High Anisotropic Critical Current Densities in MgB$_2$ Tapes

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Abstract. We present a study on the angular dependence of the critical current density ($J_c$) of SiC-doped MgB$_2$ tapes fabricated by powder-in tube (PIT) with stainless steel (SS) as sheath material. The microstructure and texturing of the tapes are analysed to understand the effects of the rolling degree on the superconducting properties. The microstructure is studied by x-ray diffraction and scanning electron microscopy, while $J_c$ is measured by transport in liquid He and Ne. We found that for Si-doped tapes the alignment degree augmented with increasing deformation but tends to saturate for aspect ratios larger than 5, while the anisotropy factor $J_{AF}=J_c(90°)/J_c(0°)$ continued increasing for larger aspect ratios. This increase is attributed to a pinning decrease for the applied field parallel to the tape direction. Very high $J_c$ values were obtained for Si-doped tapes, whose cores are as dense as those from wires processed using hot isostatic pressing (HIPing) by optimizing the rolling degree.

Index Terms— MgB$_2$, critical currents anisotropy, material synthesis effects.

1. Introduction

A new generation of superconductors based on MgB$_2$ is expected to substitute some of the Nb-based conventional materials because this compound has an excellent combination of characteristics, such as a high critical temperature ($T_c$~39K), low cost of raw materials and low weight. [1] The standard and low-cost fabrication method to make wires and tapes is the Powder In Tube (PIT), which involves filling a metallic tube with superconducting powder (ex-situ) or precursors (in-situ) and drawing it into a wire and/or rolling into a tape.[2] MgB$_2$ crystallizes in the hexagonal AlB$_2$ type structure (space group $P6/mmm$), and the anisotropic structure has given the motivation to investigate formation of texture by different deformation processes. Several groups have already reported anisotropy in MgB$_2$ tapes prepared by PIT, with different sheath materials.[3][5] Lezza et al. [6] observed a strong dependence of the critical current density ($J_c$) anisotropy with grain size of initial powder. Kovác et al. [3],[7] showed the variation in $J_c$ angular dependence of tapes with Nb, Ta, and Fe as sheath materials at different rolling degrees, doping levels and applied fields. These studies indicated that $J_c$ anisotropy increases with the sheath hardness and decreases with doping.
For most applications, it is necessary to improve $J_c$ by introducing more pinning centers (or defects) and enhancing the grain connectivity. This improvement can be achieved in many ways, i.e. by doping, nanoparticles addition, effective heat treatments such as Hot Isostatic Pressing (HIPing) and combinations of these methods.[8]-[10] Besides, the figure of merit of a superconductor not only depends on $J_c$, but also on the engineering critical current density $J_e = fJ_c$, where $f$ is the filling factor that reflects the ratio of the superconductor to the total cross section of the wire.

This work is focused in the study of tapes prepared by PIT with stainless steel (SS) as sheath material. We prepared a series of samples to explore the influence of the nano SiC addition and rolling degree on the $J_c$ angular dependence of MgB$_2$ tapes. We found that the alignment degree ($\bar{\Lambda}$) augmented with increasing deformation but tends to saturate for aspect ratios larger than 5, while the $J_c$ anisotropy factor ($J_{AF}$) continued increasing even for aspect ratio ~8.1. The $J_{AF}$ increase is attributed to a pinning decrease in the applied field parallel to the tape direction. Very high $J_c$ values were obtained for Si-doped tapes with larger aspect ratios, whose cores are as dense as those from wires processed using hot isostatic pressing (HIPing). A rapid increase in the anisotropy with applied field was observed for lower SiC addition and for higher temperature.

2. Experiment Details

MgB$_2$ commercial powder was packed, adding 5\% at Mg powder as extra source of magnesium. In some of them 2.5\% at SiC nanopowder (20-30 nm grain size) was added to the initial mixture (see composition in Table 1). All powders were ball-milled for one hour inside a glove-box under nitrogen atmosphere before filling the SS tubes (inner and outer diameters 4.6 and 6.4 mm). The tubes were cold-drawn into round wires with external diameters of 1.4 mm, and then rolled to obtain MgB$_2$ tapes with different deformation degrees. Several intermediate annealing treatments at temperatures ranging between 600-900°C were performed to release the cold working stress, while the final annealing was carried out at 850-900°C during 30 min. The MgB$_2$ cores of all SS tapes maintained very similar filling factors $f ~ 45\%$ (i.e. $J_c = 0.45 J_e$) and no reaction between the sheath and the superconductor in agreement with previous observations in SS PIT wires. [11] The different characteristics of the analyzed tapes are listed in Table 1.

