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Enhanced Connectivity and Percolation in Binary and Doped *in situ* MgB_2 Wires after Cold High Pressure Densification

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***Abstract*—The cold high pressure densification technique (CHPD) was recently developed in Geneva for improving the in-field critical current density J_c of *in situ* binary and alloyed MgB_2 wires and tapes [1, 2]. J_c of CHPD treated square wires alloyed with malic acid ($C_4H_6O_5$) was enhanced by a factor 2 at 10 T and 4.2 K, the behavior being almost isotropic. In order to understand the fundamental mechanism behind this strong improvement of J_c, the properties of binary and alloyed MgB_2 wires have been investigated before and after CHPD, using resistivity and specific heat measurements in the temperature range between 5 and 35 K at magnetic fields up to 15 T. In particular, a deconvolution of the specific heat data was used to determine the distribution of T_c in the samples.**

We have found that the effect of the densification process on the electrical and transport properties is related to the improved grain connectivity and percolation. By combining the results arising from the analysis of the T_c distribution and those from resistivity measurements, it follows that the minimum superconducting volume fraction needed for the percolation of a superconducting path is strongly reduced in samples treated by CHPD.

***Index Terms*—MgB_2, cold densification, connectivity, percolation, specific heat, T_c distribution.**

I. INTRODUCTION

THE relatively high T_c, the absence of weak-links at grain boundaries and the abundance of starting materials render MgB_2 a promising material for industrial applications, not only in view of cryogen-free devices operating between 20 and 30K, but also at 4.2K as a replacement for the more expensive Nb_3Sn wires in the field region between 9 and 12T. At present, powder-in-tube (PIT) MgB_2 wires are produced in km lengths either by the *ex situ* or the *in situ* technique. Further developments focus on the improvement of the current carrying capability at increasing temperatures and magnetic fields.

The improvement of the transport critical current of powder based MgB_2 wires can be achieved:

(i) enhancing B_{c2}, by alloying MgB_2 with Carbon in elemental form [3] or by decomposition of C-based compounds, e.g. SiC, B_4C [4,5] or of various carbohydrates, e.g. malic acid [6];

(ii) improving the grain connectivity and thus increasing the effective cross section for the conduction of transport current [7]. The improvement of connectivity is accompanied by an enhanced contact area of neighbouring grains and thus by an increased number of flux pinning centres. Low connectivity and porosity are an inherent problem of MgB_2 wires prepared by the *in situ* technique, the typical filament mass density being as low as 45% of the theoretical density (2.62 g/cm^3) due to the reduction of volume during the reaction between magnesium and boron [1].

Recently, cold high pressure densification (CHPD) was introduced at GAP in Geneva as a new route for enhancing the filament mass density of *in situ* MgB_2 wires [1,2], alternative to the high temperature densification techniques [8]. A high pressure step ($p > 1.5$ GPa) is performed on square wires at the end of the deformation process, in order to densify the Mg+B mixture before the reaction heat treatment. After reaction, this corresponds to an enhancement of the MgB_2 filament mass density up to 73% of the theoretical value.

Using this technique, a considerable enhancement of the critical current density was reported [1,2]. After applying 1.5 GPa to malic acid ($C_4H_6O_5$) added wires, the highest J_c values so far reported for *in situ* MgB_2 wires were obtained, as J_c(4.2K)=10^4 A/cm^2 at 13.8 and 13.4 T (1 μV/cm criterion) for parallel and perpendicular field, respectively [9].

The aim of the present work is the understanding of the fundamental mechanisms behind this improvement of J_c. We report on the effects of CHPD on the fundamental superconducting properties, T_c distribution, B_{c2} and B_{irr}, as well as on the parameters influencing the effective cross-sectional area for the current transport, i.e. percolation threshold and grain connectivity.

