

# A new generation of *in situ* MgB<sub>2</sub> wires with improved $J_c$ and $B_{irr}$ values obtained by Cold Densification (CHPD)

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**Abstract-** By means of Cold High Pressure Densification (CHPD), the critical current density,  $J_c$ , of binary and alloyed MgB<sub>2</sub> wires has been enhanced by more than a factor 2 at 4.2K and at fields up to 19 T. The relative MgB<sub>2</sub> mass density of binary MgB<sub>2</sub> wires was enhanced to ~ 54 % after applying 2.5 GPa at 300 K before reaction. In C<sub>4</sub>H<sub>6</sub>O<sub>5</sub> (malic acid) alloyed wires, densification also caused the enhancement of  $B_{irr}$ , as a consequence of a slightly enhanced C content, determined by X ray diffraction. Almost isotropic  $J_c$  values were obtained for C<sub>4</sub>H<sub>6</sub>O<sub>5</sub> added wires of 1 x 0.6 mm<sup>2</sup> cross section, the values of  $J_c(4.2K)=1 \times 10^4$  A/cm<sup>2</sup> for parallel and perpendicular fields being obtained at 13.8 and 13.4 T, respectively (1  $\mu$ V/cm criterion). The corresponding values for 20K were both close to 6.2 T. The value of  $B_{irr}$  at 20K was 11 T. The positive effects of cold densification on  $J_c$  and  $B_{irr}$  on MgB<sub>2</sub> was also observed on 150 mm long wires alloyed with C<sub>4</sub>H<sub>6</sub>O<sub>5</sub> or with SiC, by the succession of 6 overlapping pressure steps. This process can be extended to long wire lengths: by means of a newly developed prototype machine with an automatic press/release/advance sequence, a first wire length of 1 m was densified at 1.5 GPa, yielding  $J_c(4.2K) = 1 \times 10^4$  A/cm<sup>2</sup> at 13.1 T. Further improvements are expected after optimization.

**Index Terms**—MgB<sub>2</sub>, cold densification, critical current density, upper critical field, electrical resistivity.

## 1. INTRODUCTION

Alloyed MgB<sub>2</sub> wires can be produced at low costs and may be used for various applications, e.g. low field NMR magnets, the replacement of Nb<sub>3</sub>Sn wires in high field magnets in the field range between 9 and 12 T, fault current limiters and in poloidal field coils for fusion devices. A major requirement for the use of MgB<sub>2</sub> wires in all mentioned applications is a high critical current density, and important efforts are presently being undertaken for enhancing this quantity.

Today the fabrication of MgB<sub>2</sub> wires follows three main powdermetallurgical routes: the *ex situ*, the *in situ* and the *infiltration* technique. The first two - *in situ* and *ex situ* - have already been used industrially; they are described in detail in the review of Collings *et al.* [1]. The third one, the internal Mg diffusion process (IMD) (Togano *et al.* [2]) has the potential

for large scale production. The main difference between the *in situ* and *ex situ* technique resides in the choice of the precursor powder particles, which are MgB<sub>2</sub> or Mg + B, respectively. The *in situ* technique is more appropriate for applications at higher fields, due to the easier introduction of additives. In the following, our attention will be concentrated on the *in situ* technique. All wires mentioned in the present article are prepared using powders mixed by low energy ball milling.

From the wealth of published data it follows that the main requirements for optimized transport properties in MgB<sub>2</sub> wires are high purity and small size of the constituents B and Mg as well as of the Carbon based additives. The size aspect is of great importance, the reaction kinetics leading to final MgB<sub>2</sub> grains well below 100 nm [3,4]. The most appropriate initial particle size should thus ideally be in the nanosize range. However, due to the formation of MgO at the surface, submicron Mg powders are difficult to handle, while high purity nanosize B powders are still quite expensive. It is now commonly accepted that the substitution of C on the B lattice sites enhances the residual resistivity  $\rho_o$  and thus the upper critical field,  $B_{c2}$ , as well as the irreversibility field,  $B_{irr}$ , of MgB<sub>2</sub>. Thus, the enhancement of  $\rho_o$  with lattice disorder in alloyed MgB<sub>2</sub> wires is the dominant effect for the enhancement of the critical current density,  $J_c$ , at high magnetic fields.

From the work of Collings *et al.* [1], who analyzed more than 35 additives, it follows that the pinning behavior of MgB<sub>2</sub> is only little affected by the additives. This is confirmed by the relaxation data of Senatore *et al.* [5], who found for bulk samples with SiC and Carbon additives that the pinning energy  $U_o$  of alloyed MgB<sub>2</sub> is unchanged with respect to the one of the binary compound. Very recently, SiCl<sub>4</sub> additives were reported to cause enhanced pinning by Zhang *et al.* [6], but the effect is very small when compared to the enhancement of  $J_c$  at high fields due to disorder.

