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Fabrication of textured YBCO bulks with artificial holes

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Abstract

The recently reported hole-patterned $YBa_2Cu_3O_y$ (Y123) bulks *with improved superconducting properties are* highly interesting from the aspects of the material qualities and application various points of view. It is well known that the core of plain bulk superconductors needs to be fully oxygenated and some defects like cracks, pores and voids must be suppressed in order that the material can trap high magnetic field or carry high current densities. To minimize the above defects, we have used the infiltration and top seed growth (ITSG) process to prepare regularly perforated $YBa_2Cu_3O_y$ (Y123) bulk superconductors. This process involves less shrinkage during annealing and a uniform distribution of Y211 inclusions. The texture was evidenced by neutron pole figure measurements. Flux mapping was used to verify the superconducting homogeneity of the samples and to investigate the field trapping ability. Large increase of the trapped field in pulsed magnetization up to 60% in comparison to the hole-free parent sample with the same thermal history has been obtained. In addition, the textured drilled samples were reinforced using resin or metal impregnation and the influence of the different processing steps on the hardness of the materials is investigated.

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Keywords: YBaCuO drilled; Infiltration; Texture; Mechanical properties

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1. Introduction

It is well established that the critical current density-J_c is closely related to the microstructural features in the melt processed $REBa_2Cu_3O_y$ (RE = rare earth) samples [1–4]. Dense materials with oriented single-domains, free from cracks, voids or porosity are necessary to obtain high J_c. The conventional melt processing of $REBa_2Cu_3O_y$ involves shrinkage [5] of the preform up to 25% due to the liquid phase outflow, resulting in distortions and macrodefects as reported in other systems [6,7]. Several works [8–14] have been reported for bulk $REBa_2Cu_3O_y$ using a similar infiltration-growth process. It is also well known that during the oxygen annealing inducing superconductivity, cracks open at the surface layer and propagate into the bulk [15,16]. To overcome such problems, the perforated geometry offers a significant potential in helping the oxygenation process, avoiding crack developments and increasing mechanical reinforcement.

In this study, we have used the infiltration and top seed growth (ITSG) process to prepare textured Y123/Y211 composites. Additionally, textured bulks YBCO with multiple holes were processed and flux mapping, neutron diffraction and mechanical properties have been investigated after reinforcement.

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2. Experimental

The details of infiltration and top seed growth (ITSG) and multiple holes process of $YBa_2Cu_3O_y$ (Y123) are reported elsewhere [13,17]. Basically, The $Y_2Ba_1Cu_1O_y$ (Y211) block or pellet was placed on top of the liquid phase ($3BaCuO_2 + 2CuO$) or $Ba_3Cu_5O_8$ (Y035) to allow the liquid infiltration driving by capillarity force. A Sm123 seed as a nucleation center was placed on the top of the Y211 preform. The holes into the preform were realised by drilling cylindrical cavities with diameter 0.5–2 mm through the circular or square shaped sample. The holes are arranged in a regular network in the plane of the samples (Fig. 1).