Resistivity and specific heat measurements were performed on binary and malic acid added MgB_2 wires, non-densified and after CHPD, in magnetic fields up to 15 T. Variations in connectivity and critical fields were evaluated from the $\rho(B,T)$ data. A deconvolution of the specific heat data was used to determine the distribution of T_c in the samples. This analysis, in combination with the results of the resistivity measurements, allowed us to estimate the minimum

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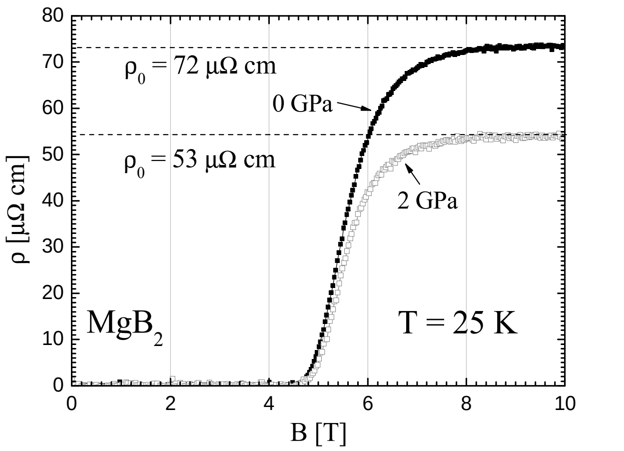

Fig. 1. Resistivity versus magnetic field at T = 25 K for the non-densified binary MgB_2 sample (solid symbols) and for the binary MgB_2 sample after densification at p = 2 GPa (open symbols). The normal state resistivity is reduced for the sample submitted to CHPD.

TABLE I Sample Specifications

Wire Sample	**Composition**	**Densification pressure** [GPa]
Binary0	Mg+2B	0
Binary2	Mg+2B	2
Malic0	Mg+2B+10wt% $C_4H_6O_5$	0
Malic2	Mg+2B+10wt% $C_4H_6O_5$	2

superconducting volume fraction needed for percolation with zero resistivity and the influence of CHPD on its value.

II. Sample Preparation

Binary and malic acid doped MgB_2 wires were fabricated in Geneva by the PIT technique using an *in situ* reaction process. The powders used for the present wires were Mg from Alfa Aesar (99%, 44 μm) and amorphous Boron (99.9%, average size 1 μm, distribution between 0.05 and 5 μm). For the binary wires preparation, the powders were mixed by a low energy ball milling process in agate balls and vials, during 1 hour at a rotating speed of 500 rpm in air.

A different procedure was used in the preparation of the precursor powders for the malic acid added wires. After dissolving 10 wt.% $C_4H_6O_5$ (malic acid, 99%) in C_7H_8 (toluene, 99.5%), the solution was mixed with an appropriate amount of B powder in a planetary mill for 1 hour (100 rpm). The slurry was dried at 150°C in a vacuum oven to form a coating around the B powder particles. This uniform composite was then mixed with an appropriate amount of Mg (99%) powder and mixed in a planetary mill (500 rpm) for 3 hours.

Precursor powders were filled into pure Fe tubes with an outer diameter of 15 mm and an inner diameter of 12 mm. After swaging to 4.2 mm and drawing to 2.75 mm, the wires were deformed by two axial rolling to obtain a 2 mm × 2 mm square cross section.

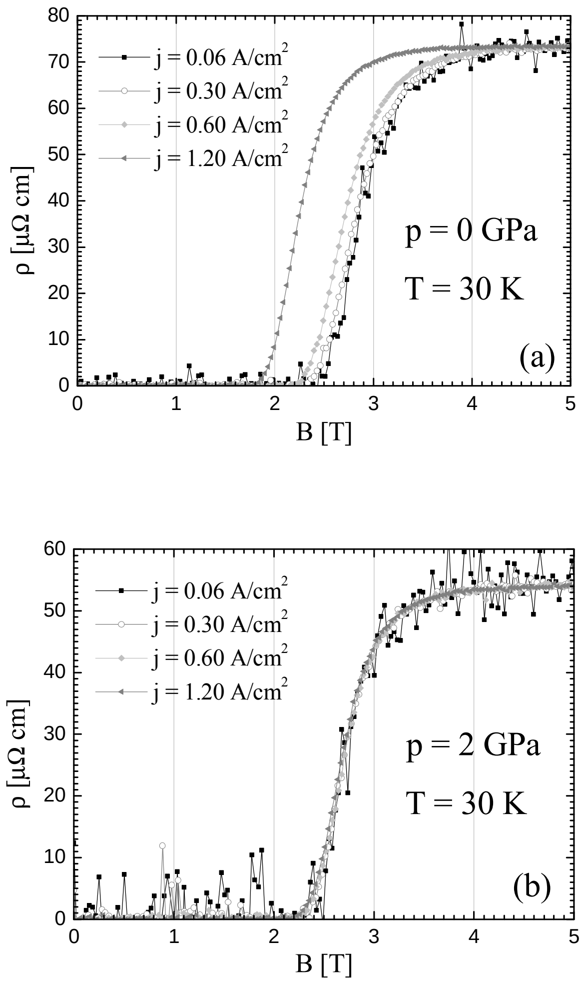

Fig. 2. Variation of $\rho(B)$ at T = 30 K as a function of the probing current density J for the non-densified binary MgB_2 sample (a) and for the binary MgB_2 sample after densification at p = 2 GPa (b). In the case of the non-densified sample, the transition is shifted at lower magnetic fields for increasing J.

