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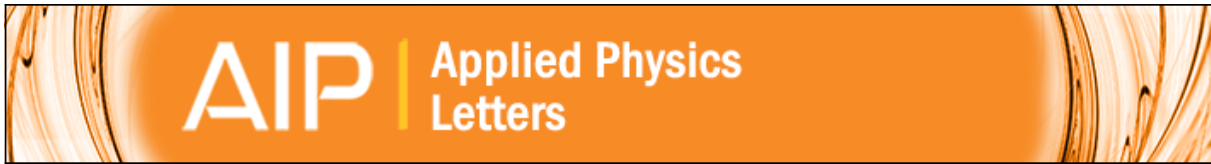
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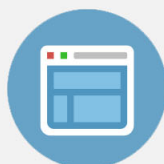
Magnetic field processing to enhance critical current densities of MgB$_2$ superconductors

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A magnetic field of up to 12 T was applied during the sintering process of pure MgB$_2$ and carbon nanotube (CNT) doped MgB$_2$ wires. The authors have demonstrated that magnetic field processing results in grain refinement, homogeneity, and enhancement in $J_c(H)$ and H_{irr}. The extent of improvement in J_c increases with increasing field. The J_c for a 10 T field processed CNT doped sample increases by a factor of 3 at 10 K and 8 T and at 20 K and 5 T, respectively. H_{irr} for the 10 T field processed CNT doped sample reached 9 T at 20 K, which exceeded the best value of SiC doped MgB$_2$ at 20 K. Magnetic field processing reduces the resistivity in CNT doped MgB$_2$, straightens the entangled CNTs, and improves the adherence between CNTs and the MgB$_2$ matrix. © *2006 American Institute of Physics.* [DOI: 10.1063/1.2388126]

MgB$_2$ superconductor has made a significant impact on superconductivity research and development since its discovery.[1] Chemical doping has been used to enhance the critical current density J_c and the upper critical field H_{c2}.[2–5] Carbon and silicon carbide doping resulted in a significant increase of in-field J_c and H_{c2}, and these records still stand for MgB$_2$.[5–7] To further advance the development of MgB$_2$ for applications we report a method of combining the magnetic field processing and carbon nanotube (CNT) doping. Magnetic field processing technology has proved to be a powerful tool to produce aligned carbon nanotubes (CNTs) in composites and macroscopic membranes[8,9] and control the phase transformation and behavior of the melts during condensation processes, resulting in major improvements in material properties.[10,11] Magnetic field processing has also been used to achieve the desired texture and improved J_c performance in high temperature superconductors.[12–15] In processing of MgB$_2$ bulk and wires the reaction *in situ* technique in combination with the powder-in-tube method has been used to produce the wires with the best field performance.[16] In the *in situ* reaction process, Mg melts before the MgB$_2$ formation by solid state reaction, provided that the heating rate is high enough. The presence of a liquid phase provides a window of opportunity for applying a magnetic field processing technique to achieve crystalline refinement, homogeneous distribution of additives and inclusions, and possible alignment of both matrix materials and additives.

In this work, a standard powder-in-tube method was used for Fe clad MgB$_2$ wire.[17] Powders of magnesium (99%) and amorphous boron (99%) were well mixed with 0 and 10 wt % of multiwall CNTs [outside diameter (o.d.) of

20 nm and length of 0.5–2 μm] and thoroughly ground. The Fe tube had an o.d. of 10 mm and a wall thickness of 1 mm and was 10 cm long with one end of the tube sealed. The mixed powder was packed into the tube, and the remaining end was blocked using an aluminum bar. The composite was drawn to a 1.4 mm diameter wire. Several short samples 2–3 cm in length were cut from the same wire. Some wires were rolled into tapes for study of anisotropy. These pieces were sealed in Fe tubes, then sintered in a tube furnace at 800–900 °C for 30 min in magnetic fields up to 12 T (H_a), with a high heating rate of 20 °C/min, and finally furnace cooled to room temperature. The processing field was applied parallel or perpendicular to the wire axis before the temperature was ramped up. The same processing conditions were used to treat the wire samples with field on and field off for comparison.

The magnetization of cores taken from the wires was measured at 5 and 20 K using a Quantum Design Physical Property Measurement System with a magnetic field sweep rate of 50 Oe/s and amplitude up to 9 T. Since there is a large sample size effect on the magnetic J_c for MgB$_2$ fabricated by the reaction *in situ* process, all the samples for

TABLE I. Transition temperature, resistivity, and lattice parameters for undoped MgB$_2$ and CNT doped MgB$_2$ wires with and without field processing.

