CURRENT MgB$_2$ WIRE PERFORMANCE AND THEIR INDUSTRIAL DEVELOPMENT

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Abstract

Although just recently discovered as a superconductor, Magnesium Diboride has already demonstrated its clear potential for an effective use in a variety of applications. With its critical temperature of about 40 K, MgB$_2$ gives the opportunity to operate devices at cryogenic temperatures of the order of 20 K, allowing for the use of alternative, cost effective, and simpler technologies than liquid helium. Suitable processes for long lengths wire manufacturing have been demonstrated in the recent years. This work will focus on the preparation of MgB$_2$ based superconducting wires by the Powder-In-Tube method, using the so-called 'ex-situ' process. The main physical and structural characteristics of these wires will be finally reported and briefly discussed.

INTRODUCTION

Just a few months after the discovery of superconductivity in Magnesium Diboride MgB$_2$, it became clear that this material would have been competing in the near future with the existing superconductors in industrial applications, in particular in the field of superconducting magnets. To date, only seven years later, the impressive advancement of industrially manufactured wires and prototype magnets based on MgB$_2$ is a practical demonstration that those initial ideas were basically accurate.

A number of different routes have been developed to improve the wire processing and to achieve high critical current densities J$_c$ in MgB$_2$. Several groups [1–4] have followed the so-called ex situ technique, while the majority [5–10] have preferred to use the in situ technique. Both ways are based on the powder in tube (PIT) method but while the first uses fully reacted MgB$_2$ powders, the latter starts from a mixture of unreacted Mg and B.

The work presented in this paper refers entirely to conductors manufactured by the ex situ PIT process. Indeed, while the in situ route presents several advantages as low cost, high fill factor, high speed process, low temperature reaction processes, and relatively easy MgB$_2$ nanoparticle doping, the ex situ technique currently appears to be more suitable for the development of long conductors and complex multifilamentary wire geometry, allowing for a better control of the powder granulometry and purity degree as well, and finally it leads to more robust conductors that can be readily employed to realize magnets by the Wind and React process.

Due to the aforementioned reasons, the J$_c$(B) behaviour of the ex situ conductors has not been always as high as in the in situ case. Therefore, a deeper development of the starting MgB$_2$ powders in the ex situ process is needed in order to further enhance J$_c$, at least at magnetic fields of the order of 2–4 T at 20 K and 5–10 T at 4.2 K. In fact these are most likely the typical operating conditions for the conductors when we consider their most widespread use in superconducting magnets.

Nevertheless, especially from the point of view of industrial applications, improving the starting MgB$_2$ powders for the ex situ fabrication process seems to be a feasible way to make this material definitely competitive. The doping and the granulometry control are two straightforward ways to run along at the beginning of such optimisation process.

In this paper, an overview of the results reached in the recent past on MgB$_2$ conductors is briefly reported. First, we will present the technique developed for the fabrication of mono- and multifilamentary tapes in long lengths and an overview of their main properties. Secondly, we will show how the tape performances can be improved by modifying the properties of the starting MgB$_2$ powders: in this case, the transport J$_c$ vs. B behaviour of monofilamentary tapes fabricated through the ex situ technique will be reported up to very high fields. We will focus on the addition of SiC nanoparticles to the B before the reaction with Mg, on the high energy ball milling of MgB$_2$ powders alone and with the addition of SiC or C.

PREPARATION OF EX-SITU MgB$_2$ WIRES

Through the ex situ technique, pre-reacted MgB$_2$ powders are filled inside metallic tubes in order to manufacture both mono- and multifilamentary conductors. The powders are currently prepared from a mixture of commercial amorphous B (95– 97% purity) and Mg (99% purity), heat treated at about 900 °C in Ar atmosphere, producing rather pure MgB$_2$ with some residual traces of MgO in a quantity well below 10 wt%. Pure Ni tubes are filled with such reacted powders with a packing density of about 1.3 g/cm^3, and are subjected to wire drawing down to a diameter of about 2 mm, followed by several steps of cold rolling and a final sintering stage at 900–1000 °C in Ar.

In the case of multifilamentary tapes, monocore wires prior to cold rolling are packed again inside a new Ni tube with a multifilamentary layout. The multifilamentary configuration has allowed to obtain wires and tapes that still show similar J_c values than the monocore ones, but capable of sustaining a larger bending strain together with a reasonably limited fluctuation of the superconducting cross-section along their entire length.

