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Synthesis of superconductor $MgCNi_3$ with carbon nanotubes*

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$MgCNi_3$, an intermetallic compound superconductor with a cubic perovskite crystal structure, has been synthesized using fine Mg and Ni powders and carbon nanotubes (CNTs) as starting materials by the conventional powder metallurgy method. The composition, microstructure and superconductivity are characterized using x-ray diffraction (XRD), energy dispersive x-ray (EDX) analysis, scanning electron microscopy (SEM), and superconducting quantum interference device (SQUID) magnetometer. The results indicate that the phases of the synthesized samples are $MgCNi_3$ (major phase) and traces of C and MgO. The $MgCNi_3$ particle sizes range from several hundreds of nanometres to several micrometres. The onset superconducting transition temperature T_c of the $MgCNi_3$ sample is about 7.2 K. The critical current density J_c is about 3.44×10^4 A/cm^2 calculated according to the Bean model from the magnetization hysteresis loop of the slab $MgCNi_3$ sample at 5 K and zero applied field.

Keywords: $MgCNi_3$ superconductor, synthesis, carbon nanotubes
PACC: 7400, 7470, 7460J

1. Introduction

Since the remarkable discovery of superconductivity at 39 K in MgB_2,[1] intermetallic compounds have renewed the interest in the search for new superconductor materials. The recently discovered superconductor $MgCNi_3$[2] with a perovskite structure has attracted attention,[3−21] because it is regarded as a bridge between conventional superconductor and high-T_c cuprate, and it is thought to be very close to ferromagnetism due to a large portion of Ni in each unit cell.[7−10] Experiments have shown that the superconductivity of MgC_xNi_3 is very sensitive to the content of carbon and disappears below about $x = 0.88$.[2,17] Synthesis of $MgCNi_3$ superconductor with different carbon sources, including glassy carbon spherical powder,[2,10,14,15,17] graphite,[11−13] amorphous carbon,[8] and liquid paraffin,[12] has been proposed. CNTs have been used as a template for fabricating superconducting polycrystalline Nb nanowires[22] and as a carbon resource for synthesis superconductor NbC.[23] To the best of our knowledge, there have been no reports on the synthesis of $MgCNi_3$ superconductor with carbon nanotubes. In this paper, we report the synthesis of $MgCNi_3$ superconductor bulk material by using CNTs as a source of carbon via the conventional powder metallurgy method. Composition, microstructure and superconductivity of the synthesized $MgCNi_3$ superconductor are investigated.

2. Experiment details

To obtain a perfectly stoichiometric composition $MgCNi_3$ with a higher T_c, excess carbon is required.[2−10,13−17] Due to the volatility of Mg during synthesizing this compound, excess Mg is needed. In this study, samples with the nominal formula $Mg_{1.2}C_{1.45}Ni_3$ were prepared. The starting materials were fine Mg powders (99% pure, –400 mesh), fine Ni powders (99% pure, 1.4 μm), and commercial CNTs (95% pure, outer diameter 40–60 nm, length 0.5–2 μm, available from Shenzhen Nano-Technologies Port Co. Ltd.). The starting materials were mixed and ground thoroughly, and the operation was carried out in a glove box with an Ar environment, in which the content of oxygen and water was as low as a few ppm. Then the mixed powders were pressed

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into tablets with 10 mm in diameter and several millimetres in thickness under an appropriate pressure. The tablets were in turn wrapped with a Ta foil and enclosed in an evacuated quartz tube; the quartz tube was placed in the heating zone of the horizontal furnace tube. The samples were heated for half an hour at 600°C and 3 hours at 900°C, and then cooled to room temperature by furnace cooling. The composition and the microstructure analyses were carried out using an x-ray diffractometer (XRD) with Cu Kα radiation and scanning electron microscopy (SEM) with energy dispersive x-ray (EDX) analysis. DC magnetization measurements were made with a superconducting quantum interference device (SQUID) magnetometer, using small pieces of $MgCNi_3$ rectangular slab.

3. Results and discussion

Figure 1 shows the powder XRD patterns for the $MgCNi_3$ superconductor sample. The phase analysis of the XRD patterns reveals that $MgCNi_3$ is a major phase in the sample. It can be seen that the diffraction peaks at high degrees are splitting for the powder sample, which is due to the Cu Kα2 radiation effect.[19] All impurity peaks are attributable to the traces of C and MgO.

