High Pressure Research
Vol. 00, No. 00, Month 200x, 1–6

This is an Author's Accepted Manuscript of an article published in High Pressure Research: An International Journal, DOI:10.1080/08957959.2012.722211, published 10 Sep 2012, [copyright Taylor & Francis], available online at: http://dx.doi.org/10.1080/08957959.2012.722211

Improvement of critical properties of undoped, multifilamentary MgB_2 wires in Nb/Cu after annealing under high gas pressure

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(*Received 00 Month 200x; in final form 00 Month 200x*)

Raw MgB_2 wires have been manufactured by Hyper Tech Inc. Each of their 18 cores contained undoped in situ powder surrounded by a Nb barrier and placed in a Cu sheath, cores were coated in a common Monel sheath. Wires of diameters 0.63 mm and 0.83 mm were annealed under high argon pressure (up to 1 GPa). Parameters of such HIP process (temperature, time, pressure) were varied in order to determine their optimal values.

Superconducting properties of such samples were investigated by means of four probe critical current j_c measurement. Bitter magnet producing magnetic field up to 14 T was used. It provided place for samples 70 mm long in parallel field and 20 mm long in perpendicular field. Results include critical current j_c and pinning force density F_p dependencies on magnetic field as well as Kramer plots. Critical current of $10^4 A/cm^2$ was achieved at 12 T magnetic field.

SEM pictures of wires cross sections were also taken to determine quality of Nb barrier and microstructure of superconducting material.

Keywords: Superconductivity, superconducting wires, magnesium diboride, high pressure.

1. Introduction

Superconducting wires of MgB_2 are presently one of the most promising solutions [1] for such devices as MRI, inductive heaters, flywheels, ITER and many other where superconducting magnets are needed. The main advantages of MgB_2 wires is low cost and high availability of Mg and B, simple composition, ease of large scale production. Operation temperature of about 20 K allows usage of such wires in liquid hydrogen or cryocoolers, avoiding need of liquid helium.

High pressure helps to densify core material, which is essentially important [3],[4] as volume of Mg and B is reduced by ca. 25% during reaction to MgB_2. Moreover, a solid state reaction is possible with high pressures, which leads to smaller grain sizes and better pinning capabilities of final superconducting cores [5].

Hyper Tech wires of undoped *in situ* powder with 18 cores surrounded by Nb barriers and sheathed in Cu [2], then in Monel, were annealed under high Argon pressure. Various pressures, temperatures and times of heat treatment were used in order to determine various possibilities for annealing. A novel approach for temperature progress was tested – annealing in slowly increasing temperature. At first, in lower temperature, small MgB_2 grains are formed, that later can prevent

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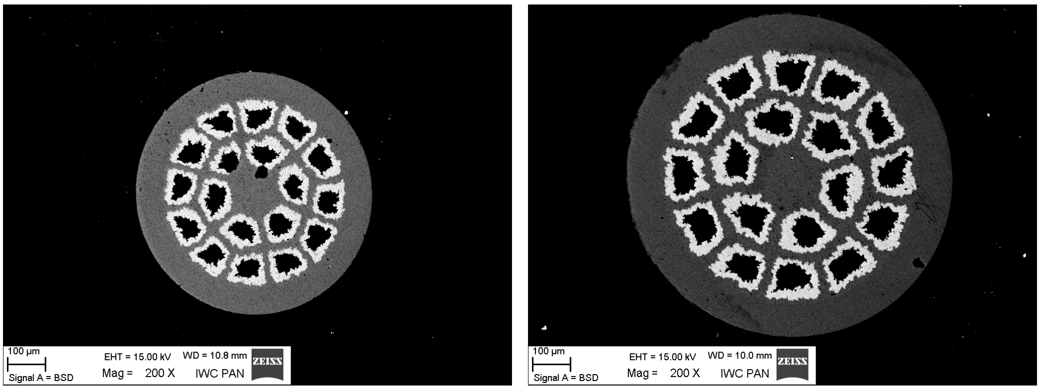
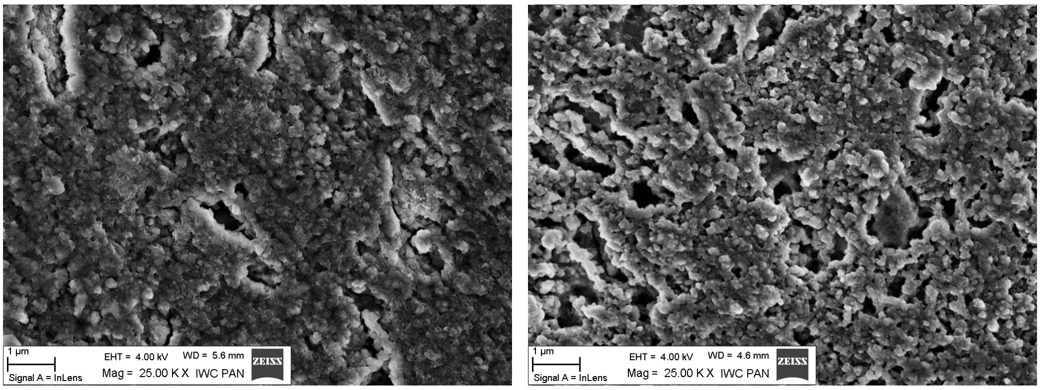
ISSN: 0895-7959 print/ISSN 1477-2299 online

