Journal

A Reliable Method for Recycling (RE)-Ba-Cu-O (RE: Sm, Gd, Y) Bulk Superconductors

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Single grain (RE)-Ba-Cu-O (RE: Sm, Gd, Y) high-temperature superconductors are able to generate high magnetic fields. However, the relatively high cost of the raw materials and the low yield of the manufacturing process have impeded the development of practical applications of these materials to date. This article describes a simple, reliable, and economical method of recycling failed bulk (RE)-Ba-Cu-O (RE: Sm, Gd, Y) samples. Sixty-four failed bulk samples, with diameters up to 31 mm, were recycled with a yield of 90%. The key innovation in this recycling process involves reintroducing the liquid phase into the melt process, which is normally lost during the primary peritectic processing of these materials. This enables the direct re-growth of failed samples from solid form without the need for re-grinding into powder. We also demonstrate that the superconducting performance and microstructure of the recycled samples is similar to that of the primary grown samples.

I. Introduction

BULK single grain rare-earth cuprate superconductors of the (RE)-Ba-Cu-O [(RE)BCO] family, where RE is Nd, Sm, Gd, Y, Yb etc., can generate magnetic fields that are an order of magnitude higher than those achievable using conventional permanent magnets.[1] However, one of the key factors preventing the wide-scale adoption of these materials is the relatively high cost of the raw materials and the low yield of the crystal growth process typically employed.

The top seeded melt growth (TSMG) process[2,3] used to fabricate single grain (RE)BCO superconductors is based on slow, controlled recrystallization following heating through a peritectic decomposition temperature. In the YBCO system, for example, Y_2BaCuO_5 (Y-211) and $Ba_3Cu_5O_8$ (liquid phase, L) react to form $YBa_2Cu_3O_{6.5}$ (Y-123) and O_2 at about 1002°C, with Y-123 forming the superconducting phase matrix.[4,5] Numerous dopants, such as Y-211,[6,7] CeO_2,[8–11] Pt,[12,13] 2411(M),[14] and ZrO_2,[15] have been investigated in an attempt to enhance the properties of (RE)BCO bulk superconductors. Silver, in particular, has been added to improve the mechanical properties[16] of the single grains (which are ceramic in nature). However all these dopants complicate the growth process significantly and nontrivial adjustments to the processing parameters are often required based on limited theory and experience.

Due directly to the complexity of TSMG, the growth process is very sensitive to changes in a large number of process variables, and this makes the likelihood of the failure of single grain growth high. Such failure often results in the formation of multigrain samples. Grain boundaries form physical barriers to the flow of supercurrent, which leads to samples that are effectively useless for practical applications (these samples invariably contain Pt and Ag, which are generally expensive). The cost effectiveness of the TSMG process is limited further by the high-purity nature of the complex rare-earth cuprate precursors (99.9%) required to fabricate single grains, which are also expensive. The ability to recycle failed bulk materials is therefore highly desirable and important, both in terms of potential economic benefit and environmental sustainability of the manufacturing process.

It has been reported that failed YBCO[17,18] and GdBCO-Ag[19,20] bulk superconductors can be refabricated through a recycling process. Unfortunately, the success rate of these recycling methods has not been reported specifically,[17–20] although, from our experience,[20] the recycling success rate is typically ~20%. Inspired by the infiltration growth method,[21] we have developed a reliable method for recycling any failed (RE)BCO sample by replenishing the liquid phase lost during the primary growth process without first reprocessing the failed samples into powders. To date, we have successfully recycled 58 bulk (RE)-Ba-Cu-O (RE: Sm, Gd, Y) samples (including 16 YBCO, 1 GdBCO, 16 GdBCO-Ag, and 25 SmBCO-Ag) of diameters up to 31 mm using this new technique. In this study, we describe the new recycling method and report analyses of the microstructures, compositions, and superconducting properties of the recycled samples. The mechanisms that may underlie the high success rate of this recycling process, compared to primary growth, are explored.

