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S. K. Chen
University of Cambridge, UK

K. S. Tan
University of Cambridge, UK

B. A. Glowacki
University of Cambridge, UK

W. K. Yeoh
University of Wollongong

Saeid Soltanian
University of Wollongong, saeid@uow.edu.au

See next page for additional authors

Authors
S. K. Chen, K. S. Tan, B. A. Glowacki, W. K. Yeoh, Saeid Soltanian, J. Horvat, and S. X. Dou

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Effect of heating rates on superconducting properties of pure MgB_2, carbon nanotube- and nano-SiC-doped *in situ* MgB_2/Fe wires

S. K. Chen, K. S. Tan,[(a)] and B. A. Glowacki

Department of Materials Science and Metallurgy, University of Cambridge, Pembroke Street, Cambridge CB2 3QZ, United Kingdom and IRC in Superconductivity, University of Cambridge, Madingley Road, Cambridge CB3 0HE, United Kingdom

W. K. Yeoh, S. Soltanian, J. Horvat, and S. X. Dou

Institute for Superconducting and Electronic Materials, University of Wollongong, Northfields Avenue, Wollongong, New South Wales 2522, Australia

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The influence of heating rates and annealing temperatures on the transition temperatures (T_c) and critical current densities (J_c) of pure MgB_2, carbon nanotube- and nano-SiC-doped *in situ* monofilamentary MgB_2/Fe wires was investigated. It was found that higher J_c was obtained for pure MgB_2 samples when heat treated with slower heating rates. SiC-doped samples also have higher J_c with slower heating rates, but the J_c is less sensitive to annealing temperatures. However, the J_c of the carbon nanotube-doped wire was found to be insensitive to heating rates. The variation in T_c and J_c with heating rate, and the different behaviors of differently doped MgB_2/Fe wires, make it essential to carefully select the optimum heating rates for heat treatment. ©2005 *American Institute of Physics*

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With the relatively recent discovery of superconductivity in MgB_2, efforts to produce high-performance, economically viable wires have been revitalized to exploit this low-cost and weak-link-free material with a higher transition temperature, T_c.[1,2] In this regard, the powder-in-tube (PIT) process remains one of the most promising and commonly used techniques for the development of low-cost MgB_2 conductors.[3,4] So far, little attention has been devoted to the effect of heating rates on the properties of MgB_2 wires. Variation in the J_c of MgB_2 is often reported worldwide, which can be attributed in part to different thermomechanical processing, and not just differences between the precursor powders.[5,6,7] Sintering temperatures and durations have a strong effect on critical current density.[8,9] Meanwhile, Serquis *et al.*[10] have shown a strong influence of annealing conditions on microstructural development that correlates to J_c. In Cu-clad MgB_2 wires, the heat treatment is crucial due to the extensive reaction between the superconducting cores and the sheath materials, leading to the formation of Cu_2Mg.[11,12] These wires require a fast formation method which causes less reaction between the Mg and the sheath materials, and markedly improves the critical current density.[11] Because of the lack of substantial reaction between Fe and MgB_2, the importance of heating rates in Fe-sheathed wires is often underestimated.[13] However, for magnet fabrication by a wind and react technique, the heating rate is very important from a practical point of view when considering the large thermal capacity of a coil. The importance of heating rates for the well-established A15 conductors used in magnetic resonance imaging and nuclear magnetic resonance magnet applications has been clearly established, and it has prompted the development of *in situ* optimization techniques.[14]

In this letter, we report a systematic study of the influence of heating rates on the superconducting properties of *in situ* MgB_2/Fe wires. For comparison, carbon nanotube (CNT)- and SiC-doped MgB_2/Fe wires were also studied. Due to the high volatility of Mg during annealing, the melting point of Mg (650 °C) was chosen as one of the two isothermal annealing temperatures to test; the other, 850 °C, was chosen to be significantly higher, as the kinetics of phase formation plays an important role in affecting J_c.

