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# Effect of size, morphology and crystallinity of seed crystal on the nucleation and growth of single grain Y-Ba-Cu-O

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### **Abstract**

The effect of size, morphology and crystallinity of seed crystals on the nucleation and growth of large grain Y-Ba-Cu-O (YBCO) bulk superconductors fabricated by top seeded melt growth (TSMG) has been investigated. Seeding bulk samples with small, square shaped seed crystals leads to point nucleation and growth of the superconducting YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-y</sub> (Y-123) phase that exhibits the usual square habitual growth symmetry. The use of triangular and circular shaped seed crystals, however, modifies significantly the growth habit geometry of the grain. The use of large area seeds both increases the rate of epitaxial nucleation of the Y-123 phase and produces relatively large crystals in the incongruent melt, which decreases significantly the processing times of large grain samples. The present study is relevant to decreased processing times of samples with both preferred or no growth sectors and for multiple seeding of large grain samples which contain clean grain boundaries.

**Key words:** Grain growth, Microstructure, Oxide superconductors, Superconductivity, Magnetic properties.

### 1. Introduction

Microstructural texture is an important parameter in the fabrication of bulk high temperature superconductors (HTS) for practical applications. Conventional ceramic processing techniques, however, are unable to produce the grain alignment and microstructural features required for high critical current densities (J<sub>c</sub>) and associated magnetic levitation forces in these materials. Fortunately, the peritectic reaction that takes place in the Y-Ba-Cu-O (YBCO) system at around 1000°C enables various solidification techniques to be employed to produce textured materials with the desired properties. In addition to the conventional directional solidification technique, various isothermal, or melt, processing techniques have been developed for fabrication of textured YBCO bulk superconductors [1].

Melt processing techniques generally involve the slow solidification of non-superconducting  $Y_2BaCuO_5$  (Y-211) and a Ba-Cu-O liquid phase mixture through the  $YBa_2Cu_3O_{7-y}$  (Y-123) peritectic temperature ( $T_p$ ) to temperatures of around  $T_p-40^{\circ}C$  to produce the superconducting Y-123phase [2]. Of these techniques, the *top seeded melt growth processes* (TSMG) is the most successful and is now used routinely to process YBCO in the form of bulk single grains. The TSMG process involves seeding a YBCO preform with a chemically and structurally compatible, higher melting point seed crystal,

such as SmBCO, NdBCO or MgO. The seed crystal initiates the nucleation and further growth of the Y-123 phase in the incongruently molten green body, which solidifies wholly into a single grain during controlled cooling. Due to the intrinsic difficulties of solidification process, however, the fabrication of high quality bulks of diameter exceeding a few cm has proved difficult. The relatively slow rate of the peritectic reaction at the site of the seed crystal is one limiting factor in extending the TSMG process to larger sized grains. The use of multiple seeds in an attempt to decrease the processing time and increase the grain size, on the other hand, has also not yet produced samples with sufficiently good properties for engineering applications, despite numerous attempts. This is due mainly to the formation of unclean grain boundaries and to the lack of precise control in nucleating grains with similar orientations within the bulk preform. To further improve the microstructure and processing speeds, therefore, it is essential to study the role played by the seed crystal in initiating and further growth of the YBCO grain. In this paper we investigate the effect of size, morphology and crystallinity of the seed on the nucleation and growth of large grain, bulk YBCO.

## 2. Experimental

Precursor powder of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-y</sub> enriched with 28 mol % Y<sub>2</sub>BaCuO<sub>5</sub> and containing 0.1 wt. % of Pt was pressed uniaxially into cylindrical pellets of various sizes from 15 mm to 50 mm in diameter and around 10 to 25 mm in height. Top seeded melt growth was performed in a box or horizontal tube furnace in ambient atmosphere. Seed crystals of various sizes from 1mm x 1mm and up to 20 mm x 15 mm of different shapes were cut from a single grain melt textured Sm123 bulks produced in a separate process. A typical thermal profile using for TSMG process involved heating the pellet and seed rapidly to 1045 °C, holding for 45 minutes, cooling rapidly to 1010 °C (the peritectic temperature of Y-123) and finally more slowly at 0.3 °C/h up to 975 °C. The resulting YBCO grains were photographed for further analysis and their size measured precisely. The samples were polished using diamond paste and their microstructure analysed under polarized light using a Leitz optical microscope.

