Improved critical current property in MgB_2/Fe tapes by acetone doping

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1. Introduction

The discovery of superconductivity at 39 K in the MgB_2 compound has generated great interest in the field of applied superconductivity, especially in low temperature (<25 K) applications, such as a potential magnetic resonance imaging (MRI) magnet conductor [1]. A number of techniques have been developed to improve the J_c and the upper critical field (H_{c2}). It has been demonstrated that the J_c of MgB_2 can be significantly enhanced by carbon-based compounds doping [2, 3]. In these samples, C can be incorporated into the MgB_2 crystal lattice by replacing boron. Another technique to enhance the J_c is the ball-milling method [4], which is effective to refine the precursor powders, resulting in the decrease of MgB_2 grain size and the increase of flux pinning centers.

Recently, a combined process of chemical doping and mechanical ball milling has been conducted to significantly enhance the superconducting properties of MgB_2. The J_c of MgB_2 bulk prepared with both milled and glycerin-treated B was strongly increased [5]. However, for this method, the process of chemical doping and mechanical ball milling was complex, which increases the cost. Herrmann *et al* [6] obtained MgB_2 tapes with the excellent J_c properties by ball milling the nano-carbon with the precursor powders in Ar for 50h. However, it is a great challenge to achieve homogeneous distribution of a small amount of nano-additives and the matrix materials. The homogeneous doping and the excellent high-field performance were achieved by ball milling the starting powders within the liquid additives. More recently, we found that the J_c-B properties of Fe-sheathed MgB_2 tapes have been strongly enhanced by just ball-milling the precursor powders within acetone [7]. This process shows several advantages such as production of highly reactive C, avoidance of agglomeration for the starting powders, avoidance of the precursor powders adhering to the milling tools, easily filling into the metal tube and low cost. In this paper, we report our findings regarding the microstructure, magnetic field dependence of J_c at 4.2 K for acetone doped MgB_2 tapes.

2. Experimental

MgB_2/Fe tapes were prepared with spherical magnesium powders (10 μm, 99.5%) and boron powders (1-2 μm, 99.99%) by the in-situ PIT process. The magnesium powder and boron powder at chemical stoichiometry were thoroughly mixed within the acetone. The amounts of acetone (CH_3COCH_3) added into the powders were 0, 5, 10, and 15 wt.%, respectively. In order to avoid the evaporation of the acetone, the powders were mixed using the ball-milling method. The ball-milling process was carried out for 1 h with a rotating speed of 400 rpm under air in a planetary ball mill using agate vials and balls. The mixed powders were filled into pure Fe tubes of 8 mm outside diameter and 1.5 mm wall thickness in air. The tubes were subsequently rotate swaged, drawn and rolled into tapes. Finally, short samples of about 40 mm were heat-treated at 600-900°C for 1 h, and then followed by cooling down to room temperature in the furnace. The argon gas was allowed to flow into the furnace during the heat-treatment process to reduce the oxidation of the samples.

The phase identification was performed by x-ray diffraction (XRD) using Cu Kα radiation. For study of tapes, the Fe sheaths were mechanically removed to

expose the core. Microstructural observation was carried out by scanning electron microscopy (SEM) and transmission electron microscopy (TEM). The DC magnetization measurement was performed with a superconducting quantum interference device (SQUID) magnetometer. The T_c value was determined by taking the first deviation point from linearity that signifies the transition from the normal to superconducting state. The resistance measurements were carried out on a physical properties measurement system (PPMS) up to 9 T, using small pieces of a MgB_2 layer obtained by removing the sheath material. Transport critical current (I_c) of the samples were measured at 4.2 K and its magnetic field dependence were evaluated at High Field Laboratory for Superconducting Materials (HFLSM) in Sendai, by using a standard four-probe technique, with a criterion of 1 μV/cm. A magnetic field up to 14 T was applied parallel to the tape surface.

