Pressure-induced property improvement of magnesium diboride wire

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Abstract. The 4.2 K non-barrier transport $J_c$ values of the powder-in-tube (PIT) in-situ MgB$_2$ wires have been enhanced by cold high pressure densification (CHPD). With respect to the control wire, the 1.5 GPa pressure induced the $J_c$ improvement of the wire at 4.2 K and 10 T by 25%. The $J_c$ enhancement induced by the CHPD may result from two aspects. First, the CHPD resulted in the reduction of the transverse MgB$_2$ core area. Second, the grain connectivity of the PIT in-situ MgB$_2$ wire was improved by the CHPD, which is reflected by the reduced porosity.

1. Introduction

Compacted-powder pellets and the cores of powder-in-tube (PIT) wires are inherently porous. Thus the packing densities of sintered ex-situ processed wires, based geometrically on the packing density of a uniform-sized starting powder fill, would be around 65% (although powders of mixed sizes will fill space more efficiently). The expected density of in-situ processed material is complicated by shrinkage associated with the Mg+2B→MgB$_2$ reaction. As a result it can be shown that a mixture of equi-sized Mg and B particles will after reaction lead to an MgB$_2$ density of 49%, although the use of coarse Mg particles could raise the final density to 62% [1]. Porosity in the starting powder fill (and staring porosity plus reaction in the case of in-situ material) impedes current flow and prevents the $J_c$ of the bulk or wire from attaining its intrinsic intragranular value. Current transport between grains may be reduced by the presence of insulating partial coatings [2]. These effects can combine to yield very low connectivities [3], for example 20~40% [1] which would offer the possibility of transport $J_c$ increases by factors of 5~2.5. The problems of porosity and intergranular coatings call for separate solutions. Many researchers have addressed the former and, as reviewed below, have attempted to squeeze out porosity by applying high pressures to bulk and wire samples both during the reaction heat treatment as well as prior to it.

Shields et al. [4] reported that hot isostatic pressing (HIP) induced the magnetic critical current densities as high as 1.3 $\times$ 10$^6$ A/cm$^2$ at 0 T and 9.3 $\times$ 10$^5$ A/cm$^2$ at 1 T of the MgB$_2$ bulks at 100 MPa and 950 °C, 20 K. Serquis et al. [5] presented that the $J_c$ values of HIP – processed PIT MgB$_2$ wires reached 10$^6$ A/cm$^2$ at 5 K and 0 T and the order of 10$^4$ A/cm$^2$ at 1.5 T and 26.5 K. Susner et al. [6] reported that a MgB$_2$ monofilamentary strand hot-pressed by 458 MPa achieved the $J_c$ value of 1.81 $\times$ 10$^5$ A/cm$^2$ at 4.2 K and 5 T, with respect to 0.86 $\times$ 10$^5$ A/cm$^2$ of the unpressed strand.
Cold high pressure densification (CHPD) has been considered as a method for improving the grain connectivity of the PIT in-situ MgB\textsubscript{2} wire as evidenced by the enhancements of relative mass density, electrical resistivity and transport \(J_c\) [7, 8]. Flükiger and Hossain [7] indicated that the CHPD induced the \(J_c\) improvement by 53 % at 4.2 K and 10 T for a binary Fe/MgB\textsubscript{2} round wire. Flükiger et al. [9] also presented that the \(J_c\) value of monofilamentary in situ MgB\textsubscript{2} wire doped with C\textsubscript{4}H\textsubscript{6}O\textsubscript{5}, heat-treated with 4.0 h/600 °C, and cold-densified by the pressure of 1.5 GPa reached about \(4.3 \times 10^4\) A/cm\(^2\) at 4.2 K and 10 T, with respect to \(2.2 \times 10^4\) A/cm\(^2\) of the control wire. In this paper, the influence of the CHPD on the current-carrying capacity of the 2 % - C doped PIT in-situ MgB\textsubscript{2} wires was investigated. The MgB\textsubscript{2} wire heat-treated by 1.0 h/675 °C and cold-densified by the pressure of 1.5 GPa was successfully prepared with 4.2 K non-barrier transport \(J_c\) value of \(3.6 \times 10^4\) A/cm\(^2\) at 10 T and the order of \(10^5\) A/cm\(^2\) at 7 T.

2. Experimental
2.1 Sample Preparation
Several coils of 2 % carbon-doped unreacted PIT in-situ MgB\textsubscript{2} wires were manufactured and provided by the Hyper Tech Research, Inc. The outer sheath and chemical barrier of the MgB\textsubscript{2} wire are copper and niobium, respectively. The outer diameter of the MgB\textsubscript{2} wire is 0.834 mm; the fill factor of MgB\textsubscript{2} core is 25.7 % and the ratio of Mg:B is 1:2.

