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Md S. Hossain
University of Wollongong, shahriar@uow.edu.au

C Senatore

M Rindfleisch
Nuclear Hyper Tech Research, Inc

R Flukiger

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Improvement of J_c by cold high pressure densification of binary, 18-filament *in situ* MgB_2 wires

M S A Hossain[1], **C Senatore**[1], **M Rindfleisch**[2] **and R Flükiger**[1]

[1] Group of Applied Physics (GAP), University of Geneva, 1211 Geneva 4, Switzerland
[2] Hyper Tech Research, Inc., Columbus, OH 43210, USA

E-mail: Rene.Flukiger@unige.ch

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Abstract

Cold high pressure densification (CHPD) has recently been found to be a promising way to improve the critical current density, J_c, in single-core MgB_2 wires prepared by *in situ* processing. In this work, the CHPD process was also applied to multifilamentary, binary MgB_2 wires, leading again to a strong enhancement of the transport J_c. The fields $B(10^4)^{\parallel}$ and $B(10^4)^{\perp}$ where the transport J_c at 4.2 K reaches the value 1×10^4 A cm^{-2} for parallel and perpendicular fields were determined as 8.5 and 8.2 T, respectively (0.1 μV cm^{-1} criterion). The behaviour of J_c versus B at 20 and 25 K was almost isotropic, the corresponding $B(10^4)^{\parallel}$ values being 4.7 and 3 T, respectively. The observed enhancement of J_c by a factor 2.3 at 4.2 K at all applied fields up to 10 T in densified samples is directly correlated with the observed enhancement of the mass density and microhardness. The decrease of the electrical resistivity of the densified wire reflects improved grain connectivity in the filaments. The improvement of J_c by CHPD was still observed after sequential pressing with overlapping regions, up to a total wire length of 150 mm. This result is promising as regards practical applications.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The transport properties of MgB_2 wires have the potential to make this material a possible solution for applications up to 20 K where costs and/or weight are relevant constraints. For large scale applications, it is imperative to explore not only the material, but also the conductor configuration for MgB_2 wires and tapes. MgB_2 superconducting wires have already demonstrated their potential for DC applications such as in MRI magnets, and the credit goes to its simple and inexpensive production process. However, further efforts have still to be made in order to extend their use to AC applications such as in current limiters, motors and transformers, a major concern being related to the reduction of AC losses. In parallel with a broad research thrust directed to achieving higher critical current densities, J_c, several manufacturers have also developed multifilamentary wires, following two different powder-in-tube (PIT) routes [1]: (a) *ex situ* processing, working with already reacted MgB_2 powders and (b) *in situ* processing, where the wire deformation is performed on metallic tubes filled with Mg and B powder mixtures.

Currently, MgB_2 based multifilamentary wires fabricated by the PIT technique can be considered as a promising industrial product. Wires with lengths exceeding 1 km are now fabricated by two manufacturers, Hyper Tech Research (HTR; *in situ* processing [13]) and Columbus Superconductors (*ex situ* processing [9]). Several doping procedures have been developed in order to enhance the values of J_c in the presence of magnetic fields [1]. Most works have been performed on alloyed wires produced by the *in situ* process [2–4], the number of works on alloyed *ex situ* wires being smaller [5]. The clear application prospect has prompted the development of various techniques for producing multifilamentary wires [6–10] and cables [11, 12]. The stabilized multifilamentary tapes produced by Columbus Superconductors have been successfully employed to realize low field MRI magnets [14] operating at 20 K, while *in situ* wires have been used as a basis for a number of

Table 1. Characterization of the binary 18-filament wires (Monel sheath, Nb/Cu barrier) before and after pressing using four hard metal anvils (650 °C/1 h).