CHPD was performed by means of 4 hard metal anvils acting simultaneously in a prototype cell at room temperature. Pressure up to 2 GPa was applied on the square wires by a high precision 4 columns press, the pressed length being 29 mm. After densification the shape of the wires became rectangular.

The reaction heat treatment was performed at 650°C for 1 hour for the binary wire and at 600 °C for 4 hours for the malic doped wire, in Ar atmosphere.

In order to perform resistivity and specific heat measurements, the superconducting MgB_2 cores were extracted from the Fe sheath by electrical discharge machining.

Four samples were studied in the present work, two binary MgB_2 wires from the same batch, non-densified (Binary0 in table I) and densified at p = 2 GPa (Binary2), and two 10 wt.% $C_4H_6O_5$ doped wires from the same batch, non-densified (Malic0) and densified at p = 2 GPa (Malic2). Specifications of the samples are reported in tables I and II.

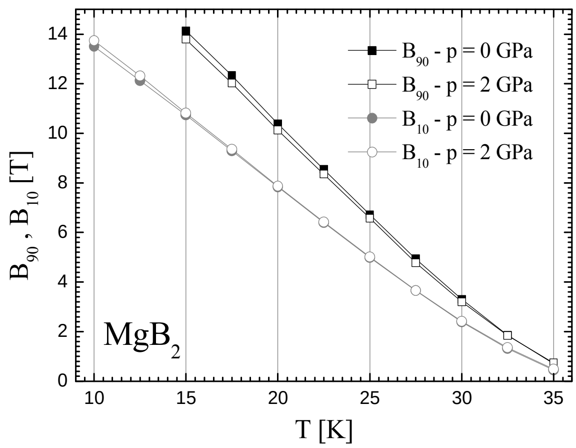

Fig. 3. Temperature dependences of B_{90} (squares) and B_{10} (circles) for the non-densified binary MgB_2 sample (solid symbols) and for the binary MgB_2 sample after densification at p = 2 GPa (open symbols). B_{90} is slightly reduced for the sample submitted to CHPD.

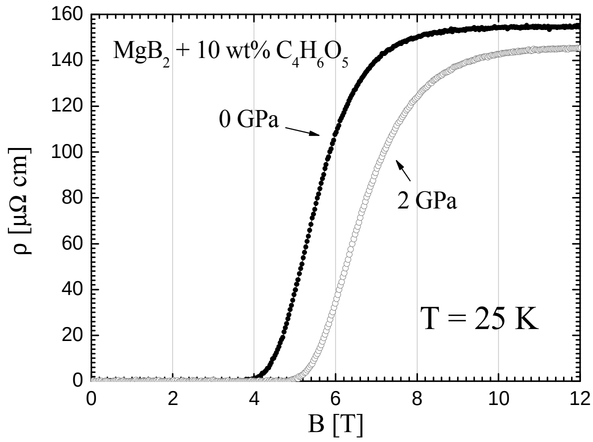

Fig. 4. $\rho(B)$ at T = 25 K for the malic acid doped samples, non-densified (solid symbols) and densified at p = 2 GPa (open symbols). Densification causes a reduction of the normal state resistivity and a shift of the transition towards higher magnetic fields.

TABLE II RESISTIVITY, RRR AND J_C

Wire Sample	ρ(40K) [μΩcm]	RRR	J_c(4.2K,10T) [A/cm^2]	J_c(4.2K,15T) [A/cm^2]
Binary0	72	1.67	910	
Binary2	53	1.73	1327	
Malic0	154	1.40		978
Malic2	146	1.33		2095

III. RESISTIVITY MEASUREMENTS

Resistivity versus field measurements were performed in magnetic fields up to 15 T, varying the temperature between 5 and 35 K. The magnetic field was swept at a rate of 0.5 T/min. while temperature was stabilized to ≤ 5 mK. The leads for the 4-point measurements were glued with silver paint on the surface of the superconducting filaments after extraction from the matrix.