A completely different approach for enhancing the transport properties of MgB<sub>2</sub> wires consists in enhancing the mass density of the filaments. The approach involving densification is particularly adapted to filaments produced by powder metallurgy, which have considerably lower mass densities compared to their theoretical value: *in situ* MgB<sub>2</sub> filaments have been reported to exhibit quite low mass densities, of the order of 45 % of the theoretical value, 2.6 g/cm<sup>3</sup> [1]. Various attempts have been undertaken for enhancing the mass density of bulk MgB<sub>2</sub> under high pressure/high temperature conditions. Prikhna *et al.* [7] reacted bulk alloyed samples at T > 1'000 °C in a multianvil device under pressures  $\leq$  2 GPa, while Yamada *et al.* [8] performed hot pressing on SiC alloyed *in situ* tapes at 630°C under 100 MPa. In the bulk samples [7],

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the mass density was enhanced to values close to 100 %, yielding  $B(10^4) = 11$  T at 4.2K, where  $B(10^4)$  is the field at which  $J_c$  reaches a value of  $1 \times 10^4$  A/cm<sup>2</sup>. In the hot pressed tapes [8], the enhancement of  $J_c$  was markedly higher, the extrapolated value of  $B(10^4)^{1/2}$  being close to 14T ( $B(10^4)^{1/2}$  was unfortunately not specified). The large difference in  $J_c$  between the hot pressed bulk samples [7] and the hot pressed tapes [8] is at least partly due to the fact that the deformation of tapes by rolling leads to a certain degree of texturing. Texturing is an inherent feature of MgB<sub>2</sub> wires and tapes produced by multistep deformation. On the basis of MgB<sub>2</sub> (002) rocking curves obtained by means of synchrotron X ray diffraction, Hässler *et al.* [9] have recently reported that the tape rolling procedure creates a texture in the Mg phase, which is transferred to the MgB<sub>2</sub> crystallites during reaction. This is possible, since the reaction already starts at temperatures where Mg is still solid. The directional morphology of the Mg is always visible in cold deformed Mg+B powder mixtures, as can be seen in Fig. 1.

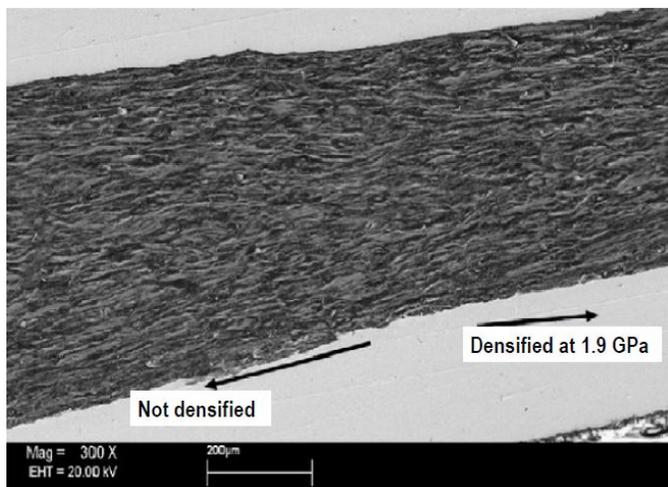


Fig. 1. SEM picture of a monofilamentary MgB<sub>2</sub>/Fe wire, as drawn (left) and densified at 1.9 GPa (right). The elongation the Mg particles is visible.

## 2. COLD HIGH PRESSURE DENSIFICATION

As large quantities of MgB<sub>2</sub> wire are necessary for industrial applications, any high temperature/high pressure processing steps are not desirable, not only from an economical point of view but also because they would limit the subsequent deformability of the wire. It is clear then that the high pressure steps should be applied at low temperature, preferably at room temperature, thus allowing winding and cabling before the reaction heat treatment. A room temperature processing technique was recently developed at GAP in Geneva, namely the Cold High Pressure Densification (CHPD) [Flükiger *et al.* [10] and Hossain *et al.* [11]]. This method is based on a prototype cell, designed for the application of high pressures at room temperature, with solid anvils acting simultaneously on all four sides of a square wire. This densification step is followed by a pressure release allowing a recovery of the wire without damage. The operation of our initial device (particularly the pressure release) is very time consuming, but it was very useful for deciding whether it was appropriate to develop an automatic

device for continuous CHPD processing of long wires (see later in this paper).

The densification step has the effect of enhancing the mass density of the unreacted Mg+B filament, letting the degree of texturing unchanged [10,11]. This is in contrast to the effect of pressing of a tape between two walls, as for example in Ref. [8]: the tape flows in the direction parallel to the pressing walls, with the consequence that the degree of texturing is enhanced, the mass density changing only slightly [7,8].

