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Fabrication of a high magnetization YBCO bulk superconductor by a bottom-seeded melt growth method

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Abstract

A large grain YBCO bulk superconductor is fabricated by the top-seeded melt growth (TSMG) method. In the TSMG process, the seed crystal is placed on the top surface of a partially melted compact and therefore the seed crystal is frequently tilted during the melt process due to intrinsic unstable nature of Y211 particle +liquid phase mixture. In this work, we report the successful growth of single-domain YBCO bulk superconductors by a bottom-seeded melt growth (BSMG) method. Investigations on the trapped magnetic field and the microstructures of the synthesized specimens show that a bottom-seeded melt growth method has hardly affected on the crystal growth behavior, the microstructure development and the magnetic properties of the large grain YBCO bulk superconductors. The bottom-seeded melt growth method is clearly beneficial for the stable control of seed orientation through the melt process for the fabrication of a large grain YBCO bulk superconductor.

Keywords: REBCO, bulk superconductors, bottom seeding, trapped field

1. INTRODUCTION

High magnetization single-grain $REBa_2Cu_3O_{7-y}$ (RE123, RE: rare-earth elements) bulk superconductors have been fabricated by a top-seeded melt growth (TSMG) process [1-3]. In TSMG processes, seeds are placed at a center of the top surfaces of the compact [1-8]. Top-seeding technique makes it possible to obtain a single grain bulk superconductor that the c-axis of the grain is perpendicular to the specimen surface. In order to grow a well-controlled single grain specimen, several parameters have been controlled systematically; single crystal seed, slow cooling, the suppression of the subsidiary grains. Subsidiary grains have random orientation and therefore leads to the degradation of the magnetic properties of the bulk superconductor along with the reduction of the grain size. Kim et. al. [9] has successfully suppressed the nucleation of the subsidiary grains at the compact surfaces by applying a surface coating of Yb_2O_3 powder and a bottom seeding method on the YBCO compact surface in order to suppress the nucleation of the subsidiary grains.

However, some issues have been remained unsolved yet. One of these issues is a seed-tilting. Melt growth process involves the high temperature heat treatment above the melting temperature of REBCO superconductor. During the annealing at high temperature, REBCO decomposes into a mixture of RE_2BaCuO_5 (RE211) solid particles and $BaCuO_3$ + CuO liquid phase by a peritectic decomposition of

$$\text{RE123} \rightarrow \text{RE211} + \text{liquid phase}$$

The mixture of RE211 solid particles and $BaCuO_3$ + CuO liquid phase (so called “quasi-melt”) has both properties of solid and liquid and deforms by a viscous flow due to a presence of liquid phase. The viscous deformation leads to the shape changes of REBCO compact. Thereby, the seed crystal is deviated from its starting position and is tilted. Seed tilting is rather serious for the specimens that use a buffer pellet between REBCO compact and a seed crystal in order to reduce the contamination of Sm in YBaCuO specimen from Sm-123 seed [10]. The seed tilting results in the deviation of the orientation of a melt-processed crystal from a target orientation. Recently, Lee [11] has reported that the seed orientation directly affects on the growth behavior and the magnetic properties of melt processed single grain REBCO bulk superconductors. The seed tilting is inevitable as far as a seed is placed on unstable quasi-melt specimen in TSMG method. One of the way to avoiding from seed tilting is to place a seed on a stable substrate and put the compact onto the seed; i.e., a bottom-seeded melt growth that crystal starts to grow from a seed at bottom surface and grows toward the top surface. Then, the shape changes from a viscous deformation of the compact may not affect on the seed orientation during the bottom-seeded melt growth because the seed crystal is placed on the stable substrate and pressed downward by the weight of the specimen.

