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Trapped magnetic field and vortex pinning properties of MgB_2 superconducting bulk fabricated by a capsule method

Tomoyuki Naito[1,2], Tomohisa Sasaki[1] and Hiroyuki Fujishiro[1,2]

[1] Department of Materials Science and Engineering, Faculty of Engineering, Iwate University, Morioka 020-8551, Japan
[2] Soft-Path Engineering Research Centre (SPERC), Faculty of Engineering, Iwate University, Morioka 020-8551, Japan

E-mail: tnaito@iwate-u.ac.jp

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Abstract
We have studied the trapped magnetic field and the critical current density for an MgB_2 superconducting bulk disc fabricated by a capsule method. The maximum trapped magnetic fields at the centre of bulk surfaces 20 and 30 mm in diameter were, respectively, 1.43 T at 13.4 K and 1.50 T at 16.4 K. The critical current densities, J_cs, were estimated to be 5.5×10^4 and 4.2×10^4 A cm^{-2} at 20 K for the 20 and 30 mm diameter bulks, respectively, from the measured trapped field at the top surface, by using the analytical equation. The estimated J_c was as small as about 30–60% of the measured J_c in the self-field which was determined from the magnetization hysteresis loop of small pieces cut from the bulk. The results suggest that an MgB_2 bulk magnet over 3 T can be realized at 20 K by means of the enhancement of both the J_c values and the mass density.

(Some figures may appear in colour only in the online journal)

1. Introduction

A type II superconductor can trap quantized vortices and realizes a strong quasi-permanent magnet after the magnetizing process. Bean's critical state model [1] suggests that the trapped field, B_T, depends linearly on both the diameter and the critical current density, J_c, of the superconducting bulk disc. Therefore a larger superconducting bulk with higher J_c is preferable.

Single-domain RE–Ba–Cu–O (REBCO; RE: rare earth element) superconducting bulks grown by a melt process [2–4] have been intensively studied with a view to achieving higher trapped field, B_T, because of their high critical temperature, T_c, above 90 K, their high J_c and the moderate J_c deterioration under magnetic fields. The introduction of fine RE_2BaCuO_5 (RE211) particles and substitutional elements, and control of the crystallographic orientation enable us to enhance J_c. The values of J_c and B_T for REBCO bulks have already exceeded 10^5 A cm^{-2} and 3 T, respectively, at 77 K in the self-field [5]. The latter value was one order of magnitude higher than that of a Nd–Fe–B permanent magnet. Furthermore, a YBCO bulk (26.5 mm in diameter and 15 mm in thickness), which was reinforced with an epoxy resin in order to overcome the high yield stress, recorded the highest trapped field of 17 T at 29 K [6]. REBCO bulks 45–60 mm in diameter are commercially available, can be used to produce tesla-order quasi-permanent magnets, and have already been used in prototypes for novel applications such as drug delivery systems [7], sputtering cathodes [8] and magnetic separation [9]. Although a large single-domain bulk over 100 mm in diameter was difficult to fabricate, a large bulk has been realized by a multi-seeding technique [10] and in a microgravity environment in space [11].

MgB_2 superconductors have mainly been studied for practical application in superconducting tapes and thin films owing to their high T_c of about 40 K and their having a high upper critical field among metallic superconductors. The typical J_c value of the polycrystalline MgB_2 samples is over 10^5 A cm^{-2} below 20 K in the self-field [12, 13], which is as high as that of the single-domain REBCO bulks at 77 K [14]. Furthermore, because of their long coherence length, ξ, the problem of weak links at the grain boundaries can be ignored even for the polycrystalline samples [15]. These results enable us to produce better and larger polycrystalline MgB_2 bulk magnets below 20 K. The most important feature of MgB_2 is light weight, compared to REBCO; that is, the ideal mass densities of MgB_2 and YBCO are 2.62 and 6.38 g cm^{-3}, respectively. The large polycrystalline MgB_2 bulk has an advantage as regards being cheap and suited to mass production. Since the operating temperature of 20 K is easily obtained by using liquid H_2 or cryocooler cooling, MgB_2 bulk magnets are expected to be applicable in magnetically levitated (MAGLEV) trains and so on.