II. Experimental Details

The method described here is typical of that used for melt-processing of all 64 failed RE(Sm, Gd, Y)BCO-Ag samples. A representative batch of 17 failed, bulk YBCO samples were recycled in this work, 16 of which were of diameter 25 and one was of diameter 31 mm. The top and bottom surfaces of the failed samples were polished prior to recycling to produce a clean and flat surface, and the weight of each polished sample was recorded. Figure 1 shows a photograph of the seven polished and unpolished samples, from which it can be seen that the failed samples vary significantly in their appearance (indicating different failure mechanisms for growth in the form of a single grain). The compositions of the failed YBCO samples in the primary process are in the range of (75–70 wt% Y-123 + 25–30 wt% Y-211) + 0.5–1.0 wt% CeO_2. Typical symptoms of single grain growth failure include the presence of multigrains, randomly oriented macro cracks and the formation of large voids, particularly in the vicinity of the seed (i.e., the center of the sample), as can be seen in Fig. 1. To improve the success rate of seeding, a buffering technique was adopted in YBCO, GdBCO-Ag, and SmBCO-Ag recycled process, for example, small pellets
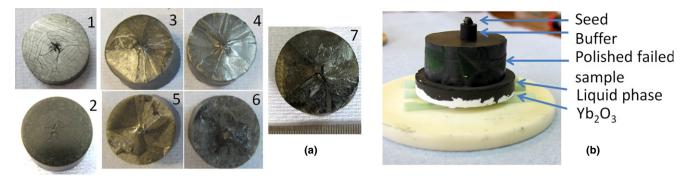
Fig. 1. Procedure for recycling failed Y-Ba-Cu-O samples.

Table I. Summary of the Trapped Fields and Weight Changes of the Recycled Y-Ba-Cu-O Samples

Sample number	YR01	YR02	YR03	YR04	YR05	YR06	YR07
Sample diameter (mm)	25	25	25	25	25	25	31
Sample weight before melt-processing (g)	44.0	44.0	38.1	44.0	44.0	44.0	75.0
Sample (failed) weight after melt-processing (g)	41.32	41.30	35.41	40.77	40.66	41.11	71.17
Weight loss (%)	6.1	6.1	7.1	7.3	7.6	6.6	5.1
Sample weight before recycling (g)	35.53	34.80	30.58	36.40	34.08	32.93	61.32
Sample weight after recycling (g)	37.26	36.87	30.03	38.20	34.76	Cut for polishing, T_c, J_c	63.90
Weight gained (%)	4.9	5.9	$-1.8^\dagger$	4.9	2.0		4.2
Samples' thickness (mm)	11.95	11.98	10.37	12.35	11.8		13.46
Trapped fields at 77 K (T)	0.64	0.73	$0.56^\dagger$	0.66	0.67		0.67

†Excess solidified liquid phase was removed from the bottom of this sample after recycling, which accounts for the weight loss and low trapped field.

(referred to as buffer pellets) of mass 0.3 g and diameter 5 mm of (RE)BCO (without Ag_2O) mixed powder were pressed uniaxially and placed on the top surface of each failed sample, as shown in Fig. 1(b), to aid the seeding process and to prevent contamination of the seeded pellets.[22–24] A conventional NdBCO seed was then placed at the center of the top surface of each buffer pellet.

The composition of the liquid phase pellet was Yb_2O_3: CuO: $BaCuO_2$ = 1: 6: 10 in molar ratio, corresponding to the composition reported for the infiltration and growth process.[21,25] A pressed pellet of this composition was placed directly beneath the failed sample, as shown in Fig. 1(b). An additional layer of Yb_2O_3 was included at the bottom of the arrangement to provide support for the liquid phase pellet and to prevent loss of shape during processing. The weight of the liquid phase pellet and Yb_2O_3 phases for failed samples of diameter 25 mm were 11.0 and 4.0 g, respectively. The diameter of the liquid phase pellet was 30 mm. The weight of the liquid phase pellet and Yb_2O_3 phases for failed samples of diameter 31 mm were 18.0 and 6.0 g. In this case, the diameter of the liquid phase pellet was 38.5 mm. The weight of the liquid phase pellet chosen in this study was approximately 25% of the weight of the original (failed) sample, corresponding to 44 and 75 g for the failed samples of diameter 25 and 31 mm, respectively. The liquid loss of a given (RE)BCO sample during melt growth lies usually within the range 2–15 wt% of the original weight of the sample prior to the growth process, although this varies from sample to sample. It has been our experience that the failed samples usually exhibit a higher weight loss than fully grown, single grains. As a result, the use of 25% of the weight of the original sample for the liquid phase content is believed to be adequate for the re-melt processing of failed samples. The weights of the failed samples before and after recycling are listed in Table I.

