Published in: Zeitschrift Fur Physik B-Condensed Matter (1996), vol.100, iss.4, pp.551-555
Status: Postprint (Author's version)

Field percolation and high current density in 80/20 $DyBa_2Cu_3O_{7-x}$/Dy_2BaCuO_5 bulk magnetically textured composite ceramics

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Abstract: We measured the AC susceptibility of magnetically textured (123) 80%/211(20%) DyBaCuO composite in a special set-up in order to enhance the intergrain contribution. The synthesis process led to very clean "weak links" at grain boundaries. At the percolation threshold bulk shielding paths were such that the intergrain critical current density J_C was above 10^5 A/cm^2. The field dependence of J_C was understood through an analytical form indicating a distribution of currents similar to the law of clusters at fracture/percolation thresholds.

PACS: 74.72.Bk; 74.60Jg; 74.60.Ge; 74.80.Bj

I. Introduction

Liquid phase processing methods performed on YBaCuO bulk materials at sufficiently high temperature, i.e. above the peritectic decomposition of the 123 phase, lead to an increase of intragrain J_C. This is due to Y_2BaCuO_5 (211) precipitates within the 123 superconducting grains [1]. It is also known that the addition of the so-called 211 phase in excess in Y-123 oxide in the same synthesis condition improves further the critical current density J_C, but also enhances the oxygen diffusion in the 123 matrix [2]. Moreover, due to the fact that the peritectic reaction does not go to completion, there is an excess liquid phase ($BaCuO_2$ + CuO) in the system in such a way that 211 added to this system offers the possibility to consume this excess liquid (i) by forming additional YBaCuO phase, but more importantly, (ii) by avoiding the presence of liquid phase at the grain boundaries, whence "cleaning" them very much.

Recent works published on 123-211 composite systems show that their physical properties strongly depend on their microstructure [3]. Differences in preparation conditions lead to the formation of specific defects that complicate the understanding of the composite physical properties. Sandiumenge et al. discussed such a problem from detailed microstructural investigations on 123-211 composites [4]. These authors observed that up to 20 vol% in excess of 211 phase leads to a polygonalization dislocation-type process of 123 grains creating low angle grain boundaries in the 123 matrix. Studies of the inter-granular regions of polycrystalline samples are thus of importance in order to understand their electrical as well as their magnetic properties. Also examination of the 211-123 interfaces could explain the strong magnetic field dependence of the intragrain critical current densities J_{Cg}. Other studies have probed the role of superconducting layers or interfaces like in Bi-2223-Ag sheated tapes, [5] even in 2212 Bi-based single crystal [6] or in Hg compounds. [7-8] We recently studied a 2223-2212 mixture prepared by the glass route. [9] Thus it is of interest to further probe the intergranular coupling of such 123-211 composites. We do so here for 123-211 magnetically textured composite materials through electrical and magnetic susceptibility measurements as a function of magnetic field.

The behavior of the magnetic field dependence of the real and imaginary parts of the susceptibility for granular ceramics is also well-known [10] : first, during the temperature lowering, the real part χ' of the susceptibility undergoes a tread which occurs at T_{Con} corresponding to the superconducting transition of the individual grains. At a lower temperature (T_{Coff}) the percolation transition is marked by a second step, associated with zero resistance when the optimum phase coupling arises between the superconducting grains. It will be seen that the technique which was used in the present work, and in [9], provides informations strictly on the intergrain coupling component of the susceptibility, thus on the percolation transition only.

In practice, an AC magnetic field was applied parallel to the a-b planes of magnetically bulk textured composite sintered through a process similar to that described in [11]. The shielding currents are thus created in the a-c and/or b-c planes of 123 superconducting grains. Due to the low critical current density expected along the c-axis, an *interlayer shielding path* cannot be established near T_{Con}. This geometry thus avoids to establish a perfect diamagnetic state in the 123 superconducting grains above T_{Coff}. At the percolation threshold however, the induced currents are able to establish a *bulk shielding path* through the sample by intergrain phase coupling. We emphasize that the field geometry used for this system, and in [9] for 2223-2212 mixture, allows us to study one and only one isolated transition, i.e. that at T_{Coff}.

