Effects of SPS pressure on the mechanical properties of high packing ratio bulk MgB₂ superconductor

A Murakami¹, A Iwamoto² and J G Noudem³

¹ National Institute of Technology, Ichinoseki College, Takanashi, Hagisho, Ichinoseki, Iwate 021-8511, Japan
² National Institute for Fusion Science, 322-6 Oroshi-cho, Toki, Gifu 509-5292, Japan
³ CRISMAT-CNRS UMR 6508/UNICAEN-ENSICAEN, 6 bd Marechal Juin, Caen Cedex 04, 14050, France

E-mail: amura@ichinoseki.ac.jp

Abstract. In order to investigate the effects of SPS pressure on the mechanical properties of MgB₂ bulk, MgB₂ bulk samples were processed by SPS under different pressures. Mechanical properties of these bulk samples were evaluated at room temperature through the bending tests for specimens cut from the bulk samples. Stress-strain behaviours of these bulk samples were almost linear until the fracture. No significant difference was observed for the Young’s modulus values among the bulk samples. The average bending strength values of these bulk samples were also similar to each other. However, the bending strength data of the samples processed under higher pressure scattered widely in comparison with those of the sample processed under lower pressure. The bending strength values were related with the pore sizes observed on the tensile side fracture surfaces.

1. Introduction

Enlargement of bulk superconductor is effective in improving the performance of superconducting devices. Although the superconducting transition temperature of MgB₂ is lower than that of REBaCuO (RE: Y or rare-earth elements), both MgB₂ and REBaCuO bulks are promising for liquid-helium-free bulk superconductor. REBaCuO bulks have to be a single-grain to trap strong magnetic field. REBaCuO bulks are commonly fabricated by melt-processing using a seed crystal. It is difficult to obtain large single-grained REBaCuO bulk piece due to the undesirable nucleation apart from the seed crystal. On the other hand, MgB₂ bulk does not have to be a single-grain [1-12]. Thus, it is not so difficult to obtain large bulk piece for MgB₂ in comparison with REBaCuO. Sintering for the MgB₂ bulk is commonly carried out under the ambient pressure [1-4]. However, the fracture strength value of a low packing ratio MgB₂ bulk sintered under the ambient pressure [13] is lower than those of REBaCuO bulks [14,15]. Since bulk superconductors are subjected to electromagnetic force and thermal stress in the superconducting devices [16,17], improvements of the mechanical properties are important for the development of high performance superconducting devices. It has been reported that the superconducting properties of MgB₂ bulks processed by high-pressure sintering, such as hot-isostatic pressing (HIP) and spark plasma sintering (SPS), are excellent [5-12]. The present authors
previously reported that the fracture strength of a high packing ratio MgB$_2$ bulk processed by SPS was significantly higher than those of MgB$_2$ bulks fabricated by other sintering processes [18]. Thus, spark plasma sintered MgB$_2$ bulk is promising for high-performance bulk superconductor. However, the effects of SPS conditions on the mechanical properties of MgB$_2$ bulk have not been understood extensively. The present authors previously investigated the effects of SPS temperature on the mechanical properties of MgB$_2$ bulk through the bending tests for specimens cut from MgB$_2$ bulk samples processed at 950, 1000 and 1100 °C, respectively [19]. The mechanical properties of the bulk sample processed at 1100 °C were superior to those of other bulk samples [19]. In this study, the effects of SPS pressure on the mechanical properties were investigated through the bending tests for specimens cut from MgB$_2$ bulk samples processed at 1100°C.

2. Experimental procedure

Table 1 presents the SPS conditions for the fabrication of the MgB$_2$ bulk samples. In this study, two spark plasma sintered MgB$_2$ bulk samples were prepared under the pressure of 75 and 100 MPa. They are denoted as Samples 75 and 100, respectively, as shown in Table 1. Mechanical properties of these bulk samples are compared with those of a MgB$_2$ bulk sample processed in the previous study under the pressure of 50 MPa [19]. This bulk sample is denoted as Sample 50. Commercial MgB$_2$ powder from ABCR GmbH (Karlsruhe, Germany) was used for the fabrication of these bulk samples. The SPS process was carried out in the dynamic vacuum atmosphere (10$^{-3}$ bar) under the pulsed electric current of 2000 A. A graphite die was used for the SPS process. The diameter and thickness of the bulk samples were about 30 and 8 mm, respectively. Details of the fabrication process are reported in [12].