After CHPD on short samples the mass density of binary MgB<sub>2</sub> monofilaments was enhanced from 0.44% to 0.54% of the theoretical mass density after applying 2.5 GPa [10]. At the same time, a marked decrease of electrical resistance was observed on densified wires, reflecting an improved connectivity. It should be noted that the pressures applied on the 4-wall cell reached a maximum value of 6.5 GPa. At this pressure, the mass density  $d_m$  of the unreacted Mg+B powder mixture reached 96 %, while the corresponding value  $d_f$  in the reacted MgB<sub>2</sub> filament increased up to ~73% of the theoretical value [9]. However, a reproducible enhancement of  $J_c$  was only observed up to pressures of the order of ~3 GPa. It is still unknown why the higher pressures did not lead to a further enhancement of  $J_c$ .

The first cold densification experiments were carried out on monofilamentary, 45 mm long binary MgB<sub>2</sub>/Fe wires [10]. After densification at 1.85 GPa,  $J_c$  increased by 300% at 20K and 5T with respect to the as-deformed wire of the same batch. At 4.2K and 10 T,  $J_c$  was found to increase by 53%. Later on, CHPD processing was also applied to C<sub>4</sub>H<sub>6</sub>O<sub>5</sub> alloyed MgB<sub>2</sub> wires [11]: after densifying at  $p = 2.0$  GPa a  $J_c$  of  $10^4$  A/cm<sup>2</sup> at 4.2 K was observed at  $B(10^4)^{1/2} = 12.7$  T and  $B(10^4)^{1/2} = 12.5$  T, for fields parallel and perpendicular to the surface, respectively (criterion 0.1  $\mu$ V/cm). The corresponding values at 20K were 5.9 and 5.75 T, respectively, while  $B_{irr}^{1/2}$  at 20K was ~10 T [11]. These values exceed the highest reported critical current densities on *in situ* MgB<sub>2</sub> round wires prepared without pressure: Susner *et al.* [12] reported for round SiC added MgB<sub>2</sub> wires the value of  $B(10^4) = 12$  T, using the 1  $\mu$ V/cm criterion at 4.2 K.

The aim of the present article is to report further progress of the CHPD processing of alloyed monofilamentary MgB<sub>2</sub> wires with malic acid and SiC additives. In the case of binary MgB<sub>2</sub> wires, the analysis of the change in electrical resistivity for densified wires will be discussed, showing that the reason for the enhancement of  $J_c$  and  $B_{irr}$  is the enhanced connectivity.

At the end of the article, the very first results of our densification experiments at 1.5 GPa by means of a prototype automatic machine on a 1 m long C<sub>4</sub>H<sub>6</sub>O<sub>5</sub> doped wire are presented. The highest  $J_c$  achieved by means of this machine is almost the same as obtained by the pressure cell for the short wires. This shows that the transfer of the CHPD technique to industrial wire lengths is possible.

## 3. EXPERIMENTAL DETAILS

### 3.1. CHARACTERIZATION OF THE WIRES

The binary MgB<sub>2</sub> wires for the determination of the electrical resistivity were prepared at the University of Geneva using Mg from Alfa Aesar (99%, 44  $\mu$ m) and amorphous Boron (99.9%, size between 0.05 and 5  $\mu$ m). The

preparation follows the description given in Ref. 10. The ball milled powders were filled into pure Fe tubes with an outer diameter of 15 mm and an inner diameter of 12 mm. After packing, the tubes were swaged to rods of 8 mm diameter before being further deformed by turk's head rolling to wires of  $2 \times 2 \text{ mm}^2$  size. The square wires were subsequently densified at various pressures up to 2 GPa. After reaction at  $650^\circ\text{C}$  for 1 hour, the critical current density was measured and then the Fe sheath was removed by spark erosion in preparation of the electrical resistivity measurements. The final cross section of the bare  $\text{MgB}_2$  rod was  $1.5 \times 1.5 \text{ mm}^2$ . Pieces of the same wire were further deformed to a diameter of 1 mm, in order to compare their  $J_c$  values with the earlier values [10].

The alloyed wires described in this work ( $\varnothing 0.83 \text{ mm}$ ) were supplied by Hyper Tech Research, Inc., Columbus, OH, USA. The sheath material was Nb encased in Monel, while the additives were  $\text{C}_4\text{H}_6\text{O}_5$  and SiC. One of the  $\text{C}_4\text{H}_6\text{O}_5$  added wires was multifilamentary, with 18 cores. The reaction conditions were  $600^\circ\text{C}$  for 4 hours (heating rate  $2.5^\circ\text{C}/\text{min}$ ). All monofilamentary wires are prototypes and did not contain a Cu stabilization. In the present work, densification of short wires was performed under the same conditions as in Refs. 10 and 11, the sample length being 45 mm, the pressing tool having a length of 29 mm.