Several groups have already reported the trapped field on the MgB_2 bulk [16–18]. The B_T value of about 2.3 T at 6 K was obtained for the MgB_2 bulk (28 mm in diameter and 11 mm in thickness) which was sintered under a high pressure of 2 GPa [16]. Recently, Yamamoto *et al* reported a 1.2 T MgB_2 magnet (20 mm in diameter and 5 mm in thickness) at 17 K [17]. Dense MgB_2 bulks fabricated by the reactive Mg liquid infiltration (Mg-RLI) technique have also been reported [19], on which the maximum B_T was about 1.3 T at 15 K after field cooling under a magnetic field of 1.4 T for an MgB_2 disc (55 mm in diameter and 15 mm in thickness) [18]. The reported B_T values for MgB_2 bulks are smaller than those of REBCO bulks at the present stage. However, they are promising materials for producing a strong and large superconducting bulk magnet, because the B_T value must be enhanced by the improvement of the filling factor and the connectivity between the grains [20] and by the enlargement of the diameter. MgB_2 has usually been fabricated in a vacuum-sealed quartz or metal tube to prevent the oxidization and/or nitridization of Mg and B; however this method is not suitable for large bulk, several centimetres in diameter.

For the fabrication of large polycrystalline MgB_2 bulk, we proposed a simple and convenient capsule method [21], which does not require special techniques such as sealing-off or welding and gives us a kind of hot-press annealing effect by suppressing the expansion of the precursor pellet. We demonstrated that a 1.65 T MgB_2 bulk magnet 30 mm in diameter was successfully produced at 14 K by this method; however the relationship between the trapped field and the vortex pinning properties has not been examined in the previous report [21]. In this paper, we measure the trapped field, B_T, for large MgB_2 bulks 20–30 mm in diameter and the critical current density, J_c, of small pieces cut from the bulk, and discuss their correlation.

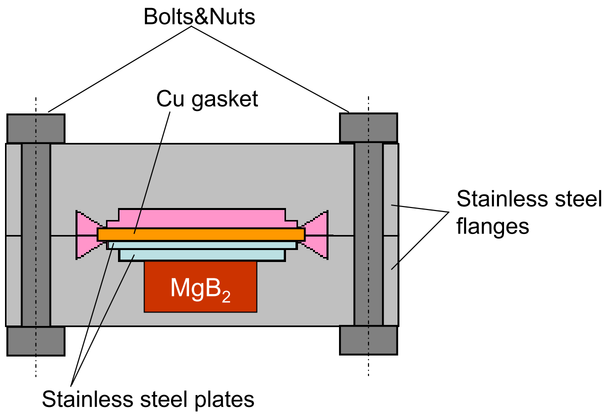

Figure 1. Schematic illustration of the closed capsule.

2. Experimental details

2.1. The process of fabrication of MgB_2 bulks

MgB_2 bulks were fabricated by the *in situ* capsule method [21]. A schematic image of the capsule is shown in figure 1. The capsule consists of two commercial stainless steel flanges (70 mm in diameter and 10 mm in thickness; ICF flange), two stainless steel plates and a copper gasket. Raw powders of Mg (99% purity, $\leq$180 μm grain size, Kojundo Chemical Laboratory Co., Ltd) and amorphous B (99% purity, 300 mesh, Furuuchi Chemical Corp.) were weighted at 1.1:2.0 in molar ratio and ground using an agate mortar and pestle for 1 h. The mixed powder was pressed into pellets 20–30 mm in diameter and 5–9 mm in thickness under uniaxial pressure of about 12 MPa. The precursor pellet was set in the hole of the stainless steel flange, covered by the stainless steel plates, and finally closed in by means of the other flange with the copper gasket, using the bolts and nuts. The stainless steel plates were inserted to prevent reaction of Mg with Cu. The precursor pellet was prepared in air and the capsule was sealed in an Ar atmosphere using a glove box. The MgB_2 precursor in the closed capsule was sintered at 800 °C for 6 h in a box furnace and cooled down to room temperature by furnace cooling. The MgB_2 bulks 20 and 30 mm in diameter were named MB-20 and MB-30, respectively. Sample specifications are listed in table 1. Figure 2 shows a photograph of the MB-20 bulk with the stainless steel flange after the sintering.

