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in Melt Textured $YBa_2Cu_3O_x$** OSTI

By

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Abstract

Conventional polycrystalline HTS possess extremely low transport critical current density, which has been attributed to grain boundary related deficiencies. We have developed a processing method to eliminate secondary phases and to reduce the amount of microcracks at the grain boundaries in melt-textured YBCO superconductors. In this process, a fraction of the liquid phase is removed during melt-texturing which leads to cleaner grain boundaries. These boundaries have been found to carry high currents (3×10^4 A/cm² at self field and 1.5×10^4 A/cm² at 1.5 T) at 77 K in spite of the relatively high misorientation angles. Also, the weak dependence of J_c on magnetic field indicates that these grain boundaries are strongly coupled. In addition, the linear I-V characteristics of these samples suggest that flux flow along these boundaries may be the J_c limiting mechanism. These results give promise to manufacturing of polycrystalline YBCO superconductor with high current carrying capability.

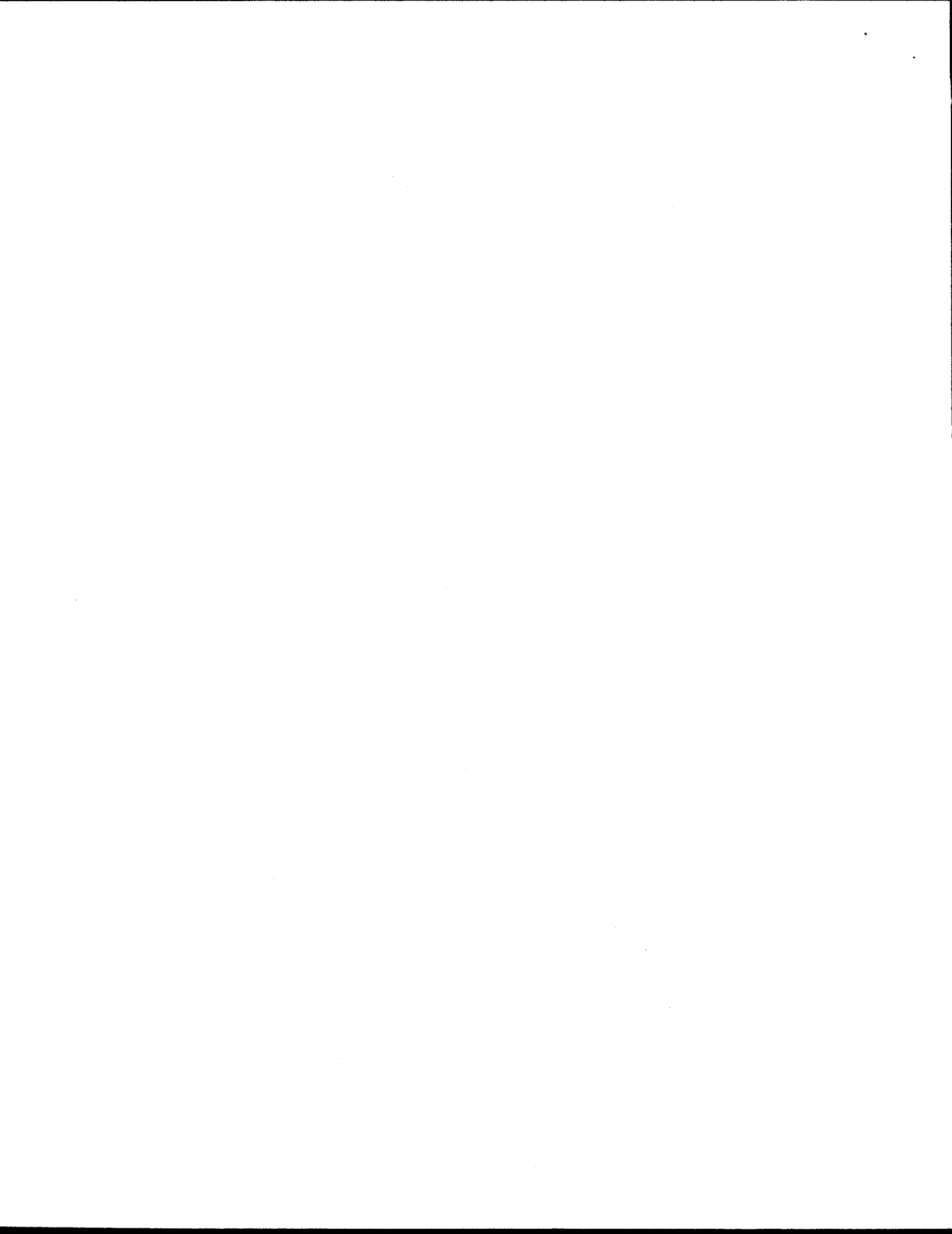
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PROCESSING AND PROPERTIES OF HIGH J_c GRAIN BOUNDARIES IN MELT TEXTURED $YBa_2Cu_3O_x$

