THE REACTIVE Mg-LIQUID INFILTRATION TO OBTAIN LONG SUPERCONDUCTING MgB_2 CABLES

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Abstract

An alternative “in situ” process to the MgB_2 wire manufacturing is represented by the Reactive Mg-Liquid Infiltration (Mg-RLI) process [1], in which the precursor wire is constituted by a metallic sheath encasing a central Mg rod, surrounded by the B powders. We demonstrated that this peculiar “internal Mg” assembly is able to produce very dense superconducting material of high critical current density, with an acceptable fill factor, up to 0.28. Furthermore the Mg-RLI allows also to easily dope the MgB_2 material either by carbon or nanoSiC powders. In order to realize long cables with this technique, two different approaches may be applied. The first one relies on the assembly of thin wires, fine enough that the liquid Mg cannot freely percolate along the wire during the reaction, and the second one relies on the assembly of thick hollow wires, reacted with a continuous supply of Mg to avoid deficiency of Mg in some part of the precursor wire. Both techniques have been demonstrated feasible and the relative usefulness is discussed. As far as the large superconducting magnets are concerned, either for future physics applications or for fusion reactors, it will be evident the great advantage of the low weight of the MgB_2 wires, other than their good performances at intermediate high magnetic fields: typically of the order of 4T at temperatures of about 20K.

INTRODUCTION

The MgB_2 conductors are generally manufactured by a Powder in Tube process, performed either by “in situ” or by “ex situ” routes, so named in order to distinguish where the MgB_2 is formed. These two alternative processes gives rise to wires or tapes with different superconducting properties. Today the most performing MgB_2 wires have been obtained by the “in situ” route [2], but the realization of a long superconducting cable from the wire is a process which is not without difficulties. Furthermore, in order to build superconducting magnet prototypes one needs the more demanding “wind and react” technology. On the other hand, long superconducting tapes have been manufactured with the “ex situ” route. This route, even if less performing, has demonstrated its applicability in the manufacturing of large magnet prototypes through the more friendly “react and wind” technology [3]. Notwithstanding the strong competition in the field of the superconducting magnets, based on a field/temperature performance rather than on a cost/performance base, attempts to maximize the current density of the wires in higher magnetic fields or at higher temperatures have been done. For this reason the “in situ” route appears as the most appealing long term alternative to MgB_2 conductors, especially in view of the possibility, in a near future, to easily manufacture long superconducting cables. In the framework of the “in situ” processes, the Mg-RLI technology, applied to the MgB_2 superconducting wire manufacturing, presents important benefits, related to the high level of the superconducting properties of the resulting material, to the relatively easy processing scheme, that can be applied to several cabling options that will guarantee high mechanical strength, crucial for large magnets manufacturing.

THE Mg-RLI TECHNOLOGY FOR MgB_2 DENSE WIRES

In the conventional solid state reaction route, to obtain MgB_2, a mixture of B and Mg fine powders is usually reacted. Indeed, this procedure conducts to poorly sintered materials, when not used appropriate hot high pressure conditions. The presence of porosity has been well documented in the MgB_2 bulk and wires manufacturing, if the “in situ” case is applied in a conventional way, i.e. with powders mixing. This porosity is due to the poor sintering ability of the MgB_2 material together with the intrinsic volume reduction, of about 25% , during the reaction. The final product, at the best, will have a density of 75% of the theoretical value and this happens, no matter will be the size of the initial powders. To avoid this pitfall, we have introduced an innovative way to react the B and Mg: allowing the infiltration of liquid Mg inside a preform of B powders [4]. In such a way it is possible to obtain an almost full dense MgB_2 without external pressure. The possibility to realize the infiltration reaction inside the boron powder, even for high deepness, is based on unexpected chemical behaviour of the reactants. From our experience, two main driving forces favour this behaviour: a) the aforementioned volume contraction which induce more liquid to reach the reacting zone; b) the existence of a precursor phase of the reaction, when crystalline boron is used: a boron rich Mg boride, Mg_2B_{25}, that we have discovered and fully structurally characterized inside our MgB_2 material [5,6]. After reaction, the zone where the Mg was located remains practically void and the zone where the B powders ware stacked results fill of MgB_2. In the peculiar case of the wires manufacturing, it was possible to obtain, directly from the reaction, either hollow or Mg filled superconducting wires [7].

