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On the influence of magnetic field processing on the texture, phase assemblage and properties of low aspect ratio Bi$_2$Sr$_2$CaCu$_2$O$_x$/AgMg wire

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Abstract

Bi$_2$Sr$_2$CaCu$_2$O$_x$/AgMg conductors are potentially important for many applications up to 20 K, including magnets for cryogen-free magnetic resonance imaging and high field nuclear magnetic resonance research. One promising approach to increased critical current density is partial-melt processing in the presence of a magnetic field which has been shown to enhance c-axis texturing of wide, thin tape conductors. Here, we report on low aspect ratio rectangular conductors processed in an 8 T magnetic field. The magnetic field is applied during different stages of the heat treatment process. The conductors are electrically characterized using four-point critical current measurements as a function of magnetic field and magnetic field orientation relative to the conductor. The superconductive transition and magnetization hysteresis are measured using a SQUID magnetometer. The microstructures are characterized using scanning electron microscopy and energy dispersive spectroscopy and analyzed using digital image processing. It is found that the presence of a magnetic field during split melt processing enhances the electrical transport and magnetic behavior, but that the anisotropy is not consistently affected. The magnetic field also affects development of interfilamentary Bi$_2$212 bridges, and that this depends on the initial shape of the Bi$_2$212 filament. At least two behaviors are identified; one impacts the oxide phase assemblage and the other impacts textured growth.

Keywords: magneto science, Bi$_2$212, texture, critical current density

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The development of cryogen free superconducting magnets has contributed significantly to an expanding role of magnetic fields in materials synthesis and processing and magnetic field has been recognized not only as a parameter for studying physical properties but also as a tool to control microstructure and properties [1, 2]. Owing to the magnetic anisotropy of certain crystal structures, grain alignment and crystal orientation by the application of a magnetic field has led to interesting phenomena. The experimental research has demonstrated that magnetic field can have a significant effect on the solidification [3–5], solid phase transformations [6, 7], grain growth and grain boundary
solidification in a thermal gradient \([Y-Ba-Cu-O]\) and \([Bi-Sr-Ca-Cu-O]\) superconductors, including several texturing techniques have been successfully used on is typically an important aspect of conductor processing. Strongly dependent on grain alignment and long-range texture is typically an important aspect of conductor processing. Recently, however, the focus for Bi2212 R&D has shifted to round wire and low aspect ratio (1–2) geometries [20, 21]. For reasons not yet fully understood, these conductors now show isotropic electromagnetic behavior and higher \(J_c\) than the wide tape conductors. From the perspective of magnet construction, they are preferable because they are likely to result in significantly higher fill-factor than wide, thin tape conductors [22].

Powder-in-tube manufacturing and partial-melt processing are typically used techniques for producing Bi2212 tapes and wires [23]. Powder-in-tube manufacturing is a batch process whereby the starting oxide powder is packed within a pure Ag tube and mechanically drawn into wire via traditional wire drawing technology. Typically, the outermost sheathing is a Ag-alloy, such as AgMg. Partial melt-processing consists of melting the material above its peritectic temperature, slow cooling to a temperature below \(T_{solidus}\), and subsequently sintering within the Bi2212 solid state. Note that because this heat treatment has a peak temperature above the peritectic temperature, the starting powder melts incongruently, forming several non-superconducting crystalline phases within a liquid phase, resulting in densification but also some phase segregation. When this is cooled properly, well-connected, well-aligned Bi2212 grains form from reactions between the liquid and the crystalline phases present in the partial-melt. During subsequent solid-state growth, Bi2212 grain alignment can result while the size of non-superconducting grains is reduced.