The sample arrangement shown in Fig. 1 was melt-processed in a box furnace. No purposely designed heating profiles were used for the recycling process, which, other than the sample composition and arrangement, was achieved using only

a standard TSMG processing technique.[2,3] Indeed, the samples for recycling were placed in the box furnace together with other, primary samples of the same size or larger (i.e., those were being melt processed for the first time). The heating profiles used for different sample batches were similar, but within a well-defined range. This involved heating the samples to 1055°C, holding for 1.0 h, cooling to 1015°C at a rate of 120°C/h, cooling slowly to 1008°C at between 0.8°C/h and 1.0°C/h, cooling to 985°C at between 0.4°C/h and 0.2°C/h and, finally, furnace cooling to room temperature. These samples were oxygenated subsequently at temperatures of between 450°C and 420°C for 10 d. Figures 2 and 3 show photographs of the top surfaces of fully grown, recycled samples.

Trapped magnetic field, which is the most important superconducting property for practical applications, was measured for each sample at 77 K using a rotating array of hall probes following field cooling (FC). One of the seven recycled samples was chosen randomly for cutting into subspecimens for microstructural examination. Chemical composition analysis of the cross section of this sample and of a fully grown, standard sample was performed using an energy dispersive X-ray spectroscopy analyzer (Hitachi S-3400, Hitachi High-Technologies, Singapore). Measurements were taken over an area of 40 μm × 40 μm, with the average composition, recorded in the same way as reported previously,[26] to allow for the presence of defects, which produce fluctuations in the local proportions of individual elements during analysis. These measurements began at a position approximately 1 mm under the position of the seed, and were then repeated at regular vertical intervals as the detector was moved through the thickness of the sample (i.e., along the *c*-direction of the sample).

III. Results and Discussion

(1) A Simple and Economical Method with High Yield
Seventeen failed YBCO bulk superconductors (one 31 mm in diameter and the others 25 mm in diameter) have been recycled, of which 16 were grown into single grains and one
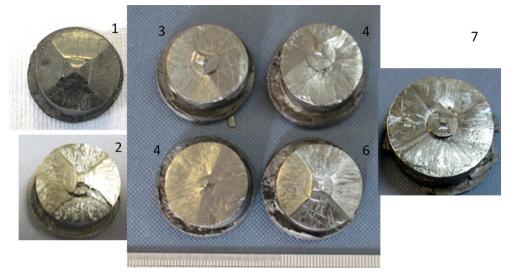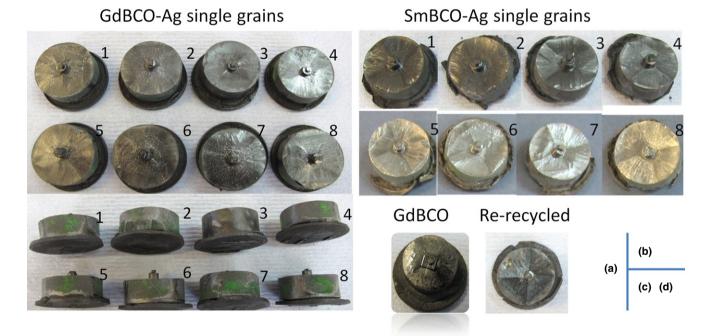
Fig. 2. Fully grown, recycled Y-Ba-Cu-O samples.

