Synergetic combination of LIMD with CHPD for the production of economical and high performance MgB2 Wires

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Abstract
We propose an economical fabrication concept, the localized internal magnesium diffusion (IMD) method. Instead of using a single magnesium (Mg) rod in the center of a metal sheath tube, we use large-sized Mg particles (20-50 mesh) mixed well with cheap 97% crystalline boron powder to fill the metal sheath tube. After a repeated drawing process, the coarse Mg is elongated along the core wire axis of the metal sheath tube. Textured MgB2 grains are then formed during the sintering process. In the localized IMD process, however, there is still a need to improve the overall density. In order to increase the density of the composite, a modified cold high pressure densification (CHPD) technique has been applied before the reaction. It is found that the critical current density (Jc) of the sample made from large-sized Mg with crystalline boron powder and treated by CHPD is increased significantly, so that it is quite comparable with the Jc values of samples made from expensive small magnesium and nanosized amorphous boron powder. At 4.2 K and 8 T, the Jc value of the wire in this work with the cheapest starting materials reaches 10 000 A/cm², which is similar to reported values for samples made by the powder-in-tube and IMD processes with expensive nanosized amorphous boron powder. A possible mechanism is proposed, and the microstructure is analyzed to explain this interesting feature. The main goal of this work is to develop a novel and cost-effective fabrication technique by combining the localized IMD process with CHPD and using cheap crystalline boron powder to manufacture MgB2 superconductor wires with electromagnetic performance superior to that of low-temperature Nb-Ti superconductors. 2002-2011 IEEE.

Keywords
mgb2, high, economical, performance, cphpd, limd, production, synergetic, wires, combination

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Synergetic Combination of LIMD with CHPD for the Production of Economical and High Performance MgB₂ Wires

Minoru Maeda, Md. Shahriar Al Hossain, Ashkan Motaman, Jung Ho Kim, Anna Kario, Matt Rindfleisch, Mike Tomsic, Shi Xue Dou

Abstract—We propose an economical fabrication concept, the localized internal magnesium diffusion (LIMD) method. Instead of using a single magnesium (Mg) rod in the centre of a metal sheath tube, we use large-size Mg particles (20-50 mesh) mixed well with cheap 97% crystalline boron powder to fill the metal sheath tube. After a repeated drawing process, the coarse Mg is elongated along the core wire axis of the metal sheath tube. Textured MgB₂ grains are then formed during the sintering process. The localized IMD process, however, there is still a need to improve the overall density. In order to increase the density of the composite, a modified cold high pressure densification (CHPD) technique has been applied before the reaction. It is found that the critical current density \( J_c \) of the sample made from large-size Mg with crystalline boron powder and treated by CHPD is increased significantly, so that it is quite comparable with the \( J_c \) values of samples made from expensive small magnesium and nanosize amorphous boron powder. At 4.2 K and 8 T, the \( J_c \) value of the wire in this work with the cheapest starting materials reaches 10,000 A/cm², which is similar to reported values for samples made by the powder-in-tube (PIT) and IMD processes with expensive nanosize amorphous boron powder. A possible mechanism is proposed, and the microstructure is analyzed to explain this interesting feature. The main goal of this work is to develop a novel and cost effective fabrication technique by combining the localized IMD process with CHPD and using cheap crystalline boron powder to manufacture MgB₂ superconductor wires with electromagnetic performance superior to that of low temperature Nb-Ti superconductors.

Index Terms—Critical current density, crystalline boron, localized diffusion, elongated Mg.

