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Role of carbon in enhancing the performance of MgB_2 superconductor

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Abstract

The enhancement of the critical current density ($J_c(H)$) of carbon and *nano*-SiC doped MgB_2 is presented and compared. The upper critical field (H_{c2}) being determined from resistivity under magnetic field experiments is though improved for both C substitution and *nano*-SiC addition the same is more pronounced for the former. In $MgB_{2-x}C_x$ carbon is substituted for boron that induces disorder in the boron network and acts as internal pinning centres. The optimal $J_c(H)$ values are obtained for $x = 0.1$ sample. In case of *nano*-SiC doped in MgB_2, the $J_c(H)$ improves more profoundly and two simultaneous mechanisms seems responsible to this enhancement. Highly reactive *nano*-SiC releases free carbon atom, which gets easily incorporated into the MgB_2 lattice to act as *intrinsic* pinning centres. Further enhancement is observed for higher *nano*-SiC concentrations, where the un-reacted components serve as additional *extrinsic* pinning centres.

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1. Introduction

Recently, there have been several reports describing high performance *nano*(*n*)-particle doped MgB_2 superconductor where the *n*-SiC/*n*-diamond/*n*-carbon-tubes/*n*-carbon have yielded high dividends [1–5]. One wonders if the enhanced flux pinning is indeed solely due to these small size (<10 nm) additives or whether carbon plays any additional important role.

Soon after the discovery of superconductivity in MgB_2 at T_c nearly 40 K [6], its various interesting physical properties were found to be intimately linked with: (a) the unexpected transparency of its grain-boundaries [7,8] to passage of electric current, and (b) to the existence of unusual two-band structure [9]. Because of its higher T_c, MgB_2 clearly holds a distinct advantage over most other superconductors in that it can conveniently function at a relatively higher temperature of, say 20 K, easily produced using a conventional close cycle cooling system. Although the T_c of high temperature superconductors (HTS) is much higher, i.e., around 90–130 K, their small coherence length ξ (around 0.3–2.0 nm), large anisotropy and ceramic nature have posed formidable problems for conductor development for practical applications. It is nearly impossible to optimally pin the flux vortices in HTS via foreign additives because of very small ξ. On the other hand, the

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relatively larger ξ of MgB_2 (about 5–10 nm) allows optimum pinning by *n*-particles for enhancement of the $J_c(H)$ performance. Although this effect has been studied for a variety of different *nano*-particle additives, e.g., *n*-Dy_2O_3 [10], *n*-SiO_2 [11], *n*-carbon [2,4], *n*-tubes of carbon [3] and *n*-diamond [5], the best performance yet seen, is with nano-sized carbon derivatives. In fact it might be interesting to intercompare the high field critical current (J_c,H) behaviour resulting from carbon, when substituted for boron ($MgB_{2-x}C_x$) with that when it (or its derivatives) are introduced as additive. We found here, that in the $MgB_{2-x}C_x$ system, though T_c decreases with x, the high field $J_c(H)$, when measured at lower temperatures, is markedly higher for low x values. For $MgB_2 + n$-SiC the T_c decreased slightly but the $J_c(H)$ goes on improving more profoundly till 7 wt% addition. We thus conclude that the enhancement in $J_c(H)$ is a result of two parameters: the existence of pinning centres due to addition of *n*-SiC and, simultaneously, the presence of disorder in boron networks caused by carbon substitution. Our present results demonstrate the importance of disorder originated from carbon substitution.

2. Experimental

Our polycrystalline MgB_2–*n*SiC ($n = 0\%$, 5%, 7% and 10%) and $MgB_{2-x}C_x$ ($x = 0$, 0.04, 0.08 0.10 and 0.20) samples were synthesized by solid-state reaction route with ingredients of Mg, B, *n*-SiC and C powders. The Mg powder used is from *Reidel-de-Haen* of 99% purity, insoluble in HCl and with Fe impurity of less than 0.05%. Amorphous B powder used is from *Fluka* (of 95–97% purity). The *n*-SiC powder is from Aldrich with average particle size (APS) of 5–12 nm. For synthesizing the samples, the stoichiometric amounts were ground thoroughly, palletized, and put in a quartz tube inserted at 850 °C (heating rate 425 °C per hour) under a flow of argon at ambient pressure. This temperature was held for 2.5 h, and subsequently cooled under Ar, to room temperature over a timeframe of 6 h. The X-ray diffraction pattern of the compound was recorded by using CuK_α radiation. *Jeol*-200KV transmission electron microscope was used for bright/dark field images. The magnetization measurements were carried out using *Quantum Design MPMS-XL* magnetometer.