Bottom-seeded melt growth has been applied previously [9, 12]. However, it was hard to investigate the effect of the bottom seeding independently because they used a double

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seeded technique; top seed was placed on a top of the REBCO compact and a bottom-seed was embedded in Yb_2O_3 powder [9] or Y211 powder [12]. The dual seeds technique also leads to the formation of the liquid trapped interface inside of the specimen because two grains, which started to grow from both of top seed and bottom seed, meet at the middle of the specimen. The liquid trapped interface may provide a mechanically-weak interface which may result in the failure of the specimen during a field service.

In this article, single grain REBCO bulk superconductors have been prepared by a bottom-seeded melt growth (BSMG) method. The specimen fabricated by a BSMG method has been characterized and its microstructure and magnetic property have been compared with those of the specimen prepared by a TSMG method.

2. EXPERIMENTALS

$Y_{1.8}Ba_{2.4}Cu_{3.4}O_{7-y}$ (hereafter Y1.8) powder was used as a raw material in this study. Y1.8 powder, which was purchased from Superconductor Components Inc., was mixed with 1 wt. % CeO_2 powder in order to refine Y211 particles. An appropriate amount of Y1.8 powder was put into a steel mold with a diameter of 25 mm (or 30 mm). Compacts were prepared by a uni-axial pressing and then cold isostatic pressed.

Fig. 1 shows the schematic drawings how to arrange the seed and compact for the TSMG and the BSMG, respectively. Thin Yb_2O_3 pieces (~1.57 mm-thick) were placed on an alumina plate. For TSMG, Y1.8 compact was placed on the Yb_2O_3 pieces and then a small-sized Sm-123 seed was put on the top surface of the Y1.8 pellet at last. For BSMG, Sm-123 seed was put on the alumina plate and then Y1.8 compact was placed on the Yb_2O_3 pieces. The height of Sm-123 seed was little bit higher than that of the Yb_2O_3 pieces in order to assure as the Sm-123 seed to contact to the bottom of the Y1.8 compact.

The heat treatment procedure for melt growth (MG) was similar to those reported in the literature [5]. The cooling rate controlled with $0.25 ℃ h^{-1}$ at the temperature regime for the growth of Y123 grains. After the MG heat treatment, Y1.8 samples were heated to 500℃ at a rate of $200 ℃ h^{-1}$ in flowing oxygen for oxygenation, held at this temperature for 50 h, cooled to 400–500℃ at a rate of $100 ℃ h^{-1}$, held at this temperature for 200-300 h, and then cooled to room temperature at a rate of $200 ℃ h^{-1}$.

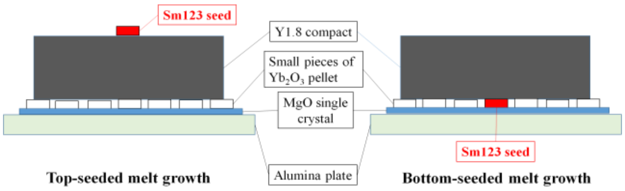

Fig. 1. Schematic drawings illustrating the arrangement of Sm123-seed and Y1.8 compact for TSMG method and BSMG method, respectively.

Trapped magnetic fields at 77 K were measured for the slab surface and the cross section of the field-cooled samples. A trapped magnetic field (B) measurement was performed on field-cooled samples. Permanent magnet with a diameter of 50 mm and a surface field of 4.9 kG was placed on the sample, and liquid nitrogen was poured to cool the sample to 77K (field cooling, FC). When the temperature of the sample reached 77 K, the permanent magnet placed on the superconductor was removed, and the magnetic force trapped in the superconductor was measured on the top surface using a hall probe. The microstructures of the melt-processed specimen were investigated using optical microscope (OP).