Using criteria of 90% and 10% of the normal state resistivity $\rho_0 = \rho$(40K), the $\rho(B,T)$ curves were used to determine the temperature dependences of B_{90} and B_{10}, which are representative of B_{c2} and B_{irr}, respectively.

A. Binary Wires

As indicated in table II, the application of 2 GPa on the original square binary MgB_2 wire caused an enhancement of J_c at 4.2 K and 10 T by ~50%. The increase of J_c with the applied pressure is accompanied by a reduction of the normal state resistivity. Fig. 1 shows the comparison of the $\rho(B, T$ = 25 K) curves for the binary MgB_2 samples reacted in the as-drawn state and after cold densification. The probing current density J, defined as the ratio between the probing current and the total sample cross section, was 0.6 A/cm^2.

The resistivity of the MgB_2 core at the superconducting transition decreases from 72 μΩcm for the non-densified sample to 53 μΩcm for the wire densified at 2 GPa. The

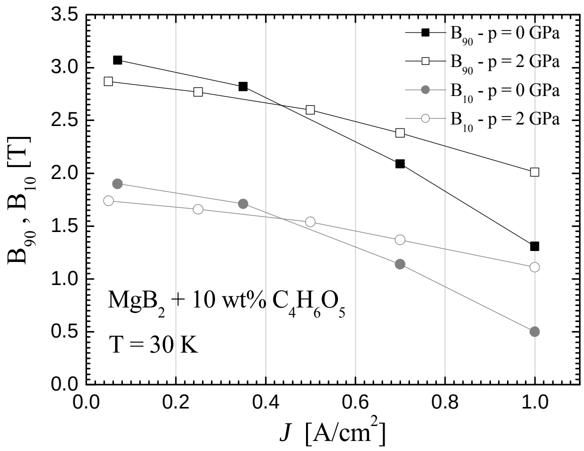

Fig. 5. Dependences of B_{90} (squares) and B_{10} (circles) at T = 30 K on the probing current density J for the malic acid doped samples, non-densified (solid symbols) and densified at p = 2 GPa (open symbols). The variations of B_{90} and B_{10} with J are less pronounced for the densified sample.

decrease of ρ with pressure represents a strong evidence of the improvement of the grain connectivity. The correlation between electric resistivity and grain connectivity is discussed in a seminal article of Rowell [10]. Rowell defines the connectivity as

$$K = \frac{\Delta\rho_{SC}}{\Delta\rho},$$

where $\Delta\rho = \rho(300K) - \rho(40K) = \rho(40K)(RRR-1)$ is the phononic term of the electric resistivity, RRR $= \rho(300K)/\rho(40K)$, and $\Delta\rho_{sc}$ is the corresponding value for a fully connected single crystal. Rowell assumes $\Delta\rho_{sc}$ = 4.482, using the single crystal results from Eltsev *et al.* [11].

According to this model, the connectivity increases by ~25% in the sample densified at p = 2 GPa, the values of K being $K(p$ = 0 GPa) = 9.3% and $K(p$ = 2 GPa) = 11.5%.

As a probe for the homogeneity of the superconducting layer, the changes in $\rho(B)$ were studied varying the measuring current density J between 0.06 and 1.20 A/cm^2. As shown in

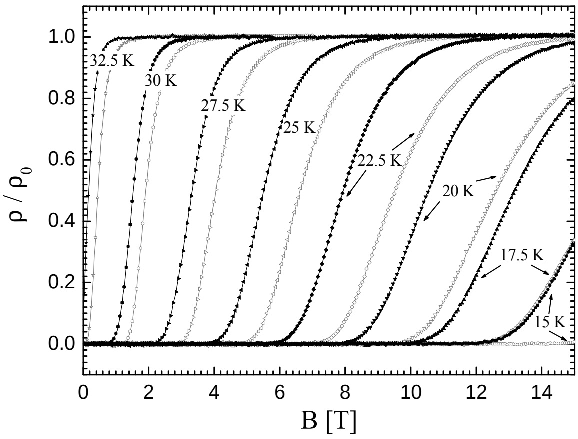

Fig. 6. Reduced resistivity as a function of magnetic field, at various temperatures between 15 and 32.5 K for the malic acid doped samples. Solid symbols correspond to the non-densified wire, open symbols to the wire pressed at p = 2 GPa.

