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Silicon oil: a cheap liquid additive for enhancing in-field critical current density in MgB2

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Abstract

In this work, the authors report on significant flux pinning enhancement in MgB_2 that has been easily obtained using a liquid additive, silicon oil. MgB_2 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_2Si 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 $R(300\ K)/R(T_c)$ values, accompanied by a significant enhancement of $J_c(H)$, H_{irr}, and H_{c2}.

Keywords

Silicon, oil, cheap, liquid, additive, for, enhancing, field, critical, current, density, MgB2

Disciplines

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Silicon oil: A cheap liquid additive for enhancing in-field critical current density in MgB_2

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In this work, the authors report on significant flux pinning enhancement in MgB_2 that has been easily obtained using a liquid additive, silicon oil. MgB_2 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_2Si 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 $R(300\ \mathrm{K})/R(T_c)$ 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_2 superconductors. An effective way to improve the flux pinning is to introduce flux pinning centers into MgB_2. It has been found that chemical doping with nonmagnetic materials appears to be the most suitable approach to increase the ability of MgB_2 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_2 with nanoparticles, such as SiC, Si, and C.[1–8] 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.[1,2,9] It should be pointed out that the improvement of flux pinning enhancement is controlled by the sizes of the particles doped into MgB_2. 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_2. This homogeneity of mixing is very crucial in determining the flux pinning ability for MgB_2 made by the *in situ* reaction method. Recently, it has been reported that aromatic hydrocarbon addition to MgB_2 can enhance the flux pinning in MgB_2 at low sintering temperatures.[10] 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_2[11] has also been reported to enhance the flux pinning in MgB_2. 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_2 at atomic scale, even when sintering is short and at low temperature. In this letter, we report that a significant flux pinning enhancement in MgB_2 can be easily achieved using a liquid precursor, silicon oil, which can produce Si and C at atomic level when reacting with MgB_2.

Commercial silicon oil, $(-SiC_2H_6O-)n$, 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_2 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_2.[12] A high purity argon gas flow was maintained throughout the *in situ* sintering process to avoid oxidation. An undoped MgB_2 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_2 structure as the major phase. Slight amounts of MgO and Mg_2Si are also present in silicon oil doped samples. The amount of Mg_2Si 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:[13]

$$(-SiC_2H_6O-)n \rightarrow SiO + 2C + 3H_2 \rightarrow SiC + CO.$$

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_2 grains together with Mg_2Si and also substitute into B sites in

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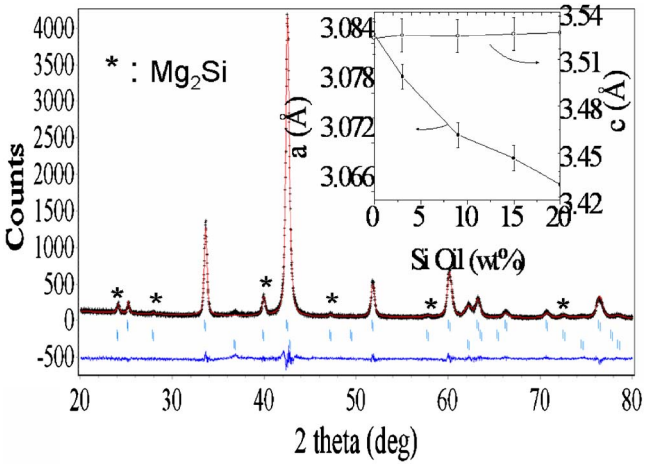

FIG. 1. Observed (crosses) and calculated (solid line) diffraction profiles and their difference (bottom solid line) at 300 K for MgB_2 with 10 wt % silicon oil added. Top, middle, and lower peak markers relate to MgB_2, Mg_2Si (also marked as *), and MgO, respectively. Inset is the lattice parameters vs silicon oil content.

the MgB_2 crystal lattice, as has been observed in nano-SiC, Si, and C doped 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 Mg_2Si 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 MgB_2 to 64 $\mu\Omega$ cm for the 10 wt % silicon oil doped MgB_2.

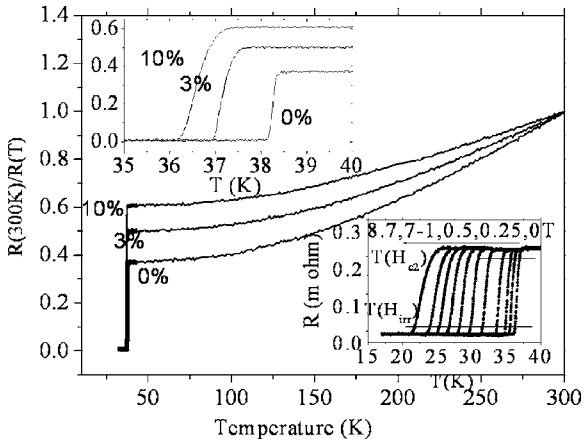

FIG. 2. Temperature dependence of the normalized resistance (R-T) for pure and silicon oil doped samples. The inset (right) shows R-T in the fields up to 8.7 T for the sample doped with 10 wt % silicon oil.