### 3. Results and Discussion

The TSMG technique effectively reduces the thermodynamic barrier to the nucleation of the Y-123 phase in the Y-211 and Ba-Cu-O liquid melt, which, as a result, produces a delay in the grain growth process. This, inevitably, results in a finite incubation time during which no crystal nucleation occurs. The presence of a heterogeneous nucleation site, such as an externally placed seed crystal, lowers this barrier. If the solidification is limited to within the nucleation barrier window, the heterogeneously nucleated Y-123 grain at the seed position will grow during further solidification without competition from random nucleation elsewhere in the molten sample [2]. A seed crystal of square or rectangular planar geometry of size around 1 mm is generally used in the TSMG process with the c-axis parallel to the seed thickness. In this case the growth of the YBCO grain nucleating from the seed crystal follows characteristically a square growth habit with orthogonal facet lines, due to the formation of various growth sectors within the single grain [3], as shown in figure 1(a). This growth

habit, however, is influenced significantly by the morphology of the seed crystal. Figure 1 shows the samples grown with different shaped seed crystals. It can be seen in figure 1(b), for example, that the initial growth pattern produced by a triangular shaped seed crystal is not square. The departure from a square initial growth pattern is even more evident for a circular shaped seed crystal, where the initial growth geometry is convex, as shown in figure 1(c). No distortion of the square habitual growth plane is observed, however, for an irregular seed shaped seed, as shown in figure 1(d). In this case the local small irregularities of the seed edges are not effective in influencing the initial growth morphology.

Figure 2(a) shows a cross-section of a sample grown using a large area seed. The Y-123 phase is observed to nucleate epitaxially over the full surface of the seed and the entire preform is transformed into a single grain. Significantly, the orientation of the seed is replicated in the sub-grains within the bulk of the YBCO grain, as shown in figure 2(b). The use of a large area seed crystal suppresses the formation of growth sectors, which could be useful in controlling the homogeneous dispersion of pro-peritectic Y-211 and other externally added particles, where their distribution is effected generally by the presence of growth sectors [3,4].

Figure 3 shows typical examples of samples that have been grown using different sized seed crystals under otherwise identical growth conditions. Each sample was cooled to 990 °C and its growth terminated. In this case, increasing the size of the seed crystal increased the size of the YBCO grain, which can be explained by two different YBCO growth modes. The first involves the rapid expitaxial nucleation of the Y-123 phase beneath the seed crystal, followed by usual peritectic Y-123 growth from solid Y-211 and Ba-Cu-O liquid phases in the incongruent melt. Secondly, the subsequent production of a large size grain into the melt by the large surface area of the seed crystal contributes further to large, increased grain growth. Generally it has been observed that the roughly linear growth rate of the grain at a given supersaturation is size-dependent, with larger crystals growing faster than smaller ones [5]. A more detailed investigation of this pheonomenon is underway.

### 4. Conclusions

The effect of seed crystal geometry on the nucleation and growth of large YBCO grains by the TSMG process has been investigated. The use of triangular and circular shaped seed crystals has been observed to modify the morphology of the initial growth sector geometry. The use of a large area seed decreases the processing time of single grain YBCO bulks due to rapid epitaxial nucleation of Y-123 phase and the production of a relatively large crystal into the incongruent melt. The observations made in this study could lead potentially to improving the microstructure and decreasing the processing times of large single grain YBCO bulk superconductors.

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References

- 1. Reddy E.S. and G.J. Schmitz, Near *net-shaped objects of RE123* superconductors by *Infiltration Processing*, Published by Tsinghua Science and Technology, 2003, **8**, 292-306.
- 2. Cardwell D.A., *Processing and properties of large grain (RE)BCO* Mat. Sci. Eng B-SOLID, 1998, **53**, 1-10.
- 3. Diko P., Zmorayova K, Granados X., Sandiumenge F., Obradors X. Growth related Y2BaCuO5 particle concentration micro-inhomogeneity in the growth sectors of TSMG YBa2Cu3O7/Y2BaCuO5 bulk superconductor Physica 2003, **384**, 125.
- 4. Honjo S, Cima M.J., Flemings M.C., Ohkuma T, Shen H, Rigby K., Sung T.H., Seeded crystal growth of YBa2Cu3O6.5 in semisolid melts, J. Mater. Res. 1997b, 12, 880-890.
- 5. Simeone M. Palomba M. and Volpicelli G., *Size-dependent growth rate of gypsum crystals By video-enhanced optical microscopy*, In proceedings of 15<sup>th</sup> International symposium on industrial crystallization 15 18 September 2002, Sorrento, Italy.