3. Results and discussions

Figure 1 shows the typical XRD patterns of the superconducting cores for MgB_2 samples with different acetone doping levels after heat-treatment. Clearly, MgB_2 is found to be the main phase in all samples, with minor impurity phases of MgO present. The dashed lines plotted in Fig. 1 show the position of the (002) and (110) peaks of pure MgB_2 sample. The position of the (002) peak shows almost no shift in acetone milled samples. Whereas, the position of (110) peak of all acetone doped samples shifts to higher angles, suggesting the decrease of the *a*-axis lattice parameter. The inset of Fig. 1 shows the lattice parameters obtained from Rietveld refinement. As can be seen, the *a*-axis lattice parameter of doped sample was reduced as compared to the pristine sample, while the *c* lattice parameter remained almost unchanged. The shrinkage of *a* lattice constant is attributed to the substitution of C for B. However, the decrease of the a-axis length is almost independent of the amount of acetone, indicating that adding more acetone did not proportionally lead to more carbon substitution. This result is in good agreement with the results reported by other groups through using benzene [8] as dopants.

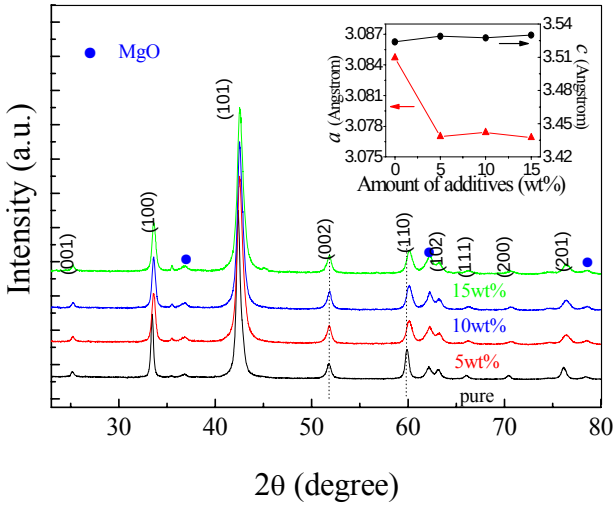

Fig. 1 XRD patterns of pure and doped tapes sintered at 700 °C. The inset shows the lattice parameters *a* and *c* for various acetone doping levels. The data were obtained after peeling off the Fe-sheath.

Table 1 summarizes the characteristic data of the pure and acetone doped samples. Assuming a linear dependence of the *a* parameter on carbon content, the actual carbon content, *x*, in the MgB_2 lattice was estimated by fitting our data to the Lee et al results [9]. As can be seen, the estimated actual amounts of carbon (x in $Mg(B_{1-x}C_x)_2$) within the MgB_2 lattice are approximately 0.017, 0.016 and 0.018 for 5, 10 and 15 wt.% acetone doped samples, respectively. The slight changes of the actual carbon content in the acetone-doped sample might be due to the experimental errors from both XRD measurement and Rietveld refinement. According to the formula of acetone (CH_3COCH_3), doping 5 wt.% acetone yielded a nominal composition of $MgB_2C_{0.119}$. Clearly, the actual carbon content incorporated into the MgB_2 lattice is much smaller than the nominal value. This suggests that a saturation of carbon substitution for boron might have been reached, which may also explain why increasing the amount of acetone to 15 wt.% did not lead to more carbon substitution for boron. Table 1 also shows that the full width at half maximum (FWHM) values of acetone-added samples are larger than that of pure one, indicating that the MgB_2 grain size was reduced by

acetone doping. Interestingly, only slight changes of FWHM values were observed as increasing the acetone doping level. This result suggests that the MgB_2 grain size seem not to decrease further with the increase of acetone amount. As shown in Table 1, the contents of MgO phase, as estimated by the peak intensity ratios, $I_{MgO(220)}/I_{MgB2(110)}$, increase with the acetone doping levels. If MgO impurities are present between MgB_2 grains as insulating precipitates, it will reduce the effective cross-sectional area of the sample, and thereby block supercurrents.