Fig. 3. Recycled (RE) -Ba-Cu-O-Ag (RE: Y, Gd, Sm) single grains (a) top and side views of one batch of 8 recycled Gd-Ba-Cu-O-Ag single grains 25 mm in diameter (b) top views of 8 Sm-Ba-Cu-O-Ag single grains 25 mm in diameter (c) top view of a recycled Gd-Ba-Cu-O single grain 20 mm in diameter (d) top view of re-recycled Y-Ba-Cu-O single grain 25 mm in diameter.

failed. Figure 2 shows seven such recycled YBCO single grains. Some of the features of the initially failed samples remain visible on the top surface after processing. However, these features disappear when the samples are polished to allow trapped field measurements, with any cracks visible on the top surface of the sample becoming fused during recycling. Fourfold, growth facet lines are clearly visible on the top surfaces of the samples and extend to the sides of the cylindrical shaped samples, which is an established indication of successful single grain growth and quality. These crystal-related growth features indicate that all seven samples constitute single grains after recycling.

In addition to YBCO, we have also successfully reprocessed other types of (RE)BCO-Ag systems (where RE = Gd and Sm). One GdBCO, 16 GdBCO-Ag, and 25 SmBCO-Ag failed samples have been recycled successfully by using the same technique applied to YBCO. The initial compositions of these failed samples on primary growth were 75 wt% Gd-123 + 25 wt% Gd-211 + 1 wt% BaO_2 + 0.1 wt% Pt for GdBCO, (75 wt% Gd-123 + 25 wt% Gd-211) + 1 wt% BaO_2 + 0.1–0.5 wt% Pt + 10–15 wt% AgO_2 for GdBCO-Ag and (75 wt% Sm-123 + 25 wt% Sm-211) + 2 wt% BaO_2 + 1 wt% CeO_2 + 10 wt% AgO_2 for SmBCO-Ag. Exactly the same composition for the liquid phase pellet was used in each case aside from that Yb_2O_3 was replaced by Y_2O_3 in the composition of the thin pellet containing liquid phase for GdBCO system. Seeding was supported via the use of a buffer pellet (0.3 g in weight and 5 mm in diameter[27]), to prevent Ag from diffusing into the generic seed.[28,29] The compositions of the two types of buffers used were 75 wt% Gd-123 + 25 wt% Gd-211 and 75 wt% Sm-123 + 25 wt% Sm-211 for GdBCO-Ag and SmBCO-Ag systems, respectively. The heating profiles of GdBCO, GdBCO-Ag, and SmBCO-Ag were adjusted according to their different

melting temperatures and growth rates relative to YBCO and were identical to those used in the primary growth process.[27,30] Figure 3 shows a selection of these recycled samples, all of which are single grains aside from sample 3 in the GdBCO-Ag batch, which did not grow fully to the edge of the sample. The formation of distinct facet lines in the side elevation of each sample is further indication of the success of the growth process, even though the upper surfaces retain some features of the originally failed sample. In total 64 recycled samples, 16 YBCO, 1 GdBCO, 16 GdBCO-Ag, and 25 SmBCO-Ag bulk superconductors have been grown in the form of single grains, corresponding to a success rate of over 90%.

SmBCO-Ag single grains are particularly challenging to grow by a primary TSMG process and, consequently, a batch process of single grains with good superconducting properties has not yet been realized. It should be noted that even failed, recycled samples can be re-recycled by using this method. Of 58 well-grown, recycled single grains, two samples were selected for re-recycling after failure following initial recycling by an original recycling method reported in.[20] Figure 3(d) shows one such re-recycled YBCO sample. The yield rate of this recycling process was higher than that observed for primary growth for both YBCO and SmBCO-Ag systems when primary growth and recycling processes were performed together in the same furnace using the same heating profile. This success rate in recycling indicates that the current method is very tolerant to a wide variety of failure mechanisms in the primary TSMG process and can be applied generally to potentially recycle any failed (RE)BCO bulk high-temperature superconductor. Understanding this recycling process would help further understand and improve the primary TSMG process for the different bulk compositions.