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ABSTRACT

Conventional polycrystalline HTS possess extremely low transport critical current density, which has been attributed to grain boundary related deficiencies. We have developed a processing method to eliminate secondary phases and to reduce the amount of microcracks at the grain boundaries in melt-textured YBCO superconductors. In this process, a fraction of the liquid phase is removed during melt-texturing which leads to cleaner grain boundaries. These boundaries have been found to carry high currents (3×10^4 A/cm² at self field and 1.5×10^4 A/cm² at 1.5 T) at 77 K in spite of the relatively high misorientation angles. Also, the weak dependence of J_c on magnetic field indicates that these grain boundaries are strongly coupled. In addition, the linear I-V characteristics of these samples suggest that flux flow along these boundaries may be the J_c limiting mechanism. These results give promise to manufacturing of polycrystalline YBCO superconductor with high current carrying capability.

1. Introduction

Bulk polycrystalline superconductors contain large number of grain boundaries, inhomogeneities and other defects which lead to a poor current carrying capability. This is a major hurdle to many practical applications of these materials.¹ The fact that grain boundaries act as weak links stresses the need for developing new processing methods to manufacture superconductors capable of transporting high currents. Grain boundaries have been found to be weakly linked because of the presence of secondary phases, micro-cracks and atomic disorder which gets manifested in the form of misorientation at the grain boundary interface. While the first two problems can be overcome by processing methods to a large extent, the last one can be alleviated by optimizing the processing conditions such that the severity of the problem is considerably reduced by decreasing misorientation angles between grain boundaries.²

In a preliminary work carried out by Willén and Salama,² a fraction of the liquid phase formed during melt-texturing was removed and the grain boundary microstructure was improved. Following these encouraging results, further efforts were directed to test and characterize the grain boundaries in samples processed using this method.^{3,4} It was found that grain boundaries with misorientation angles up to 27° are capable of transporting large current densities ranging from 10 kA/cm² to 30 kA/cm² in self field and also in the presence of external magnetic field of 1.5 T. This indicates that the grain boundaries, in samples produced using the above method, are relatively clean and therefore are strongly coupled.

2. Processing

During melt texturing sintered YBCO undergoes a peritectic reaction and decomposes into 211 and liquid phase and is then slowly cooled to allow the formation of textured 123. The 211 is rich in Yttrium while the liquid phase is rich in Barium and Copper. Upon slow cooling the textured 123 consists of peritectic 211 which has not recombined and the excess liquid phase which is trapped at the grain boundaries. This excess liquid phase is usually composed of BaCuO_2 and CuO .⁵

