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Enhancement of Critical Current Density and Flux Pinning in Acetone and La_2O_3 Codoped MgB_2 Tapes *

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MgB_2 tape samples with simultaneous additions of acetone and La_2O_3 were prepared by an in-situ processed powder-in-tube method. Compared to the pure and single doped tapes, both transport J_c and fluxing pinning are greatly improved by acetone and La_2O_3 codoping. Acetone supplies carbon into the MgB_2 crystal lattice and increases the upper critical field, while the La_2O_3 reacts with B to form LaB_6 nanoparticles as effective flux pining centers. The improvement of the superconducting properties in codoped tapes can be attributed to the combined effects of improvement in H_{c2} and flux pinning.

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The $J_c - H$ performance of MgB_2 has been improved greatly by a number of experimental techniques since its discovery as a superconductor in 2001.[1−4] However, the critical current density J_c values of MgB_2 tapes are still below the practical level due to the low upper critical field (H_{c2}) and poor flux pinning. Therefore, it is necessary to increase the critical current density to develop useful MgB_2 superconducting wires for practical applications. Up to now, doping with carbon seems to be most effective to improve the H_{c2}. The critical current density J_c of MgB_2 tapes can easily exceed the level 10^4 A/cm^2 at 4.2 K and 10 T through chemical doping with carbon containing materials, such as nano-SiC, nano-C, and carbohydrate.[2−6] Recent results[7] show that the $J_c - H$ performance of MgB_2 wires and tapes with carbohydrate dopant can be even better than nano-SiC and nano-C dopants. It is believed that the enhancement of superconducting properties should be mainly attributed to the effective substitution B by the highly reactive C released from carbohydrate decomposition. On the other hand, the nano and submicron particles of rare earth (RE) oxides such as Y_2O_3, Dy_2O_3 and Ho_2O_3 have been used to introduce the nano-pinning-centers,[8−10] which significantly improve the pinning behavior of MgB_2.

The substitution of boron by carbon is known to enhance the critical field of MgB_2. In addition, the pinning behavior is expected to be improved by doping of RE oxides. Therefore, in order to study the combined effect of both C substitution and RE oxides doping on the structural and superconducting properties of MgB_2, we have synthesized the acetone and La_2O_3 codoped MgB_2 tapes via an in-situ powder-in-tube method. We find that the transport $J_c - H$ properties of MgB_2 are greatly improved by the simultaneous additives.

A range of MgB_2 tapes was prepared as follows: (A) no additions, (B) addition of 5 wt% La_2O_3, (C) 10 wt% acetone and (D) double additions: 5 wt% La_2O_3+10 wt% acetone. For samples A and B, the raw powders were ground by hand with a mortar and pestle for one hour. In order to avoid the evaporation of the acetone, the raw powders of C and D were mixed using the ball-milling method. The ball-milling process was carried out for 30 min in a planetary ball mill using agate vials and balls. These mixed powders were packed into Fe tubes. After packing, the tube was rotary swaged and then drawn to wires of 1.75 mm in diameter. The wires were subsequently rolled to tapes. These tapes were heated at 700°C for 1 h, and then followed by a furnace cooling to room temperature. The high purity argon gas was allowed to flow into the furnace during the heat-treatment process to reduce the oxidation of the samples.

The phase identification and crystal structure investigation were carried out using x-ray diffraction (XRD). Microstructure was studied using a scanning electron microscope (SEM). The dc magnetization measurements were performed with a superconducting quantum interference device (SQUID) magnetometer. The transport current I_c at 4.2 K and its magnetic field dependence were evaluated at the High Field Laboratory for Superconducting Materials (HFLSM) at Sendai, by a standard four-probe resistive method, with a criterion of 1 μV·cm^{-1}, a magnetic field up to

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14 T was applied parallel to the tape surface. For each set of doped tapes, the I_c measurement was performed for several samples to check the reproducibility.

Figure 1 shows the results of x-ray diffraction measurements for the samples studied in this work. For the undoped and acetone doped samples, MgB_2 was obtained as a main phase with less amounts of MgO. LaB_6 and MgO were detected as impurity phases in La_2O_3 doped and La_2O_3+acetone codoped samples. It can be concluded that the La_2O_3 can react with B to form LaB_6. No La_2O_3 can be detected, suggesting that the reactions of Mg and B with La_2O_3 are nearly complete.