3. Results and discussion

Fig. 1 depicts the X-ray diffraction (XRD) patterns of some of the studied $MgB_{2-x}C_x$ and *n*-SiC added MgB_2 samples and the lattice parameters are listed in Table 1. The pristine MgB_2 compound carries a small amount of un-reacted MgO (marked as #). In $MgB_{2-x}C_x$ the single phase is retained up to $x = 0.2$. When *n*-SiC is added to MgB_2, the phase purity is maintained up 3 wt%. For 5 wt% (assigned as *n*-SiC5) the SiC derivative viz. Mg_2Si lines are clearly seen in the XRD spectrum (marked by *). Lattice parameters are given in Table 1. For $MgB_{2-x}C_x$

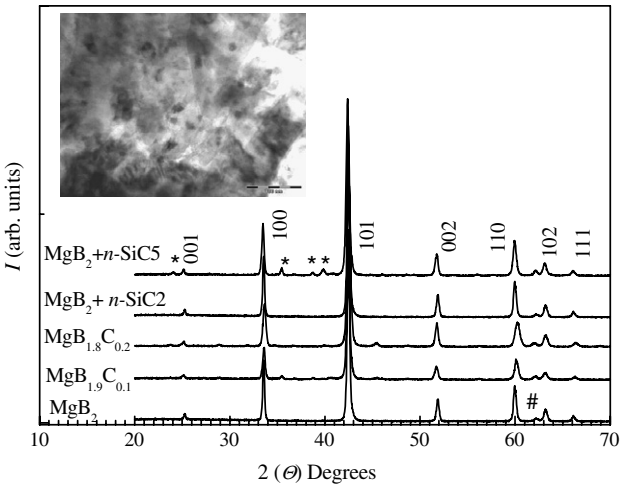

Fig. 1. X-ray diffraction patterns at room temperature of various MgB_2–*n*SiC and $MgB_{2-x}C_x$ samples. The MgO impurity is marked as # and Si derivatives with *. The inset shows the TEM image with scale bar of 100 nm, for $MgB_2 + 7$ wt% *n*-SiC sample, highlighting the presence of *nano*-particles in the matrix.

system, though the *a*-lattice parameter decreases, the *c* increases slightly with increasing x up to $x = 0.2$. The same is true for *n*-SiC added samples except that some unreacted lines are seen above 5-wt% addition. This is consistent with previous reported data on $MgB_{2-x}C_x$ [12]. This indicates clearly that in both cases carbon enters to the matrix. The nano *n*-SiC reacts with Mg (leaving free silicon or Mg_2Si) and release highly reactive free C which can be easily incorporated into the lattice of MgB_2 and substitute into the boron sites [13]. For higher *n*-SiC concentrations, the unbroken *n*-SiC still acts as additional pinning centres. The inset in Fig. 1 shows the presence of *nano*-particles in bright field TEM image with average particle size of around 10 nm.

The zero-field-cooled (ZFC) and field-cooled (FC) magnetic susceptibility curves of the $MgB_{2-x}C_x$ and *n*-SiC added MgB_2 samples are shown in Fig. 2. The T_c values for all the studied samples are given in Table 1. The ZFC branch of pristine MgB_2 exhibits a sharp diamagnetic superconducting transition (T_c) at 38.5 K, whereas the FC signal is very small due to intrinsic pinning. Intrinsic pinning in MgB_2 due to various *nano*-structural defects was highlighted by us very recently [14] and is not discussed here. For $MgB_{2-x}C_x$ system, the T_c values of 34.5 and 32 K for with $x = 0.10$ and 0.20 are obtained, which are in agreement with the previous reports [12]. For *n*-SiC added MgB_2 materials the T_c obtained for 5 wt% is 36 and 35 K for 10 wt% *n*-SiC added sample. The relatively constant T_c in *n*-SiC added MgB_2 samples for 5 wt% up to 10 wt% indicates that at higher concentrations, only a fraction of C presumably substitutes for boron in MgB_2, which would justify the marginal decrease in T_c. The rest of *n*-SiC is available in the matrix and serves as pinning centres.

Fig. 3 depicts the $J_c(H)$ results for *n*-SiC added MgB_2 samples at 10 and 20 K up to relatively high fields of