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Significant enhancement of critical current density by effective carbon-doping in MgB_2 thin films

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Abstract

The pure and carbon (C)-doped MgB_2 thin films were fabricated on Al_2O_3 (0001) substrates at a temperature of 650 °C by using hot-filament-assisted hybrid physical–chemical vapor deposition technique. The T_c value for pure MgB_2 film is 38.5 K, while it is between 30 and 35 K for carbon-doped MgB_2 films. Expansion in *c*-axis lattice parameter was observed with increase in carbon doping concentration which is in contrast to carbon-doped MgB_2 single crystals. Significant enhancement in the critical current density was obtained for C-doped MgB_2 films as compared to the undoped MgB_2 film. This enhancement is most probably due to the incorporation of C into MgB_2 and the high density of grain boundaries, both help in the pinning of vortices and result in improved superconducting performance.

Keywords : MgB_2 film, carbon doping, HFA-HPCVD

1. INTRODUCTION

The remarkably high transition temperature (T_c = 39 K) of MgB_2 makes it practically more advantageous over conventional metallic superconductors [1]. The strongly linked nature of the intergrains with a high charge carrier density in this material makes it further an attractive candidate for the next generation of superconductor applications. For most of the practical applications, high critical current density (J_c) in the presence of a magnetic field is required [2]. The J_c of undoped MgB_2 is high enough 10^6–10^7 A/cm^2 at low magnetic fields for practical application, however, J_c drops rapidly with increasing magnetic field due to the low H_{c2} and the lack of the effective pinning sites in MgB_2 [3]. Therefore the improvement of J_c under magnetic field is indispensable for the development of MgB_2 material for magnet applications. The substitution of carbon atoms into the boron sites of MgB_2 are known to be the most effective way to improve pinning properties in MgB_2 and hence J_c performance under high magnetic fields [4]. The current-carrying performance has been improved by carbon in all the form of MgB_2 superconductors, such as polycrystalline bulk, single crystals, wires, tapes, filaments, fibers and thin films. Despite much research done on enhancement of J_c in MgB_2 bulk samples, only a little work has been reported for improving J_c in MgB_2 films [5]. Few reports have shown the improvement in J_c by means of carbon doping in MgB_2 films [6, 7]. Here, we are reporting the effect of carbon-doping on the superconducting properties of MgB_2 thin films. The carbon-doped MgB_2 films were fabricated by using hot-filament-assisted hybrid physical–chemical vapor deposition (HPCVD) using methane as the doping source.

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2. EXPERIMENTAL

The carbon-doped MgB_2 thin films were fabricated on Al_2O_3 (000*l*) substrates by using a modified HPCVD system. Inspite of capable of producing high-quality MgB_2 films, it is difficult to dope the MgB_2 films using the original HPCVD system [3]. Therefore, additional dopant methane (CH_4) gas line was installed in the original HPCVD system for doping MgB_2 thin films. A separate heater, a Kanthal-super filament, was also installed to decompose CH_4 gas. In this hot-filament-assisted (HFA)-HPCVD process, the Al_2O_3 substrate was placed on the top surface of a susceptor and Mg chips were placed around it. The reactor was firstly evacuated to a base pressure of ~10^{-3} Torr using rotary pump and purged several times by flowing high purity argon and hydrogen gases. Prior to the carbon-doped MgB_2 film growth, the susceptor along with substrate and Mg chips were inductively heated towards the set temperature under a reactor pressure of 100 Torr in H_2 atmosphere. Upon reaching the set temperature, a boron precursor gas, B_2H_6 (5% in H_2) and CH_4 (100%) gas were introduced into the reactor to initiate the film growth. Finally, the fabricated C-doped MgB_2 film was cooled down to room temperature in a flowing H_2 carrier gas. The flow rates were 100 sccm for the H_2 carrier gas and 50 sccm for the B_2H_6/H_2 mixture. CH_4 gas of different flow rates was added to the carrier gas to dope the film with different carbon concentrations. All the carbon-doped MgB_2 films were fabricated at a substrate temperature of 650 ^oC. CH_4 was decomposed by the hot filament at 900 ^oC. A pure MgB_2 film was also prepared under the same conditions for comparison. The nominal carbon concentrations were determined using the correlation between gas concentrations and flow rates. For example, for the flow rates of 50, 100, and 5 sccm of B_2H_6, H_2, and CH_4, respectively, the calculated nominal carbon concentration is 3.2%.

The crystal structures of pure and carbon-doped MgB_2 films were investigated by X-ray diffraction (D8 discover, Bruker AXS) using Cu Kα as an X-ray source. Scanning electron microscopy (SEM) was used for measuring the surface morphologies and the thicknesses of the films. The standard four-probe method was used to measure the temperature dependence of resistivity for all the prepared films. The magnetization hysteresis (*M–H*) measurements were carried out using a magnetic property measurement system (XL-5S, Quantum Design).

