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Y. Zhang
University of Wollongong, yz268@uow.edu.au

S. Zhou
University of Wollongong, sihai_zhou@uow.edu.au

Alexey Pan
University of Wollongong, pan@uow.edu.au

S. X. Dou
University of Wollongong, shi@uow.edu.au

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Effect of Sucrose ($C_{12}H_{22}O_{11}$) Doping on the Critical Current Density of MgB_2

Y. Zhang, S. H. Zhou, A. V. Pan, and S. X. Dou

***Abstract*—MgB_2 bulk samples doped with sucrose were fabricated with the formula $MgB_{2-x}C_x$ ($x = 0$, 0.08, 0.12, 0.2, 0.3, 0.5). Appropriate amounts of sucrose corresponding to the desired carbon content were added. The doping effect of sucrose was observed. It was found that samples doped with $x = 0.2$ sucrose have optimized current density at high magnetic field. The optimized sintering temperature for high current density was found to be 850°C. H_{c2} and H_{irr} were both found to be improved due to the sucrose doping.**

***Index Terms*—Critical current, doping, magnesium diboride, sucrose.**

I. Introduction

MgB_2 has been regarded as one of the most promising superconductor materials since its discovery in the year 2001 [1]. This superconductor has already been fabricated in the bulk, wire and film states. The properties of MgB_2 have been studied extensively [2]–[8]. On the one hand, its high T_c, low material cost, and good weak-link tolerance [5], [9] are very advantageous for practical applications. On the other hand, several issues exist that urgently need to be solved, such as its low H_{c2} and rapid decrease in critical current density (J_c) under magnetic field compared to Nb-based superconductors. High critical current density and good $J_c(\mathrm{H})$ performance are crucial for the application of this material in the so called "strong electrical application" field. Intensive studies have been focused on the improvement of critical current density (J_c) and H_{c2} since the discovery of MgB_2. So far, a number of experimental techniques, including chemical doping (addition or substitution), proton irradiation [2], [10], and various types of thermomechanical processing, have been attempted to realize these purposes.

Currently, from more practical and scalable considerations, addition or substitution of nanoparticles into MgB_2, which causes chemical and nanostructural changes, seem to be an effective way to induce flux pinning centers in MgB_2, thereby improving J_c and H_{c2} [11]. Among the types of chemical doping, nanosize SiC doping has achieved a considerable J_c improvement. The J_c of SiC doped MgB_2 has achieved $4 \times 10^4\ \mathrm{A/cm^2}$ at 20 K and 4 T [12]. Some other nanosize dopants have also shown J_c improvement to different levels [13]–[15]. However, doping with nanomaterials gives rise to two concerns: one is agglomeration of the particles when they are mixed with MgB_2 raw materials, which make it hard to achieve an uniform structure [16], and the other factor is the high cost of nanomaterial, incurring economic disadvantages.

Carbon has been proved to be an effective dopant element for improving H_{c2} and J_c of MgB_2 superconductor. Tiny carbon particles and carbon nanotubes have shown a strong enhancing effect on the superconducting properties, although agglomeration is still a problem [17]–[20]. Carbohydrate is a very good carbon source for C doping into MgB_2. Carbohydrate materials provide carbon as an element when they are heated above the decomposition temperature. This freshly obtained carbon is distributed more uniformly than with carbon nanotubes. Commercial sugar has been used recently as a dopant in MgB_2 because it is cheap and easy to find. We have shown that sugar doping has benefits for $J_c(\mathrm{H})$ improvement [21]. In this paper, the fabrication process was studied more systematically and the parameters were optimized.

II. Experimental Detail

MgB_2 bulks were prepared by the in-situ solid state sintering method. Powders of Mg (99%), amorphous B (99%), and 98% sucrose were used as starting materials to achieve the formula of $MgB_{2-x}C_x$ ($x = 0$, 0.08, 0.12, 0.2, 0.3, 0.5). Appropriate amounts of sucrose based on the corresponding carbon content were added in to the MgB_2. In order to investigate the effects of sucrose doping on MgB_2 bulks, B powder with 100 nm particle size was mixed with sucrose with the help of de-ionized water. The slurry was dried in a vacuum oven. After drying, the B powder was coated with a sucrose layer. This obtained B and sucrose mixture was mixed with Mg powder. The powder was ground by hand with a mortar and pestle. Appropriate mixtures of these powders were pressed into pellets and sealed in Fe tubes. This packing process was carried out in air. The samples were heated from room temperature to 780°C–1050°C in a tube furnace under Ar atmosphere at ambient pressure with a $5^\circ\mathrm{C/min}$ heating rate, and kept at the set temperature for 1 hour before being furnace cooled to room temperature.

