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Superconducting Performance, Microstructure and SEM by EDX Analysis of IG Processed $YBa_2Cu_3O_y$ Bulk Superconductors by Top and Interior Seeding Methods

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Abstract. The top-seeded and interior seeded methods, together with infiltration growth (IG) technique were used to produce $YBa_2Cu_3O_y$ (Y-123) samples with Y_2BaCuO_5 (Y-211) secondary phase particles. T_c (onset) was around 91.5. K. When interior seeding process was used, a complete growth of Y-123 single grain starting at the lower part of the bulk was observed by optical microscopy. The Y-211 particles dispersion was quite uniform in lower and upper parts of the samples, both in the *a*- and *c*-axis growth sectors, both at the beginning and end of the grain growth. In the sample produced by infiltration growth and interior seeding the critical current density at 77 K with *H*//*c*-axis was 44,000 A/cm^2 and 7,750 A/cm^2 in self-field and 2 T, respectively. It is a good basis for optimization of processing conditions, which can further improve the superconductor's performance and enable to grow large-size Y-123 bulks.

1. Introduction

Bulk $REBa_2Cu_3O_y$ (RE: Y, Nd, Sm, Gd, NEG) superconducting magnets can trap magnetic fields by order of magnitude higher than the best classical hard magnets and are therefore promising as permanent magnets for use in magnetic drug delivery system (MDDS), for construction of small mobile diagnostic devices, for water cleaning technologies etc. [1-6]. The drug delivery system could be controlled by the superconducting magnetic force in the body. As a result, a high drug concentration can be delivered in a controlled way to the target diseased tissue [2]. For all these applications good quality samples are needed, with a uniform performance and a high critical current density [7,8]. For this purpose, single domain bulk samples are produced among other techniques by infiltration-growth (IG) [9]. It allows for fabrication of near-net-shape products, uniform microstructure and performance [10,11]. Furthermore, the density of pores is quite low in IG samples, which results in a uniform distribution of the RE-211 particles and a reduced amount of cracking. However, due to limitations in the processing, one cannot produce thick samples. To overcome this drawback, we tried to produce the bulk $YBa_2Cu_3O_y$ material using the interior seeded method, together with infiltration growth (IG) technique and compared the results of top-seeded and IG + internal seed processed samples. The results are presented in this paper. They indicate that high quality single-domain material can scale up from laboratory to industrial production by using the interior-seeded infiltration growth process. A further processing optimization is needed to prepare a high performance Y-123 bulk materials by means of this method.

2. Experimental details

High-purity commercial powders of Y_2O_3, $BaCO_3$ and CuO were mixed in a nominal composition of Y_2BaCuO_5 and calcined at 900º C for 24 h. In parallel, the powders of BaO_2 and CuO were mixed in a nominal composition of $Ba_3Cu_5O_8$ (Y-035). Eventually, the pre-forms of Y-211 were placed on Ba_3Cu_5O and then an Nd-123 melt-textured seed was placed at the centre of the top surface of the Y-211 pre-forms for the growth of single Y-123 grains. We call it a cold top-seeded method combined with the infiltration growth (IG) (see Fig.1, left). In the case of the internal seeded infiltration growth (IG), the seed is placed at the top centre of the RE-211 preform, too, but covered by another RE-211 preform (see Fig.1, right). Yb_2O_3 and MgO plates were used to suppress liquid loss and spontaneous Y-123 nucleation. The samples 20 mm in diameter and 8 mm in thickness were produced by the IG process. The sintering temperature profile for large-grain Y-123 production was then designed based on the isothermal results reported elsewhere. The sample was heated at a rate of 100 ºC/h to 880 ºC and held there for 15 min, and further heated in 5 h to the temperature of 1040 ºC and held there for 50 min. Then, the temperature was lowered in 60 min to 1000 ºC, and slowly cooled to 925 ºC at a cooling rate of 0.3 ºC/h. Finally, the temperature was lowered with a cooling rate of 100 ºC/h to 100 ºC, followed by furnace cooling to room temperature.