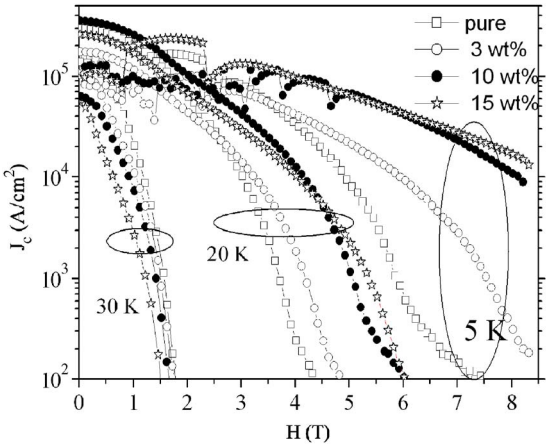

FIG. 3. Field dependence of J_c at 5, 20, and 30 K.

The T_c values and residual resistivity ratios, $R(300\ \mathrm{K})/R(T_c)$, 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 A/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 $(1-2)\times10^4$ A/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[14] as a result of the two band superconductivity in 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 $r_{\mathrm{irr}}=H_{\mathrm{irr}}(\mathrm{doped})/H_{\mathrm{irr}}(\mathrm{undoped})$ or $r(H_{c2})=H_{c2}(\mathrm{doped})/H_{c2}(\mathrm{undoped})$ with T/T_c. Both ratios are about 1.25 and 1.5 for the 3 wt % and the 10 wt % silicon oil doped MgB_2, respectively. The above results reveal that 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.

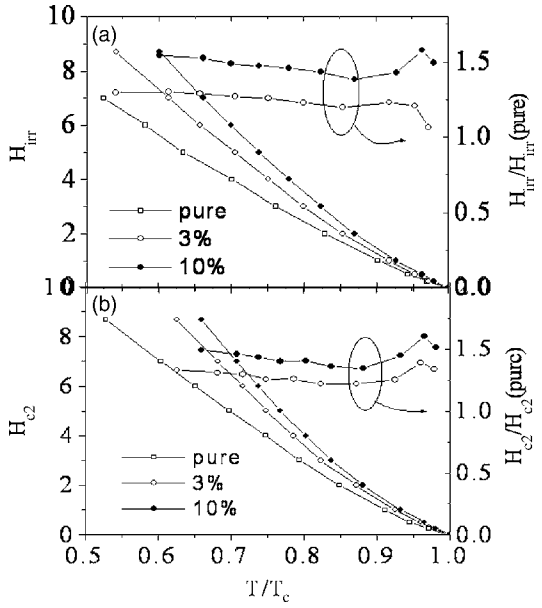

FIG. 4. Normalized T/T_c vs H_{irr} (a) and H_{c2} (b) for pure and silicon oil doped MgB_2.

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.[15] 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.[9] Therefore, the enhanced flux pinning, H_{c2}, H_{irr}, and $J_c(H)$ observed in our 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}.

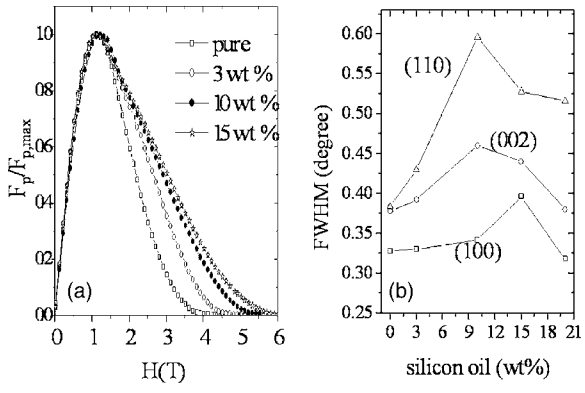

FIG. 5. (a) Field dependence of the volume pinning force of all samples at 20 K. The F_p is normalized by the maximum volume pinning force, $F_{p,max}$. (b) FWHM of various diffraction peaks as a function of the amount of silicon oil.

The data on SiC nanopowder added MgB_2 prepared using hot pressing method presented in our previous work[1,3] 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 $R(300\ \mathrm{K})/R(T_c)$ values, resulting in a significant enhancement of $J_c(H)$, H_{irr}, and H_{c2}.

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