The SEM images in Figs.2(a) and 2(b) illuminate two typical regions of the $MgCNi_3$ sample. Figure 2(a) is the image of the surface of the prepared $MgCNi_3$ sample, and Fig.2(b) is the image of the cross section of the $MgCNi_3$ sample. Both of them show that the $MgCNi_3$ particle sizes range from several hundreds of nanometres to several micrometres. The small particle size is attributable to the fine powder of the starting materials and CNTs. More SEM observations show that there exist also some fluffy regions on the surface and the cross section of the prepared sample. These fluffy regions show unreacted and partly reacted carbon nanotubes by EDX analysis (Fig.3). However, these fluffy regions are smaller than the regions of the un-reacted carbon of the synthesized $MgCNi_3$ superconductor from other carbon source, such as graphite.[13,16]

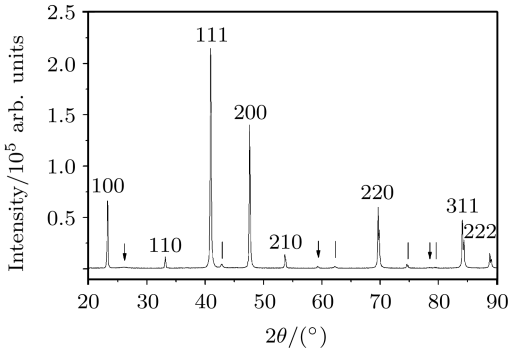


Fig.1. Powder XRD patters of the $MgCNi_3$ sample. The arrows and the lines indicate traces of C and MgO.

The superconducting transition temperature of the sample was determined by magnetization measurement with zero-field cooling (ZFC) at $H = 1200$ A/m. The T_c value was chosen to be the value at the jump point of the dM/dt[14] as shown in Fig.4, and the T_c is about 7.2 K.

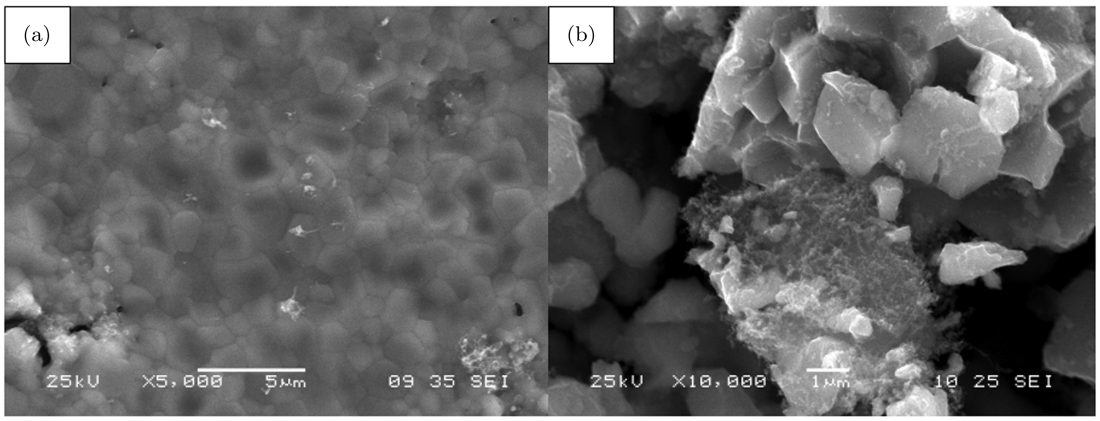


Fig.2. Microstructure of $MgCNi_3$ sample surface (a) and microstructure of $MgCNi_3$ sample cross section (b). Both of them show that the average sizes of grains range from several hundreds of nanometres to several microns by SEM observation.

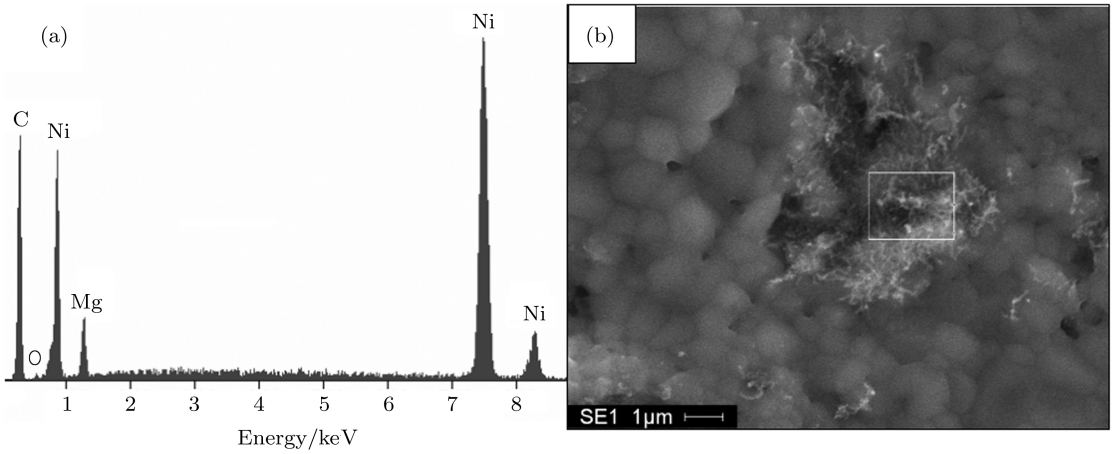


Fig.3. EDX element analysis on the dispersive fluffy regions (a) and SEM image of $MgCNi_3$ sample surface (b).

