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The Effect of MWCNT Addition on Superconducting Properties of MgB_2 Fabricated by High-Pressure Combustion Synthesis

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Abstract—We successfully prepared superconducting powders of magnesium diboride doped with carbon nanotubes by the method of combustion synthesis under high Ar pressure. Powders of magnesium, boron, and multi-walled carbon nanotubes (MWCNT) were used as starting materials. X-ray diffraction analysis showed the presence of MgB_2 and MgO in combustion products. The temperature dependence of magnetization showed a sharp superconducting transition at around 38.5 K. The critical current density can be estimated from the hysteresis of magnetization curve by using the Bean's formula. MgB_2 doped with MWCNT (1%) showed the best value of high critical current density, 1.4×10^8 A/cm^2 at 5 K, in zero magnetic fields.

Keywords: combustion synthesis, type II superconductor, binary metallic compound, MWCNT doping

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INTRODUCTION

Superconductivity in the binary metallic compound MgB_2 was discovered by Akimitsu et al. [1] and since then a lot of activities both in the physics interests and in the application aspects have stimulated considerable interest in this system. MgB_2 has a transition temperature, T_c, of about 40 K, a hexagonal AlB_2 structure, and is a type II superconductor.

The crystal structure of MgB_2 consists of honeycombed boron layers and magnesium layers located in between the boron layers. The hexagonal unit cell has the lattice parameters $a = 3.086$ Å and $c = 3.524$ Å [1]. Synthesis of bulk MgB_2 directly from the elemental powders via the $Mg + 2B \rightarrow MgB_2$ process seems an attractive alternative to the existing methods for processing this material. Since the reaction of MgB_2 formation is accompanied by a large negative volume change (~25%), the final product is expected to be porous if no external pressure is applied.

The synthesis reaction may proceed in an isothermal, diffusion-controlled regime or in a non-isothermal, self-sustained manner. The latter is called SHS, or combustion synthesis [2, 3]. While the advantages of SHS include very high reaction rates and elimination of the need for high temperature furnaces due to self-generation of heat, its major limitation is the high porosity of combustion products. In the present paper, we report on preparation of bulk MgB_2 by SHS in a mode of thermal explosion under Ar pressure in a single-step process. Earlier, such approach was successfully used for the fabrication of MgB_2 with high critical current density in an Ar atmosphere [4].

The aim of the present investigation was to elucidate the influence of the high-pressure synthesis conditions and doping with carbon nanotubes on the superconducting properties of MgB_2 superconductor. We succeeded in the synthesis of doped MgB_2-based materials with a critical current density (J_c) higher than those reported by Kim et al. [5] and Serquis et al. [6]. When the inert gas argon is used in a hot hydrostatic compression method, the melting and boiling points of the magnesium are increased, and it becomes possible to synthesize MgB_2 at higher temperatures than in the sealed vacuum tube synthesis method or the method involving low temperature synthesis in a stream of argon gas [7].

EXPERIMENTAL

The high-pressure setup (see Fig. 1) with a 45 cm^3 working volume was used to produce bulk MgB_2. The main element of the setup is a thick walled metallic spherical reactor vessel without welded parts, with a wall thickness of 60 mm and a capacity of 45 liters. Corps is mounted on a metal frame, made of corner 60 × 60 mm, and provided with an upper and a lower cap which are fastened with nuts on eight studs 35 mm in

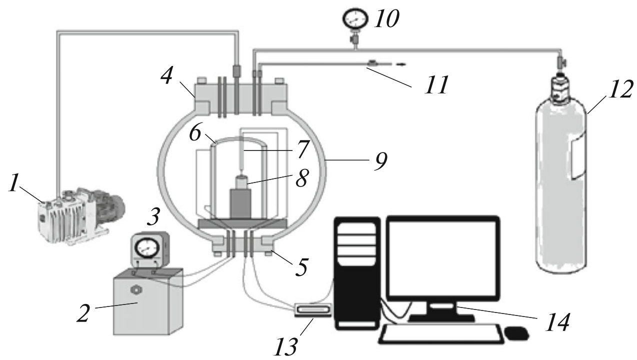

Fig. 1. High-pressure reactor: *1* vacuum pump, *2* transformer, *3* ammeter, *4* upper cover, *5* bottom cover, *6* tubular heating furnace, *7* thermocouple, *8* sample, *9* reactor vessel, *10* pressure gauge, *11* intake and exhaust valves, *12* nitrogen cylinder, *13* data collection system LTR-U-1, and *14* computer.