Practical applications considering the use of a superconducting material, however, require further precautions and protective measures in case of a quench. This scope is usually achieved by closely adding OFHC (oxygen-free high conductivity) Copper in parallel with the superconducting wire. Due to the chemical incompatibility of MgB_2 with most of the highly conductive metallic pure elements, however, the best solution we have found to date is the insertion of a diffusion barrier with the purpose of separating the superconducting filaments from the highly conductive metal.

We have concretize this approach by introducing a copper core in the centre of the conductor, properly coated by a thin barrier to limit its chemical diffusion towards MgB_2. The resulting composite is incorporated in the central part of a superconducting wire comprising a plurality of MgB_2 filaments arranged in a ring around the core. The barrier has to be made of a material capable of chemically, but not electrically insulate the copper core and it has to be compatible with MgB_2.

Mostly for economical reasons we are currently using pure iron for the barrier material in our standard materials. In Fig. 1, a typical transverse cross-section of this type of multifilamentary conductor is reported.
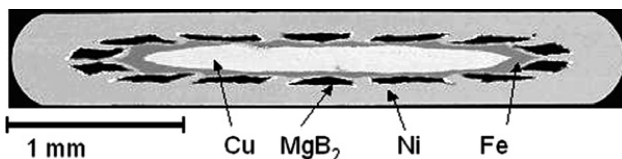

Fig. 1. Cross-section of a Cu-stabilized multifilamentary tape with 14 filaments.

The cross-section is 3.5 x 0.65 mm^2 (width x thickness) and the superconducting filling factor is just 8-9% of the entire cross section, i.e. about 0.2 mm^2. The first example of long length (1.53 km) of multifilamentary conductor was achieved in April 2005; at present, Columbus Superconductors is able to produce up to 1.78 km in a single piece by a reproducible process that has been already successfully completed more than 100 times. The picture of one of such unit lengths is shown in Fig. 2.

Fig. 2. 1.6 km standard stabilized multifilamentary MgB_2 tape in only one piece.

SUPERCONDUCTING PROPERTIES OF THE 'STANDARD' MULTIFILAMENTARY TAPES

The transport critical current I_c was measured using the standard four-probe method in varying magnetic field applied both perpendicular and parallel to the tape surface direction and at different temperatures. In Figs. 3 and 4 the I_c behaviour as a function of the temperature and of the magnetic field is reported as measured by Kitaguchi at NIMS in Tsukuba, Japan, on short multifilamentary conductors cut from a unit length exceeding 1.6 km.

The I_c value drops quickly as the magnetic field increases; nevertheless there is direct evidence of the reproducibility of the tape properties: the measurements performed on two different standard tapes cut at different places from the same batch of conductor give almost identical I_c values in magnetic field and at all temperatures (Fig. 4).

A number of practical applications would benefit from the possibility of employing an MgB_2 conductor to realize windings that can be operated in persistent mode. The potential for a superconducting material of sustaining relevant persistent currents for a long period of time is usually judged by analyzing the exponential n-factor of the *V–I* characteristics. A high *n*-factor (above 30) is generally considered as a proof for a highly homogeneous superconductor with rather strong pinning centres, and therefore low flux line relaxation should occur.
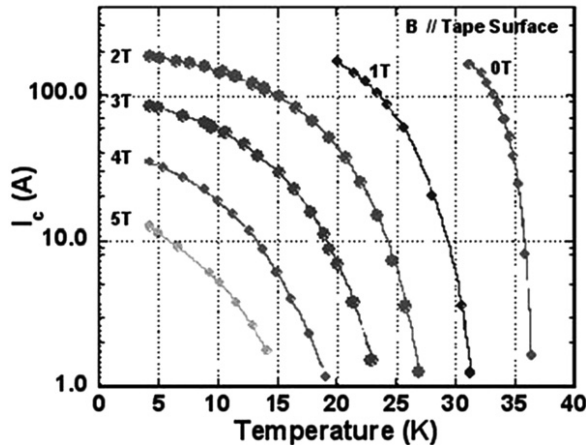
Fig. 3. I_c vs. T at different magnetic fields (parallel to the tape surface) for a standard multifilamentary MgB_2 tape.
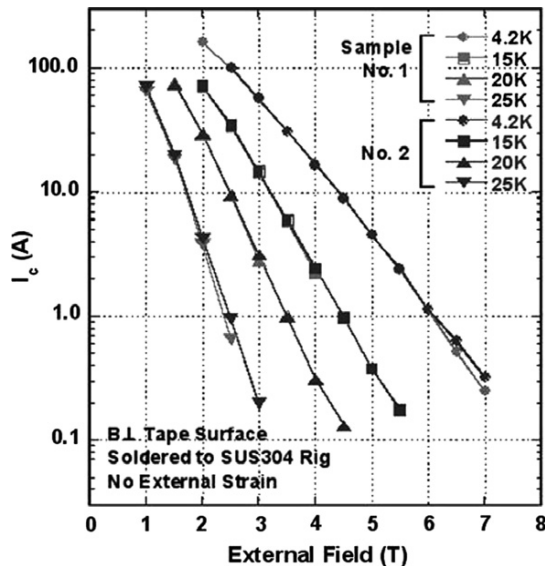