### 3.2. $J_c$ OF DENSIFIED $\text{MgB}_2$ WIRES WITH ADDITIVES

The transport  $J_c$  values were measured as a function of applied magnetic field up to 19 T at 4.2 K and 20 K in a He flow cryostat using a four-probe technique, with currents up to 250 A. The temperature was measured on a current lead positioned close to the sample. The voltage taps were 10 mm apart, and the voltage criterion used was in most cases  $0.1 \mu\text{V}/\text{cm}$ . The irreversibility field  $B_{\text{irr}}$  was determined using an extrapolation of the  $J_c$  vs.  $B$  curve to  $100 \text{ A}/\text{cm}^2$ . In order to study the effect of CHPD on binary  $\text{MgB}_2$  wires, the critical field values were also determined from the magneto-resistivity measurements on the filaments after removing the Fe matrix (a more detailed description is given by Senatore *et al.* [13]), the criteria being 90% and 10% of the resistance in the normal state,  $R_N$ , for  $B_{c2}$  and  $B_{\text{irr}}$ , respectively. A dc probe current density between 0.05 and  $1.0 \text{ A}/\text{cm}^2$  was used in the temperature range between 10 and 30 K. The crystal structure analysis was performed by X-ray diffraction and the lattice parameters  $a$  and  $c$  were obtained from Rietveld refinement. The microstructure of the filaments was investigated using SEM and optical microscopy.

### 3.3. MALIC ACID AND SiC ADDITIVES TO $\text{MgB}_2$

The first results about the effects of densification in  $\text{MgB}_2$  wires with 10wt.%  $\text{C}_4\text{H}_6\text{O}_5$  additives have been recently published [10]. In the meantime, the pressing parameters have been improved, leading to substantially higher  $J_c$  values, as shown in Figs. 2 and 3. The  $J_c$  values at 4.2 K are represented in Fig. 2, those at 20 and 25 K in Fig. 3, the initial dimensions of the wires before densification being the same as in Ref. 10:  $0.6 \times 1.0 \text{ mm}^2$ . From Fig. 2, the values for  $B(10^4)_{\perp}$  and  $B(10^4)_{\parallel}$  can be extracted as 13.4 and 12.9 T, respectively. The small difference between these two values confirms the almost

isotropic behavior of the critical current density in densified wires of aspect ratios  $\leq 2$  already observed by Hossain *et al.* [11]. A fully isotropic behavior is expected for wires with aspect ratios closer to 1.

Fig. 2 also shows a considerable effect of the  $J_c$  criterion: if  $1 \mu\text{V}/\text{cm}$  instead of  $0.1 \mu\text{V}/\text{cm}$  is chosen, the value of  $B(10^4)$  is raised by 0.45 T. These data are the highest published so far for *in situ*  $\text{MgB}_2$  wires. At 4.2 K and 8T, a value slightly above  $1 \times 10^5 \text{ A}/\text{cm}^2$  is measured, which corresponds to that of NbTi. The effect of pressure at 4.2 K can be summarized by saying that 1.5 GPa correspond to an enhancement of  $J_c$  by a factor 2.1 at all fields between 8 and 17 T.

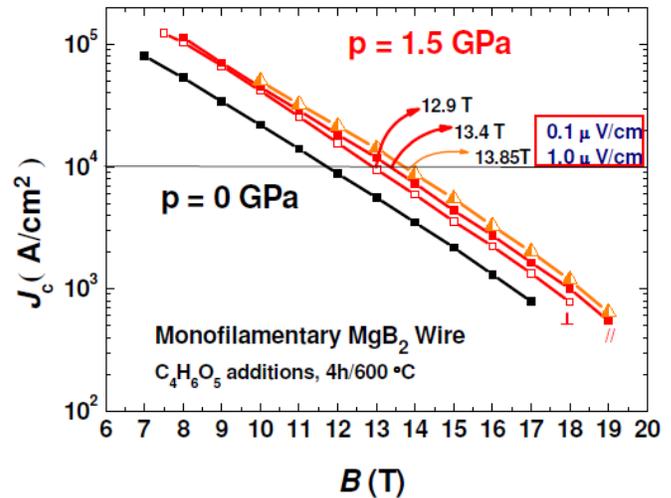


Fig. 2.  $J_c$  vs.  $B$  at 4.2 K for monofilamentary  $\text{C}_4\text{H}_6\text{O}_5$  alloyed  $\text{MgB}_2$  wires, at  $p = 0$  and after CHPD at 1.5 GPa. The  $B(10^4)$  values are 13.4 and 12.9 for  $\parallel$  and  $\perp$  fields. Note the influence of the criterion.