Wires features

The typical feature of a monofilament precursor wire, used in the Mg-RLI process, is displayed in Figure 1A. After reaction, two peculiar arrangements of the resulting monofilament superconducting wires are possible: a wire with the hole filled by Mg metal, Figure 1B, and a wire with unfilled hole, Figure 1C. The fill factor, related to the MgB_2 superconducting material, is in both cases about 25%. Other features that may represent key variables in the wire processing and on the final wire properties, are:

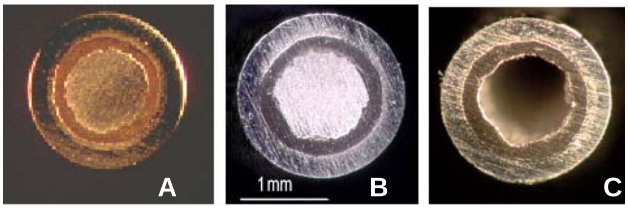

Figure 1: Optical microscope images of : A) precursor wire with internal Mg surrounded by B powder; B) superconductive MgB_2 wire with Mg-filled hole; C) superconductive MgB_2 wire with unfilled hole

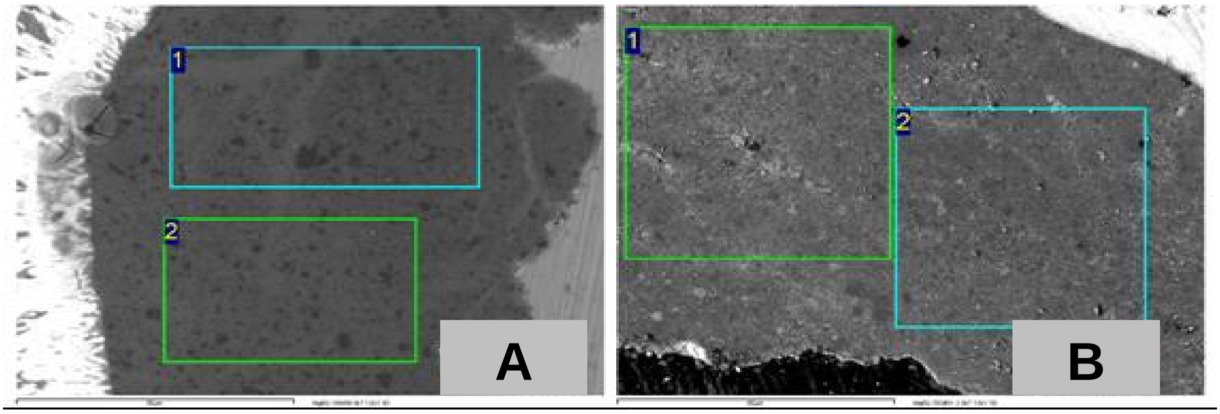

Figure 2: SEM images of the MgB_2 material : A) wire with Mg_2B_{25} impurity , B) wire without Mg_2B_{25} impurity

- the material of the external metallic sheath, made by Fe or, preferably, by Monel;
- the presence or not of a thin interlayer of Nb, between the external sheath and the boron;
- the addition of appropriate doping elements to the B powders;
- the purity of the B powders.

The Mg filled wires have many interesting advantages: an higher thermal stability, an electrical shunt that can prevent damages during a quench, a more robust structure that protect the MgB_2 part from mechanical damaging .