3. RESULTS AND DISCUSSION

The X-ray θ–2θ scans of pure and carbon-doped MgB_2 thin films with different nominal carbon concentrations of 2.6, 3.2, 3.8, and 5.0 % are shown in Fig. 1a. The XRD patterns show only (000*l*) peaks of MgB_2 indicating that the films are *c*-axis oriented. Expansion in *c*-axis lattice parameter was observed with increase in carbon doping concentration, which is similar to carbon-doped films [6] but in contrast to carbon-doped single crystals, where the *c*-axis remains almost constant for all the carbon concentrations [8]. Compared to the undoped film, the MgB_2 (000*l*) peaks are suppressed as carbon concentration increases. The magnification of MgB_2 (0001) peak is shown in Fig. 1b. Moreover, there is no indication of any secondary phases, such as Mg, MgO, and MgB_4.

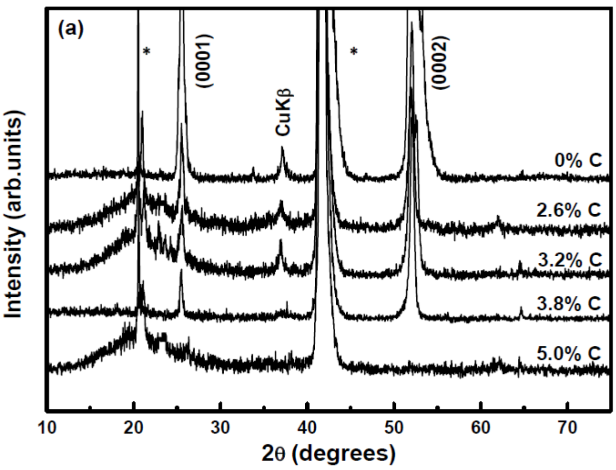

Fig. 1. (a) XRD patterns of pure and carbon-doped MgB_2 thin films.

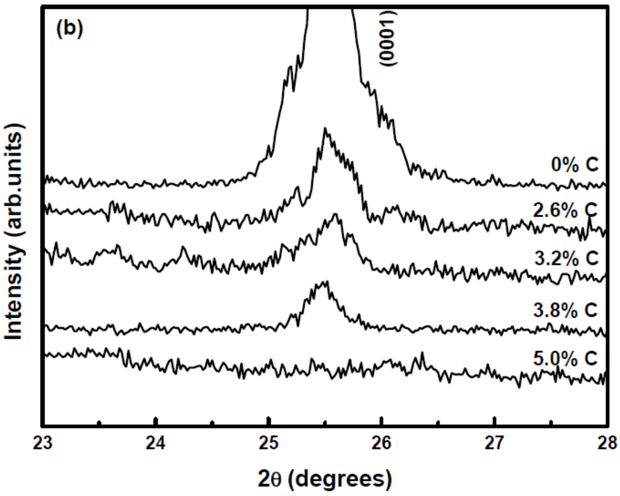

Fig. 1. (b) Magnified view of 0001 peak of MgB_2, suppression of peak was observed as the doping concentration increases.

The resistivity versus temperature curves for pure and carbon-doped MgB_2 films are plotted in Fig. 2. The inset shows the magnified view near the superconducting transition temperature (T_c). The T_c value for pure MgB_2 film is 38.5 K, while it is between 30 and 35 K for carbon-doped MgB_2 films. The reduction in T_c could also be an indicator of carbon substitution at boron sites of MgB_2. The carbon-doped films exhibit a systematic increase in resistivity with increasing carbon concentration, except for 3.2% C-doped sample. The residual resistivity ratio (RRR = $\rho_{300\ K}/\rho_{40\ K}$) values for the pure and C-doped films with concentrations of 2.6, 3.2, 3.8, and 5.0 % are 4.3, 1.4, 1.3, 1.5, and 1.4 respectively. It indicates that the impurity scattering is stronger in carbon-doped MgB_2 films, and hence the decreased RRR values.

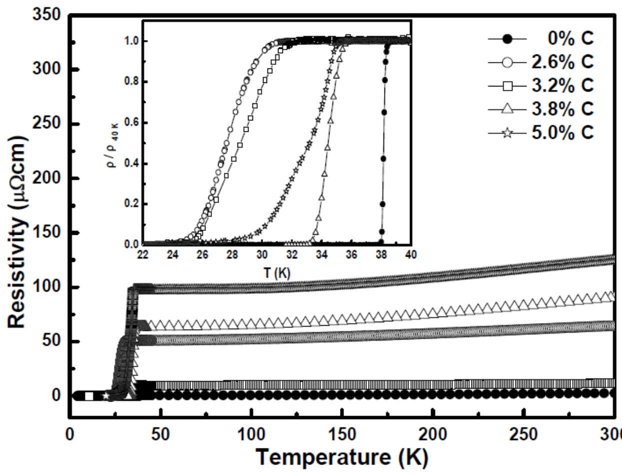

Fig. 2. Resistivity versus temperature curves of pure and carbon-doped MgB_2 films with different nominal carbon concentrations of 2.6, 3.2, 3.8, and 5.0 %.