Table 1. Spark plasma sintering conditions.

| Sample | 50$^{[19]}$ | 75  | 100 |
|--------|-------------|-----|-----|
| Temperature [°C] | 1100 | 1100 | 1100 |
| Pressure [MPa] | 50  | 75  | 100 |
| Duration [min] | 20  | 20  | 20  |

Bending test specimens were cut from the bulk samples as schematically shown in Figure 1. Wire-electrical discharge cutting machine was used for the cutting. Since spark plasma sintering process is carried out under the uniaxial pressure, two types of specimens were prepared to investigate the effects of the bending loading directions on the bending test results. These specimens are denoted as Specimens A and B, respectively. 4-point bending load was applied at room temperature in the 2.1 mm direction of the specimens as schematically shown in Figure 2. Outer supporting span and inner loading span of the 4-point bending test were 21 and 7 mm, respectively. While the loading direction for the bending tests of the Specimens A was parallel to the compression direction in the SPS process, the loading direction for the bending tests of the Specimens B was perpendicular to it. One strain gauge was glued to the specimens to measure the strain caused by the loading. The strain gauge length was 0.2 mm. The loading speed was 0.2 mm/min. 4-point bending stress $\sigma$ was calculated by the following equation.

$$\sigma = \frac{3P(L - l)}{2wt^2} \quad (1)$$

Where $P$ is applied load, $L$ is outer supporting span (21 mm), $l$ is inner loading span (7 mm), $w$ and $t$ are width and thickness of the specimens (2.8 and 2.1 mm), respectively. After the bending tests, fracture surfaces were observed by using a scanning electron microscope.
3. Superconducting properties

To investigate the superconducting properties, the small specimens with dimensions of 2×2×2 mm³ were cut from the bulk MgB₂ samples in order to measure their critical temperature ($T_c$) and record their magnetization hysteresis loops ($M-H$ loops) at various temperatures (5, 20 and 30 K) between –5 and +5 T with a SQUID magnetometer (Quantum Design, model MPMS5). The critical current density, $J_c$ values of the sample processed at 75 MPa were estimated using the extended Bean critical state model for a rectangular sample [20].

The zero field cooled magnetization curve of Figure 3 performed on the sample processed at 75 MPa, shows a very narrow transition with an onset at 38.5 K. The critical current density, $J_c$, was estimated from magnetization, $M-H$ cycle at different temperatures (Figure 4) on the basis of the extended Bean model [20] leading to an estimated critical current density $J_c \approx 2.2 \times 10^5$ A/cm² in self field. While $J_c$ decreases as the temperature and the external field increases respectively, its value with 1T applied field ($J_c = 1.1 \times 10^5$ A/cm²) is still interesting for practical applications. This high $J_c$ value could be related to the excellent connectivity between grains and is similar to published results [11] in which the samples were prepared using the same processing conditions under high applied pressure (80 MPa).

4. Bending test results

Figure 5 presents stress-strain curves of the specimens cut from the Samples 75 and 100. Data points represent the fracture of the specimens. Stress-strain curves of the Specimens A and B are not distinguished in this figure. Since MgB₂ is commonly brittle and both the Samples 75 and 100 are quite dense as mentioned in the followings, stress-strain behaviours of these bulk samples are almost linear until the fracture. These stress-strain behaviours are similar to the behaviour observed for the
Sample 50 in the previous study [19]. On the other hand, it has been reported that stress-strain behaviours of the specimens cut from a low packing ratio MgB$_2$ bulk are not linear [13].

