J. Am. Ceram. Soc., **99** [9] 3111–3119 (2016)
DOI: 10.1111/jace.14315
© 2016 The Authors. *Journal of the American Ceramic Society* published by Wiley Periodicals, Inc. on behalf of American Ceramic Society (ACERS)
This is an open access article under the terms of the Creative Commons Attribution License, which permits use, distribution and reproduction in any medium, provided the original work is properly cited.

Journal

Microstructure and Composition of Primary and Recycled Single Grains of YBCO, GdBCO-Ag, and SmBCO-Ag Bulk Superconductors

Jasmin V. J. Congreve,† Yunhua H. Shi, Antony R. Dennis, John H. Durrell, and David A. Cardwell

Department of Engineering, University of Cambridge, Cambridge CB2 1PZ, UK

The widespread use of single grain RE-Ba-Cu-O [(RE)BCO] bulk superconductors, where RE is typically Sm, Gd, or Y, is, in part, limited by the relatively high costs of precursor powders and the low success rate of the manufacturing process. Both these problems can be addressed by recycling primary-processed grains in which the initial growth process has failed in some way. Key to the use of recycled grains in practical applications is an assurance that their properties and performance are not inferior to those of primary grown grains. In this work, we describe the differences between the growth process, microstructure, and properties of primary and recycled (RE)BCO single grains. We observe that the mechanism of growth is the same for both primary and recycled single grain samples in all three RE-based systems investigated. In the recycling process additional liquid-rich phase powder is provided beneath a failed sample, whereby this liquid phase infiltrates upwards and contributes a sufficient concentration of additional RE species at the growth front to enable samples to grow relatively easily in the form of single grains by producing a more uniform composition at the growth front, which leads directly to an increased tolerance to the presence of Ag and Ce-rich agglomerates. Importantly, we observe that the recycled samples have a much more uniform composition, and therefore exhibit more uniform superconducting properties, than single grain samples fabricated by a primary grown process.

Keywords: *grain growth; microstructure; particle size distribution; superconductors*

I. Introduction

High-temperature, single grain RE-Ba-Cu-O [(RE)BCO] bulk superconductors, where RE is a rare-earth element or Y, have many potential applications, including magnetic bearings, flywheel energy storage systems, and rotating electrical machines[1–3] due to their ability to trap large, stable magnetic fields far in excess of those possible in permanent magnets.[4] The field trapping ability of bulk (RE)BCO depends on the macroscopic critical current (J_c) which, in turn, depends upon both the flux pinning ability, provided by nanosize defects and inclusions, and the absence of high angle grain boundaries in the single grain bulk microstructure. High angle grain boundaries reduce significantly the maximum achievable critical current density by providing physical barriers to the flow of superconducting current.[5] Therefore, bulk superconductors must be produced in the form of large single grains (or quasi single crystals) if they are to support large critical current densities and generate large magnetic fields.

Single grains are fabricated commonly by the so-called top seeded melt growth (TSMG)[5,6] process in which the sample is heated initially above its peritectic temperature (T_p), leading to the decomposition of the initial solid phase to a secondary solid phase and a BaCuO liquid phase, and then cooled slowly through T_p to allow recrystallization under controlled thermal conditions. The undercooling, ΔT (= $T - T_p$), is limited during the grain growth process to ensure that growth occurs only from a seed crystal placed on the top surface of the sample. The peritectic reaction for the YBCO system, $Y_2BaCuO_5 + Ba_3Cu_5O_8$ (liquid) → $YBa_2Cu_3O_{6+\delta}$,[7] for example, occurs at a temperature of about 1000°C. This produces a superconducting $(RE)Ba_2Cu_3O_7$ (RE-123) phase matrix that contains a distribution of discrete, nonsuperconducting $(RE)_2BaCuO_5$ (RE-211) inclusions that contribute directly to flux pinning.[8,9] The TSMG process is characterized by a relatively large number of processing variables, the effect of many of which on the growth process is not fully understood. The growth of single grains is very sensitive to changes in the growth variables, which makes the likelihood of failure of the single grain growth process relatively high, resulting frequently in the production of multigrain samples. These multigrain samples are usually discarded as waste product given that they contain grain boundaries, and hence exhibit low J_c, and are therefore generally useless for practical applications.

The rare-earth precursor powders used in the TSMG process are of high purity (99.9%) and so are relatively expensive. In addition, expensive alloying elements such as Ag and Pt are also used typically to improve the properties of (RE)BCO superconducting grains. Waste samples that fail to grow in the form of single grains, therefore, is both an expensive and an inefficient use of material resources. As a result, recycling of these failed samples is highly desirable and increases both the economic viability and environmental sustainability of the production of bulk (RE)BCO single grains. A successful recycling process will, in turn, increase significantly the viability of bulk superconductors for use in practical applications.

A technique to recycle failed single grains has been developed by the Cambridge Bulk Superconductivity Group that is both cheap and easy to apply, and which has been found to be reliable in the fabrication of complete single grains.[10] The reasons for the success of the recycling technique are not clear, however, so we have repeated systematically the recycling process for the YBCO, GdBCO-Ag, and SmBCO-Ag systems. The superconducting properties of the recycled samples have been measured to confirm the viability of the recycling process and compared with those of single grains fabricated by a primary growth process. A detailed analysis of the microstructure and composition has been undertaken at discrete points along both the *a* and *c* axes of the single grain samples using scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDX) for both primary grown and recycled samples from all three RE systems. This has enabled the trend in distribution of the RE-211 phase, liquid phase and RE-123 phase and RE-211 particle

A. Goyal—contributing editor

Manuscript No. 37923. Received December 15, 2015; approved April 20, 2016.
†Author to whom correspondence should be addressed. e-mail: jvjc2@cam.ac.uk

size to be analyzed. Conclusions about the success of the recycling growth process have been drawn based on the results of these investigations and possible further research to improve the primary grown process has been identified.

II. Experimental Details

(1) Sample Growth by TSMG and Recycling

The recycling technique is based on the TSMG process in air,[6,7] which is used widely for primary single grain processing. The TSMG technique has drawn inspiration from the infiltration growth process.[11,12] The initial stage of the recycling process involves infiltration growth, which involves placing a precursor pellet of the liquid-rich phase composition beneath the failed sample to provide additional liquid phase at elevated temperature. This, in turn, enables a small amount of infiltration growth to occur during recycling to replenish liquid lost in the failed primary grown process.