The morphology observations and microanalysis of the samples were done on a Philips 515 SEM microscope fitted with an EDAX 9900 energy dispersive spectrometer (EDS). The possible reaction between the sheath material SS and MgB$_2$ was investigated through EDS analysis and backscattering images. X-ray diffraction (XRD) measurements were made with a Bragg–Brentano Phillips diffractometer using Cu K$\alpha$ radiation.

Table 1. Characteristics and alignment degree ($\bar{\Lambda}$) of SS MgB$_2$ tapes

| Sample | Composition : MgB$_2$ +… | Aspect Ratio | $\bar{\Lambda}_{100/002}$ |
|--------|-------------------------|-------------|---------------------|
| T1     | 5\%Mg                   | 2.9         | 0.34                |
| T2a    | 2.5\%SiC +5\%Mg         | 1.9         | 0.14                |
| T2b    | 2.5\%SiC +5\%Mg         | 5.4         | 0.45                |
| T2e    | 2.5\%SiC +5\%Mg         | 8.1         | 0.53                |

D.C. transport critical current $I_c$ was measured using a 1 $\mu$V criterion at $T = 4$ and $T = 26.5$K, with the wires immersed in liquid He and Ne, respectively, and the field $H$ applied perpendicular to the current flow (maximum Lorentz force configuration) forming the angle $\Theta$ with the normal vector to the tape (see the inset of Fig. 1). Reliable data could only be obtained for applied magnetic fields above certain field
depending on the sample due to heating problems at the contacts as we discussed in previous works. [11]-[12]

3. Results and discussion

Figure 1(a) shows $J_c(H)$ curves at 4 K with applied field parallel to a-b plane ($\Theta = 90^\circ$), for all samples listed in table 1. $J_c(H)$ values of doped samples are higher than the undoped sample for all measured fields. For comparison we have included a 5%wt Si-doped tape with an aspect ratio of ~5.2 and Fe-Ni sheath from Ref [7]. The $J_c$ of T2b, our tape with similar aspect ratio, is almost twice the reported value for the Fe-Ni tape, even containing half of SiC addition, which may be due to a higher core density as shown later.

In order to evaluate the influence of rolling degree in $J_c$ at high fields, $J_c(\Theta=90^\circ)$ and $J_c(\Theta=0^\circ)$ vs. aspect ratio of the tapes at $H = 6.9T$ are shown in Figure 1 (b). Doped SiC samples with high aspect ratio ($b/a \geq 5$) have the highest $J_c$. In particular, $J_c$ increases by a factor of 10 at 4 K and 6.9 T when comparing T1 (undoped) and T2c, while it only varies less than a factor 2 when comparing different aspect ratios of doped samples. $J_c(\Theta=90^\circ)$ tends to saturate while $J_c(\Theta=0^\circ)$ decreases for values of $b/a \geq 5$ indicating that a higher rolling degree will not improve much further the superconducting properties.

![Graph a)](image1.png) ![Graph b)](image2.png)

Figure 1. (a) Transport Critical Current Densities ($J_c$) as function of applied field at 4 K for all samples described in Table 1. $J_c$ data of a 5%wt Si-doped tape from Ref [7] has been added for comparison. (b) $J_c(\Theta=90^\circ)$, $J_c(\Theta=0^\circ)$ as function of the aspect ratio of the tapes at 4 K, 6.9 T. The inset shows $\Theta$ definition.

The $J_c$ angular dependences of Si-doped samples measured at 4 K and 6.9 T are displayed in Fig. 2. The sample T2c has both the highest aspect ratio (8.1) and angular variation of $J_c$ in comparison with samples T2a and T2b. It can also be seen that the $J_{AF}=J_c(90^\circ)/J_c(0^\circ)$ continues increasing with the aspect ratio degree and does not saturate, in contrast with the $J_c(90^\circ)$ (see Fig. 1 (b)).

Figure 3 shows a detail of the X-ray diffraction (XRD) patterns of various tapes in comparison with the initial MgB$_2$ powder. The peak (100) decreases whereas the peak (002) increases with the aspect ratio indicating the presence of textured grains in direction (00l). Lotgering [14] proposed an expression for alignment degree using the integrated intensity of the x-ray reflections of a textured sample, and a simplified expression used by Handstein et al. [13] is:
where $I$ and $I_0$ are the correspond to the peak intensities of textured and isotropic samples, respectively. The inset of Fig. 3 displays $\tilde{A}$ values for the different tapes as a function of aspect ratio, which suggest a saturation for aspect ratios larger than 8. An increment in $J_{AF}$ for larger deformations which is not correlated with the alignment degree may indicate the presence of some correlated defects or a pinning decrease for $\Theta=0$. The second option is more plausible according to the results shown in Fig. 1(b). The reduced pinning may be explained by a diminution in grain boundary pinning for $H//T$ to the normal of the tape when MgB$_2$ plate-like shaped grains are oriented. This also explains the large increase in $J_{AF}$ (from 10 to 30) for larger starting powder particle sizes (from 10 to 50 $\mu$m) as reported by Lezza et al [6].