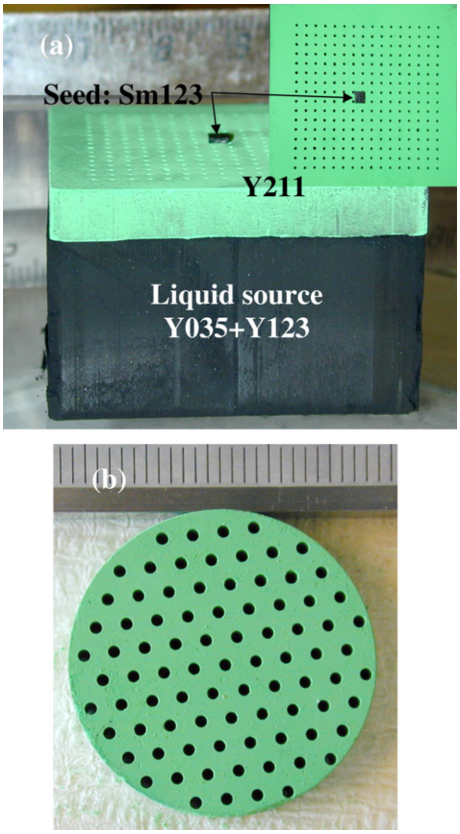

Fig. 1. (a) Configuration (cross-section) used for converting the mechanically drilled Y_2BaCuO_5 into a single phase of $YBa_2Cu_3O_{7-\delta}$ (Y211) by the infiltration and growth process. The Sm123 seed crystal used for nucleation is placed on the surface of the perforated structure. Inset showing the top view with artificially patterned holes ($\Phi = 0.5$ mm). (b) Top view of the multiple holes ($\Phi = 1$ mm) Y211 pellet.

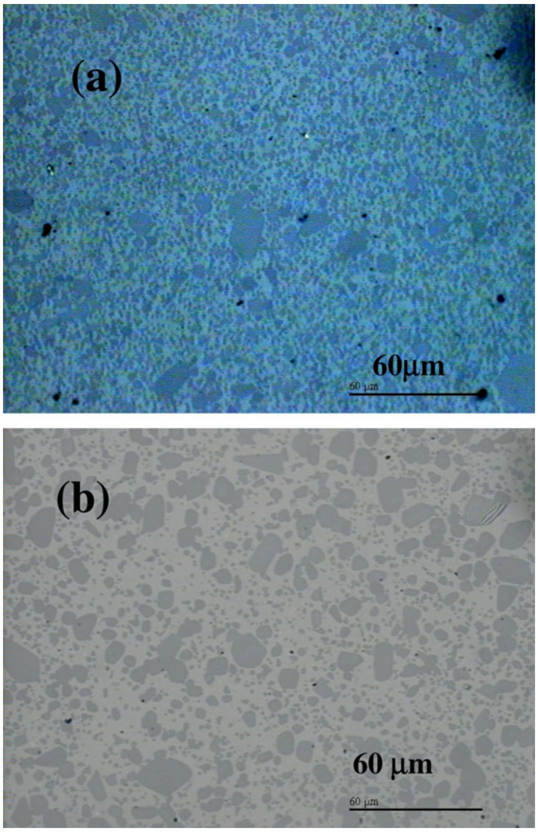

Fig. 3. Microstructures of the samples prepared by (a) ISTG and (b) TSMTG process.

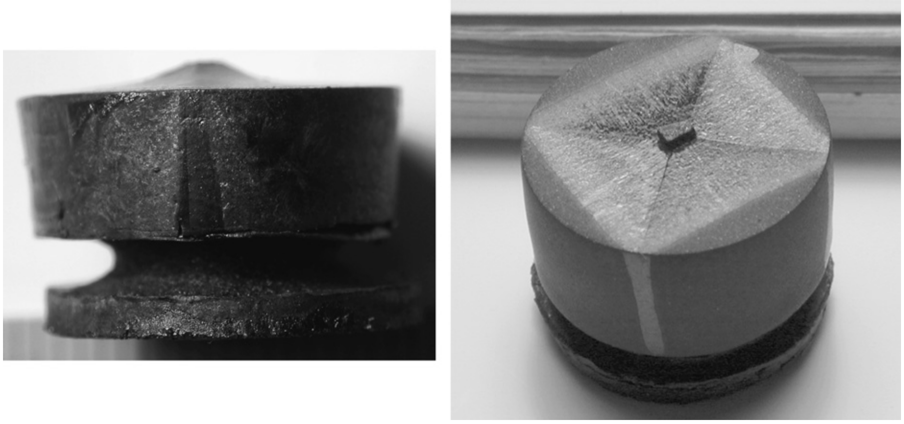

Fig. 2. As-grown bulk sample obtained from ITSG-process showing the strong shrinkage of the source and the single-grain growth along the whole height of the pellet, evidenced by the contrast on the section.