Sheath material	P (GPa)	Wire cross section (mm × mm)	Filament cross section (mm^2)	Fill factor (%)	Mass density (Mg + 2B) (g cm^{-3})	Relative mass density (%)	Vickers microhardness (GPa)
Monel/Nb/Cu	0	0.61 × 1.00	0.0957	15.5	1.015	51	1.4
Monel/Nb/Cu	1.5	0.57 × 1.01	0.0829	14.3	1.413	69	3.6

prototype magnets. From the industrial point of view, a fundamental role is played by the material sheath and the design architecture of the conductor as they have to provide suitable mechanical properties and appropriate thermal and electrical stabilization [15, 16].

A key requirement for the use of MgB_2 wires in all aforementioned applications is a high critical current density; significant efforts are still being made to enhance this property. A new approach for enhancing the transport properties of MgB_2 wires consists in enhancing the mass density of the filaments and is particularly adapted to filaments produced by powder metallurgy. Indeed, *in situ* MgB_2 filaments have been reported to exhibit quite low mass densities, of the order of 45% [1, 20] of the theoretical value, 2.6 g cm^{-3}. Higher mass densities have so far only been reported by Togano *et al* [19], who used another route, the so-called internal Mg diffusion process. Higher mass densities have also been obtained in bulk MgB_2 upon treating at $T \geqslant 1000$ °C under pressures >2 GPa [17–20].

A sizable enhancement of the mass density in *in situ* MgB_2 wires was obtained by the cold high pressure densification or CHPD process developed in Geneva [20]. Pressures exceeding 1 GPa are applied to MgB_2 wires at room temperature, just before the final reaction heat treatment. For binary *in situ* MgB_2 wires of rectangular shape, an enhancement of J_c at 4.2 K by a factor of 2 was reported [20]. In the following, it was shown that the CHPD process could also be successfully applied to alloyed MgB_2 wires [21, 22]. However, these works were limited to monofilamentary wires. The aim of the present article is to extend the CHPD processing to multifilamentary MgB_2 wires and to longer wire lengths. Sequential densification was applied to 18-filament wires of 150 mm total length, with overlapping pressure regions.

2. Experimental details

The present work was performed on 18-filament MgB_2/Nb/Cu/Monel composite wires of 0.83 mm diameter with a Nb/Cu barrier and a Monel outer sheath, produced at Hyper Tech Research, Inc. (HTR), following the *in situ* route previously described by Sumption *et al* [7]. The densification of short wires (length: 45 mm) was performed under the same conditions as were described in [20–22], the pressure being transmitted by four hard metal anvils of 29 mm length. In order to check the applicability of the CHPD method to industrial wires, a wire of 150 mm total length was sequentially pressed, the anvils being shifted longitudinally by lengths up to 14 mm between two subsequent pressing steps. As a consequence, each wire portion was pressed twice. The parameters were chosen in such a way that subsequent pressing steps were performed on overlapping pressure zones, up to the whole length of the wire. As a consequence, the whole wire length was subjected to at least two pressing steps. The reaction heat treatment was performed at 650 °C for 1 h in an argon atmosphere at 1 bar.

The transport J_c values were measured as a function of applied magnetic field in a 17 T magnet at 4.2, 20 and 25 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 14 mm apart, and the voltage criterion used was 0.1 μV cm^{-1}. In some cases, the criterion of 1 μV cm^{-1} was used for comparison.

