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S X. Dou

University of Wollongong, shi@uow.edu.au

Saeid Soltanian

University of Wollongong, saeid@uow.edu.au

J. Horvat

University of Wollongong, jhorvat@uow.edu.au

Xiaolin Wang

University of Wollongong, xiaolin@uow.edu.au

S. H. Zhou

University of Wollongong, sihai@uow.edu.au

See next page for additional authors

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Authors

S X. Dou, Saeid Soltanian, J. Horvat, Xiaolin Wang, S. H. Zhou, M. Ionescu, Hua-Kun Liu, P. Munroe, and M. Tomsic

Enhancement of the critical current density and flux pinning of MgB₂ superconductor by nanoparticle SiC doping

S. X. Dou,^{a)} S. Soltanian, J. Horvat, X. L. Wang, S. H. Zhou, M. Ionescu, and H. K. Liu
*Institute for Superconducting and Electronic Materials, University of Wollongong, Northfields Avenue,
 Wollongong, NSW 2522, Australia*

P. Munroe

Electron Microscopy Unit, University of New South Wales, Sydney, NSW 2001, Australia

M. Tomsic

Hyper Tech Research Incorporated, 110 E. Canal Street, Troy, Ohio 45373

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Doping of MgB₂ by nano-SiC and its potential for the improvement of flux pinning were studied for MgB_{2-x}(SiC)_{x/2} with $x=0, 0.2,$ and 0.3 and for 10 wt% nano-SiC-doped MgB₂ samples. Cosubstitution of B by Si and C counterbalanced the effects of single-element doping, decreasing T_c by only 1.5 K, introducing intragrain pinning centers effective at high fields and temperatures, and significantly enhancing J_c and H_{irr} . Compared to the undoped sample, J_c for the 10 wt% doped sample increased by a factor of 32 at 5 K and 8 T, 42 at 20 K and 5 T, and 14 at 30 K and 2 T. At 20 K and 2 T, the J_c for the doped sample was 2.4×10^5 A/cm², which is comparable to J_c values for the best Ag/Bi-2223 tapes. At 20 K and 4 T, J_c was twice as high as for the best MgB₂ thin films and an order of magnitude higher than for the best Fe/MgB₂ tapes. The magnetic J_c is consistent with the transport J_c which remains at 20 000 A/cm² even at 10 T and 5 K for the doped sample, an order of magnitude higher than the undoped one. Because of such high performance, it is anticipated that the future MgB₂ conductors will be made using a formula of MgB_xSi_yC_z instead of pure MgB₂. © 2002 American Institute of Physics. [DOI: 10.1063/1.1517398]

The critical current density (J_c) in MgB₂ has been a central topic of research since superconductivity in this compound was discovered.¹ High J_c values of 10^5 to 10^6 A/cm² have been reported for MgB₂ by several groups. However, J_c drops rapidly with increasing magnetic field due to its poor flux pinning. To take advantage of its high T_c (39 K), improvements in the irreversibility field (H_{irr}) and $J_c(H)$ were achieved by oxygen alloying of MgB₂ thin films² and by proton irradiation of MgB₂ powder.³ However, for practical applications, the pinning centers should be introduced by a simple process, such as chemical doping. Most of element substitution studies were aimed at raising T_c and were thus limited to a low doping level. Improvement of flux pinning was also attempted, using doping by chemical compounds. Here, the results are largely limited to the addition of the compounds, rather than substitution of Mg or B by the compounds. Additives alone appear to be ineffective for the improvement of pinning at high temperatures.^{4–6} Recently, we found that chemical doping of nano-SiC into MgB₂ can significantly enhance J_c in high fields with only slight reductions in T_c up to a doping level of 40% of B.⁷ This finding clearly demonstrated that cosubstitution of SiC for B in MgB₂ induced intragrain defects and a high density of nanoinclusions as effective pinning centers. However, the processing conditions used were far from optimized and the sample density was only 50% of the theoretical value. Therefore, it was not possible to assess the full potential of nano-

SiC doping for the improvement of J_c . In this work, we show that nanometer size SiC-doped MgB₂ gives the highest J_c values in high magnetic fields at 20 K reported for any form of MgB₂, including thin films.

MgB₂ pellet samples were prepared by an *in situ* reaction method, which has been described in detail elsewhere.⁷ Powders of magnesium (99% pure) and amorphous boron (99% pure) were well mixed with SiC nanoparticle powder (size 10 to 100 nm) with the atomic ratio of MgB_{2-x}(SiC)_{x/2}, where $x=0, 0.2,$ and $0.3,$ for samples designated as 1 to 3, respectively. A sample with 10 wt% of SiC addition to MgB₂ was also made as sample 4. Pellets 10 mm in diameter and 2 mm in thickness were made under uniaxial pressure, sealed in Fe tubes and then heated at temperatures of 700–900 °C for 1 h in flowing high-purity Ar. This was followed by furnace cooling to room temperature. The same powders used in samples 1 and 4 were made into wires using the powder-in-tube method.¹⁰ These are designated as samples 5 and 6, respectively.