DC magnetization measurements were performed on square-shaped samples, selected from the single domains, using a SQUID magnetometer to determine the critical transition temperature, T_c, and critical current density, J_c, under magnetic field. J_c was estimated using the extended Bean critical state model [18].

In order to estimate the influence of the holes and the impregnation phases on the mechanical properties of the samples, we have determined the Vickers hardness of hole-free specimens, of non-impregnated-drilled samples (holes diameter of 0.5 and 1 mm) and drilled samples impregnated by metal (holes diameter of 0.5 and 1 mm) and resin (holes diameter of 0.5 mm). The Vickers indentations were introduced on polished surfaces by means of a Zwick testing machine. Prior to the test, a thin gold layer (500 Å) was sputtered on the polished surfaces of the samples, in order to obtain a clean-looking impression. A load of 14.7 N was applied for 15 s and the hardness was determined from the usual relationship:

$$H_v = \frac{1.854P}{4a^2}$$

where "a" is the mean half-diagonal of the square-shaped hardness imprint (see Fig. 9).

For the drilled specimens (impregnated or not), the influence of the stress states around the holes on the hardness was investigated by performing tests in the "bulk" of the drilled samples, between two holes (+ in Fig. 9) and between four holes (× in Fig. 9). "Bulk" refers to central positions far away from the holes, but also not too close to the center as the values could be influenced by the seed.

3. Results and discussion

3.1. Infiltration-growth single-domain

A typical as-grown bulk sample obtained from ITSG-process is shown in Fig. 2. The trace of the faceted growth on the surface reveals that a single-domain pellet has been successful processed. The vertical shine trace on the side of the pellet demonstrates that the domain has grown on all thickness of the pellet.

Polished surface (Fig. 3) of the samples reveals the typical microstructure features known from melt processed bulk materials. In the present ITSG process, no specific efforts like doping [19,20] or irradiation [21] have been made to optimize the Y211 particle size and defect density in further improving the critical current densities. The optical micrographs of the ITSG sample along with a reference microstructure of that of Y123 prepared by conventional melt processing method is shown in Fig. 3b. The average