(2) Superconducting Properties

Superconducting properties of the recycled YBCO single grains were measured to examine the viability of the recycled single grains. The superconducting transition temperature T_c and critical current density J_c of small sections cut from the recycled single grains were similar to those observed for primary grown YBCO, for example, onset T_c 90 K with transition width less than 1 K and $J_c(0)$ about 5×10^4 A/cm^2, respectively. The trapped field, which is the most important property for practical applications of high-temperature superconducting single grains, for all six YBCO single grains in

Fig. 2 were measured. The peak trapped fields of the samples recycled here, which are listed in Table I, have an average value of 0.66 ± 0.07 T. This is 5%–10% lower than the value observed typically in conventional, primary grown samples using identical precursor powders from the same powder manufacturers. However, these values are good enough for many applications, including magnetic levitation where device performance is generally limited by the properties of a permanent magnet.

Figure 4 shows contour profiles of the trapped field of six recycled single grain GdBCO-Ag samples out of the total number of 16. This consists generally of roughly concentric circles, which provides further evidence that the samples have been recycled into single grains. The average trapped field for the recycled GdBCO-Ag single grains in this study is 0.60 ± 0.07 T. Based on the average value of trapped field for the batch processed, single grain GdBCO-Ag samples (25 mm in diameter and 12 mm in height) of 0.9 ± 0.05 T[30] observed to date in the Cambridge Bulk Superconductivity Group, we would estimate that the trapped fields of the recycled samples (25 mm in diameter and 9 mm in height) is 5%–15% lower compared to primary, single grain samples taking into account the fact that recycled samples are typically 3 mm thinner (although this approximation may be not accurate because there is no well-defined the relation between the trapped field and sample height).

The trapped field of 18 of 25 recycled SmBCO-Ag single grains was also measured as part of this study. These values fluctuated between 0.35 and 0.81 T for samples 25 mm in diameter and 8 mm in height. Clearly, the effects of Sm/Ba substitution in SmBCO system processed in air are severe,[31] which is a common, unsolved problem for the batch processed, primary growth of these materials, for which the reported trapped fields are generally low. 0.81 T is actually a record value for a SmBCO-Ag single grain of 25 mm in diameter and 8 mm in height, including all primary grown samples grown in air reported to date.

(3) Microstructural Differences Between Recycled and Primary Melt Processed Samples

Figures 5(a) and (b) show the microstructures of single grain samples grown successfully in a single process and recycled YBCO sample 6 at a position 5 mm away from the seed along the c-direction (i.e., through the sample thickness). It can be seen that the particle size of the Y-211 inclusions in the recycled sample has increased (the inclusions in the
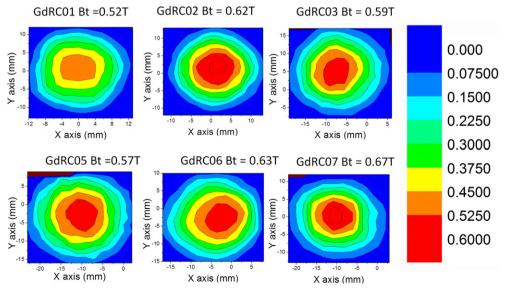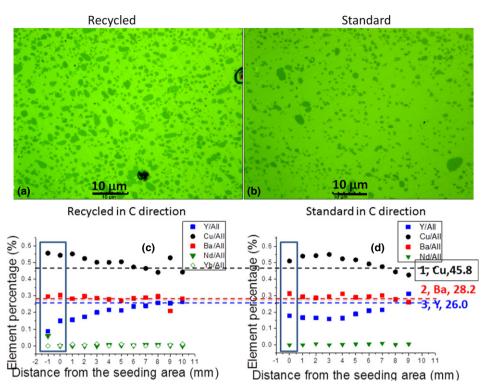

Fig. 4. Contour maps of the trapped field, B$_t$, of recycled Gd-Ba-Cu-O-Ag samples.

Fig. 5. Microstructures of recycled (a) and primary grown standard (b) Y-Ba-Cu-O single grains at a position 5 mm away from the seed along the *c*-axis. Variation in chemical composition with distance from the seed to the bottom of recycled sample (c) and primary grown standard sample (d) at intervals of 1 mm measured along the *c*-axis (dashed lines demonstrate the original composition in the mixed precursor powder, line 1, 45.8 for Cu; line 2, 28.2 for Ba; and line 3, 26 for Y. The vicinity of the seed is indicated by the rectangles).