The failed samples for recycling were prepared as reported elsewhere.[10] A single crystal seed was placed centrally on top of a precursor buffer pellet of the appropriate composition for the growth of a single grain.[13–18] Conventional NdBCO seeds were used for the YBCO single grain samples, whereas NdBCO-MgO generic seeds were used for the GdBCO-Ag and SmBCO-Ag samples due to their higher melting temperatures.[19]

In each case, the assembly was heated in air using thermal profiles employed for the conventional TSMG process.[5,6] These were the same as those used for the primary grown TSMG process, which enabled primary grown samples of the same diameters to be melt processed in the furnace alongside the recycled samples. Following successful growth in the form of single grains, the samples were then annealed in an oxygen atmosphere in a tube furnace using the following thermal conditions: YBCO, 420°C for 10 days; GdBCO-Ag, 400°C for 12 days; SmBCO-Ag, 360°C for 15 days.

In total 14 YBCO, 19 GdBCO-Ag and 30 SmBCO-Ag failed samples were recycled as part of this investigation. In addition, 2 YBCO, 3 GdBCO-Ag, and 5 SmBCO-Ag primary grown samples were produced using the TSMG process to allow comparison between the recycled and primary grown samples. Figure 1 shows a selection of the samples examined as part of this investigation. Single grain samples that had been recycled successfully were identified visually by the presence of characteristic, fourfold facet lines that are visible on both the top surface of the sample and which extend to the sides of the single grains.

(2) Measurements of Superconducting Properties

The trapped field of each sample at 77 K was measured after field cooling in an external magnetic field of approximately 1.4 T, supplied by a DC electromagnet. The maximum trapped field at both the top and bottom surface of each sample was measured using a rotating array of 20 Hall probes at a distance of approximately 1 mm from the surface of the sample for a random selection of successfully recycled single grain samples for each of the three systems.

One recycled and one primary grown single grain sample for each RE system was chosen randomly to be cut into subspecimens for measurement of T_c and J_c, observation of the microstructure and composition analysis as a function of position within each single grain. An additional single, recycled SmBCO-Ag grain was cut and analyzed separately due to the large variation in microstructure and phase content observed in primary grown samples of this composition.[20] A minimum of three subspecimens of approximate dimensions 1.5 mm × 2.0 mm × 1.2 mm corresponding to a slice

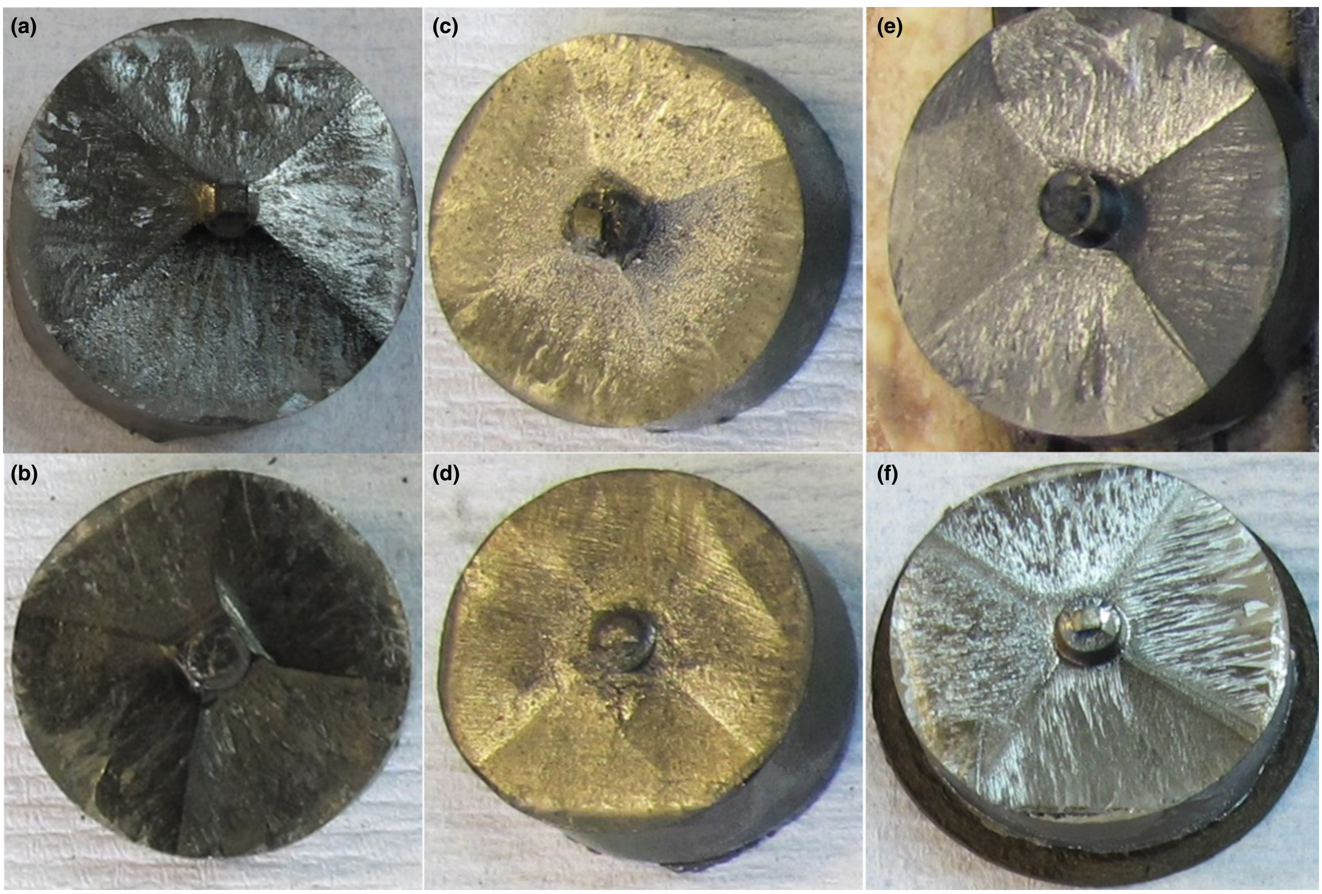

Fig. 1. Superconducting single grains of diameter 25 mm: (a) YBCO primary grown, (b) YBCO recycled, (c) GdBCO-Ag primary grown, (d) GdBCO-Ag recycled, (e) SmBCO-Ag primary grown, (f) SmBCO-Ag recycled.