DOI: 10.1080/08957959.YYYY.xxxxxx
http://www.informaworld.com



(a) (b)

Figure 1. SEM images of raw wires with (a) d=0.63 mm and (b) d=0.83 mm. 200 x magnification



(a) (b)

Figure 2. SEM images of raw wires with (a) d=0.63 mm and (b) d=0.83 mm. 25 k x magnification

growth of large grains in higher temperatures. Additionally, finishing the process in higher temperature provides a full reaction of all *in situ* material, that in lower temperature could be achieved only by a significant increase of time of annealing.

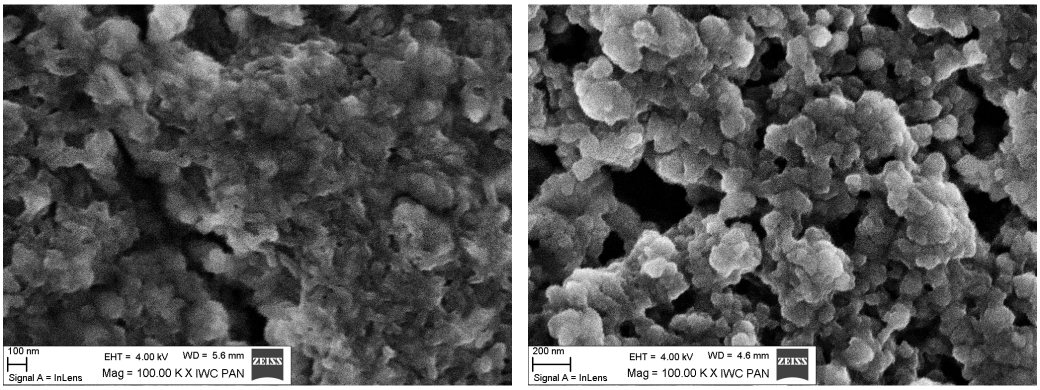
2. SEM results

Raw wires were investigated via SEM imaging to determine their general composition, Nb barrier continuity, powder density and grain sizes. Three magnifications (200x, 25kx and 100kx) are shown separately for wire of diameter d=0.63 mm and diameter d=0.83 mm (Fig. 1, 2 and 3). On pictures one can distinguish *in situ* material (darkest areas), Nb barrier (lightest areas) and sheaths as a whole. Copper and Monel parts of sheath are indistinguishable.

The SEM pictures show that barrier is sometimes broken in the thinner wires (Fig. 1a), which rather does not occur in the thicker ones. Apart from that, powder in the thinner wires is less porous (compare Fig. 2a and 2b), this should lead to a higher critical current density. The grain sizes are not affected by the wire diameter and are of about 100 nm.

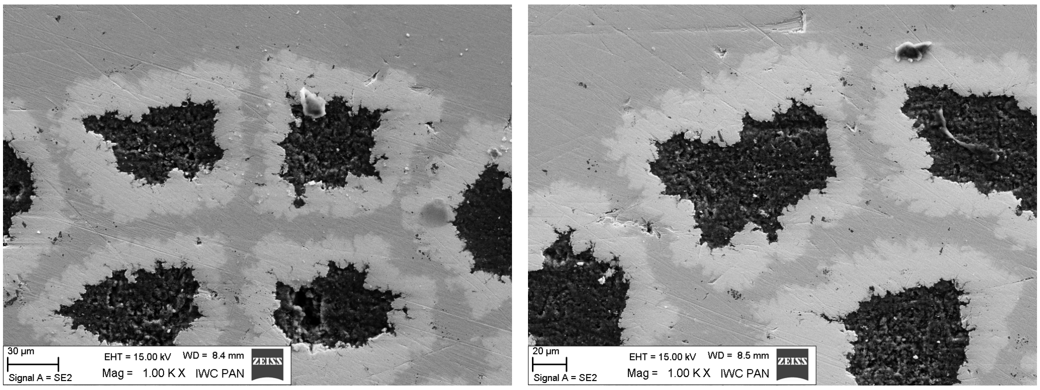
Samples after annealing under high pressure have dense superconducting cores, difference in density between wires of d=0.63 mm and d=0.83 mm is no longer visible (Fig. 4 and 5). An advantage of HIP annealing is that grain sizes (compare Fig. 3 and 5) did not increase during the heat treatment.