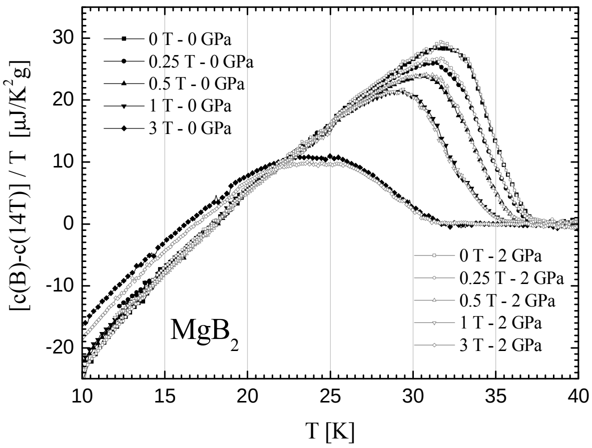

Fig. 8. Superconducting contribution to the specific heat for B =0, 0.25, 0.50, 1, and 3 T for the binary MgB_2 samples. Solid symbols correspond to the non-densified wire, open symbols to the wire pressed at p = 2 GPa.

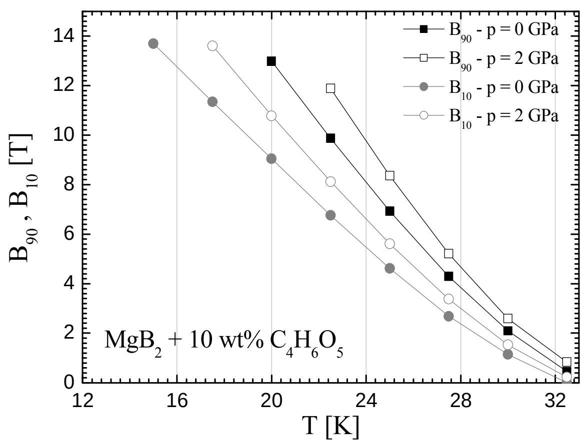

Fig. 7. Temperature dependences of B_{90} (squares) and B_{10} (circles) malic doped samples, non-densified (solid symbols) and densified at p = 2 GPa (open symbols). Both B_{90} and B_{10} are enhanced for the sample submitted to CHPD.

Fig. 2a for the non-densified sample, the superconducting transition in the $\rho(B, T = 30$ K) curve is shifted to lower magnetic fields for increasing J, thus indicating the presence of a distribution of J_c within the sample. On the other hand, the same variation of the probing current density does not influence the superconducting transition of the sample submitted to CHPD (see Fig. 2b). This increased homogeneity of the transport properties is also reflected by the current-voltage (I-V) characteristics, which exhibit a higher n value for the densified wires [9].

The comparison of the $B_{90}(T)$ and $B_{10}(T)$ curves for the two samples is illustrated in Fig. 3. After CHPD, binary MgB_2 exhibits a slightly reduced B_{90} (-2.5% with respect to the non-densified wire). This could be interpreted in the frame of the dirty limit superconductivity, taking into account the reduction of the normal state resistivity associated with the densification process.

B. *Malic Acid Doped Wires*

After densification at p = 2 GPa , the malic acid added MgB_2 wire exhibits an enhancement of J_c at 4.2 K and 15 T by >100% (see table I). In the case of doped MgB_2, densification produces two simultaneous effects on the resistivity versus magnetic field, as shown in Fig. 4:

- the resistivity at the superconducting transition is reduced, but the variation of ρ_0 is less pronounced compared to the effect on the binary sample. ρ_0 after CHPD decreases by 7% for the doped sample and by 35% for the binary MgB_2;
- the superconducting transition is shifted to higher magnetic fields, i.e. the upper critical field is increased.

These two phenomena are the consequence of the interplay of two competing mechanisms determined by densification:

- connectivity is improved, leading to the reduction of ρ_0;
- the precursor powders in the densified filaments are better packed and thus the reaction paths become shorter. In doped wires this makes the C substitution more effective, leading to a reduction of T_c, an enhancement of B_{c2} at low temperatures, accompanied by an increase of ρ_0.

Unfortunately, the combination of improved connectivity and increased C content does not allow for the doped samples the estimation of the variation in K using the Rowell method.