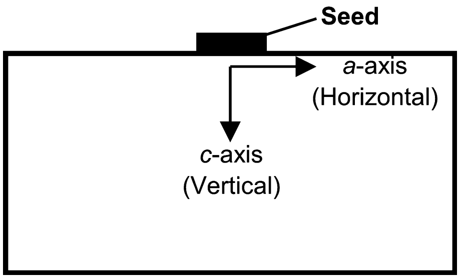

Fig. 2. Schematic illustration of the orientation of the axes within the single grain.

directly below the seed were cut from the parent single grain and analyzed to identify variations in superconducting properties across the sample. A SQUID (superconducting quantum interference device) magnetometer (Quantum Design MPMS XL, San Diego, CA) was used to measure J_c as a function of applied magnetic field at constant temperature and T_c at a constant field of 20 Oe. The value of T_c for each subspecimen was established, with the value of J_c determined from the measured hysteresis of the magnetic moment using the Bean critical state model.[21]

The other half of the batch of seven samples used to measure J_c and T_c were prepared for microstructure and composition analysis. The microstructure was observed using an optical microscope [Nikon Eclipse ME600 optical microscope (Nikon, Tokyo, Japan) with a Motic Multicam Pro 282A camera (Motic, Hong Kong, China)] at a magnification of 500× at intervals of 1 mm along both the *a* and *c* axes of the single grain samples. The orientation of the single grain axes is illustrated schematically in Fig. 2. The microstructure of these samples was also observed for areas of approximately 40 μm × 40 μm at intervals of 1 mm at a distance approximately 1 mm directly below the seed using a scanning electron microscope (SEM). The average composition of each of these areas was recorded using an energy dispersive X-ray spectroscopy analyzer (s-3400; Hitachi, Tokyo, Japan).

The variation in composition with distance from the seed was used to illustrate the trend in composition through the bulk single grain along both the *a* and *c* axes directions. These data were also used to identify the differences in composition between the primary grown and recycled samples of each of the three RE systems investigated. These data were normalized according to atomic percent of the constituent RE (Y, Gd, Sm), Ba, and Cu elements. The expected stoichiometric atomic percentages of RE, Ba, and Cu were also plotted to allow comparisons to be made. The amount of RE-211 phase present correlates directly with the ratios RE/Ba and RE/Cu, so these data can be used to identify trends in the RE-211 inclusion content throughout the single grain.

III. Results and Discussion

(1) Superconducting Properties

The superconducting properties of recycled single grained samples were measured to ensure viability in comparison with primary grown samples.

Single grains exhibit a characteristic trapped field profile consisting of a single peak that decreases radially with distance from the position of the seed with a continuous, smooth gradient. A typical trapped field profile for a single grain sample of SmBCO-Ag can be seen in Fig. 3, with typical values of maximum trapped field for the recycled samples given in Table I. These values are similar to those measured previously for recycled samples.[10] It can be concluded that the recycled samples typically exhibit a maximum trapped field of approximately 15% lower than primary grown samples which is in agreement with previous results.[10]

The values of T_c were measured for a number of subspecimens cut from the central cross section of the relevant single

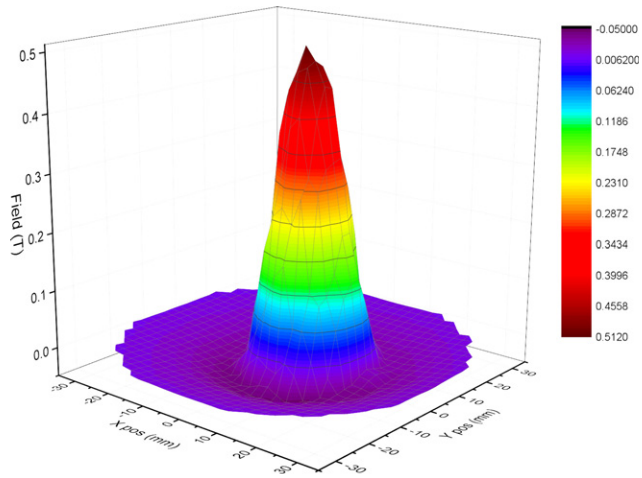

Fig. 3. A typical example of a trapped field profile for a single grain, SmBCO-Ag recycled sample.

Table I. Typical Values of Maximum Trapped Field and Average T_c and J_c of Recycled Samples of YBCO, GdBCO-Ag, and SmBCO-Ag

	Maximum trapped field (*T*)	T_c (K)	J_c (A/cm^2)
YBCO	0.607	90.3	40259
GdBCO-Ag	0.751	92.9	23069
SmBCO-Ag	0.717	93.5	38470

grain at different locations relative to the seed and the average over all specimens was calculated for each sample. The average values of T_c are given in Table I. The recycled samples exhibited a lower average value of T_c than the primary grown samples for all the systems studied, except for the recycled SmBCO-Ag sample, which had a T_c value that was 0.7 K higher than the primary grown sample.

The values of $J_c(0)$ were measured for each of the subspecimens used to measure T_c. The average value of $J_c(0)$ in all three systems was higher for the primary grown sample than for the recycled sample, as can be seen in Fig. 4. The YBCO samples exhibit only a small difference in the average value of J_c between the primary and recycled samples, whereas in the GdBCO-Ag and SmBCO-Ag systems the average values of J_c are significantly lower for the recycled sample than for the primary grown sample. The average value of J_c is very different for the two recycled samples and for the recycled and primary grown samples for the SmBCO-Ag system. The reduction in T_c and $J_c(0)$ for recycled samples in comparison to primary grown samples is in agreement with previous results.[10]

The J_c values showed a much smaller variation throughout the bulk single grain for the recycled samples, except in the case of GdBCO-Ag where fewer subspecimens were analyzed. The variation in the values of J_c throughout the sample for the primary grown samples was much larger than the variation across the subspecimens for the recycled samples in the YBCO and SmBCO-Ag systems. This suggests that the distribution of flux pinning centers in the recycled samples is likely to be more uniform. The superconducting properties of the recycled (RE)BCO-Ag are similar to those reported elsewhere.[22]

(2) Microstructural Analysis

Images were taken at 1 mm intervals along both the *a* and *c* axes of all seven samples using an optical microscope and using a scanning electron microscope. Examples of the images produced using the SEM are shown in Fig. 5.