MgB_2 microstructure

An example of the quality of the MgB_2 material resulting from the Mg-RLI process, applied to the wires, is illustrated in Figure 2, where the lack of porosity is highlighted. The pictures show the typical morphologies of the MgB_2 material deriving from two different boron powders. The wire A was obtained by using microcrystalline β-rhombohedral boron and the SEM image shows the presence of darker grains, few microns in size, embedded in the pure MgB_2 materials. These grains are constituted by a mixture of MgB_2 and of the impurity phase Mg_2B_{25}, being the superconducting phase the continuous matrix. With the aid of an aerial X-ray fluorescence microanalysis [6] we estimate a mean presence of the Mg_2B_{25} phase of about 7mol%, with a max percentage of 40% inside the darker grains. The wire B was obtained by using really amorphous boron: the corresponding SEM image shows very little amount of the boron-rich phase and also the aerial compositional analysis indicates a low percentage, about 0.3 mol%, of Mg_2B_{25} with respect to the MgB_2. In both wires there is no visible porosity in the MgB_2 part, at a SEM resolution, like in the cases when the material is prepared with high pressure apparatus.

SUPERCONDUCTING PROPERTIES

The most important superconductive characteristic is represented by the critical current density as a function of magnetic field and of the temperature. Challenging conditions for the application of MgB_2 material may be a temperature of 20 K at a maximum magnetic field of 4T or a temperature of 4.2 K at a magnetic field of 10 T. The measurement of the offset of the R(T) curve, at different magnetic flux, gives a detailed information of the irreversibility field, B_{irr}, i.e. the upper limit of the practical magnetic flux that the material can sustain. The measured B_{irr}, for several wires obtained by Mg-RLI, shows a clear effect of the impurities on the improving of the irreversibility field, as expected by the increase of the pinning centres. For example, at 20 K, it has been evaluated the B_{irr} values of 4.8 , 6 and 6.1 T, respectively for the wires derived from amorphous B (no impurity), microcrystalline B (Mg_2B_{25} impurity) and amorphous B added by 6 at.% C [7].

As far as the critical current density is concerned, the actual performance of the Mg-RLI wires are on the edge of the best published results, even if a distinction must be done between short wires and commercial-like long wires or tapes. In Table 1 are collected some recent J_c values measured for the best short wires or tapes produced by different research groups, taking as reference conditions (20 K @ 4 T) and (4.2 K @ 10 T). The large spread of values reflects many variables in the preparation: the use of B powders of different purity or grain size, different additives and different thermal treatments used by the various groups. In particular, among the Mg-RLI type wires, the very recent data of a Japanese team has substantially improved our old data by using 99.99% pure boron, a peculiar SiC additive and an optimized thermal treatment [8].

The best J_c values of the MgB_2 wires, at 4.2 K and 10 T, now compares favourably also with the best NbTi wires that amounts to about 200 A/mm^2 [9]

Table 1 - Best J_c values of MgB_2 short wires or tapes

MgB_2 short sample	**Jc (20K@4T)**	**Jc (4.2K@10T)**	**Ref.**
nanoSiC dop/PIT (Karlsruhe)	**200**	**200**	2
B4C+SiC dop/PIT (Geneve)	**75**	**150**	2
HE mill+nanoC/PIT(Dresden)	**240**	**600**	10
SiC dop/PIT (Ohio)	**200***	**200**	11
HE mill/PIT (Genova)	**8**	**40***	12
C dop/Mg-RLI (Milano)	**60**	**80**	15
SiC dop/Mg-RLI (Tsukuba)	**-**	**400**	8

Values in A/mm^2 *** extrapolated value**

As far as the km long wires or tapes are concerned, there are two products: i) a tape of Columbus Superconductors (I) and ii) a wire of Hypertech Research (US, Ohio). The tape is of the "ex situ" type and, at the moment, has been used to wind a magnet for MRI with the "react and wind" technique [12]. The wire is of the "in situ" type and has been used to wind some prototype magnets with the "wind and react" technique [13].

In Figure 3 the behaviour of $J_E(B)$ of these two conductors, at 4.2 K, is drawn, together with our Mg- RLI undoped monofilament wire of external diameter of 0.52 mm, measured on a meter long wire wound in an ITER barrel [14].

