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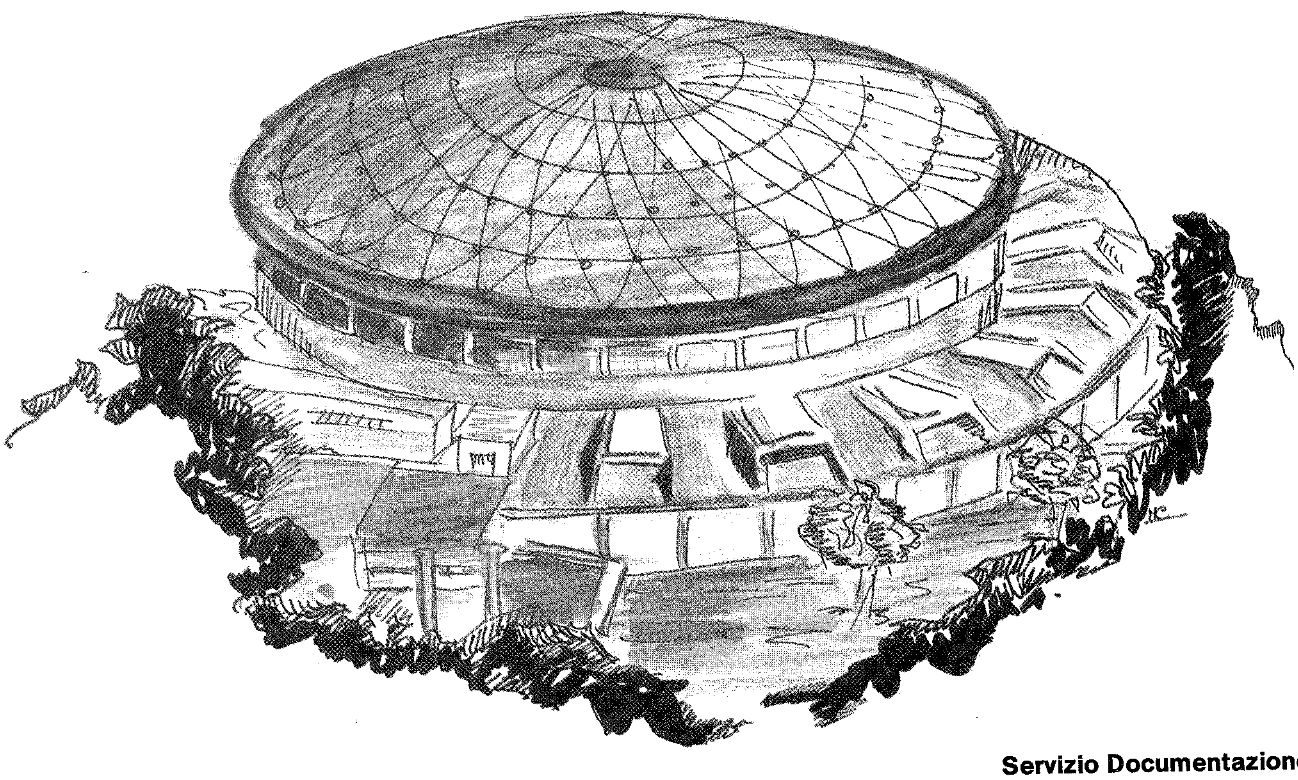

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Fabrication process and superconducting behavior of sintered $Y_1Ba_2Cu_3O_7$ ceramic samples

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Abstract

Electric and superconducting properties of $Y_1Ba_2Cu_3O_7$ sintered samples have been measured at different temperatures and in the presence of a magnetic field. The effects of some fabrication parameters, e.g. sintering temperature and time, on these properties have been investigated. The limiting effect on the critical current density through the $Y_1Ba_2Cu_3O_7$ samples is discussed in terms of the inter-grain coupling. Highest density samples sintered at temperature above 950 °C sustain lower critical current densities, and are more sensitive to the magnetic field.