Figs. 5(a) and (b) are Y-211 particles and the matrix is the Y-123 phase), as has been observed in recycled samples reported previously using other processing techniques.[17,20] This is understandable given that the Y-123 phase is re-melted during recycling to form new Y-211 phase, and that these new Y-211 particles can either nucleate directly from the decomposition of the Y-123 phase (i.e., to form completely new particles) or nucleate on the pre-existing Y-211 particles. The liquid phase reacts with both the new Y-211 particles (which are smaller in size) and the old Y-211 particles (larger in size) to form the Y-123 single phase when a grain re-grows during the recycling process. These growth processes result, inevitably, in the formation of larger residual Y-211 particles in the microstructure of the recycled single grains.

(4)   Reasons for the High Success Rate

In addition to the use of buffers to ensure the success of seeding in all systems investigated, the provision of an adequate quantity of liquid phase ($Ba_3Cu_5O_8$) through a thin pellet with appropriate composition for RE (Y, Gd and Sm) to diffuse to re-form RE(Y, Gd, and Sm)-123 lattice is the main reason for the high success rate of the crystal growth in the recycling process.

The crystal growth process involved in the fabrication of bulk (RE)BCO superconductors is unique compared to other crystal growth processes. The peritectic reaction that drives the process is shown below.

$$Y_2BaCuO_5(Y\text{-}211)$$
$$+ Ba_3Cu_5O_8(\text{liquid state}) \overset{1002°C}{\Longleftrightarrow} 2YBa_2Cu_3O_{7-\delta}(Y\text{-}123)$$
$$+ (\delta - 0.5)O_2 \; 0 < \delta < 0.5$$

This peritectic reaction occurs during the growth of the Y-123 crystal phase in air. The chemical reaction proceeding from left to right is a necessary condition for the Y-123 growth process. As a result, extra Y-211 phase is usually provided in the precursor powder to ensure that the chemical reaction occurs in this direction and to help retain the shape of a pressed pellet, as well as to improve the superconducting properties of the fully processed single grain. The yttrium flux required for the peritectic process is provided by the Y-211 phase particles during growth of the Y-123 phase, which reacts with the Ba-Cu-O liquid phase to form Y-123 and thus advance the crystal growth front. Liquid phase, therefore, carries the Y element through diffusion and provides the driving force for (RE)BCO single grain growth.[32,33] Adequate availability of the liquid phase provides a sufficient quantity of Y for the Y-123 crystal to grow easily. Conversely, the crystal growth will cease if the concentration of the liquid phase present falls below a critical level. This rather obvious observation has been generally neglected in previous research of the TSMG technique, and only a limited number of attempts have been made to engineer and control the liquid phase presence in developing the melt process.

The loss of liquid, which is usually caused by gravity during the single grain growth process, can be reduced by adjusting the heating profile[26] but cannot be avoided completely. However, liquid phase can migrate from the bottom to the top of the precursor sample arrangement in the infiltration process.[21,25] Indeed, we observed recently that liquid can infiltrate from a position distant to the seed to a position in the vicinity of the seed along both the *a* and *c* crystallographic directions to sustain crystal growth and nucleation from the position of the seed through energy-dispersive X-ray spectroscopy.[26] These observations imply that liquid phase travels to the regions where it is required to sustain the crystal growth process.

The extent of liquid loss is generally larger in failed samples (the weight loss for some failed samples can be as high as 20 wt%) than in fully grown, primary single grain samples. This study is based on the premise that the loss of liquid in failed samples can be compensated for by providing adequate liquid phase during the sample growth from the