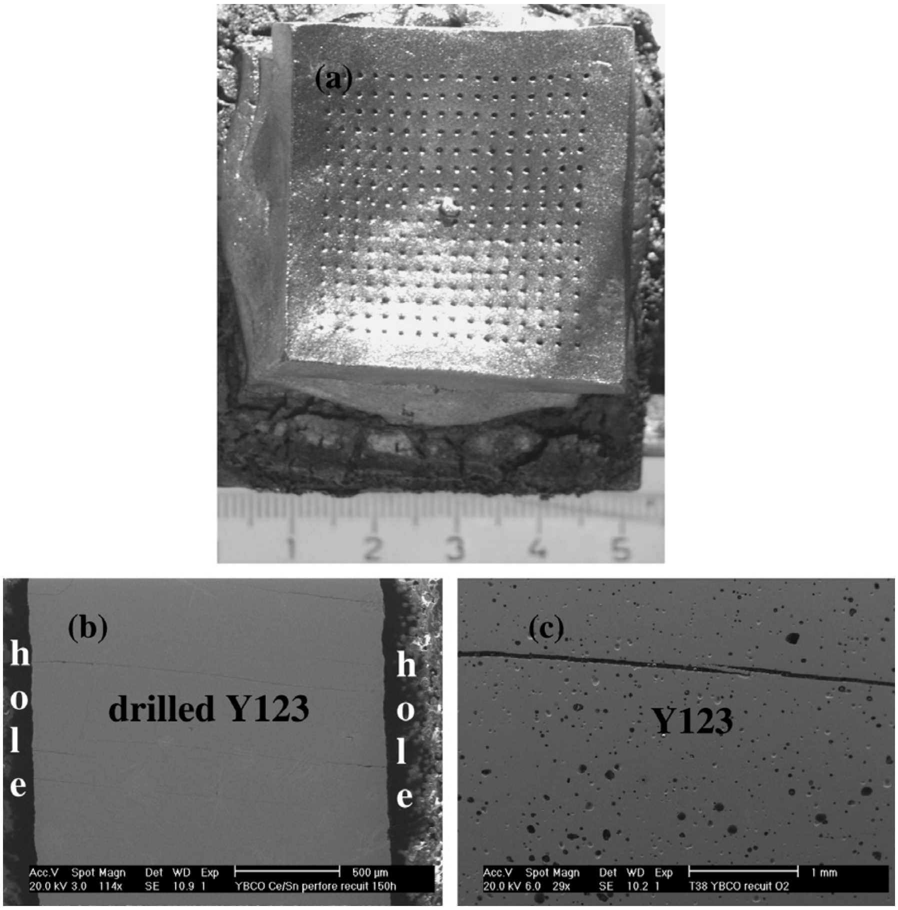

Fig. 4. (a) AS-process drilled square form with 0.5 mm hole, the microstructures of the drilled (b) and (c) plain sample, respectively.

size of Y211 particles was determined to be 1μm for ISTG sample compared to 2.4 μm for conventional top seeding melt texture growth (TSMTG) one. A uniform distribution of Y211 inclusions can be seen in the ITSG microstructure. The small Y211 particle obtained without the dopants is clearly one of the advantages of ITSG method. This refinement of Y211 inclusions is considered for inducing a good flux pinning and high critical current density. Magnetization hysteresis for all samples was measured at 77 K by using a SQUID magnetometer. The magnetic field was applied parallel to the axis of pellet. The magnetic, J_c, values are estimated for this direction from M–H cycles by applying the extended Bean critical-state relation [18]. At 77 K and self field, the J_c values are 86,000 A/cm^2 for ITSG sample compared to 10,000 and 90,000 A/cm^2 for the TSMTG without and with dopants, respectively. These measured values are comparable with those reported in the literature [22–24].

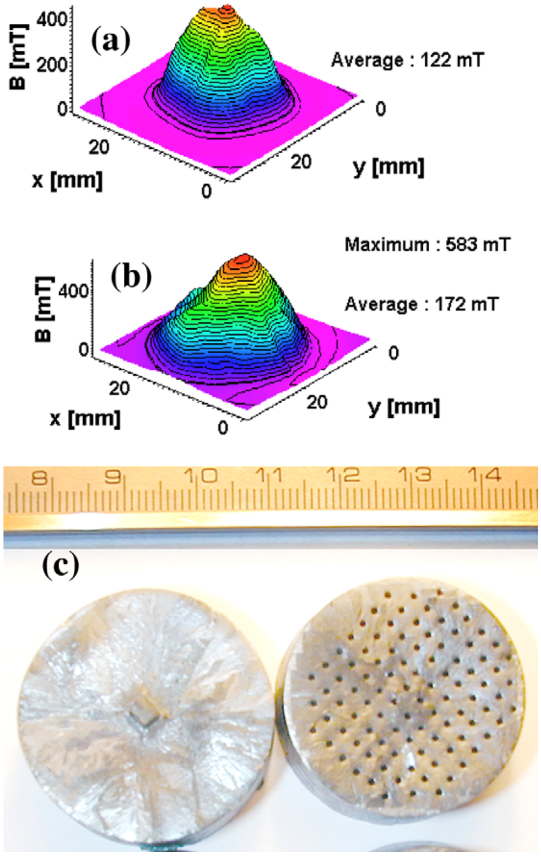

Fig. 5. (a) and (b) Flux trapped measurements on (c) perforated and plain pellets.

3.2. Perforated samples

The square or circular-shaped pellets of Y211 were grown into single grain by TSMTG or ITSG-process. Optical macrographs of as-grown samples with 0.5 mm holes are shown in Fig. 4a–c illustrate the cross sections of plain and perforated samples. The porosity is drastically reduced for the drilled sample. For the plain sample, a large porosity and crack zones are noticeable. The scanning electron microscopy between two holes shows (i) the compact, free-of-crack microstructure and (ii) an uniform distribution of fine Y211 particles into the Y123 matrix (like in Fig. 5).