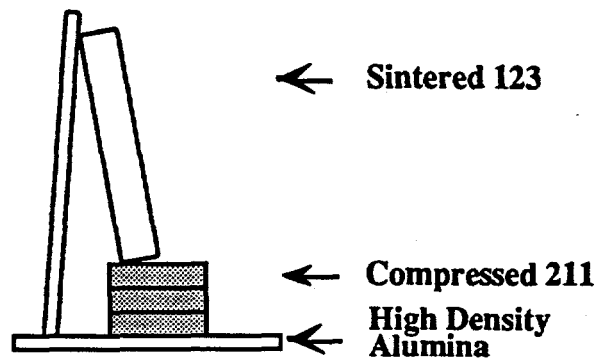


Figure 1. Assembly used to fabricate grain boundaries free of secondary phases in melt textured YBCO.

If a fraction of this liquid phase is removed, there will not be any excess liquid phase trapped at the grain boundaries and the 211 content in the melt textured 123 will increase. This increase in 211 content serves as a mechanical reinforcement between the 123 grains and platelets, and reduces the amount of micro-cracks in the bulk specimen. This improvement can be achieved by absorbing some liquid phase, using 211 as absorbant, during melt texturing. The arrangement used to manufacture 123 samples with high current carrying capability is shown in Figure 1. In this arrangement the 211 is compressed in the form of blocks and placed in contact with the 123 beam during melt texturing. Because of the porosity in the 211 blocks the liquid phase is absorbed easily with the high capillary forces. A removal of 40 % liquid phase is found to produce samples with grain boundaries free of secondary phases.² The samples fabricated using this approach consists of large domains with length extending over 15 to 20 mm, width from 8 to 10 mm and thickness of about 5 mm. Samples in the form of bars of lengths ranging from 50 mm to 100 mm and cross sectional area from $10 \times 5 \text{ mm}^2$ to $10 \times 10 \text{ mm}^2$ have been successfully processed.

Specimens for grain boundary characterization are cut from these samples after selecting the regions of interest by observing the microstructure. The final dimensions of the grain boundary specimens after cutting and polishing usually are $10\text{-}15 \text{ mm} \times 0.5 \text{ mm} \times 0.4 \text{ mm}$.

3. Characterization

Macroscopic studies of the samples processed revealed that these bars possessed several large domains. These domains are misoriented to each other and therefore the bars are polycrystalline in nature. The transport current carrying capability of these bars is tested

at 77 K in self field and in magnetic field by the four-point probe method. Current and voltage leads are applied using silver wires and silver colloidal liquid. The bars and the electrical contacts are annealed in flowing oxygen atmosphere at 500 °C for 2-6 days.

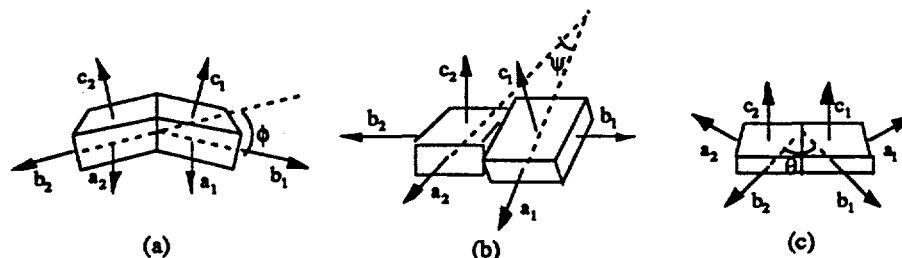


Figure 2. Description of misorientation angles at a grain boundary; (a) a-axis tilt, (b) b-axis twist and (c) c-axis tilt.

The misorientation between two adjacent grains where stacks of parallel plates meet edge to edge is determined by optical microscopy using a combination of three different misorientation angles shown in Figure 2.³ The misorientation between two grains is also characterized by X-ray pole figure analysis using two angles namely the kink angle γ between the c-axes of both the grains, and the angle β between the b-axis of the first grain and the projection of the b-axis of the second grain on the a-b plane of the first grain.^{3,6,7} The transport critical current density across the grain boundaries is measured in self field and also in the presence of magnetic field. The DC current is transported through the specimen and the voltage drop across the voltage leads, that are bonded on either side of the boundary, is measured.