It will be shown that the critical current density associated to that intergrain decoupling is as high as 10^5 A/cm^2 in zero field at 40 K, a large value specific to the presently examined textured 123-211 composite materials with clean grain boundaries.

In the Dy-123 systems without 211 excess particles, the real part of the susceptibility usually presents two plateaus, suggesting that complete shielding process occurs in two steps. These steps result from various types of inter-grain coupling or weak links. The critical current densities associated to those weak links are then not higher than 10^4 A/cm^2 in absence of magnetic field.

II. Experimental details

Background and synthesis procedure

Unoriented polycrystalline materials are characterized by very low J_C. The existence of grain boundary current barriers are responsible for these low J_C values. It is thus necessary to texture the material in order to favor high-J_C properties. Three basic techniques are of interest: (i) creep sintering, (ii) melt texture growth, and (iii) magnetic field orientation [12]. The first is quite rapid but it produces quite compact samples which can be hard to reoxygenate. The second one, and its variants, are known to be the most effective at this time whereas the third is successful for aligning ab planes of grains dispersed in epoxy resin.

Consequently, we combined the latter two techniques [11] to prepare materials having high quality strongly coupled superconducting grains. To reach such a goal we sintered a 80/20% weight Dy-123/211 composite in the presence of its own liquid phase in a 0.6 T magnetic induction. We chose to sinter Dy-123 because this compound is magnetically anisotropic and has a susceptibility larger than that of Y-123.

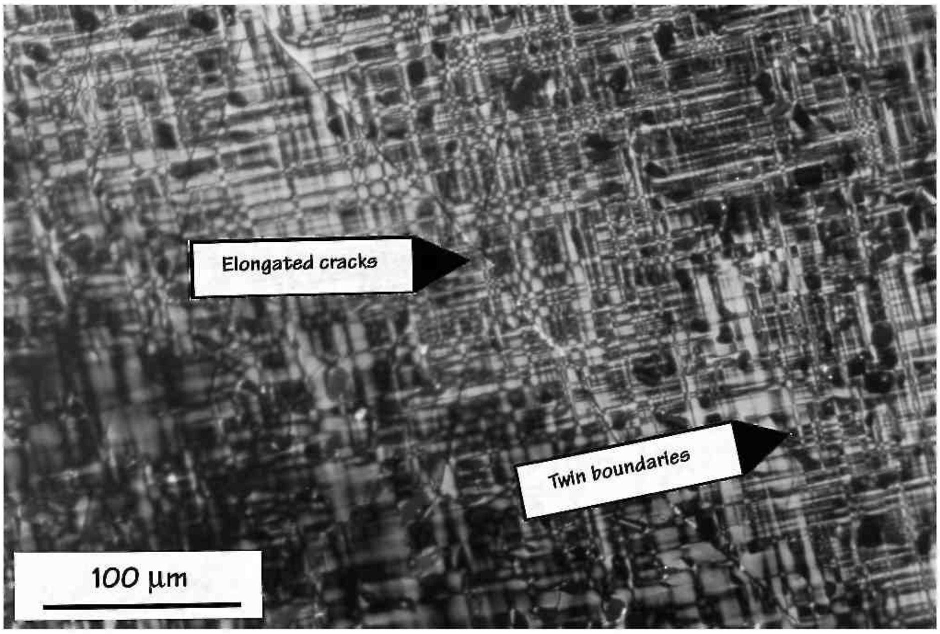
The synthesis of 123 and 211 powders started respectively from a corresponding stoichiometric mixing of Dy_2O_3, $BaCO_3$, and $CuCO_3 \cdot Cu(OH)_2$ pretreated at 920 °C for 48 h, including two intermediate grindings. The quality of the powder was checked by X-ray diffraction analysis. The Dy-123 and 211 powders were mixed together in the appropriate ratio, compacted into a strip (the sample dimensions were ca. 15 × 10 x 3 mm), and transfered into an alumina crucible which was vertically inserted in a specially built furnace placed between the polar heads of a magnet. A magnetic induction of 0.6 T which was generated horizontally was applied perpendicularly to the largest face of the sample during the whole process. The thermal cycle started at room temperature with heating to 1035°C at a rate of 150°C/h. A slow decrease of 2°C/h over several hours to 980°C is followed by a cooling process at 50°C/h to room temperature under oxygen atmosphere.