Fig. 5 presents the flux trapping obtained on plain and perforated samples (36 mm in diameter and 15 mm in height) after conventional oxygenation at 450 °C for 150 h. The samples (Fig. 5b) were previously magnetized at 1 T, 77 K, using Oxford Inc. superconducting coil (CRETA, CNRS, Grenoble). The 3D representation of the magnetic flux shows the single dome in the both case

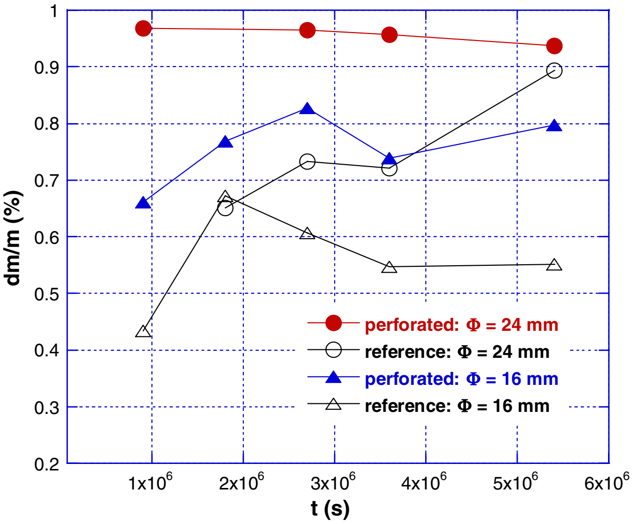

Fig. 7. Influence of oxygen annealing showing the oxygen uptake into the drilled and plain samples.

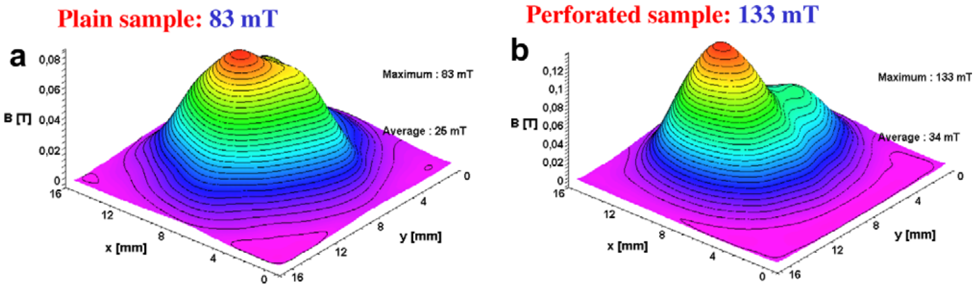

Fig. 6. Contour map of the trapped magnetic field of the (a) drilled and (b) plain samples as reference after pulse magnetization at 77 K.

corresponding to signature of a single-domain. The network of the holes has not affected the current loops at the large scale. This result was confirmed by the neutron diffraction measurements (D1B line at ILL, France) showing only one single domain bulk orientation with mean *c*-axis parallel to the pellet axis. The trapped field value is higher in the perforated pellet (583 mT) than in the plain one (443 mT). This represents an increasing of 32% for the drilled sample than the plain one in agreement with our previous report [25]. This increasing of the trapped field value is probably due to: (i) the better oxygenation and/or less cracks and porosities for the drilled pellet as illustrated on Fig. 4b, (ii) the strong pinning because the hole could be favourable the vortices penetration (iii) enhancement of the cooling, because the sample with holes offer the large and favourable surface exchange into the liquid nitrogen bath.

On the other hand, the pulse magnetization was used to perform the drilled and plain pellets. Both samples (16 mm diameter samples, 8 mm thick) were tested with a series of pulse magnetisation experiment. A Helmholtz coil was used to generate homogeneous magnetic field. The maximum amplitude of the magnetic field is 1 T and the raising time of the pulse was 1 ms and the decay time was 10 ms. After the pulse the trapped field was mapped with a hall sensor probe at 0.5 mm above the sample. The result shows (Fig. 6) that for 1 T pulse applied the trapped field increases up to 60% for drilled pellet with respect to the plain one. This is an interesting result for such kind of new geometry, demonstrating the ability of the textured Y123 with multiple holes to trap a high field.