Table 1 Superconducting properties of tapes

Sample	*a*(Å)	*c*(Å)	Actual C in $Mg(B_{1-x}C_x)_2$	FWHM of (100)	T_c (K)	$I_{MgO(220)}/I_{MgB2(110)}$
Pure	3.0841(4)	3.5238(6)	-	0.322	35.63	0.427
5 wt.%	3.0770(6)	3.5289(9)	0.017	0.489	34.08	0.749
10 wt.%	3.0774(7)	3.5275(9)	0.016	0.514	33.58	0.770
15 wt.%	3.0769(7)	3.530(1)	0.018	0.503	33.08	0.861

Figure 2 summarizes the J_c at 4.2 K in magnetic fields up to 14 T for Fe-sheathed MgB_2 tapes with various amounts of acetone doping from 0 to 15 wt.% that were heat treated at 700 °C. It is noted that all the doped tapes showed an enhancement of J_c-B performance in fields above 9 T. The highest J_c value of the Fe-sheathed tapes was obtained at the 5 wt.% acetone addition. At 4.2 K, it reached up to 2.4×10^4 A/cm^2 at 10 T and 1×10^4 A/cm^2 at 12 T, respectively. This value was higher than most of the carbon compound added MgB_2 tape samples. And then further increasing the additive content caused a strongly reduction of J_c in magnetic fields below 14 T. More insulating MgO impurities may account for this.

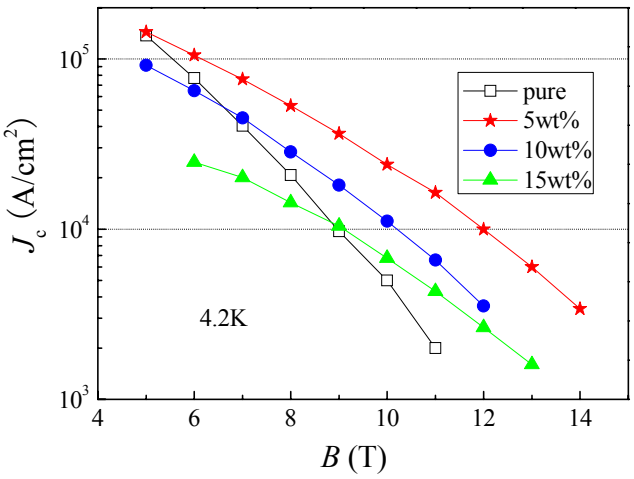

Fig.2 J_c–B properties of different acetone doped level samples heated at 700 °C.

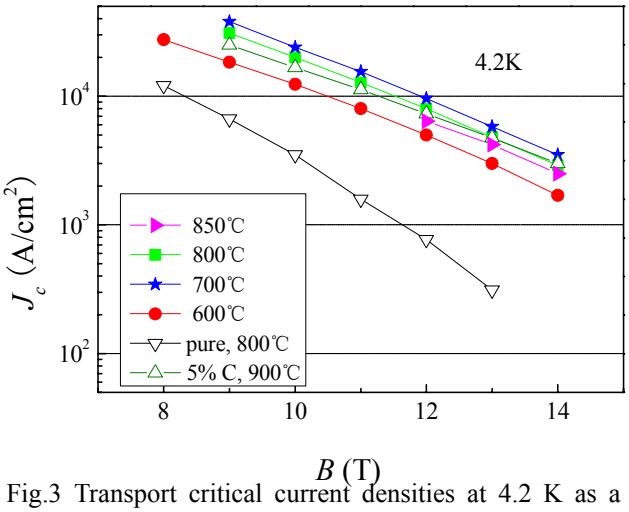

Fig.3 Transport critical current densities at 4.2 K as a function of magnetic field for MgB_2/Fe tapes milled with and without acetone. The measurements were performed in magnetic fields parallel to the tapes surface.