2.2. Measurements

The MgB_2 bulk was magnetized by field cooling (FC) in a magnetic field of 5 T parallel to the thickness direction using a 10 T cryogen-free superconducting magnet (JMTD-10T100, Japan Superconductor Technology (JASTEC), Inc.) and then the applied magnetic field was decreased to 0 T at a rate of -0.22 T min^{-1}. The bulk was set on the cold-head of a Gifford–McMahon type helium refrigerator by inserting a thin In plate to achieve good thermal contact. The trapped field was measured by using a cryogenic Hall sensor (BHT-921, FW Bell Inc.) mounted on the centre of the bulk surface using

Table 1. Specifications of the MgB_2 bulks. T_c is the critical temperature, ΔT_c is the width of the transition and B_T is the trapped field.

Sample name	Diameter (mm)	Thickness (mm)	T_c (K)	ΔT_c (K)	Max. of B_T
MB-20	20.2	5	38.2	0.9	1.43 T at 13.4 K
MB-30	30.4	9	38.4	1.0	1.50 T at 16.4 K

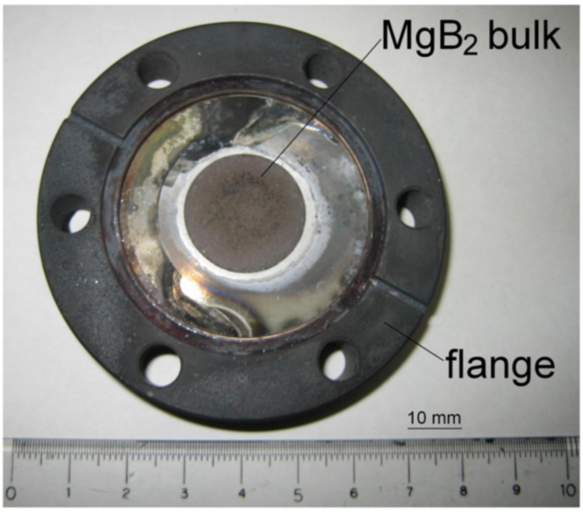

Figure 2. Photograph of the MB-20 MgB_2 bulk (centre circle) with the stainless steel flange.

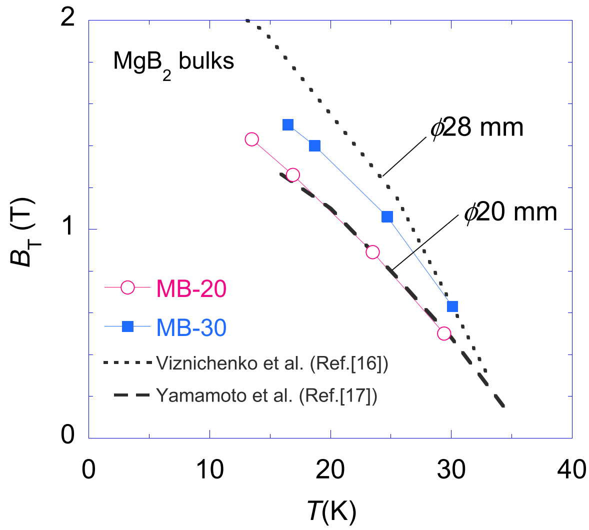

Figure 3. Temperature dependence of the trapped magnetic field, $B_T(T)$, for the MgB_2 bulks. The reported $B_T(T)$ lines are also presented.

GE7031 varnish. During FC, the temperature of the bulk was controlled by a Cernox thermometer which adhered beside the Hall sensor on the bulk surface. After the FC magnetization experiments, the bulks were cut into several small pieces, and we evaluated the superconducting properties such as the critical temperature, T_c, and the critical current density, J_c, by measuring the magnetization, M, using a commercial SQUID magnetometer (MPMS-XL, Quantum Design Inc.). J_c was estimated from the hysteresis loop using the extended Bean model, $J_c = 20\Delta M/a(1 - a/3b)$, where ΔM is the width of the hysteresis loop, and a and b ($a < b$) are the cross-sectional dimensions of the sample perpendicular to the applied magnetic field.