Riassunto

Sono state misurate le proprietà elettriche e superconduttive di campioni sinterizzati di $Y_1Ba_2Cu_3O_7$ superconduttore a differenti temperature ed in presenza di campo magnetico. Sono stati esaminati gli effetti sulle caratteristiche elettriche del campione di alcuni parametri di fabbricazione, come la temperatura e la durata del processo di sinterizzazione. Sono discussi gli effetti che limitano la densità di corrente critica attraverso i campioni di $Y_1Ba_2Cu_3O_7$ in termini di accoppiamento tra i grani. I risultati delle nostre misure mostrano che campioni a densità elevata ottenuti a temperature superiori a 950 °C hanno una minore densità di corrente critica, e che quest'ultima è maggiormente influenzata dalla presenza di un campo magnetico.

1 - Introduction

The critical current density and its behavior as a function of the magnetic field, are important features of superconductors for practical applications. In particular high critical current density in strong magnetic field is a demand in large scale applications such as superconducting magnets. Bulk sintered samples of policrystalline $Y_1Ba_2Cu_3O_7$ show transport critical current values between 10^2-10^3 A/cm^2 at 77 K in zero magnetic field, and they exhibit a strong reduction in magnetic field /from 1 to 4/. These values are much lower than critical currents measured by magnetization cycles. This latter method may lead to an over estimation of the critical current density in policrystalline samples, because of the granular nature of these materials, which limits the transport properties of the bulk material. In fact the grains are coupled each other by weak links, which are strongly sensitive to the magnetic fields. In this work we deal with the critical current density, directly measured by transport current through the samples, and its dependence both on applied magnetic field and on the temperature, as a function of the sample density. The density range of the tested samples is between 0.5 and 0.9 of the theoretical density, assumed to be 6.3 g/cm^3. These densities have been obtained both varying the temperature and duration of the sintering process.

2 - Experiments

The samples have been prepared starting from an appropriate mixture of Y_2O_3, BaO_2, and CuO powders, at ENEA CRE, Casaccia. Three calcination steps at 700 °C, 800 °C, and 900 °C have been performed, each one followed by an intermediate grinding. Oxygenation of all the samples has been made at 650 °C and 400 °C, during the slow cool down. The sintering process lasted different times, and has been carried out at two temperatures (950 °C and 970 °C). In the sintering process the resistivity of the samples has been recorded in-situ, by a four contacts method. In Figs. 1a) and 1b) the relative sample densities as a function of the sintering process parameters, and the ambient temperature resistivity of the samples, after the process, versus their relative density, are respectively shown. Highest density samples have been obtained for long sintering time, at 970°C. The samples which show lower resistivities are those having the highest density sintered at 950 °C.

Further measurement to investigate the superconducting properties of the samples have been carried out at ENEA CRE, Frascati. The critical current density J_c has been measured in samples having the geometry of an half disc with a restriction in the center, in order to preserve for self-heating effects due to large currents. Moreover, to improve the electric connections to the sample, silver pads, consisting of 6000 Å thick films, in a linear four probe configuration have been deposited by electron beam on the sputter cleaned surface of the specimen. Both current and voltage wires have been soft welded with indium. By using this process we achieved current contact resistances lower than 0.3 Ω. Measurements have been carried out in a gas flow variable temperature cryostat, using both helium and nitrogen as coolant, provided with a temperature controller. Temperature is sensed by a silicon diode thermometer in close thermal contact with the sample. The magnetic field up to 150 Gauss, perpendicular to the current direction, is generated by a split coil magnet at ambient temperature. Further details on the measurement apparatus are given in ref. /5/. The behavior of the sample resistance lowering the temperature is recorded using a current bias of 1 mA. In Fig. 2 the temperature dependence of the resistance for a sample of density d=4.23 g/cm^3 is reported. In Fig. 3 the resistivity ratio between 270 °K and 110 °K as a function of the sample density is shown. From these data we observe that the resistivity ratio increases with density, except for the samples sintered at 970 °C. In Fig. 4 we report the results obtained from critical current density measurements versus the sample density. Critical current data have been recorded in a liquid nitrogen bath at 78 K. The critical current density J_c of the sample is computed from the current value which gives rise to the appearance of 1 μV across the voltage contacts 5 mm spaced. The largest value of J_c does not correspond to the highest density samples. In fact, although the samples sintered at 970 °C showed higher geometrical densities, they sustain lower current densit /6/. The effect of the inter grain coupling can be evidenced recording the behavior of the critical current when a weak magnetic field B is applied /2,7/. In Fig. 5 it is shown the typical behavior of the critical current I_c versus the applied magnetic field at temperature of liquid nitrogen.