The wires to be pressed were held between the jaws of a six-inch Kurt vice. The jaws were faced with blocks of heat treated M2 steel and plates of tungsten carbide as shown in Figure 1. Held between this assembly was the wire to be pressed and upper and lower blades of tool steel 0.75 mm thick. Also located between the jaws was a 10 ton hydraulic short-body ram. Vertical pressure was applied to the tool-steel blades, and hence the subject wire, by a 20-ton hydraulic ram. Vertical pressures of 1.0, 1.5, and 2.0 GPa were applied to the wires which at the same time were exposed to 2.0 GPa horizontal pressure, Table 1.

Figure 1. The assembly used for applying pressures on the MgB\textsubscript{2} wires. The directions of vertical pressure and horizontal pressure were defined.
During vertical pressing there is a tendency for the wires to expand horizontally and to extrude past the blades as shown in Figure 2.

A fixed 2.0 GPa horizontal pressure continually applied prevented this from happening and enabled clean rectangular cross sections to be maintained, Figure 3.

All the wires were encapsulated in quartz tubes under 200 torr Ar and heat treated for 1h/675°C.

![Figure 2](image1.png)  
**Figure 2.** The extrusion of the wire vertically cold-pressed by 2.0 GPa.

![Figure 3](image2.png)  
**Figure 3.** Transverse cross sectional areas of the MgB₂ wires (A) W00 (B) W10 (C) W15 (D) W20.

| Sample No. | Vertical Pressure (GPa) | Horizontal Pressure (GPa) |
|------------|------------------------|---------------------------|
| W00        | 0.0                    | 0.0                       |
| W10        | 1.0                    | 2.0                       |
| W15        | 1.5                    | 2.0                       |
| W20        | 2.0                    | 2.0                       |

2.2 Measurement
The transport critical current densities of the wires were measured as a function of applied magnetic fields up to 12 T at 4.2 K in a pool of boiling liquid helium by the four-probe technique. The wires were 5 cm long, the gauge length between voltage taps was 5 mm, and the voltage criterion was 1.0 µV/cm.

3. Result and Discussion
3.1 Transport Result
Figure 4 shows the 4.2 K non-barrier transport critical current densities of the cold-pressed and control wires as a function of applied magnetic field up to 12 T. All cold-densified wires have $J_c$ values above $3.0 \times 10^4$ A/cm² at 10 T. Compared with control wire, the 4.2 K non-barrier transport $J_c$ values of W10
(1.0 GPa cold-pressed wire) and W15 (1.5 GPa cold-pressed wire) have been enhanced at all applied magnetic fields; for example, the 4.2 K, 10 T non-barrier transport $J_c$ values of the W10 and W15 are 15% and 25% higher than that of W00. The best $J_c$ values were obtained by the wire W15 at all applied magnetic fields and are found to be $3.6 \times 10^4$ A/cm$^2$ at 10 T and $1.1 \times 10^5$ A/cm$^2$ at 7 T.

![Figure 4. Non-barrier transport critical current density $J_c$ of the MgB$_2$ wires as a function of applied magnetic field at 4.2 K.](image4)

![Figure 5. Average transverse MgB$_2$ core area of the PIT in-situ MgB$_2$ wire with error bars versus applied pressure.](image5)

![Figure 6. Average $n$-values of the MgB$_2$ wires with error bars as a function of applied magnetic field at 4.2 K.](image6)

![Figure 7. Average porosity of the PIT in-situ MgB$_2$ wire with error bar versus applied pressure.](image7)
The $J_c$ enhancements of the cold-pressed MgB$_2$ wires are partially due to the reduction of the transverse MgB$_2$ core area induced by the CHPD as shown in Figure 5. The pressure of 2.0 GPa led to the largest reduction of the transverse MgB$_2$ and was found to be 5.5 %. If the reduced transverse MgB$_2$ core area is the unique factor to inducing the $J_c$ enhancements of the MgB$_2$ wires, the $J_c$ of W20 will be improved by a factor no higher than 5.8 %. However, the $J_c$ enhancement of W20 was at least 10.4 % at the range of 7 to 12 T indicating that the CHPD has the potential of improving other properties of the PIT in-situ MgB$_2$ wire as well as reducing transverse MgB$_2$ core area leading to the enhancements of its $J_c$ values. It can be seen that the 4.2 K non-barrier transport $J_c$ values of W20 were lower than those of the W15 at all applied magnetic fields suggesting the too large pressure might...
introduce defects in the MgB$_2$ wire and therefore induced the $J_c$ degradation of the MgB$_2$ wire. W20 had the 4.2 K non-barrier transport $J_c$ values of $3.4 \times 10^4$ A/cm$^2$ and $1.1 \times 10^5$ A/cm$^2$ at 10 T and 7 T, respectively, which were still higher than those of W00. Figure 6 shows the 4.2 K $n$-values of the cold-densified and control wires as a function of applied magnetic field up to 12 T. The largest error in the measurement of $n$-value is 11%. At 10 T the cold-densified wire has a 34% decrease in $n$-value, but the $n$-value of W00 is close to those of the cold-densified wires at 8 T. The field dependence of $n$-value suggests that the CHPD might not induce higher level of extrinsic defects in PIT in-situ MgB$_2$ wire.