Fig. 4. Comparison between I_c vs. B at different temperatures for two samples cut from two different long conductors: different samples have reproducible performances.

So far, MgB_2 tapes present n-factor between 30 and 100 at fields below 5 T at 4.2 K, making it possible to conclude that the critical current density of MgB_2 tapes is not severely limited by microstructural defects and inhomogeneities along their length.

IMPROVEMENT OF THE PERFORMANCES OF THE CONDUCTOR

It is well known that in the production process of MgB_2 conductors, several parameters have a direct influence on J_c in the superconducting filaments, and that relevant transport properties can be achieved only after optimising the superconducting and/or microstructural properties of the initial powders and of the constituents composing the conductor itself.

The first example of long length (1.53 km) of multifilamentary MgB_2 conductor reached a transport critical current of about 110 A @ 20 K, 1.2 T (5×10^3 A/cm^2 of engineering J_c). Considering that the amount of superconducting material in this conductor was very low (filling factor of 10%), its J_c at 20 K, 1.2 T on the MgB_2 fraction was about 5×10^4 A/cm^2. To date, after about three years, the average results are in the order of 320 A @ 20 K, 1.2 T, i.e. almost a factor of three higher than at the beginning, translating into a J_c at 20 K, 1.2 T larger than 10^5 A/cm^2.

In parallel with the optimisation of the production parameters, an intense research activity on the powder production to achieve higher critical current densities by the ex situ process has been also carried out [11].

In particular, positive results were obtained (a) by the addition of SiC nanoparticles to B before the reaction with Mg, (b) by high-energy ball milling of MgB_2 powders alone and (c) together with the addition of SiC or C. As a test of the effective improvement of these modified powders, monofilamentary tapes were fabricated following the ex-situ route of the PIT method and their I_c values were measured.

The first attempt to improve the magnetic field performance of MgB_2 has been pursued through the SiC nanoparticles inclusion in the precursors during the MgB_2 powder synthesis. In Fig. 5 the transport I_c at 4.2 K in a magnetic field up to 13 T is reported with the field perpendicular and parallel to the tape surface for monofilamentary tapes prepared with undoped and SiC-doped powders.
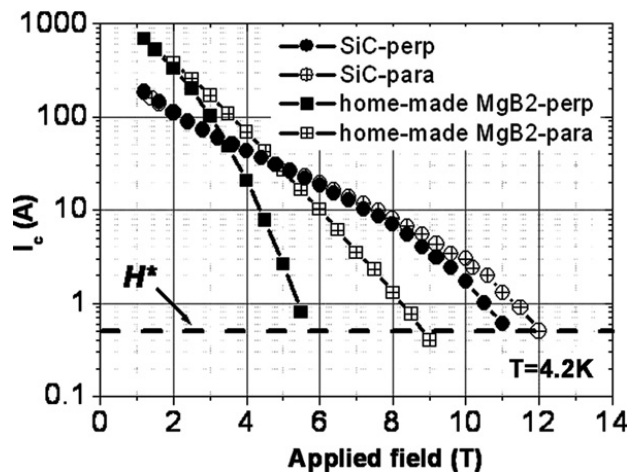

Fig. 5. Transport I_c vs. magnetic field at 4.2 K in both orientations for undoped MgB_2 and MgB_2 prepared from SiC-doped B.