The MgB_2 pellets were then taken out by cutting the iron tube open. The phase analysis was carried out by X-ray diffraction (XRD) in a Phillips PW1730 Model diffractometer using Cu $\mathrm{K}\alpha$ radiation. The compositional analyses were performed in an energy dispersive X-ray spectroscopy (EDX) system. The magnetization as a function of temperature T and magnetic field H applied along the longest sample dimension was measured

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The authors are with the Institute for Superconducting and Electronic Materials, University of Wollongong, Wollongong, NSW 2522, Australia (e-mail: yz268@uow.edu.au; sihai@uow.edu.au; pan@uow.edu.au; shi@uow.edu.au).

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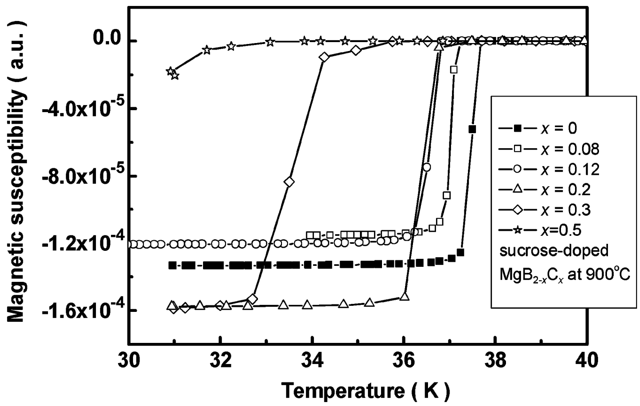

Fig. 1. Magnetic AC susceptibility for $MgB_{2-x}C_x$ as a function of temperature for different carbon doping content. Carbon added was in the form of sucrose ($C_{12}H_{22}O_{11}$). As the doping content increases, the onset transition temperature decreases.

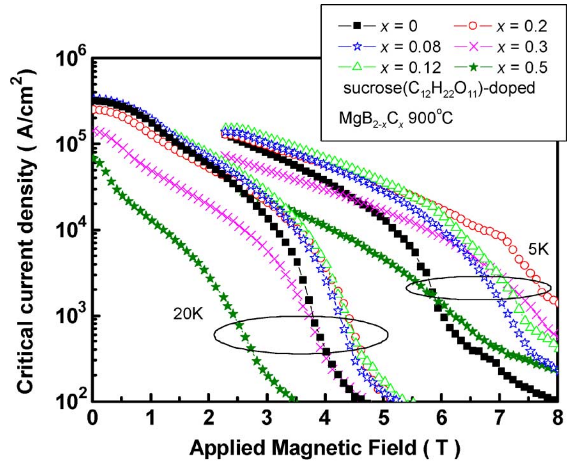

Fig. 2. Critical current density, J_c, as a function of applied magnetic field for all sucrose-doped samples of $MgB_{2-x}C_x$ at 5 K and 20 K.

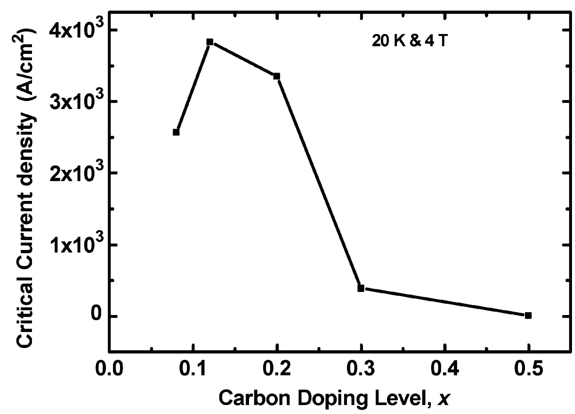

Fig. 3. Critical current density as a function of x for $MgB_{2-x}C_x$, with sucrose as the carbon source.

using Quantum Design Magnetic Property and Physical Property Measurement Systems within the field range $H < 9$ T, and within the temperature range of $5 \text{ K} < T < 30 \text{ K}$. The magnetic J_c was derived from the half-width of the magnetization difference between the descending branches (M^+) and ascending branches (M^-) of the magnetization loop, using the following critical state model formula: $J_c = k\Delta M/d$, where $k = 12w/(3w-d)$ is a geometrical factor and $\Delta M = (|M^+| + |M^-|)/2/V_{l\times w\times d}$, with l, d and w being the sample length, thickness and width, respectively. The typical dimensions of the samples used for magnetization measurements are $3 \times 2 \times 1 \text{ mm}^3$. The T_c was determined by measuring the real part of the ac susceptibility at a frequency of 117 Hz and an external magnetic field of 0.1 Oe. T_c was defined as the onset of diamagnetism.