3. Results and discussion

Figure 3 shows the temperature dependence of the trapped field, $B_T(T)$, for two MgB_2 bulks. The $B_T(T)$ lines reported previously [16, 17] are also shown for comparison. For both bulks, $B_T(T)$ increases with decreasing temperature; this effect originates from the enhancement of J_c at low temperatures. The highest $B_T(T)$ values for MB-20 and MB-30 bulks, respectively, are 1.43 T at 13.4 K and 1.50 T at 16.4 K. The magnitude of B_T for the MB-30 bulk is approximately 1.2–1.3 times larger than that for the MB-20 bulk at the same temperature, which is 13–20% smaller than the 1.5 expected from the ratio of the diameters; B_T should be proportional to the diameter in terms of Bean's critical state model if J_c is identical in both bulks. The results suggest that the J_c value for the MB-30 bulk is smaller than that for the MB-20 bulk. The $B_T(T)$ value for the MB-20 bulk almost corresponds to the one reported (20 mm in diameter and 5 mm in thickness) [17], but those for the MB-30 bulk are obviously smaller than those for the MgB_2 bulk reported on, which is 28 mm in diameter [16]. This might arise from the fact that the density (approximately 1.3–1.4 g cm^{-3}) of the MB-30 bulk is lower than that of the bulk in [16] fabricated under high pressure. Higher B_T could be realized by increase of the density of the bulk.

The superconducting properties of the MgB_2 bulks were evaluated using small pieces cut from the bulk after the measurements of the trapped field. Figure 4 shows the powder x-ray diffraction patterns taken at the room temperature using Cu Kα radiation. The observed peaks were mainly indexed to the MgB_2 phase, and small amounts of impurity phases such as Mg, MgO and $MgCu_2$ were also observed. The small peak at $2\theta = 37°$ for the MB-20 bulk represents the (101) plane of Mg, originating from the unreacting Mg. The peak at $2\theta = 63°$ for both bulks represents the (111) plane of MgO, which might be created in the mixture grinding or the sintering process. The peaks at $2\theta = 22°$ and 44° found only for the MB-30 bulk arise, respectively, from the (111) and (222) planes of $MgCu_2$, resulting from the reaction of liquid Mg and the Cu gasket. The reason for the $MgCu_2$ phase appearing only in the MB-30 bulk is that the distance between the precursor pellet and the Cu gasket for the MB-30 bulk is less than that for the MB-20 bulk, i.e., the liquid Mg easily reacts with Cu.

Figure 5 presents the temperature dependence of the normalized magnetization, $M(T)$, in a magnetic field of 0.4 mT after zero-field cooling. Six pieces were cut from

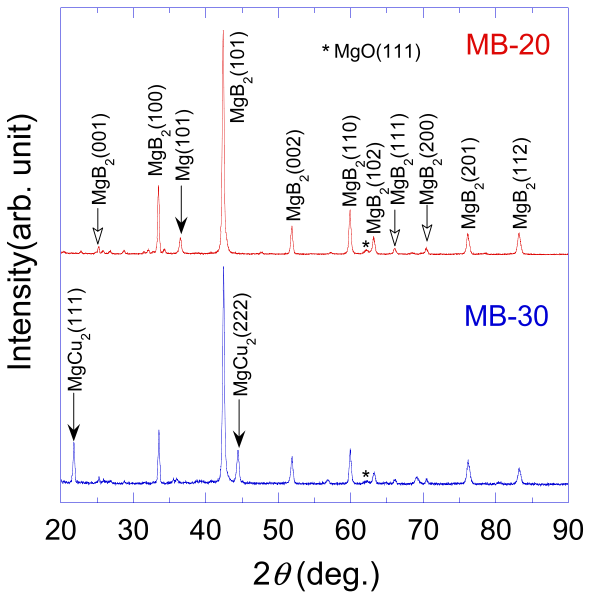

Figure 4. X-ray diffraction patterns taken at room temperature for the MgB_2 bulks.

the centre and the edge of each bulk; the positions are schematically shown in the figure. We named the samples from higher, at and below the bulk centre C1, C2 and C3, respectively. The edge samples, E1, E2 and E3, are also named in a similar manner. The sample dimensions were typically 2 × 2 × 2 mm^3. All the small samples show the sharp superconducting transition, although the C1 and E1 samples of the MB-20 bulk show a broad tail. The critical temperatures, T_cs, defined at the mid-point of the transition are about 38.2 K for the MB-20 bulk and 38.4 K for the MB-30 bulk. The widths of the transition, ΔT_cs, are about 0.9 K for the MB-20 bulk and 1.0 K for the MB-30 bulk. The result that T_c is slightly lower than the optimal T_c of 39 K may arise from the existence of the impurity phases. It is noteworthy that the $MgCu_2$ phase seems not to degrade the T_c for the MB-30 bulk.

