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# Silicon oil: a cheap liquid additive for enhancing in-field critical current density in MgB<sub>2</sub>

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# Silicon oil: a cheap liquid additive for enhancing in-field critical current density in MgB<sub>2</sub>

## Abstract

In this work, the authors report on significant flux pinning enhancement in MgB<sub>2</sub> that has been easily obtained using a liquid additive, silicon oil. MgB<sub>2</sub> bulk samples with 0 up to 30 wt % silicon oil added were prepared by an in situ reaction. Results showed that the Si and C released from the decomposition of the silicon oil formed Mg<sub>2</sub>Si and substituted into B sites, respectively. Increasing the amount of the Si oil up to 15 wt % has resulted in the reduction of the lattice parameters, as well as T<sub>c</sub> and R(300 K)/R(T<sub>c</sub>) values, accompanied by a significant enhancement of J<sub>c</sub>(H), H<sub>irr</sub>, and H<sub>c2</sub>.

## Keywords

Silicon, oil, cheap, liquid, additive, for, enhancing, field, critical, current, density, MgB<sub>2</sub>

## Disciplines

Engineering | Physical Sciences and Mathematics

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## Silicon oil: A cheap liquid additive for enhancing in-field critical current density in MgB<sub>2</sub>

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In this work, the authors report on significant flux pinning enhancement in MgB<sub>2</sub> that has been easily obtained using a liquid additive, silicon oil. MgB<sub>2</sub> bulk samples with 0 up to 30 wt % silicon oil added were prepared by an *in situ* reaction. Results showed that the Si and C released from the decomposition of the silicon oil formed Mg<sub>2</sub>Si and substituted into B sites, respectively. Increasing the amount of the Si oil up to 15 wt % has resulted in the reduction of the lattice parameters, as well as  $T_c$  and  $R(300\text{ K})/R(T_c)$  values, accompanied by a significant enhancement of  $J_c(H)$ ,  $H_{irr}$ , and  $H_{c2}$ . © 2007 American Institute of Physics. [DOI: 10.1063/1.2435321]

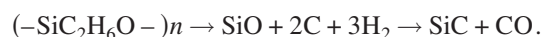
For practical applications that require carrying large supercurrents in the presence of magnetic fields, improvements in the critical current density ( $J_c$ ), the upper critical field ( $H_{c2}$ ), and the irreversibility field ( $H_{irr}$ ) have been the key topics of research on MgB<sub>2</sub> superconductors. An effective way to improve the flux pinning is to introduce flux pinning centers into MgB<sub>2</sub>. It has been found that chemical doping with nonmagnetic materials appears to be the most suitable approach to increase the ability of MgB<sub>2</sub> to carry large currents for practical applications. Numbers of additives have been examined for  $J_c$ ,  $H_{c2}$ , and  $H_{irr}$  improvements. It has already been shown that a  $J_c$  enhancement by more than one order of magnitude in high magnetic fields can be easily achieved with only slight reduction in  $T_c$  through adding MgB<sub>2</sub> with nanoparticles, such as SiC, Si, and C.<sup>1–8</sup> It has been shown that SiC addition significantly enhances the  $H_{c2}$  and  $H_{irr}$  in polycrystalline bulks, as well as in wires and tapes.<sup>1,2,9</sup> It should be pointed out that the improvement of flux pinning enhancement is controlled by the sizes of the particles doped into MgB<sub>2</sub>. However, the requirement for finer nanoparticles brings some dilemmas such as higher cost and some technical problems in fabricating the much finer nanoparticles. Because the nanoparticles are in solid state form, another problem is agglomeration of nanoparticles, which will certainly limit the homogeneity of mixing with MgB<sub>2</sub>. This homogeneity of mixing is very crucial in determining the flux pinning ability for MgB<sub>2</sub> made by the *in situ* reaction method. Recently, it has been reported that aromatic hydrocarbon addition to MgB<sub>2</sub> can enhance the flux pinning in MgB<sub>2</sub> at low sintering temperatures.<sup>10</sup> However, the enhancement is not greater than in nano-SiC doped samples, and this organic solvent is very volatile at ambient pressure. In addition, solid state malic acid addition into MgB<sub>2</sub><sup>11</sup> has also been reported to enhance the flux pinning in MgB<sub>2</sub>. However, the sintering temperature used was as high as 900 °C. To solve the problem of nanoparticle agglomeration, the best way is to use liquid precursors that contain Si and C and are able to introduce both Si and C into MgB<sub>2</sub> at atomic scale, even when sintering is short and at low temperature. In this letter, we report that a significant flux pinning enhance-

ment in MgB<sub>2</sub> can be easily achieved using a liquid precursor, silicon oil, which can produce Si and C at atomic level when reacting with MgB<sub>2</sub>.