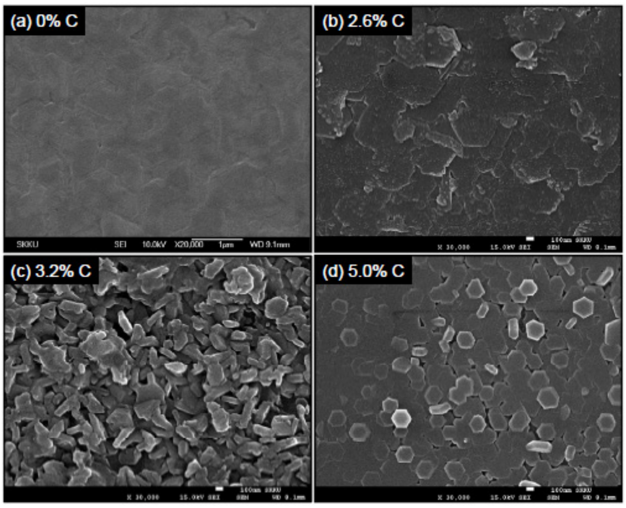

Fig. 3. SEM images of pure (a) and C-doped MgB_2 films with different nominal carbon concentrations of (b) 2.6 %, (c) 3.2 %, and (d) 5.0 %.

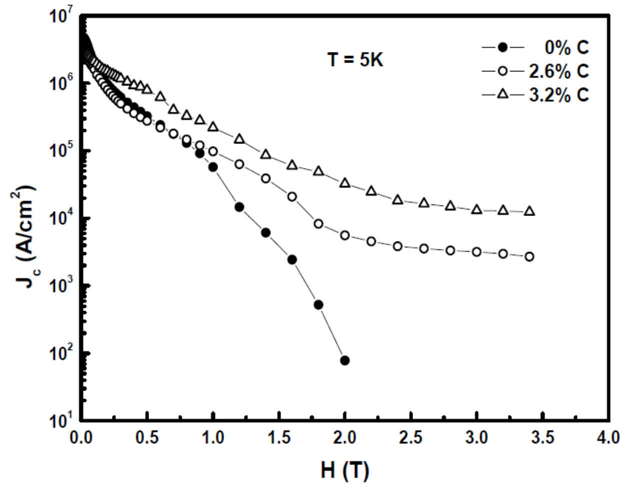

Fig. 4. $J_c(H)$ curves at 5 K for pure and C-doped MgB_2 films with different nominal carbon concentrations of 2.6 and 3.2 %.

The surface morphologies of pure and doped MgB_2 films with different carbon concentrations are shown in Fig. 3a–d. The dense and smooth morphology was observed for pure MgB_2 thin film. As the pure film was doped with carbon, the surface morphology changes from smooth to a planar structure for 2.6 % C-doped MgB_2 film. With further increase in carbon doping concentration, the average grain size was found to decrease from 450 nm for 2.6 % C-doped film to 200 nm for 5.0 % C-doped film. It indicates that the carbon doping suppresses the MgB_2 grain growth and reduces the grain size. Both the carbon doping and the high density of grain boundaries which are known to be the main pinning source in MgB_2, are expected to be beneficial for improving the J_c performance of MgB_2 superconductor.

The critical current density (J_c) is evaluated from the magnetic hysteresis (*M–H*) loops by using the Bean critical state model. Fig. 4 shows the magnetic field dependence of J_c at 5 K for pure and C-doped MgB_2 films. At self-field both the C-doped films show nearly comparable J_c to that of pure sample. This is in contrast to the reported data on C-doped MgB_2 films, where one order of magnitude of reduction in J_c was noticed at self-field and 5 K [6]. It indicates that the inter-grain connectivity for C-doped films is denser and comparable to the pure one. For the undoped film, J_c drops rapidly in the presence of magnetic field and it is 10^2 A/cm^2 at an applied field of 2 T. On the other hand, the 3.2% C-doped MgB_2 film exhibits a J_c of the order of 10^5 A/cm^2 at the corresponding field. Significantly high J_c values at high fields are obtained for the C-doped films. This enhancement in J_c is most likely due to the incorporation of C into MgB_2 and the high density of grain boundaries, both help in the pinning of vertices and result in improved superconducting performance.

4. CONCLUSION

The effect of carbon-doing on microstructure and superconducting properties of MgB_2 thin films were investigated. As compared to pure MgB_2 film, reduction in T_c, decrease in RRR values and increase in resistivity were observed for C-doped MgB_2 films. The average grain size was found to decrease with increase in carbon doping concentration. It indicates that the carbon doping suppresses the MgB_2 grain growth and reduces the grain size. At high magnetic fields the C-doped MgB_2 films exhibit considerably larger critical current density as compared to undoped MgB_2 film. The enhanced J_c could be attributed to the strong flux pinning achieved by the incorporation of C into the MgB_2 as well as by the high density of grain boundaries. These results imply that the HFA-HPCVD would be a promising technique to fabricate C-doped MgB_2 superconducting wires and tapes with high J_c values for large scale applications.

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