Figure 6 depicts the magnetic field dependence of the critical current density, $J_c(H)$, at 20 K. The samples measured are the same as those used for making the $M(T)$ measurements. For the MB-20 bulk, as found in figures 6(a) and (b), the J_c is about 1.2–1.6 × 10^5 A cm^{-2} at $\mu_0 H = 0$ T, and decreases monotonically with increasing magnetic field. The J_c curves hardly depend on the sample position except for above 2.5 T. The irreversibility field, $\mu_0 H_{irr}$, defined at the $J_c = 10$ A cm^{-2} value is 3.3–3.6 T. For the MB-30 bulk, as shown in figures 6(c) and (d), the $J_c(H)$s also decrease monotonically with increasing magnetic field. The properties of the vortex pinning of the edge region are slightly inferior to those of the centre region; the $J_c(0\ \mathrm{T})$ and $\mu_0 H_{irr}$ values, respectively, are 1.1–1.3 × 10^5 A cm^{-2} and 3.3–3.5 T for the centre samples and are 0.7–1.0 × 10^5 A cm^{-2} and 3.2–3.3 T for the edge samples. Both the $J_c(0\ \mathrm{T})$ and $\mu_0 H_{irr}$ values for the MB-30 bulk are somewhat small compared with those for the MB-20 bulk, which can explain the relatively low B_T value of the MB-30 bulk. The $MgCu_2$ impurity phase in the MB-30 bulk seems not to act as a pinning centre.

We compare the microscopic J_c (micro-J_c) determined from the magnetization of the small pieces with the macroscopic J_c (macro-J_c) which gives the trapped field. Under the assumption of a uniform J_c distribution in the bulk, which is supported by the uniform two-dimensional B_T distribution for the other MgB_2 bulk in our previous paper [21], we estimate the macro-J_c value of the bulks from the experimentally obtained B_T using the following analytical

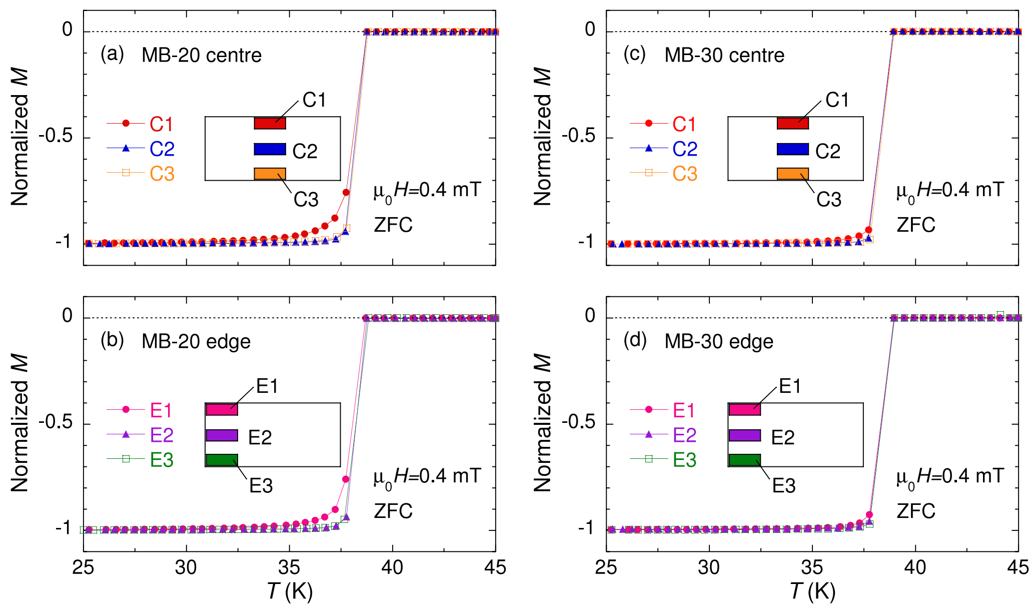

Figure 5. Temperature dependence of the normalized magnetization in the magnetic field of $\mu_0 H = 0.4$ mT after zero-field cooling for the MB-20 ((a) and (b)) and MB-30 ((c) and (d)) bulks. The sample positions are shown in the figure.