Microstructural characterization

Microstructure analysis was performed with a HITACHI S2500 scanning electron microscope with a 15 kV accelerating voltage.

Optical polarized light microscopy was also used in order to emphasize both (i) the 211 particle distribution and (ii) the oxygenation stage by looking at the twin patterns. Figure 1 shows the typical microstructure observed for such a magnetically oriented Dy-123/211 textured composite. Large twinned grains of 1 mm^2 size can be seen. A large number of cracks are also seen through the sample. It is remarkable that very clean grain boundaries exist as compared to Dy-123 systems without 211 excess particles. X-ray diffraction analysis showed that only (001) peaks were recorded through, and the c-axis of most grains was thus perpendicular to the largest face of the sample, i.e. parallel to the applied field during the synthesis.

Fig. 1: *Optical polarized light micrograph of 80/20% 123/211 composite material*



Electrical and magnetic investigations of the intragranular medium

Resistivity measurements were performed using an automated sequence and acquisition procedure. The sample was in a helium exchange gas cryostat. The classical four-point resistivity measurement method was used. Magnetic susceptibility measurements were made in a home-made susceptometer based on a CTI-21 model cryocooler. An AC field (h_{ac} = 0.1, 5.0, and 31G respectively; f = 271 Hz) was applied parallel to the sample long axis, i.e. the field was perpendicular to the c-axis of magnetically textured Dy-123 grains in such a way that the shielding currents were located in the a-c and/or b-c planes at the surface of most of the Dy-123 grains. A DC field was superimposed perpendicular to the sample long axis, i.e. parallel to the c-axis of magnetically textured Dy-123 grains. The field cooling method from room temperature was used. The critical currents were deduced by the AC Campbell method which is based on the flux profile acquisition. [13-16] By measuring the AC flux entering the sample, it was possible to obtain the field flux profile as a function of the AC field amplitude assuming the Bean model from which intragrain and/or intergrain currents can be deduced. [13,14] This can be done following some generalization towards granular materials, beyond the original homogeneity hypothesis.

III. Results and discussion

The temperature dependence of the AC susceptibility (χ' - $i\chi''$) in zero and finite DC magnetic field is shown in Fig. 2. The resistivity measurement in zero field as a function of temperature is shown above the χ' and χ'' data in order to emphasize the critical temperatures and their role. *Only one smooth step* in χ' and one maximum in χ'' are visible for the zero DC field case. *They both appear at the temperature for which there is zero resistivity.* This is markedly the intergrain coupling transition which is only determined by the quality of the intergrain coupling when a bulk shielding path and a percolation path are both established through out the sample. This T_{Coff} temperature (79 K) is relatively low as compared to classical 123 systems. This is probably related to the deficient oxygen concentration in the studied region where non-uniform 211 particle distribution occurs in 123-grains as e.g. shown in Fig. 3. In the latter figure, a lack of 211 particles at grain surfaces is indeed clearly observed. The relative lack of oxygen is however uniform since the resistive transition is rather narrow.

Moreover, one should again emphasize that there is no intrinsic susceptibility peak in χ'' for this material at T_{Con}, peak which should coincide with the initial drop in resistivity. This absence of peak can be explained by the impossibility to maintain a perfect diamagnetic state within each Dy-123 grain. Indeed, when the AC field is applied parallel to the sample axis and thus is parallel to the a-b planes, the shielding currents must have a component along the c-axis of the Dy-123 grains. Due to the low critical current density value generally expected along the c-axis at 77 K, a bulk shielding path can then be established only by coupling with neighboring grains. This cannot occur at "high temperature" in the mixed state because of the lack of coherence of the wave function between neighboring grains. However, at lower temperature, when the wave function phase

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coherence occurs, a complete shielding path can exist, whence the sharp transition in χ' occurs T_{Coff}.