The SEM images of the GdBCO-Ag recycled sample in Fig. 5 reveal that the density of Gd-211 particles increases

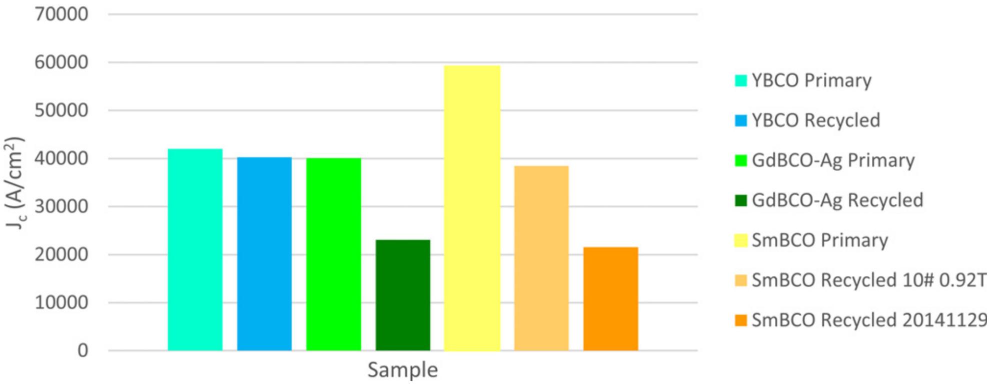

Fig. 4. Average value of $J_c(0)$ for each sample analyzed.

with distance from the seed in the horizontal direction, as predicted by particle pushing/trapping theory.[6] A similar increase in RE-211 particle density is also observed in the vertical direction in all the primary grown (RE)BCO and (RE)BCO-Ag single grain samples. The large dark region indicated in the 0 mm image is a pore, whereas the large, brightly colored regions with a small black-in-contrast adjoining region, as seen in the 5, 7, and 11 mm images (and labeled in the image at 5 mm), are Ag-rich agglomerates as confirmed by EDX.

The magnitude of the critical current density is determined by the ability of the sample microstructure to pin magnetic flux (i.e., a fluxoid), with a larger macroscopic J_c producing a larger trapped field.[23] Both nanosize defects and nonsuperconducting inclusions act as particularly effective pinning centers in bulk (RE)BCO superconductors. It has been reported that un-reacted, un-dissolved small Y-211 particles are typically trapped in the as-grown Y-123 single grain phase matrix during peritectic solidification, which creates Y-211/Y-123 interfaces that are considered generally to correlate with the formation of effective flux pinning centers and to improve J_c of the single grains.[24] In particular, J_c is reported to increase with the V_{211}/d_{211} ratio, where V_{211} is the Y-211 volume fraction and d_{211} is the average size of the Y-211 particles.[25,26] The more uniform the distribution of (RE)-211 particles, therefore, the more uniform the critical current, and the higher the density of small-sized (RE)-211 and the larger the magnitude of the critical current.

The density of RE-211 particles has been observed to increase with distance from the seed in all the systems investigated in this study. However, the distribution of RE-211 particles is more uniform for the recycled samples and the uniformity of the distribution of RE-211 increases from Y to Gd to Sm. This suggests that the provision of adequate liquid phase during the grain growth process, as is provided during the recycling process, enables the peritectic reaction to continue to progress even at the edge of the sample. This, in turn, enables growth of the single grain to continue to the edge of the sample to produce a more uniform sample, which is highly desirable for practical applications.[23]

The RE-211 inclusions become both larger with increasing distance from the seed and noticeably elongated during the recycling process. This is expected since the RE-123 phase is remelted during recycling to form new RE-211 phase. This RE-211 phase may either nucleate on preexisting RE-211 particles or nucleate directly to form new RE-211 phase particles. The liquid phase reacts with these RE-211 particles to form RE-123 when a grain regrows during the recycling process. An increase in the size of the flux pinning inclusions for the same density of RE-211 phase present, therefore, will reduce the number of pinning centers present, and the increase in pinning due to the increase in pinning centre size will be less than that if there were a larger number of smaller pinning centers (of equivalent volume). This is likely to be the reason why the recycled samples exhibit a lower maximum critical current density than the primary grown samples from the same system, as is apparent in Fig. 4.

(3) Compositional Analysis

The average composition of each area imaged using the SEM analysis was measured, along with the composition of a number of the phases present. The data for all three systems for the recycled and primary grown samples are shown in Fig. 6.

Composition analysis in conjunction with the SEM images has been used to identify the phases present in the microstructure, including cerium-rich, silver-rich, RE-123, and RE-211 phases. This has revealed that the average composition at each location in the single grain samples fluctuates around the original stoichiometric composition for most of the points analyzed across the full sample range. Any large fluctuations at a given point in the single grain microstructure can be explained using the SEM. In many cases the SEM images reveal the presence of large agglomerates, large pores, cracks, or regions of abnormal microstructure. For example, a large change in normalized composition is observed at 1 mm from the position of the seed in the horizontal direction for the primary grown SmBCO-Ag sample, corresponding to the presence of a large, Ag-rich agglomerate.

(4) Relation Between Composition and Growth

Each position within a single grain sample fabricated by TSMG at a given distance from the location of the seed along either the *a* or *c* axes direction corresponds to an equivalent point in time during the solidification of the growth front. The growth of (RE)BCO single grains is usually interpreted by a "black box" approach given that changes in composition during the growth process itself are difficult to detect. For this reason, it is assumed commonly that the local composition on completion of grain growth is the same as that present during the growth process.

The general trend in composition seen in Fig. 6 is that of a reduction in the normalized atomic percentage of Cu, indicating the shortage of liquid phase (liquid phase composition: $Ba_3Cu_5O_8$) and an increase in the atomic percentage of RE element with increasing distance from the seed for both the horizontal and vertical direction. The level of barium, however, remains constant throughout the sample, suggesting that the atomic percentage of RE-211 phase increases with distance from the seed, whereas the atomic percentage of the RE-123 phase decreases with distance from the seed. This can also be seen in the optical microscope and SEM images of the single grain samples.

