Fabrication of extruded wire of MgB\textsubscript{2}/Al composite material and its superconducting property and microstructure

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Abstract. Superconductive MgB\textsubscript{2}/Al composite material with low and high volume fractions of particles were fabricated by our special pre-packing technique and 3-dimensional penetration casting (3DPC) method. The composite material showed homogeneous distribution of MgB\textsubscript{2} particles in the Al-matrix with neither any aggregation of particles nor defects such as cracks or cavities. The critical temperature of superconducting transition ($T_C$) was determined by electrical resistivity and magnetization to be about 37 $\sim$ 39K. A billet of the superconducting composite material was successfully hot-extruded, forming a rod of 10 mm and wires of 3 and 1 mm in diameter. These intermediate products showed the onset $T_C$ of electrical resistivity also at 37-39 K. Microstructures of these samples have been observed by TEM and SEM method to establish morphology and crystallography of particles. The magneto-optic (MO) imaging method has been also applied to measure magnetic flux through this composite material when in external field.

1. Introduction

Studies on MgB\textsubscript{2} have been focused on application for superconducting magnets similarly as Nb-based intermetallic compounds [1,2], and many projects aiming at fabrication of wires and/or sheets are actively being pursued [3]. Superconducting magnets have been, among others, successfully applied to medical magnetic-resonance imaging (MRI) systems, and this market sector is extending. In our previous studies we reported fabrication of composite materials based on Al or age-hardenable Al alloys matrix reinforced by ceramics particles, and investigated their hardening behaviours, microstructures, and aging properties [4]. Advantage of our special technique for fabricating
composite materials is in homogeneous dispersion of particles in the matrix without any aggregation, and in control of their volume fraction within the range of 4 – 40 %, even with a particle size below 1 \( \mu m \).

In the present work, we applied our technique to fabrication of billets of the composite materials consisting of Al matrix reinforced by MgB\(_2\) particles, which were then successfully extruded into 10, 3 and 1 mm rod and wires. Their microstructures and superconductive and thermal properties were also evaluated.

2. Experimental
MgB\(_2\) powders were provided by Kojundo Chemical Laboratory Co., Ltd., at purity higher than 99% and with size smaller than 40 \( \mu m \). Received powders were gently ground in an agate mortar to break any aggregation. The procedure for forming a composite material billet was described in our recent report in detail [5]. When using this method, the molten Al can penetrate into the pre-form from all directions so we refer to this method as to the 3-dimensional penetration casting (3DPC) method. After cooling, the billet is removed from the steel mold by cutting. The volume fraction \( V_f \) can be controlled within 10 – 50%. In billets used in this study, \( V_f \) of MgB\(_2\) powders was about 40 % - such billet will be referred to as a high \( V_f \) sample. In order to evaluate the effect of varying \( V_f \), a low \( V_f \) sample was also fabricated by means of the melt spray method [4] with \( V_f \) of about 10 %. Also this billet was extruded by a hot-extruding machine of 50 t or 400 t to a rod 10 mm in diameter, and to 3 and 1 mm wires.

Superconducting and thermal properties were measured by means of the Physical Property Measurement system (PPMS, Quantum Design, Co., Ltd.). Samples for these measurements were cut from the composite material billets in the form of 1 mm cubes. Electrical resistivity was measured by a DC 4-terminal method at a direct current of 1.0 mA. Magnetization was measured by SQUID (Quantum Design, Co., Ltd.) using an applied magnetic field of 100 G. The microstructures of composite materials were observed by a scanning electron microscope (SEM), scanning low energy electron microscope (SLEEM), and transmission electron microscope (TEM) in order to establish arrangement and size of particles, chemical composition and crystallography. The magneto-optic (MO) imaging method [6] was applied to investigate the magnetic flux behaviour of this composite material in the applied field.

3. Results and discussion
Fig. 1(a) shows macrostructure of the high \( V_f \) billet of the MgB\(_2\)/Al composite material. The billet was cut along its vertical direction. No remarkable shrinkages, cracks, large aggregations of powders or any other defects are observed. Gray and bright contrasts appearing in this figure correspond to a reinforced region and to pure Al without particles, respectively. The Al region was observed at the bottom side of the steel mold, indicating the molten Al sufficiently penetrating through the preform of MgB\(_2\) to the bottom, from where it could be turned back to the preform by applied pressure. The region indicated by the rectangle in Fig. 1(a) was observed by SEM - see Fig. 1(b). Particles showed homogeneous distribution and no cracks between particles and the Al matrix are observed at this magnification.