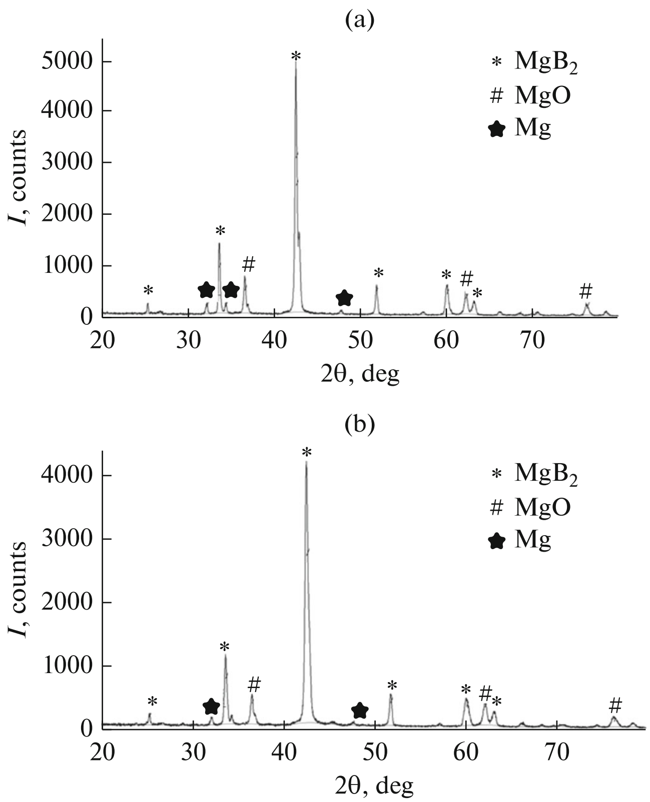

Fig. 2. Diffraction patterns of MgB_2 doped with (a) 1% and (b) 3 wt % MWCNT (P_{Ar} = 2.5 atm).

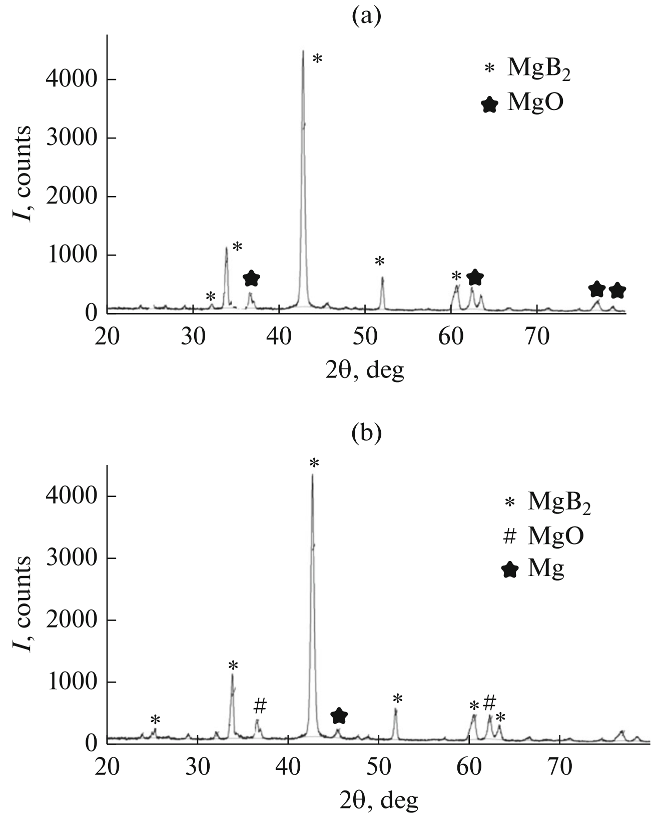

Fig. 3. Diffraction patterns of MgB_2 doped with (a) 5% and (b) 7% MWCNT (P_{Ar} = 2.5 atm).

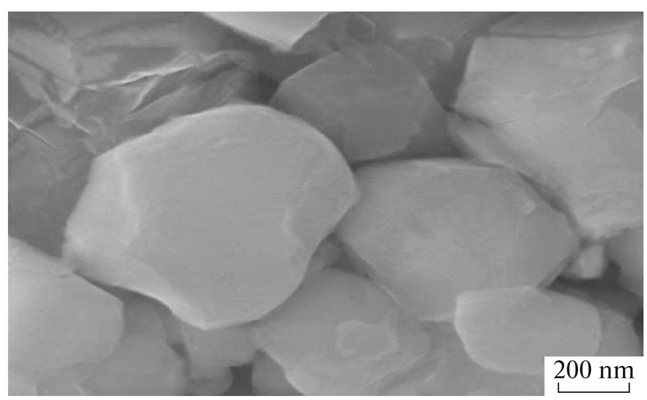

Fig. 4. Typical SEM image of the MgB_2 grains in resultant bulk material (P_{Ar} = 2.5 atm).

diameter. For the thermocouple wires and the power supply, cover current fittings are installed in the bottom. A tubular heating furnace is placed inside the high pressure reactor that allows preheating the sample up to 1000°C. The heater furnace is made of refractory ceramic tube with the diameter of 70 mm and the height of 250 mm. The furnace power was 1.2 kW. The results of temperature measurements were computer-processed.