Due to the difficulties in extracting undamaged MgB_2 filaments from the Monel/Nb/Cu sheath, the effects of densification on connectivity were studied indirectly, by comparing the changes of electrical resistance before and after densification of wires. The microstructure of the filaments was investigated using SEM and optical microscopy. The determination of the mass density was already described previously [20] for monofilamentary wires. As shown in table 1, the relative mass density in multifilamentary wires with a Monel sheath upon densifying at 1.5 GPa was found to increase from 51 to 69%, the uncertainty being of the order of 5%. These values are markedly higher than for the previous Fe sheathed wires [20], which could be correlated with the considerably harder Monel matrix. The variation in cross section and fill factor of the present wires as well as the Vickers microhardness and mass density of the filaments with and without densification are listed in table 1. The rectangular cross sections of the same wire at the end of the drawing process and after applying 1.5 GPa have been shown in figure 1. The aspect ratio b/a after densification is 1:1.75 and the reduction of the cross sectional area of each one of the 18 filaments is clearly visible in figure 1. The fill factor after densification at 1.5 GPa had decreased from 15.5 to 14.3% (table 1). The decrease of the filament volume and thus the enhancement of the (Mg + 2B) filament mass density, d_m, in densified wires can easily be found, the Nb/Cu/Monel volume being unchanged. The mass density of the unreacted (Mg + 2B) filaments, d_m, reached 1.41 g cm^{-3} at 1.5 GPa, thus corresponding to 69% of the theoretical (Mg + 2B) density. The mass density values were determined taking into account the slight elongation of the wire upon CHPD. The microhardness values for densified filaments were found to increase from 1.4 to 3.5 GPa (mass: 50 g).

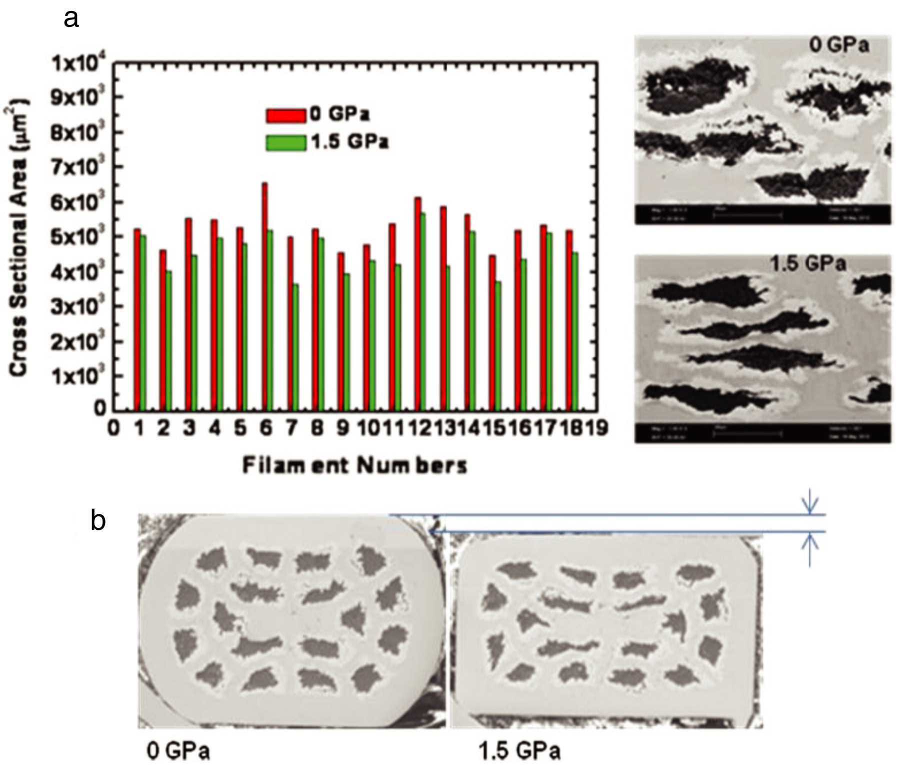

Figure 1. (a) Cross section of unreacted binary 18-filament MgB_2 wires (Monel sheath, Nb/Cu barrier) after cold densification at 0 and 1.5 GPa. All the filament cross sectional areas are noticeably reduced after applying the pressure of 1.5 GPa. (b) The width of the wires is given by the width of the pressure cell, the pressure effect being thus visible in the thickness reduction.