The starting powders were magnesium (99%, -325 mesh), amorphous boron (99%, 1–2 μm), multiwalled CNTs (>94%, outer diameter 20–30 nm, length 0.5–2 μm) and SiC nanoparticle powder (99%, <20 nm). Three types of wire were prepared with powders of nominal composition—MgB_2, $MgB_{1.8}C_{0.2}$, and MgB_2+10 wt % SiC, respectively—using the standard PIT method. The powders were packed into Fe tubes and then drawn to about 1.4 mm in diameter. Short samples (~5 cm each) were cut from the wires and heat treated in a tube furnace which was being constantly pumped down to a vacuum (10^{-6} Torr) during the heat treatments. Two heating rates of 100 °C/h and 900 °C/h were used, followed by isothermal annealing at 650 °C or 850 °C for 30 min, and then cooling in the furnace to room temperature.

X-ray diffractometry (XRD) was conducted using Cu $K\alpha$ radiation in the Bragg–Brentano configuration to determine the phase composition of the samples. Superconducting transition temperature, T_c and critical current density, J_c of the superconducting cores were measured using a dc superconducting quantum interference device magnetometer. T_c was defined as the onset of the diamagnetism, and magnetic J_c was determined using the Bean model. The magnetic field was applied parallel to the wire so that the core material was directly exposed to the field, and the shielding effect from the iron sheath was much reduced compared to the perpendicular field orientation. For T_c measurement, the sample was first zero-field-cooled to 15 K and centered in an applied field of 20 Oe. In order to avoid the ferromagnetic effect from the iron sheath, the sample was then heated to above the T_c of MgB_2 without field, followed by demagnetizing the iron sheath by applying fields of decreasing magnitude and alternating polarity. The sample was then zero-field-cooled to 15 K and the magnetic moment was measured with increasing temperature in a field of 20 Oe. This ensured that the temperature dependence of the magnetic moment reflected the behavior of the superconducting core, although the magnitude of the moment included was influenced by the sheath. Since the saturation field of the iron sheath is lower than 1 T, the magnetization hysteresis loop for J_c measurement above 1 T was not affected. In addition, the reported overcritical state in iron-sheathed round wires is not observed for the longitudinal field orientation.[15,16]

Figure 1 shows the T_c for pure, CNT and SiC doped MgB_2/Fe wires annealed at 650 °C or 850 °C for 30 min with two different heating rates of 100 °C/h and 900 °C/h. The T_c is generally higher for wires heat treated at 850 °C than 650 °C regardless of heating rates except for CNT doped wires, for which the T_c is lower than expected for 850 °C with slow heating (100 °C/h). As T_c is depressed in proportion to how much carbon is substituted in a given sample,[17] this may suggest that slow heating enhances the substitution of boron for carbon.

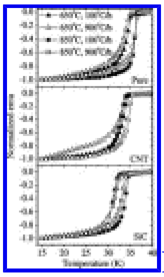
Figure 1.

The effect of heating rates and annealing temperatures on J_c of each type of MgB_2/Fe wires are presented in Fig. 2. It shows that the J_c of pure MgB_2 is strongly influenced by heating rate, in addition to the differences caused by annealing temperature; for either annealing temperature, better J_c is obtained on slow heating. This is because the slower heating rate gave the samples more time at temperatures below the melting point of Mg, allowing more MgB_2 to form through solid-state diffusion and hence minimizing the amount of free Mg on attaining 650 °C. In contrast, the Mg loss becomes more significant when heated at fast rates and to high temperatures, hence causing Mg deficiency in the sample.

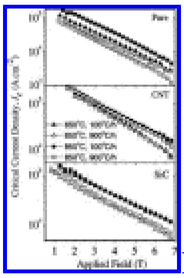
Figure 2.

For CNT-doped samples, the J_c is insensitive to the heating rate. Figure 2 shows that samples heated at 850 °C have similar J_c, and a higher J_c than samples heated at 650 °C regardless of heating rates. A high annealing temperature promotes the C substitution reaction for B, thus enhancing flux pinning in MgB_2.[18] As can be seen from Fig. 2, the high-field J_c is improved in the samples heat treated at a higher temperature. Occupation of an intragranular site by an intact CNT and the unique geometry of the CNT are responsible for effective pinning,[18,19] and the resulting J_c enhancement dominates over any negative effects of magnesium loss during rapid heating.