![Image](image1.png)

**Figure 12.** The longitudinal cross section area of W00.

![Image](image2.png)

**Figure 13.** The longitudinal cross section area of W15.

### 3.2. Microstructures

Figure 7 shows the average porosity of the wire versus the applied pressure. It can be seen that the wire processed by the CHPD have lower porosity than the control wire. Figure 8 - Figure 11 demonstrate the SEM images with secondary - electrons (SE) and back – scattered electrons (BSE) modes of the transverse MgB$_2$ core area of W00 and W15. In the BSE SEM image as in Figure 9 and Figure 11, the dark parts represent the voids existing in the wires and light parts represent the MgB$_2$ stringers in the wires. The SEM images in BSE mode were taken under same brightness and contrast. Consequently, the porosity of the wire can be obtained by calculating the area fraction of the dark parts in the BSE mode. The SEM images in SE mode as shown in Figure 8 and Figure 10, there exists some large voids in W00, but no large void was observed in W15. Based on these results, it can be concluded the CHPD has the ability to squeeze out the porosity of the PIT in-situ MgB$_2$ wire suggesting the enhanced grain connectivity of the MgB$_2$ wire. On the other hand, the CHPD has the limited ability to further reduce the porosity of the wire when the applied pressure was larger than 1.0 GPa. For example, the reduction of the average porosity of the MgB$_2$ wire was only 8.5 % when the applied pressure was increased from 1.0 GPa to 2.0 GPa; however, it was reduced by 35.6 % when the applied pressure was increased from 0.0 GPa to 1.0 GPa. The longitudinal cross section areas of W00 and W15 are shown in Figure 12 and Figure 13. Susner et al. [10] described that continuous MgB$_2$ stringers in PIT in-situ wire were aligned along drawing direction and elongated voids which were left
behind by melted Mg powder. It can be seen that the CHPD does not alter the morphology of the continuous MgB$_2$ stringers in the PIT *in-situ* MgB$_2$ wire.

4. Conclusions
Cold high pressure densification can improve the 4.2 K non-barrier transport $J_c$ of the PIT *in-situ* MgB$_2$ wire in present paper. The best 4.2 K non-barrier transport $J_c$ performance was obtained by W15 at all applied magnetic fields. The $n$-values of all cold-densified wires are above 30 when the applied magnetic field is below 9 T. The pressure dependence of the porosity of the PIT *in-situ* MgB$_2$ wire was discussed. The reduced porosity of the MgB$_2$ wire processed by the CHPD reflects that the $J_c$ enhancement induced by the CHPD might result from the improved grain connectivity of the PIT *in-situ* MgB$_2$ wires.

5. References
[1] Collings E W Sumption M D Bhatia M Susner M A and Bohnenstiehl S D 2008 *Supercond. Sci. Technol.* 21 103001
[2] Jiang J Senkowicz B J Larbalestier D C and Hellstrom E E 2006 *Supercond. Sci. Technol.* 19 33-6.
[3] Yamamoto A Shimoyama J Kishi K and Matsushita T 2007 *Supercond. Sci. Technol.* 20 658-66.
[4] Shields T C Kawano K Holdom D L and Abell J S 2002 *Supercond. Sci. Technol.* 15 202-5.
[5] Serquis A Civale L Hammon D L Liao X Z Coulter J Y Zhu Y T Jaime M Peterson D E Mueller F M Nesterenko V F and Gu Y 2003 *Appl. Phys. Lett.* 82 2847-9.
[6] Flükiger R Hossain M S A and Senatore C 2009 *Supercond. Sci. Technol.* 2 085002.
[7] Susner M A Sumption M D Bhatia M Tomsic M J Rindfleisch M A Collings E W 2008 *AIP Conference Proceedings* 986 375.
[8] Hossain M S A Senatore C Flükiger R Rindfleisch M A Tomsic M J Kim J H and Dou S X 2009 *Supercond. Sci. Technol.* 22 095004
[9] Flükiger R Hossain M S A Kulich M and Senatore C 2011 *Physica C.* 471 1119-23.
[10] Susner M A Daniels T W Sumption M D Rindfleisch M A Thong C J and Collings E W 2012 *Supercond. Sci. Technol.* 25 065002.