The superconducting powders of doped magnesium diboride were synthesized from the following powder reactants: metallic magnesium (98.0% purity, 200–250 μm), amorphous boron (94% purity, 1–5 μm) and multi-walled carbon nanotubes (MWCNT, 95%, 10–20 nm). The powders were mixed in the following proportions (wt %):

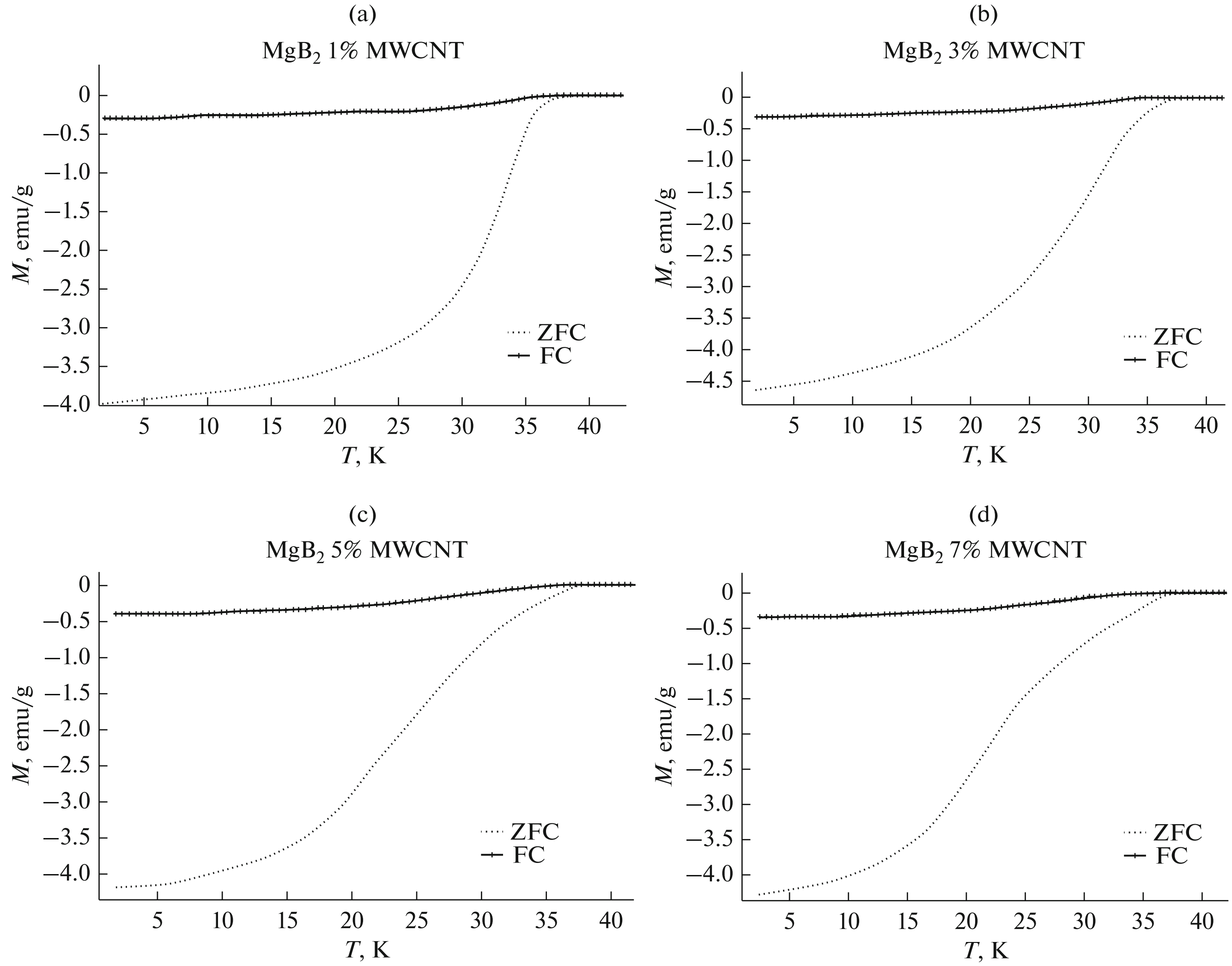

Fig. 5. Temperature dependence of magnetization *M* for MWCNT-doped MgB_2 at H=100 Oe: MWCNT = 1 (a), 3 (b), 5 (c), and 7 wt % (d).