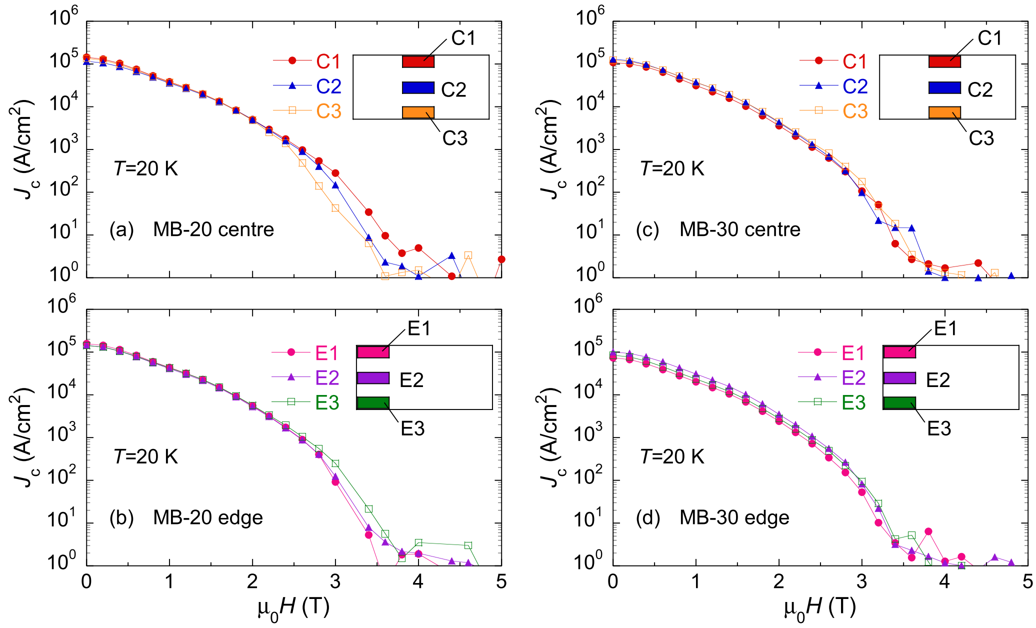

Figure 6. Magnetic field dependence of the critical current density, $J_c(H)$, at $T = 20$ K for the MB-20 ((a) and (b)) and MB-30 ((c) and (d)) bulks. The sample positions are shown in the figure.

equation based on the Biot–Savart law:

$$B_T(z) = \frac{\mu_0 J_c}{2}\left((z+t)\log\frac{(D/2)+\sqrt{(D/2)^2+(z+t)^2}}{z+t} - z\log\frac{(D/2)+\sqrt{(D/2)^2+z^2}}{z}\right), \quad (1)$$

where D and t are the diameter and thickness of the disc-shaped superconductor and z is the distance from the centre of the top surface of the bulk. For the MB-20 bulk, the macro-J_c at 20 K is estimated to be 5.5×10^4 A cm^{-2} using the experimentally obtained B_T ($z = 0$) of 1.09 T. For the MB-30 bulk, on the other hand, the estimated macro-J_c at 20 K is about 4.2×10^4 A cm^{-2} from the measured B_T ($z = 0$) of 1.33 T. The estimated macro-J_c value is about 30–60% of the measured micro-J_c for both bulks; similar results were also observed for the REBCO bulks [10]. The small macro-J_c compared to the micro-J_c is probably caused by the degree of connectivity at the interface of the grains [20], because it prevents the supercurrent from circulating. The results of analysis suggest that the 3 T class MgB_2 bulk magnets 20–30 mm in diameter can be produced at 20 K by improving the macro-J_c, which is as large as the micro-J_c.

4. Summary

We measured the trapped magnetic field and the critical current density of the large MgB_2 bulks (20–30 mm in diameter and 5–9 mm in thickness) fabricated by the capsule method. The important results are described below.

(i) The maximum of the trapped field at the top surface was 1.43 T at 13.4 K for the MgB_2 bulk 20 mm in diameter and was 1.50 T at 16.4 K for the one that was 30 mm in diameter.

(ii) The critical current density, J_c, which was measured using small pieces cut from the bulk was approximately 1×10^5 A cm^{-2} at 20 K in the self-field.

(iii) The macroscopic J_c was estimated to be 5.5×10^4 A cm^{-2} for the 20 mm diameter MgB_2 bulk at 20 K from the trapped field of 1.09 T and was 4.2×10^4 A cm^{-2} for the 30 mm diameter one at 20 K, from the trapped field of 1.33 T, using the analytical equation. The estimated J_c values were as small as about 30–60% of the microscopic J_c which was measured using the small pieces cut from the bulk.

(iv) To produce a strong and large MgB_2 bulk magnet over 50 mm in diameter, improvement of the critical current density, mass density and connectivity is needed.

Acknowledgments

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