The ratios of RE/Cu and RE/Ba at different points in the single grains, which usually represent the trend of RE-211 concentration, were plotted against position within the sample, as shown in Fig. 7. These data show that both ratios

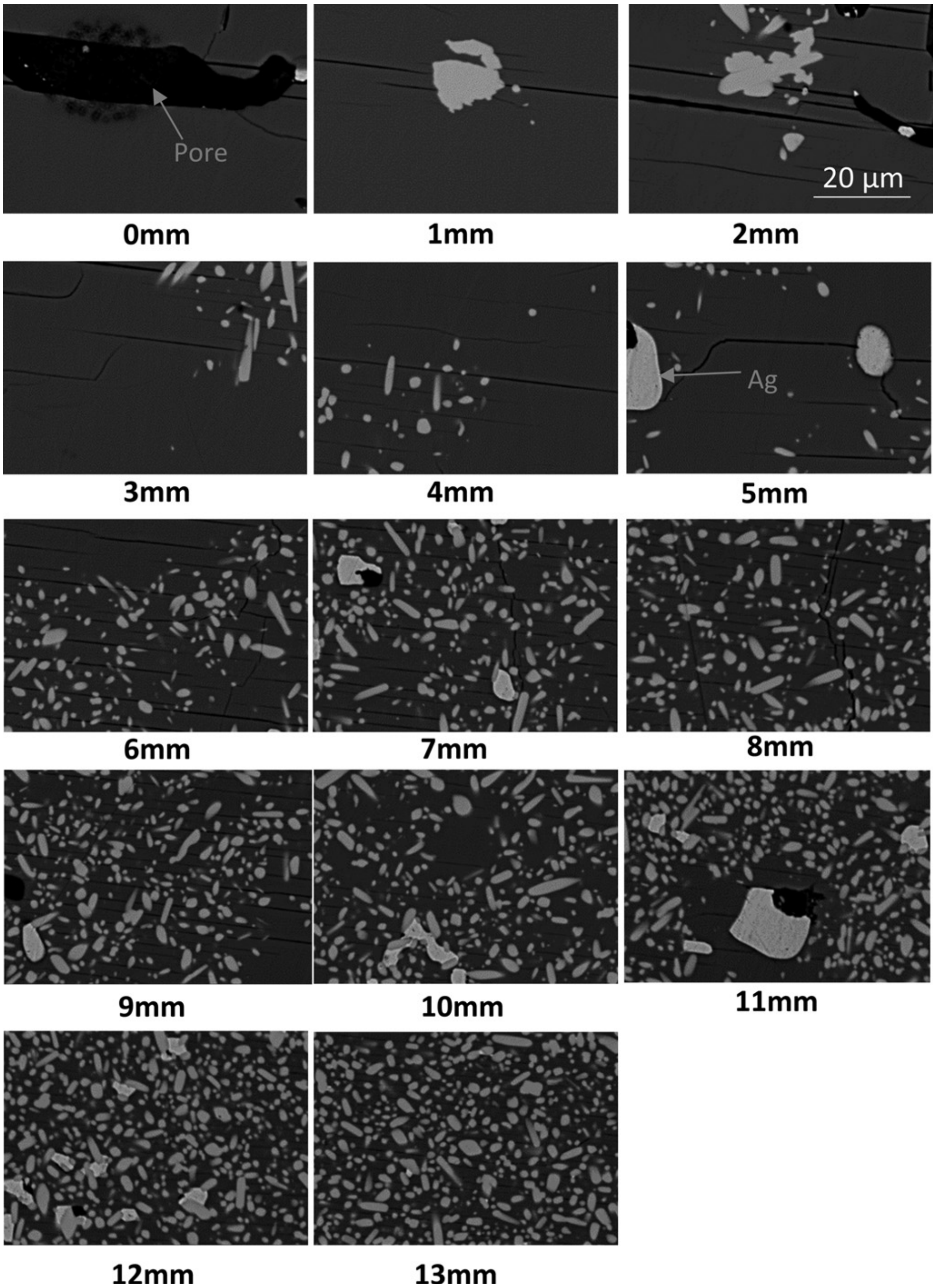

Fig. 5. SEM images of the recycled GdBCO-Ag grain with increasing distance from the seed in the horizontal direction. The dark gray matrix is the Gd-123 superconducting phase, whereas the lighter colored sections are the Gd-211 phase, which provides flux pinning.

increase with distance from the seed and that this increase is much larger for all systems in the horizontal direction than in the vertical direction. This suggests that there is a much greater change in the ratio of RE-123 and RE-211 in the horizontal direction and, therefore, there is greater grain growth in the a-b direction (horizontal direction) than in the c-direction in the samples fabricated in this study. This is a consequence of the liquid phase draining much more quickly in the vertical direction, under the influence of gravity, than in the horizontal direction during the growth of the single grains.

It can be seen that the ratios of RE/Cu and RE/Ba for the recycled grains follow the same trend much more closely than those for the primary grown samples (the curves in the former are more clearly parallel with each other). This may suggest that the overall compositions of the RE-123 and RE-211 phases are more uniform throughout the sample, which may lead, in turn, to more uniform superconducting properties of the material. As discussed above, an adaptation of infiltration growth provides additional liquid phase during the recycling process, this additional liquid phase is responsible for the greater uniformity in the composition of the recycled samples.

From Fig. 7 it can be seen for the better of the two SmBCO-Ag recycled single grains that, in contrast to the data for the other samples, the trend of the RE/Ba and RE/Cu fluctuates about an average value but does not show a trend that increases or decreases with distance from the seed. As this sample exhibits a very high trapped field, this suggests that the uniformity in the sample composition ratios may have a direct influence on the achievable value of this important parameter.

The large Ag-rich, Ce-rich and combined Ce and Ag-rich clusters apparent in Fig. 5 may be one of the reasons