Figure 6 presents the relationship between the Young’s modulus and SPS pressure for the Specimens A and B. The Young’s modulus values were obtained from the slope of the stress-strain curves shown in Figure 5. Solid and open symbols represent the data of the Specimens A and B, respectively. While the loading direction for the bending tests of the Specimens A was parallel to the thickness of the bulk sample, the loading direction for the bending tests of the Specimens B was perpendicular to it (see Figures 1 and 2). The average Young’s modulus values of the Sample 50 reported elsewhere [19] are also shown in Figure 6 for reference. There is no significant difference in the average Young’s modulus values among the SPS pressures of 50, 75 and 100 MPa. It is mainly attributable to that the packing ratios of these bulk samples are similar to each other. Since the net cross-sectional area of the specimen increases with the increase of the packing ratio, the Young’s modulus commonly increases with increase of the packing ratio. The increase of the net cross-sectional area commonly suppresses the deformation of the specimen and thus improves the Young’s modulus. There is no significant difference in the Young’s modulus values between the Specimens A and B of the high pressure Samples 75 and 100, which is similar to the bending test results of the low pressure Sample 50.

Figure 7 presents the relationship between the bending strength and SPS pressure. The bending strength means the stress at fracture. Figure 8 presents the Weibull plots of the bending strength data. Weibull plots are commonly carried out to evaluate the scatter of the data. The slope obtained through the linear fitting of the data points represents the Weibull coefficient m; larger Weibull coefficient value means smaller scatter of the data. Since the dependence of the bending strength data on the loading directions has not been observed distinctly in Figure 7, the data of the Specimens A and B are not distinguished in Figure 8. The average bending strength values of the Samples 50, 75 and 100 are similar to each other (see Figure 7). However, the bending strength data of the Samples 75 and 100 scatter widely in comparison with those of the Sample 50 (see Figure 8).

Figures 9 and 10 present scanning electron micrographs of the fracture surfaces of the Specimens A and B cut from the Sample 75. Bottom of the micrographs corresponds to the tensile side where the fatal crack initiates by bending loading. Although some extraordinary pores are observed on the fracture surfaces as marked by arrows, the specimens cut from the Sample 75 are quite dense. The sample 100 is also dense. There is no significant difference in the fracture surface morphologies between the Specimens A and B. Fracture surface morphologies of these specimens are typical of ceramic materials; the tensile side fracture surface is relatively flat. Based on the fracture surface morphology, it is deduced that unstable fracture occurred. Fracture surface morphologies of the
specimens cut from the Samples 75 and 100 are similar to each other. One extraordinary pore is clearly observed on the tensile side fracture surfaces of the specimens shown in Figures 9 (a) and 10 (a). The pore sizes of the specimens shown in Figures 9 (a) and 10 (a) are similar to each other. The bending strength values of these specimens are also similar to each other, 257 and 252 MPa, respectively. On the other hand, few pores are observed on the fracture surface of the specimen shown in Figure 10 (b). The bending strength value of this specimen is 394 MPa, which is the maximum value among the specimens of the Sample 75. One small pore is slightly observed on the fracture surface of the specimen shown in Figure 9 (b). The bending strength value of this specimen is slightly
lower than that of the specimen shown in Figure 10 (b). Although it has not been clearly understood the reason why the bending strength data of the high pressure bulk samples (Samples 75 and 100) scatter widely in comparison with those of the low pressure bulk sample (Sample 50) at present, it is deduced that the bending strength values are related with the pore sizes observed on the tensile side fracture surfaces.

Figure 11 presents the relationship between the bending strength and porosity of MgB$_2$ bulk materials. Data of other MgB$_2$ bulk materials processed by SPS under different temperature [19] and by other sintering techniques such as capsule method CAP and HIP [13] are shown for reference. The packing ratio increases with increase of the SPS temperature. Quite dense bulk samples are obtained by SPS at 1100°C. The packing ratio largely affects the bending strength. The relationship between the average bending strength value and packing ratio can be approximated exponentially [13]. The average bending strength increases with increase of the packing ratio. In this study, in order to reduce the scatter of the bending strength data of the quite dense bulk, effects of the SPS pressure on the bending strength were investigated for the bulk samples processed at 1100°C. However, the bending strength data of the bulk samples processed under higher pressure, 75 and 100 MPa, scattered widely in comparison with the sample processed under 50 MPa as mentioned above. Future work will investigate the effect of dwell time during SPS process on the scatter of the strength data.