Commercial silicon oil,  $(-\text{SiC}_2\text{H}_6\text{O}-)_n$ , is composed of the elements C, H, O, and Si, and it is a colorless, odorless, chemically inert lubricant, with excellent thermal stability. The starting materials we used in this work are amorphous boron powder with a purity of 99.9%, Mg powder with a purity of 99%, and commercial high temperature silicon oil from Sigma Aldrich. B and Mg powders at chemical stoichiometry were thoroughly mixed with diluted silicon oil in acetone. The amounts of silicon oil added into the MgB<sub>2</sub> samples were 3, 10, 15, 20, and 30 wt %, respectively. Pellets of 13 mm in diameter and 2 mm in thickness were made under uniaxial pressure. These pellets were then sealed in an iron tube and sintered in a tube furnace at 750–780 °C for 10 min only, as we have found that short sintering is as good as long sintering in terms of flux pinning for MgB<sub>2</sub>.<sup>12</sup> A high purity argon gas flow was maintained throughout the *in situ* sintering process to avoid oxidation. An undoped MgB<sub>2</sub> sample was also prepared under the same *in situ* processing conditions as a reference sample.

From x-ray diffraction (XRD) experiments, we observed that all the samples crystallized in the MgB<sub>2</sub> structure as the major phase. Slight amounts of MgO and Mg<sub>2</sub>Si are also present in silicon oil doped samples. The amount of Mg<sub>2</sub>Si is increased by increasing the silicon oil content. However, the tiny amount of MgO phase remains the same for the undoped and all the doped samples as determined by XRD.

The decomposition of pure commercial silicon oil possibly follows the following reaction at 800 °C.<sup>13</sup>



It is obvious that the above decomposition of silicon oil took place below 800 °C in our samples, as all the samples were sintered at 780 °C. Si and C released as a result of the decomposition of silicon oil may not form SiC, as no detectable SiC phase was observed from XRD. It is believed that the chemically active Mg reacted with Si and that this caused the decomposition of silicon oil at relatively low temperatures. The remaining C would then embed itself into the MgB<sub>2</sub> grains together with Mg<sub>2</sub>Si and also substitute into B sites in

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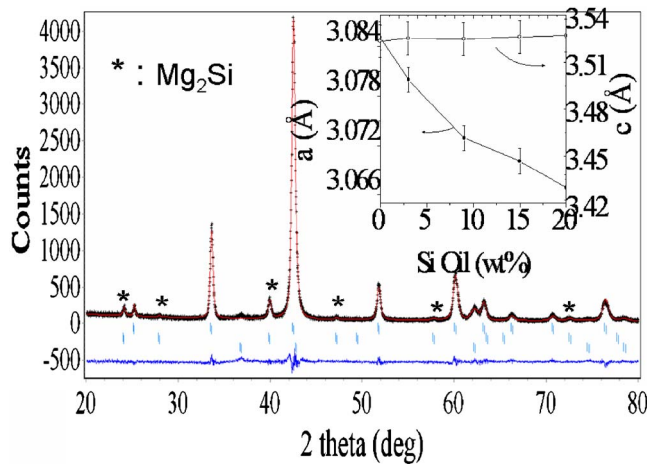


FIG. 1. Observed (crosses) and calculated (solid line) diffraction profiles and their difference (bottom solid line) at 300 K for  $\text{MgB}_2$  with 10 wt % silicon oil added. Top, middle, and lower peak markers relate to  $\text{MgB}_2$ ,  $\text{Mg}_2\text{Si}$  (also marked as \*), and  $\text{MgO}$ , respectively. Inset is the lattice parameters vs silicon oil content.

the  $\text{MgB}_2$  crystal lattice, as has been observed in nano-SiC, Si, and C doped  $\text{MgB}_2$ .

The calculated XRD patterns using Rietveld refinement fit very well with the observed ones. The refined and observed XRD patterns for the 10 wt % silicon oil added sample are shown in Fig. 1. The lattice parameters obtained by the refinement revealed that the  $a$  lattice parameter is reduced from 3.085 to 3.065 Å for the pure and 15 wt % silicon oil doped samples, respectively; while the  $c$  lattice parameter is only slightly increased as illustrated in the inset.

The significant reduction in the  $a$  lattice parameter indicates that carbon has been doped into the B sites in the crystal lattice and caused the reduction in  $T_c$ . Both C doping and the inclusion of  $\text{Mg}_2\text{Si}$  can enhance the electron scattering, as proved by the decreased residual resistivity ratio (RRR) values, and, in turn, enhance the flux pinning.