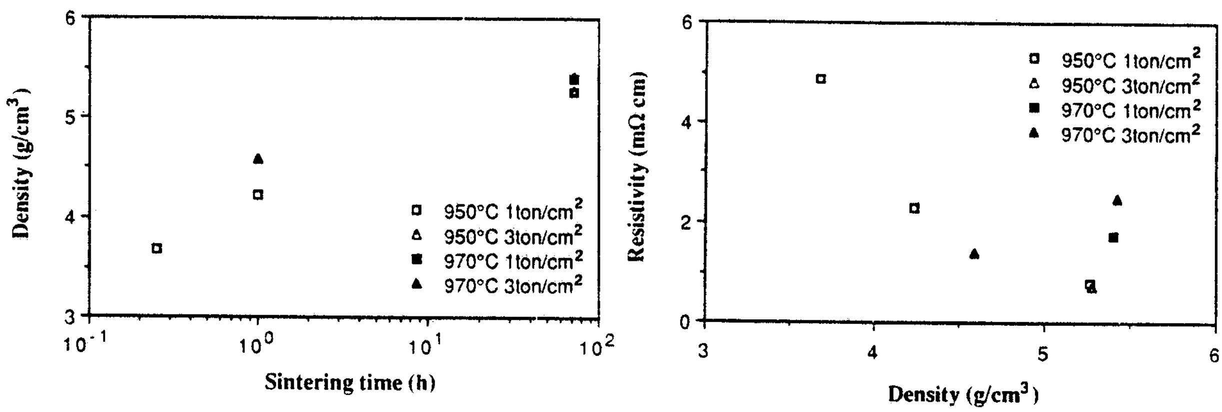

FIG. 1 - a) Sample density as a function of the sintering time; b) Sample resistivity measured at ambient temperature as a function of the sample density.

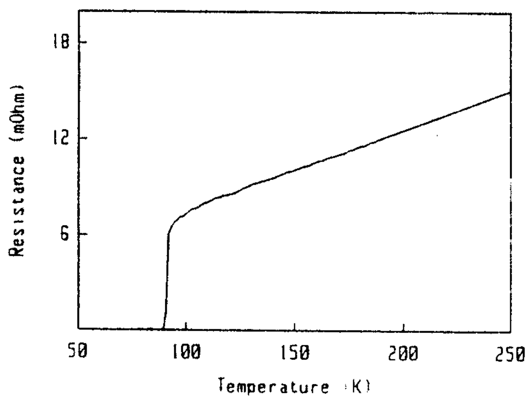

FIG. 2 - Resistance versus temperature in the sample with d=4.23 g/cm^3.

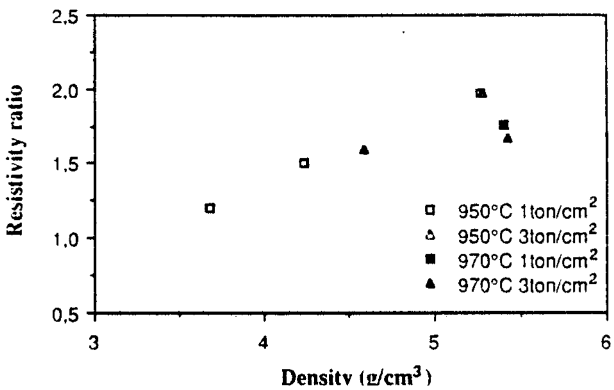

FIG. 3 - Resistivity ratio between 270 °K and 110 °K of our samples as a function of their geometrical density.