3.3. Effect of oxygenation duration

According to their thin wall geometry, the drilled samples should be well oxygenated in comparison to the plain samples. The oxygen diffuses easily through the tube channels. The thermogravimetry technique was selected to compare the oxygenation quality of different pellets. The oxygen uptake was related to the increase of sample weight. In this study the pellets of 16 and 24 mm in diameter have used and the network of 30 holes has been perforated. For each diameter, five drilled and five plain pellets have been processed at the same heat treatment. All the samples have been weighted before and after the oxygenation and the percentage of the weight gain has been evaluated according to the following relation:

$$m\ (\%) = 100(m_{\text{final}} - m_{\text{initial}})/m_{\text{initial}}$$

The measurements have been realized twice to check the reproducibility. For that, the samples after the first measurement have been de-oxygenated at 900 °C, half an hour and follow by the quench step and then re-oxygenated. After the second measurement, the average values of the weight were estimated and plotted on Fig. 7. It is difficult to oxygenate the bulk sample with big diameter in this case the oxygen should diffuse into the core of the bulk. Generally the big samples are annealed under oxygen at 400–450 °C between 150 and 600 h [15,25–27]. These annealing dwell time are so long in order to allow the oxygen diffusion until the core of the monolith bulk materials. The drilled samples seem to offer an advantage (a saving of time) for annealing under oxygen of the superconductor bulk. This advantage is clearly shows in Fig. 9 where 25 h is sufficient to obtain the full oxygenated sample in the order word maximum weight gain is quickly reaching or achieve.

3.4. Reinforcement of the drilled samples

The Y123 domain with open holes could be reinforced, e.g. by infiltration with a low temperature melting alloy, to improve the mechanical properties that are useful for levitation applications or trapped field magnets. The perforated Y123 bulk with 1 or 2 mm diameter holes were dipped into the molten Sn/In alloy or epoxy wax at 70 °C for 30 min in a vessel after evacuating it with a rotary pump and venting air to enable the molten alloy or liquid resin to fill up the holes. After cooling, the impregnated bulk materials were polished. Some samples have been impregnated with BiPbSnCd-alloy using the process described elsewhere [28]. Fig. 8 shows the top surface and the cross sectional view of the impregnated Y123 bulk samples. We can notice

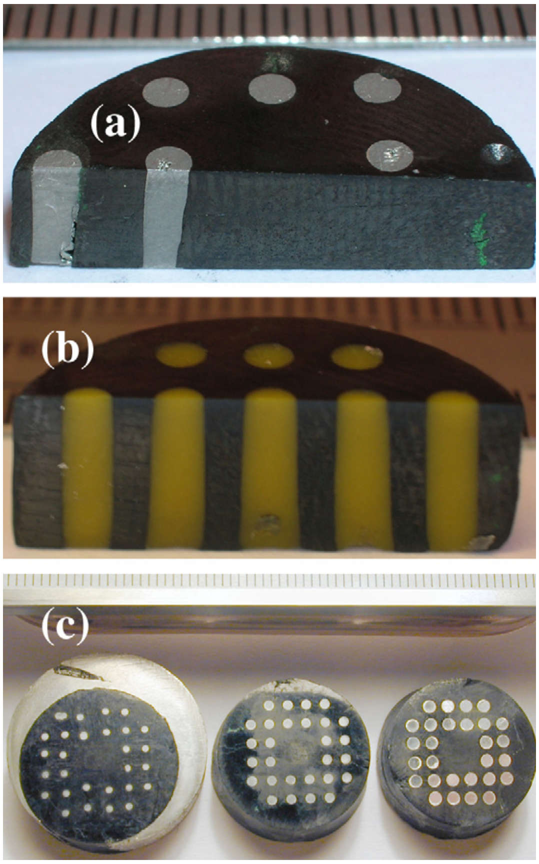

Fig. 8. Reinforcement of the drilled samples. (a) A view of cross section impregnated with Sn/In alloy, (b) with wax resin and (c) the top view of the samples filled with BiPbSnCd-alloy.

