



Effect of combined addition of graphene oxide and citric acid on superconducting properties of MgB₂



Sudesh^a, S. Das^b, C. Bernhard^b, G.D. Varma^{a,*}

^a Department of Physics, Indian Institute of Technology Roorkee, Roorkee 247667, India

^b Department of Physics and Fribourg Centre for Nanomaterials-FriMat, University of Fribourg, Chemin du Musée 3, CH-1700 Fribourg, Switzerland

ARTICLE INFO

Article history:

Received 27 November 2014

Accepted 20 December 2014

Available online 30 December 2014

Keywords:

MgB₂ superconductor

Upper critical field

Critical current density

Flux pinning

ABSTRACT

In the present work, polycrystalline samples with compositions MgB₂ + 3wt% GO + *x* wt% C₆H₈O₇ (*x* = 0, 5 and 10) have been synthesized to study the effect of combined addition of graphene oxide (GO) and citric acid (C₆H₈O₇) on superconducting properties of MgB₂. X-ray diffraction studies show the formation of hexagonal crystal structure of MgB₂ with space group P6/mmm in all synthesized samples. We observe that the addition of GO in the sample improves the grain connectivity and consequently enhances the critical current density significantly with no substantial change in *T_c*. However for this sample, there is no significant improvement in *H_{c2}* and *H_{irr}*. With the combined addition of GO and citric acid, the *J_c(H)*, *H_{c2}* and *H_{irr}* are observed to improve substantially as compared to the pristine MgB₂ and GO added MgB₂ samples. For example *J_c*(10 K, 5 T) of sample *x* = 10 has improved by a factor of ~15 as compared to pure MgB₂ sample and by a factor of ~5.5 as compared to the *x* = 0 sample. Furthermore, *H_{c2}*(0) for *x* = 10 sample has enhanced by 13 T as compared to pure MgB₂ while it has increased by ~10 T in comparison to *x* = 0 sample. Enhanced flux pinning has been observed with the combined addition of GO and citric acid.

© 2014 Elsevier B.V. All rights reserved.

1. Introduction

Superconductivity in MgB₂, a binary intermetallic compound, was discovered by Akimitsu's group in 2001 [1]. This simple compound has a high superconducting critical temperature (*T_c*) of ~40 K. Its low anisotropy, absence of weak links and low fabrication cost make this material promising over the conventional low *T_c* superconductors for various technological applications. However, the rapid drop of critical current density (*J_c*) under the application of magnetic field is disadvantageous for taking this material into many practical applications. An extensive research has been done in order to further enhance the superconducting properties of MgB₂, such as critical current density (*J_c*), upper critical field (*H_{c2}*) and irreversibility field (*H_{irr}*). The substitution of C at the B-site by using various forms of C-containing compounds, e.g. SiC [2,3], C [4–6], B₄C [7], carbohydrates [8,9] and some organic compounds [6,10–12], have been found to be very effective in improving *H_{c2}*. However, the chemical substitution in MgB₂ leads to reduction in critical temperature, *T_c* [13,14]. More recently, it

has been reported that the *J_c* values have been improved significantly by adding graphene [15,16], reduced graphene oxide (rGO) [17] and graphene oxide (GO) [18] into MgB₂ without affecting *T_c* much. It is studied that the addition of graphene, rGO or GO lead to improved inter-grain connectivity in the samples and thus leading to better conduction and hence *J_c*. In the previous study by our group [18], it has been found that with the addition of GO into MgB₂, *J_c* improves significantly with no substantial change in *T_c*. However, only a marginal improvement in *H_{c2}* and *H_{irr}* was observed. This is possibly due to negligible amount of C substitution at the B-site and hence less lattice distortion and consequently lesser charge carrier scattering. This has been proved from the XRD data which shows no significant change in lattice parameters on GO addition. In our previous study [6], we have found significant improvement in *H_{c2}* of citric acid (C₆H₈O₇) added MgB₂. In the present work, we have added GO and citric acid together in MgB₂ with the aim to improve *J_c(H)*, *H_{c2}* and *H_{irr}*. Polycrystalline samples with compositions MgB₂ + 3 wt% GO + *x* wt% C₆H₈O₇ (*x* = 0, 5 and 10) have been prepared using the standard solid-state reaction route. We observe that with the combined addition of GO and citric acid, the *J_c(H)*, *H_{c2}* and *H_{irr}* improve substantially as compared to the pristine MgB₂ and GO added MgB₂ samples. Enhanced flux pinning has been observed with the combined addition of GO and citric acid.

