Published in: Superconductor Science and Technology (2005), vol. 18, iss. 4, pp.508-512
Status: Postprint (Author's version)

Silver paint as a soldering agent for DyBaCuO single-domain welding

J-P Mathieu[1], J-F Fagnard[2], Ph Laurent[2], B Mattivi[2], C Henrist[3], Ph Vanderbemden[2], M Ausloos[4] and R Cloots[1,2]

[1] *SUPRATECS, Chemistry Institute B6, University of Liège, Sart-Tilman, 4000 Liège, Belgium*
[2] *SUPRATECS, Department of Electrical Engineering and Computer Science B28, University of Liège, Sart-Tilman, 4000 Liège, Belgium*
[3] *CATμ, University of Liège, Sart-Tilman B6, B-4000 Liège, Belgium**
[4] *SUPRATECS, Physics Institute B5, University of Liège, Sart-Tilman, 4000 Liège, Belgium*

Abstract

Silver paint has been tested as a soldering agent for DyBaCuO single-domain welding. Junctions have been manufactured on Dy-Ba-Cu-O single domains cut either along planes parallel to the c-axis or along the *ab*-planes. Microstructural and superconducting characterizations of the samples have been performed. For both types of junctions, the microstructure in the joined area is very clean: no secondary phase or Ag particle segregation has been observed. Electrical and magnetic measurements for all configurations of interest are reported ($\rho(T)$ curves, and Hall probe mapping). The narrow resistive superconducting transition reported for all configurations shows that the artificial junction does not affect significantly the measured superconducting properties of the material.

1. Introduction

Nowadays, the production of a large number of good quality (RE)BaCuO (RE = rare earth) single-domain bulk samples by the top-seeded melt textured growth (TSMTG) process is well established [1]. Nevertheless, applications based on superconducting technology require complex-shaped superconducting single domains. For example, a superconducting ring is required for making flywheels [2-4]. More complex shapes are required for applications involving superconducting rotating machines [5] or magnetic field shields [6].

In order to produce complex-shaped samples, the growth of large single grains which would be drilled and machined is not the best solution [7]. The main reason is that the sample inhomogeneity increases with increasing size. These inhomogeneities mainly affect the transport current; consequently, large samples usually display a smaller critical current density, J_c, than smaller ones. Moreover, machining ceramics, and especially superconducting (RE)BaCuO single domains, is very difficult because of the poor mechanical properties of such materials; they are very brittle [8].

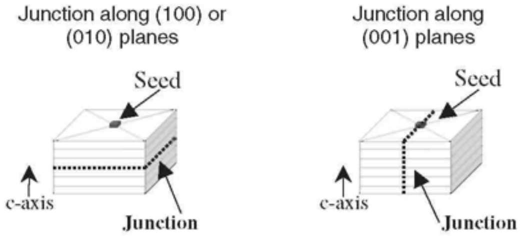
An alternative way to produce complex-shaped superconducting materials is to use the infiltration and growth (IG) process instead of the TSMTG process. The IG process is based on the infiltration of a barium and copper rich liquid phase into a RE-211 preform, which could have a complex shape, even a foam structure, at a temperature near the peritectic temperature of the RE-123 phase [7, 9-17]. This technique allows us to obtain high density materials with small size RE-211 particles well dispersed in the bulk [9]. Moreover, a near-net shape material can be obtained. Indeed, the process leads to a limited shrinkage which reduces cracks and distortions in the superconducting matrix.

