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Enhanced Critical Current Density of MgB_2 Superconductor Synthesized in High Magnetic Fields

Yanwei MA*, Aixia XU, Xiaohang LI, Xianping ZHANG, Satoshi AWAJI[1] and Kazuo WATANABE[1]

Applied Superconductivity Lab., Institute of Electrical Engineering, Chinese Academy of Sciences, P.O. Box 2703, Beijing 100080, China
[1]*High Field Laboratory for Superconducting Materials, Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan*

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The effect of high magnetic fields on the current carrying properties of magnesium diboride (MgB_2) bulks and Fe-sheathed tapes was investigated following different thermal sequences. It is found that application of a large magnetic field during processing results in the quite uniform microstructure and the better connectivity between the MgB_2 grains. As a result, the J_c of these samples has shown much higher value than that of the MgB_2 samples in the absence of magnetic field. The possible mechanism of the J_c enhancement under an external magnetic field is also discussed. [DOI: 10.1143/JJAP.45.L493]

KEYWORDS: MgB_2, magnetic field processing, grain connectivity, critical current density

Magnesium diboride (MgB_2) with its superconducting transition temperature at 39 K has generated much interest in promising applications potential in high field magnets and magnetic resonance imaging.[1] At present, research efforts have been directed towards either improving critical current density (J_c) by improving grain connectivity, or improving in-field performance through introducing pinning centers. To achieve such goals, doping with elements or compounds,[2–7] hot isostatic pressing,[8] and irradiation with heavy ions[9] has been investigated.

Processing in an external magnetic field is a well-proven technique to enhance the degree of grain alignment and critical current density for the case of high-T_c oxide superconductors (HTSs).[10–12] The mechanism operating to achieve such improvement is understood to be due to the anisotropy in the paramagnetic susceptibility. Hence, when high-T_c superconductors are placed in a magnetic field in its normal state, the magnetic energy is minimized when the axis of maximum susceptibility is parallel to the magnetic field. On the other hand, MgB_2 exhibits a strong anisotropy in the B–B lengths: the distance between the boron planes (c-axis) is significantly longer than the in plane B–B distance (a-axis).[1] Furthermore, it is known that the J_c properties of MgB_2 are quite sensitive to preparation and annealing conditions. If a magnetic field is applied during the MgB_2 fabrication process, the enhancement of critical current density and other field effects are expected. In this letter, we have found that the magnetic field sintering (MFS) effectively improves the current carrying properties of both MgB_2 bulks and Fe-clad tapes.

Powders of Mg (99.8%, 325 mesh) and B (amorphous, 99.99%) were well mixed and ground in air for 1 h, using an agate mortar and pestle. Pellets 10 mm in diameter and 2 mm in thickness were made under uniaxial pressure. Fe-sheathed MgB_2 tapes with a thickness of ~0.5 mm and a width of ~3.5 mm were prepared by the standard powder-in-tube method. Subsequently, the pellet or tape samples were wrapped in Zr foil, placed in a vertical tube furnace, then heated in vacuum in applied magnetic fields (H_a) up to 14 T following different thermal sequences. As for the tapes, several groups of samples were prepared in this experiment, based on that the surface of the tapes was oriented whether parallel to or perpendicular to the magnetic field. A description of the samples is presented in Table I.

The samples were sintered in an electrical furnace in a vacuum of about 10^{-4} Pa. The furnace was installed in a 15 T cryogen-free type superconducting magnet.[13] For the samples sintered in a magnetic field, first, the magnetic field was raised up to the set value; second, the temperature was heated up to the target value and the samples were sintered at the set temperature in a magnetic field for 1 h; then the samples were furnace-cooled down to room temperature; finally the magnetic field was decreased to zero. MgB_2 samples were also prepared in the absence of a magnetic field and used as the standard. Several different samples were made in separate reaction runs to check for reproducibility.

Phase identification was performed by X-ray diffraction (XRD) using Cu Kα radiation. For study of tapes, the Fe sheaths were mechanically removed by peeling off the sheath to expose the core. Microstructural observation was carried out by scanning electron microscopy (SEM). Transport critical current densities (J_c) of some tape samples were measured at 4.2 K using a conventional four-probe method. The criterion for the I_c definition was 1 μV/cm. A magnetic field was applied parallel to the tape surface. The hysteretic magnetization ΔM of samples was also measured in a superconducting quantum interference device magnetometer, from which the critical current density J_c was calculated assuming fully connected samples using the extended bean model: $J_c = 20\Delta M/[a(1 - a/3b)]$ where a and b are the width and thickness of a rectangular section bar.

