Pinning properties of Ag/MgB$_2$ bulk system

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ABSTRACT

Bulk MgB$_2$ containing nano-particles of Ag–Mg alloys were sintered from the mixtures of Mg-, B- and Ag$_2$O-powder with the mole ratio of Mg:B:Ag$_2$O = 1.6 $(1-x)$:2$(1-x)$:x/2, where x was the nominal concentration of Ag from 0 to 0.1. The solid reaction of the powder mixture was performed in the electric furnace at 900 °C for 30 min under Ar-atmosphere with 10% H$_2$ gas. The X-ray diffraction patterns and images of transmission electron microscope showed the production of Ag–Mg nano-particles in bulk MgB$_2$, except for MgO impurity. For x = 0.01 and 0.02 samples, the enhancement of J_c was observed without T_c reduction, which means the efficiency of the nano-particles of Ag–Mg as the pinning centers in MgB$_2$.

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1. Introduction

Many authors have tried to enhance the pinning force by the doping of several kinds of element in the MgB$_2$ superconductor with the T_c of about 40 K [1]. Especially, the critical current density J_c and the irreversible field H_{irr} have been paid attention for the application of MgB$_2$ at 20 K of liquid hydrogen temperature.

The powder in tube (PIT) method to fabricate the superconducting wire or tape have been developed using Ag sheath [2–7], in which the improved J_c's of 2–3 $\times$ 10^4 A/cm^2 under 1 T at 20 K were observed without T_c-reduction. By scanning electron microscopy (SEM) measurement, the formation of Ag–Mg alloys were confirmed between MgB$_2$ cores and the Ag sheaths [5]. Therefore the Ag–Mg alloy can be the candidate to form strong pinning centers in MgB$_2$. Jin et al. [8] transferred the mixture of Ag powder with 1–10 μm size and MgB$_2$ powder into Fe tubes under Ar atmosphere, and the Fe-clad ribbons of MgB$_2$ including 5 at% Ag were fabricated by sintering at 900 °C for 30 min. Little effect on T_c suggested the Ag is not incorporated into the MgB$_2$ superconductor phase. However the deterioration of J_c was only observed in this case.

The superconductive properties were also studied for nominal Ag$_x$(MgB$_2$)$_{1-x}$ bulk system [9–13], where x is the mixing rate of Ag and MgB$_2$. Sun et al. [9] used the mixtures of commercial MgB$_2$ and Ag-powder in different mole ratios from 100:2.13 to

100:38.3. They were sintered in a vacuumed quartz ampoule at 950 °C for 2 h, and quenched to room temperature. In these samples, Ag reacted with MgB$_2$ to form the impurities of AgMg and MgB$_4$. The T_c remained at about 39 K, but the J_c dropped from that of MgB$_2$ for all of the samples.

Recently, Shimoyama et al. [13] reported that a small amount of Ag addition decreased the reaction temperatures of Mg and B without degradations in T_c and J_c. For example, a sample with about 3 at% and 5 at% Ag showed the J_c of 5 $\times$ 10^4 A/cm^2 at 20 K and 1 T by heating at 500 °C for 72 h. However there is a problem of very long time sintering must be needed in this method.

Here we will present the improvement of pinning property of bulk MgB$_2$ by mixing small amount of Ag and rapid sintering for 30 min. As these results, MgB$_2$ compound, containing AgMg and AgMg$_4$ nano-particles, showed the enhancement of J_c greater than that in pure MgB$_2$ at 20 K and 1 T.

2. Sample preparation

The Ag/MgB$_2$ bulk samples were sintered from the mixture of Mg-, B- and Ag$_2$O-powder with the mole ratio of Mg:B:Ag$_2$O = 1.6$(1-x)$:2$(1-x)$:x/2, where x was 0–0.1. The excess amount of Mg from the stoichiometry was determined empirically to form high quality MgB$_2$ by the present rapid sintering method. Therefore the above mole ratio effectively means the nominal composition of Ag$_x$(MgB$_2$)$_{1-x}$ when Ag$_2$O is reduced to 2Ag. The reduction of Ag$_2$O and the solid phase chemical reaction among the Ag, Mg and B occurred in the electric furnace at 900 °C for 30 min under Ar-atmosphere with 10% H$_2$ gas, being followed by the cooling in a
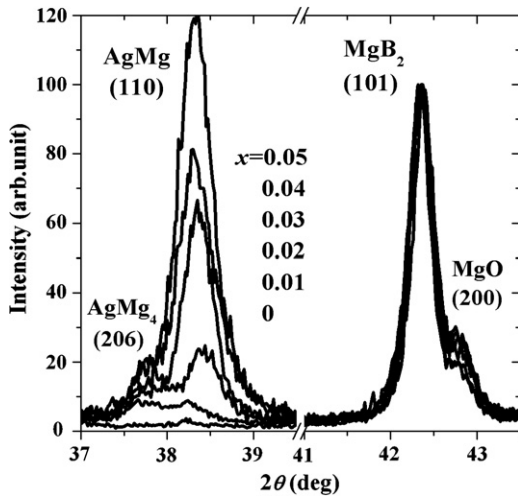
Fig. 1. XRD patterns of nominal compounds of nominal $Ag_x(MgB_2)_{1-x}$ for $x = 0$–0.1.
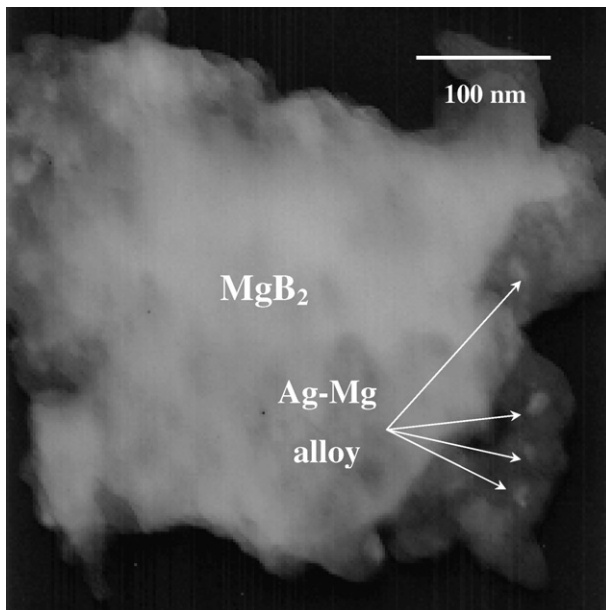
Fig. 2. TEM image of $x = 0.01$ sample. Nano-particles of Ag–Mg alloys were contained in the MgB_2 matrices.