* Corresponding author at: Department of Physics, Indian Institute of Technology Roorkee, Roorkee 247667, Uttarakhand, India. Tel.: +91 1332 285353; fax: +91 1332 273560.

E-mail address: gvarfph@iitr.ernet.in (G.D. Varma).

2. Experimental

All the bulk polycrystalline MgB_2 samples were synthesized by employing the standard solid-state reaction method. Graphene oxide (GO) used for the addition has been synthesized using the method developed by Marcano et al. [19] as explained earlier [18]. High quality Mg (Sigma Aldrich, 99.9% pure, $\sim 100 \mu\text{m}$ grain size) and B (Sigma Aldrich, amorphous, 99%, submicron grains size) powders were weighed in appropriate quantities to form MgB_2 . The mixture was properly ground and the resulting powder was pressed into rectangular pellets. We further synthesized the samples with compositions $\text{MgB}_2 + 3 \text{ wt\% GO} + x \text{ wt\% citric acid}$ ($x = 0, 5$ and 10). For all samples, an extra 5 wt% quantity of Mg was added to compensate for the Mg loss during sintering. The pellets were put in a Fe-tube with holes in the cap (for the inlet of Ar/H_2 gas) for sintering. The samples were sintered at 850°C in Ar/H_2 (9:1) for 3 h followed by quenching at 650°C to room temperature. The samples are henceforth named as MB (MgB_2), MBG ($\text{MgB}_2 + 3\text{wt\% GO}$), MBGC05 ($\text{MgB}_2 + 3 \text{ wt\% GO} + 5 \text{ wt\% citric acid}$) and MBGC10 ($\text{MgB}_2 + 3 \text{ wt\% GO} + 10 \text{ wt\% citric acid}$). The X-ray diffraction (XRD) pattern was recorded for phase identification of samples with a diffractometer (Bruker AXS, D8 ADVANCE) using $\text{Cu K}\alpha$ radiation. The microstructure of all the samples was studied using field emission scanning electron microscope (FESEM). Resistivity measurements were carried out using standard four-probe technique under applied magnetic field up to 8 T. The DC magnetization measurements were done using the Vibrating Sample Magnetometer (VSM) facility of PPMS (Quantum Design) performed at University of Fribourg, Switzerland.

3. Results and discussion

The XRD patterns of MB, MBG, MBGC05 and MBGC10 samples are shown in Fig. 1. All the samples show MgB_2 main phase with trace amounts of MgO impurity phase. The indexing of characteristic peaks corresponding to MgB_2 is shown in Fig. 1. The peaks corresponding to MgO are marked by *. The presence of MgO is unavoidable with the present synthesis method due to some entrapped air before the sample is enclosed in the iron tube for sintering and also due to oxide nature of dopants. Refinement of the XRD patterns was done using X'pert HighScore to obtain the structural parameters such as lattice parameters, a and c and FWHM. The obtained parameters are listed in Table 1. The volume percentage of MgO as obtained from the refinement of the XRD data are shown in Table 1 for all samples. The inset in Fig. 1 shows the expanded view of peaks (110) and (002). It is clear, within the accuracy of measurements, that with increasing wt% of citric acid the shifting of (110) peak is more prominent than that of (002)

peak, which indicates a significant change in the in-plane lattice parameters. We observe only a small change in the lattice parameters of MBG sample with respect to MB sample. However, when citric acid is added in the samples together with GO, a substantial change in the lattice parameters a and c with respect to MB sample is observed as listed in Table 1. From the variation of lattice parameters, the content of carbon entering into the lattice is calculated using the relation: $y = 7.5 \times \Delta c/a$, where $\Delta c/a$ is the change in c/a value as compared to that of the pure MgB_2 sample and y is the exact value of C (atomic wt%) substituted for B in the sample [13]. The obtained values (see Table 1) show that in case of GO addition, the amount of C entering into the lattice is very small, while in case of citric acid and GO addition, there is larger amount of C substituting for B into the lattice. This is possibly due to lack of dissociation of GO into its constituents at the sintering temperature, thus less carbon is available for doping.

