2008

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Recommended Citation

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Influence of disorder on the in-field J_c of MgB$_2$ wires using highly active pyrene

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In this work, we report on significantly enhanced critical current density (J_c) in MgB$_2$ superconductor that was easily obtained by doping with a hydrocarbon, highly active pyrene (C$_{16}$H$_{10}$), and using a sintering temperature as low as $\sim$600 °C. The processing advantages of the C$_{16}$H$_{10}$ additive include production of a highly active carbon (C) source, an increased level of disorder, and the introduction of small grain size, resulting in enhancement of J_c. © 2008 American Institute of Physics. [DOI: 10.1063/1.2838756]

The discovery of superconductivity in MgB$_2$ has aroused a great deal of interest in the relevant scientific and technological communities. Specifically, its simple binary composition, lack of weak intergranular links, critical temperature (T_c) of 39 K, and inexpensive starting materials make MgB$_2$ a promising candidate for applications at operating temperatures around 20 K. However, pristine MgB$_2$ exhibits weak flux pinning, which results in a strong field dependence of the critical current density (J_c) and a low irreversibility field (B_{irr}), compared to commercial low temperature superconductors such as the Nb-based ones.[1] It is, thus, necessary to further improve the J_c and B_{irr} in MgB$_2$ for magnet applications.

Several techniques, such as mechanical alloying,[2,3] proton irradiation,[4] mechanical processing,[5] and using nanosized starting materials[6] have been employed in order to improve flux pinning, J_c, and B_{irr}, but with limited success. Significant results were obtained by adding carbon (C) compounds to MgB$_2$, resulting in enhancement of both J_c and B_{irr}. In particular, SiC nanoparticles significantly enhanced the J_c under magnetic fields around 8 T.[7] However, the addition of SiC to MgB$_2$ has been limited by local agglomerations (with a size of $\sim$30 nm) of unreacted SiC or C, so it is difficult to achieve uniform dispersion within the matrix. In our previous work, we reported on the C source possibilities of carbohydrate (malic acid, C$_4$H$_6$O$_5$) doping through a chemical solution route to achieve homogeneous mixing.[8] It was estimated that intragranular J_c reached 2.5×10^4 A cm^{-2} at 5 K and 8 T for MgB$_2$+10 wt % C$_4$H$_6$O$_5$ bulk when sintered at 900 °C, with a T_c reduction of only 1.5 K. B_{irr} at 20 K was about 7 T with a J_c criterion of 100 A cm^{-2}. The advantages of the chemical solution route using various carbohydrates have now been confirmed by other groups.[9–11] However, our understanding of the cause of the significantly enhanced J_c and B_{irr} has still remained unclear due to the complicated reactions involved. In addition, although we conducted evaporation processing to eliminate the oxygen (O) content, the fraction of MgO due to included O from the carbohydrate

increased gradually with increasing doping level.[12] If we can reduce the MgO fraction within the matrix and find a highly active C source that will be effective at lower sintering temperatures 600 °C, the same as for MgB$_2$ formation, we speculate that the resulting high disorder due to C substitution, as well as strong flux pinning due to the small crystalline size, can have simultaneous positive effects on the in-field J_c. A higher sintering temperature is usually necessary for most of the other C sources, leading to crystalline growth and worse pinning.[13] The use of low temperature processing is also crucial as it allows selection of various sensitive sheath materials and the reduction of production costs.

Although Yamada *et al.*[14] prepared Fe sheathed tape samples by using a liquid aromatic hydrocarbon together with ball milling, it is very difficult to control the composition during ball milling because of its volatile properties. In this study, therefore, we suggested a different hydrocarbon, pyrene (C$_{16}$H$_{10}$), which has no oxygen content, as the C source. This is a colorless solid and consists of four fused benzene rings, resulting in a large, flat aromatic system. At around a temperature of 145–148 °C, it decomposes into benzene (C$_6$H$_6$), other hydrocarbons, and C. The released fresh C enables the improvement reactivity. In addition, it can be dissolved in a proper solution for homogeneous mixing. To clearly understand the J_c behavior and superconductivity of MgB$_2$ using C$_{16}$H$_{10}$, the lattice parameters, the actual amount of C substituted, the lattice strain, the MgO fraction, and the transport J_c were evaluated and compared to pristine MgB$_2$/Fe wires. Magnesium (Mg, 99%), boron (B, 99%), and pyrene (C$_{16}$H$_{10}$, 98%) powders were used as starting materials. The mixed powders were packed into an Fe tube, which was then drawn to an outer diameter of 1.42 mm. The fabricated wires were sintered at different temperatures within a wide range from 600 to 800 °C for 30 min to 4 h under high purity argon gas and then cooled in the furnace. The heating rate was 5 °C min^{-1}.