4. Results and Discussion

4.1 Self field measurements of grain boundaries

Table 1 contains the critical current density in self field at 77 K for 13 specimens studied in the previous work. The misorientation angles ϕ , ψ and γ characterizing the boundary through which the current was passing varies from 0° to 47° for the a-axis tilt ϕ and 0° to 83° for the b-axis twist ψ and the kink angle γ . The grain boundary specimens with misorientation angles lower than 27.8° are found to transport large currents and have current densities up to 26,300 A/cm² while for those with higher misorientation angles the critical current density drops by approximately two orders of magnitude.

4.2 Self field measurements of bars

The current-voltage characteristics of the melt-textured bars of dimensions 50 mm \times 10 mm \times 5 mm were measured in self field at 77 K. The bars are found to carry currents up to 30-40 Amps before losing superconductivity. As the current is increased further, the voltage across the voltage leads increases. The reason for studying this dissipative regime was to observe how the resistivity changes with higher currents. These results are displayed in Figure 3. From Figure 3 it can be seen that the I-V behavior is ohmic. The voltage increases linearly as a function of the current. Using the Pulse DC current measurement technique, the bars were tested up to 1000 Amps.

Table 1 Misorientation angles of grain boundary specimens and the critical current density in self field at 77 K.

| Specimen | No. of grain boundaries | Misorientation Angles | | | J_c (A/cm ²) |
|----------|-------------------------|-----------------------|----------------------|-------------------------|----------------------------|
| | | a-tilt ϕ° | b-twist ψ° | kink ang γ° | |
| 1 | 0 | 0.0 | 0.0 | 0.0 | 47,600 |
| 2 | 1 | 4.5 | 4.5 | 6.4 | 11,100 |
| 3 | 2 | 3.4 | 9.1 | 9.7 | 26,300 |
| | | 11.0 | 3.2 | 11.5 | |
| 4 | 2 | 7.2 | 7.1 | 10.1 | 22,600 |
| | | 9.3 | 4.4 | 10.3 | |
| 5 | 1 | 3.1 | 10.2 | 10.7 | 10,300 |
| 6 | 2 | 4.2 | 9.5 | 10.4 | 7,600 |
| | | 15.8 | 4.2 | 16.3 | |
| 7 | 1 | 16.2 | 2.5 | 16.4 | 11,300 |
| 8 | 1 | 23.0 | 16.0 | 27.8 | 19,300 |
| 9 | 1 | 30.4 | 32.1 | 43.0 | 115 |
| 10 | 1 | 40.0 | 20.0 | 44.0 | 216 |
| 11 | 1 | 9.7 | 45.0 | 45.8 | 304 |
| 12 | 1 | 47.0 | 18.4 | 49.7 | 605 |
| 13 | 1 | 42.3 | 81.5 | 83.7 | 213 |