We notice that even if I_c is clearly lower at low magnetic fields, the magnetic field dependence is significantly improved at high fields, while the anisotropy

of I_c, visible in our undoped tapes for magnetic field above 2–2.5 T, is significantly suppressed.

High-energy ball milling was performed on undoped MgB_2 powders in order to lower their particle size significantly and to improve the performances of the conductors in high magnetic fields by adding more effective grain boundary pinning. In Fig. 6, J_c as a function of the magnetic field is shown at 4.2 K (a) and 20 K (b) as extracted from the M–H loops with the applied field perpendicular to the tape surface, measured with a commercial 5.5 T MPMS Quantum Design Squid Magnetometer [12].
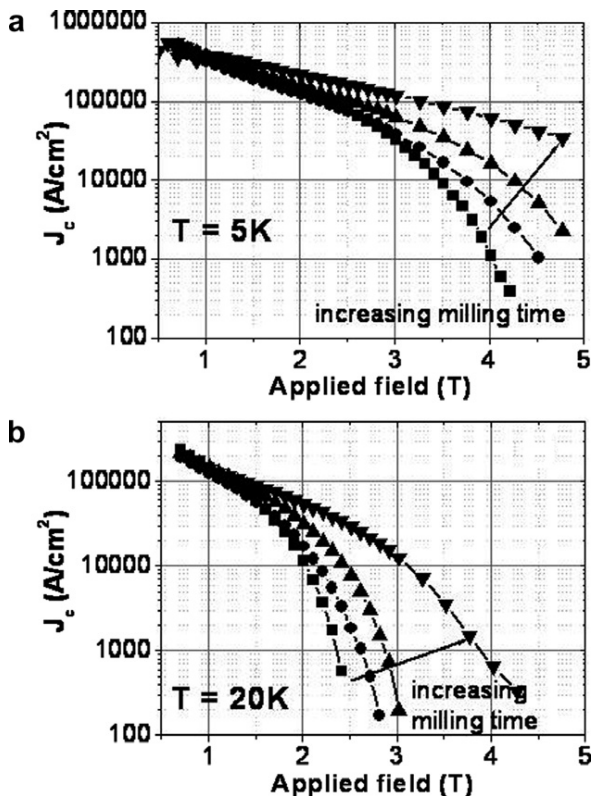

Fig. 6. Magnetic J_c vs. magnetic field at (a) 5 K and (b) 20 K in perpendicular orientation for undoped MgB_2 by increasing the milling time.

The magnetization measurements were performed at 5 K and 20 K on portions of conductors selected from the same wire batch used for the transport measurements. The magnetic field was applied both perpendicular and parallel to the tape surface. We notice that, while J_c is only slightly changing at low fields, a substantial improvement is obtained at higher fields by increasing the milling time, together with a significant decrease of the anisotropy.

This is better represented in Fig. 7, where the magnetic J_c is reported in both perpendicular and parallel orientation for the not milled MgB_2 and the sample prepared with the powders milled for the longest time, both at 5 K and 20 K.
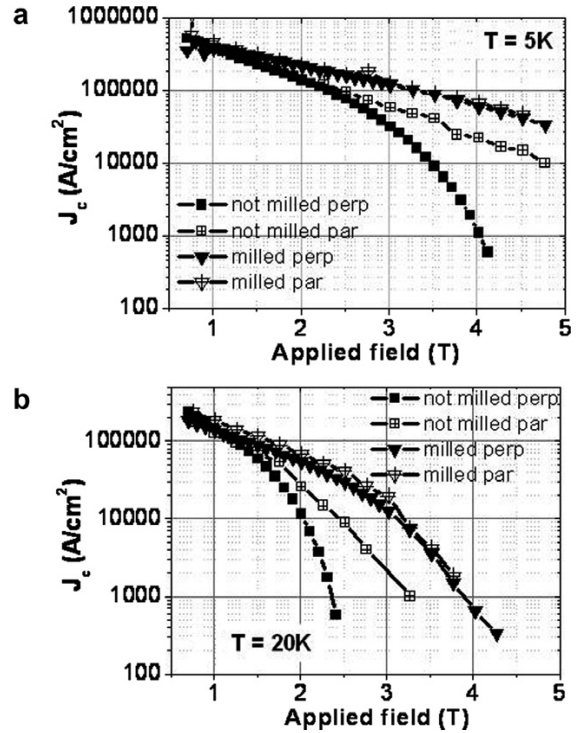

Fig. 7. J_c data extracted from the M–H loops in perpendicular and parallel orientation for the not milled and the highest-time milled samples at (a) 5 K and (b) 20 K.