The magnetization of samples was measured over a temperature range of 5 to 30 K using a physical property measurement system (PPMS) (Quantum Design) in a time-varying magnetic field with sweep rate 50 Oe/s and amplitude up to 8.5 T. All the samples were cut to the same size of $0.56 \times 2.17 \times 3.73$ mm³ from as-sintered pellets. A magnetic J_c was derived from the height of the magnetization loop ΔM using a Bean model: $J_c = 20\Delta M/[a(1 - a/3b)]$. Irreversibility field (H_{irr}) was obtained from measuring the field-cooled and zero-field-cooled magnetic moments as a function of temperature for several values of the

^{a)}Author to whom correspondence should be addressed; electronic mail: shi_dou@uow.edu.au

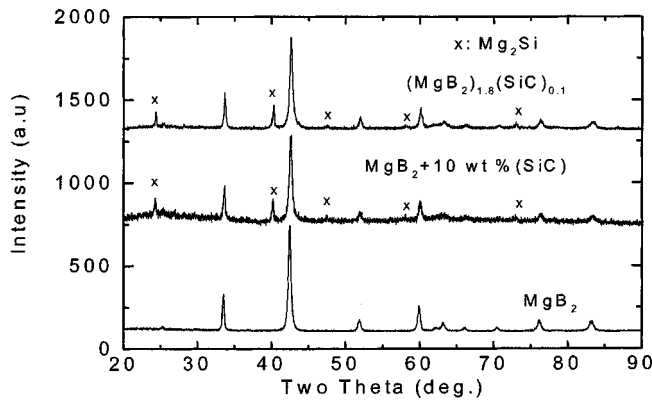


FIG. 1. XRD patterns for the nondoped and SiC-doped samples.

field. The critical temperature (T_c) was obtained as the onset of the diamagnetic transition in the magnetic ac susceptibility measurements. The transport J_c was measured with the four-probe method using a pulsed current source.

Figure 1 shows x-ray diffraction (XRD) patterns for the SiC doped and nondoped samples. The XRD pattern for the nondoped sample (sample 1) reveals about 5% MgO, beside MgB₂ as the main phase. Samples 2 and 3 consist of MgB₂ as the main phase, with Mg₂Si as the major impurity phase (crosses in Fig. 1). The estimated fraction of Mg₂Si was 10%. The energy dispersive spectroscopy (EDS) analysis results showed that the Mg:Si ratio was identical over the entire sample area, indicating a homogeneous phase distribution.

Figure 2 shows magnetic $J_c(H)$ curves for the SiC-doped MgB₂ samples at 5 K, 20 K, and 30 K, for different doping levels. It is noted that all the $J_c(H)$ curves for doped samples show a crossover with the nondoped samples at higher fields. Although SiC doping caused a slight reduction of J_c in low fields, it is much larger than for the nondoped samples in high fields for all the measured temperatures. Compared to the nondoped sample, J_c for the 10 wt % doped sample increased by a factor of 32 at 5 K and 8 T, 23 at 15 K and 6 T, 42 at 20 K and 5 T, and 14 at 30 K and 2 T. This is the best $J_c(H)$ performance ever reported for MgB₂ in any form. It is noted that the $J_c(H)$ curves for the nondoped sample showed a rapid drop in high fields and a plateau near H_{irr} . Earlier, we ascribed this phenomenon to the grains de-

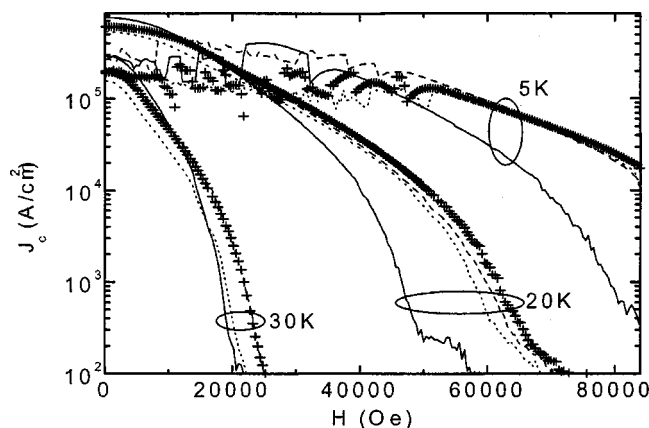


FIG. 2. The magnetic J_c dependence at 5, 20, and 30 K for samples 1, 2, 3, and 4, shown by solid, dashed, and dotted lines, and crosses, respectively.