Figure 2 shows the resistance versus temperature curves ( $R$ - $T$ ) for three samples at zero external magnetic field over a temperature range of 30–300 K. It can be seen that the scattering increases with increasing silicon oil content. The resistivity at 40 K increased from 24  $\mu\Omega$  cm for the pure  $\text{MgB}_2$  to 64  $\mu\Omega$  cm for the 10 wt % silicon oil doped  $\text{MgB}_2$ .

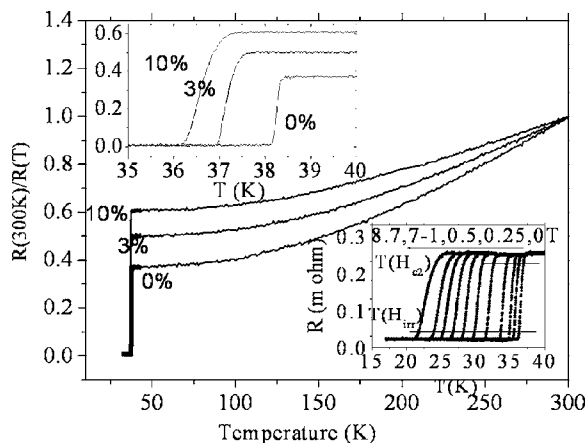


FIG. 2. Temperature dependence of the normalized resistance ( $R$ - $T$ ) for pure and silicon oil doped samples. The inset (right) shows  $R$ - $T$  in the fields up to 8.7 T for the sample doped with 10 wt % silicon oil.

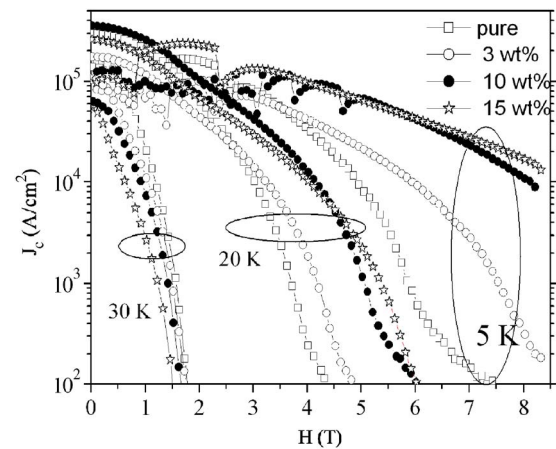


FIG. 3. Field dependence of  $J_c$  at 5, 20, and 30 K.

The  $T_c$  values and residual resistivity ratios,  $R(300\text{ K})/R(T_c)$ , were obtained to be 38.2, 37, and 36.2 K and 2.72, 2.0, and 1.67, for the 0%, 3%, and 10% silicon oil samples, respectively.

The magnetic field dependence of  $J_c$  at 30, 20, and 5 K is shown in Fig. 3. It should be noted that the  $J_c$  values in high fields are significantly enhanced for all the doped samples. The  $J_c$  of the un-doped sample dropped to 100  $\text{A}/\text{cm}^2$  at 7 T and 5 K. However, the  $J_c$  values at the same field are increased by more than one or two orders of magnitude for the 3, 10, and 25 wt % silicon oil added samples. At 8 T and 5 K, the  $J_c$  values of the 10 and 15 wt % doped samples are over  $(1-2) \times 10^4$   $\text{A}/\text{cm}^2$ , more than one order of magnitude higher than for the 3 wt % doped one. It should also be noted that there was no degradation in self-field  $J_c$  values for the 10 and 15 wt % silicon oil doped samples.