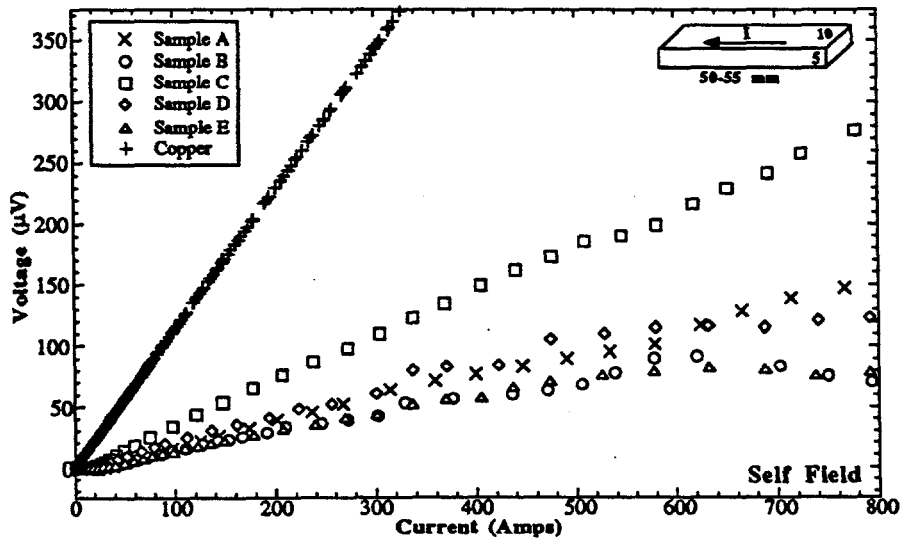


Figure 3 Self field measurements of bulk polycrystalline melt textured bars at 77 K.

The decrease in voltage at higher currents is due to the inductive voltage which is negative in nature. In Figure 3, the voltage across a copper bar of dimension 50 mm × 10 mm × 5 mm, of grade B187, at 77 K is also plotted. The resistance of this bar at 77 K for these dimensions is 1.14 $\mu\Omega$.⁸ This shows that these superconducting bars have a lower resistance than that of an industrial grade electrical copper conductor. Therefore it is possible to reach currents much larger than the critical value and still maintain a resistivity that is very low compared with that of copper at the same temperature. Some of these bars have resistance as low as 0.15 $\mu\Omega$. Hence the heat losses can be reduced by as much as 1/7th of that of copper. Also since the thermal conductivity of YBCO is 1/50th that of copper, these bars are very appealing as current leads and bus bars.

The low critical current density of these bars can be attributed to the high angle grain boundaries between the grains of these polycrystalline samples. The low resistance, however, demonstrates the low resistivity of the boundaries. Many applications like current leads and interconnects can benefit from these bars due to their low electrical resistance in addition to their low thermal conductivity.

4.3 Magnetic field measurements of grain boundaries

In order to study the effect of magnetic field on the current passing through a grain boundary, measurements were made with the current flowing along the length of the specimen and the magnetic field applied in the plane perpendicular to the current direction. The critical current density is then measured when the field is applied at various orientations. The critical current density is found to behave anisotropically and has maximum and minimum values corresponding to certain orientations of the magnetic field.³

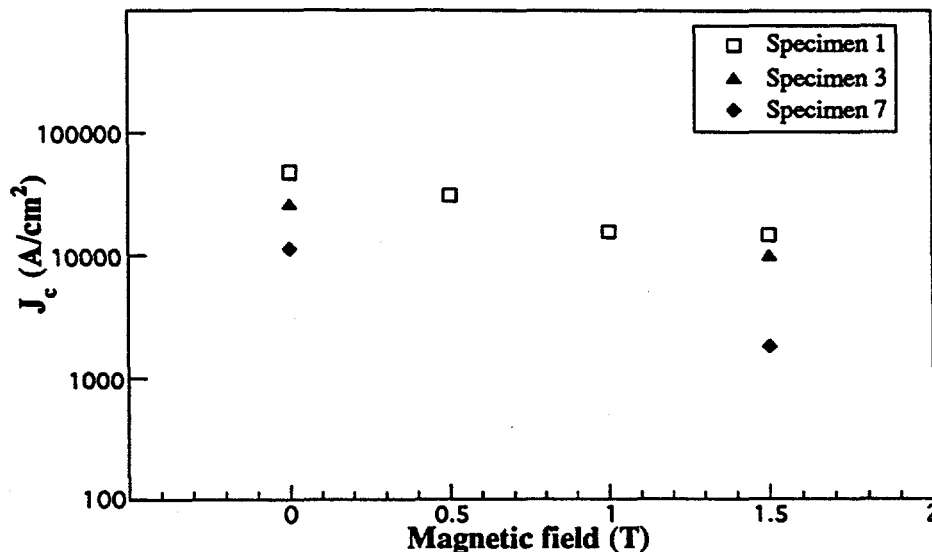


Figure 4 Maximum critical current density observed during magnetic field measurements of grain boundaries at 77 K.