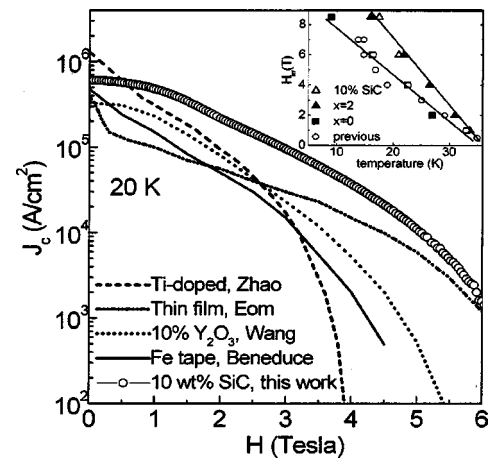


FIG. 3. A comparison of magnetic $J_c(H)$ at 20 K for the 10 wt % SiC-doped sample (sample 4) and for samples that were: Ti doped (see Ref. 4), Y₂O₃ (see Ref. 5) doped, thin film with strong pinning (see Ref. 2) and Fe/MgB₂ tape (see Ref. 9). Inset: temperature dependence of the irreversibility field for SiC-doped MgB₂ with different SiC content (triangles and squares) and for previously prepared nondoped MgB₂ (round symbols).

coupling at higher fields, as a consequence of impurities at the grain boundaries.⁸ In contrast, none of the SiC-doped samples show this phenomenon, as either the substitutions or the induced nano-inclusions are incorporated into the grains.

Figure 3 shows a comparison of magnetic $J_c(H)$ for a 10 wt % SiC-doped sample at 20 K with data reported in literature. J_c for this sample exhibits a better field performance and higher values of J_c in high field than any other element doped samples⁴⁻⁶ or nondoped wires.⁹ Our SiC-doped MgB₂ is even better than the thin-film MgB₂ (Fig. 3), which had exhibited the strongest reported flux pinning and the highest J_c in high fields to date. At 20 K, the best J_c for the 10 wt % SiC-doped sample was 10⁵ A/cm² at 3 T, which exceeds the J_c values of state-of-the-art Ag/Bi-2223 tapes. At 20 K and 4 T, J_c was 36 000 A/cm², which is twice as high as for the best MgB₂ thin films² and an order of magnitude higher than for the state-of-the-art Fe/MgB₂ tapes.⁹

The temperature dependence of H_{irr} for nano-SiC-doped MgB₂, as well as for the pellets and tapes prepared previously (round symbols), is shown in the inset to Fig. 3. Apparently, H_{irr} for $x=0$ overlaps with H_{irr} for the previous samples, even though the latter had significantly smaller values of J_c . Doping with SiC significantly improved H_{irr} . For example, H_{irr} for the SiC-doped samples reached 7.3 T at 20 K, compared to 5.7 T for the nondoped one. This is consistent with improvement of the field dependence of J_c with the doping. Because H_{irr} for the nondoped control sample ($x=0$) is the same as for the previously prepared samples, the improvement of $J_c(H)$ definitely occurred because of the improvement of flux pinning by the doping and not because of improved sintering of MgB₂.

Figure 4 shows the transport $J_c(H)$ values for the undoped and 10 wt % SiC-doped MgB₂ wires (samples 5 and 6) at 5 K, 10 K, and 20 K. It is evident that the transport J_c results for both the undoped and doped wires are in excellent agreement with the magnetic J_c . It is also clear that the enhancement in transport J_c due to SiC doping is consistent with the magnetic J_c . The transport I_c for the 10 wt % SiC-doped MgB₂/Fe reached 660 A at 5 K and 4.5 T (J_c

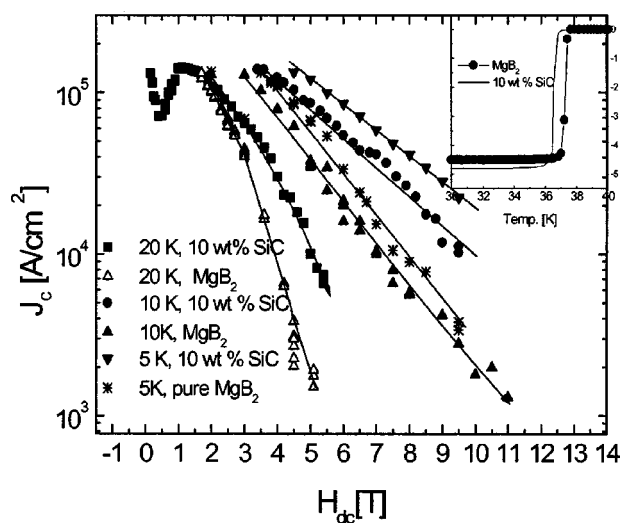


FIG. 4. Transport J_c for SiC doped (sample 5) and undoped MgB_2 (sample 6) wires (the lines only serve for guiding the eyes). Inset shows T_c for these samples.