We should also emphasize that the field range is very relevant for such considerations. At high and moderate fields one can easily understand that grains can be decoupled. For such a field range the decoupling is markedly depending on the texture, and is quite anisotropic. For small fields however, the shielding supercurrents are not so anisotropy limited. The field they create ensures that the local magnetic field vanishes except in regions of the size of the penetration depth. When the thickness of the grains along the c-direction is of the order of the penetration depth in that direction, grains should be decoupled [17]. Therefore such an alternative explanation has to be considered, and might be an additional cause of the lack of peak in χ'' at high temperature. Our estimate is that the penetration depth along the c-direction ranges between 150 and 180 nm in the temperature and field range investigated here. The size of the grains in the c-direction is somewhat of that order of magnitude in such polycrystal-line samples.

Fig. 2: *Resistivity and ac susceptibility measurements (h_{ac} = 0.1, 5.0, and 31G respectively; f = 271 Hz) as a function of temperature for 80/20% 123/211 composite material. Insert: zoom of the ac susceptibility transition for hac = 0.1 G*

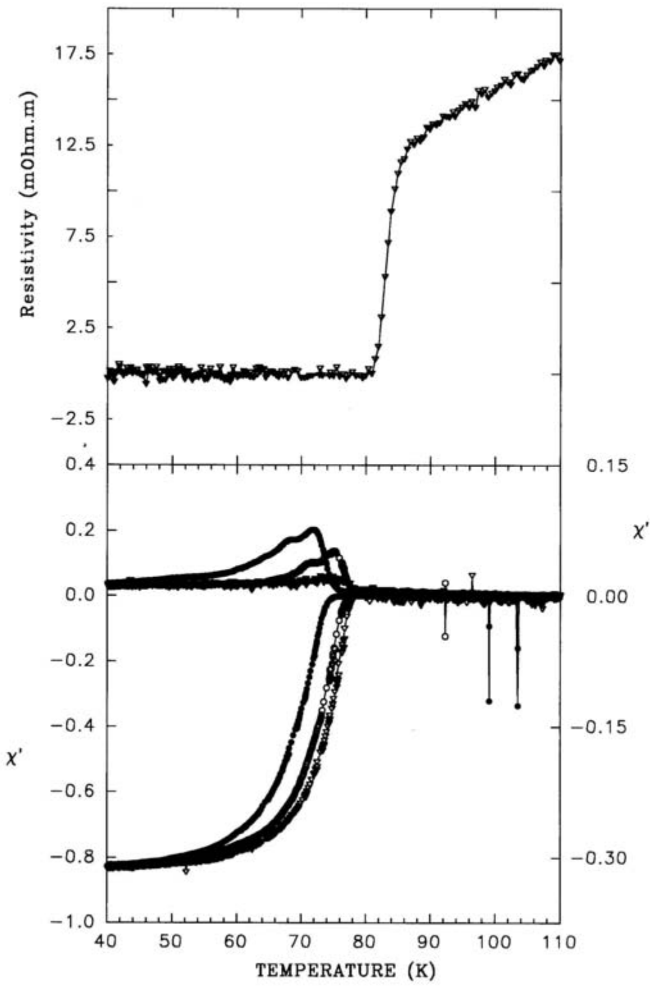


Fig. 3: *Optical polarized light micrograph of 80/20% 123/211 composite material showing inhomogeneous distribution of 211 particles inside 123 grains*