The variation of full width at half maximum (FWHM) of all the samples obtained for the peaks (002) and (110) of MgB_2 are shown in Fig. 2. The FWHM represents the peak broadening in the sample and is influenced by crystallite size and lattice strain [20]. We observe from this figure that on addition of citric acid, the increase in FWHM of peak (110) is more than that of the peak (002). As the (110) plane corresponds to the in-plane lattice, so the broadening of (110) indicates increased distortion in the ab -plane due to C-substitution at the B-site of MBGC05 and MBGC10 samples. The lattice strain and crystallite size of the samples have been calculated using the Williamson-Hall [21] plots for all the samples. The calculated values for strain are shown in Fig. 2. It is observed that as compared to MB sample, the lattice strain has increased significantly in MBG, MBGC05 and MBGC10 samples. This further confirms the increased lattice distortion in MBG, MBGC05 and MBGC10 samples. The crystallite sizes for all the samples are given in Table 1. We see that the crystallite size of the samples has decreased with addition of citric acid.

FESEM micrographs of all the samples are presented in Fig. 3. We observe that the grain size has reduced with increased citric acid doping in the sample which confirms the XRD results of increased FWHM values. Also it is clearly observed from the micrographs that in the samples added with GO, grain connectivity is improved in comparison to the pure MgB_2 sample as is clear from the improved mass density (see Table 1). This feature has also been reported earlier for graphene, rGO and GO added samples [16], [17]. However, in the GO and citric acid added samples, connectivity is better than the pure MgB_2 sample but less than the GO added sample.

The temperature dependence of resistivity, normalized with its room temperature value ($\rho(T)/\rho(300 \text{ K})$), of all the polycrystalline samples is shown in Fig. 4(a). Fig. 4(b) shows the superconducting transition in the temperature range 35–40 K. The critical temperatures (T_c), defined as the onset of superconducting transition, are listed in Table 2. The T_c is 38.84 K for pure MgB_2 . For the GO added sample, we observe no significant change in T_c (=38.67 K). However, T_c decreases for the citric acid added samples, $T_c = 38.20 \text{ K}$ and 37.92 K for MBGC05 and MBGC10 samples, respectively. This is due to the increased substitution of C at the B-site in the lattice. The normal state resistivity ($\rho_{40 \text{ K}}$) for GO doped sample is almost the same as that of the pure MgB_2 sample while with combined addition of GO and citric acid $\rho_{40 \text{ K}}$ has increased, this is due to the enhanced impurity scattering with C substitution as well as the increased MgO content in the MBGC05 and MBGC10 samples. In polycrystalline samples, the grain connectivity is a major factor to affect the conduction in the material [22]. The porous nature of MgB_2 and the presence of MgO impurity phase tend to decrease the grain connectivity. The grain connectivity in MgB_2 can be estimated by knowing the value of $\Delta\rho$ ($\rho_{300 \text{ K}} - \rho_{40 \text{ K}}$) as suggested by Rowell [22]. From this parameter, the effective area cross

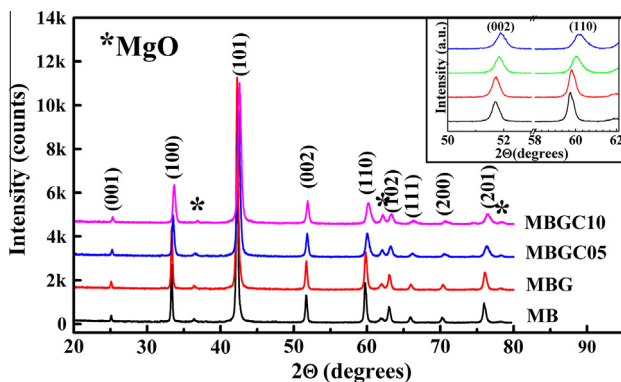


Fig. 1. X-ray diffraction patterns for MB, MBG, MBGC05 and MBGC10 samples. * Represents the impurity phase MgO in all the samples.

Download English Version:

<https://daneshyari.com/en/article/1817782>

Download Persian Version:

<https://daneshyari.com/article/1817782>

[Daneshyari.com](https://daneshyari.com)