The  $H_{c2}$  and  $H_{irr}$  were also enhanced, as proved by the data determined from the  $R$ - $T$  curves, which are shown in the inset of Fig. 2. The inset shows the resistance versus temperature ( $R$ - $T$ ) measured at different applied magnetic fields up to 8.7 T for the 10 wt % doped sample. The  $H_{c2}$  values versus normalized temperature  $T/T_c$  obtained from the 90% or 10% values of their corresponding resistive transitions are shown in Fig. 4. The  $H_{c2}$  values of the undoped sample are also included for comparison. Significantly enhanced  $H_{irr}$  and  $H_{c2}$  for the silicon oil doped sample are clearly observed. As we can see, the  $H_{c2}$  curves of all the samples show a positive curvature near  $T_c$ <sup>14</sup> as a result of the two band superconductivity in  $\text{MgB}_2$ . Also, all the doped samples have larger  $dH_{c2}/d(T/T_c)$  values compared to the undoped sample. The evolution of the enhancement of flux pinning is shown clearly in the variation of the ratio  $r_{irr} = H_{irr}(\text{doped})/H_{irr}(\text{undoped})$  or  $r(H_{c2}) = H_{c2}(\text{doped})/H_{c2}(\text{undoped})$  with  $T/T_c$ . Both ratios are about 1.25 and 1.5 for the 3 wt % and the 10 wt % silicon oil doped  $\text{MgB}_2$ , respectively. The above results reveal that  $\text{MgB}_2$  with silicon oil added exhibits higher  $H_{irr}$  values compared to the undoped samples that were processed under the same fabrication conditions. The field dependence of the normalized volume pinning force  $F_p = J \times B$  at 20 K for all the samples is shown in Fig. 5(a). It can be seen that the pinning force for the silicon oil added samples is significantly higher than for the undoped sample at  $B > 1.5$  T.

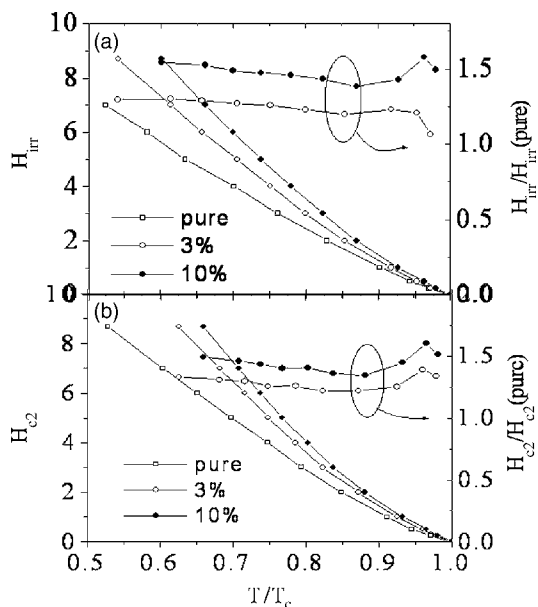


FIG. 4. Normalized  $T/T_c$  vs  $H_{irr}$  (a) and  $H_{c2}$  (b) for pure and silicon oil doped  $MgB_2$ .

The XRD diffraction peaks are observed to become broadened with an increasing amount of silicon oil. Figure 5(b) shows the full width at half maximum (FWHM) for the (100), (002), and (110) peaks for all the samples. It can be seen that the values of the FWHM of the (100) peak increase monotonically for all samples with an amount of Si oil up to 15 wt %. The FWHM values also increase for the (002) and (110) peaks for the 3 and 10 wt % silicon oil samples. The peak broadening in our samples likely arise from nonuniform strain that is mainly caused by C doping on B sites.<sup>15</sup> The grain sizes, which could also affect the peak width, have been observed to be very similar under scanning electron microscopy. However, a further study on the grain sizes and crystal defects using high resolution transmission electron microscopy is needed. The presence of  $Mg_2Si$  impurity phase is also responsible for the peak broadening, as the  $Mg_2Si$  is believed to act as a grain refiner in  $MgB_2$ .<sup>9</sup> Therefore, the enhanced flux pinning,  $H_{c2}$ ,  $H_{irr}$ , and  $J_c(H)$  observed in our

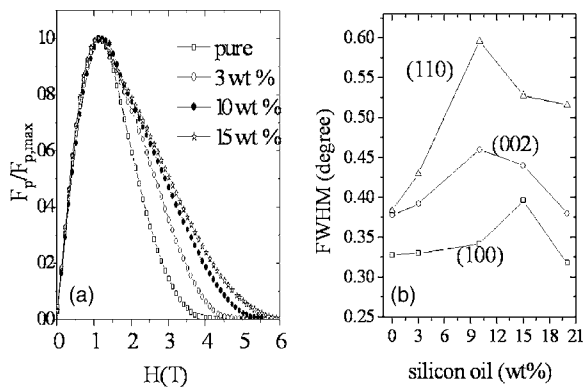


FIG. 5. (a) Field dependence of the volume pinning force of all samples at 20 K. The  $F_p$  is normalized by the maximum volume pinning force,  $F_{p,max}$ . (b) FWHM of various diffraction peaks as a function of the amount of silicon oil.

silicon oil added  $MgB_2$  are due to the C-doping effect and inclusions of  $Mg_2Si$ . It is believed that the large distortion of the crystal lattice caused by both carbon substitution for B and inclusion of  $Mg_2Si$  leads to enhanced electron scattering and enhancement of  $H_{c2}$ .