When chemical Nb barrier contains defects, reactions between the core and the



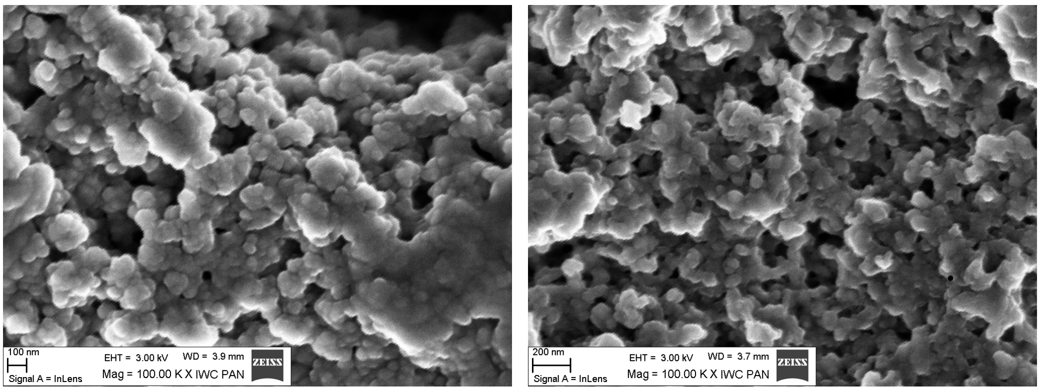
(a) (b)

Figure 3. SEM images of raw wires with (a) d=0.63 mm and (b) d=0.83 mm. 100 k x magnification



(a) (b)

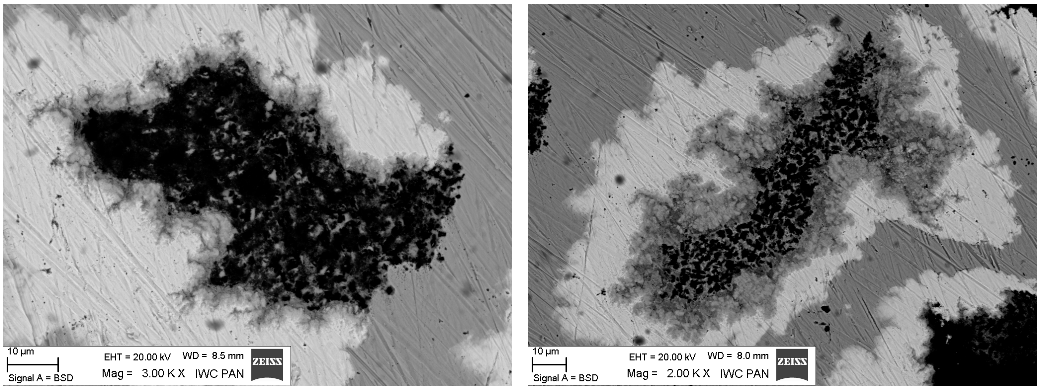
Figure 4. SEM images of wires after HIP (1 GPa, 660-705^{o}C, 15 min) with (a) d=0.63 mm and (b) d=0.83 mm. 1 k x magnification



(a) (b)

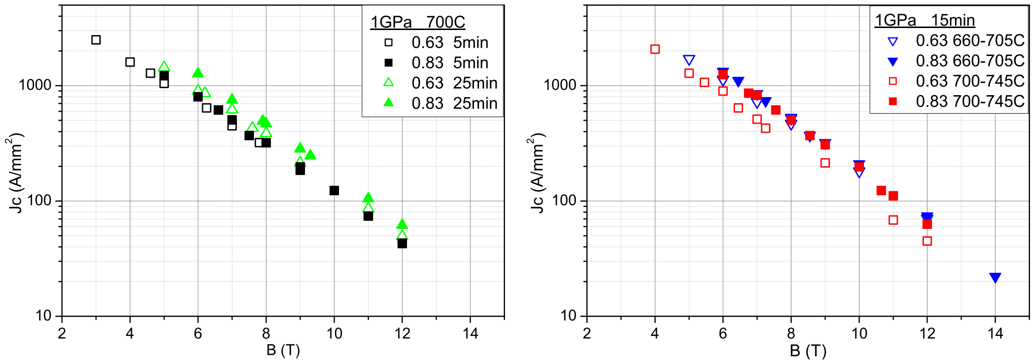
Figure 5. SEM images of wires after HIP (1 GPa, 740^{o}C, 15 min) with (a) d=0.63 mm and (b) d=0.83 mm. 100 k x magnification