As already observed in the case of binary wires, the homogeneity of the superconducting layer is improved after CHPD. This is shown in Fig. 5, where the variations of $B_{90}(T$ =30 K) and $B_{10}(T$ =30 K) with the probing current J, between 0.05 and 1.0 A/cm^2, are reported for the samples reacted in the as-drawn state and after CHPD. As a consequence of the improved homogeneity and in spite of the fact that CHPD determines a reduction of T_c, the densified sample exhibits higher values of $B_{90}(T$ =30 K) and $B_{10}(T$ =30 K) for J > 0.4 A/cm^2.

Fig. 6 shows the field dependence of the normalized resistivity ρ / ρ_0 at different temperatures in the range between

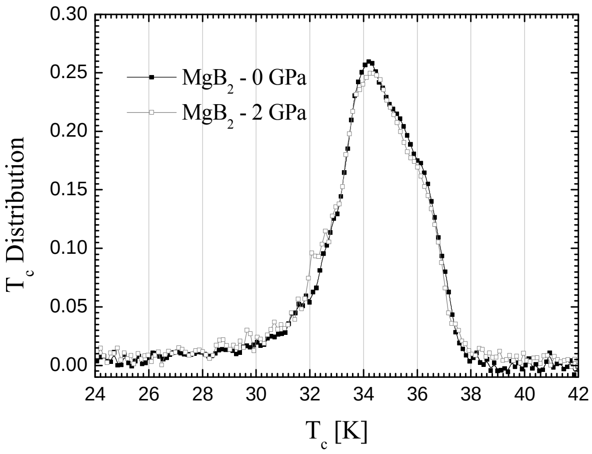

Fig. 9. Distribution of T_c obtained by the deconvolution of the calorimetric data for the non-densified binary MgB_2 sample (solid symbols) and for the binary MgB_2 sample after densification at p = 2 GPa (open symbols). It is seen that the T_c distribution is not influenced by CHPD in the case of binary MgB_2.

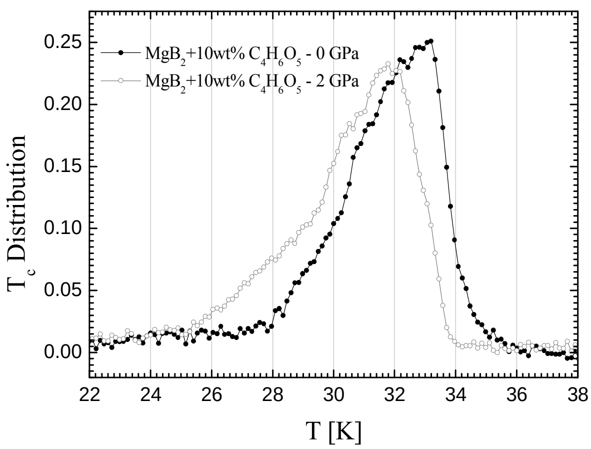

Fig. 10. Distribution of T_c for the malic doped samples, non-densified (solid symbols) and densified at p = 2 GPa (open symbols). CHPD determines a higher substitution rate of carbon, resulting in a reduced onset T_c and a broader T_c distribution.

12.5 and 32.5 K, measured at J = 0.70 A/cm^2. The resulting temperature dependences of B_{90} and B_{10} are shown in Fig. 7 for the two samples. As expected, the enhancement of B_{90}, and thus of B_{c2}, is accompanied by a large increase of the irreversibility field B_{irr}, represented by B_{10}.

IV. Specific Heat Measurements

The temperature dependence of the specific heat was measured from to 2 to 45 K for the four MgB_2 samples in table I, at various fields between 0 and 14T. Measurements were performed using the long relaxation technique [12]. A Cernox chip was employed as sample holder and thermometer/heater.

The mass of the samples was 8.786 mg (Binary0), 8.407 mg (Binary2), 8.811 mg (Malic0) and 8.065 mg (Malic2).

The calorimetric superconducting transition in the field region between 0 and 3 T was isolated from the phononic background by subtracting the specific heat curves measured at 14 T from those measured at lower fields.

The superconducting contributions to specific heat were then analysed by means of a particular deconvolution method in order to determine the distribution of T_c [13]. The advantage of calorimetry respect to other techniques is that the T_c distribution is obtained for the whole sample volume, ruling out percolation and/or shielding effects.

