oxygen diffusion in large Y123 grains and to establish the optimum oxygenation process for large

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doi:10.1016/j.physc.2011.05.078

sized single grain Y123 superconductors. Y_2O_3 additions to a Y123 powder was attempted in order to control the Y211/Y123 interfacial areas. The influence of Y_2O_3 additions on microstructures, T_c and J_c of TSMG processed Y123 bulk superconductors are reported in this article.

2. Experimental procedure

Single grain Y123 superconductors were fabricated by a TSMG process using Y123 powders (Solvay, Germany, 99.9%) with 0.05 mole or 0.3 mole Y_2O_3 addition (BM-chem hi-tech Co., China, 99.9%). Based on yttrium composition, the nominal compositions of Y123-Y_2O_3 powder mixtures are $Y_{1.1}Ba_2Cu_3O_{7-y}$ (herein called Y1.1) and $Y_{1.6}Ba_2Cu_3O_{7-y}$ (Y1.6). The purpose of Y_2O_3 addition is to produce Y211/Y123 interfaces in Y123 grains which may act as oxygen diffusion paths [18] and flux pinning medium [10]. 1 wt.% CeO_2 powder was added to the Y123-Y_2O_3 powder mixtures so as to refine Y211 particles. The powder mixtures were deposited in a plastic jar with ZrO_2 balls and solvent (ethanol), milled for 24 h and then dried in a vacuum oven. The dried powders were pressed in a 15 mm dia. steel mold into pellets. To increase the compact density, the pellets were pressed isostatically in a water chamber. For the growth of a single Y123 grain, the Sm123 seed (melt-texture, single grain) was placed on the top surface of pellets. Yb_2O_3 coated MgO substrate (single crystal) was used as a bottom plate for the seeded pellets. The seeded pellets were positioned at the center of a box furnace and subjected to melt growth heating cycles with the cooling rate of 1 °C/h from 1020 °C to 980 °C. The detailed heating schedule applied in this study was well described in the literature [4]. At the partially melted state where Y123 powder decomposes into Y211 and melt, the added Y_2O_3 powders act as Y211 nucleation promoter and then produce additional Y211 particles according to Eq. (1). Consequently, a Y123 sample with Y_2O_3 addition has more Y211 particles in a Y123 grain in comparison with a Y123 sample without Y_2O_3 addition.

$$\begin{aligned} YBa_2Cu_3O_{7-y} + xY_2O_3 \rightarrow\ & Y_2BaCuO_5(\text{decomposed}) \\ & + xY_2BaCuO_5(\text{additional}) \\ & + \text{melt}(Ba_3Cu_5O_z) \end{aligned} \quad (1)$$

After the TSMG process, the prepared samples were heated to 450 °C at a rate of 100 °C/h in flowing oxygen, held at this temperature for 200 h for oxygen embedding and cooled to room temperature at a rate of 100 °C/h. The oxygen diffusion aspect from sample surfaces to centers was inferred from T_c values estimated for parts of samples.

The microstructures of samples were examined using scanning electron microscopy (SEM). The measurement of superconducting properties (T_c and J_c) was taken for the three different regions of surface (*s*), intermediate (*i*) and center (*c*) regions of TSMG processed Y1.1 and Y1.6 samples (see Fig. 1). The T_c and J_c for the selected regions were measured using a dc superconducting quantum interference device (SQUID) magnetometer in a time-varying magnetic field with an amplitude of 7 T. T_c was measured in zero-field-cooled mode and defined as the onset of a diamagnetism. The magnetic J_c for rectangular samples of approximately $1.3 \times 1.9 \times 2.4$ $(a \times b \times c)$ mm^3, estimated for $H//c$-axis at 77 K, was derived using the extended Bean's critical model [19].

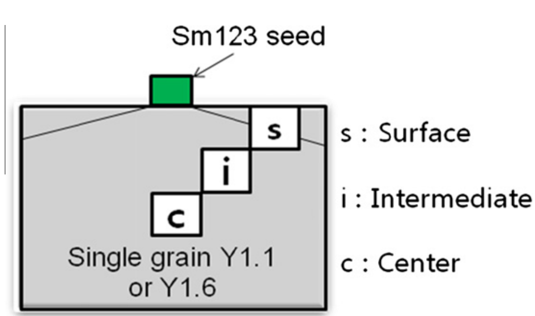

Fig. 1. Three different regions selected for property measurement: *s*, *i* and *c* denote surface, intermediate and center regions, respectively.

3. Results and discussion

Fig. 2a and b shows the top surface views of TSMG-processed Y1.1 and Y1.6 samples, respectively. Both top surface views show the successful growth of single rectangular Y123 grains which covered almost entire top surfaces. The side length of the rectangular grain of sample (a) was 10 mm, which is slightly larger than 9 mm of sample (b). The growth fronts of sample (a) were rough and irregular, whereas the growth fronts of sample (b) were relatively flat. The flat growth fronts suggest that the mass supply for the Y123 growth was more uniform due to the higher yttrium concentration in the melt.

Fig. 3a and b shows the SEM micrographs of grain interiors of TSMG processed Y1.1 and Y1.6 samples, respectively. Y211 particles were found to be trapped within Y123 grains with a large density difference between the two samples. The Y211 density (the number (*N*) of Y211 particles per unit area) of sample (b), estimated from the microstructure, was 5.23×10^7 N/cm^2, which is higher than 9.02×10^6 N/cm^2 of sample (a). The higher Y211 density of sample (b) is attributed to the formation of more Y211 particles by the larger amount Y_2O_3 addition. Additionally, the Y211 size of sample (a) was much larger than that of sample (b).

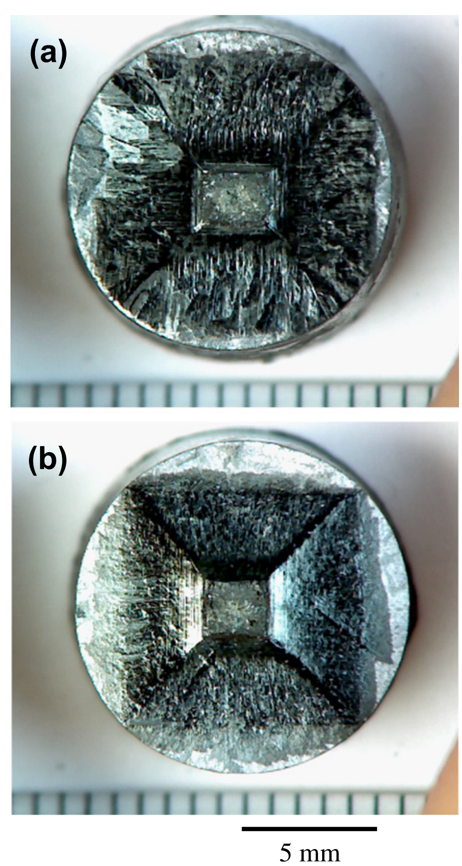

Fig. 2. Top surface views of TSMG processed (a) Y1.1 and (b) Y1.6 samples.