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Microstructure and critical current density in MgB_2 bulk made of 4.5 wt% carbon-coated boron

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Abstract. Superconducting performance and its uniformity was studied in the single-step sintered MgB_2 bulk prepared with 4.5 wt% of carbon in the carbon-encapsulated boron. The 20 mm in diameter MgB_2 pellet was cut into several pieces from bottom to top and the microstructure, superconducting transition temperature (T_c onset), and critical current density at 20 K were studied. DC magnetization measurements showed a sharp superconducting transition with onset T_c at around 35.5 K in all positions. SEM analysis indicated a dispersion of grains between 200 and 300 nm in size, as the main pinning medium in this MgB_2 superconductors. The critical current density at 20 K was quite uniform, around 330 kA/cm^2 and 200 kA/cm^2 at self-field and 1 T, respectively, for all measured positions. The results indicate that the *carbon-encapsulated* boron is very promising for production of high quality bulk MgB_2 material for various industrial applications.

1. Introduction

Practical applications of bulk MgB_2 magnets are similar to those of melt-textured Y-123, namely magnets for nuclear magnetic resonance (NMR), magnetic resonance imaging (MRI), fault current limiters, non-contact bearings for liquid pumping, and for magnetic shielding screens [1-3]. To fully utilize the bulk MgB_2 material, especially for superconducting super-magnets, the optimization of the flux pinning and further improvement in critical current density (J_c) are very important. To improve the critical current density of bulk MgB_2 material a variety of processing techniques has been developed [4-8]. As for all functional materials, the microstructure control is crucial. In MgB_2 it concerns mainly the grain size, governing the low field critical current density [9]. In our recent report we showed that J_c and the trapped magnetic field in bulk MgB_2 materials were dramatically improved when the material was processed at around 800 ºC for an optimized sintering time [9]. Under optimized conditions uniformly distributed nanometer sized grains formed and a coarsening of the MgB_2 grains was prevented, resulting in an enhanced grain boundary pinning [10]. The critical current density was dramatically improved, especially with doping by metals like Ag, Bi, Sn, Si, Te, Cu etc. [11-14]. On the other hand, the high-field critical current performance was enhanced utilizing carbon-based dopants [15-19]. These dopants improve the high-field flux pinning by substituting carbon into boron lattice sites [20]. The resulting point-like arbitrary pinning disorder improves superconducting properties such as J_c, H_{irr}, and H_{c2} [20]. A recent report clarified that the optimized sintering temperature combined with carbon-encapsulated boron powders dramatically improved flux pinning and critical current density in bulk MgB_2 materials [19]. As a result, the highest J_c values at 20 K, of 375 kA/cm^2 and 220 kA/cm^2, in the self-field and 1 T, respectively, were achieved in the MgB_2 sample with 2.8% of carbon-encapsulated boron.

These results are very promising. However, for trapped-field applications of bulk MgB_2 materials it is very important to achieve such properties uniformly in the whole superconductor pellet. In the presented study we synthesized a bulk MgB_2 material utilizing the carbon-encapsulated boron powder with 4.5 wt% carbon and checked the effect of carbon distribution over a large bulk MgB_2 material on the superconducting performance. The original pellet was cut into several pieces and x-ray diffraction, superconducting transition temperature, and critical current density at 20K were studied on the small samples from different positions.

2. Experimental details

Commercial powders (Furu-uchi Chemical Corporation, Japan) of Mg (99.9% purity, 200 meshes) and carbon-encapsulated amorphous B powder with 4.5% carbon (98% purity, 300 meshes) were mixed in the nominal ratio of Mg: B = 1: 2. The carbon-encapsulated B powders were produced at PAVEZYUM, Advanced Chemicals, Turkey. More details of the carbon-encapsulated B powder production can be found in Ref. [21]. The starting powders were thoroughly ground in a glove box in argon atmosphere. The powder mixture was pressed into pellets of 20 mm in diameter and 7 mm thick using a uniaxial press of 200 MPa. The consolidated pellets were then wrapped in titanium foils and subjected to the heat treatment in Ar atmosphere in a tube furnace. All samples were heated to the target sintering temperature of 805 °C and kept there for 3 h in flowing argon gas. Finally, the temperature was lowered to room temperature at a cooling rate of 100 °C/h. The constituent phases of the samples were identified with a high-resolution automated X-ray powder diffractometer (RINT2200), using Cu-Kα radiation generated at 40 kV and 40 mA. The microstructure of these samples was studied with a scanning electron microscope (SEM).