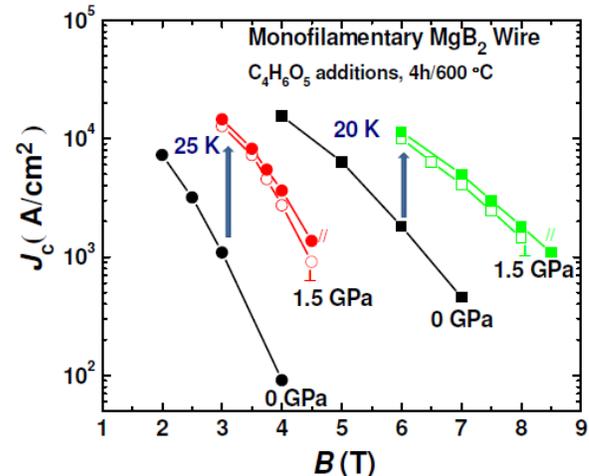


Fig. 3.  $J_c$  vs.  $B$  at 20 and 25 K  $\text{C}_4\text{H}_6\text{O}_5$  alloyed  $\text{MgB}_2$  wires, at  $p = 0$  and after CHPD at 1.5 GPa. At 20 K and 25 K, the  $B(10^4)$  values are 6.2 and 3.4 T, respectively. The  $J_c$  behavior is almost isotropic.

The effect of densification at 20 and 25 K is particularly interesting in view of MRI applications. The enhancement of  $J_c$  at 1.5 GPa is considerably higher than at 4.2 K: at 20 and 25 K, the value is enhanced by a factor 8 and 20, respectively. The behavior of  $J_c$  at 20 and 25 K is almost isotropic, as for 4.2 K, and the values for  $B(10^4)_{\perp}$  are 6.2 and 3.4 T, respectively. It is interesting that the C content in densified  $\text{MgB}_2$  wires with  $\text{C}_4\text{H}_6\text{O}_5$  additives increases, as can be seen from the variation of the lattice parameters in Table I. This

reflects that the shorter reaction path in the denser powder mixture leads to an enhanced reaction.

TABLE I  $MgB_2 + C_4H_6O_5$ : SAMPLE SPECIFICATIONS

p (GPa)	Lattice parameters (Å)		C (x) in $Mg(B_{1-x}C_x)_2$	$T_c$ (K)
	a	c		
0	3.0749	3.524	0.0232	34.6
1.5	3.0733	3.5237	0.0264	33.4
2	3.0722	3.5189	0.0293	

In view of the densification of longer wire segments, we had to ask ourselves whether the overlapping between two sequential pressing steps has a negative effect on the transport properties of the filaments. This question was studied on  $MgB_2$  wires with two different additives,  $C_4H_6O_5$  and SiC. In both cases, wire lengths of 150 mm were chosen, along which the densification was performed, with 20 mm forward motion between two subsequent pressings. The results for an applied pressure of 1.9 GPa are shown in Figs. 4 and 5. Both additives have a very similar behavior. The enhancement of  $J_c$  for this set of measurements is not as high as for a single densification step, the enhancement factors being only 1.6 and 1.4 for  $C_4H_6O_5$  and SiC, respectively. As will be shown in Fig. 8 of this article, a progress has been meanwhile accomplished in connection with the overlapping problem.

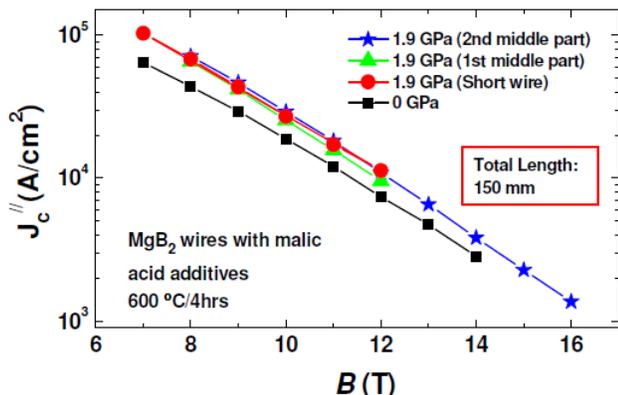


Fig. 4.  $J_c$  vs.  $B$  at 4.2 K for  $C_4H_6O_5$  alloyed  $MgB_2$  wires, at  $p = 0$  and after CHPD at 1.9 GPa. The pressure was applied sequentially, with an overlapping width of 20 mm.

### 3.4. CONFIGURATION OF DENSIFIED WIRES

Envisaging the densification of long lengths of multifilamentary  $MgB_2$  wires, it is important to know how the filament morphology and positions change after CHPD. Such a study has been started with a binary  $MgB_2$  multifilamentary wire of 0.83 mm. Fig. 6 shows the cross sections of a 18 filament wire rolled in advance to match the size of the pressure cell (1 mm), reacted without densification ( $p = 0$ ) and after densification at 1.25 GPa.