Due to the highly anisotropic crystal structure and electromagnetic behavior of HTS materials, the transport \(J_c\) is strongly dependent on grain alignment and long-range texture is typically an important aspect of conductor processing. Several texturing techniques have been successfully used on \([Y-Ba-Cu-O]\) and \([Bi-Sr-Ca-Cu-O]\) superconductors, including solidification in a thermal gradient [24–26], in a high magnetic field [18, 27–30], floating zone [31, 32], composite reaction texturing processes [33], and templated growth on a textured substrate [34–37]. Among the different techniques for producing grain alignment, processing in an external magnetic field is a demonstrated technique that also improves \(J_c\). HTS materials. Many efforts have been carried out for texture development in Bi2212 bulks and thick films or tapes by magnetic melt processing (MMP), during which the partial melt and solidification steps of the Bi2212 heat treatment are performed within a large magnet. It was demonstrated that a uniformly high degree of texture with the \(c\)-axis parallel to the direction of magnetic field applied during MMP is formed throughout the thickness of monocore tapes and bulk samples [28, 38]. Misalignment angles are reduced in multifilamentary Bi2212 tapes textured in a magnetic field [29]. The underlying mechanisms for obtaining improved texture and properties by magnetic field processing of Bi2212 remain uncertain. One possibility is related to the existence of anisotropy in the paramagnetic susceptibility. The source of such anisotropy, however, is unclear, particularly considering that the processing involves peritectic melting. It is also unknown if magnetic field processing affects Bi2212 processing through mechanisms other than texturing. For example, it is unknown if the presence of magnetic field alters the phase assemblage or the phase diagram.

Traditionally there are two approaches for manufacturing superconducting magnets. The react\&wind (R&W) approach is typically used for NbTi magnets while the wind\&react (W&R) approach was developed for Nb3Sn due to its intrinsic strain sensitivity. For Bi2212, both methods have been used [14, 16] but each has fundamental limitations. To improve upon Bi2212 magnet manufacturing, a new approach has recently been developed, react-wind-sinter (RWS), to avoid the primary issues that limit W&R and R&W manufacturing. For this manufacturing approach, a two-step process (‘split melt processing’) is used. Initial results show that this approach has significant benefits and further development is on-going [39–41]. One of the primary differences between SMP and conventional processing is that the Bi2212 undergoes two heat treatments. In the first, the Bi2212 is partially melted and solidified. It is then cooled to room temperature (at which point a magnet can be wound) and then reheated for subsequent solid-state sintering. This is illustrated in figure 1.

Here we report on investigations on split melt-processing of Bi2212/AgMg low aspect ratio multifilamentary wires using an 8 T magnetic field. Experiments are performed including the four permutations of magnetic field ‘on’ and ‘off’ during SMP as illustrated in figure 1. After heat treatment, the electromagnetic properties of the wires are characterized, including the dependence of the transport critical current \((I_c)\) on magnetic field and magnetic field angle, superconducting transition temperature, and magnetization hysteresis, in order to determine the effects of magnetic field during different stages of the heat treatment process. These measurements are used to quantify the anisotropy in terms of electrical and magnetic behavior. Microstructures were examined using scanning electron microscopy (SEM) and image analysis is used to begin understanding if magnetic fields alter the phase assemblage.
Figure 1. Split melt process used for the magnetic field processing experiments. The four processes, A, B, C and D refer to the various permutations of using magnetic field as illustrated.

Figure 2. Cross-section of the rectangular wire before heat treatment. The curves drawn on the image roughly separate the regions of round filaments from regions of aspect filaments. The scale bar shown equals 200 µm. The neighboring diagram illustrates the definition of the angle θ, which is the angle between wide plane and the magnetic field applied during measurements.

2. Experimental approach

Single restack powder-in-tube Bi2212/AgMg rectangular wire with an aspect ratio equal to two is provided by Supercon, Inc. [42]. The wire consists of 502 filaments with cross-sectional dimensions of 1.2 × 0.6 mm² and is shown in figure 2 in the as-drawn state (i.e. before heat treatment). Note that there are two distinct regions within the wire, separated by an hour-glass shape, as illustrated on the image in figure 2. In the outer portions, the filaments remain round and are ~25 µm in diameter, while in the center, top and bottom, the filaments are elongated and ~45 µm × 10 µm, which is a larger aspect ratio than that of the wire itself.

For each heat treatment run, 12 samples, each 45 mm long, are cut from one batch of wire, placed in a ceramic sample holder and introduced into a vertical furnace within a superconducting magnet. During all heat treatments, the magnet either provides an 8 T magnetic field parallel to the long axis of the furnace or it is off. The experimental setup was described in detail previously [29]. A version of the split melt process, consisting of a reaction step and a sintering step, is employed as illustrated in figure 1. Details of the temperatures, times and ramp rates, corresponding to the heat treatment in figure 1, are given in table 1. Also shown in figure 1 is the direction of magnetic field during heat treatment, which is not varied in these experiments. For the runs with the magnetic field on, the field is first increased to 8 T and the furnace temperature subsequently increased to the desired value. After heat treatment, the temperature is decreased to room temperature and only then is the magnetic field returned to zero. All heat treatments are performed in flowing oxygen. Note that, before processing, the furnace is profiled to obtain the temperature distribution within the furnace at the peak processing temperature. The sample holder is such that, for each run, the 12 samples result in six sets of two samples that have seen the same peak temperature. All 12 samples are measured and the pair with the highest performance for each magnetic-field process (A, B, C and D in figure 1) are studied in detail and reported here. This overcomes any potential run-to-run variation within the furnace that may otherwise influence the interpretation of the effects of magnetic field processing.

