Evaluation of connectivity, flux pinning, and upper critical field contributions to the critical current density of bulk pure and SiC-alloyed MgB_2

A. Matsumoto,[a)] H. Kumakura, and H. Kitaguchi
National Institute for Materials Science, 1-2-1 Sengen, Tsukuba 305-0047, Japan

B. J. Senkowicz, M. C. Jewell, E. E. Hellstrom, Y. Zhu, P. M. Voyles, and D. C. Larbalestier
Applied Superconductivity Center, University of Wisconsin-Madison, Madison, Wisconsin 53706

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Measurement of critical current density J_c, normal state resistivity ρ_n, and upper critical field H_{c2} on pure and 10% SiC-doped MgB_2 bulks show systematic enhancement of H_{c2} by SiC addition and by lowering reaction temperature. H_{c2}(10 K) exceeds 33 T, while the extrapolated zero temperature value exceeds 40 T. The Rowell [Supercond. Sci. Technol. **16**, R17 (2003)] analysis suggests that only 8%–17% of the MgB_2 cross section actually carries current. Higher reaction temperature enhances the connectivity but degrades H_{c2} and flux pinning, making the measured J_c a complex balance between connectivity, H_{c2}, and flux pinning induced by grain boundaries and precipitates. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2357027]

Magnesium diboride[1] (MgB_2) is a potential competitor to Nb-base superconductors due to its lower raw material cost and good H_{c2} values. However, some confusion often surrounds the meaning of measured H_{c2} values in untextured samples, which are almost always the higher value $H_{c2}^{\parallel}$, where the applied field H is parallel to the *ab* planes. Sometimes the irreversibility field H_{irr} is defined by the bottom of this $H_{c2}^{\parallel}$ transition, making MgB_2 look very good in comparison to an isotropic superconductor such as Nb_3Sn. But the real measure should be the H_{irr} defined by the extrapolation of the critical current density $J_c(H)$ to zero, which is typically only about half of $H_{c2}^{\parallel}$, because H_{irr} is determined by the breadth of the lower, perpendicular H_{c2} transition, $H_{c2}^{\perp}$. At 4.2 K, measured $H_{c2}^{\parallel}$ values[2–7] now are very similar to those of Nb_3Sn, ~25–27 T,[8] but H_{irr} values are only 12–17 T due to suppression of current flow in those grains oriented with Mg and B planes perpendicular to the applied field, which explains why defining H_{irr} by the bottom of the $H_{c2}^{\perp}$ transition is more appropriate for untextured samples than the bottom or 10% point on the small-current-density, resistive H_{c2} transition which corresponds to $H_{c2}^{\parallel}$. However, the resistive H_{c2} transition is still useful for measuring the breadth of the parallel H_{c2} transition ΔH, which may be indicative of inhomogeneity in composition in the sample. Hopes for expanding the useful range of MgB_2 are encouraged by earlier work that has shown that $H_{c2}^{\parallel}(0)$ can exceed 70 T in C-doped MgB_2 thin films,[2] but so far the highest $H_{c2}(0)$ of C- or SiC-doped wires or bulks is ~35 T,[3,4,9,10] only half this value. Since H_{c2} and H_{irr} enhancement is crucial for magnet applications, we have here systematically studied the H_{c2} transition and $J_c(H,T)$ behavior of pure and SiC-doped bulks. Irrespective of this high-field perspective on MgB_2, we should also point out that $J_c(H)$ falls off only slowly in the 10–30 K range, making MgB_2 useful for lower field applications without liquid He.

Our previous reports[6,7] showed that higher J_c values were obtained in tapes using MgH_2 rather than Mg powder. Nano-SiC addition improved the high-field J_c at low temperatures and produced a measured H_{c2} value of 23 T at 4.2 K. Here we present a more detailed study of MgB_2 samples cut from this same tape measured without any extraneous sheath material.