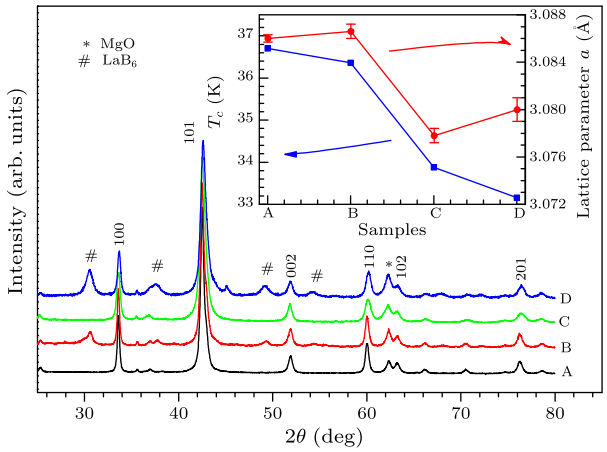

Fig. 1. XRD patterns of in situ processed pure and doped tapes sintered at 700°C. Inset: the variation of lattice parameter a and critical temperature for different samples.

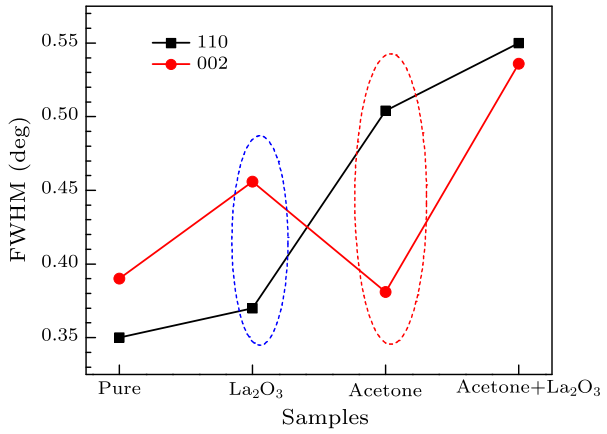

Fig. 2. FWHM values of various diffraction peaks for the samples.

The variation of the lattice parameter a and critical temperature as a function of the additives are shown in the inset of Fig. 1. The obvious decrease of a-axis lengths for the acetone doped and La_2O_3+acetone codoped tapes indicates that carbon has been doped into the B sites in the crystal lattice and caused the reduction in T_c. The substitution of boron by carbon is known to enhance the electron scattering, and in turn, to improve the critical field. Besides a slight enlargement of c lattice parameter in the La_2O_3+acetone codoped tapes, the c lattice parameters obtained from Rietveld refinements did not almost change within the limit of calculation error for other studied samples. The reason of small change in the c lattice parameter of La_2O_3+acetone codoped samples is unclear now. This may be due to the strain induced by the nanoparticles, or the co-substitution effects. Figure 2 shows the FWHM values of various diffraction peaks as a function of samples. It can be seen that the FWHM values of peak (110) and (002) were selectively improved by acetone and La_2O_3, respectively. While the codoped sample exhibits a combination effect. The broadening in peaks may suggest the occurrence of grain size refinement or lattice distortion in out-of-plane and in-plane at the same time.

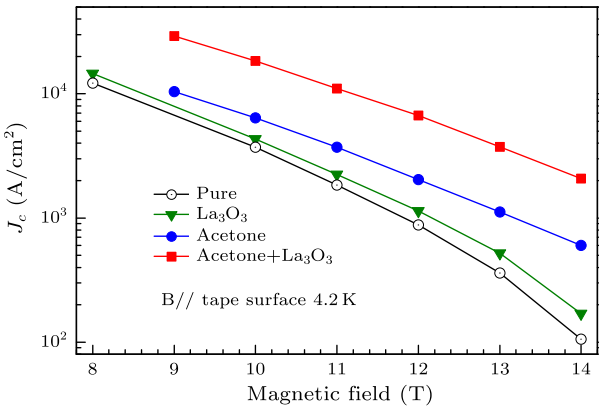

Fig. 3. $J_c - H$ properties of Fe-sheathed pure and doped tapes. The measurements were performed in magnetic fields parallel to the tapes surface at 4.2 K.

Figure 3 shows the transport J_c at 4.2 K in magnetic fields up to 14 T for all MgB_2 tapes. It can be seen that all the doped samples exhibit superior J_c values compared to the pure one. Even for the sample doped with 5 wt% La_2O_3, it still shows less sensitivity of J_c to the magnetic field. This suggests that effective flux pinning centers are possibly introduced by the La_2O_3 doping, which increases the J_c performance of MgB_2 tapes in higher magnetic fields. The striking result of Fig. 3 is that the La_2O_3+acetone codoped sample exhibits the cumulative effects of La_2O_3 and acetone. For example, at 4.2 K and in a field of 14 T, the J_c value of the codoped sample reaches 2.1×10^3 A/cm^2, i.e. 12 and 3.5 times larger than those of La_2O_3 doped and acetone tapes, respectively.