I. INTRODUCTION

So far as practical applications are concerned, the preparation methods play a very important role in achieving good performance. The ‘in situ’ method has been used successfully to make MgB₂ wires and tapes. In most cases, both high purity crystalline or amorphous boron (B) powder and small size Mg are used to make the MgB₂ conductor [1-4]. If this wire is to be applied in an industrial application, however, the costs of the raw materials will be significantly increased and need to be taken into serious consideration. The material cost could be decreased significantly by using low-grade precursors. High purity (99%) amorphous B powder is about ten times more expensive than low purity (96%) crystalline powders. Alternative lower purity materials need to be tested to find out if they can be used to prepare MgB₂ without much deterioration in the superconducting properties. Recently, Kim et al. [5] proposed a very cost effective fabrication process that involved tailoring the starting material for high performance MgB₂ wire. It was reported that using carbon (C)-encapsulated crystalline boron powder and coarse Mg as precursors worked to improve the MgB₂ grain linkage by controlling the shape and direction of the voids; however, grain connectivity is still affected by the presence of elongated voids. To date, the low density within the filament cores has been a serious obstacle to reaching high critical current density \( J_c \) values in MgB₂ wires fabricated by the in situ processing technique. In addition, the reaction of Mg and B to form MgB₂ involves a volume contraction that produces a final density limited to about 50 % of the theoretical density (2.36 g/cm³). The high porosity is well known to be difficult to avoid. As a result, the grain connectivity is hampered by the presence of voids in the finished wires. The typical effective superconductor area in the wire is only about 10% [6, 7]. Recently, cold high pressure densification (CHPD) has been suggested as an alternative to enhance mass density and probably grain linkage [8, 9]. The \( J_c \) at 4.2 K and 10 T has been reported to be increased to ~40,000 A·cm⁻² for a malic acid doped square conductor produced using CHPD. The first successful CHPD of a dense MgB₂ conductor in a short-length (<= 1 m) was reported by the University of Geneva in 2009 [8]. The density increased from 50% for normal wire to 73% for the CHPD wire after heat treatment. The \( J_c \) increase was particularly impressive at 20 K, amounting to more than 300%. Another very effective densification technique is called the infiltration method, first reported by Giunchi et al., in which the MgB₂ wire was fabricated using a composite billet composed of a steel pipe internally lined with an Nb tube filled with a coaxial internal pure Mg rod and B powder [10]. The \( J_c \) performance from the
reported work was not outstanding, however, as there was no chemical doping associated with the technique. Following this work, an internal Mg diffusion (IMD) process using a pure Mg rod as a core surrounded by B powder and SiC additive was developed. The composite was cold worked into a wire and heat-treated at temperatures above the melting point of Mg (650 °C). During the heat treatment, liquid Mg infiltrated into the B layer and reacted to form MgB2 phase. The best Jc for this IMD SiC doped MgB2 wire was 100,000 A·cm−2 at 8 T [11], exceeding the performance of commercial Nb-Ti wires and making it satisfactory for all technical applications. However, there is a serious limitation for scaling-up the IMD method. In this process, the Mg infiltration into the B layer forms a hollow wire with only a thin layer of MgB2 in the interior of the sheath tube. The total superconductor cross-sectional area is small. It is obvious that for this technique to be useful in large-scale production, an innovative process must be developed, and this is the process we propose in this work.

The main goal of this work is to develop a novel fabrication technique by combining the localized IMD process (LIMD) with CHPD and using large size Mg instead of Mg rod and cheap crystalline B powder for the manufacture of economical MgB2 superconductor wires with electromagnetic performance suitable for industrial application.

II. EXPERIMENTAL PROCEDURE

All wires were fabricated by the in situ powder-in-tube (PIT) process. The starting Mg:B atomic ratio was chosen to be 1:2. Mg powders with different particle sizes of 325 mesh, 100 mesh, and 20-50 mesh were mixed with 97% crystalline boron supplied by Specialty Material Inc (SMI), USA. For comparison, two extra batches of wires were prepared from 325 mesh Mg/99% nanosize amorphous boron supplied by SB Boron USA and 325 mesh Mg/crystalline boron supplied by H.C. Starck, Germany. The compacted mixtures were inserted into Fe sheaths 9.5 mm in outer diameter and 5.5 mm in inner diameter. The Fe sheaths were cold-rolled into wires 3.5 mm in diameter using a two-axial grooved roller and inserted into Monel sheaths 6.0 mm in outer diameter and 3.5 mm in inner diameter. Afterwards, the composites were cold-rolled and drawn into wires 1.0 mm in diameter. The wires were then subjected to the CHPD process with the applied pressure of 1.5 GPa for core densification. All wires were sintered at 600 °C for 16 hours in a flowing argon gas environment. The transport critical currents of short pieces (6 cm) of straight wires were measured against magnetic field in the low temperature laboratories at the Karlsruhe Institute of Technology (KIT), Germany. Samples were immersed in a liquid helium bath and exposed to an external transverse magnetic field ranging from 2 up to 12 T. Current–voltage (I–V) characteristics were recorded by using a standard four-point measurement, where the potential taps were placed in the middle of the wire and spaced 2 cm from each other. The critical currents were estimated from the I–V curves using the criterion of 1 μV/cm. Grain morphology and microstructure were investigated by scanning electron microscopy (SEM).
Fig. 3: Transport critical current densities at 4.2 K as a function of magnetic field for binary MgB$_2$/Fe/Monel wires fabricated from Mg powders with different particle sizes of 325, 100, and 20-50 mesh, and crystalline boron powder from Specialty Materials, Inc. For comparison, the wires were also subjected to the CHPD process at 1.5 GPa.