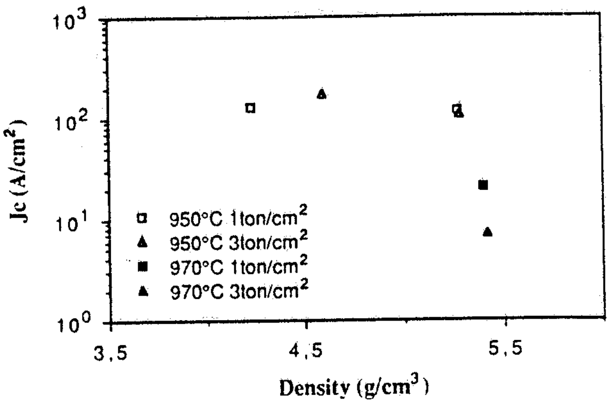

FIG. 4 - Critical current density at 78 °K as a function of the sample density.

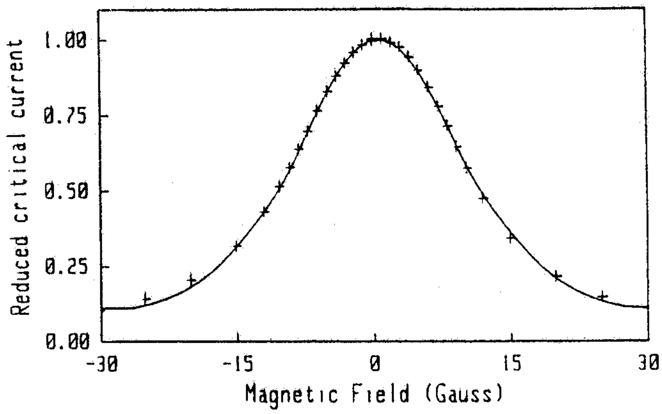

FIG. 5 - Critical current behavior as a function of magnetic field for a sample having d = 4.23 g/cm^3: (+) experimental data; solid line is eq. 1 computed for B_0 = 18 Gauss.

Let us observe that the magnetic field value is always kept much lower than the estimated first critical field $H_{c1}\approx$100 Gauss /3/. The dependence shown can be accounted for by a simple model a parallel array of Josephson junctions . In this case the total critical current is given by /2/:

$$\frac{I_c(B)}{I_c(0)} = \frac{1}{N}\sum_{i=1}^{N}\left|\frac{\sin \pi B/B_0F_i}{\pi B/B_0F_i}\right|$$

where the factor F_i accounts for the different effective area of each junction. In Fig. 5 the solid line represents eq. (1) for B_0=18 Gauss and N=26, using for the coefficients F_i the simple expressions:

$F_i = 0.5 + 0.05\ i$ (i=0, 1, ..., 25)

The particular choice of the coefficients F_i describes a physical situation in which the effective area between grains has a spread of a factor 4. In Fig. 6 the magnetic field pattern of two different samples having geometrical density of d=4.23 g/cm^3 and d= 5.28 g/cm^3 respectively are shown.

As can be noted the highest density specimen has the reduced I_c vs B pattern which exhibits a stronger dependence upon the applied magnetic field. This feature can be related both to the larger grain sizes in higher density samples, and to the presence of a liquid phase among grains, occurring when the sintering temperature approaches to the melting temperature /8/. More experimental data are given in ref. /6/.

The critical current density as a function of temperature has been also investigated. A typical temperature dependence of the critical current for one of our samples is shown in Fig. 7, where T_c is the transition temperature. In the same figure the usual $J_c \approx const * (1-T/T_c)^{3/2}$ Ginzburg - Landau dependence is reported as solid line a), while solid line b) is the typical $J_c \approx const * (1-T/T_c)^2$ behavior valid for a superconducting - normal - superconducting (S-N-S) junction /9/. The S-N-S dependence shows better agreement with experimental data approaching T_c, thus indicating the presence of normal zones between grains. Furthermore the I_c vs B dependence has been measured at different temperatures /7,2/. Since the critical field B_0 is inversely proportional to the penetration depth λ, eq. 1 would imply a temperature dependence of B_0 given by:

$$B_0 (T) = const * \{1 - (T/T_c)^4\}^{1/2}$$

The measured half-width of the I_c vs B patterns at different temperatures shows a reasonable agreement with the temperature dependence of B_0 (T) given by the last expression /6/.