$=133\,000\text{ A/cm}^2$) and 540 A at 20 K and 2 T ($J_c = 108\,000\text{ A/cm}^2$). The transport J_c for the $10\text{ wt}\%$ SiC-doped MgB_2 wire is more than an order of magnitude higher than for the best Fe-sheathed MgB_2 wire reported to date at 5 K and 10 T and 20 K and 5 T ,^{5,9} respectively.

The inset in Fig. 4 shows T_c for the nondoped and $10\text{ wt}\%$ doped samples. The T_c for the undoped sample is 37.6 K . For the doped samples, T_c decreased with increasing doping level. The transition width was typically 0.5 K . It is striking to note that T_c has only dropped by 1.5 K for the $10\text{ wt}\%$ SiC-doped sample. In contrast, T_c was depressed by almost 7 K for $10\text{ wt}\%$ C substitution for B in MgB_2 .¹⁰ These results suggest that the higher tolerance of T_c to SiC doping in MgB_2 is attributable to the cosubstitution of B by C and Si. This is because the atomic radii of C (0.077 nm) and Si (0.11 nm) atoms are close to that of B (0.097 nm). Codoping with SiC counterbalanced the negative effect on T_c of the single element doping.

Regarding the mechanism behind the enhancement of J_c at higher fields, it is necessary to recognize the special features of SiC doping. First, in contrast to previous work on doping for improving J_c , SiC doping has no densification effect, as evidenced by the fact that the density of doped samples is 1.2 g/cm^3 , independent of the doping level. In addition, SiC doping takes place in the form of substitution and/or addition,⁸ while in the previously reported work,^{4–6} the doping was in the form of additives, not incorporated into crystalline lattice of MgB_2 .

The transmission electron microscopy images showed a high density of dislocations and a large number of $\sim 10\text{ nm}$ inclusions inside the grains (Fig. 5). Their concentration increased with the doping level. EDS analysis of the grains revealed the presence of uniformly distributed Mg, B, C, Si, and O (inset to Fig. 5). This, and the results of XRD, suggests that the inclusion nanoparticles were made of Mg_2Si , or unreacted SiC. All the intragrain defects and the inclusions act as effective pinning centers. Our results suggest that a combination of substitution-induced defects and highly dispersed additives are responsible for the enhanced flux pin-

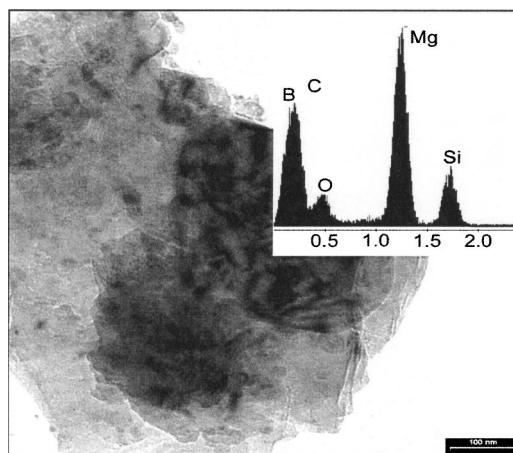


FIG. 5. TEM image showing the intragrain dislocations and nanoparticle inclusions within MgB_2 grains. Inset: EDS element analysis of MgB_2 grains.

ning. When SiC reacts with liquid Mg and amorphous B at the sintering temperatures, the nanoparticles of SiC will act as nucleation sites to form MgB_2 and other nonsuperconducting phases which can be included within the grains as inclusions. Thus, the reaction-induced products are highly dispersed in the bulk matrix.

Given the ease of production of SiC-doped MgB_2 , our results significantly strengthen the position of MgB_2 as a competitor to more expensive conventional and high-temperature superconductors. It is evident that future MgB_2 conductors will be made using a formula of $\text{MgB}_x\text{Si}_y\text{C}_z$ instead of pure MgB_2 . In summary, we have demonstrated that both the transport and magnetic J_c , the irreversibility field and the flux pinning of MgB_2 are all significantly enhanced through nano-SiC doping, significantly improving the potential of MgB_2 for many applications.

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