3. RESULTS AND DISCUSSION

Fig. 2 shows the digital pictures of the melt-processed YBCO bulk specimen prepared by a BSMG method. It is seen that the specimen is consisted of a single grain. Rectangular shape pattern, which is appeared in the top-view, matched with the a-c growth sector. In case of TSMG method, the remaining liquid is absorbed in Yb_2O_3 substrate or has been flown down at the bottom of the specimen. Therefore, the rectangular pattern usually is not observed at the bottom of the specimen which has been prepared by a TSMG specimen. Fig. 3 shows the schematic drawing illustrating the interior-seeding method developed by Kim et al. [13]. It is seen that the upper half of the interior-seeded specimen should be the same with the bottom-seeded specimen as shown in the schematic drawing of Fig. 1. It is also understood that a low half section is the same feature of top-seeded growth as shown in Fig. 1. Thus, it is well understood that a rectangular pattern also has been reported for the specimen prepared by an interior seeding technique [13, 14]. Radusovska *et. al.* [14] has reported that the formation of the rectangular pattern is explained by so-called edge melt distribution (EMD) model. EMD model suggests the transport of liquid phase (melt) from the a-b growth sector to the a-c growth sector because the growth rate is higher in c-axis that in a-axis and b-axis. However, it is thought that the long-range transport of melt may not be necessary for the formation of the rectangular pattern on the top surface. Recently, Lee [11] has shown that the shape change of a melt-processed bulk specimen is simply related with the interfacial energy between the melt and the growing Y123 crystal. Chen et al. [15] has also reported based on the real time observation of the melting behavior of Y123 film that the liquid phase is

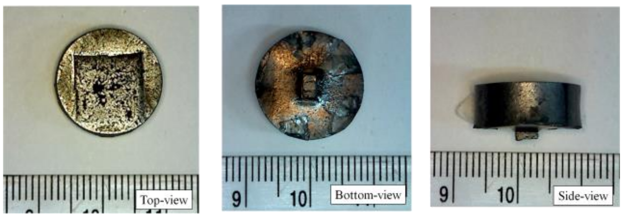

Fig. 2. Digital pictures of the melt-processed YBCO bulk specimen prepared by a BSMG method.

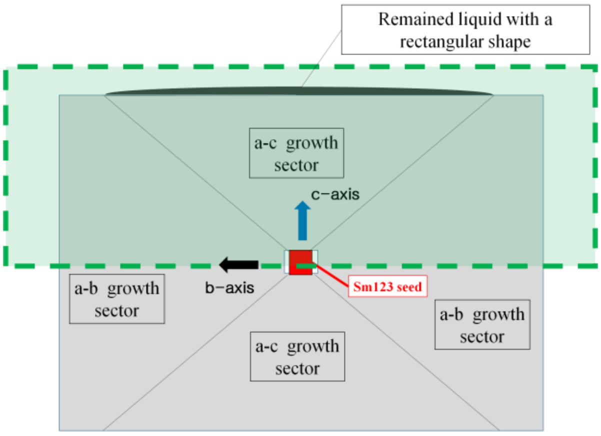

Fig. 3. Schematic drawing illustrating the interior- seeding method.

hard to wet on the (001)-oriented surface (the plane normal to the c-axis of Y123 crystal). Top surface is consisted of both of a-b growth sector and a-c growth sector and only a-c growth sector is covered with the remained liquid. The crystallographic plane of both a-b growth sector and a-c growth sector is (001). Therefore, other factors should be considered in order to explain why only a-c sector is covered with the remained liquid phase. Contrary to (001) plane, (100) or (010) plane of RE123 has a good wettability with the liquid phase. Fig. 4 depicts the schematics showing the growth procedure just before and after the growing Y123 crystal has reached to the top surface of the specimen. Until Y123 crystal has reached to the top surface of the specimen, all the sides of Y123 crystal is covered by the melt consisted of the Y211 + liquid phase as in Fig. 4(a). Fig. 4(b) shows just after the Y123 crystal has reached the top surface. This is the situation that a-b sector appears on the top surface for the first time. At this step, the sides of Y123 crystal is (100) and/or (010) surface that has good wettability with the liquid phase while top surface of a-b growth sector has a poor wettability with the liquid phase. Therefore, the remained melt at the later stage of the crystal growth (Fig. 4(b)) is isolated at the a-c growth sector. The remained melt at the top surface has a lenticular cross section due to the force balance relation between the interfacial energies as follows;