Sample	T_c (K)	$\rho_{40\ \mathrm{K}}$ ($\mu\Omega$ cm)	$\rho_{300\ \mathrm{K}}$ ($\mu\Omega$ cm)	RRR
MgB$_2$ (0 T)	37.7	28.0	63.0	2.2
MgB$_2$ (5 T)	37.7	23.5	58.0	2.4
CNT (0 T)	35.6	50.8	81.0	1.6
CNT (10 T)	36.7	34.3	63.0	1.9
CNT (0 T)	35.4	61.6	96.6	1.6
CNT (12 T)	36.1	43.5	75.6	1.7

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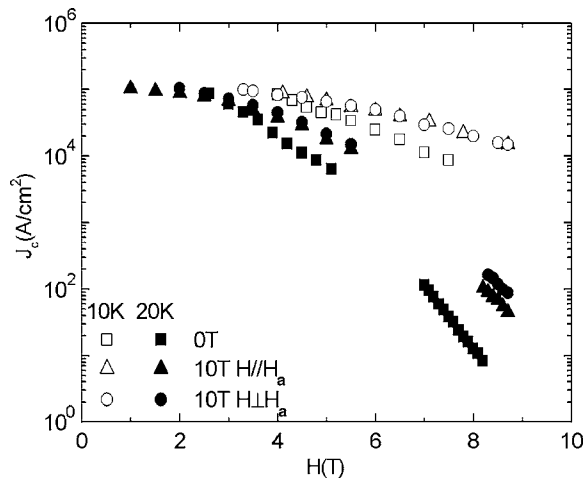
FIG. 1. J_c vs H for 10% CNT doped MgB$_2$ tapes, processed at 900 °C for 30 min without magnetic field and with a 10 T field, respectively.
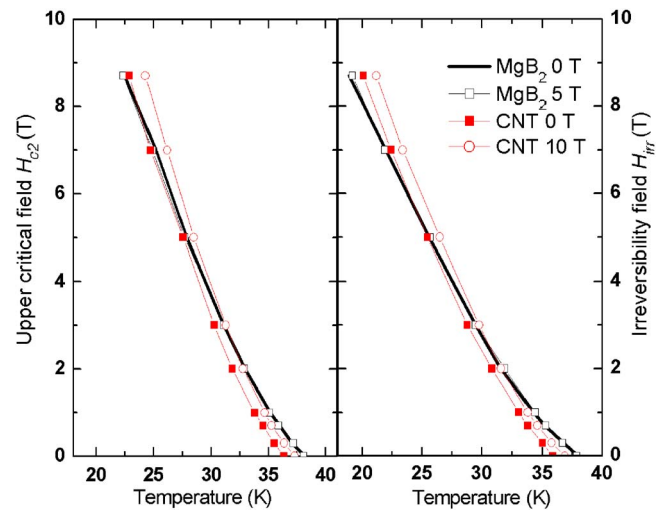

FIG. 2. (Color online) Upper critical field H_{c2} (a) and irreversibility field H_{irr} (b) as a function of temperature for undoped and 10% CNT doped MgB$_2$ wires.

measurement were made to the same size for comparison. The magnetic J_c was derived from the height of the magnetization loop using the critical state model. Samples were measured with the field H applied perpendicular and parallel to the wire axis. The transport J_c was measured with the four-probe method, using a pulsed current source for critical currents down to about 70 A. In the high field region, which was characterized by critical currents lower than 1 A, a dc source was used to measure the transport J_c.

Table I lists the basic data for the undoped MgB$_2$ and 10 wt % CNT doped MgB$_2$ wires processed with and without magnetic field. The T_c for the undoped MgB$_2$ remained unchanged with field processing, while there is a small increase in the T_c for the two pairs of CNT doped samples which were processed without applied field and with 10 and 12 T fields, respectively. The resistivity was reduced by 20%–30% with field processing for both the undoped and CNT doped samples. Accordingly, there is an increase in the residual resistivity ratio (RRR) value for both samples. There is no noticeable change in lattice parameters for both the undoped and CNT doped samples with field processing. The interaction between magnetic field and materials will generate magnetization force, which can act as pulling force for magnetic and paramagnetic materials or repulsing force for diamagnetic materials. The magnetization force will also tend to rotate the crystals to align with the field direction if there is anisotropy in the overall magnetic susceptibility. As a result of these forces the magnetic processing has several beneficial effects, including improved interaction between components, reduced size of crystallites, homogenization, reduction of impurities, and alignment of parent or additive materials. The observed reduction in resistivity for both the undoped and CNT doped samples may be attributable to the improvement in convection and diffusion of components in the applied magnetic fields during Mg melting. There are no data on the magnetic property at high temperatures for Mg, B, and CNT. At room temperatures, B and MgB$_2$ were found to be diamagnetic,[18,19] while CNT could be diamagnetic or paramagnetic depending on the helical structure, magnetic field, and orientation.[20–22] As many metals, Mg could become paramagnetic in strong fields. It is evident that the magnetization force on B, CNT, and Mg will be different. It is the difference in magnetization force that will increase the migration among these components and hence improve the

interaction between them, which, in turn, enhance the crystallinity and grain connectivity.