A. Binary Wires

The superconducting contribution to the specific heat at different magnetic fields between 0 and 3 T is reported in Fig. 8 for the two undoped samples in table I. The $c(B)/T$ curves measured on the filaments extracted from the non-densified wire (solid symbol) and from the wire densified at 2 GPa (open symbols) exhibit the same transition widths and field dependence in the field region below 3 T. In particular, from the data at B = 0 T, we determined the T_c distributions shown in Fig. 9. It follows that CHPD does not influence the width of the distribution, which in the present samples is > 8 K.

The underlying mechanism behind this broad T_c distribution in binary MgB_2 remains still unclear. MgB_2 is a line compound and thus compositional variations of B and/or Mg are not expected to influence the value of T_c. Previous measurements performed on MgB_2 bulk samples annealed at various temperatures between 650 and 1000°C [14,15] show that the width of the T_c distribution decreases with increasing annealing temperatures. The heat treatment temperature influences the grain size and thus the internal strains, arising from the random orientation of the grains and the anisotropic thermal expansion coefficients of MgB_2, as well as the lattice disorder. However, the present results are not conclusive and further experiments are being performed in order to understand the reasons for the broad T_c transition.

TABLE III Superconducting Volume Fraction

Wire Sample	Onset T_c [K]	$T(\rho = 0)$ [K]	*Supercond. Volume Fraction for Percolation* [%]
Binary0	37.4	36.2	14.9
Binary2	37.5	36.5	8.4
Malic0	34.6	33.0	25.3
Malic2	33.8	32.6	12.5

B. Malic Acid Doped Wires

Fig. 10 shows the calorimetric distributions of the critical temperature for the samples doped with 10 wt.% of malic acid. After densification the onset T_c is reduced of about 1 K. As already pointed out in Sec. III.B, CHPD causes the substitution of a higher content of C on the B sites. This explains the lower onset T_c and the slightly broader T_c distribution for the densified sample.

The conventional methods to increase the substitution level of C on the B sites involve a higher annealing temperature and/or a longer annealing time. In both cases, the higher C content is accompanied by an increase of the grain size and thus by a reduction of the grain boundary density. On the other hand, CHPD enhances the amount of substituted C, thanks to a reduced reaction path under the same reaction conditions, i.e. 600°C/4h.

V. Effects of CHPD on Percolation

A common feature observed in binary as well as in malic acid doped wires is the reduction of the electric resistivity with densification.

From the resistivity versus temperature measurements, it follows that after densification the width of the resistive transition for a given probing current density J is reduced. In table II we report the onset T_c and the temperature corresponding to $\rho = 0$, $T(\rho = 0)$, for the four samples examined in this paper and measured for $J = 0.6$ A/cm^2.

Combining the analysis of the T_c distribution with the results of the $\rho(T)$ measurements, we can obtain insights on the influence of CHPD on percolation. In particular, by integrating the T_c distributions in Fig. 9 and Fig. 10 between the onset T_c and $T(\rho = 0)$ in table III we determined for the first time the minimum superconducting volume fraction needed for a full percolation path with zero resistivity. For the non-densified binary sample, 14.9% of the filament volume needs to become superconducting in order to get percolation with zero resistivity. This fraction decreases to 8.4% after CHPD.

By analogy, percolation with zero resistivity in the malic acid doped wires is obtained for a superconducting filament volume of 25.3% in the non-densified sample. This threshold is lowered to 12.5% for the sample densified at $p = 2$ GPa.

This sizeable effect of CHPD on the percolation properties is a direct consequence of the improved connectivity between the MgB_2 grains.

VI. Summary

The cold high pressure densification method (CHPD) induces a strong enhancement of J_c in binary and doped MgB_2 wires. This process consists in submitting the wires to high pressures ($p > 1.5$ GPa) at room temperature after the final drawing and before reaction treatment. The present work was performed on binary and malic acid doped MgB_2/Fe wires in order to investigate the fundamental physical mechanisms behind the enhancement of the electrical transport properties after CHPD. The main results are the following ones:

- after CHPD, the electric resistivity is considerably reduced, reflecting an improvement of connectivity between the MgB_2 grains;
- the homogeneity of J_c in the superconducting layer is improved by densification;
- for the first time we have determined the minimum superconducting volume fraction needed for full percolation with zero resistivity. This volume fraction is strongly reduced after densification;
- in the doped wires the average C content is enhanced, thanks to the reduced reaction path of the precursor powders in the densified filaments. This is accompanied by a reduction of T_c and an enhancement of the upper critical field B_{c2}.