For all Ic measurements are at 4.2 K in a liquid helium bath using the four-point method and a 1 V cm⁻¹ electric field criterion. Transport measurements are made using a probe with a rotating sample holder to allow for measurement at varying field angles from 0 to 90° in magnetic fields up to 5 T. This is discussed in detail in [29, 30]. The definition of the magnetic field angle during measurement, θ, is illustrated in figure 2. Note that the applied magnetic field is perpendicular to the transport current for all magnetic field
Figure 3. $I_c(4.2 \text{ K})$ in self-field and at $5 \text{ T}$, $\theta = 0^\circ$, for the four sample types. Values are provided atop each bar in $\text{A min}^{-2}$.

Figure 4. Dependence of $I_c(4.2 \text{ K})$ on magnetic field angle at $5 \text{ T}$ for all four sample types. The angle dependence is seen by plotting the critical current normalized to its peak value.

The presence of magnetic field only during sintering (process D) has no impact on the transport behavior. Figure 4 shows the dependence of the normalized $I_c$ on the magnetic field angle for all four sample types. A strong dependence exists for all samples. With the application of magnetic field during split melt processing, however, the field angle dependence on $I_c$ is increased, especially for the wire with the magnetic field applied during both heat treatments (process B) as compared to those without magnetic field.

Figure 5 shows the average misalignment angle, $\varphi_{\text{avg}}$, of samples treated with and without magnetic field as calculated using (1) and following the approach detailed in [30, 43]:

$$\varphi_{\text{avg}} = \arctan \left( \frac{B_y}{B_x} \right). \quad (1)$$

In this formulation, which is derived in [30], $B_x$ and $B_y$ are defined such that $I_c(B_x, \theta = 0^\circ) = I_c(B_y, \theta = 90^\circ)$, where $\theta$ is the angle between the magnetic field and the plane of the tape as illustrated in figure 2. $\varphi_{\text{avg}}$ range from 24.3° to 27.2° for process A, 17.7° to 21.0° for process B, 19.8° to 24.4° for process C, and 20.0° to 26.0° for process D.
Figure 5. The average grain misalignment angle of samples of all four sample types.

Figure 6. Superconducting transition for all four sample types as measured by magnetization in a 100 G magnetic field in a SQUID magnetometer. The data on the left (a) is for $\theta = 0^\circ$ and that on the right (b) is for $\theta = 90^\circ$.

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Figure 7. Width of the magnetization hysteresis loop as a function of magnetic field for all four sample types (a) 4.2 K, $\theta = 0^\circ$; (b) 4.2 K, $\theta = 90^\circ$; (c) 10–40 K, $\theta = 0^\circ$; (d) 10–40 K, $\theta = 90^\circ$.

Figure 8. Width of the magnetization hysteresis loop as a function of temperature for $\theta = 0^\circ$. (a)–(d) Correspond to processes A–D, respectively.
difference, however, is significantly greater for \( \theta = 0^\circ \). These results indicate that the presence of magnetic field during processing impacts not only the crystallographic texture, but also grain-to-grain coupling and the volume fraction of Bi2212 phase formed within the oxide filaments [46]. There are also small differences between processes B, C and D, with process B having the sharpest transition and largest diamagnetic moment.

The magnetization hysteresis has been measured as a function of temperature for \( \theta = 0^\circ \) and \( 90^\circ \) and temperatures ranging from 4.2 to 40 K. Figures 7–9 show the width of the magnetization hysteresis loops, \( \Delta M \), as a function of magnetic field and temperature for all four sample types. Consistently seen in all cases, regardless of temperature, magnetic field or magnetic field angle, is that the samples processed in magnetic field have significantly higher \( \Delta M \) than the process A samples. Comparing the \( \Delta M \) data to the transport results shown in figure 3, it is particularly interesting to note that the transport enhancement from magnetic field processing was only 8.9–9.7%, whereas the enhancement in \( \Delta M \), which is proportional to the intragranular critical current density and the size of the current loop, increases by about 80% for process B and about 60% for processes C and D. The differences between processes B, C and D are relatively small, and in particular the differences between C and D, which only vary significantly for \( \theta = 0^\circ \).