In this study, we selected large size Mg following the concept proposed by Kim et al. [5], which was in contrast to the internal magnesium diffusion (IMD) method reported by the National Institute of Materials Science (NIMS) group [11]. We used large size Mg particles (100 mesh, 20-50 mesh) rather than using a single Mg rod in the centre of a metal sheath. By this technique, the Mg diffusion process was localized after long hours of heat treatment (600 °C/16 h) to allow a more uniform distribution of elongated voids generated during the cold working along the wire axis rather than a single big hollow, as is found in the typical IMD wires after reaction. Kim et al. demonstrated the validity of this concept [5].

Although the localized IMD process only uniformly distributes the voids and changes the voids to an elongated shape, it slightly improves the overall density. Mg is ductile, and it can be stretched along the wire direction [12] during the drawing process, resulting in a stringy and fibrous structure, to which is attributed the improved superconducting area fraction. If the size, shape and distribution of the voids can be correctly identified, then it will be easier to find a solution to avoid it completely. CHPD is one of the most successful solutions for improving the density of the filament. Although voids can be minimized by CHPD, they are not completely avoided. The shape and direction may be controlled and more aligned, however, so that the MgB$_2$ grain linkage and area fraction can be improved. A schematic diagram of this mechanism is presented in Fig. 2.

Fig. 3 shows the effect of CHPD on the transport $J_c$ at 4.2 K in magnetic fields up to 12 T for binary wires fabricated from various sizes of Mg powders and crystalline boron, and sintered at 650 °C/16 h. Only data above 3 T are shown, because in the lower field region, $J_c$ was too high to be measured due to the limitations of the current supply source and the lack of proper electrical stabilization. It was found from Fig. 3 that the binary wire made from the smaller size (325 mesh) Mg powder usually showed higher values of $J_c$ than the samples made from relatively larger sizes of Mg powder (100 and 20-50 mesh), which was consistent with literature data [13], but surprisingly, this behavior was changed completely when CHPD was applied. The $J_c$ of wires fabricated from large size Mg (20-50 mesh) was significantly enhanced by applying CHPD due to the flat and elongated voids in the pressed region (Fig. 2), which might be attributed to the higher densification and hence, better connectivity in the filaments, as evidenced by the SEM images (Fig. 4). The aligned fibrous structure along the wire direction might also be
the possible reason for the enhancement of the effective area fraction. Such grain alignment did not seem to be severely degraded inside the wire by applying CHPD (1.5 GPa), and the boundaries between filaments may also act as strong pinning centers. This work is still ongoing, and there is plenty of room for further investigations of this interesting feature of void engineering in the MgB₂ filaments after the reaction heat treatment.

The critical current density as a function of magnetic field was compared in this study with the reported best performance binary PIT in situ and IMD processed wires fabricated from expensive nanosize amorphous boron, small size Mg (325 mesh), MgH₂ [14], and Mg rod. It is clear from Fig. 5 that the wire fabricated in this work from the cheapest starting materials treated with CHPD was well comparable in terms of performance with reported binary wires fabricated from expensive precursors. The performance of the densified wire made from cheap crystalline boron and large-size Mg shows similar \(J_c\)-B properties to wires using expensive nanosize materials. The critical current density of the wire in this work reaches \(> 10^5\) Acm\(^{-2}\) at 4.2 K and 4 T, which is within the feasible range for application in low-field magnetic resonance imaging (MRI) machines.

IV. Conclusion

We have shown the most economical fabrication route and an alternate solution to the IMD method for the development of high performance, cost-effective MgB₂ wires for industrial application. The \(J_c\) of CHPD treated wires made from crystalline boron and large-size Mg shows the best performance among the other wires in this study. The possible reason for increased \(J_c\) may be the flat, directional, and elongated voids that arise after heat treatment from large size ductile Mg during the cold working process. The core is densified by the application of CHPD without any severe deterioration of the filament inside the wire. The most noticeable point in this study is that such a good quality binary conductor using cheaper crystalline boron and large-size Mg can be equally useful and cost effective for industry, even without the use of nanoparticle dopants.

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