In order to manufacture large single domains, it is also possible and efficient to weld small high-quality single domains together. In such a case, the junction has to display the highest possible critical current density. Various techniques for joining (RE)BaCuO single domains have been developed: (i) a natural joining using a multi-seeding technique [18-23]; (ii) diffusion bonding of polished surfaces under uniaxial pressure in the absence of soldering agents; and (iii) artificial joining by using low peritectic temperature rare earth cuprate ceramics, like $YbBa_2Cu_3O_7$, $ErBa_2Cu_3O_7$ or $TmBa_2Cu_3O_7$, as soldering agents [24-29]. Silver has been also used to reduce locally the peritectic temperature of the RE-123 phase [20, 30, 31]. Unfortunately, using low melting point RE-123 compounds leads to microstructural inhomogeneities which locally affect the critical current density. YBCO/Ag used as a soldering agent gives better results. However, the processing time is long because YBCO/Ag single domains have to be prepared prior to manufacturing the junction.

* www.catmu.ulg.ac.be

Published in: Superconductor Science and Technology (2005), vol. 18, iss. 4, pp.508-512
Status: Postprint (Author's version)

Figure 1: *Description of the junction configurations.*



Recently, Iliescu *et al* [32] have proposed an alternative method for single-domain welding along (100) or (010) planes. The authors suggest the use of a silver foil as a welding agent. This set-up gives good results and allows scaling up the production of welded samples.

In this paper, we have studied the efficiency of silver paint as a soldering agent. The main advantage of silver paint compared with the silver foil is a greatly simplified processing. Pieces to be welded quickly stick together after the evaporation of the solvent. The manipulation of the samples is thus made easier. More precisely, we have constructed junctions of DyBaCuO single domains either along the (100) or (010) planes (planes parallel to the c-axis) or (001) planes (planes perpendicular to the c-axis); see figure 1. These junctions have been characterized by microscopy and electrical measurements; all configurations of interest are reported.

2. Experimental details

DyBaCuO single domains were at first produced by a top-seeded melt-textured growth process using a $Sm_{1.8}Ba_{2.4}Cu_{2.4}O_x$ single-grain seed. The details of the single-domain synthesis are described elsewhere [8].

Single-domain samples were cut along a plane parallel to the c-axis or along an *ab*-plane (plane perpendicular to the c-axis) with a diamond disc fretsaw. The so-obtained faces were finely polished in order to obtain flat surfaces for welding.

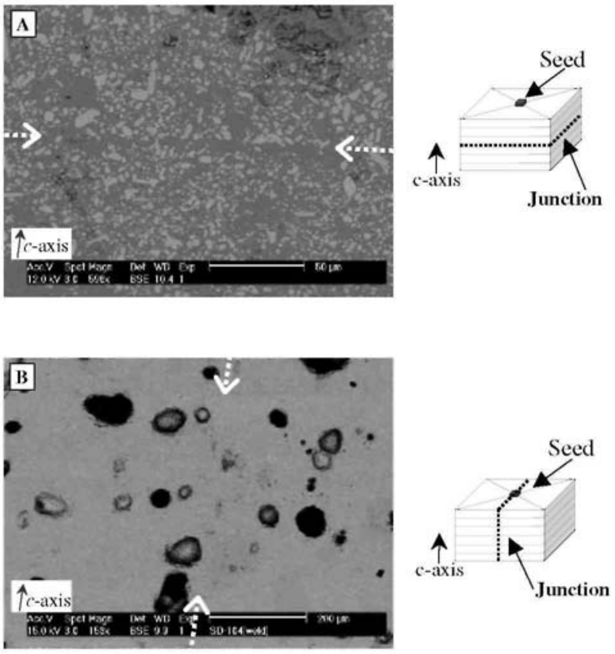
A silver paint, consisting of silver particle suspension in iso-butyl methyl ketone solvent, was used to cover the faces to join. These faces were quickly manually pressed against each other before the solvent evaporated. Since the silver paint makes the different pieces stick together, complex-shaped samples with multi-junctions can be easily constructed. The thickness of the 'silver layer' was optically observed to be about 50-60 μm depending on the painting conditions.

The sample was then subjected to a heat treatment. The sample was first heated rapidly to a temperature of 990°C and maintained at this temperature for 1 h. This step was followed by a slow cooling ramp (0.5 ° C h^{-1}) to 950 °C in order to induce an efficient recrystallization of the material around the junction. Subsequently the system was cooled down at 200 °C h^{-1} up to room temperature. Oxygen annealing at 450 °C during 200 h was applied afterwards.