The dependence of maximum J_c on magnetic field applied perpendicular to specimen axis for specimens Nos. 1, 3 and 7 is shown in Figure 4. The drop in the value of J_c is about 66 % which is similar to that usually observed in melt-textured single grain

samples.⁷ Specimen 3 ($\gamma = 9.7^\circ$) shows a similar drop of only 62 % at 1.5 T from its zero field value even with the presence of two grain boundaries. Specimen 7 ($\gamma = 16.4^\circ$) shows a drop of almost 80 % but still has a J_c of 2,000 A/cm² at 1.5 T which is one order of magnitude higher than J_c in polycrystalline sintered samples. These results indicate that the grain boundaries in these specimens are strongly coupled and are not easily penetrated by magnetic flux even in high magnetic fields. The dependence of minimum J_c on magnetic field applied perpendicular to the specimen axis in these three specimens is shown in Figure 5. The drops in the J_c values are comparable or even higher than that observed in single grain specimens and again indicates that these grain boundaries are strongly coupled and are capable of carrying relatively high currents in the presence of large external magnetic fields.

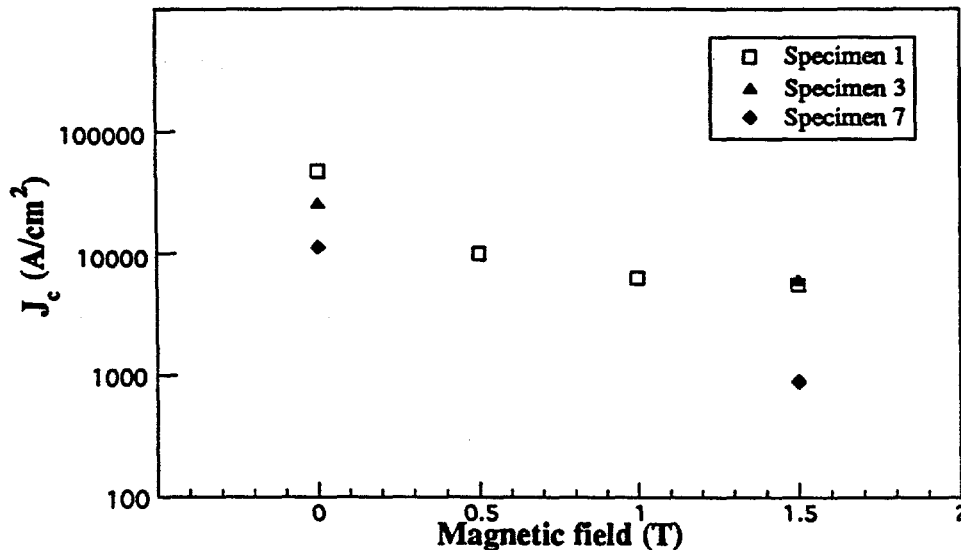


Figure 5 Minimum critical current density observed during magnetic field measurements of grain boundaries at 77 K.

4.4 Magnetic field measurements of bars

In the presence of magnetic field, the bars exhibit an ohmic behavior very similar to the self field. Due to the magnetic field, the critical current density across the bars decreases. However as the magnetic field increases, no substantial change is observed in the I-V behavior (see Figure 6). This indicates that the I-V behavior is insensitive to the increasing magnetic field and the bars behave like a normal electrical conductor. The increase in resistance of these bars in the presence of magnetic field is apparent because of the poor quality of current contacts. As the current is transported continuously, heat is generated at the contacts between the current leads and the superconductor bars. This heat generation gives rise to additional resistance. This is absent in the case of Pulse DC measurements. Improving the quality of current contacts will decrease this apparent resistance and the I-V behavior in magnetic field should follow the I-V behavior in self field as it does for sample B. For this sample, the I-V behavior at 0.5 T, 1.0 T and 1.5 T is almost similar and the resistance is equal to that measured in self field using the Pulse DC

current measurement technique. These observations give promise to the use of these bars for carrying high currents with minimal heat loss in the presence of large magnetic fields.