![Figure 2. $J_c$ angular dependence of of Si-doped samples measured at 4 K and 6.9 T.](image)

![Figure 3. X-ray diffraction patterns details for samples T1, T2c and the initial MgB$_2$ powder. The inset displays the texture anisotropy factor $\tilde{A}$ values (see text) as function of aspect ratio.](image)

The microstructural observation of the cross section core and MgB$_2$/SS interface along and perpendicular to the tapes has been done by SEM. Figures 4(a), (c) and (d) show SEM images of the core structure at the micron scale of samples T2a, T2b and T2c, respectively. It can be observed that the density and connectivity between grains increases as the aspect ratio of tapes increases. Sample T2a, with the lowest rolling degree still presents both porous and dense regions as illustrated in Figure 4(b), in contrast with the relatively flat and dense aspect of MgB$_2$ cores of tape T2b and T2c. The high density of these tapes is typical of ex-situ PIT and large hardness of sheath materials like SS. This important improvement of the MgB$_2$ core quality is believed to be responsible for the higher transport $J_c$ in these tapes. In contrast, in-situ PIT tapes showed high core porosity due to the volume shrinking taking place during the transformation of the Mg + B mixture into the MgB$_2$ compound.[7]

EDAX analysis and backscattered SEM images (see inset of Fig. 4(d)) indicate the absence of a significant diffusion in both directions across the interface. Another SEM observations of the cross section along the tape showed a homogeneous interface between the MgB$_2$ core and SS.
Figure 4. MgB$_2$ core SEM micrographs of T2a (a) and (b), T2b (c) and T2c (d) tape samples. The insets show SS/MgB$_2$ interface of secondary (c) and backscattered (d) electron images.

The $J_c$ angular dependences of sample T2c measured under different applied fields at $T = 4$ and 26 K are shown in Fig. 5. It is apparent that the $J_c$ anisotropy factor increases with applied field and temperature. A faster increase in the $J_c$ anisotropy factor with applied field was reported by Lezza et al [6] for undoped samples with an aspect ratio ~10. On the other side, samples with very low $J_{AF}$ field dependence have been reported by Kováč et al [5] for 10% wt SiC addition with an aspect ratio ~5. The $J_{AF}$ of our 2.5% Si-doped samples falls in between in agreement with the observed tendency. Therefore a degradation in the pinning for $\Theta=0$ and, consequently a faster increase in the anisotropy was observed for lower SiC addition and for higher rolling degree.

4. Summary
We prepared a series of samples to explore the influence of nano SiC addition and rolling degree on the $J_c$ angular dependence and texture of MgB$_2$ PIT tapes fabricated with stainless steel (SS) as sheath material. Alignment of the c-axis along the direction perpendicular to the tape were found in all samples as confirmed by X-ray diffraction. The alignment degree $\Theta$ in SiC samples augmented with increasing deformation but tends to saturate for aspect ratios larger than 5, while $J_c$ anisotropy factor $J_{AF} = J_c(90^\circ)/J_c(0^\circ)$ at 6.9 T continued increasing even for aspect ratio ~8.1. This increase is attributed to a pinning decrease in the applied field parallel to the tape direction. A faster increase in $J_{AF}(H)$ is observed for the undoped sample in agreement with previous studies. Besides, $J_{AF}$ field dependence also increases for Si-doped samples at high temperatures.
Figure 5. $J_c$ angular dependence of T2c sample measured under different applied fields at (a) $T = 4\, \text{K}$ (b) $T = 26.5\, \text{K}$. The inset shows (a) $J_{AF} = J_c(90^\circ)/J_c(0^\circ)$ as function of the applied field for sample T2c in comparison with T1 and others from Refs [4,5]; (b) $J_c(\Theta=90^\circ)$ and $J_c(\Theta=0^\circ)$ as a function of the applied field at 26.5 K.

We showed that very high $J_c$ and $J_e$ values ($J_c(6.9\, \text{T}, T=4\, \text{K}) \sim 10^4\, \text{A/cm}^2$ and $J_e =0.45\, J_c$) were obtained for Si-doped tapes with larger aspect ratios, whose cores are as dense as those from wires processed using hot isostatic pressing (HIPing). Therefore, there is an optimum rolling degree beyond which the superconducting properties cannot be further improved due to degradation in the pinning properties for $\Theta=0$ (with the applied field parallel to the tape direction).

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