$$\gamma_{(001)/V} = \gamma_{(001)/L} + \gamma_{V/L} \cos\theta$$

where $\gamma_{(001)/V}$ is an interfacial energy between (001) plane of Y123 crystal and air, $\gamma_{(001)/L}$ is an interfacial energy between (001) plane of Y123 crystal and liquid phase, $\gamma_{V/L}$ is an interfacial energy between liquid phase and air and θ is a contact angle of the liquid phase with (001) surface of Y123 crystal.

Fig. 5 shows the digital pictures of the melt-processed YBCO bulk specimens prepared by a TSMG method (Fig. 5(a)) and a BSMG method (Fig. 5(b) and (c)). It is seen that the x-shaped facet lines are evident for a TSMG specimen. However, the x-shaped facet lines are not clear for as-processed BSMG specimen (Fig. 5(b)) partly because the Yb_2O_3 substrate pieces are attached on the bottom surface of the specimen. Fig. 5(c) is a picture of the bottom-seeded grown specimen that ~ 1mm of the bottom surface has been polished. It is seed that single grain has been well grown with an x-shaped growth pattern which is one of the characteristics of a single grain growth of melt-processed REBCO bulk superconductor.

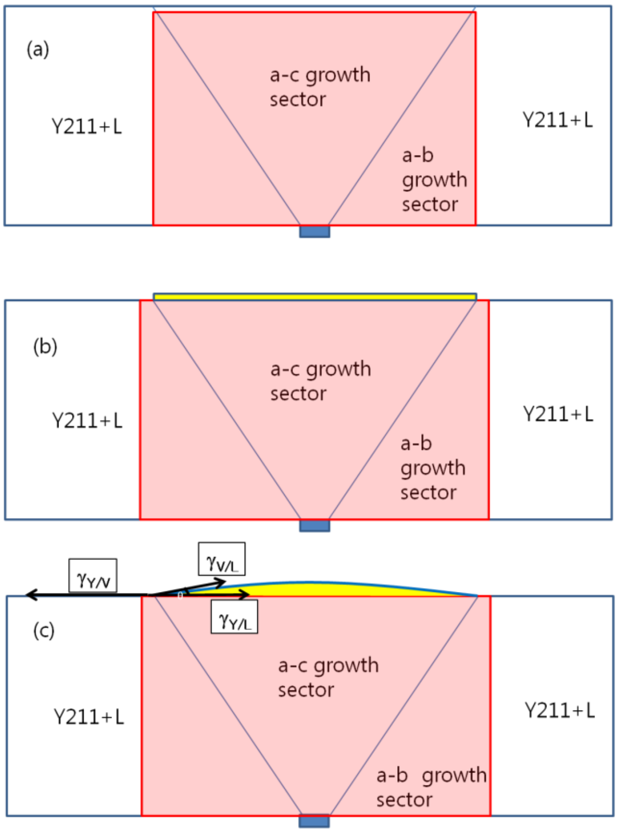

Fig. 4. Schematic drawings showing the crystal growth procedure and the formation of the cap of the remained liquid at the top surface for the specimen prepared by a BSMG method.

Fig. 6 shows the trapped magnetic field of the specimens which have been prepared by a TSMG method and a BSMG method. It is seen that both specimen shows single peak contour lines which is one of the characteristics of a single grain REBCO bulk superconductor. Bottom-seeded specimen shows better magnetization behavior but the difference is not large.