To confirm the enhanced flux pinning ability in doped MgB_2 tapes, Fig. 4 presents the normalized volume pinning force $F_p(B)/F_p^{\max}$ as a function of magnetic field at 5 and 20 K. It is clear that the pinning force of all the doped samples shows higher vortex pinning strength at higher magnetic fields than the pure samples. The samples with the acetone and La_2O_3 codoping show the highest flux pinning force among the samples studied, consistent with the excel-

lent $J_c - H$ performance of the sample as shown in Fig. 3.

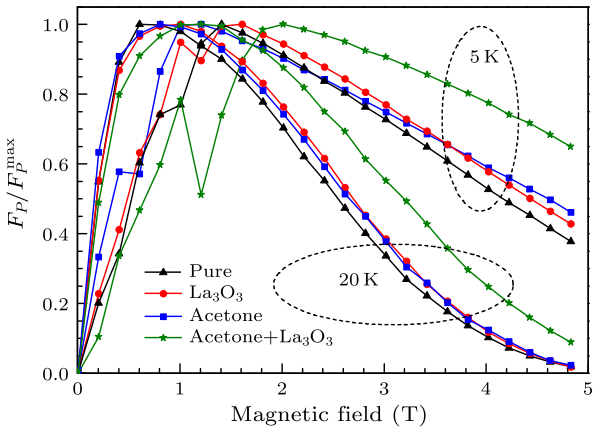

Fig. 4. The normalized volume pinning force ($F_P/F_P^{\max}$) versus magnetic field at 5 K and 20 K for pure and doped tapes.

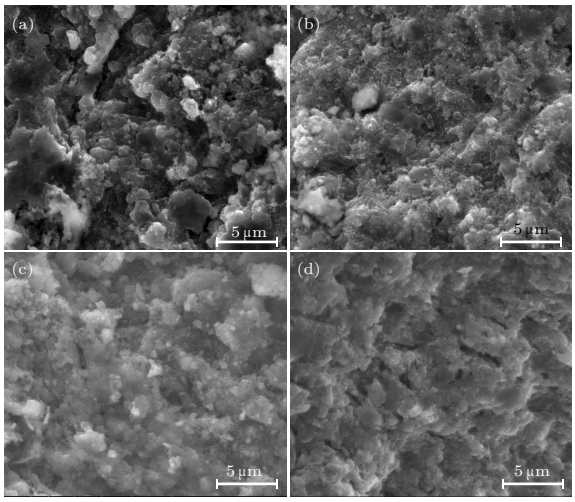

Fig. 5. SEM images for the pure (a), 5 wt% La_2O_3 (b), 10 wt% acetone (c) and 5 wt% La_2O_3+10 wt% acetone codoped samples (d) after peeling off the Fe sheath.

Figure 5 shows the typical SEM images of the fractured core layers for pure and doped tapes. From the images, it can be seen that the MgB_2 core of undoped sample is quite inhomogeneous with lots of voids. However, both doped tapes have a quite uniform microstructure with few voids, which may improve the J_c values.[14] Furthermore, the grain size of MgB_2 is reduced by doping. The reduction in grain size is probably explained by the inhibition of grain growth by finely second phases introduced during the MgB_2 phase formation. Obviously, the fine grains mean more grain boundaries, which can act as strong pinning centers,[15] resulting in better J_c performance.

The improvement of $J_c - H$ performance in the codoped samples can be attributed to the combinations of improvement in upper critical field and flux pinning. Doping with carbon containing materials is the most effective way to improve the $J_c - H$ performance of MgB_2.[2–7] When acetone is heated above the decomposition temperature, the highly reactive C released from decomposition can effectively substitute into B sites. Substitution of C for B induces disorder on the lattice sites, which leads to the enhancement of the H_{c2}. On the other hand, the reaction induced nanosize precipitates LaB_6 and/or the MgO with a size smaller than the coherence length of MgB_2 can act as effective flux pinning centers and hence improve the J_c performance of codoped samples. Therefore, a combination of C substitution induced H_{c2} enhancement as well as the strong flux pinning centers induced by nanosize precipitates are responsible for the superior $J_c - H$ performance of the codoped samples.

In summary, we have demonstrated that codoping of acetone and La_2O_3 is a promising way to enhance the J_c–H properties of MgB_2 tapes. The improvement in J_c performance mainly comes from both the pinning and scattering enhancements. A further enhancement of transport J_c is expected when using an optimized doping level or different combinations of additives.

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