sheath material may occur during annealing. Size of barrier breakages as well as HIP parameters determine to what extent a given core reacts with copper. Fig. 6 shows effects of such reactions. This effect is more common for the thinner wires, thus both pictures are for d=0.63 mm.



(a) (b)

Figure 6. SEM images of reacted cores of a d=0.63 mm wire after HIP (1 GPa, 725^{o}C, 15 min). (a) A partially and (b) a fully reacted core. Magnification 2-3 k x



(a) (b)

Figure 7. Critical current vs. wire diameter for (a) different times of annealing and (b) different temperatures

3. Critical current results

Critical current measurements were conducted with the four-probe method in a Bitter magnet, with increasing current or with increasing magnetic field [6]. The maximal available magnitude of magnetic field was 14 T and maximal current was 150 A. Samples were measured in 4.2 K temperature, in either parallel (samples 70 mm long) and perpendicular (20 mm) magnetic field. The distance between measuring probes was similar with both orientations. Various comparisons are presented, most of them for parallel orientation.

It is usually expected that thinner wires have better critical performance [7]. It was already shown on SEM images that drawing of wires to smaller diameters reduces powder porosity. However, the results in Fig. 7 show that wires of d=0.63 mm are at best equal to wires of d=0.83 mm. Advantage of the thicker wires is more significant when integral of temperature over time during heat treatment is larger (i.e. higher temperatures and/or longer treatment). This can be explained through problems with Nb barrier in the thinner wires, that lead to numerous reactions and nonsuperconducting cores.

For both wire diameters it is not enough to heat samples for 5 minutes (Fig. 8). Better results are obtained for treatments of 15 min or 25 min, however, there is no visible difference between those two under 700^{o}C. Optimal time of annealing should be considered within this range.

Samples measured in a perpendicular magnetic field show worse critical properties when compared with samples oriented parallelly (Fig. 9a). This effect is more