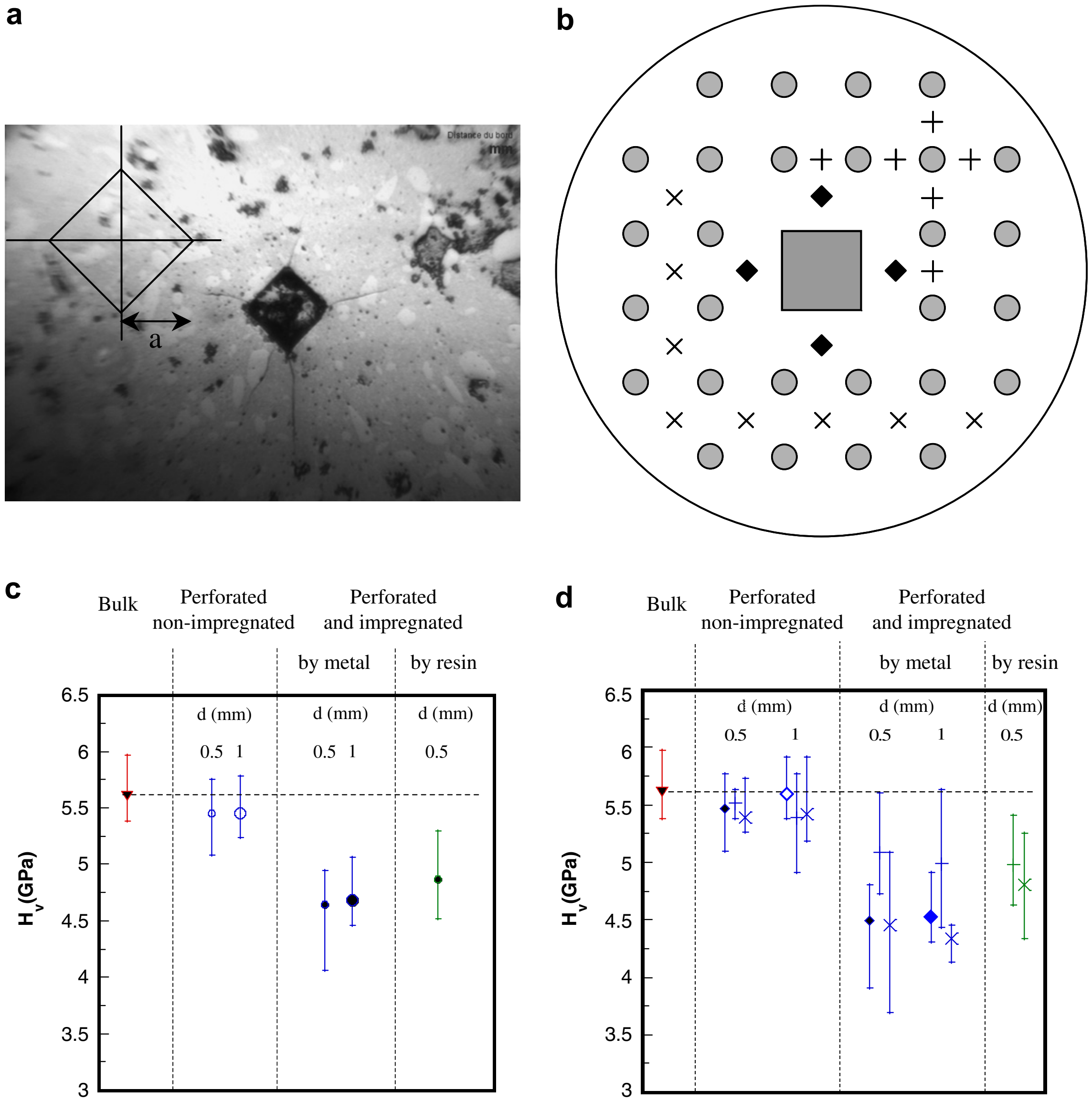

Fig. 9. (a) Optical micrograph showing a Vickers indentation in a hole-free and impregnation-free sample, the insert shows a sketch of the Vickers indentation. (b) Sketch of the different positions of the indentations on the drilled specimens (♦: in the bulk, +: between 2 holes, ×: between 4 holes) (seed print is the darkened square in the middle of the sample). (c) Hardness values determined for the different samples. (d) Influence of the indentation location on the hardness values (♦: in the bulk, +: between 2 holes, ×: between 4 holes).

the dense and homogeneous infiltration of the wax epoxy and Sn/In alloy. The magnetic flux mapping of the sample filled with BiPbSnCd-alloy has been investigated. The same trapped field of 250 mT before and after impregnation have been measured. Presently, it is important to develop the specific shapes of bulk superconductor with mechanical reinforcement for any practical application. Different papers have already reported and demonstrated the reinforcement of bulk superconducting materials. Krabbes and co-workers [29,30], for example, have observed that reinforcing Y123 monoliths with steel bandages and banding with high strength steel rings enables large grain samples to trap magnetic fields of 16 T at 24 K. In addition, Tomita and Murakami [28] reported trapped fields of 17 T at 29 K for a YBCO monolith after resin impregnation and wrapping the sample in carbon fiber.

3.5. Hardness anisotropy

The measured hardness of the bulk sample (i.e. non-impregnated hole free-material) is about 5.6 GPa, which is in a good agreement with different values reported in the literature [31,32]. Fig. 9 shows the average values of the hardness for the different studied samples. The large scatter of the experimental values is explained by the coarse microstructure of the materials and the used local investigation technique (micro indention). Comparison of the hardness values of the bulk to those obtained from non-impregnated drilled-samples indicates that the presence of the holes has no significant influence, whatever the diameter of the holes, in the investigated size range. The impregnation of the drilled samples by metal alloy or resin results in a decrease of the hardness of the samples. This softening was expected as a consequence of the introduction of a