There might be several explanations for this effect. First of all, the effect of the cold working procedure on the grain orientation is probably less relevant if the grains are much smaller. Furthermore, the current is determined by percolative paths that might be more relevant in the milled powders in which the grains are smaller, giving a stronger effect at higher field where the direct superconducting paths are suppressed.

Due to the strong improvement of the tape performances in magnetic field in samples prepared with ball milled powders, the procedure was also repeated with SiC nanoparticles and carbon additions.

In Fig. 8, the transport I_c is shown for a standard not milled sample and two tapes prepared with milled MgB_2 powders for the same duration but respectively with and without SiC addition, measured both in perpendicular and parallel orientation. We can see that the magnetic field behaviour is significantly improved in the doped powders with respect to the undoped case, and the anisotropy is strongly suppressed.
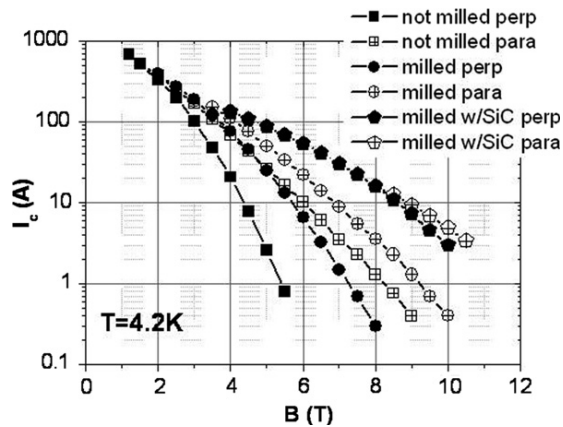
Fig. 8. Transport I_c in magnetic field at 4.2 K for tapes prepared with not milled powders compared with a tape prepared with milled MgB$_2$ powders and MgB$_2$ powders milled the same time with SiC nanoparticles.

The same procedure was followed with Carbon addition, that was ball-milled together with the MgB$_2$. In Fig. 9 the transport I_c is shown at 4.2 K in magnetic field for tapes made from the undoped powders, the milled powders, the MgB$_2$ milled for the same time and twice this time with addition of C. The effect of milling with C is even stronger concerning the behaviour at high magnetic fields.
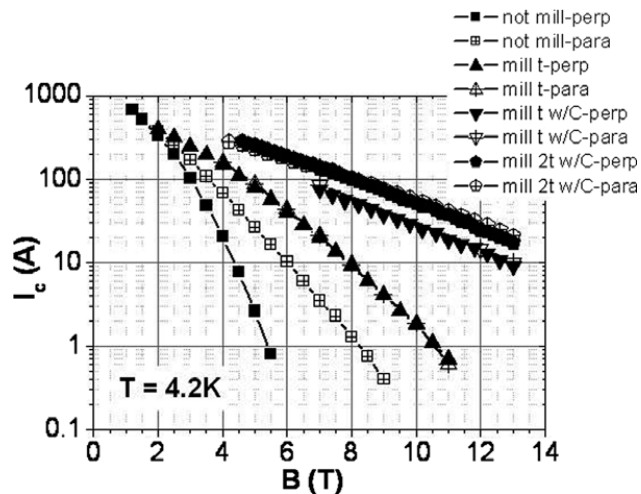

Fig. 9. Transport I_c in magnetic field at 4.2 K for tapes prepared with not milled powders compared with a tape

prepared with milled MgB$_2$ powders, MgB$_2$ powders milled the same time and twice the time with carbon.

In the best sample, a critical current of 20 A – which corresponds to about 10^4 A/cm^2 – is reached at 13 T.

All these positive results – in particular, the use of milled powders with or without carbon – are being extended now to the production of large powder batches for the fabrication of long length multifilamentary conductors with improved properties.

The result of this R&D work confirms the possibility of a commercial use of MgB$_2$ in the high field region, considering in particular the very promising level of J_c of about 10^4 A/cm^2 reached at 13 T in liquid helium (Fig. 9).

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