# **Figure Captions:**

- **Figure 1:** YBCO samples showing the nucleation and growth of a grain from seed crystals of different morphologies. Initial growth patterns from c-axis oriented seed crystals of (a) square, (b) triangular, (c) circular and (d) irregular seed geometries.
- **Figure 2:** (a) Cross-section of a YBCO single grain grown using a seed crystal of size larger than that of the preform. (b) The orientation of sub-grains, marked by arrows, from the seed crystals are observed to replicate that of the seed throughout the entire grain.
- **Figure 3:** YBCO grains grown using different size seed crystals under otherwise identical thermal conditions. Increasing the size of the seed crystals results in increasing the size grains within the same processing times.

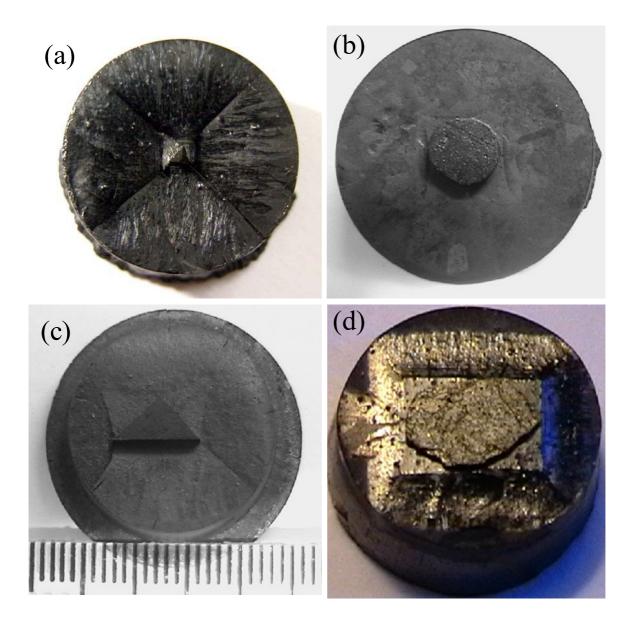


Figure 1

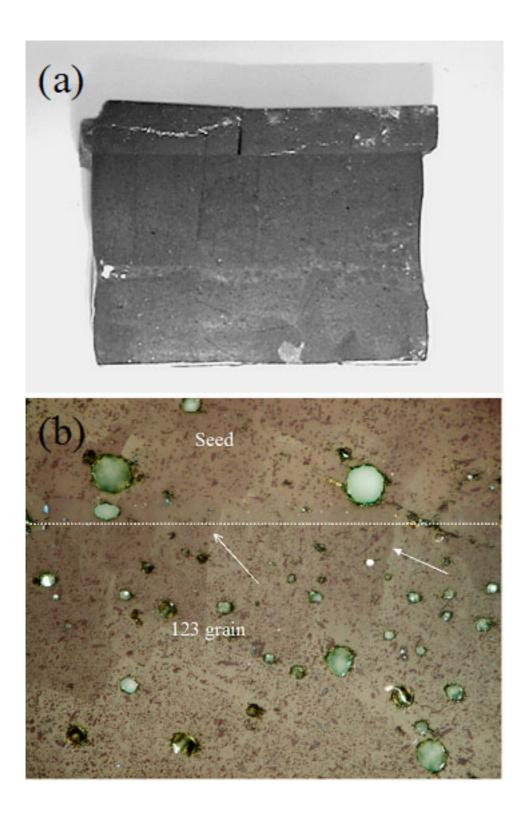


Figure 2

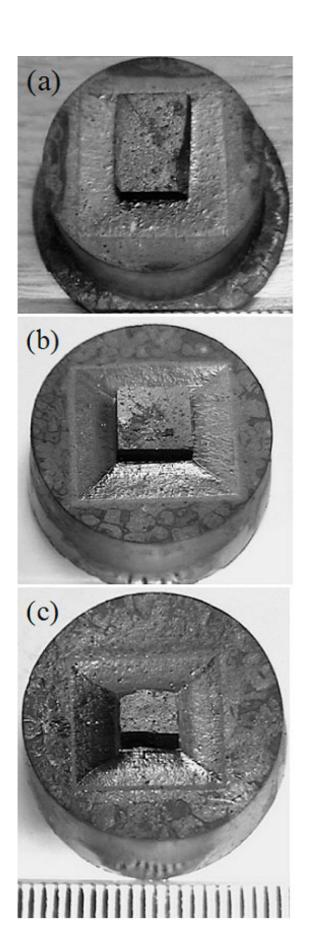


Figure 3