Small specimens with dimensions of $1.5\times1.5\times0.5$ mm^3 were cut from MgB_2 pellets and subjected to the measurements of critical temperature (T_c) and magnetization hysteresis loops (*M-H* loops) in applied magnetic fields from –1 to +5 T at 20 K using a SQUID magnetometer (Quantum Design, model MPMS5). The magnetic J_c values (in MA/cm^2) were estimated based on the extended Bean critical state model using the relation

$$J_c = 20\,\Delta m/[a^2 d(b-a/3)] \quad (1)$$

where d is the sample thickness, a, b are cross sectional dimensions, $b \geq a$, (a, b, d in mm) and Δm (in emu units, 1 emu≡10^{-3} Am^2) is the difference of magnetic moments during increasing and decreasing field in the *M-H* loop [22].

3. Results and discussion

To study the bulk MgB_2 performance at various positions, small samples were cut from the pellet as shown in Fig. 1 (left). 3 pieces were selected in top and 3 pieces in bottom of the pellet (see Fig. 1, left). Fig. 1 (right) shows the X-ray diffraction pattern for the sample from the top centre. It can be seen that the sample mainly consists of the MgB_2 phase and a small quantity of the MgO phase. The latter phase was found to arise during the samples pressing into pellets after the samples being wrapped into tantalum foils before loading them into the tube furnace [23]. Several pieces selected from various positions on the pellet exhibited the same structure, with the main phase of MgB_2 and a small trace of MgO. It follows from the XRD results that the 4.5 wt% of carbon in the carbon-encapsulated boron does not destroy structure of the bulk MgB_2 material. To support this statement, we measured transition temperature and critical current density at 20 K on the samples selected at top and bottom of the pellet. The results are presented in following section.

Temperature dependence of magnetization of the samples selected from top (T1, T2, T3) and bottom (B1, B2, B3) of the pellet is presented in Fig.2. All samples exhibited a sharp superconducting transition with the onset T_c around 35K. The T_c (onset) values for the T1, T2, T3 positions were 35.5 K, 35.8 K, and 36 K, respectively (Fig.2., left). T_c of 35 K, 35.5 K, and 36.1 K was observed at B1, B2, and B3 positions, respectively (Fig.2., right). Compared to a pure MgB_2 material, there is only a slight

depression in T_c (onset) in the present samples, in accord with our earlier reports [10]. The results indicate that a uniform high quality MgB_2 material can be produced utilizing the carbon-encapsulated B powder with 4.5 wt% of carbon.

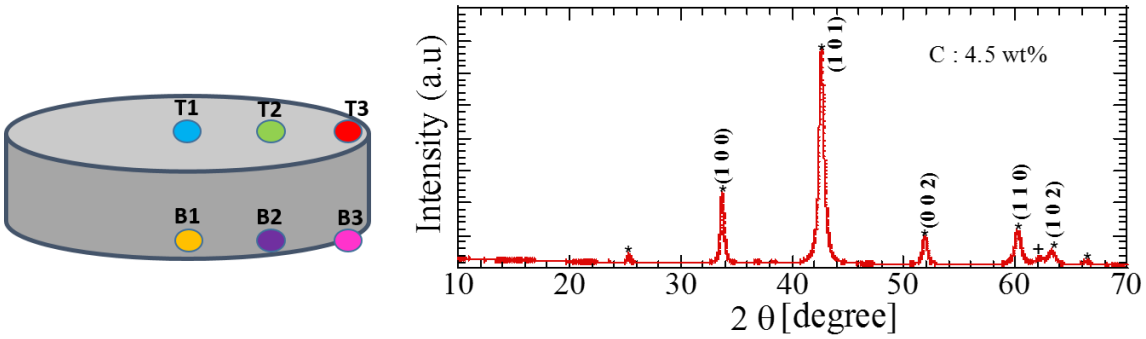

Figure 1. Scheme of the bulk MgB_2 pellet showing the sample positions selected for magnetization measurements (left). X-ray diffraction pattern of the sample from the top centre (right, *MgB_2 phase and + MgO phase).