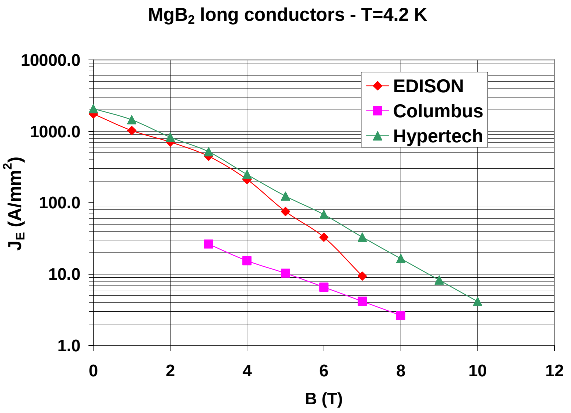

Figure 3: Engineering critical current density of a 0.53 mm diameter Mg-RLI monofilament wire , compared to the reported long commercial-like conductors

The rapid decrease of J_E at high field, for the reported Mg-RLI wire, is due to the absence in it of any additive to improve the pinning strength. Better results are expected with C or SiC addition, as already found in short samples [15].

Mg-RLI WIRES POTENTIALITY

Key characteristics of the practical superconducting wires, other than a high fill factor and a good mechanical strength, are a high thermal stability and low AC losses. One of the golden rules to implement both last characteristics is the reduction of the superconducting material thickness. In the conventional NbTi metallic superconductors this is achieved by a multi-filamentary assembly and by large reduction in the wire drawing operations. The brittle materials like A15 superconductors and High Temperature Superconductors, MgB_2 included, cannot be thinned in the superconducting state, so it is a common practice to make the precursor ductile wires thinner, and then to react them after drawing. This procedure exposes the resulting superconducting wires to mechanical failures, during the magnet winding or during its full in current operation, due to the magnetic forces. The thickness of the superconducting parts in such cases is limited and also the possibility to obtain stranded cables is confined to the thinner wires, excluding the tapes. We have verify the capability of the Mg-RLI wires to be thinned and stranded.

A Mg-RLI monofilament precursor wire, having the external sheath in Monel/Nb, has been successfully reduced to an external diameter of 0.250 mm, for a length of 6 m [14] maintaining the peculiar circular symmetry, as reported in Figure 4, of the original wire. In the corresponding reacted wire the MgB_2 has a thickness of about 20 micron and a SC area of about 8x10^{-3} mm^2, corresponding to a fill factor of about 16%.

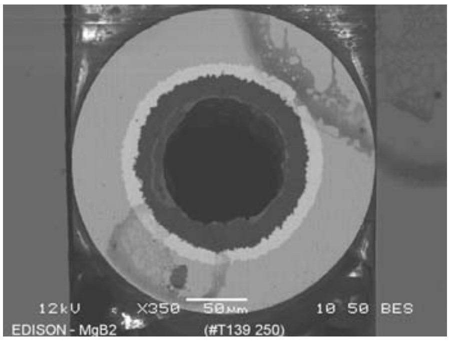

Figure 4: Thin Mg-RLI wire cross section

The drawability of the Mg-RLI precursor wire was quite unexpected, due to the well known brittleness of the metallic Mg. We have realized that the B powders acts as a lubricant for the Mg and prevent the crack initiation on its surface.

Concerning the stranded cables, we have tested a prototype cable made of 3 x 7 Mg-RLI monofilaments of 1 mm thickness each, with a twist pitch of about 50 mm, for a total cable length of 2 m. The precursor strand is shown in Figure 5. After reaction, the transport properties of the cable where measured by inductive method [16].