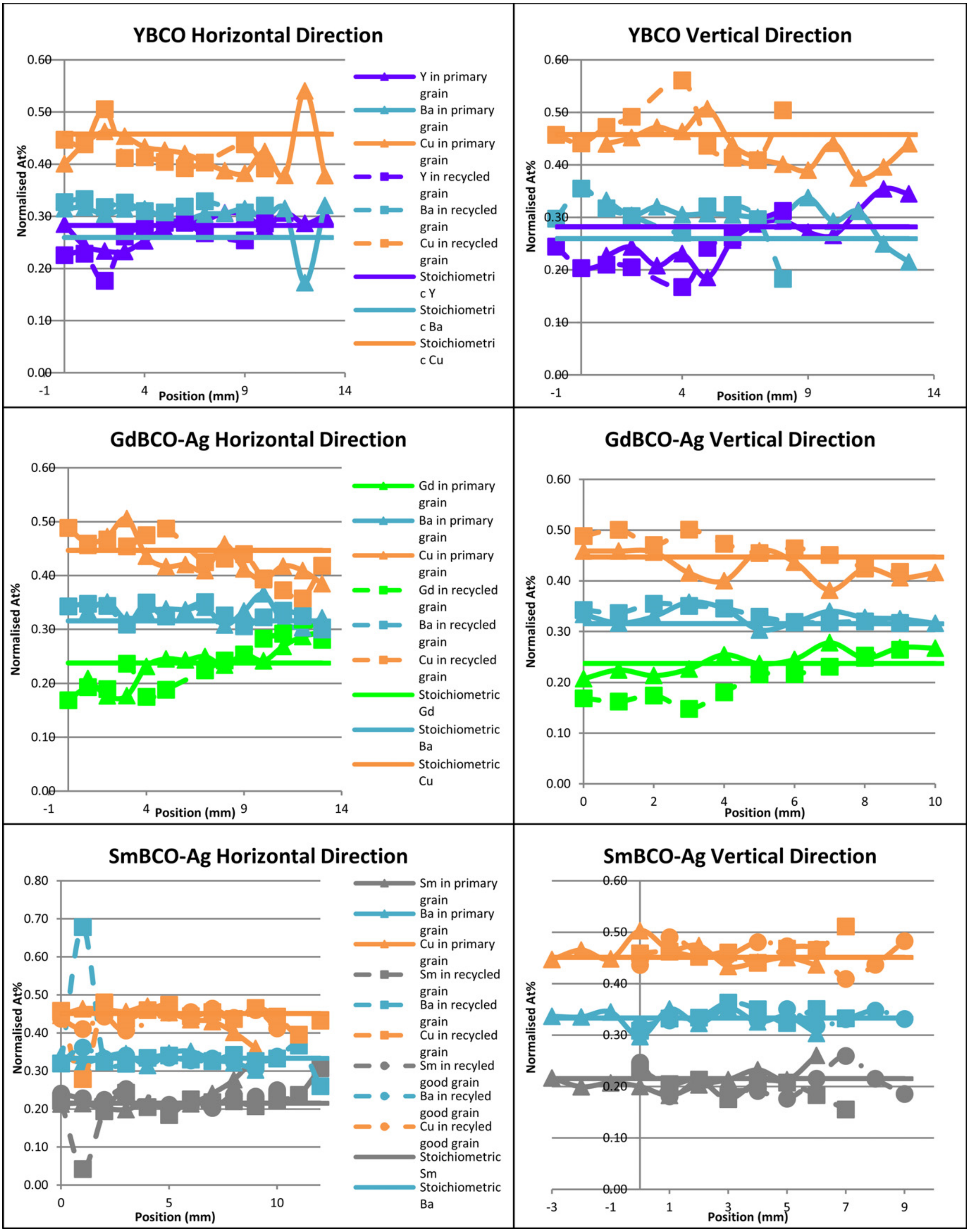

Fig. 6. Graphs of the variation in normalized composition with position for the primary grown and recycled samples in each of the three systems.

responsible for the failure of single grain growth. These clusters may be generated due to poor mixing of the precursor powders used in the primary growth process. The high success rate of the recycling process is likely to be because this process is tolerant to the presence of these Ag, Ce and Ag- and Ce-rich clusters due to appropriate liquid phase that provides additional Y, Gd, or Sm, which is made available at the growth front. This additional liquid phase allows the grain to continue to grow as a single grain, although a large change in the liquid phase composition

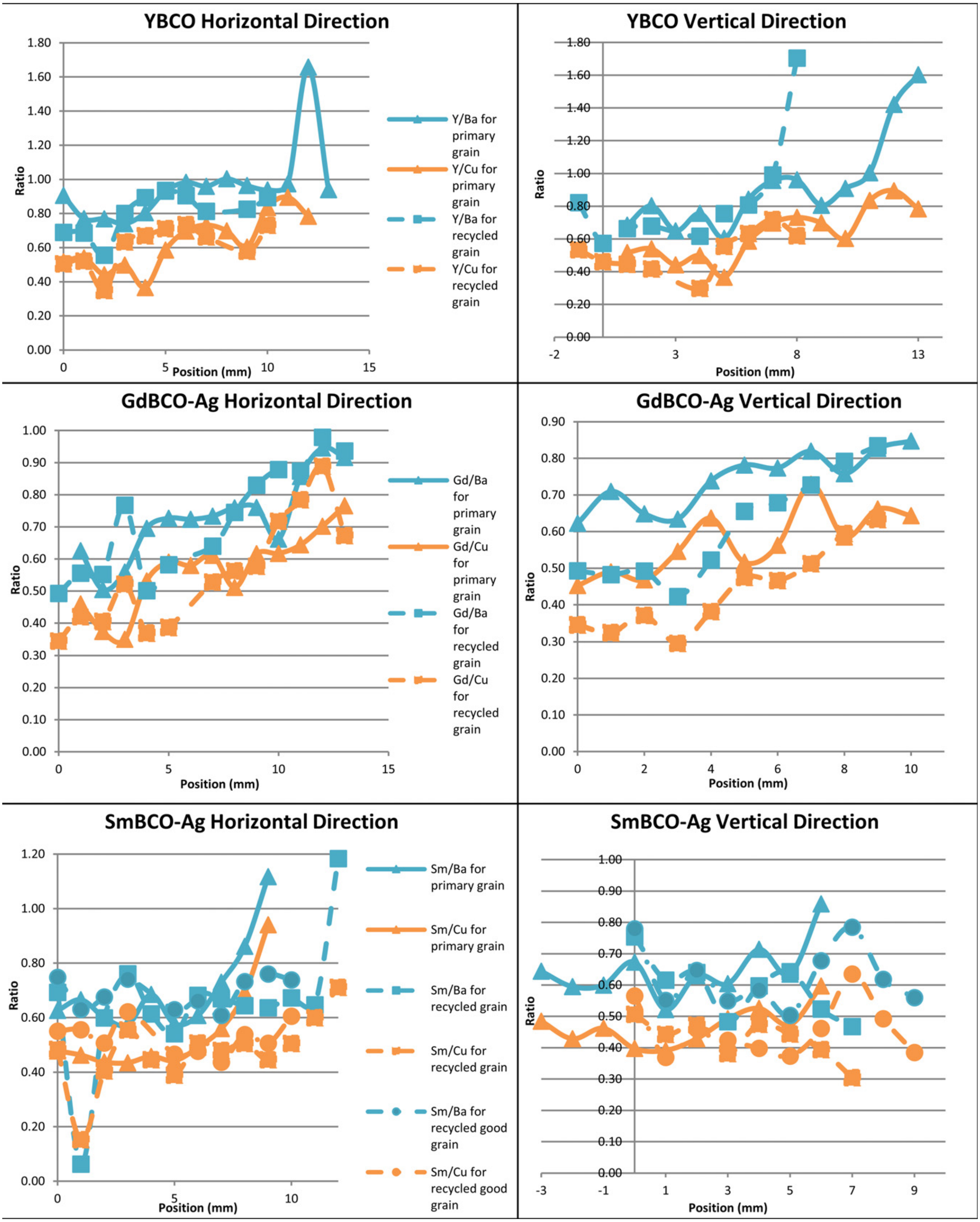

Fig. 7. The variation in the ratio of RE/Ba and RE/Cu with position for the primary grown and recycled samples in each of the three RE systems.

occurs at the growth front where these large clusters have formed.