3. Results and discussion

The variation of the J_c values for the present multifilamentary wires at 4.2 K upon densification at 1.5 GPa is represented in figure 2, while the variation at 20 and 25 K is shown in figure 3. From figure 2, it follows that the field value at which J_c reaches the value 10^4 A cm^{-2} is strongly enhanced: for the initial wire, $B(10^4)^{\|} = 7.3$ T increased to $B(10^4)^{\|} = 8.5$ T; for the final wire, the corresponding value of $B(10^4)^{\perp}$ became 8.20 T (the dimensions of the rectangular cross sections before and after densification are given in table 1). The small difference between the two orientations confirms the almost isotropic behaviour of the critical current density in densified wires of aspect ratios $\leqslant 2$ already observed in our previous papers [20–22]. A fully isotropic behaviour is expected for wires with aspect ratios closer to 1.

Figure 2 also shows the considerable effect of the J_c criterion: for 1 μV cm^{-1} (this criterion is used in most literature data), instead of 0.1 μV cm^{-1}, the value of $B(10^4)^{\|}$ is raised from 8.4 to 8.8 T. The observed enhancement of J_c at 1.5 GPa for the present wire is higher than for the short wire: the enhancement of $J_c(p)/J_c(0)$ at 4.2 K and at all fields between 6 and 10 T was 2.3, compared to a ratio of 2 for short wire samples with an Fe matrix (see figure 2). As mentioned above, this difference could be caused by the choice of the matrix materials, but this question was not further studied here.

The effect of densification at 20 and 25 K is particularly interesting in view of the cryogen-free MRI applications. The behaviour of J_c at 20 and 25 K was found to be almost isotropic, the difference in $B(10^4)$ for $\|$ and $\perp$ fields being of the order of 0.1 T. For this reason, only the J_c values for the $\|$ orientation are represented in figure 3.

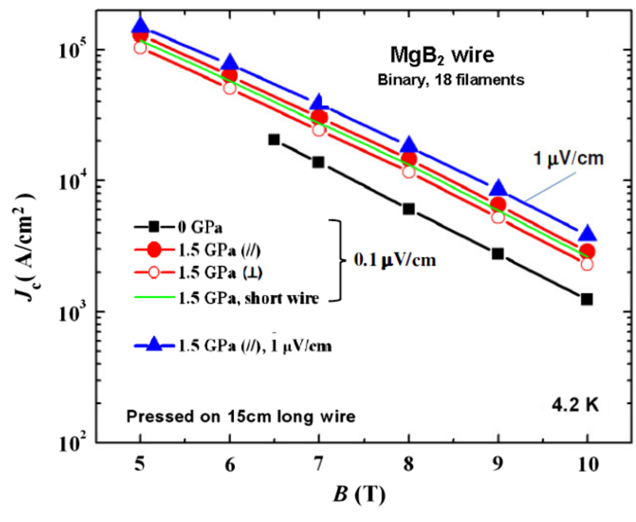

Figure 2. J_c versus B at 4.2 K for binary 18-filament MgB_2 wires, at $p = 0$ and after CHPD at 1.5 GPa. The $B(10^4)$ values are 8.48 and 8.18 T for $\|$ and $\perp$ field orientation with respect to the wider surface. Criterion for J_c: 0.1 μV cm^{-1} (triangles: 1 μV cm^{-1}).

After pressing at 1.5 GPa, the value of $J_c^{\|}$ at 20 and 25 K exhibited a strong increase, by a factor 4.5 in the field range between 4.5 and 5 T, the corresponding enhancement at 25 K