Figure 1 shows the XRD patterns of the superconducting cores of Fe sheathed tapes processed in magnetic fields H_a of 0 and 10 T at 600 °C for 1 h (Group I). As we can see, both the zero field and 10 T samples compose of almost a single phase of MgB_2 containing a small amount of MgO. However, the relative intensities of the (001) and (002) diffraction peaks from MgB_2 for the 10 T tapes are lower than those of the 0 T samples. These results indicate that the c-axis grain alignment of MgB_2 was deteriorated by the magnetic field sintering.

Figure 2 shows the transport critical current densities at 4.2 K as a function of magnetic fields for our 0 and 10 T processing MgB_2/Fe tapes. The tapes processed in a 10 T magnetic field exhibit higher J_c values than those processed

*E-mail address: ywma@mail.iee.ac.cn

Table I. Description of samples used in this work.

Group	Sample type	Sintering temperature/time	Applied field during sintering	Sample surface and field direction during sintering	T_c (K)
Group I	Fe clad tape	600 °C/1 h	10 T	Parallel	35.2
	Fe clad tape	600 °C/1 h	0 T		35.5
Group II	Fe clad tape	700 °C/1 h	14 T	Perpendicular	—
	Fe clad tape	700 °C/1 h	0 T		—
Group III	Pellet	800 °C/1 h	8 T	Perpendicular	36.9
	Pellet	800 °C/1 h	0 T		37.1

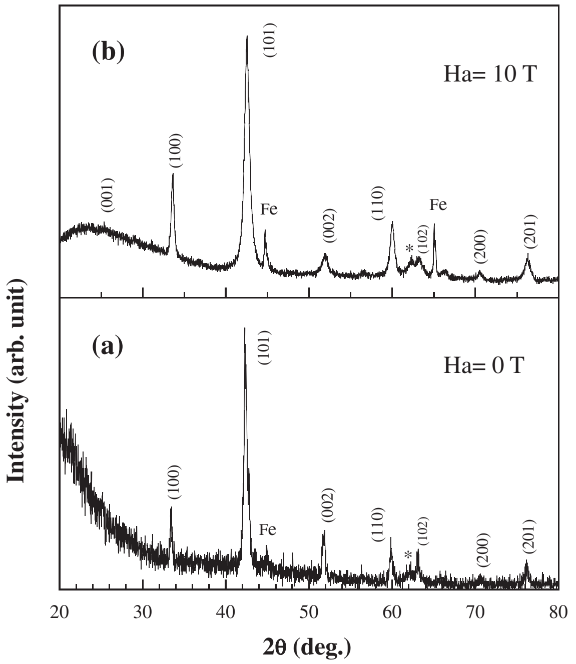

Fig. 1. X-ray diffraction patterns of the MgB_2 tapes processed in magnetic fields H_a of (a) 0 and (b) 10 T. The data were obtained after peeling off the Fe-sheath. The peaks of MgO are marked by asterisks. The peaks of Fe were contributed from the Fe sheath.

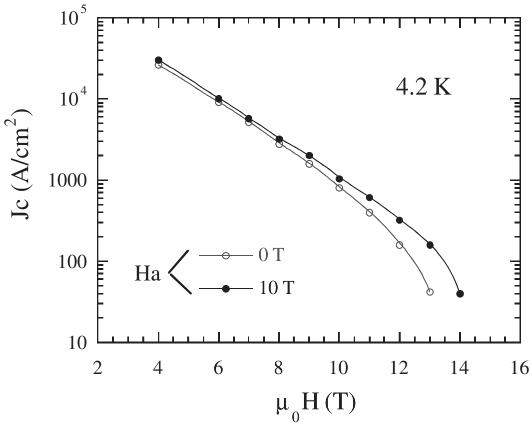

Fig. 2. Field dependence of the transport critical current density at 4.2 K for the tapes processed in magnetic fields H_a of 0 and 10 T. The measurements were performed in magnetic fields parallel to the tape surface.

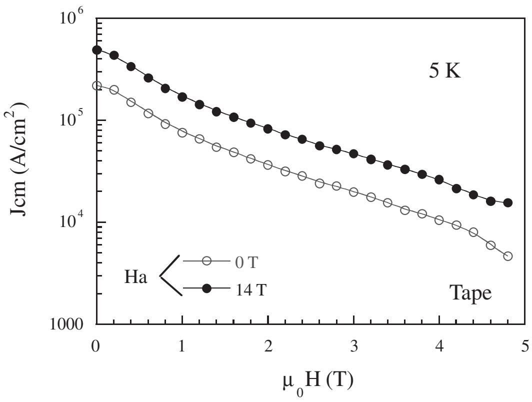

Fig. 3. Magnetic J_c dependence at 5 K for the tapes processed in magnetic fields H_a of 0 and 14 T, as measured by magnetization. The measurements were performed in magnetic fields parallel to the tape surface.