MgB_2 bulk samples were prepared by conventional *in situ* powder-in-tube method with commercial MgH_2 and amorphous B powders which were mixed and packed into a pure Fe tube in air.[7] 5 or 10 mol % of ~30 nm SiC powder[5] was added for the doped samples. The filled tubes were groove rolled into 2 mm square rods and then flat rolled into 0.5 mm thick by 4 mm wide tapes. 50 mm long samples were heat treated at 600, 700, 800, and 900 °C for 1 h under Ar atmosphere making the 12-sample set.[7] After peeling away the Fe sheath, resistivity curves were measured with 5 mA transport currents in a 9 T Quantum Design physical properties measurement system, the 33 T Bitter magnet at the National High Magnetic Field Laboratory (NHMFL) in Tallahassee, and the 60 T short pulse magnet at the NHMFL in Los Alamos National Laboratory. The 10% and 90% points on the resistive transition curves were used to define a transition breadth ΔH and $H_{c2}^{\parallel}$. Magnetization properties were measured in an Oxford Instruments vibrating sample magnetometer, from which the critical current density $J_c(H,T)$ was calculated assuming fully connected samples using the expression $J_c(H,T)=0.5\Delta M12b/(3bd-d^2)$, where b and d are the width and thickness of the rectangular section bar. Extrapolation of $J_c(H)$ to zero allowed extraction of H_{irr}. However, following Rowell,[11] we believe that the connected cross section $1/F$ of our samples is much less than unity, based on calculations of $1/F$ using the relation $\rho(T)=F[\Delta\rho_{sc}(T)+\rho(0)]$, where ρ_n is the measured normal state resistivity and $\Delta\rho_{sc}(300-50\text{ K})=7.3\ \mu\Omega$ cm (Table I).

Table I provides an overview of the properties of the four samples. SiC additions depress T_c, raise H_{c2}, broaden the H_{c2} transition, and raise ρ_n. $1/F$ factors are variable and much less than 100%, implying that the variation in measured ρ_n in Table I is only partly due to scattering. The best connected sample was 900 °C pure at only 17%, while the two 600 °C samples were only 8%–9% connected. Reaction temperature also played a large role: T_c was lower, ρ_n higher,

[a)]Author to whom correspondence should be addressed; electronic mail: matsumoto.akiyoshi@nims.go.jp

TABLE I. Superconducting properties of the MgB_2 samples as a function of reaction temperature and SiC addition.

Sample	T_c^{zero} (K)	ρ_n(40 K) ($\mu\Omega$ cm)	RRR	Active cross section (%)	J_c(5 T,4.2 K) ($\times 10^4$ A/cm^2)	J_c(1 T,25 K) ($\times 10^4$ A/cm^2)	ΔH(10 K) (T)	H_{c2}(10 K) (T)
600 °C pure	36.0	116	1.70	9.0	3.5	4.2	6.3	24.7
900 °C pure	37.5	31	2.40	16.6	12	4.9	3.5	17.2
600 °C 10% SiC	32.0	324	1.27	8.2	3.3	0.6	10.2	31.6
900 °C 10% SiC	35.5	100	1.58	12.7	7.9	5.3	3.8	26.7

$H_{c2}^{\parallel}$ higher, and H_{c2} transition broader for the 600 °C reaction.

Figure 1 shows $J_c(H)$ measured at 4.2 and 25 K but the values are not easy to systematize because of the competing influences of connectivity, H_{c2}, and flux pinning. At $H > 6$ T, the SiC-doped samples have higher J_c(4.2 K) than the pure samples, consistent with their higher H_{c2} (see Fig. 4). At low H where H_{c2} is not important, J_c(4.2 K) for the 600 °C-SiC sample drops below the 900 °C-SiC sample, consistent with its smaller connectivity (8% vs 12%). By contrast, J_c(4.2 K) of the pure samples is higher for the 600 °C reaction than for the 900 °C reaction for all H, a striking contrast with the expectation from the approximate halving of connectivity (9% vs 17%) seen in Table I. However, transmission electron microscopy (TEM) analysis[12] shows that the MgB_2 grain size is significantly smaller after 600 °C rather than after 900 °C reaction (Fig. 2). Consistent with Nb_3Sn where fine grain pinning dominates,[13] the pure MgB_2 samples show a flux-line lattice shear shape of F_p curve. The high J_c data of the very fine grain pure 600 °C reaction are consistent with strong vortex pinning by grain boundaries, which have been shown[14] to be an important pinning center in MgB_2. However, the low connectivities in Table I also suggest that the local, vortex pinning J_c of our samples is up to an order of magnitude higher than indicated in Fig. 1. Finally we note that $J_c(H, 25\text{ K})$ shows significant effects from T_c. For example, the 600 °C-SiC sample has the lowest $J_c(H)$ and the lowest T_c(32 K).