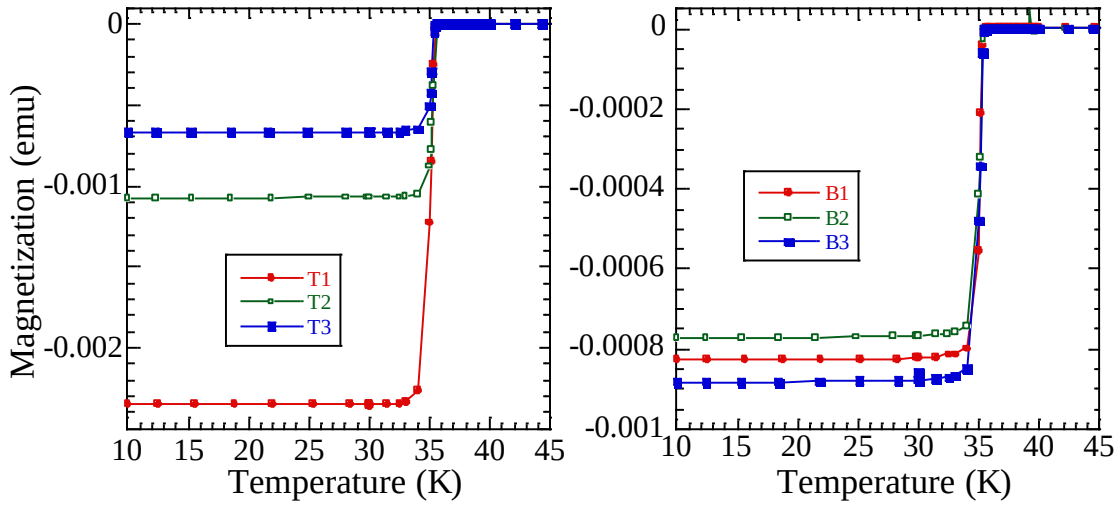

Figure 2. Superconducting transitions in the samples selected from top (left) and bottom (right) of the MgB_2 pellet. The MgB_2 was produced using carbon-encapsulated B powder with 4.5 wt% carbon and sintered at 805 ^oC for 3h in argon.

To see the effect of carbon distribution on critical current density, the magnetic J_c values were determined from magnetization hysteresis loops (*M-H* loops) detected by a SQUID magnetometer in applied magnetic fields from –1 to +5 T at 20 K. Figure 3 shows J_c values at 20 K as a function of applied magnetic field for the samples selected from the top (Fig. 3., left) and bottom (Fig. 3., right) of the MgB_2 pellet. All the curves show almost same critical current density values. The high-field J_c values and the irreversibility field were same in all positions. The self-field critical current density between 300 kA/cm^2 and 330 kA/cm^2 was observed in all the samples. At 1 T and 2 T the critical current density reached the values of around 200 kA/cm^2 and 75 kA/cm^2, respectively. These results again indicate that the performance of the bulk MgB_2 material is nearly same in various positions of the large bulk. It is a good message for mass production of the MgB_2 super-magnets.

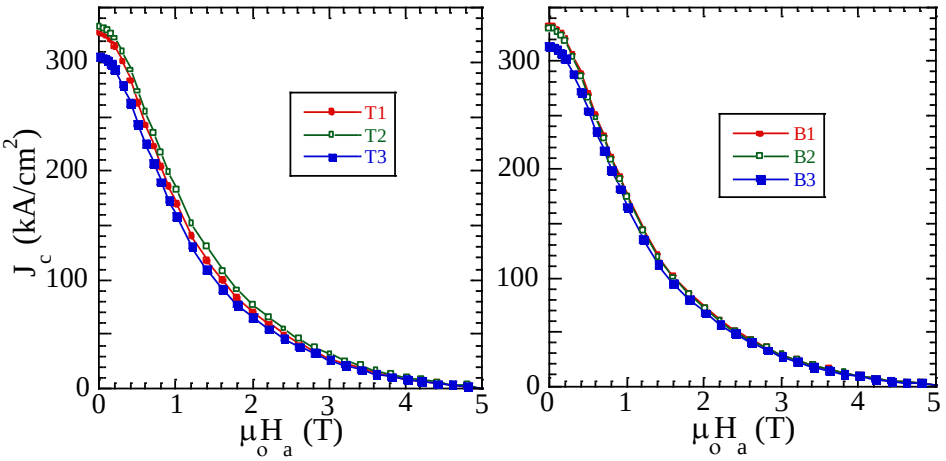

Figure 3. Field dependence of critical current density (T = 20 K) of the MgB_2 samples from different positions in top (left) and bottom (right) of the pellet.