For SiC doped samples, the J_c for samples heated at 650 °C or 850 °C with a slow ramp rate (100 °C/h) are very similar and are both higher than samples heated with at the faster rate (900 °C/h). XRD data show that the main impurity phases are Mg_2Si and some unreacted SiC. In the previous report, Mg_2Si was found to be the major impurity phase in a SiC-doped sample with an estimated volume fraction of 10%.[20] This suggests that intragrain nanoparticle inclusions consisting of Mg_2Si or unreacted SiC may act as effective pinning centers.[20] Matsumoto *et al.*[21] showed that the formation of Mg_2Si takes place at around 500 °C; therefore, slow heating will enhance the formation of Mg_2Si as the dwell time below the MgB_2 phase formation temperature is much longer compared to fast heating.

Figures 3,4 compare the J_c at 6 and 20 K for all samples annealed at 650 °C and 850 °C, respectively. Overall, SiC-doped samples exhibit the best pinning behavior among the wires with a more gradual drop of J_c with increasing field. Improvement in the pinning properties of CNT-doped samples at a higher annealing temperature results in better J_c over a wider field range than the SiC-doped samples. The plots also show that the J_c of the pure MgB_2 sample can be much enhanced if an appropriate heating rate is chosen.

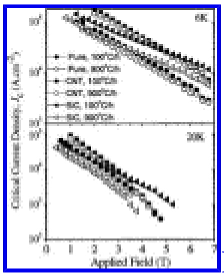
Figure 3.

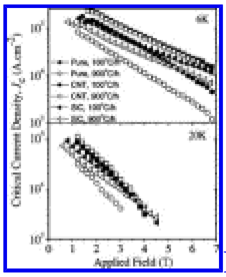
Figure 4.

In summary, pure MgB_2/Fe wires have higher J_c with slower heating rates. The J_c of CNT-doped samples is relatively insensitive to the heating rate, while SiC-doped samples have a higher J_c with slower heating rates and the J_c is less sensitive to the annealing temperature. Therefore, heating rates should be taken into careful consideration during heat treatment of the MgB_2 wires and tapes in order to achieve optimum superconducting properties for a given application. Experiments assisted by *in situ* resistometry during heat treatments, in conjunction with differential thermal analysis, are being carried out to further clarify the influence of heating rates on the kinetics of phase formation of all the wires discussed in this letter.

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FIGURES

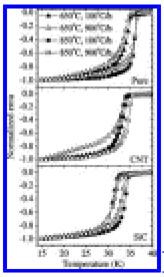
Full figure (19 kB)

Fig. 1. Magnetic moment measurements as a function of temperature for pure, CNT-, and SiC-doped MgB_2/Fe wires annealed at 650 °C or 850 °C for 30 min with two different heating rates of 100 °C/h and 900 °C/h. First citation in article

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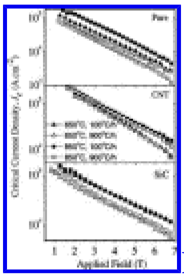
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Fig. 2. Critical current density, J_c as a function of magnetic field at 6 K for pure, CNT-, and SiC-doped MgB_2/Fe wires annealed at 650 °C or 850 °C for 30 min with two different heating rates of 100 °C/h and 900 °C/h. First citation in article

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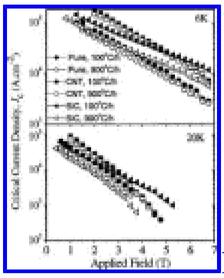
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Fig. 3. Comparison of J_c at 6 and 20 K for pure, CNT-, and SiC-doped MgB_2/Fe wires annealed at 650 °C for 30 min with two different heating rates of 100 °C/h and 900 °C/h. First citation in article

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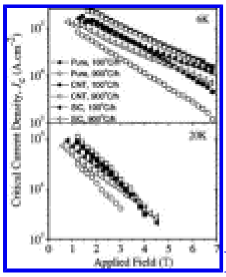
Full figure (14 kB)

Fig. 4. Comparison of J_c at 6 and 20 K for pure, CNT-, and SiC-doped MgB_2/Fe wires annealed at 850 °C for 30 min with two different heating rates of 100 °C/h and 900 °C/h. First citation in article

FOOTNOTES

[a]Electronic mail: kst26@cam.ac.uk

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