Because of the additional rolling operation to bring the round wire to a size matching the pressure cell, a larger distortion of the filaments is expected in the direction of the narrow side. The comparison between the two cross sections reflects this larger contraction, the central filaments being slightly flattened. In spite of the strong pressure and of

the initial cross section showing rounded corners, only a small change in the configuration is observed. An even smaller change should be obtained choosing more appropriate conditions for the initial cross section.

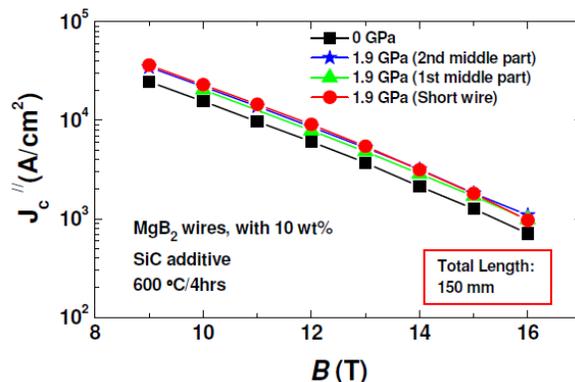


Fig. 5.  $J_c$  vs.  $B$  at 4.2 K for SiC alloyed  $MgB_2$  wires, at  $p = 0$  and after CHPD at 1.9 GPa. The pressure was applied sequentially, with an overlapping width of 20 mm.

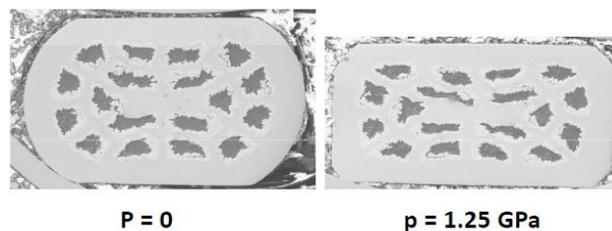


Fig. 6. Effect of densification at 1.25 GPa on the configuration of a  $MgB_2$  wire with 18 filaments.

An aspect ratio closer to 1 is expected to lead to a more homogeneous stress distribution in the wire during densification. The conditions of this present study were not ideal, the fixed width of the wire being not appropriate to the diameter of the received wires, 0.83 mm. A more detailed description of the effects on  $J_c$  of multifilamentary wires will be published at a later date.

### 3.5. THE EXPONENTIAL $n$ FACTOR AFTER DENSIFICATION

In view of MRI or low field NMR applications, the exponential  $n$  factor plays an important role. It is known that at 4.2 K and low fields,  $n$  of  $MgB_2$  wires can reach very high values. However, the strong decrease of  $n$  with field is a limiting factor when envisaging applications at higher fields. As shown in Fig. 7 for the malic acid doped wire, densification leads to a considerable increase of the  $n$  factor. The data set is not complete yet, but it can be seen that the increase of  $n$  after densification at 1.48 GPa is around 35%. For example, the field at which  $n$  takes the value 30 is higher in densified wires, the average enhancement being estimated to  $\sim 1$  T at 20K and  $\sim 2$  T at 4.2 K. This argument is of industrial importance.

Although  $n$  is an empirical factor which depends on a variety of effects which are not all well defined, it reflects in a certain way the homogeneity of a filament. The present enhancement of  $n$  can thus be interpreted as an improved homogeneity of the densified filament (this is also confirmed by the analysis of the electrical resistivity presented later in this paper (see Fig. 9)).

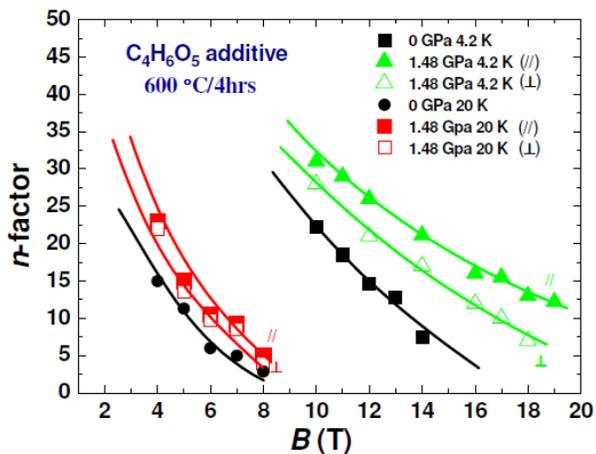


Fig. 7. Variation of the exponential  $n$  factor as a function of applied field, at 4.2 and 20 K. Note the strong increase of  $n$  at all temperatures and fields.