The microstructural quality of the joined samples was analysed with a polarized-light optical microscope (Olympus Vanox AHMT3) and a SEM-EDX (Philips XL30 FEG-ESEM).

The superconducting properties were characterized by micro Hall probe mapping in liquid nitrogen. The Hall probe active area was 0.1 mm by 0.1 mm and the measurements were performed with a 0.5 mm step on a 25 mm x 25 mm scanned surface at 1 mm from the sample top surface. The sample is magnetized following a field cooling (FC) procedure using an air coil providing an applied magnetic field of $\mu_0 H$ = 380 mT in liquid nitrogen. The $\rho(T)$ transport measurements were obtained using a conventional four-point technique in a quantum design physical property measurement system (PPMS) with applied magnetic fields, $\mu_0 H$, ranging between 0 and 3 T. The resistivity measurements were performed following a zero-field cooling (ZFC) procedure in a range between 60 and 100 K with a sweep rate of 0.3 K min^{-1}. The temperature step was 0.1 K and the applied current was 10 mA.

Figure 2: *SEM micrographs of the welded samples. (A) Junction along a (100) or (010) plane; (B) junction along a (001) plane. The observed plane is indicated by a dashed line on the schematic representation of the sample.*



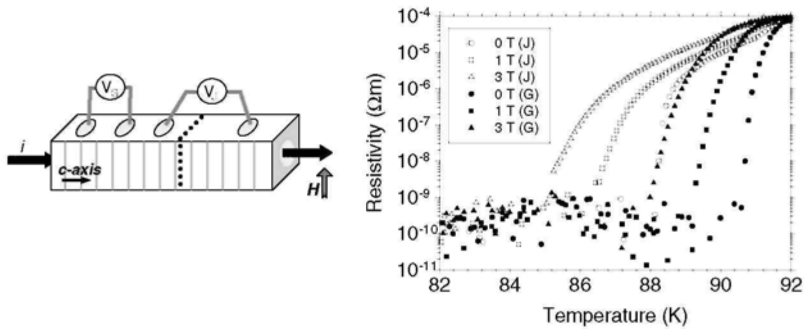
3. Results and discussions

3.1. Microstructural characterizations

SEM micrographs of welded samples are presented in figure 2. A junction along a plane perpendicular to the c-axis (usual configuration of junction) is shown in figure 2(A), and one in an *ab*-plane in figure 2(B). In both cases, it can be observed that the junctions are very clean. No secondary phase has been observed. Polarized light microscopy confirms that the junctions are well textured. The junction porosity is a little higher in the central part than near the edge of the single domain. This porosity is probably due to impurities contained in the silver paint, to residual solvent traces, or to air trapping during the pressing.

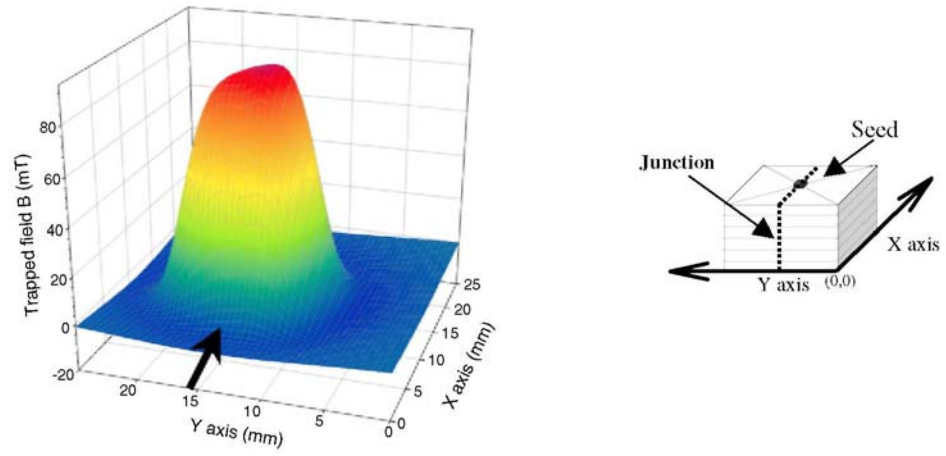
In addition, by performing electronic microscopy in back-scattered electron mode (BSE), no silver particles were found. Furthermore, EDX measurements did not highlight the presence of silver in the ceramics near the junction. This might be due to the fact that the sensitivity of EDX detection is however limited for light elements, such as silver, in a heavy matrix, here DyBaCuO. Nevertheless, both results suggest that the silver content in the sample is very small, probably smaller than 1-2 at.%, and well distributed.