Figure 1 shows the transport $J_c(H)$ curves for the 10% CNT doped MgB$_2$ tape samples processed without field and with a 10 T field at 900 °C. A pronounced J_c enhancement was observed in the entire field region at both 10 and 20 K for the field processed sample. For the CNT doped sample, the resistivity $\rho_{40 K}$ was reduced by 30% for the field processed sample, and there was a moderate increase in T_c due to the field processing (Table I). The XRD data indicate that there is no change in the lattice parameters upon field processing for both the undoped and CNT doped samples. Thus, the improvement in the resistivity may be attributable to the improved grain connectivity and the connectivity between CNTs and MgB$_2$ grains. The extent of the enhancement in J_c due to the field processing increases with increasing measurement field H. For example, the J_c for a 10 T field processed CNT doped sample increases by a factor of 3 at 10 K and 8 T and at 20 K and 5 T, respectively. It is particularly worth noting that the irreversibility field (H_{irr}) for the field processed sample reached 9 T, which exceeded the record value for the SiC doped MgB$_2$ at 20 K.[5] The J_c for the CNT doped sample shows a small anisotropy when the measurement field H is applied perpendicular ($H \perp H_a$) and parallel to the processing field H_a ($H//H_a$). As the MgB$_2$ crystals are not aligned in the magnetic field, the anisotropy in the CNT doped MgB$_2$ may be attributable to some extent to CNT alignment in the field. Figure 2 shows the upper critical field H_{c2} and irreversibility field H_{irr} versus temperature T for the undoped and CNT doped samples processed with field and without field. The field processing showed no effect on H_{c2} and H_{irr} for the undoped sample but a small improvement for the CNT doped sample within the limited field up to 9 T.

As reported in previous work, the added CNTs were entangled in the MgB$_2$ matrix for the non-field-processed sample and appeared as mainly bare CNTs that were not well bonded to the MgB$_2$ matrix.[8] In contrast, all the CNTs in the magnetic field processed sample are straightened and embedded in the MgB$_2$ matrix as shown in Fig. 3(a). Figure 3(b) shows a focused ion beam (FIB) assisted SEM for the CNT doped MgB$_2$ processed with a 5 T pulsed magnetic field ap-
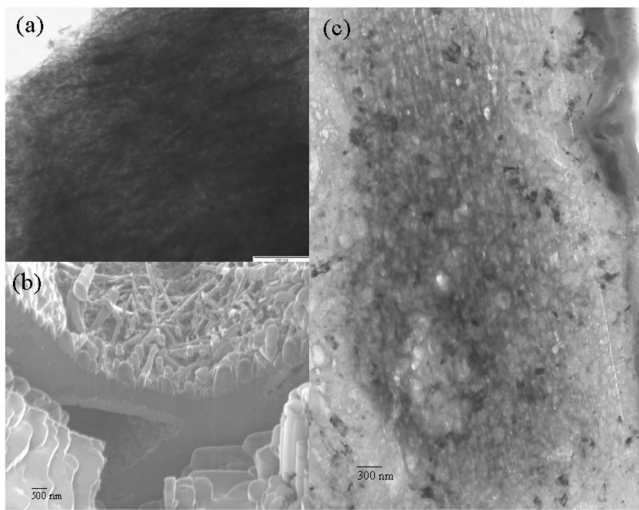
FIG. 3. Transmission electron micrographs for CNT doped MgB_2 processed with a 10 T field (a), FIB-SEM micrograph of straight MgB_2 whiskers formed on the CNTs in 5 T field (b), and TEM image of aligned CNTs embedded in the MgB_2 matrix in 5 T field (c).

plied along the wire axis direction. The magnetic field processing converts the CNT agglomerates to whiskers with lengths of about $2-5$ μm and diameters of $200-400$ nm, which is ten times that for CNTs. These whiskers are formed under magnetic field with CNTs as nucleation centers for the MgB_2 growth in the area of CNT agglomerates. These whiskers are mostly straight with good contact among them. These MgB_2 whiskers would be good conductors, accounting for the enhancement of J_c in the low field region. Figure 3(c) is a transmission electron microscopy (TEM) image showing a number of aligned and straight CNTs embedded in the MgB_2. The aligned CNTs will improve the grain connectivity and act as efficient flux pinning sites. This observation is consistent with the observed strong anisotropy in $J_c(H)$ for field processed CNT doped MgB_2. Such anisotropy is lacking for pure MgB_2. Ajiki and Ando,[20] Lu,[21] and Ramirez *et al.*[22] found that the susceptibility χ of CNTs is large enough for them to be aligned in certain magnetic fields. The melting of Mg could create some liquid environment for the rotating force to align CNTs in field. However, there is substantive solid B particles in the composite. Thus, the extent of CNT movement due to magnetization force will be very limited.

The advantages of the magnetic field processing include the provision of a unique window of opportunity to control the microstructure and the behavior of various additives that have a response to magnetic field. We can extend the conventional materials processing variables of pressure, composition, and temperature, $f(P, C, T)$, to include field (H) and magnetization (M) of the matrix and additives,

$f(P, C, T, H, M)$. We can manipulate various combinations of the parameters such as field strength, processing temperature and time, and the properties of additives to achieve an optimal enhancement in $J_c(H)$. As the formation of MgB_2 in the reaction *in situ* technique is a rapid process, magnetic field processing can be used in large-scale production.

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