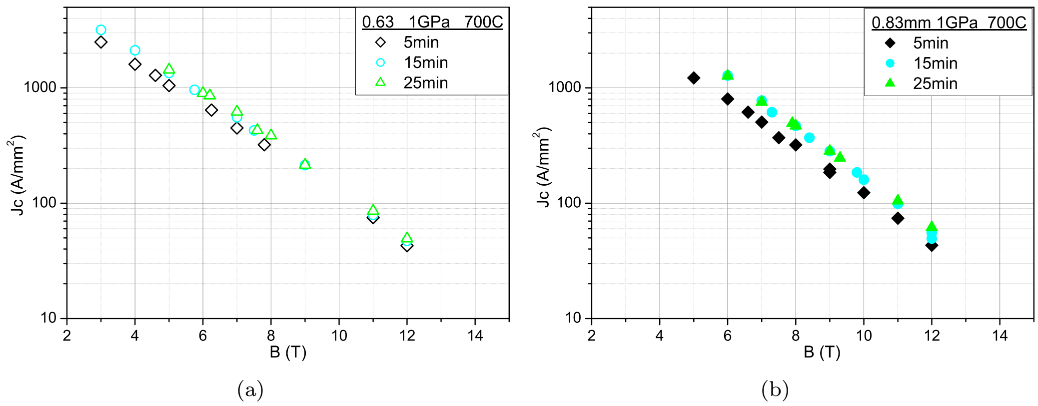


Figure 8. Critical current vs. time of annealing, (a) d=0.63 mm, (b) d=0.83 mm

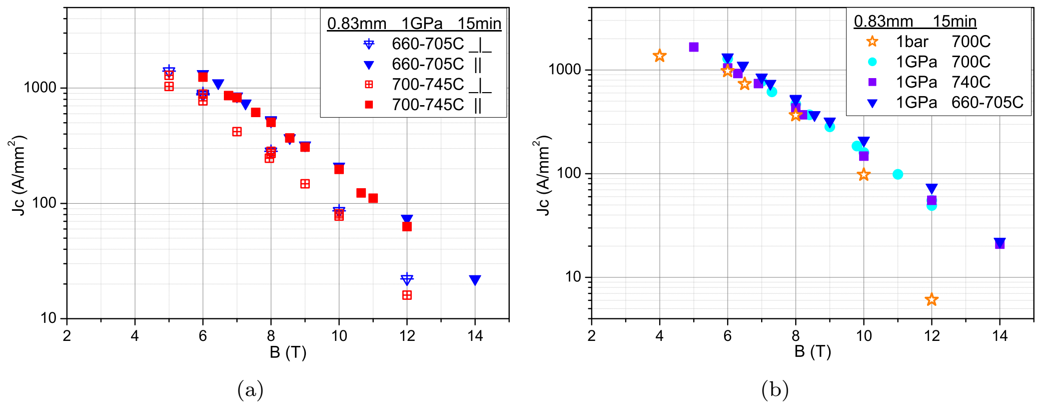


Figure 9. (a) Critical current vs. magnetic field orientation, (b) comparison of critical currents for samples annealed at 1 bar and after HIP

significant in higher magnetic fields. Those results might be biased by the fact that perpendicular samples are shorter. The distances between current and measuring probes are short and current may not be able to fully penetrate into superconducting core at points of voltage measure.

Comparison with samples annealed under atmospheric pressure is essential to evaluate usefullness of high pressure for MgB_2 wires. Such comparison for previous wire strand 961 was presented in [8], however for only a single setup of HIP parameters, and showed a significant increase in critical properties. A more detailed research with a new strand 1019 of the same composition also shows an improvement of critical properties in reference to sample annealed at atmospheric pressure (Fig. 9b). This positive effect was achieved with numerous samples and for different setups of HIP parameters and is at a level of 50-100 % in intermediate magnetic fields. For magnetic field of 12 T critical current increased ca. 12 times.

Based on critical current results, pinning force density (Fig. 10a) was calculated and a Kramer plot (Fig. 10b) was prepared. Due to limitation of current source, maximum of pinning force was not measured and it is difficult to comment changes in pinning mechanisms [9, 10] with high pressures. The Kramer analysis shows an increase in irreversibility field for HIP-ed samples.

4. Conclusion

High pressure is a promising technique that allows an improvement of critical properties of superconducting MgB_2 wires. Annealing under argon pressure of

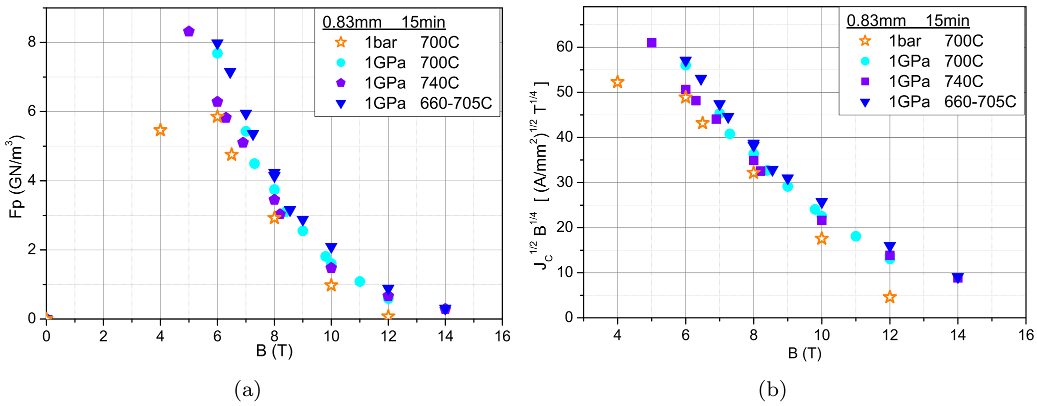


Figure 10. (a) Pinning force and (b) Kramer plot for samples annealed at 1 bar and after HIP

1 GPa provides both densification of superconducting material and small grain sizes (grain sizes after HIP annealing are comparable to those of powder in raw wires). Improvement of critical current by 50-100% was achieved in intermediate magnetic fields and by a factor of 10 in high magnetic field of 12 T.

Raw wires of smaller diameter d=0.63mm have higher density of core powder compared to 0.83mm. This, however, does not improve their critical propertiees, mainly due to problems with barrier continuity that leads to reactions of superconducting core with sheath Cu material. Such problems are more often when integral of temperature over time is large (long time, high temperature).

Quality of Nb barrier is an essential factor in preparation of discussed wires, as any cracks and discontinuities quickly lead to reaction of Mg in *in situ* material with Cu in the sheath.

Acknowledgements

This work was partially financed by Polish Ministry of Science and Information, Technology Project no. N N510 278738 and Project no. N N510 35747.

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