Fig. 2(a) shows appearance of the extruded 10 mm rod and in Fig. 2(b) we see macrostructure of this rod cut along its longitudinal direction. The top end of the rod (region A) is obviously of pure Al while the region B of 50 mm in length is of the composite. Fig. 3 shows appearance and microstructure of the extruded 3 mm dia. wire from the high \( V_f \) MgB\(_2\)/Al composite material. Figs 2 and 3 show no remarkable defects on cross sections cut parallel to the extruded direction. SEM-EDS analysis and XRD measurement were also applied to the billet, rod and wire in order to reveal impurities or reaction products like Mg-Al or Mg oxides but no defects have been detected.
Figure 1. Macro- and micro-structure of the MgB$_2$/Al composite material billet: (a) Macroscopic image of the high-$f_v$ MgB$_2$/Al composite material, and (b) SEM image of a region indicated by the square in (a).

Figure 2. Macrostructure of the extruded 10 mm dia. rod from the high-$f_v$ MgB$_2$/Al composite material: (a) appearance of the extruded sample, and (b) its cross section cut parallel to the extruded direction.
Figure 3. Macrostructure of the extruded wire 3 mm in diameter from the high-$V_f$ MgB$_2$/Al composite material: (a) appearance of the extruded sample, and (b) SEM image of its cross section.

The measured temperature dependence of electrical resistivity for the 3 mm wires of the high- and low-$V_f$ MgB$_2$/Al composite materials is shown in Fig. 4. Behavior of decreasing resistivity is similar to results obtained for the billet and 10 mm rod [5]. Namely the resistivity of the 3 mm high-$V_f$ wire dropped down at 39 -37 K (onset $T_c$) to 0 Ωm.

Figure 4. Temperature dependence of electrical resistivity of the extruded 3 mm high- and low-$V_f$ MgB$_2$/Al composite material wires and of the pure aluminum.
Figure 5. MO images of the extruded, 3 mm in diameter, high-$V_f$ MgB$_2$/Al composite material wire at $T = 4$ K at increasing magnetic field: a) $B_a = 8.5$ mT, b) $B_a = 79$ mT. The side of the square sample is 2.5 mm long.

Magnetic shielding of the 3 mm high-$V_f$ wire at 4 K was examined by the MO imaging method - see Fig. 5. Here the image brightness represents the local value of magnetic field. At low applied fields (Fig. 5(a)) the sample interior appears dark, which indicates the magnetic flux not allowed to penetrate. Thus, the sample bulk is well shielded by induced supercurrents flowing in the volume as a result of contacts between the MgB$_2$ grains. At higher fields (Fig. 5(b)) magnetic flux penetrates the inter-granular space, creating dark diagonal lines which are characteristic for partial shielding of a square shaped superconductor. At the same time, the numerous dark “particles” remain completely shielded by currents induced inside each grain. Thus, in this composite material the inter-grain current density is considerable, although smaller than the intra-grain current.

Fig. 6 shows the extruded 1 mm in diameter, high-$V_f$ MgB$_2$/Al composite material wire. Its surface was smooth without any cracks, however, behavior of the electric resistivity and existence of $T_c$ was not so clear now because of inhomogeneous distribution of powders in this thin wire. For thin wires further technology improvements are obviously needed.

Figure 6. Appearance of the extruded 1 mm, high-$V_f$ MgB$_2$/Al composite material wire.

4. Conclusion
1. MgB$_2$-dispersed aluminum matrix composite materials containing high and low volume fractions of MgB$_2$ powders were successfully fabricated. No remarkable defects were observed in these composite materials. SEM-EDS and XRD methods did not detect any reaction products like Mg-Al or Mg oxides.

2. Extruded rod 10 mm in diameter and wires 3 and 1 mm in diameter were successfully obtained without remarkable defects from the pre-fabricated billet. 10 mm rod and 3 mm wire of high $V_f$ showed remarkable onset of electrical resistivity at $T_C$ of about 37 - 39 K, similarly as the high-$V_f$ billet.

3. MO imaging detected magnetic shielding in the 3 mm wires at 4 K in an applied field of 8.5 mT.

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