Figure 1(a) shows the calculated results for pristine MgB$_2$ and MgB$_2$+C$_{16}$H$_{10}$/Fe wires as a function of the amount of pyrene, C$_{16}$H$_{10}$. All wire samples were sintered at 650 °C for 30 min. We observed that the a-axis parameter of MgB$_2$+C$_{16}$H$_{10}$/Fe wires decreased gradually with increasing

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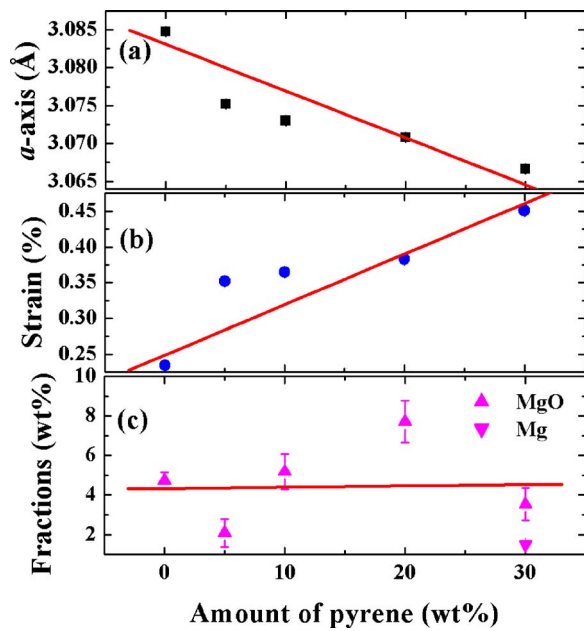FIG. 1. (Color online) (a) The *a*-axis lattice parameter, (b) lattice strain, and (c) fraction of MgO/Mg for pristine MgB$_2$ and MgB$_2$+C$_{16}$H$_{10}$/Fe wires as a function of the amount of pyrene. All wire samples were sintered at 650 °C for 30 min. The solid line show a linear fit to the data.

FIG. 2. (Color online) (a) The *a*-axis lattice parameter, (b) *c*-axis lattice parameter, and (c) actual amount of carbon (C) substitution (*x*) in the composition of Mg(B$_{1-x}$C$_x$)$_2$ for pristine MgB$_2$ and MgB$_2$+10 wt % C$_{16}$H$_{10}$/Fe wires as a function of sintering temperature. The wire samples were sintered at 600 °C for 4 h, 620 °C for 1 h, 650 °C for 30 min, 700 °C for 30 min, 800 °C for 30 min, frespectively. The solid line show a linear fit to the data.

amounts of C$_{16}$H$_{10}$. The shrinkage of the *a*-axis lattice parameter can be attributed to the substitution of C with small atomic size, which causes disorder in the MgB$_2$ structure that can potentially change the properties of MgB$_2$.[13,15] What is interesting is that C substitution into B sites took place even at the low sintering temperature of ~650 °C. To further clarify the effects of C substitution, we calculated the lattice strain of the structure as a function of the amount of C$_{16}$H$_{10}$, as can be seen in Fig. 1(b). Lattice strain is usually an interesting parameter for quantifying disorder. Estimated strain values at different doping levels showed the opposite trend to the changes in the *a*-axis parameter. This indicates that more C substitution has introduced higher disorder into the MgB$_2$ structure. However, the MgO fraction does not increase with increasing doping level. Figure 1(c) shows the calculated MgO fraction of pristine MgB$_2$ and of MgB$_2$+C$_{16}$H$_{10}$/Fe wires on the basis of Rietveld refinement. At the very least, the MgO fraction for MgB$_2$+10 wt % C$_{16}$H$_{10}$/Fe wires is less than or similar to that found in the pristine MgB$_2$ under our experimental conditions. Thus, the MgO effect on the degradation of J_c is negligible.