Figure 3 shows the normalized pinning force at 25 K of the four samples. The SiC-doped samples show higher vortex pinning strength at higher magnetic fields than the pure samples. Microstructural analysis[12] shows that the SiC samples contain a significant density of various nanoscale precipitates, consistent with this additional pinning. The pinning force F_p curve shapes were rather independent of temperature.

The full range of $H_{c2}^{\parallel}$ data is not easy to show in one plot due to the breadth and overlap of the data sets, so in Fig. 4 we show the range ΔH between the 10% and 90% points on the H_{c2} transition as a function of measurement temperature only for the 900 °C-pure and 600 °C-SiC samples, which have the narrowest (4.5 T at 2 K) and the broadest (18 T at 2 K) transitions of our sample set. The 600 °C-SiC sample has $H_{c2}(0\text{ K})=42$ T, the highest in our set, considerably higher than in C-doped single crystals[10] or filaments,[9] and exceeded only by some thin films.[4]

It is becoming increasingly clear that most MgB_2 bulk samples have rather inhomogeneous electromagnetic properties. High porosity associated with the ~40% volume contraction of the reaction $MgH_2+B=MgB_2$ provides part of the reason for the low connectivity shown in Table I but our microstructural studies[12] show that various grain boundary phases are present in both pure and SiC-doped samples. Both

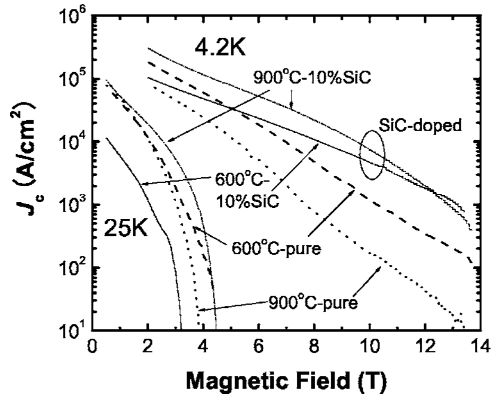

FIG. 1. J_c-H curves at 4.2 and 25 K derived from magnetization measurements.

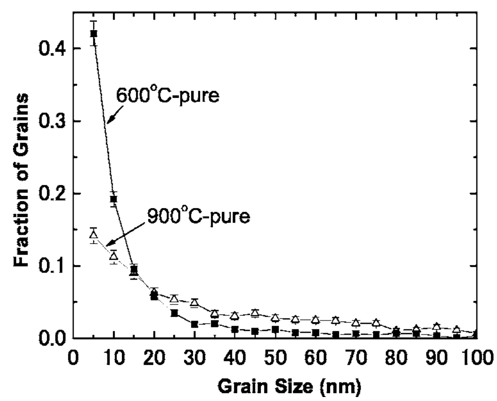

FIG. 2. Grain size distributions for the two pure samples, measured from TEM dark field images. Both samples show a preponderance of grains <50 nm in diameter, but grain growth does occur at 900 °C. Doped samples show the same trend, but analysis is complicated by SiC reaction phases. Approximately 7% of the grains in the 600 °C sample and 9% of the grains in the 900 °C sample are larger than 100 nm and are not included in the figure.

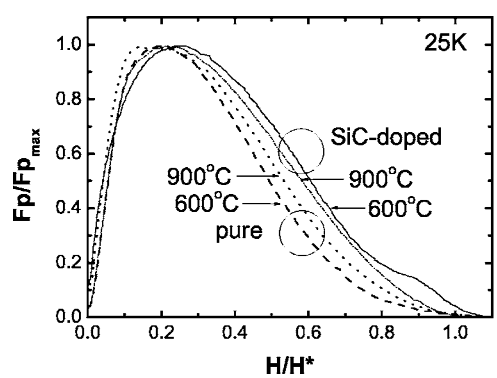

FIG. 3. Normalized pinning force of the four samples at 25 K, where the criterion for the irreversibility field is J_c=100 A/cm^2. The SiC-doped samples show distinct additional pinning in higher reduced fields, a result seen at all temperatures.