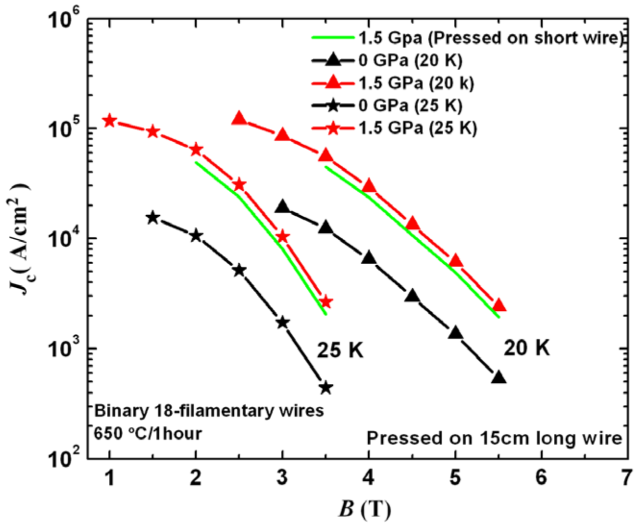

Figure 3. J_c versus B at 20 and 25 K for binary 18-filament MgB_2 wires, at $p = 0$ and after CHPD at 1.5 GPa. At 20 K and 25 K, the $B(10^4)$ values are 4.7 and 3.0 T after CHPD, respectively (only the values for $J_c\|$ have been represented, the anisotropy ratio at these temperatures being $J_c^{\|}/J_c^{\perp} < 1.05$).

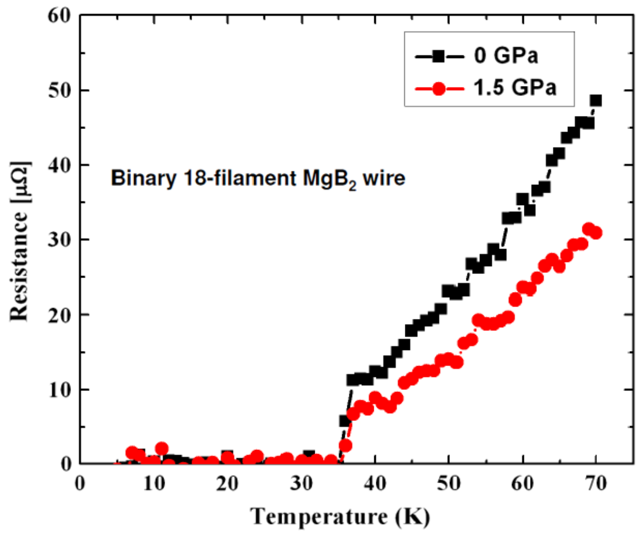

Figure 4. Variation of the electrical resistance for a binary 18-filament MgB_2 wire (Monel sheath, Nb/Cu barrier) reacted at 650 °C for 1 h, at $p = 0$ and 1.5 GPa.

at 3 T being close to 6. For comparison, the corresponding enhancement at 4.2 K is only 2.3 (figure 2). The increase of J_c at 20 K in figure 3 appears to be much stronger than that at 4.2 K (figure 2), but is in reality due to the fact that the measurements at 4.2 and 20 K were performed at very different values of the reduced field $B/B_{irr}(T)$.

In figures 2 and 3 the behaviour of J_c versus B for 150 mm long wires is shown, together with the values obtained for short lengths. The present results demonstrate that within the indicated pressure range, overlapping pressure regions do not damage the filaments in a multifilamentary configuration, the J_c values being even slightly higher than for short wires. This confirms earlier results on monofilamentary MgB_2 wires [20] and is promising as regards the extension of the CHPD process to industrial wire lengths.

The multifilamentary configuration does not allow determining a quantitative value for the connectivity, defined by Rowell [23]. The temperature dependence of the electrical resistance was measured for the two samples, non-densified and densified at 1.5 GPa, keeping the distance between the voltage taps at 10 mm (figure 4). By analogy with the data obtained on monofilamentary MgB_2 wires, the observed decrease of the normal state resistance upon densification is attributed to a reduction of the MgB_2 filament resistivity and thus to enhanced grain connectivity. The decrease of the electrical resistance for densified wires shown in figure 4 confirms the previous observation for monofilamentary binary *in situ* MgB_2 wires [20]. It also confirms the observation of Matsushita *et al* [24] that the electrical resistivity $\rho_{40\ K}$ of bulk MgB_2 samples decreased upon applying a pressure of 0.1 GPa. Table 2 shows the enhancement of the fields $B(10^4)^{\|}$ (field parallel to the wider wire surface) with increasing densifying pressures at 4.2, 20 and 25 K, thus confirming the behaviour observed earlier for monofilamentary MgB_2 wires [20].