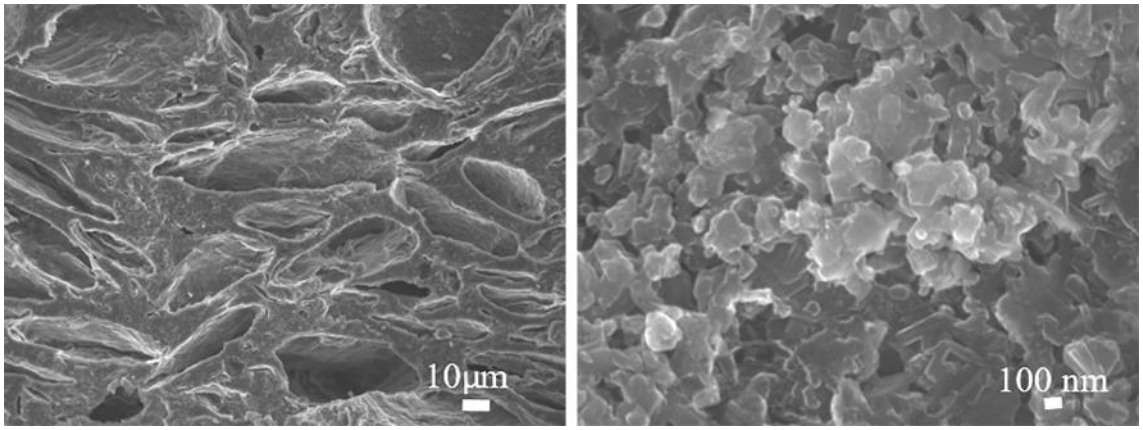

Figure 4. High and low magnification SEM images of the bulk MgB_2 sample produced utilizing the carbon-encapsulated B powder with 4.5 wt% carbon and sintered at 805 °C for 3h in argon atmosphere.

The microstructural features in the MgB_2 material were tested by scanning electron microscopy (SEM) as shown in Fig. 4. The SEM image taken at a lower magnification shows that the MgB_2 matrix contains numerous voids of various shapes and sizes (left). The porosity presents a serious problem in MgB_2 materials. To overcome this problem, numerous measures have been tested, like sintering time optimization, silver addition, use of carbon-based dopants etc. On the other hand, it is evident from the SEM micrograph (Fig. 4., right) that the size of the MgB_2 grains is very small, in nanometer size. One can see a rather dense structure of 200 to 300 nm grains on the high magnification image that are most probably responsible for the high J_c in this material. The smaller the grains, the more effectively they can act as strong flux pinning centres. These results again imply that high pinning performance of the sintered MgB_2 material can be enhanced by means of carbon-encapsulated boron along with optimum sintering temperature.

4. Conclusions

In the present work, a high quality superconducting bulk MgB_2 was produced utilizing the carbon encapsulated B powder with 4.5 wt% carbon and sintered at 805 °C for 3h in argon atmosphere. The effect of carbon encapsulated B powder distribution in the MgB_2 bulk on electromagnetic and superconducting properties was studied. Only a negligible variation was observed in the

superconducting transition temperature measured in various spots at top and bottom parts of the pellet. The critical current density and irreversibility field values were found almost same at top and bottom of the large bulk MgB_2. The best J_c value at 20 K was around 330 kA/cm^2 in self-field and 200 kA/cm^2 at 1 T. The observed material homogeneity is by far better than in cuprate melt-textured bulks [24]. These results suggest that carbon coating of the boron used by fabrication of bulk MgB_2 is very promising for production of superconducting super-magnets for several industrial applications.

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