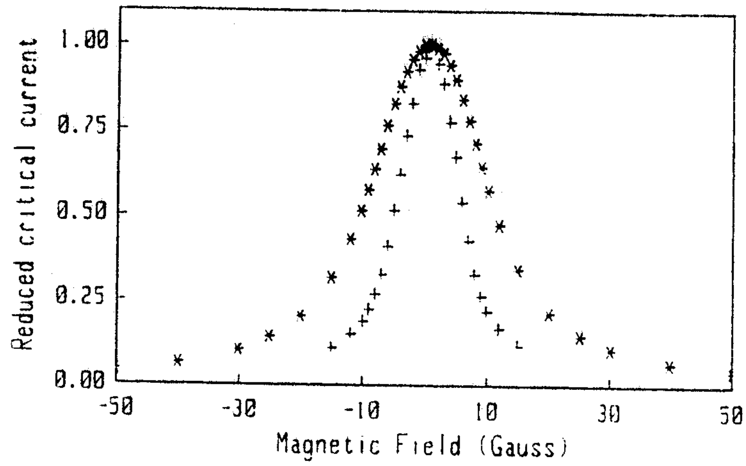

FIG. 6 - Normalized critical current behavior at 78 °K as a function of the magnetic field: (*) sample with d=4.23 g/cm^3; (+) sample with d=5.28 g/cm^3.

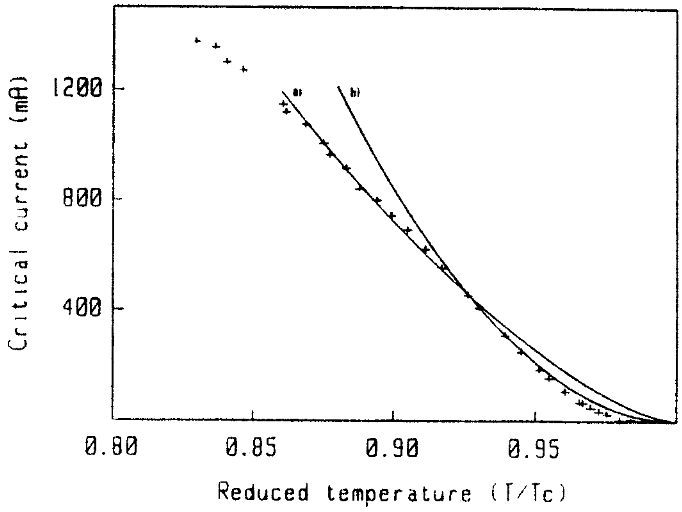

FIG. 7 - Temperature dependence of the critical current: (+) experimental data; solid line a) is the Ginzburg - Landau theoretical dependence $I_c \approx const * (1-T/T_c)^{3/2}$; solid line b) represents the S-N-S critical current behavior $I_c \approx const * (1-T/T_c)^2$.

3 - Conclusions

We have investigated the superconducting properties of $Y_1Ba_2Cu_3O_7$ sintered samples as a function of different fabrication parameters. We observed that the maximum values of critical current density are found in samples which had lower geometrical densities. This seems related to the highest temperature of 970 °C used to get the most dense samples, which allows the growth of liquid phase among the grains, thus reducing the transport current. The experimental behavior of the critical current in magnetic field at different temperatures as a function of the sample density has been studied. The magnetic and temperature behavior of the critical current are well described in terms of weak coupling between grains. We found that the highest density samples have a strongest dependence on the magnetic field. This is explained in terms of an increased junction area, due both to the larger grain size and to the non-superconducting liquid phase between grains, which makes more sensitive the decrease of the critical current in the presence of a magnetic field.

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