smoother phase into the superconductor material then usual. Moreover, as the resin is harder than the metal alloy, the hardness value of the resin-impregnated sample is higher than the metal-impregnated ones. Fig. 9b presents the hardness values as a function of the different locations in the samples. According to these results, no clear trend can be established between the hardness and the location of the test point, except for the metal-impregnated samples for which the hardness is somewhat higher between two holes than in the bulk or among holes.

4. Conclusion

Single grain Y123 superconductors have been grown by infiltrating a Y035 liquid source into a Y211 preform. The critical current density at 77 K and 0 T was determined to be 86,000 A cm^{-2}, which is close to the values of samples prepared by conventional TSMTG process. In addition, the textured Y123 bulks with multiple holes have been processed and characterized. SEM studies have shown that the holes presence does not hinder the domain growth. The perforated samples exhibit a *c*-axis grain orientation confirmed by pole figure and the single domain character is evidenced by trapped-field distribution. This new structure has a great potential for many applications with improved performances in place of Y123 hole free bulks since it should be easier to maintain at liquid nitrogen temperature during application, avoiding the hot spot apparition. It is clear that the Y123 bulks with an artificial pattern of holes are beneficial to evacuate porosity from the bulk and to uptake the oxygen.

The ability of the Y123 with multiple holes to trap a high field has been demonstrated. At 77 K, the trapped field value for drilled sample is 32% better than the plain sample one. Using pulse magnetization the trapped field increase up to 60% for drilled pellet with respect to the plain one.

Superconducting bulks with the artificial arrays of holes can be filled with metal alloys or high strength resins to improve their thermal properties, without any crucial decrease of the hardness which is needed to overcome the yielded stresses in levitation and quasi-permanent magnet applications.

Further investigations implying additional mechanical properties holes density and interconnected of regular holes are under way.

Acknowledgments

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