under a zero magnetic field, although the J_c difference is small in magnetic fields below 10 T. In particular, it is evident that the field dependence of J_c was decreased by sintering in a magnetic field for Fe sheathed tapes, namely, higher J_c in high-field region. Note that the similar result was found for the tapes made in a 14 T field, when sample's surface was placed parallel to the field direction (not shown in Table I). Furthermore, magnetization data reveal that compared to the 0 T tape, the critical temperature T_c for the samples processed in a 10 T magnetic field slightly decreased by 0.3 K. The small decrease of T_c is likely due to the slightly poor crystallinity originating from magnetic field processing, as supported by weaken XRD patterns with broad peaks.

To clarify the influence of field direction on the tape surface during MFS processing, we set horizontally the Fe sheath tapes in the furnace and obtained the Group II samples. Figure 3 shows the magnetic $J_c(B)$ curves for the MgB_2 tapes processed in magnetic fields H_a of 0 and 14 T at 700 °C for 1 h. It is noted that the tapes processed in a 14 T magnetic field has better $J_c(B)$ performance, more than two times higher than the 0 T ones. In order to investigate the reason for the J_c improvement, we studied the difference in the microstructure of the tapes with and without magnetic field. Figure 4 shows the typical SEM images of the fractured core layers for the 0 and 14 T samples. Clearly, well-developed grains can be seen in both samples. However, the 0 T sample shows a broader grain size distribution and the core is quite porous and loose [see Fig. 4(a)]. The porous microstructure of MgB_2 directly means a reduction of the effective current path. In contrast, with the application of strong magnetic field, the field sample has fewer pores and seems very dense and consequently the connections

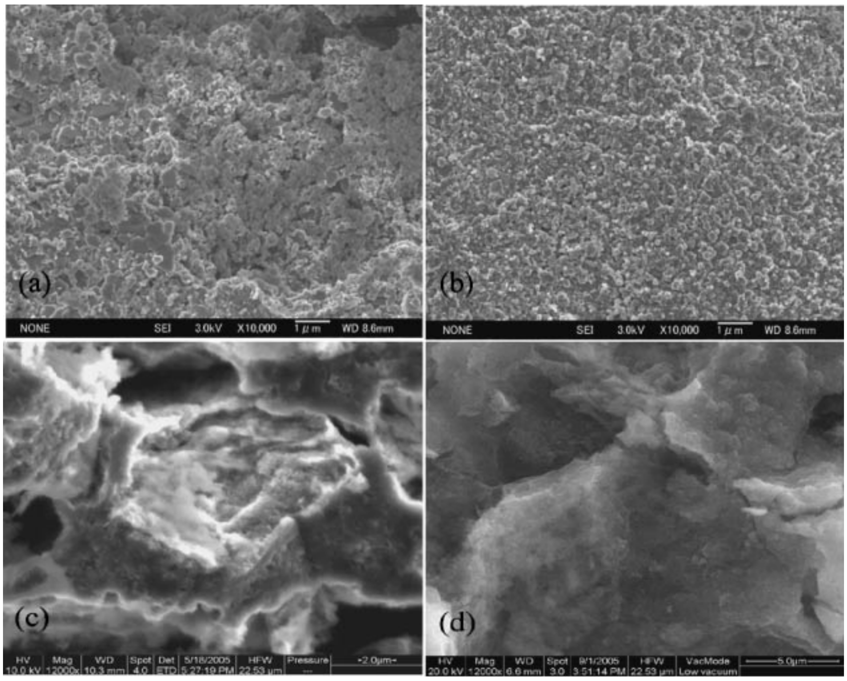

Fig. 4. SEM micrographs of samples which were processed in different magnetic fields. (a) 0 T tape and (b) 14 T tape; (c) 0 T bulk and (d) 8 T bulk.

between grains are much improved. Moreover, the quite uniform microstructure of the MgB_2 core is observed [see Fig. 4(b)]. As were demonstrated earlier,[14] the high-density MgB_2 samples with less voids have high superconducting homogeneity and strong intergranular current flow as determined by magneto-optical studies. The fact was also corroborated by many recent results, in which the J_c enhancement of MgB_2 was achieved by the improvement in the grain coupling as a consequence of densification of the tape core.[3,7,8]

It is interesting to note by comparing Figs. 2 and 3, the effect of a magnetic field seems different between Group I and II samples. For Group I (the applied field was in the tape plane), although the enhanced J_c–B characteristic was observed in high field region, however, the J_c improvement in low field area is small. On the other hand, the improved J_c by more than a factor of 2 for the field tapes of Group II was achieved. This indicates that the magnetic field works more effectively to enhance the J_c–B properties when the direction of applied fields was perpendicular to the tape surface (Group II) during processing.