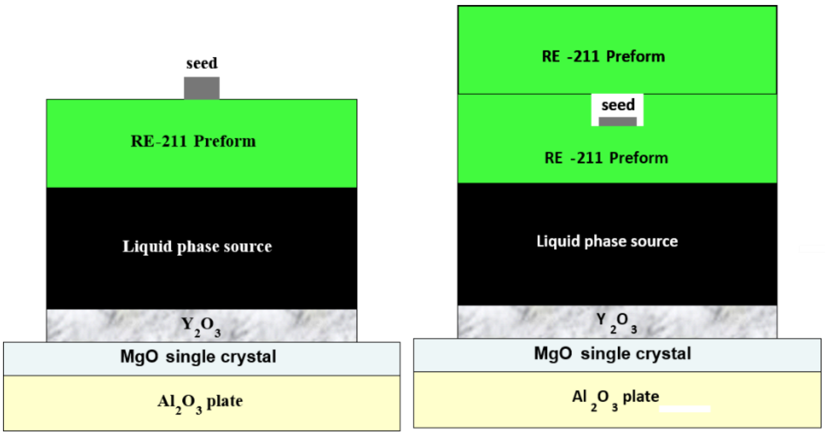

Figure 1. A typical arrangement used in the IG experiment. A source of liquid phase ($Ba_3Cu_5O_y$) is supported on a chemically inert, impervious substrate. The pre-form fabricated of 211 powder is placed on top of it. A *c*-axis Nd-123 seed crystal is placed on top (left) or interior (right) of the pre-form, at the center.

The top surface of the as-grown sample clearly indicated that sample was fully processed without multi-nucleation (see Fig. 2, right). The melt-textured samples were annealed at 450 ºC for 250 h in flowing pure O_2 gas. The microstructure of these samples was studied with an optical micrograph. Small test specimens with a size of 1.5 × 1.5 × 0.5 mm^3 were cut from the bulk samples. Measurements of the critical temperature (T_c) and magnetization hysteresis (M-H_a) loops around 77 K in fields from –1 to +5 T were done using a commercial SQUID magnetometer (Quantum Design, model MPMS5). Critical currents were estimated using the extended Bean's critical state model formula for a rectangular sample given by

$$J_c = 20\,\Delta m/[a^2 d\,(b-a/3)] \qquad (1)$$

where d is the sample thickness, a, b are transversal dimensions, $b \geq a$, and Δm is the difference of magnetic moments during increasing and decreasing field in the M-H loop [12].

3. Results and discussion

Figure 1 shows the top surfaces of the top-seeded (left) and interior seeded (middle) Y-123 materials produced from Y-211 pre-form sintered at 925 ºC, with the final melt growth performed during a slow cooling process, as described in the experimental section. All samples were 19 mm in diameter. For the seed in the interior of the sample in Fig.2 (middle), 2.5 mm bed hole was made, seen clearly after the melt growth process (Fig. 2., right).

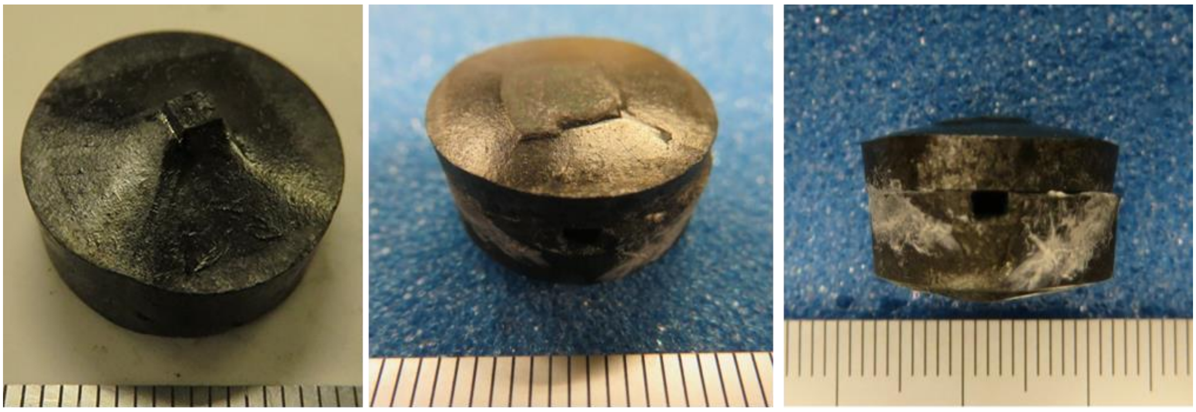

Figure 2. Top seeded IG processed Y-123 superconductor prepared in air by IG process using Y-211 pre-form (left). Interior seeded Y-123 superconductor produced by IG technique, top view (middle), and side cross-section view (right).