References

[1] R. Flükiger, M.S.A. Hossain, C. Senatore, “Strong enhancement of J_c in binary and alloyed *in situ* MgB_2 wires by a new approach: Cold High Pressure Densification”, *Supercond. Sci. Technol.*, vol. 22, 2009, 085002.

[2] M.S.A. Hossain, C. Senatore, M.A. Rindfleisch, M.J. Tomsic, J. H. Kim, S. X. Dou , R. Flükiger, “Enhancement of J_c and B_{irr} of *in situ* MgB_2 wires and tapes alloyed with $C_4H_6O_5$ (malic acid) after cold densification, *Supercond. Sci. Technol.*, vol. 22, 2009, 095004.

[3] Y.W. Ma, X.P. Zhang, G. Nishijima, K. Watanabe, S. Awaji, X.D. Bai, “Significantly enhanced critical current densities in MgB_2 tapes made by a scaleable nanocarbon addition route”, *Appl. Phys. Lett.*, vol. 88, 2006, 072502.

[4] S. X. Dou, O. Shcherbakova, W. K. Yeoh, J. H. Kim, S. Soltanian, X. L. Wang, C. Senatore, R. Flükiger, M. Dhalle, O. Husnjak, and E. Babic, “Mechanism of Enhancement in Electromagnetic Properties of MgB_2 by Nano SiC Doping”, *Phys. Rev. Lett.*, vol. 98, 2007, 097002.

[5] P. Lezza, C. Senatore, and R. Flükiger, “Improved critical current densities in B_4C doped MgB_2 based wires”, *Supercond. Sci. Technol.*, vol. 19, 2006, 1030–1033.

[6] M.S.A. Hossain, J.H. Kim, X. Xu, X.L. Wang, M. Rindfleisch, M. Tomsic, M.D. Sumption, E.W. Collings, and S.X. Dou, “Significant enhancement of H_{c2} and H_{irr} in MgB_2+$C_4H_6O_5$ bulks at a low sintering temperature of 600°C”, *Supercond. Sci. Technol.*, vol. 20, 2007, pp. L51-L54.

[7] T. Matsushita, M. Kiuchi, A. Yamamoto, J. Shimoyama, and K. Kishio, “Essential factors for the critical current density in superconducting MgB_2: connectivity and flux pinning by grain boundaries”, *Supercond. Sci. Technol.*, vol. 21, 2008, 015008.

[8] A. Serquis, L. Civale, D. L. Hammon, X. Z. Liao, J. Y. Coulter, Y. T. Zhu, M. Jaime, D. E. Peterson, F. M. Mueller, V. F. Nesterenko, and Y. Gu, “Hot isostatic pressing of powder in tube MgB_2 wires”, *Appl. Phys. Lett.*, vol. 82, 2003, pp. 2847-2849.

[9] R. Flükiger *et al.*, *IEEE. Trans Appl. Supercond.*, submitted for publication.

[10] J. M. Rowell, “The widely variable resistivity of MgB_2 samples”, *Supercond. Sci. Technol.*, vol. 16, 2003, pp. R17-R27.

[11] Y. Eltsev, S. Lee, K. Nakao, N. Chikumoto, S. Tajima, N. Koshizuka, and M. Murakami, *Phys. Rev. B.*, vol. 65, 2002, 140501.

[12] D. Sanchez, A. Junod, J.-Y. Genoud, T. Graf, and J. Muller, “Low-temperature specific heat of the 123, 124 and 247 phases of Y-Ba-Cu-O in magnetic fields up to 14 Tesla”, *Physica C*, vol. 200, 1992, pp. 1-11.

[13] C. Senatore, D. Uglietti, V. Abacherli, A. Junod, and R. Flükiger, “Specific heat, a method to determine the T_c distribution in industrial Nb_3Sn wires prepared by various techniques”, *IEEE Trans. Appl. Supercond.*, vol. 17, 2007, pp. 2611-2614.

[14] C. Senatore, P. Lezza, R. Lortz, O. Shcherbakova, W.K. Yeoh, S.X. Dou, and R. Flükiger, “Specific heat and magnetic relaxation analysis of MgB2 bulk samples with and without additives”, *IEEE Trans. Appl. Supercond.*, vol. 17, 2007, pp. 2941-2944.

[15] C. Senatore, unpublished.