### 3.6. TOWARDS LONGER $\text{MgB}_2$ WIRE LENGTHS

All results reported in the preceding Section have been obtained using the same small scale densification device. Based on the promising results, a new prototype machine was constructed, which allows the densification of longer wire lengths by means of an automatic press/release/advance mechanism. The new machine takes into account the experience previously acquired with the laboratory device, but allows a considerably faster change between the various pressing and releasing operations, combined with a forward movement of the wire. A functioning but not complete version of this machine was tested very recently, the first results being represented in Fig. 8. Due to the limited amount of available prototype  $\text{MgB}_2$  wire with  $\text{C}_4\text{H}_6\text{O}_5$  additive, the first tests were restricted to a length of 1 meter. The results in Fig. 9 confirm the results on short wire samples,  $B(10^4)^{1/2}$  for 1.5 GPa being raised from 11.1 to 13.1 T.  $J_c$  was enhanced by a factor slightly above 2 over the whole field region.

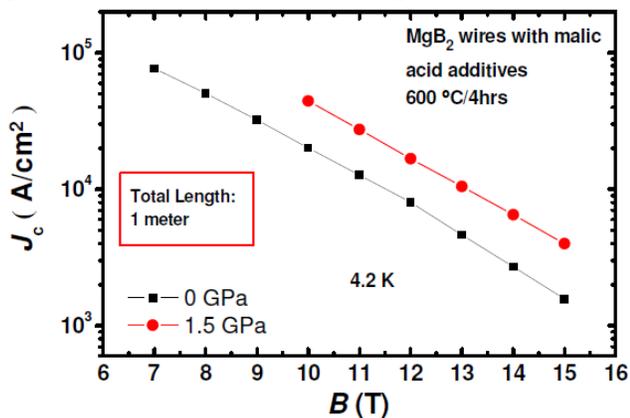


Fig. 8.  $J_c$  vs. applied field  $B$  for a monofilamentary  $\text{MgB}_2$  wire with  $\text{C}_4\text{H}_6\text{O}_5$  additions, densified with 1.5 GPa over a length of 1 m with a forward movement of 20 mm by using the new automatic press/release/advance device.

These results were obtained on various 45 mm pieces cut from the total wire length, the fluctuation being  $\leq 5\%$ . They demonstrate that a reasonable choice of the densification parameters overcomes the problems caused by the overlapping

between subsequent pressing regions. No obstacles against the densification of much longer wire lengths were encountered.

### 4. CONNECTIVITY IN DENSIFIED BINARY $\text{MgB}_2$ WIRES

The above mentioned increase of the  $n$  factor for densified  $\text{MgB}_2$  wires can be interpreted as reflecting an increase of homogeneity. However, changes due to the chemical composition are excluded in the binary compound  $\text{MgB}_2$ , due to its very narrow equilibrium phase field. A first description for the source of inhomogeneity is given by Senatore *et al.* [13], who performed a detailed characterization of densified binary and alloyed  $\text{MgB}_2$  wires by means of electrical resistivity and specific heat measurements.

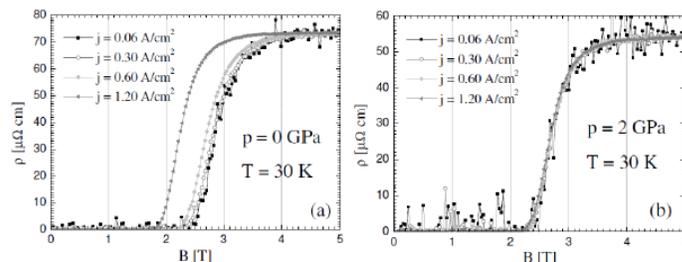


Fig. 9. Variation of  $\rho(B)$  at 30K as a function of the probing current density  $j$  for a binary  $\text{MgB}_2$  filament extracted from the Fe matrix. Left: reacted as deformed. Right: densified at 2 GPa [13].

The results for binary filaments are shown in Fig. 9, which shows a comparison between the behavior of  $\rho(B)$  for filaments of the same batch (no densification, left side) and densified at 2GPa. For reference filament, Fig. 10 shows clearly that the resistive transition depends on the probing current density. At  $T = 30\text{K}$ , the change from 0.06 to 1.20  $\text{A}/\text{cm}^2$  leads to a shift of the magnetic field by more than 0.5 T. The behavior is completely different for the densified wire, where a change of the probing current density has no effect on the transition (right side). At the same time, the electrical resistivity in the binary  $\text{MgB}_2$  filament decreases from 72 to 53  $\mu\Omega\text{cm}$  after densifying at 2 GPa, suggesting that densification improves the connectivity.

The variation of  $B_{c2}$  and  $B_{irr}$  at  $T = 30\text{K}$ , extracted from the data in Ref. [13] is shown in Fig. 10. In the range up to 1.2  $\text{A}/\text{cm}^2$ , both  $B_{c2}$  and  $B_{irr}$  do not depend on  $j$  in the densified filament, in contrast to the reference filament. The different behavior of  $B_{irr}$  and  $B_{c2}$  as function of  $j$  explains the enhancement of  $B_{irr}$  as determined with the 100  $\text{A}/\text{cm}^2$  criterion shown in our earlier article [10], where a slight increase of  $B_{irr}$  was reported for densified binary  $\text{MgB}_2$ , ( $\Delta B_{irr} = 0.5\text{T}$ ).