Figure 3: *Temperature dependence of the resistivity along the c-axis (junction perpendicular to the c-axis) with applied magnetic fields $\mu_0 H$ = 0,1 and 3 T. The field is perpendicular to the current flow and to the c-axis. Filled symbols correspond to intragrain measurements, whereas open symbols are for 'across the junction' measurements. Note the extended log scale for the resistivity axis.*



Published in: Superconductor Science and Technology (2005), vol. 18, iss. 4, pp.508-512
Status: Postprint (Author's version)

Figure 4: *Hall probe mapping at 77 K of a welded sample with junction parallel to the c-axis. The arrow corresponds to the axis of the junction. A view of the sample has been plotted in order to correlate the trapped field with the surface probed by positioning precisely the X and Y axes.*



3.2. Electrical and magnetic properties

Both kinds of junctions were characterized by $p(T)$ measurements. Results for samples with the junction along a plane perpendicular to the c-axis are presented in figure 3, i.e., the c-axis resistivity versus temperature curves with magnetic fields $\mu_0 H = 0$, 1 and 3 T applied perpendicular to the *c*-axis. Intragrain, *G* (filled symbols), and 'across the junction', *J* (open symbols), measurements are presented on the same graph using a logarithmic scale for the resistivity axis.

For intragrain and 'across the junction' measurements, the onset critical temperature, T_c, is about 92 K, which is a reasonable value for Dy-123. The resistive transitions within a grain are rather narrow whatever the applied magnetic field. For 'across the junction' measurements, the transition is widened. Remarkably, however, in the absence of magnetic field, the transition width is less than 4 K. Only a small shouldering is observed on $\rho(T)$ when measured across the junction. This behaviour is similar to that found by Doyle *et al* [33, 34]. This can be related to the junction weak link effect. Another reason for this shouldering in the $\rho(T)$ curves might be related to macrocracks in the sample. Recall that in the studied configuration the injected current flows along the c-axis (as shown in figure 2). This corresponds to a geometry in which eventual macrocracks, usually directed parallel to the *ab*-planes, may impede the superconducting current flow. Indeed, such macrocracks have been observed by optical microscopy after $\rho(T)$ measurements run between the V_J contacts and not between the V_G contacts. They may have induced a weak link effect responsible for a shouldering in the $\rho(T)$ curve.

Samples with junction in a plane parallel to the c-axis were characterized by micro Hall probe mapping at $T = 77$ K (figure 4). The shape of the trapped field distribution exhibits one single peak and no decrease of the trapped field near the junction. Although Hall probe mapping results should be interpreted with great care when analysing a single grain boundary [34], it is not possible to give evidence for a weak link effect of the junction by this technique.