Since the single grain specimens possess power-law I-V characteristics, it can be assumed that the linear I-V characteristics observed in these samples represent the dissipation mechanism in the boundary rather than in the grains themselves. This suggests that the mobility of the flux lines is higher along these grain boundaries due to lower pinning forces in these boundaries.⁹

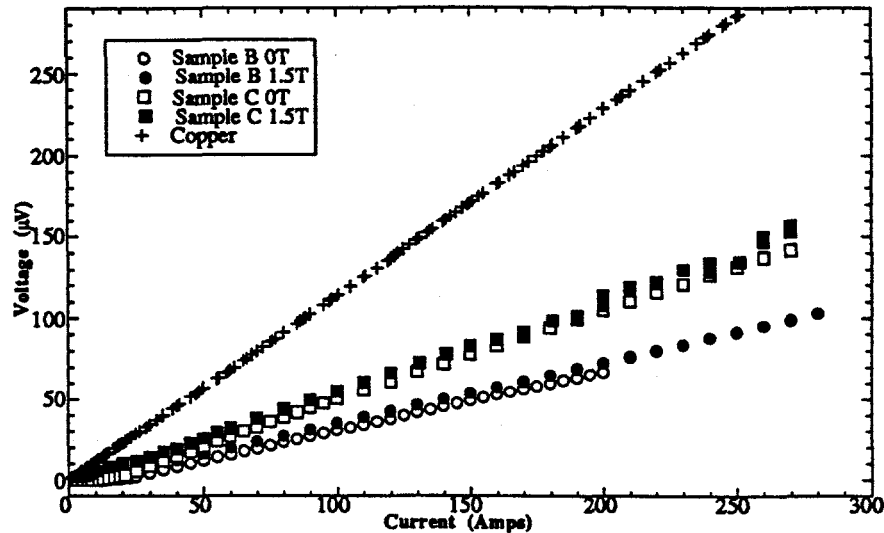


Figure 6 Magnetic field measurements of bulk polycrystalline melt textured bars at 77 K.

The reason for the decreased pinning energy along the boundary can be understood by considering that the pinning energy arises from fluxoids which typically penetrate the superconductor where the superconductivity is weakened. The creation of a normal vortex inside a superconductor requires additional energy which is equal to the difference between the energy of the normal state and the energy of the superconducting state. This difference in energy is smaller if the superconducting region is weakly superconducting. When a fluxoid is to be moved, the activation energy will then correspond to the energy that must be added to the system in order to move the normal vortex into the strongly superconducting region. If the boundary consists of a two-dimensional region with weaker superconductivity than the adjacent grains, there will be no strongly superconducting barriers between the pinning centers along the boundary. Consequently, the activation energy necessary for flux flow to occur is reduced along the boundary.

5. Summary

Melt-textured samples are processed by a liquid phase removal method to minimize the amount of secondary phases in the grain boundaries and also reduce the micro-cracks present at the interface. The critical current density is found to decrease very sharply, almost by two orders of magnitude where the misorientation angles exceed above 27.8° . Specimens capable of carrying high currents in self field at 77 K, are also found to carry high currents in 1.5 T at 77 K. Bulk polycrystalline samples 50-100 mm long and 50-100 mm² cross-section are found to transport currents up to 1000 Amps in self field and

magnetic field of 1.5 T with ohmic resistance much less than that of copper at 77 K. These results give promise to using the method of liquid phase removal for processing bulk YBCO superconductors capable of carrying high currents.

6. Acknowledgements

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