Figure 9. Fracture surfaces of Specimens A of Sample 75 fractured at (a) 257 MPa and (b) 355 MPa, respectively.

Figure 10. Fracture surfaces of Specimens B of Sample 75 fractured at (a) 252 MPa and (b) 394 MPa, respectively.
5. Summary

In order to investigate the effects of SPS pressure on the mechanical properties of MgB$_2$ bulk, 4-point bending tests were carried out at room temperature for the specimens cut from the bulk samples. Dense bulk samples were obtained by the SPS process. Stress-strain behaviours of these bulk samples were almost linear until the fracture. The Young’s modulus values were evaluated from the slope of the stress-strain behaviours of the specimens. No significant difference was observed for the Young’s modulus values among the bulk samples. The average bending strength values of these bulk samples were also similar to each other. However, the bending strength data of the sample processed under higher pressure scattered widely in comparison with those of the sample processed under lower pressure. One extraordinary pore was observed on the tensile side fracture surfaces of the specimens with lower bending strength. The bending strength values were related with the pore sizes observed on the tensile side fracture surfaces.

Acknowledgements

The authors thank Mr. J. Lecourt of CRISMAT laboratory and students of National Institute of Technology, Ichinoseki College for their relevant technical support. This work was supported in part by KAKENHI (17K06825) and NIFS Collaboration Research Program (NIFS17KECA050).

References

[1] Zhao Y, Feng Y, Cheng C H, Zhou L, Wu Y, Machi T, Fudamoto Y, Koshizuka N and Murakami M 2001 Appl. Phys. Lett. 79 1154
[2] Naito T, Sasaki T and Fujishiro H 2012 Supercond. Sci. Technol. 25 095012
[3] Muralidhar M, Ishihara A, Suzuki K, Fukumoto Y, Yamamoto Y and Tomita M 2013 Physica C 494 85
[4] Yamamoto A, Ishihara A, Tomita M and Kishio K 2014 Appl. Phys. Lett. 105 032601
[5] Shields T C, Kawano K, Holdom D and Abell J S 2002 Supercond. Sci. Technol. 15 202
[6] Tampieri A, Celotti G, Sprio S, Caciuffo R and Rinaldi D 2004 Phys. C, Supercond. 400 97
[7] Durrell J H, Dancer C E J, Dennis A, Shi Y, Xu Z, Campbell A M, Hari Babu N, Todd R I, Grovenor C R M and Cardwell D A 2012 Supercond. Sci. Technol. 25 112002
[8] Sasaki T, Naito T and Fujishiro H 2013 Phys. Procedia 45 93
[9] Schmidt J, Schnelle W, Grin Y and Kniep R 2003 Solid State Sci. 5 535
[10] Shim S H, Shim K B and Yoon J W 2005 J. Amer. Ceram. Soc. 88 858
[11] Dancer C E J, Prabhakaran D, Basoglu M, Yanmaz E, Yan H, Reece M, Todd R I and Grovenor C R M 2009 Supercond. Sci. Technol. 22 095003
[12] Noudem J, Aburras M, Bernstein P, Chaud X, Muralidhar M and Murakami M 2014 J. Appl. Phy. 116 163916
[13] Murakami A, Teshima H, Naito T, Fujishiro H and Kudo T 2014 Phys. Procedia 58 98
[14] Murakami A, Miyata H, Hashimoto R, Katgiri K and Iwamoto A 2008 Physica C 468 1395
[15] Murakami A and Iwamoto A 2017 J. Phys.: Conf. Ser. 871 012055
[16] Ren Y, Weinstein R, Liu J, Sawh R P and Foster C 1995 Phys. C, Supercond. 251 15
[17] Miyamoto T, Nagashima K, Sakai N and Murakami M 2000 Supercond. Sci. Technol. 13 816
[18] Murakami A, Noudem J, Guesmi Z, Kudo T and Iwamoto A 2015 Phys. Procedia 65 77
[19] Murakami A, Iwamoto A and Noudem J G IEEE Trans. Appl. Supercond. Submitted
[20] Chen D X and Goldfarb R B 1989 J. Appl. Phys. 66 2489