To further understand the results, the electromagnetic anisotropy has been quantified based upon the electrical transport measurements and the magnetization hysteresis measurements. For the former, the anisotropy is defined as the ratio of \( I_c(\theta = 0^\circ) \)/\( I_c(\theta = 90^\circ) \), and for the latter it is the ratio of \( \Delta M(\theta = 0^\circ) \)/\( \Delta M(\theta = 90^\circ) \). Results for the four sample types are seen in figure 10. Interestingly, the anisotropy as determined by \( \Delta M \) is not affected by magnetic field, whereas the electrical transport anisotropy is increased primarily for process B and by a small amount for processes C and D (as could also be seen from figure 4).
3.3. Microstructure

Figures 11 and 12 show backscattered electron (BSE) images of polished cross-sections of samples of all four processing types and from both the edge (initially round filaments, figure 11) and the center (initially rectangular filaments, figure 12) sections of the wire. In all cases, the microstructure consists of Bi2212, Bi2201 and non-superconducting AEC phases, and significant bridging between filaments which is typical for high performance Bi2212 wires. It appears that the initial filament shape plays a role in how the magnetic field affects Bi2212 growth. In the edge sections of all four sample types (figure 11), where the filaments were initially round as found in Bi2212 round wires, the only significant difference between the four sample types is that the samples heat treated in a magnetic field show a reduction in porosity. This is particularly true for the process B and C images. There is no noticeable change in the Bi2212 texture and the growth direction for the Bi2212 bridges that connect the filaments is random. In the center Section (figure 12), however, in addition to the reduction in porosity, there is a noticeable change in the Bi2212 interfilamentary bridges. While the bridges in process A remain randomly oriented, those in the process B and C wires have grown predominantly in the direction perpendicular to the applied magnetic field, indicating that the magnetic field affects textured growth primarily during resolidification/nucleation. The eight images in figures 11 and 12 have been analyzed quantitatively with results shown in figure 13. Plotted is the area percentage of Bi2212 within the non-Ag areas of the wire. These results clearly indicate that the presence of magnetic field during resolidification/Bi2212 nucleation increases the Bi2212 area fraction, whereas magnetic field during solid state growth does not.

4. Discussion

The effects of magnetic field on the textured growth of Bi2212 wires have been studied in geometry where, due to the large number of small Bi2212 filaments, the effects of the Bi2212/Ag interface dominate. As a result, the field effects are small relative to past studies on Bi2212 bulk material that lacked Ag altogether, and on monocore tapes, where the Bi2212 volume is large relative to the Bi2212/Ag surface. Nonetheless, some clear and noteworthy effects are observed indicating that magnetic field processing does impact the microstructure and properties of Bi2212 wires. Electrical transport measurements show an improvement in performance when the magnetic field is present during peritectic melting and resolidification. Magnetization hysteresis measurements show an improvement in all samples which experience magnetic field during at least part of the process, with the largest improvement for samples with the magnetic
Figure 12. Backscattered electron (BSE) images taken within the SEM of filaments at the center of the wire. (a)–(d) Correspond to processes A–D, respectively.

Figure 13. Percentage of Bi2212 in non-Ag areas of processed wires as determined by digital image analysis of the SEM images shown in figures 11 and 12.

field present throughout the entire process. Changes in the electromagnetic anisotropy, however, are relatively small.

Magnetic field induced texture in Bi2212 superconductors results from the anisotropy in the paramagnetic susceptibility. Therefore, when the oxide is placed in a magnetic field while in the normal state, the magnetic energy is minimized when the axis of maximum susceptibility is parallel to the magnetic field. There are at least four drivers for grain alignment due to magnetic field: (i) rotation of paramagnetic crystals within a liquid phase, (ii) grain nucleation with a preferred orientation, (iii) selective grain growth where grains aligned with their $c$-axis parallel to the magnetic field preferentially grow at the expense of grains with their $c$-axis otherwise oriented, (iv) a reduction in quantity of other phases present, resulting in less obstacles to textured growth that is driven by other texturing mechanisms (e.g. the Ag interface). It remains difficult to fully assess which of these mechanisms dominate.