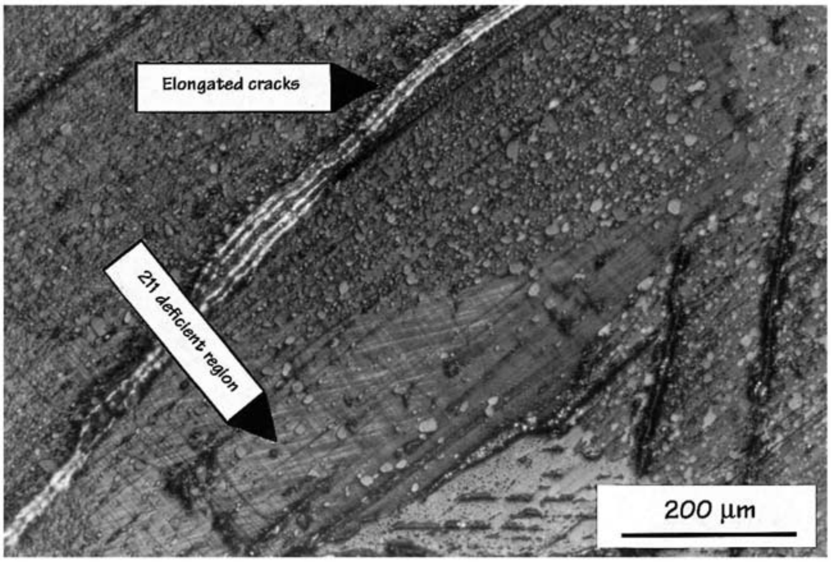
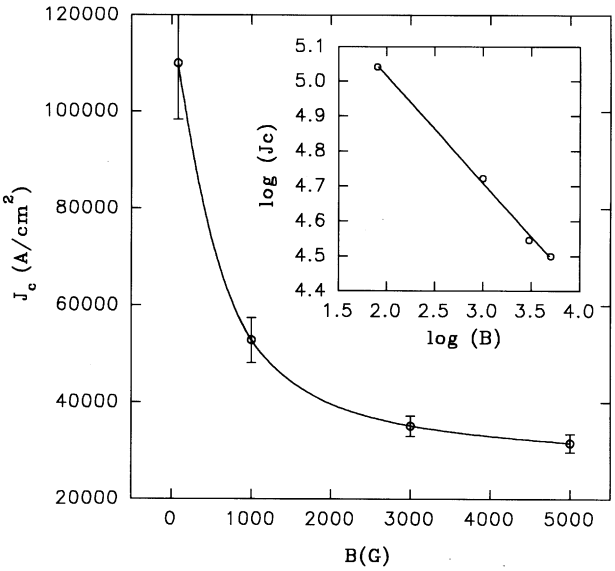


Fig. 4: *J_c as a function of the DC applied magnetic field at 40 K for a 80/20% 123/211 composite material. Insert: power law behavior of Jc(B)*



One should also note that there exist two closely spaced maxima in χ'' when the DC magnetic field increases (Fig. 3). This can be due to experimental error, but more probably understood in terms of oxygen in-homogeneity distribution in grains [18-20], or also resulting from the existence of different current shielding paths due to various grain angle boundaries.

The critical current densities as deduced from the AC Campbell method, in granular materials, [13] based on the flux profile acquisition technique are shown in Fig. 4. The variation of the deduced intergrain critical current density at 40 K as a function of the DC applied magnetic field is reported. For example, J_C at 0.1 T is quite high and equal to 5×10^4 A/cm^2. These results are in good agreement with the reported values of Sandiumenge et al. [4]

The numerical data when analyzed show a power law behavior (Insert in Fig. 4). We propose to understand the data with the following equation:

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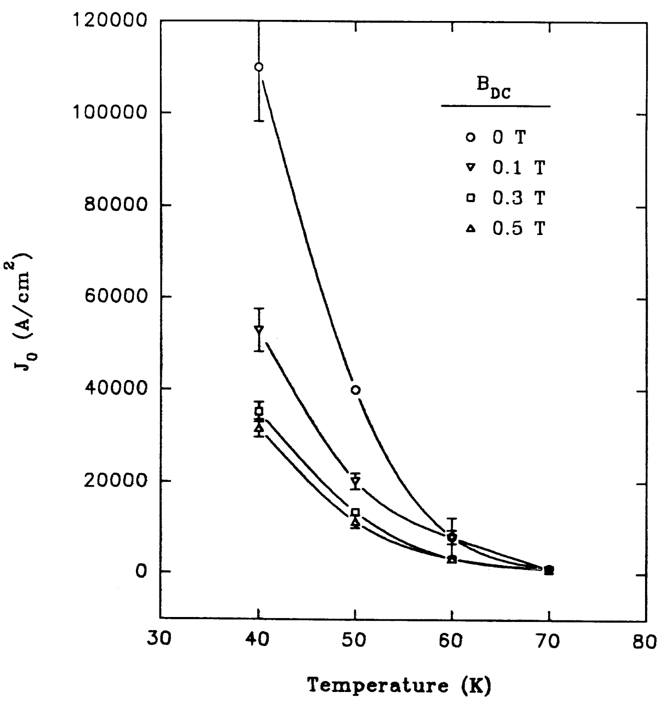
$Jc = J_{C0} \cdot B^{-q}$