Note that the four-fold growth facet lines were visible on the top surface of all Y-123 samples prepared by both seeding methods, which clearly indicated that the samples were grown into single-grains, typical for the samples processed by top-seeded melt-growth process [13-15]. Due to the latter fact, bulk single-grain Y-123 or LRE-123 materials can be routinely produced by batch production [16]. The IG-process combined with the interior seed method is new and we would like to see the performance of this new material processing in comparison with the top-seeded method.

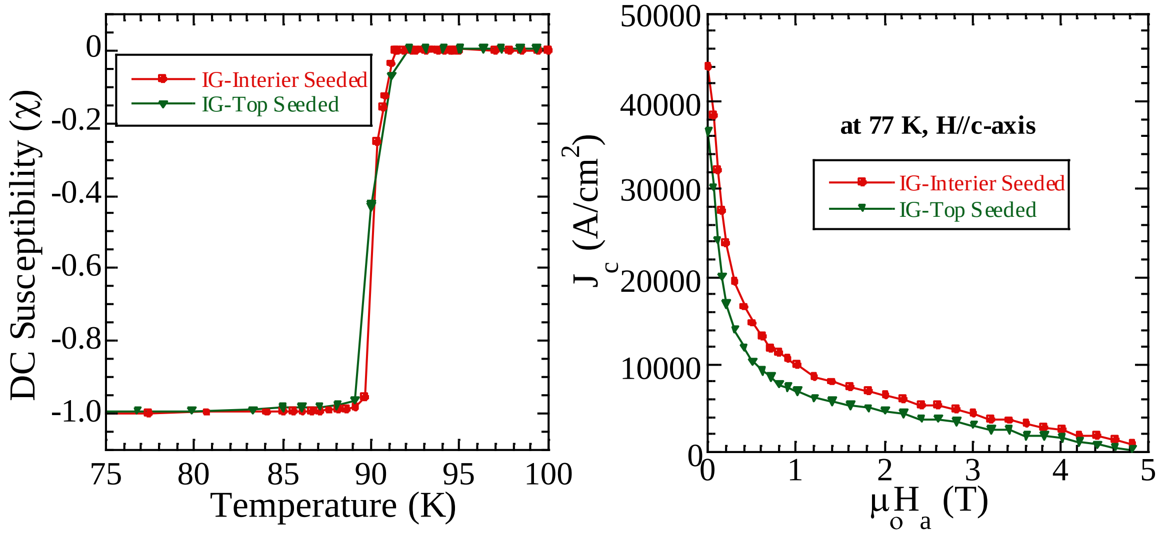

Figure 3. Superconducting transitions temperatures (left) and field dependence of the super-current density (right) at 77 K for specimens cut 2 mm below the seed of the single-domain Y-123 bulk superconductors produced by top seeding and interior seeding of Y-211 pre-form, by the infiltration growth and slow cooling in air method.

Figure 3 (left) presents the example of the temperature dependence of the dc susceptibility after zero-field-cooling (ZFC) in magnetic field of 1mT for both types of samples. A sharp superconducting transition with onset around 92.1 K and transition width of less than 1.9 K was observed, which indicated that the top-seeded IG processed sample was of an excellent performance. The other sample, produced by interior seeding, showed superconducting transition onset slightly lower, at around 91.5 K, but the transition was narrower, ≈ 1.5 K, as compared with the samples produced by top-seeded IG process. *M-H* curves were measured on the same samples at 77 K with the field applied parallel to the *c*-axis and critical current density, J_c, was calculated from the M-H curves using the extended Bean model formula, as shown in Fig. 3 (right). The interior seeded sample showed J_c at 77 K, in self-field and 2 T of about 44,000 A/cm^2 and 7,750 A/cm^2, respectively (Fig.3, right), slightly higher than that in the Y-123 sample produced by top seeding and lower than the Y-123 material produced Y-211 pre-form sintered from 900 ^{o}C to 1100 ^{o}C [17]. There, critical current density of 37,000 A/cm^2 and 6,000 A/cm^2 at self-field and 2 T, respectively, were detected at 77 K and *H*//*c*-axis. Anyway, a refinement of secondary phase particles and a processing optimization can further significantly improve the already quite good performance of the interior seeded Y-123 material.