Fig. 7 shows the optical microstructures of the bottom-seeded melt processed specimen at various positions. Low magnification picture shows that a lot of

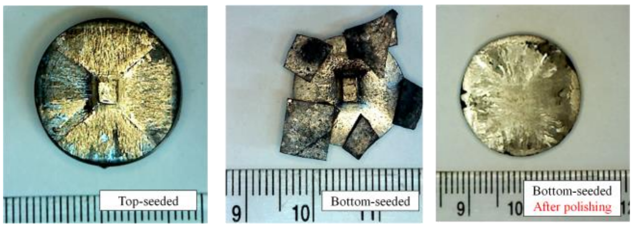

Fig. 5. Digital pictures of the melt-processed YBCO bulk specimen prepared (a) by a TSMG method, (b) and (c) by a BSMG method. (c) is the picture after polishing of (b).

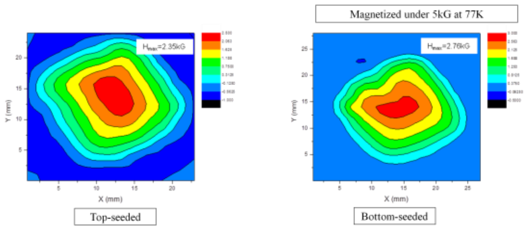

Fig. 6. Trapped magnetic field of the specimens which have been prepared by a TSMG method and a BSMG method, respectively.

pore has been formed at the middle of the specimen. High magnification pictures show that small sized Y211 particles are present through the specimen. It is also seen that the size of Y211 particles is relatively large at the early stage of crystal growth but becomes smaller with the crystal growth. Y123 crystal has started to grow from the bottom seed and ends at the top of the specimen. It is seen that a lot of pores are present in the middle of the specimen. Pores are formed because the gas evolved during the peritectic decomposition has had no chance to diffuse out. It has been reported that the artificial holes can reduce the porosity drastically in a melt-processed REBCO bulk specimen [16]. Nearly the same microstructures have been observed for a top-seeded specimen except that the size of Y211 particle becomes finer from the upper part of the specimen to the bottom of the specimen because the direction of crystal growth is reversed as from the top surface to the specimen bottom. The increase of Y211 density (i.e., the decrease of the Y211 size) with the crystal growth has been reported [17]. Endo et al. has reported that the Y211 density is increased as the undercooling is increased [18].

Summarizingly, high magnetization single grain YBCO bulk superconductor has been fabricated by a BSMG method. Therefore, it can be said that a bottom-seeding method is an alternative approach to obtain a high magnetization REBCO bulk superconductors. Additionally, we would like to point out again that a bottom-seeding technique has an advantage at the stand point of seeding stability because the orientation of seed is not changed even though there is a viscous flow of bulk specimen due to a natural behavior of quasi-melt specimen.

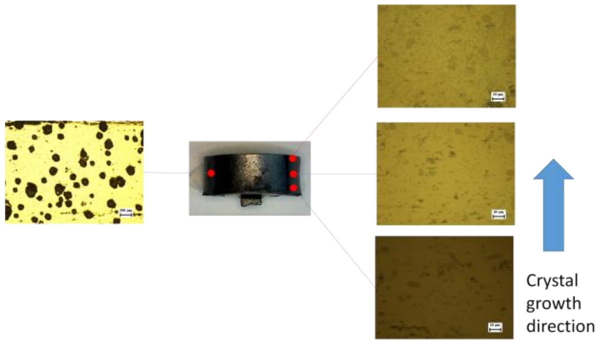

Fig. 7. Optical microstructures of the bottom-seeded melt processed specimen at various positions. Scale bars of low magnification and high magnification pictures represent 100 μm and 10 μm, respectively.

4. CONCLUSIONS

A large-grain YBCO bulk superconductor has been fabricated by a bottom-seeded melt growth method. Material characteristics of the bottom-seeded melt growth specimen are nearly the same with the top-seeded melt growth specimen. This work presents that the bottom-seeded melt growth method is beneficial for the better control of the crystal orientation of the melt-processed REBCO bulk superconductor.

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