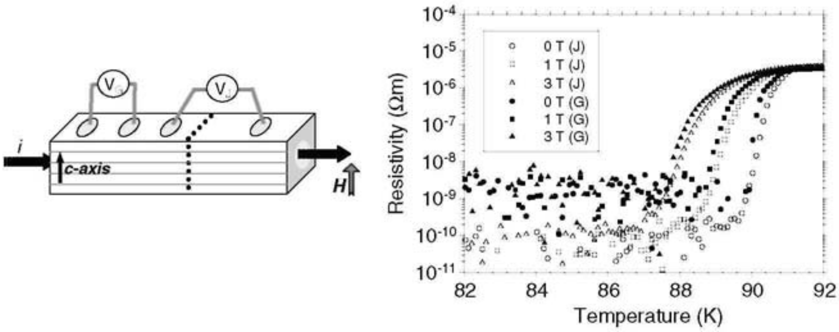
Resistivity versus temperature measurements of samples with junctions in a plane parallel to the c-axis are presented in figure 5. Intragranular (G) and 'across the junction' (*J*) $\rho(T)$ curves are plotted on the same graph, using a logarithmic scale for the resistivity axis. In this case, the current flows along the *ab*-planes in the presence of magnetic fields $\mu_0 H$ ranging from 0 to 3 T, and applied parallel to the c-axis and perpendicular to the injected current.

The transition is narrow and the onset critical temperature, T_c, is about 91 K. This T_c value is slightly lower than the T_c of samples with a junction perpendicular to the c-axis (cf figure 2). This small difference can be attributed to a better oxygenation in the case of the sample with junction perpendicular to the c-axis.

The $\rho_G(T)$ and $\rho_J(T)$ curves are very similar and very close to each other for all applied magnetic fields. No intermediate shoulder is observed on $\rho_J(T)$ curves, and these curves are only shifted by under 0.2 K. Therefore the junction does not act as a weak link nor significantly affect the critical current density.

It can be observed that the resistivity at $T > T_c$ is higher along the c-axis (figure 4) than in the *ab*-planes (figure 5). This can be related to the anisotropy of the Y-123-type structure.

Figure 5: *Temperature dependence of the resistivity along ab-planes (junction parallel to the c-axis) with applied magnetic fields $\mu_0 H$ = 0, 1 and 3 T. The field is perpendicular to the current flow and parallel to the c-axis. Filled symbols correspond to intragrain measurements, whereas open symbols are related to 'across the junction' measurements. Note the log scale for the resistivity axis.*



4. Conclusions

Silver paint has been successfully used to weld Dy-Ba-Cu-O single domains. Junctions have been performed in planes perpendicular or parallel to the c-axis. The microstructure of the junctions in both studied configurations is very clean; no secondary phase nor Ag particles can be observed. Optical microscopy allows us to conclude that the texturation occurs in all the material around the junction.

The junction parallel to the c-axis is characterized by better electrical properties than the junction perpendicular to the c-axis. For samples with the junction parallel to the c-axis, the narrow superconducting resistive transition, which is very close to the intragranular transition, shows that the junction does not alter significantly the superconducting properties of the samples.

Acknowledgments

The preparation and the characterization of materials are parts of the doctorate thesis of J-P Mathieu who thanks FRIA (Fonds pour la Formation à la Recherche dans l'Industrie et dans l'Agriculture, Brussels) for financial support. Part of this work results from research activities in the framework of the VESUVE project of the Walloon Region (RW.01.14881), and of the Supermachines project of the EC.

References

[1] Gawalek W, Habisreuther T, Zeisberger M, Litzkendorf D, Surzhenko A, Kracunovska S, Prikhna T, Oswald B and Canders W 2004 *Supercond. Sci. Technol.* 17 1185

[2] Ma K B, Chen Q Y, Postrekhin E, Zhan Y and Chu W K 2000 *Physica C* 341-348 2517

[3] Komori M and Akinage N 2001 *IEEE Trans. Appl. Supercond.* 11 1733

[4] Fang J R, Zin L Z, Yan L G and Xiao L Y 2001 *IEEE Trans. Appl. Supercond.* 11 1657

[5] Coombs T, Cansiz A and Campbell A M 2002 *Supercond. Sci. Technol.* 15 831

[6] Harvey I K 1972 *Rev. Sci. Instrum.* 43 1626

[7] Cloots R, Koutzarova T, Mathieu J P and Ausloos M 2005 *Supercond. Sci. Technol.* 18 R9