To further confirm the enhanced J_c properties with the magnetic field, we also prepared the MgB_2 bulk samples using the *in situ* reaction in a magnetic field 800 °C for 1 h. Figure 5 presents the magnetic $J_c(B)$ data at 5, 20, and 30 K for the MgB_2 pellets processed in magnetic fields H_a of 0 and 8 T. It is noted that the 8 T field produced a stronger enhancement of J_c than without the field at all temperatures and in the entire field region. The 8 T bulks show the slightly degraded T_c compared to the 0 T ones, as given in the Table I. From the SEM observations for all bulk samples [Figs. 4(c) and 4(d)], it is clear that the microstructure development with the magnetic field is consistent with the tape case. Thus, we conclude that when the MgB_2 pellets processed in a magnetic field, J_c is also significantly

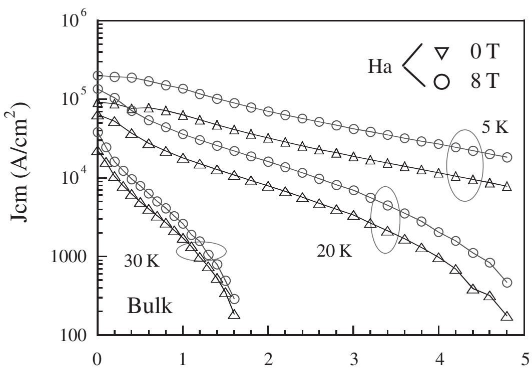

Fig. 5. Magnetic J_c dependence at 5, 20, and 30 K for the pellet samples processed in magnetic fields H_a of 0 and 8 T. The measurements were performed in magnetic fields parallel to the bulk surface.

improved due to the denser microstructure and the improved connectivity of MgB_2 grains. More details about bulks heat-treated in a magnetic field will be published elsewhere. Combined with the results of bulk and tape samples sintering in magnetic fields, our present study demonstrates that the magnetic field does enhance the J_c properties in MgB_2 significantly.

To understand the mechanism behind the J_c improvement, one may consider first the crystal orientation effect of MgB_2 during the magnetic field processing, since like HTS, the structure of MgB_2 is also strongly anisotropic. In the process of crystal growth in a magnetic field, if the anisotropic magnetic energy of crystal $\Delta E = \Delta\chi V H_a^2/2$, which promotes crystal alignment magnetically, exceeds the thermal energy, the grain alignment is obtained,[10] where V is the volume of a grain and $\Delta\chi$ is the anisotropy of the paramagnetic susceptibility. Thus, it is expected that c-axis grain alignment of the MgB_2 core might be preferred by the strong magnetic field imposed. However, studies of the magnetism of the normal state of MgB_2 show that the net susceptibility is very small, of the order of 10^{-6} emu/mol.[15] Furthermore, the average grain size of MgB_2 in the present work is quite small (about 200 nm), while the HTS has the grains with a size of $\sim$20–50 µm.[12] These suggest that the MgB_2 crystal orientation effect due to the applied magnetic field would be tiny, compared to HTSs. Other beneficial effects such as densification and homogenization of crystallites caused by Lorentz force and magnetization force must be considered.[16,17] Therefore, the clear J_c enhancement in MgB_2 is mainly due to the well-connected grains of uniform size as a consequence of densification of the core during magnetic sintering.

As revealed by microstructural analyses, it is evident that an external magnetic field can enhance the densification and the homogeneity of superconducting cores. In particular, the magnetic field effect seems to be of great significance for Group II when the applied field was perpendicular to the tape plane. We can see that the density of pores in the magnetically sintered sample are much less than that in the 0 T one. Similar results were also found by Tsurekawa *et al.*[18] when sintering iron samples in the presence of a magnetic field. This is probably because a magnetic field provides an extra driving force for grain boundary migration to break away the dragging pores. Another interesting feature is that the grain structure seems to be homogeneous, suggesting the abnormal grain growth was effectively prevented by a magnetic field.[19] Therefore, the applied magnetic field may provide a driving force for grain boundary migration that greatly contributes to densification during sintering, leading to better connections between grains, hence the J_c enhancement.

In summary, MgB_2 bulks and tapes were prepared in high magnetic fields up to 14 T. Enhanced J_c properties in comparison with their zero-field counterpart were observed. Application of a large magnetic field during processing produced the uniform and dense microstructure in MgB_2, leading to increased critical current density J_c. It is suggested that an external magnetic field is responsible for an increase in the driving force for grain boundary migration that greatly contributes to densification. The present study demonstrated that the magnetic field is very effective on yielding MgB_2 superconductors with the enhanced J_c properties.

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