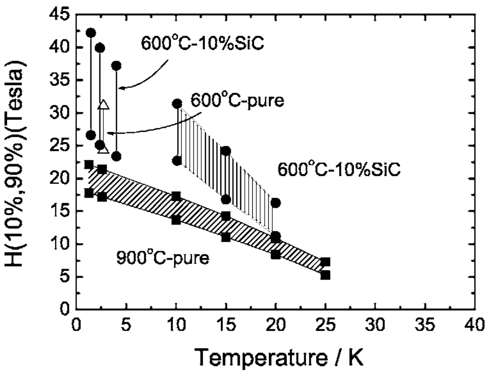

FIG. 4. 90% and 10% of normal state resistivities in the resistive transition curves for two samples of MgB_2. 90% of normal state resistivity is taken as H_{c2}. The much broader transition of $\Delta H(90\%-10\%)$ of the 600 °C-SiC sample suggests that significant inhomogeneity is introduced by alloying and by reaction at low temperatures. This sample also has the largest upper critical field.

factors may play a significant role in producing connectivity factors that lie only between 8% and 17%. Partially offsetting these low connectivities is the observation of very fine grains of <50 nm in all four samples, which appear to produce very strong flux pinning. The most frequently observed grain size in the 600 °C samples is <20 nm, a very favorable size compared to the ~150 nm grain size seen in optimum, high-J_c, high-H_{c2} Nb_3Sn.[8]

In summary our study shows that very high values of $H_{c2}(0)$ exceeding 40 T can be attained in SiC-doped bulk MgB_2 but also that extended transitions with considerable smearing exist in the higher H_{c2} samples. Strong vortex pinning is indicated both by J_c up to 10^4 A/cm^2 at 10 T, 4.2 K and by distortion of the pinning force curve at high reduced fields in SiC-doped samples.

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[1] J. Nagamatsu, N. Nakagawa, T. Muranaka, Y. Zenitani, and J. Akimitsu, Nature (London) **410**, 63 (2001).
[2] C. B. Eom, M. K. Lee, J. H. Choi, L. J. Belenky, X. Song, L. D. Cooley, M. T. Naus, S. Patnaik, J. Jiang, M. Rikel, A. Polyanski, A. Gurevich, X. Y. Cai, S. D. Bu, S. E. Babcock, E. E. Hellstrom, D. C. Larbalestier, N. Rogado, K. A. Regan, M. A. Hayward, T. He, J. S. Slusky, K. Inumaru, M. K. Haas, and R. J. Cava, Nature (London) **411**, 558 (2001).
[3] V. Braccini, L. D. Cooley, S. Patnaik, D. C. Larbalestier, P. Manfrinetti, A. Palenzona, and A. S. Siri, Appl. Phys. Lett. **81**, 4577 (2002).
[4] B. J. Senkowicz, J. E. Gienke, S. Patnaik, C. B. Eom, E. E. Hellstrom, and D. C. Larbalestier, Appl. Phys. Lett. **86** (2005).
[5] S. X. Dou, S. Soltanian, J. Horvat, X. L. Wang, S. H. Zhou, and M. Ionescu, Appl. Phys. Lett. **82**, 4996 (2003).
[6] H. Kumakura, H. Kitaguchi, A. Matsumoto, and H. Hatakeyama, Appl. Phys. Lett. **84**, 3669 (2004).
[7] A. Matsumoto, H. Kumakura, H. Kitaguchi, and H. Hatakeyama, Supercond. Sci. Technol. **16**, 926 (2003).
[8] A. Godeke, M. C. Jewell, C. M. Fischer, A. A. Squitieri, P. J. Lee, and D. C. Larbalestier, J. Appl. Phys. **97**, 093909 (2005).
[9] R. H. T. Wilke, S. L. Bud'ko, P. C. Canfield, D. K. Finnemore, Raymond J. Suplinskas, and S. T. Hannahs, Phys. Rev. Lett. **92**, 217003 (2004).
[10] S. M. Kazakov, R. Puzniak, K. Rogacki, A. V. Mironov, N. D. Zhigadlo, J. Jun, Ch. Soltmann, B. Batlogg, and J. Karpinski, Phys. Rev. B **71**, 024533 (2005).
[11] J. M. Rowell, Supercond. Sci. Technol. **16**, R17 (2003).
[12] Y. Zhu, A. Matsumoto, H. Kumakura, H. Kitaguchi, B. J. Senkowicz, M. C. Jewell, E. E. Hellstrom, D. C. Larbalestier, and P. M. Voyles, J. Appl. Phys. (to be published).
[13] J. Kramer, J. Appl. Phys. **44**, 1360 (1973).
[14] H. Kitaguchi, A. Matsumoto, H. Kumakura, T. Doi, H. Sosiati, and S. Hata, Appl. Phys. Lett. **85**, 2842 (2004).