[8] Mathieu J P, Cano I G, Koutzarova T, Rulmont A, Vanderbemden Ph, Dew-Hughes D, Ausloos M and Cloots R 2004 *Supercond. Sci. Technol.* 17 169

[9] Mathieu J P, Koutzarova T, Rulmont A, Fagnard J F, Laurent P, Mattivi B, Vanderbemden Ph, Ausloos M and Cloots R 2004 *Supercond. Sci. Technol.* 18 S136

[10] Reddy E S and Rajasekharan T 1998 *J. Mater. Res.* 13 2472

[11] Reddy E S and Rajasekharan T 1998 *Supercond. Sci. Technol.* 11 523

[12] Noudem J G, Reddy E S and Schmitz G J 2004 *Physica* C 390 286

[13] Meslin S and Noudem J G 2004 *Supercond. Sci. Technol.* 17 1324

[14] Noudem J G, Guilmeau E, Chateigner D, Lambert S, Reddy E S, Ouladdiaf B and Schmitz G J 2004 *Physica* C 408-410 655

[15] Reddy E S, Noudem J G, Tarka M and Schimtz G J 2000 *Supercond. Sci. Technol.* 13 716

[16] Reddy E S, Herweg M and Schmitz G J 2003 *Supercond. Sci. Technol.* 16 608

[17] Reddy E S, Babu N H, Shi Y H, Cardwell D A and Schimtz G J 2003 *Supercond. Sci. Technol.* 16 L40

[18] Jee Y A, Hong G W, Sung T H and Kim C J 2000 *Inst. Phys. Conf. Ser.* 167 107

[19] Kim C J, Jee Y A, Kwon S C, Sung T H and Hong G W 1999 *Physica* C 315 263

[20] Harnois C, Chaud X, Laffez I and Desgardin G 2002 *Physica* C 372-376 1103

[21] Delamare M P, Bringmann B, Jooss C, Walter H, Leenders A and Freyhardt H C 2002 *Supercond. Sci. Technol.* 15 16

[22] Kim H, Kim C J, Hong G W and Joo J 2002 *Physica* C 372-376 1159

[23] Furusawa K, Chikumoto N, Ogasawara K, Nagatomo T and Murakami M 2002 *Physica* C 378 255

[24] Delamare M P, Walter H, Bringmann B, Leenders A and Freyhardt H C 2000 *Physica* C 329 160

[25] Noudem J G, Reddy E S, Tarka M, Noe M and Schmitz G J 2001 *Supercond. Sci. Technol.* 14 363

[26] Prikhna T *et al* 2001 *Physica* C 354 333

[27] Iida K, Yoshioka J, Sakai N and Murakami M 2002 *Physica* C 370 53

[28] Prikhna T A *et al* 2002 *Physica* C 372-376 1528

[29] Noudem J G, Reddy E S, Goodilin E A, Tarka M, Noe M and Schmitz G J 2002 *Physica* C 372-376 1187

[30] Harnois C, Desgardin G and Chaud X 2001 *Supercond. Sci. Technol.* 14 708

[31] Puig T, Rodriguez P, Carrillo A E, Obradors X, Zheng H, Welp U, Chen L, Claus H, Veal B W and Crabtree G W 2001 *Physica* C 363 75

[32] Iliescu S, Granados X, Bartolomé E, Sena S, Carrillo A E, Puig T, Obradors X and Evetts J E 2004 *Supercond. Sci. Technol.* 17 182

[33] Doyle R A, Bradley A D, Lo W, Cardwell D A, Campbell A M, Vanderbemden P and Cloots R 1998 *Appl. Phys.Lett.* 73 117

[34] Vanderbemden P, Bradley A D, Doyle R A, Lo W, Astill D M, Cardwell D A and Campbell A M 1998 *Physica* C 302 257