The situation is different for CHPD treated  $\text{C}_4\text{H}_6\text{O}_5$  alloyed wires: here, the enhancement of  $B_{irr}$  is real, and is due to the fact that the Carbon content in densified wires increases, which is proven by the decrease of the lattice parameter (see Table I).

### 5. DISCUSSION

The critical current density,  $J_c$ , of *in situ* binary and alloyed  $\text{MgB}_2$  wires has been considerably enhanced since the recent introduction of the Cold High Pressure Densification (CHPD)

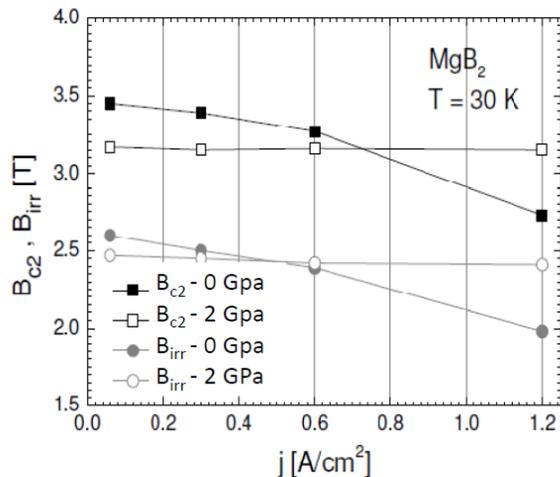


Fig. 10. Variation of  $B_{c2}$  and  $B_{irr}$  at 30K as a function of the probing current density  $j$  for a binary  $MgB_2$  filament after removal from the Fe matrix. For the densified filament,  $B_{c2}$  and  $B_{irr}$  are practically unchanged, in contrast to the behavior of the reference filament.

method [9,10], demonstrating that this method has a large potential. A reproducible enhancement of  $J_c$  was observed for pressures in the range between 1 and 2.5 GPa. Pressures up to 6.5 GPa have shown to further enhance the mass density [9], but failed in producing higher  $J_c$  values, which is attributed to a possible crack formation. From the present investigation, as well as from Refs. 10, 11 and 13, one can draw the following conclusions for wires processed by CHPD.

- After densification up to 2 GPa, a strong enhancement of  $J_c$  was observed in all measured binary and alloyed wires, at 4.2 K in the field range between 8 and 19T and at 20 K between 4 and 9 T. A correlation between mass density and  $J_c$  has been found for binary  $MgB_2$  wires. Due to the difficulties to measure the mass density of alloyed  $MgB_2$  wires, it was assumed that the mass density enhancement is similar to that of binary  $MgB_2$  wires.
- The enhancement of  $J_c$  for  $C_4H_6O_5$  alloyed  $MgB_2$  wires at 1.5 GPa and 4.2 K corresponds to a factor 2.1 at magnetic fields between 8 and 17 T. At 20 and 25 K, the values of  $J_c$  at 6 and at 3 T were enhanced by a factor 8 and 15, respectively. This result is very promising in view of future industrial applications.
- For wires with aspect ratios  $\leq 2$ ,  $J_c$  exhibits a practically isotropic behavior, with  $\Gamma = J_c^{\parallel} / J_c^{\perp} \leq 1.2$ . By changing the initial geometry, a fully isotropic behavior can be obtained.
- The electric resistivity of binary  $MgB_2$  filaments decreases substantially, thus suggesting improved connectivity. For alloyed wires, filaments have not yet been isolated, but the same behavior is expected.
- The value of  $B_{irr}$  remains essentially unchanged for densified binary  $MgB_2$  wires. The observed enhancement of  $B_{irr}$  in  $C_4H_6O_5$  alloyed  $MgB_2$  wires is due to an increase of the Carbon content in the densified wires.
- Densification leads to a sizeable enhancement of  $\geq 35\%$  of the exponential  $n$  factor of binary and alloyed wires.

## 5. CONCLUSIONS

After having obtained considerable improvements of  $J_c$  on short  $MgB_2$  wires treated by CHPD, the process has been

extended to long wires. In spite of the occurrence of overlapping pressure zones, no particular adverse effects on  $J_c$  were observed after the first experiments with the new automatic press/release/advance device. Indeed, a  $C_4H_6O_5$  alloyed wire of 1 m length, densified with an advance of 20 mm, with an overlap of 29 mm, yielded almost the same  $J_c$  values as for the short wires, the value of  $J_c$  being enhanced by a factor 2. The value of  $B(10^4)$  was 13.1 at 4.2 K, the enhancement being  $\Delta B(10^4) = 1.6$  T, a sizeable increase of the potential field range of a magnet by CHPD.

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