Figure 2 shows the calculated results for pristine MgB$_2$ and MgB$_2$+10 wt % C$_{16}$H$_{10}$/Fe wires as a function of sintering temperature. The wire samples were sintered at 600 °C for 4 h, 620 °C for 1 h, 650 °C for 30 min, 700 °C for 30 min, and 800 °C for 30 min, respectively. What is surprising is that the *a*-axis parameter of MgB$_2$+10 wt % C$_{16}$H$_{10}$/Fe wires showed a slight increasing trend from 3.0684 to 3.0724 Å with increasing sintering temperature [Fig. 2(a)]. However, this is not an ordinary phenomenon. For example, common C additives, such as C, SiC, and carbon nanotube (CNT), can achieve much more C substitution into the MgB$_2$ structure as the sintering temperature further increases, resulting in a reduced *a*-axis parameter and enhanced B_{irr} and J_c.[16] The *c*-axis parameter also mimics the change in the *a*-axis parameter [Fig. 2(b)]. Obviously, a low sintering temperature can give rise to higher disorder in the
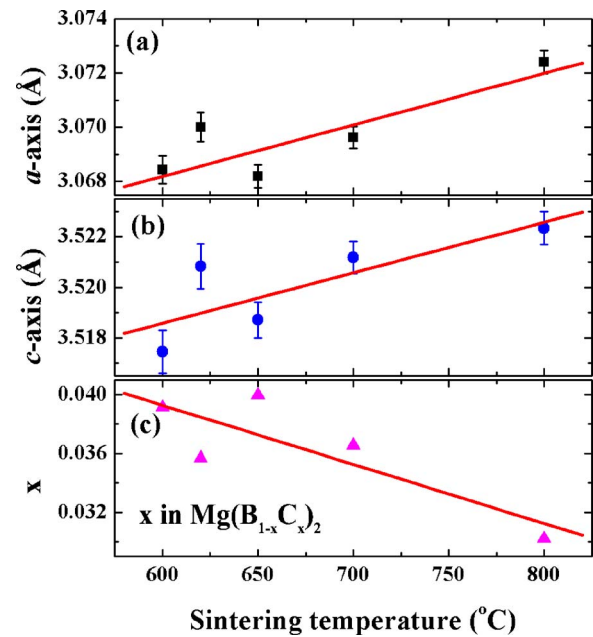
MgB$_2$ structure, leading to broadening of the diffraction peaks. This implies combined effects of crystalline size reduction and increased lattice strain. Specifically, the crystalline size in MgB$_2$ superconductor sample can be attributed to the number of grain boundaries. Based on changes in the *a*-axis parameter, the actual amount of C substitution can be estimated by comparing the calculated value with that of a single crystal. The actual C substitution[15] for the sample sintered at 600 °C was about $x \approx 0.03913$ in the composition of Mg(B$_{1-x}$C$_x$)$_2$. As a result, the maximum C substitution level can be easily achieved at a low sintering temperature of 600 °C under our experimental conditions [Fig. 2(c)]. At this temperature, the much greater amount of C substituted on the B sites introduces strong electron scattering centers that reduce the electron mean free path and, thus, could significantly enhance B_{irr}.[13]
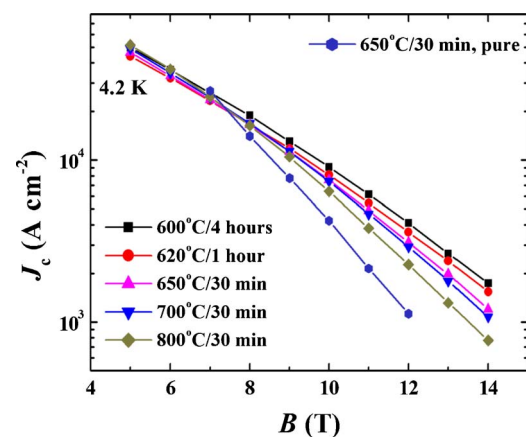
Figure 3 shows the transport J_c of pristine MgB$_2$ and

FIG. 3. (Color online) Transport critical current density (J_c) of pristine MgB$_2$ and MgB$_2$+10 wt % C$_{16}$H$_{10}$/Fe wires as a function of applied magnetic field and sintering temperature.

$MgB_2 + 10$ wt % $C_{16}H_{10}$/Fe wires sintered within a temperature range of 600 to 800 °C. The J_c of the $MgB_2 + 10$ wt % $C_{16}H_{10}$/Fe wires gradually decreased as the sintering temperature increased. This is because grain boundaries are likely to be acting as the predominant pinning centers.[17] In addition, the amount of C substitution decreased with increasing sintering temperature. Note that J_c of the $MgB_2 + 10$ wt % $C_{16}H_{10}$/Fe wires was higher than that of the pristine MgB_2 wire under magnetic field from 7 to 12 T. Transport J_c did not show any large differences among the $MgB_2 + 10$ wt % $C_{16}H_{10}$ wire samples. As a result, the $MgB_2 + 10$ wt % $C_{16}H_{10}$/Fe wire sintered at 600 °C showed better J_c performance, compared to samples doped with other forms of C and sintered at a similar temperature.[16]

In summary, using pyrene, $C_{16}H_{10}$, is effective for enhancing J_c of MgB_2 superconductor, even at sintering temperatures as low as 600 °C. Specifically, the J_c of the sample sintered at 600 °C for 4 h was $\sim 10\,000$ A cm^{-2} at 10 T and 4.2 K. In addition, the MgO fraction for $MgB_2 + 10$ wt % $C_{16}H_{10}$/Fe wires was less than or similar to that found in the pristine MgB_2 under our experimental conditions. The advantages of using $C_{16}H_{10}$ include the production of highly active C due to the decomposition of the pyrene and the introduction of small grain size due to low temperature sintering, resulting in enhancement of J_c.

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