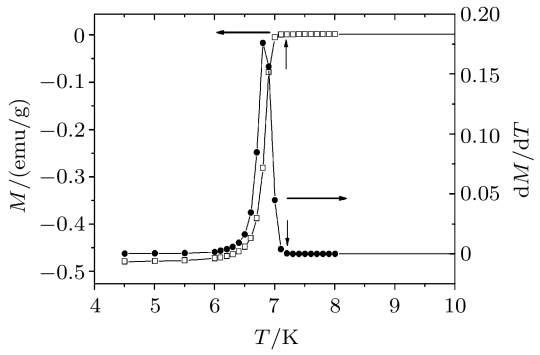


Fig.4. Magnetization of the $MgCNi_3$ sample as a function of temperature.

To estimate the critical current density J_c, the magnetization hysteresis loop of the $MgCNi_3$ sample was measured at 5 K up to a magnetic field of $H = 4 \times 10^6$ A/m (Fig.5). The rectangular slab sample of dimension $a \times b \times c = 1.5 \times 2.0 \times 6.0\,\mathrm{mm}^3$ was used for the measurement and subjected to an externally applied field parallel to the c axial direction. The J_c was calculated according to the Bean model[24−26] from the magnetization hysteresis loop. The calculated field dependence J_c is shown in Fig.6, and it is clear that the J_c are about $3.44 \times 10^4\,\mathrm{A/cm}^2$ in zero applied field, $3.36 \times 10^3\,\mathrm{A/cm}^2$ in an applied field of 8×10^5 A/m and $1.27 \times 10^3\,\mathrm{A/cm}^2$ in an applied field of 1.6×10^6 A/m. These values are larger than the values of J_c for bulk samples in the literature.[15,16] The higher values of J_c for bulk samples may be attributed to the small particle sizes of the samples and the un-reacted and partly reacted CNTs. Since CNTs have many special features such as 1) higher current-carrier density up to $10^9 - 10^{10}\,\mathrm{A/cm}^2$,[27] 2) higher thermal conductivity for isolated multi-walled CNTs estimated to be about $3000\,\mathrm{W/mK}^{-1}$,[28] and 3) very large aspect ratio that can act as a line-pinning site.[29]

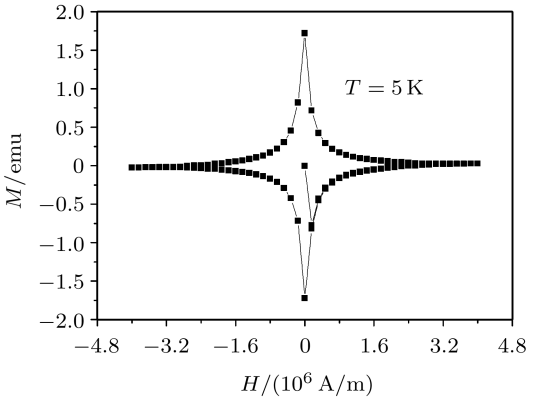


Fig.5. Field dependence of the magnetization hysteresis loop of $MgCNi_3$ sample at 5 K.

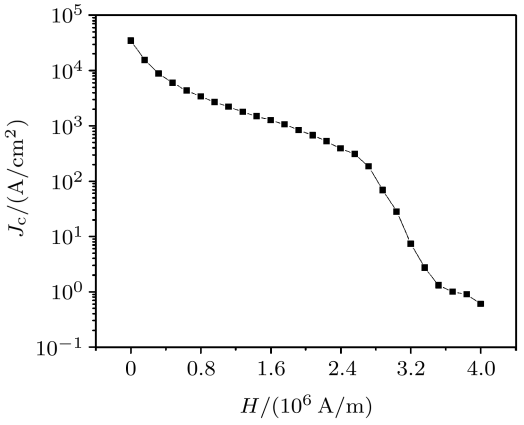


Fig.6. Field dependence of critical current density calculated by Bean model for $MgCNi_3$ slab at 5 K.

4. Conclusion

We have synthesized $MgCNi_3$ bulk materials using CNTs as starting materials by the conventional powder metallurgy method. The sample obtained is composed of $MgCNi_3$ (major phase) and traces of C and MgO with particle sizes ranging from several hundred nanometres to several micrometres which is attributed to the fine powder of the starting materials and CNTs. Its onset superconducting transition temperature is about 7.2 K obtained by magnetization measurement, and the J_c is about 3.44×10^4 A/cm^2 calculated according to the Bean model from the magnetization hysteresis loop of the slab $MgCNi_3$ sample at 5 K and zero applied field.

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