Both the horizontal and vertical directions in the YBCO system exhibit a similar distribution in composition. However, there is a lower concentration of Y in the recycled sample than in the original primary grown grain within a volume of approximately 4 mm × 8 mm × 8 mm close to the seed, compared with an area of 4 mm × 10 mm × 10 mm for the primary grown sample, suggesting that the Y-211 concentration is lower in this region. This suggests that the composition of the recycled sample is much more uniform than that of the primary grown sample.

Both the GdBCO-Ag and SmBCO-Ag systems exhibit a similar trend to that of the YBCO system described above

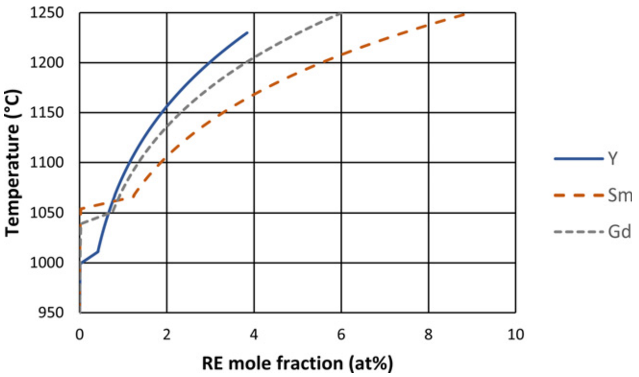

Fig. 8. Liquidous lines of RE elements in the $Ba_3Cu_5O_8$ melt under an air atmosphere.[27]

with the only difference being that the gradients of the graphs become increasingly lower from YBCO to GdBCO-Ag to SmBCO-Ag. This can be explained by the increasing growth rates from YBCO to SmBCO-Ag, which increases in this way due to the increasing solubility of the RE (Y, Gd, and Sm) element in the BaCuO liquid during growth, as shown in Fig. 8.[27] The growth rate during the growth process is controlled by diffusion of the RE element in the liquid phase and so, when the growth front proceeds, the more soluble the element the better the growth process. Therefore, the required RE element is more abundant at the growth front for SmBCO-Ag than for GdBCO-Ag and YBCO.

The compositions of the recycled samples of all of three systems investigated here are significantly more uniform than those of the primary grown samples, as indicated by the flatter slope of the composition graphs. This suggests that the liquid phase provided at the edges of the recycled samples help the superconducting RE-123 phase to form. The recycling method provides adequate liquid phase without leakage and therefore provides an adequate concentration of RE element at the growth front, which enables the recycled samples to grow fully and easily. In addition, and significantly, the recycling technique is tolerant to failed samples containing relatively large, localized agglomerations of Ce or Ag-rich phases.

It has been found that the liquid phase that carries the RE element is important to produce fully grown single grains. As some liquid phase is always lost during the primary growth TSMG process, this suggests that any method that can reduce liquid loss or compensate for the liquid loss during primary grain growth would help the fabrication of good quality single grains.

IV. Conclusions

We have demonstrated that failed YBCO, GdBCO-Ag, and SmBCO-Ag melt processed samples can be recycled successfully and reliably by the controlled replenishment of liquid phase lost during the primary growth process. SEM and EDX have been used to observe and analyze the composition of the growth front of primary and recycled single grains of all three systems postprocessing. Comparison of both sets of data has shown that the EDX analysis technique is convincing and reliable and reveals relatively large fluctuations from the composition of the precursor powders. This has been explained by a variety of processing features, including the presence of Ce and Ag-rich agglomerates and pores and cracks at specific locations within the single grain samples. The compositional analysis suggests further that the growth mechanisms are the same for both the primary grown and recycled grains in all three systems investigated.

The additional liquid phase provided in the recycling process dissolves the RE element (Y, Gd, Sm) to form the RE-123 phase at the growth front, which is fundamental to the growth of single grains. The provision of adequate liquid phase yields a sufficient RE element concentration at the growth front to enable single grain samples to grow easily, to provide a more uniform composition at the growth front and to support growth in the presence of agglomerates of Ag and Ce-rich phases. The greater change in the ratio of RE-123 and RE-211 phase content observed in the horizontal direction compared to the vertical direction suggests that there is much greater grain growth in the a–b (horizontal) direction than in the c (vertical) direction, which is a direct result of the liquid phase draining more quickly from the sample in the vertical direction under the influence of gravity.

The additional liquid phase provided in the recycling process has produced recycled samples with a more uniform composition than primary grown samples. This increased uniformity has yielded recycled samples that have more uniform superconducting properties throughout the superconducting bulk microstructure. It may be possible to adapt the method used to recycle failed samples to improve the uniformity of the superconducting properties of primary grown samples and to increase the success rate of the primary single grain growth process.

Acknowledgments

We acknowledge the Engineering and Physical Sciences Research Council (EPSRC grant ref. EP/K02910X/1) for financial support. Additional data related to this publication is available at the University of Cambridge data repository [https://www.repository.cam.ac.uk/handle/1810/252962]. All other data accompanying this publication are directly available within the publication.

References

[1]A. M. Campbell and D. A. Cardwell, "Bulk High Temperature Superconductors for Magnet Applications," *Cryogenics*, **37**, 567–75 (1997).

[2]F. N. Werfel, U. Floegel-Delor, R. Rothfeld, T. Riedel, B. Goebel, et al., "Superconductor Bearings, Flywheels and Transportation," *Supercond. Sci. Technol.*, **25**, 014007 (2012).

[3]B. Li, D. Zhou, K. Xu, S. Hara, K. Tsuzuki, et al., "Materials Process and Applications of Single Grain (RE)-Ba-Cu-O Bulk High Temperature Superconductors," *Physica C*, **482**, 50–7 (2012).

[4]J. H. Durrell, A. R. Dennis, J. Jaroszynski, M. D. Ainslie, K. G. B. Palmer, et al., "A Trapped Field of 17.6 T in Melt-Processed, Bulk Gd-Ba-Cu-O Reinforced With Shrink-fit Steel," *Supercond. Sci. Technol.*, **27**, 082001 (2014).