III. Results and Discussion

A. Effect of Doping Level

A series of samples doped with different levels of sucrose were synthesized to study the effect of the doping level on the superconducting properties, with the nominal composition $MgB_{2-x}C_x$ (where x varies from 0 to 0.5). These samples were sintered at 900°C with a 5°C/min heating rate. Fig. 1 shows the transition temperature (T_c) for the doped and undoped samples, as determined by ac susceptibility measurements. The T_c noticeably decreased with an increasing sucrose doping level. The T_c onset for the undoped samples is around 37.7 K. The $x = 0.5$ doped sample has a T_c of 32.0 K. When $x = 0.2$, T_c is 36.8 K. The T_c decrease might be a result of increased impurity phases introduced by sucrose doping or as a result of C substitution for B in MgB_2.

Fig. 2 shows the magnetic field dependence of the J_c for these samples. It can be seen the doping has strong effects on the J_c both at 5 K and 20 K. Compared with the undoped sample, samples doped with $x = 0.08, 0.12$, and 0.2 experience positive effects. At 5 K and 6 T, the $x = 0.2$ sample shows one order of magnitude improvement. Above the $x = 0.2$ level, samples such as $x = 0.3$ and $x = 0.5$ show less improvement.

This can be more clearly shown in Fig. 3, which contains the J_c performance at 20 K and 4 T. The J_c reached a peak at $x = 0.12$. The J_c decrease at higher doping levels is a result of Tc decrease, and also might be related to excessive impurity phase produced by the sucrose. These impurity phases could block the current path, thus resulting in shrinkage of the effective current conducting area. Phase analysis was carried out using X-ray diffraction, and Fig. 4 shows that the impurity phases increased with the doping level. Judging from the T_c and J_c performance, we conclude that the doping level of $x = 0.2$ is the optimized doping level.

B. Effect of Sintering Temperature

In order to optimize the sintering temperature, samples doped at $x = 0.2$ was sintered at temperatures ranging from 780°C to 1050°C. T_c of these samples is shown in Fig. 5. A pure sample sintered at 900°C was also included for reference. It was found that the T_c increased with the sintering temperature. This T_c increase is the sign of improved crystallinity.

Fig. 6 shows the $J_c(\text{H})$ performance for these samples. The sample sintered at 850°C has the best $J_c(\text{H})$ performance. At 20 K, the $x = 0.2$ sample has higher J_c both at low field and at high field. At 5 K, the sample sintered at 780°C has higher J_c than the 850°C one. Higher sintering temperatures

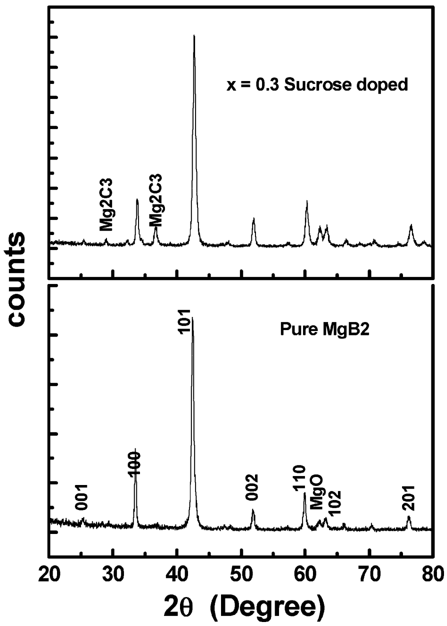

Fig. 4. X-ray diffraction pattern (Cu Kα radiation) for the pure MgB_2 and the sucrose-doped sample $MgB_{1.7}C_{0.3}$. The peaks of Mg_2C_3 and MgO are marked.