Ferreira et al [47, 48] proposed a model to explain magnetic field induced texture, which suggests that the enhancement in texture is primarily due to grain rotation during the early stages of crystal growth from the liquid. During the nucleation of crystals from the melt, when the crystals achieve a critical volume and are still surrounded by a large liquid volume fraction, the magnetic driving force is capable of introducing grain rotation. At the later stages of growth, however, the presence of nearby grains introduces grain-to-grain interactions that hinder magnetic field induced alignment. If this is correct, then in this investigation, due to the relatively small Bi2212 filaments ($\sim 25 \mu m)$, the effect of magnetic field on the Bi2212 may be limited to the nucleation and growth processes, leading to the effect of the magnetic field during processes B and C to be dominant. This is consistent with the transport results and with the
enhanced orientation of the Bi2212 bridges seen in the SEM images. Rotation of superconductor grains requires both the anisotropy of magnetic susceptibility and the coexistence of liquid which can be a lubricant for grain rotation, or at least be of low enough viscosity not to prevent rotation. While it is likely that some liquid phase is formed at the beginning of the second heat treatment during split melt processing, it is also known that multiple phases other than Bi2212 exist after the first heat treatment, including AECs, Cu-free phases and Bi2201. Thus, the mechanical interaction between superconductor grains and these second phases may obstruct Bi2212 grain rotation despite the presence of some liquid, inhibiting texturing.

The improvement in magnetization hysteresis, without any change in magnetically measured anisotropy, implies that the magnetic field during processing impacts more than just textured grain growth. Magnetic hysteresis is caused by intragranular shielding currents flowing in individual superconducting grains as well as intergranular currents flowing across grain boundaries. In the case of textured materials, the anisotropy of the properties represents an additional parameter to be considered for understanding the magnetization results. The absence of significant change in the magnetization anisotropy, with a clear increase in the bulk magnetization and an increased sharpness of the magnetic transition, is consistent with the hypothesis that the presence of magnetic field affects the phase assemblage such that a larger volume fraction of higher density Bi2212 results. This does not explain the improved texturing of the interfilamentary bridges, but the bridges represent a small fraction of the total amount of Bi2212 present and thus play a relatively small role in the magnetization behavior, even if they play a significant role in the transport behavior by interconnecting filaments. Thus, it appears that at least two mechanisms exist, one which increases texturing within filaments and of interfilamentary bridges, and another which improves the Bi2212 volume fraction and density.

It is important to recognize that the effects of magnetic field processing depend on the initial filament shape, with significant impact on the rectangular filaments and less impact on round filaments. This is understood in the context of the important role of the Ag/Bi2212 interface in Bi2212 nucleation and microstructural development. It is possible that bridges grow along certain crystallographic Ag planes which are energetically favorable and that the Ag texture may affect Bi2212 grain alignment [49]. Thus, the effect of the magnetic field on filament bridging may be a combined effect of the magnetic field on Ag, Bi2212 and the Ag/Bi2212 interface. This supports the hypothesis that the magnetic field affects the phase assemblage (supported by the image analysis) and that the modified phase assemblage (reduced non-superconducting phases) improves the oriented growth by not blocking grain growth. In this case the dominant texturing mechanism may be the Ag interface rather than the magnetic field itself. This requires further investigation. Although Liu et al have shown that the c-axis of Bi2212 grains grow parallel to the magnetic field regardless of the Bi2212/Ag interface orientation [50], this study was not performed on multifilamentary wires with small filaments, where the ratio of the interfacial area to the Bi2212 volume is large.

5. Summary and conclusions

The application of magnetic field during varying segments of split melt-processing in low aspect ratio Bi2212/AgMg multifilamentary rectangular wire resulted in enhanced electrical and magnetic behavior, small changes in the electrical anisotropy, no change in the magnetic anisotropy, and an altered microstructure, particularly in the regions of the wire with rectangular filaments. These results indicate that at least two mechanisms are at work. One mechanism is an altered phase assemblage that leads to higher volume fraction and higher density Bi2212 grains and is responsible in part for the improved magnetization hysteresis behavior. The second mechanism improves textured grain growth, particularly in the interfilamentary bridges that, despite being a relatively small fraction of the Bi2212 present, play an important role in electrical transport.

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