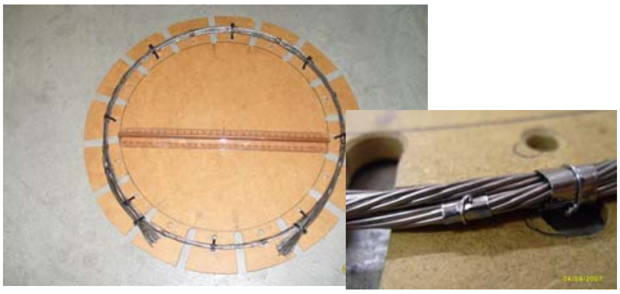

Figure 5: A 2m long stranded cable, made of 21 monofilament Mg-RLI wires

Typical values of the critical current are 17 kA and 5 kA at 14.6 K, and at 0 and 2 T field respectively. These values correspond to an engineering critical current density of the cable, at the respective conditions, of 400 A/mm^2 and 116 A/mm^2 [17].

CABLING OPTIONS

In order to build large superconductive magnets by MgB_2, useful for physics experiments or for fusion reactors, working at temperatures higher than 4.2 K, it will be crucial to design a superconducting cable that allows the "react and wind" techniques, but of substantially higher performances with respect to the actual one. In this respect the Mg-RLI technology offers, in principle, new options for cabling.

Hollow monofilament cable

This is the case in which the cable presents a hollow non superconducting core, eventually filled by stabilizing Mg. A cross section of such kind of cable is given in Figure 6. The external sheath can be Monel with a thin layer of Nb to avoid the MgB_2 contamination, during reaction. This design will be more useful in DC applications, due to the large thickness of the superconducting material.

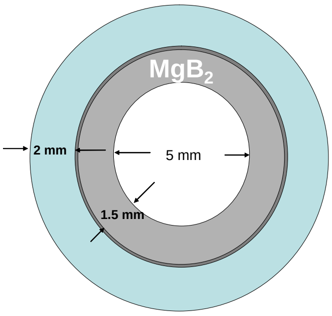

Figure 6: Cross section model of an hollow monofilament cable. The core can be filled by Mg for stabilizing purpose

The size of this cable may be tailored according to the needs. As an example if we consider the need to transport about 7 kA at 20K @ 4T, the estimation of the cable dimensions are OD= 12 mm, ID=5mm, assuming a fill factor of 30% and a critical current density optimal value of Jc = 200 A/mm^2. This cable should be manufactured starting from large composite billets, and then being reduced by drawing operations on long benches. Typically, in order to obtain a 900 m long cable one has to start from a 360 mm OD, 1 m long billet. The reaction of this cable can be done in a circular furnace of several meters in diameter, with Mg reservoir to supply Mg during reaction. The winding diameter of the cable in the furnace should be similar to the winding diameter of the final magnets, to avoid too large bending stress. .

Multifilament cable

This kind of cable is more similar to the usual multiwire cable, designed with the A15 materials. A typical cross section is modelled in Figure 7. The thin diameter of the component wires allows the use this cable also in AC conditions.

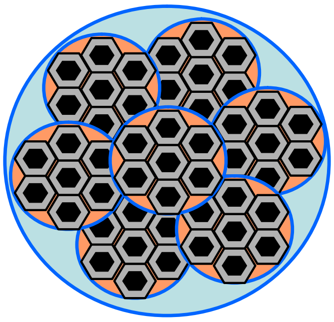

Figure 7: Model cross section of multiwire cable, made of 7x7 Mg-RLI wires

This kind of cable will have a max fill factor of 15%, due to the presence of multiple metallic sheaths. Therefore , returning to the previous example, if we assume the same OD of 12 mm and an assembly of 49 monofilaments, we can foresee the need of more performing MgB_2 wires, with J_c at least of about 400 A/mm^2 at 20 K and at 4 T, which is at the moment the limit of the MgB_2 best performances. Indeed this design will require also some stabilizing agent that could be copper braided between the MgB_2 filaments, which will introduce further penalty in the fill factor. The reaction of this kind of cable can be either similar to the previous one, with the entire cable inserted in a circular oven, or like a Cable in Conduit Conductor (CICC), where already reacted stranded wires are inserted in a tube. For this cabling design the drawing operations will not require long linear benches, but the manufacturing is expected to be more costly, due to the various assembling operations.