(55.3% Mg + 44.7% B) + 1% MWCNT

(55.3% Mg + 44.7% B) + 3% MWCNT

(55.3% Mg + 44.7% B) + 5% MWCNT

(55.3% Mg + 44.7% B) + 7% MWCNT

Homogeneous powder mixtures were compacted under a pressure of 0.5 GPa to obtain tablets 30 mm in diameter and 15 mm in thickness. High-pressure of Ar gas at 2.5 MPa has been created inside the reactor. The tablets were ignited by heating in furnace *3* (Fig. 1) located in the high pressure reactor. Self-sustaining synthesis was initiated at 650°C. The combustion temperature attained its maximum value at 1100°C. The reaction was completed in 3–5 s.

Composition and crystal structure of the product were characterized by XRD (Cu-K_α radiation, λ = 1.54056 Å) and SEM. The magnetization over a broad temperature range in magnetic fields up to 9 T were carried out with a Quantum Design PPMS EverCool-II system equipped with a Vibrating Sample Magnetometer accessory. The J_c values were estimated from magnetization hysteresis loops by using Bean's formula [8].

RESULTS AND DISCUSSION

Typical diffraction patterns of combustion products are presented in Figs. 2, 3. It follows that the products mainly consist of the MgB_2 phase. All intense peaks can be indexed assuming a hexagonal unit cell with a = 3.086 Å and c = 3.524 Å. In addition to MgB_2, one can also see the presence of some certain amount of MgB_4. Some small impurities were identified as MgO.

Figure 4 shows the typical SEM image of ultrafine MgB_2 grains in resultant bulk material. The average grain size is seen to vary within the range 300–500 nm.