Fig. 3. Temperature dependence of resistivities of $x = 0$–0.1 samples.

Fig. 4. x-Dependence of T_c, estimated from the temperature dependence of resistivity with those of other groups.

furnace. In Fig. 1, the Cu Kα X-ray diffraction (XRD) patterns are shown for the powder samples of $x = 0$–0.05, where the (1 0 1) peak intensities of MgB_2-phase were normalized for each sample. In these samples, the impurity phases of AgMg and $AgMg_4$ of about 10–15 nm size were produced within the bulk MgB_2, except for small amount of MgO. In Fig. 2, the transmission electron microscopy (TEM) image was given for $x = 0.01$ sample, where the 10–20 nm particles of Ag–Mg-alloys (AgMg or $AgMg_4$) were observed in the MgB_2 matrices.

3. Electrical resistivity

Electrical resistivities ρ were measured by DC four terminals method. The temperature dependent resistivities $\rho(T)$ were shown in Fig. 3 for $x = 0$–0.1 samples. The superconducting transition temperatures T_c (mid points) were around 39 K for all of the samples. Little change of T_c was preferable to the pinning property in this system. The x-dependence of T_c, were shown in Fig. 4 with
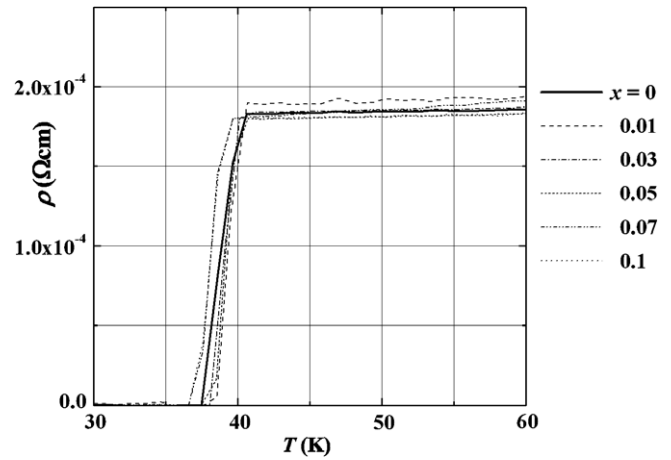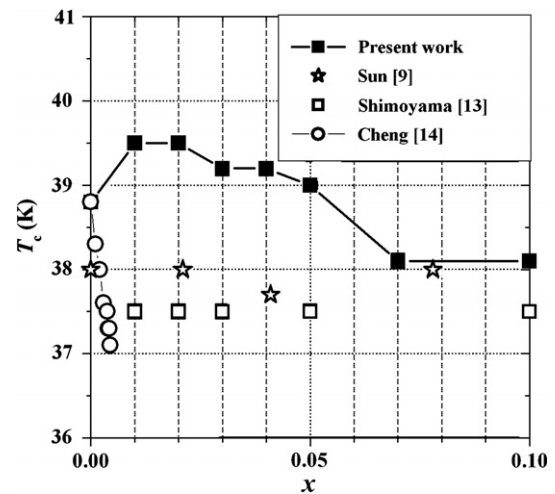
the results of the other groups. The absence of visible T_c reduction in our $x = 0.01$–0.05 samples means the coexistence of pure MgB_2-phase and nano-particles of Ag–Mg alloy, because the large reduction of T_c by the chemical pressure was observed for the solid solution of $Mg_{1-x}Ag_xB_2$ [14–16]. The substitution of Ag atoms into Mg sites in MgB_2 was realized by the sintering at relatively low temperature of 700 °C for 1–15 h in the region of $0 < x < 0.0045$ as shown in Fig. 4 [14,15]. On the other hand, the Ag atoms should be unable to substitute with Mg atoms in MgB_2 by the rapid sintering at high temperature of 900–1000 °C for 0.5–2 h [9–12].

4. Magnetization analysis

The magnetization measurements were performed by the MPMS SQUID (Quantum Design) magnetometer at 5 and 20 K between the field of −5 T and 5 T. The expressions for magnetization by extended critical state model [17–20] were fitted to the experimental data.

The equilibrium critical current density $|J_c^{eq}(H)|$ at inner sample surface was estimated by the following equation

$$|J_c^{eq}(H)| \frac{B_{eq}^*(B_{eq}^* + 2B_0)}{2\mu_0 a\{|B_{eq}(H)| + B_0\}}. \tag{1}$$