The data on SiC nanopowder added  $MgB_2$  prepared using hot pressing method presented in our previous work<sup>1,3</sup> are better than what we have achieved in this work using Si oil. However, the Si oil is easier and cheaper to enhance the flux pinning compared to SiC nanopowders. Further improvement of the flux pinning performance of  $MgB_2$  using the Si oil is highly possible by optimizing the processing conditions.

In summary, we have found that a significant flux pinning enhancement in  $MgB_2$  can be easily achieved using a liquid additive, silicon oil. Our results showed that Si and C released from the decomposition of the silicon oil formed  $Mg_2Si$  and substituted into B sites, respectively. Increasing the amount of the Si oil up to 15 wt % leads to the reduction of the lattice parameters, as well as  $T_c$  and  $R(300\text{ K})/R(T_c)$  values, resulting in a significant enhancement of  $J_c(H)$ ,  $H_{irr}$ , and  $H_{c2}$ .

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- <sup>1</sup>S. X. Dou, S. Soltanian, J. Horvat, X. L. Wang, S. H. Zhou, M. Ionescu, H. K. Liu, P. Munroe, and M. Tomsic, *Appl. Phys. Lett.* **81**, 3419 (2002); H. Kumakura, H. Kitaguchi, A. Matsumoto, and H. Hatakeyama, *ibid.* **84**, 369 (2004).
- <sup>2</sup>X. L. Wang, S. H. Zhou, M. J. Qin, P. R. Munroe, S. Soltanian, H. K. Liu, and S. X. Dou, *Physica C* **385**, 461 (2003).
- <sup>3</sup>X. L. Wang, S. Soltanian, M. James, M. J. Qin, J. Horvat, Q. W. Yao, H. K. Liu, and S. X. Dou, *Physica C* **408**, 63 (2004).
- <sup>4</sup>S. Soltanian, J. Horvat, X. L. Wang, P. Munroe, and S. X. Dou, *Physica C* **390**, 185 (2003).
- <sup>5</sup>R. H. T. Wilke, S. L. Bud'ko, P. C. Canfield, D. K. Finnemore, R. J. Suplinskas, and S. T. Hannahs, *Phys. Rev. Lett.* **92**, 217003 (2004).
- <sup>6</sup>B. J. Senkowitz, J. E. Giencke, S. Patnaik, C. B. Eom, E. E. Hellstrom, and D. C. Larbalestier, *Appl. Phys. Lett.* **86**, 202502 (2005).
- <sup>7</sup>V. Braccini, A. Gurevich, J. E. Giencke, M. C. Jewell, C. B. Eom, D. C. Larbalestier, A. Pogrebnikov, Y. Cui, B. T. Liu, Y. F. Hu, J. M. Redwing, Qi Li, X. X. Xi, R. K. Singh, R. Gandikota, J. Kim, B. Wilkens, N. Newman, J. Rowell, B. Moeckly, V. Ferrando, C. Tarantini, D. Marré, M. Putti, C. Ferdeghini, R. Vaglio, and E. Haanappel, *Phys. Rev. B* **71**, 012504 (2004).
- <sup>8</sup>S. K. Chen, M. Wei, and J. L. MacManus-Driscoll, *Appl. Phys. Lett.* **88**, 192512 (2006).
- <sup>9</sup>S. Soltanian, X. L. Wang, J. Horvat, S. X. Dou, M. D. Sumption, M. Bhatia, E. W. Collings, P. Munroe, and M. Tomsic, *Supercond. Sci. Technol.* **18**, 658 (2005).
- <sup>10</sup>H. Yamada, M. Hirakawa, H. Kumakura, and H. Kitaguchi, *Supercond. Sci. Technol.* **19**, 175 (2006).
- <sup>11</sup>J. H. Kim, S. Zhou, M. S. A. Hossain, A. V. Pan, and S. X. Dou, *Appl. Phys. Lett.* **89**, 142505 (2006).
- <sup>12</sup>X. L. Wang, S. Soltanian, J. Horvat, A. H. Liu, M. J. Qin, H. K. Liu, and S. X. Dou, *Physica C* **361**, 149 (2001).
- <sup>13</sup>V. G. Pol, S. V. Pol, A. Gedanken, S. H. Lim, Z. Zhong, and J. Lin, *J. Phys. Chem. B* **110**, 11237 (2006).
- <sup>14</sup>A. Gurevich, *Phys. Rev. B* **67**, 184515 (2003).
- <sup>15</sup>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).