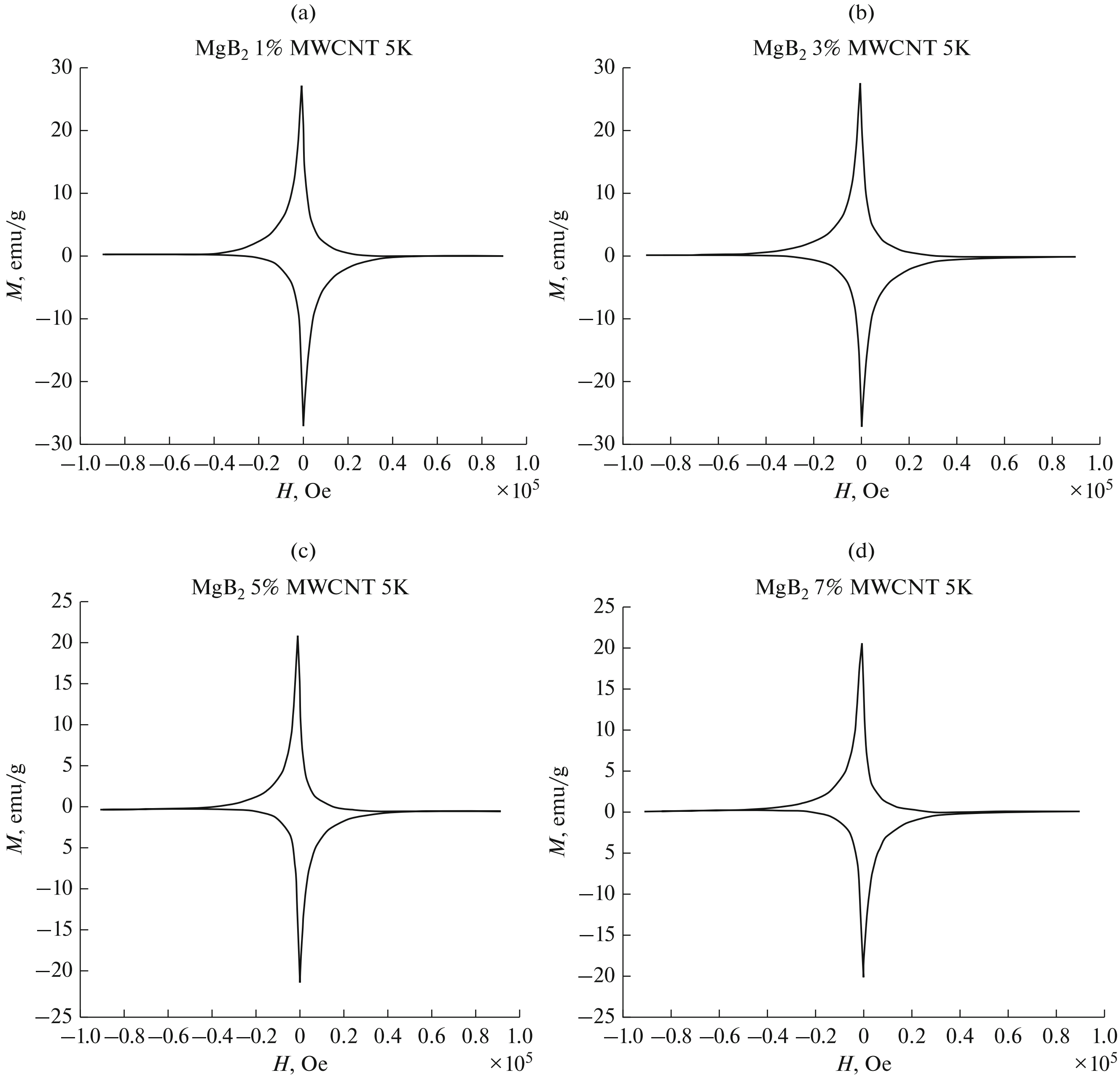

Fig. 6. Hysteresis loops of MWCNT-doped MgB_2 at 5 K: MWCNT = 1 (a), 3 (b), 5 (c) and 7 wt % (d).

Figure 5 shows the temperature dependence of magnetization M in conditions of zero field cooling (ZFC) and field cooling (FC) at H=100 Oe. The general behavior of magnetization for SHS-produced and conventional MgB_2 polycrystalline materials [1] is very similar. Negative signals in the ZFC curve clearly indicate a SC state with an onset of transition temperature at $T_C = 38.5$ K. This is indicative of true realization of superconductivity in the system under consideration. Figure 6 shows typical hysteresis loops for MWCNT-doped MgB_2 superconductors at 5 K. Such a behavior is characteristic of type II superconductors.

We also estimated the value of critical current density, J_c, using the magnetization hysteresis loops and the Bean formula $J_c = 30{*}\Delta M/d$, where J_c is the current carrying capacity expressed in A/cm^2, $\Delta M = M^+ - M^-$ is the difference between magnetization M in increasing and decreasing magnetic fields, and d is the average grain size as derived from SEM images (in our case, $d = 3 \times 10^{-5}$ cm). Using Bean's formula, we cal-

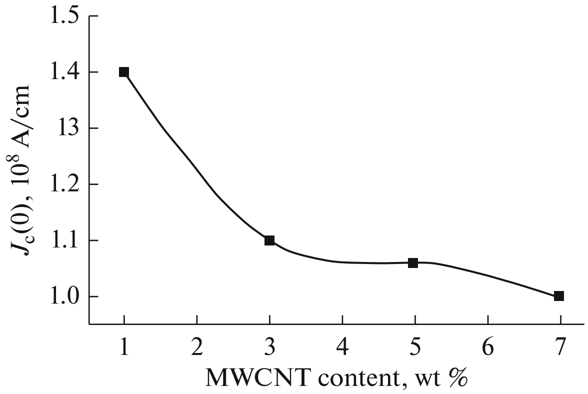

Fig. 7. Critical current density in zero field, $J_c(0)$, at 5 K as a function of MWCNT content.

culated critical current density $J_c(0)$ for MgB_2 containing different amounts of MWCNT in zero field/ The results are collected in Fig. 7. With increasing MWCNT content, $J_c(0)$ is seen to gradually decrease.

CONCLUSIONS

MgB_2 superconductor can be fabricated by thermal explosion in Mg-B blends under Ar pressure. It is shown that only appropriate conditions of SHS reaction (pressure, density) lead to generate a high superconducting property of final product. The superconducting transition takes place at a temperature of 38.5 K, as cited in the literature. Applied Ar pressure and doping with multi-walled carbon nanotubes (MWCNT) produce positive effect on critical current density J_c. A highest value of $J_c = 1.4 \times 10^8$ A/cm^2 was obtained for MgB_2 powders containing 1% MWCNT. The attained level of superconductivity of doped MgB_2 and the possibility of producing large bulk MgB_2 products from SHS-produced powders make this process very attractive for practical implementation.

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