[5]D. A. Cardwell, "Processing and Properties of Large Grain (RE)BCO," *Mater. Sci. Eng. B*, **53**, 1–10 (1998).

[6]K. Sawano, M. Morota, M. Tanaka, T. Sasaki, K. Kimura, and S. Takebayashi, "High Magnetic Flux Trapping by Melt-Grown YBaCuO Superconductors," *Jpn. J. Appl. Phys.*, **30**, L1157–9 (1991).

[7]V. Antal, M. Kaňuchová, M. Šefčiková, J. Kováč, P. Diko, et al., "Flux Pinning in Al Doped TSMG YBCO Bulk Superconductors," *Supercond. Sci. Technol.*, **22**, 105001 (2009).

[8]M. Murakami, *Melt Processed High-Temperature Superconductors*. World Scientific, Singapore, 1992.

[9]G. Krabbes, G. Fuchs, W. R. Canders, H. May, and R. Palka, *High Temperature Superconductor Bulk Materials Fundamentals – Processing – Properties Control – Application Aspects*. Wiley-VCH Verlag GmH & Co, KGaA, Weinheim, 2006.

[10]Y. Shi, D. K. Namburi, M. Wang, J. Durrell, A. Dennis, and D. Cardwell, "A Reliable Method for Recycling (Sm, Gd, Y)$Ba_2Cu_3O_{7-\delta}$ Bulk Superconductors.," *J. Am. Ceram. Soc.*, **98** [9] 2760–6 (2015).

[11]G.-Z. Li, D.-J. Li, X.-Y. Deng, J.-H. Deng, and W.-M. Yang, "Infiltration Growth and Crystallization Characterisation of Single-Grain Y-Ba-Cu-O Bulk Superconductors," *Cryst. Growth Des.*, **13**, 1246–51 (2013).

[12]D. Zhou, S. Hara, B. Li, K. Xu, J. Noudem, and M. Izumi, "Significant Improvement of Trapped flux in Bulk Gd–Ba–Cu–O Grains Fabricated by a Modified top-Seeded Melt Growth Process," *Supercond. Sci. Technol.*, **26**, 015003 (2013).

[13]T. Y. Li, L. Cheng, S. B. Yan, L. J. Sun, X. Yao, et al., "Growth and Superconductivity of REBCO Bulk Processed by a Seed/Buffer Layer/Precursor Construction," *Supercond. Sci. Technol.*, **23**, 125002 (2010).

[14]C.-J. Kim, J.-H. Lee, S.-D. Park, B.-H. Jun, S.-C. Han, and Y.-H. Han, "Y_2BaCuO_5 Buffer Block as a Diffusion Barrier for Samarium in Top Seeded Melt Growth Processed $YBa_2Cu_3O_7$-y Superconductors Using a $SmBa_2Cu_3O_7$-δ Seed", *Supercond. Sci. Technol.*, **24**, 015008 (2011).

[15]D. Zhou, K. Xu, S. Hara, B.-Z. Li, and M. Izumi, "MgO Buffer-Layer-Induced Texture Growth of RE-Ba-Cu-O Bulk", *Supercond. Sci. Technol.*, **25**, 025022 (2012).

[16]Y. Shi, A. R. Dennis, and D. A. Cardwell, "A New Seeding Technique for the Reliable Fabrication of Large, SmBCO Single Grains Containing Silver Using Top Seeded Melt Growth," *Supercond. Sci. Technol.*, **28**, 035014 (2015).

[17]N. Devendra Kumar, Y. Shi, W. Zhei, A. R. Dennis, J. H. Durrell, and D. A. Cardwell, "Buffer Pellets for High-Yield, Top-Seeded Melt Growth of Large Grain Y-Ba-Cu-O Superconductors," *Cryst. Growth Des.*, **15**, 1472–80 (2015).
[18]W. Zhao, Y. Shi, A. R. Dennis, and D. A. Cardwell, "Use of Sm-123 + Sm-211 Mixed-Powder Buffers to Assist the Growth of SmBCO and ZrO_2-Doped SmBCO Single Grain, Bulk Superconductors," *IEEE Trans. Appl. Supercond.*, **25** [3] 5801305 (2015).
[19]Y. Shi, N. Hari Babu, and D. A. Cardwell, "Development of a Generic Seed Crystal for the Fabrication of Large Grain (RE)-Ba-Cu-O Bulk Superconductors," *Supercond. Sci. Technol.*, **18**, L13–6 (2005).
[20]Y. Shi, M. Desmedt, J. Durrell, A. R. Dennis, and D. A. Cardwell, "Improving the Superconducting Properties of Single Grain Sm-Ba-Cu-O Bulk Superconductors Fabricated in Air by Increased Control of Sm/Ba Substitution Effects," *Supercond. Sci. Technol.*, **26**, 095012 (2013).
[21]D. X. Chen and R. B. Goldfarb, "Kim Model for Magnetization of Type II Superconductors," *J. Appl. Phys.*, **66**, 2489–500 (1989).
[22]Y. Shi, N. Hari Babu, K. Iida, W. K. Yeoh, A. R. Dennis, et al., "Batch-Processed GdBCO-Ag Bulk Superconductors Fabricated Using Generic Seeds With High Trapped Fields," *Physica C*, **470**, 685–8 (2010).
[23]S. Nariki, N. Sakai, M. Murakami, and I. Hirabayashi, "High Critical Current Density in RE–Ba–Cu–O Bulk Superconductors With Very Fine RE_2BaCuO_5 Particles," *Physica C*, **412–414** [1] 557–65 (2004).
[24]M. Murakami, K. Yamaguchi, H. Fujimoto, N. Nakamura, T. Taguchi, et al., "Flux Pinning by Non-Superconducting Inclusions in Melt-Processed YBaCuO Superconductors," *Cryogenics*, **32**, 930–5 (1992).
[25]D. F. Lee, V. Selvamanickam, and I. Salama, "Influences of Y_2BaCuO_5 Particle Size and Content on the Transport Critical Current Density of $YBa_2Cu_3O_x$," *Physica C*, **202**, 83–96 (1992).
[26]P. Diko, "Growth-Related Microstructure of Melt-Grown $REBa_2Cu_3O_y$ Bulk Superconductors," *Supercond. Sci. Technol.*, **13**, 1202–13 (2000).
[27]C. Krauns, M. Sumida, M. Tagami, Y. Yamada, and Y. Shiohara, "Solubility of RE Elements Into Ba-Cu-O Melts and the Enthalpy of Dissolution," *Z Phys. B*, **96**, 207–12 (1994). □