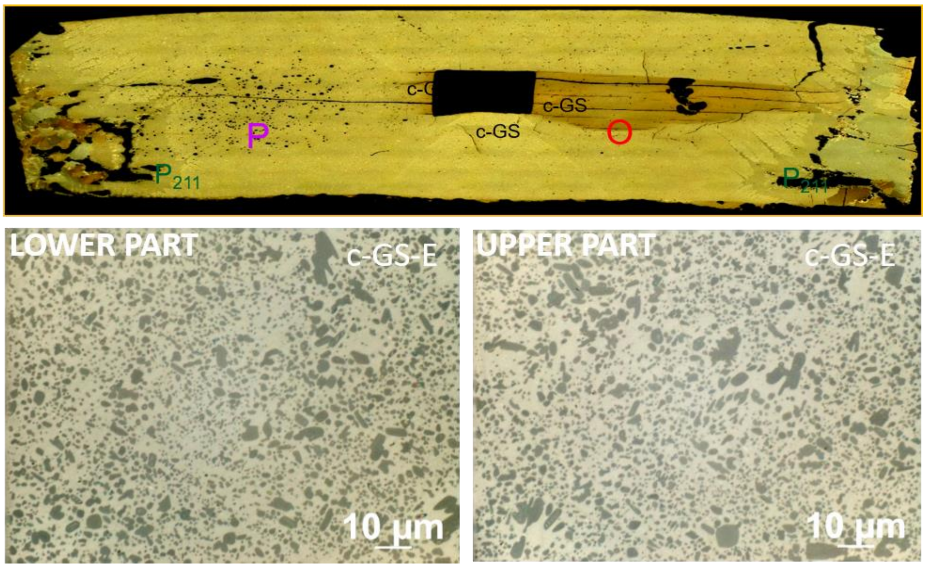

Figure 4. Optical micrograph of the Y-123 material cut along a/c- plane (top), secondary phase particles dispersion in c-GS-E at the upper and lower part of the Y-123 bulk material prepared by interior seeded method.

The structure of the interior-seeded Y-123 material is seen on the sample's cut in the a/c–plane polished up to optical smooth. Figure 4 shows the optical micrograph in polarized light of the Y-123 material produced by the interior seeding method combined with the IG technique. Figure shows the growth starting from the seed. A complicated growth of Y-123 started at the lower part of the bulk. The c-growth sectors (c-GS) are bright due to a low Y-211 content. The 211 particles were pushed away from the sample rim (P_{211}). The dark-yellow part of the sample is oxygenated (O) due to the extended cracks along a/b-planes. The black spherical objects are pores (P). The high-magnification micro-graphs of the top and bottom part of the sample clearly indicate the uniform dispersion of the secondary 211 particles (Fig. 4., bottom). Beside the fine precipitates some larger particles appear in some parts. It shows a space for a further refinement of the secondary phase, which is known to improve critical current density (J_c) in any superconductor.

4. Conclusions

We produced single-grain bulk Y-123 superconductors by infiltration growth process combined with the top and interior seeding methods. Magnetization results confirmed that samples were of good quality with superconducting transition starting at ≈ 91.5 K (interior seeding) and at ≈ 92.1 K (top seeding). The transition was 1.5 K and 1.9 K wide, respectively. The respective J_c-values at 77 K, *H*//*c*-axis, were 44,000 A/cm^2 and 37,000 A/cm^2 (self-field) and 7,750 A/cm^2 and 6,000 A/cm^2 (2 T). Although the electromagnetic and superconducting performance of the interior-seeded material was somewhat better than that of the top-seeded one, the microstructural observation by optical microscopy indicated that a further refinement of Y-211 is possible that can further improve the performance of the Y-123 material prepared in this promising way. The present results indicate that interior seeding can help to prepare thicker high-quality IG-processed Y-123 bulks, with a higher growth speed along *c*-axis direction.

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