Massive cable

This cable design may be considered as an extension of the Mg-RLI technology that we apply to the MgB_2 bulk manufacturing. In peculiar cases, where high currents of the order of several tens of Amperes are available, and DC operation are used, one can design a magnet with a low number of spires. The cable, made of massive MgB_2, may have different types of cross section: rectangular, circular or of different shape, depending on the disposition of the reacting elements B and Mg inside the metallic sheath. In Figure 8 two hypothetical model cross sections are shown.

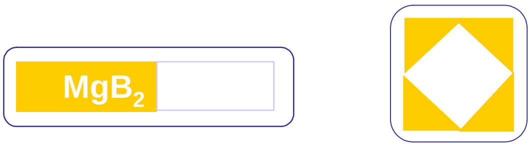

Figure 8: Model cable cross sections for massive MgB_2. The void part, inside the cable, can be alternatively filled with Mg.

With this cabling design a very high fill factor can be realized, of the order of 50%. The reaction, in such a case, will be strictly of the kind "wind and react". In order to compare the performances of this cabling with respect to the previous examples, assuming to realize the same magnetic field in the pancake, the needed Jc of the MgB_2 will be about 100 A/mm^2 at 20 K and at 4 T. The cross sectional area of the cable may be between 5-10 cm^2, transporting currents of 25-50 kA. The length of this kind of cable will be limited to about 100–200 meter.

DISCUSSION AND CONCLUSIONS

The use of the Mg-RLI technology in the manufacturing of the MgB_2 cables may open new possibilities in the design of the superconducting cables for large magnets. Here we sketched some possible designs, but many other options can be considered and analysed. Other than the cryogenic energy saving, a further key characteristics of the future MgB_2 magnets will be their low weight, with respect to the magnets made by the classical low temperature superconductors. It is possible to estimate the weight of the various MgB_2 pancake magnets obtainable by the different presented options of the Mg-RLI cables. It has been assumed as reference magnet a large pancake of 5 m of OD and with 20 x 10 cm^2 winding cross section. This kind of magnet may be the building block of an external poloidal magnet for fusion, where the operative conditions of 20 K and 4 T start to be acceptable. In Table 2 the weights of the superconducting MgB_2 magnets are compared with an equivalent copper type electromagnet.

Table 2 – Tentative superconductive pancakes of MgB_2 vs standard resistive solution (copper).

MgB_2 Cable	**L (m)**	**Turn**	**I (kA)**	**Diam (mm)**	**Fill factor (%)**	**Jc (A/ mm^2)**	**weight (kg)**
multi	1809	120	7.2	12	15	420	1750
Mono	1809	120	7.2	12	30	210	1100
Mass.	150	10	87	90x 19	60	85	830
Copper	1809	120	7.2	12	100	60	2700

As it is evident the MgB_2 solutions are all largely lighter than the copper case. Between the MgB_2 options the massive cable is far the best in term of the needed critical current density and consequently in term of weight. Nevertheless the choice between the various MgB_2 designs will be mainly driven by the applications.

It will be of practical interest to initiate explorative programs to validate the various design options and, in the mean time, also the quality of the MgB_2 materials will be improved, especially referring to the behaviour in high magnetic fields.

Last but not least are the economic issues, that favours the MgB_2 not only due to the relatively low cost of the raw materials, but also for a relative friendly manufacturing. In this respect the Mg-RLI process appears to be one of the most affordable.

ACKNOWLEDGEMENTS

The author acknowledges the interest of Prof. Bruno Coppi in the development of the Mg-RLI technology, in view of the application of MgB_2 cables to some fusion poloidal magnets for the IGNITOR project.

Mike Sumption of the Ohio State University and Mike Tomsic of Hypertech Research are acknowledged for the fine wire manufacturing and characterization.

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