bottom of the sample, in a similar way to the infiltration growth process. Hence, the composition of the pellet containing liquid phase chosen in this study was Yb_2O_3: CuO: $BaCuO_2$ = 1: 6: 10, which is similar to the liquid phase used in infiltration growth.[21,25] This composition can be also written as: Yb_2O_3: CuO: $BaCuO_2 \approx Yb_2O_3$: $Ba_3Cu_5O_8$ = 1: 3.33. Yb_2O_3 is used to help retain the shape of the liquid phase so that there is a reservoir of liquid available consistently during the single grain growth process, unaffected by gravitational leakage effects.[21] $Ba_3Cu_5O_8$ is the liquid phase in the peritectic reaction. As a result of this adequate supply of liquid, it is apparent from Table I that the failed samples in this research exhibit a weight loss of between 5 and 8 wt% during primary (failed) processing and regain between 2 and 5 wt% weight during recycling (note the weight gain is only approximate in this research, given that it is difficult to estimate the weight of the solidified liquid phase and obtain only the residual weight of the recycled sample). There is no observed correlation in this study between initial weight loss during primary melt processing and weight gain during recycling. It is understandable that the exact composition and microstructure of the failed samples are very different to one another and that each will absorb as much liquid phase as is required for growth into the Y-123 crystal phase within the processing time available. However, the amount of liquid phase provided is clearly sufficient for the recycling of each sample in this investigation and all the differently aligned multigrains in the failed primary samples corrected their orientation during secondary peritectic decomposition and solidification to form single grains. Significantly, an additional GdBCO, 18 GdBCO-Ag, and 28 SmBCO-Ag failed samples were recycled subsequently into single grains using this process (of which only two GdBCO-Ag and three SmBCO-Ag samples failed). This suggests that the provision of adequate liquid phase makes general (RE)BCO single grain growth easier and the success rate much higher. Yb_2O_3 presented in the liquid phase prevents heavy liquid ($Ba_3Cu_5O_8$) loss and crystallization from the bottom of the recycled samples due to YbBCO forming at a temperature lower than (RE)BCO(Ag) (RE = Y, Gd and Sm) but higher than the liquid ($Ba_3Cu_5O_8$) (i.e., at 900°C). This is a clear advantage over the recycling processes reported previously,[17,18,20] in which the associated composition of the replenishing liquid phase contains same RE element as that in the primary (RE)BCO system, which may cause the formation of subgrains during the TSMG recycling process.

To understand better the permeation of the liquid phase into the failed primary samples, chemical composition analyses of the cross section of recycled and fully-grown, standard samples were performed using an energy dispersive X-ray spectroscopy analyzer (s-3400), using the method reported previously (these measurements used the same samples prepared for optical microscopy).[26] It can be seen from Figs. 5(c) and (d) that the compositions of both the recycled sample and as-grown primary sample change continuously with position along the *c*-axis in the single grain, which is another indicator of the single grain nature of both samples (the measured composition would fluctuate with position in the sample if it were not a single grain). The exact composition at the bottom of the recycled sample becomes roughly constant (positions 7 and 8), which indicates that the liquid phase is indeed absorbed by the failed sample and that its composition has adjusted to that observed in conventional melt processing.

IV. Conclusions

We report a high yield recycling technique for melt processing failed, (RE)BCO-Ag bulk samples using an infiltration growth strategy without the need to first reprocess the failed samples into powder form. We believe that the technique relies on the provision of replacement liquid phase to the failed YBCO samples during a second, recycling TSMG melt process. The ytterbium in the thin pellet underneath the samples being recycled helps sustain the shape of liquid phase so that a reservoir of $Ba_3Cu_5O_8$ exists during the whole regrowth process of the failed samples. The superconducting properties of the recycled single grains are comparable to the single grains grown by a primary melt growth process, which enables the recycled (RE)BCO-Ag single grains to be used for applications. The overall recycling success rate of greater than 90% observed in this study and the low price of the replenishing liquid phase material compared to that of the primary rare-earth compounds, Pt and Ag in the initial precursor composition indicates clearly that the cost of production of bulk (RE)BCO samples can be reduced significantly. Given that the yield of the TSMG process can be fairly low, the technique we describe has the potential to have a transformative effect on the cost of production of bulk superconductors.

Supporting Information

Additional Supporting Information may be found in the online version of this article:

Data S1. Prices of the powders used in the primary growth and recycling process in the Cambridge, Bulk Superconductivity Laboratory, UK.

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