Figure 3 shows the transport J_c at 4.2 K for acetone doped samples heated at different temperatures ranging from 600 to 850 °C. The J_c value of 5 at % nano-C-doped tape made by ball milling without acetone is also included. It is noted that all the acetone doped samples sintered at different temperatures exhibited superior J_c values compared to the pure tape. The acetone doped sample sintered at 700 °C has the best J_c values in the magnetic field. At 4.2 K, the best J_c value reached 2.4×10^4 A/cm^2 at 10 T, which is even higher than that of the C-added samples heated at 900°C. At the same time, the acetone doped tapes showed a much smaller magnetic field dependence of J_c than the pure

samples. This means that effective pinning centers were introduced in acetone doped MgB_2 samples.

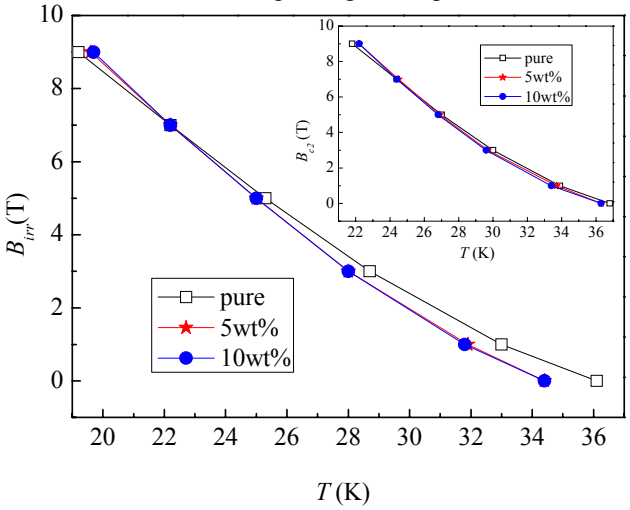

Fig.4 B_{irr} values as a function of the temperature for the different acetone doped level samples heated at 700 °C. Inset: variation of B_{c2} with temperature.

Figure 4 shows the temperature dependence of B_{irr} for the pure and acetone doped samples, obtained from the 10% values of their corresponding resistive transitions. It is noted that the B_{irr} of the acetone doped tapes increased more rapidly with decreasing temperature than that of the undoped one. Although the addition of acetone introduced a degradation of T_c, the acetone-doped tapes show a higher B_{irr} in the low temperature region. Additionally, B_{c2} has also been found to increase at temperatures below 24 K with acetone doping as shown in the inset of Fig. 4. The increase of B_{c2} is believed to be due to the substantial substitution of boron for carbon, as evidenced by the changes of the *a*-axis lattice parameter as compared to the pure sample. The large distortion of the crystal lattice caused by carbon substitution for B leads to enhanced electron scattering and enhancement of J_c.

It is also interesting to note from Fig. 4 that both the B_{irr} curves for 5 and 10 wt.% acetone doped tapes were nearly overlapped, indicating that the carbon substitution for B was almost the same with increasing the doping amount of acetone. This was confirmed by x-ray diffraction analysis (Fig. 1). No further changes in the *a*-axis lattice parameter, B_{c2} and B_{irr} are observed in 10 wt.% acetone-doped sample, suggesting that a saturated carbon substitution for boron has been reached in the 5 wt.% acetone doped sample. Thus, the electron scattering caused by carbon substitution for B was not enhanced further by increasing the acetone amount. In the meantime, according to the results of FWHM, the smaller MgB_2 grain size seems not to be obtained at higher acetone concentration (more than 10 wt.%). In contrast, more MgO contents could be seen for 10 and 15 wt.% acetone-added samples, which cause the J_c decrease especially in low magnetic field region.

4. Conclusion

In summary, we report the effect of acetone addition on *in situ* powder-in-tube processed MgB_2/Fe tapes by using acetone as liquid additive. The amount of acetone was varied from 0 to 15 wt.%. We found that a significant J_c-B enhancement was easily achieved in high field by acetone doping. At 4.2 K, the transport J_c for the best acetone added tapes (5 wt.%) reached up to 2.4×10^4 A/cm^2 at 10 T. With increasing acetone concentration, the *a*-axis lattice parameters, B_{c2} and B_{irr} were not much changed, however the insulating MgO impurities gradually increased, thereby resulting in the obvious decrease of J_c values for 10 and 15 wt.% acetone-added samples, especially within low field region.

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