with $q = 0.3$ and $J_{C0} = 4.10^5$ A/cm^2. This is far away from the record high critical current value (10^6 A/cm^2 at 77 K) in neutron irradiated YBCO single crystals, [21] but much above that of epitaxial thin films [22], of grain aligned YBCO, [23] and other zone melt YBCO with also some 211 excess particles [24].

There are several theories of the $J_C(B)$ dependence [25]. A single power law ($q = 1$) dependence is predicted in the Kim model [26] for intragrain dependence, while a q = 1/2 value is predicted for interfacial pinning. [27] Sometimes a (1 - B/B*(T)) factor is used for better fit [28] in the single vortex pinning regime (up to 10 kG). The q = 0.3 value indicates to us that the single vortex (intragrain or interfacial) pinning regime is a limiting case, in particular when an excess of 211 inclusions is introduced. In so doing, effects based on a percolation mechanism for various interacting or correlated pinning sites, including field percolation mechanisms can be thought of. In fact, this form of Jc(B) is similar to that of the distribution of paths at dielectric breakdown and of clusters at fracture threshold. [29-30] It is based on the scaling hypothesis idea for percolation. This can lead to our understanding that the intercoupling phenomenon as a function of B, whence the shielding paths, are critical in the statistical mechanics sense, or truly also of percolative nature.

Fig. 5: *J_C as a function of temperature at different DC applied magnetic fields for a 80/20% 123/211 composite material*



Also it is known [31-32] that the $J_C(B)$ dependence varies with oxygen content. A simple decay law is found for $x > 0.15$ and the fish tail behavior for $x < 0.15$. In view of the temperature dependence of the resistivity, the oxygen content seems a little bit weak indeed. This confirms the necessarily monotonous decay found here. It can be thought also that q is a function of x as in the superconductor glass model. [33]

In order to be complete, Fig. 5 gives the evolution of the critical current density at different DC applied magnetic field as a function of temperature. We do not have examined J_C at several enough temperatures to obtain the irreversibility line nor the complete (J_C, T_C, H_{C2}) technological diagram.

For completeness, let us however, mention that the irreversibility line $H_{AC}(T_P)$ for YBCO and y % excess 211 particles (y < 15) has been measured with an AC susceptibility technique. [34] No J_C value was given though.

IV. Conclusions

This study clearly indicates that the addition of 211 particles to the 123 system affects the microstructure, modifies the nature of the intergrain boundaries and the current carrying properties of the material. This results in the fact that *the* 123 *grains are strongly coupled* with clean interfaces and that a *a high value bulk shielding intergranular current can be established* at sufficiently high temperature. The processing route is thus quite optimized. Secondly, the effective pinning is large, due to large interfacial pinning force density. The role of oxygen vacancies inside the grains does not seem to be the predominant mechanism here though the vacancies might be relevant for intragrain pinning at higher fields. Finally, the variation law of J_C with field is claimed to result from the percolative structure of the materials.

Acknowledgement

This work is part of the Impulse Program on High Temperature Superconductors of the Belgium Federal Services for Scientific, Technical, and Cultural (SSTC) Affairs under contract SU/02/13. R.C. is particularly grateful to the FNRS for a research fellowship. The support of a Université de Liège ARC (94-99/174) contract is also acknowledged.

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