Table 2. Values of the fields $B(10^4)^{\|}$ at 4.2, 20 and 25 K (field parallel to the wider surface) for rectangular wires of binary, *in situ* MgB_2 wires with 18 filaments, pressed using four hard metal anvils at 1.5 GPa and reacted at 650 °C for 1 h. The values $J_c(p)/J_c(0)$ represent typical values of the enhancement of J_c at each temperature for the densified wire.

	$B(10^4)^{\|}$ (T)		
p (GPa)	4.2 K	20 K	25 K
0	7.3	3.6	2.05
1.5	8.5	4.7	3.05
$J_c(p)/J_c(0)$	2.3	4.5	6.5

Recent specific heat measurements by Senatore *et al* [25] showed that the T_c distribution in binary MgB_2 wires is not altered by the densification process, in contrast to the J_c distribution in the filaments [22, 25], the latter reflecting an improvement of the homogenization. This was demonstrated by comparing the values of B_{90} and B_{10} deduced from the electrical resistivity $\rho(B)$ at probing currents J varying between 0.06 and 1.20 A cm^{-2}. The superconducting transition in the $\rho(B)$ curve was found to be shifted towards lower magnetic fields for increasing J, thus reflecting the presence of a wider distribution of J_c within the non-densified sample. This is in contrast to the behaviour in the same wire densified at 2 GPa [22], where the superconducting transition was not influenced by the same variation of the probing current density. The 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 [22].

Along with transport current density measurements, the corresponding exponential n values for the present 18-filament wires have been determined with and without densification at different operating temperatures. As shown in figure 5, densification leads to a considerable increase of the n factor, similarly to the case for the MgB_2 wires alloyed with malic acid described earlier [22]. The data set is not complete yet, but

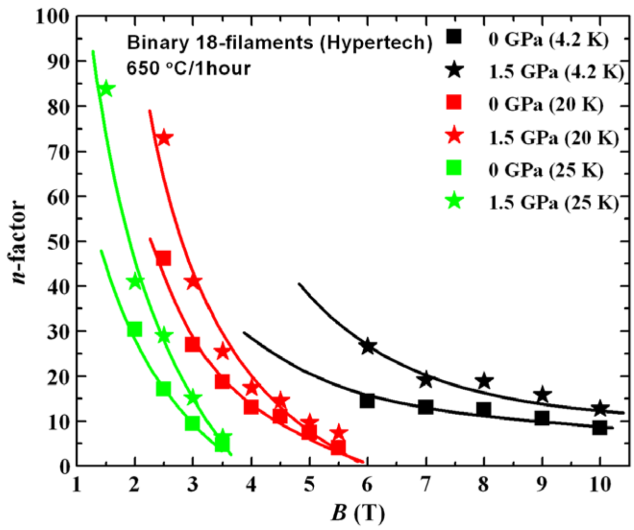

Figure 5. Variation of the exponential n factor as a function of applied field, at 4.2, 20 and 25 K. The value of n increases at all temperatures and fields upon CHPD. The factor n was defined based on the criterion 0.1 μV cm^{-1}.

an increase of n of around 30% is observed after densification at 1.5 GPa at all the operating temperatures. This argument is of industrial importance in view of the MRI and low field NMR applications.

4. Conclusions

We have demonstrated that the cold high pressure densification process (CHPD) leads to a considerable enhancement of the critical current density, J_c, of binary 18-filament MgB_2 wires produced by the *in situ* technique. The value of $B(10^4)$ for the present multifilament filament binary wire was shifted by 1.2 T, to 8.50 T at 4.2 K (0.1 μV cm^{-1}). Subsequent pressing steps with overlapping pressure zones have been performed up to a wire length of 150 mm without any degradation with respect to short wire lengths: the value of J_c at 4.2 K for the densified wires was still enhanced by a factor >2, regardless of the applied field. This result is promising as regards the application of the CHPD process to wires several hundred metres in length.

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