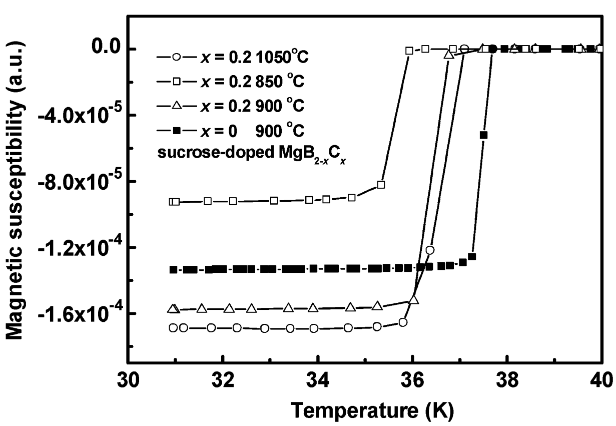

Fig. 5. Magnetic AC susceptibility as a function of temperature for the pure MgB_2 and sucrose-doped $MgB_{1.8}C_{0.2}$ sintered at various temperatures.

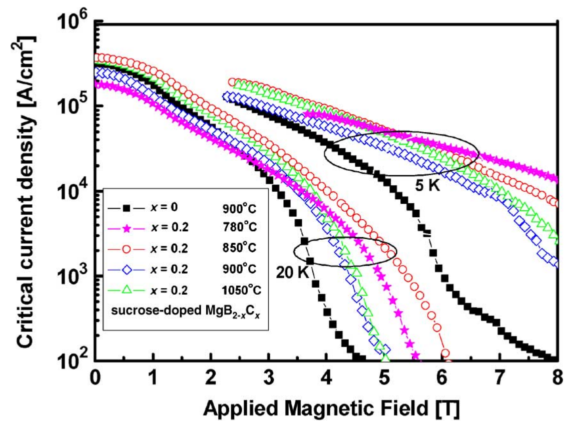

Fig. 6. $J_c(\mathrm{H})$ curves for pure MgB_2 and sucrose-doped $MgB_{1.8}C_{0.2}$ sintered at various temperatures.

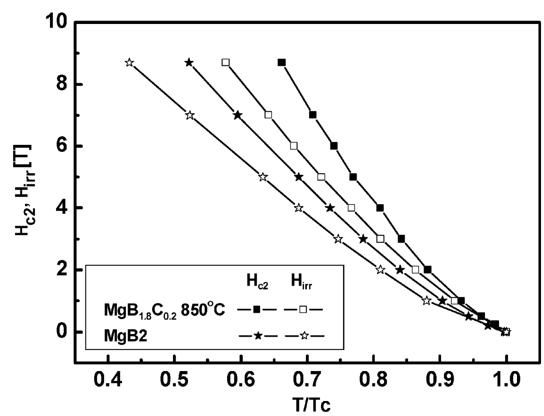

Fig. 7. Normalized temperature $(\mathrm{T}/\mathrm{T_c})$ dependence of the upper critical field, H_{c2}, and the irreversibility field, H_{irr}, for pure MgB_2 and sucrose-doped $MgB_{1.8}C_{0.2}$. The latter was sintered at 850°C.

than 850°C resulted in poorer $J_c(\mathrm{H})$ performance. In our experiments, judging from XRD results and $J_c(\mathrm{H})$ performance, J_c is related to the amount of impurity phase and grain boundary pinning. At lower sintering temperatures than 780°C, the doped carbon did not reacted fully with the MgB_2, so the doping effect of the carbon was not fully utilized, and J_c at higher field is not high. When the sintering temperature is higher than 850°C, the crystal grain size might grow larger due to the thermal effects which reduce the grain boundary area, thus reducing the grain boundary pinning, and decreasing J_c at high field due to the lack of pinning centers, as indicated in reference [22]. The 850°C sintering temperature is a compromise temperature chosen to produce the best overall $J_c(\mathrm{H})$ performance. Fig. 7 is the temperature dependence of H_{irr} and H_{c2} for the doped and undoped samples sintered at 850°C. The temperature was normalized to the samples' T_c. It is obvious that H_{irr} and H_{c2} both increased after doping with sucrose.

IV. Conclusion

Good $J_c(\mathrm{H})$ performance was found in sucrose doped MgB_2 superconductor. T_c was impeded by the doping, but can still remain reasonably high (above 34 K at doping levels less than $x = 0.2$). H_{c2} and H_{irr} both shifted towards higher field. This work has opened up way of improving MgB_2 superconducting properties by doping with a carbohydrate [23] such as sucrose. Further work is likely to produce even more promising results.

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