Characteristics of MgB_2 superconducting bulks fabricated by spark plasma sintering

Fujishiro · Naito Laboratory 20211006 Amira

1. INTRODUCTION

The intermetallic compound of magnesium diboride (MgB_2) has the highest superconducting transition temperature T_c=39 K, which are expected to be applicable in magnetically levitated trains (MAGLEVs) and wind power generators using liquid H_2 or a cryocooler system. MgB_2 superconducting bulk also has a promising potential as a quasi-permanent magnet with high-performance, light-weight and homogeneous trapped field distribution superconducting materials. MgB_2 has a long coherence length compared to RE-Ba-Cu-O (RE: rare earth elements). Therefore, the problem of weak-links at grain boundaries can be ignored and a superconducting bulk magnet using a polycrystal can be realized.

To fabricate a high-performance MgB_2 superconducting bulk, one important issue is to attain competitive values for critical current density, J_c under high applied magnetic fields. It has been reported that by enhancement of the filling rate and the decrease of the grains size, the pinning centres at the grain boundaries increase; hence we are able to improve J_c of the bulk. We have suggested that to improve the filling rate, we can control the grains size in the bulk. The smaller the grains size, the greater the number of the grain boundaries in the bulk. Mortar mixing and ball milling methods were considered which could decrease the grains size. Moreover, the Spark Plasma Sintering (SPS) method strongly influences the filling rate and grain boundaries and also produces nanograined high density bulk materials. In this study, to optimize the mixing and milling conditions, we have investigated the trapped field and superconducting properties of MgB_2 bulks fabricated by SPS method using MgB_2 *ex-situ* powder.

2. EXPERIMENTAL

MgB_2 *ex-situ* powders (99% in purity) were weighted and mixed using mortar mixing and ball milling machine. The conditions for mortar mixing samples are 1 h, 2 h, 10 h in air atmosphere, and for ball milling conditions are 1 h, 12 h, 48 h at 250 rpm in argon atmosphere. Then the powders were loaded into a graphite die (mortar mixed samples: ø=10 mm, ball milled samples: ø=20 mm) and sintered between 600 ˚C ~ 1050 ˚C for 5-10 min in vacuum under a mechanical uniaxial pressure and a pulse electrical current of SPS. The applied pressures were 3.93 kN for ø=10 mm samples and 15.71 kN for ø=20 mm samples. For sample measurement, the structural characterization was performed using XRD (X-Ray Diffraction). XRD patterns were measured before and after sintering process. The trapped fields and temperatures in the centre of the bulk surface were measured using a Hall sensor and cernox. The magnetization characteristics were measured using a SQUID (Superconductor QUantum Interference Device) and J_c was estimated from the hysteresis loop using the extended bean model. The electrical resistivity was measured by using a standard four probe technique.

3. RESULTS AND DISCUSSION

Fig.1 (a) and (b) show the Full Width Half Maximum (FWHM) of XRD patterns at the main peak of mortar mixed samples and ball milled samples. The mortar mixing effect of MgB_2 *ex-situ* powder at 1 h, 2 h and 10 h does not show much change in grains size compared to the pristine powder. On the other hand, it can be seen that from ball milling effect, remarkably the grain size decreases with the milling time increases. However the grains size remains the same after a milling time of 12 h. Fig.1 (c) shows X-ray diffraction patterns of samples after SPS using mortar mixed powder. As we can see, the MgO, MgB_4 and graphite peaks increase as the mixing time increases. We suggest MgB_2 *ex-situ* powder reacts with air and/or water during the mixing process. Nevertheless, we expect that impurities that exist could act as pinning centres which could improve J_c.

Fig. 2 compares the temperature dependence on trapped fields, B_T for the MgB_2 SPS pristine bulk, SPS mortar mixed bulk and SPS ball milled bulk with the CAP(capsule) and HIP(Hot Isostatic Pressure) method bulks between applies fields B_{ex}=5 T. The SPS bulk using ball milled powder show the highest trapped

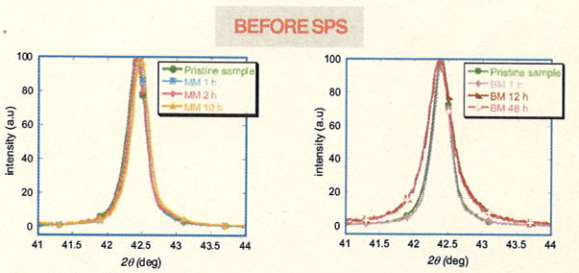


Fig.1 (a) FWHM of XRD patterns at the main peak of Mortar Mixed samples

Fig.1 (b) FWHM of XRD patterns at the main peak of Ball Milled samples

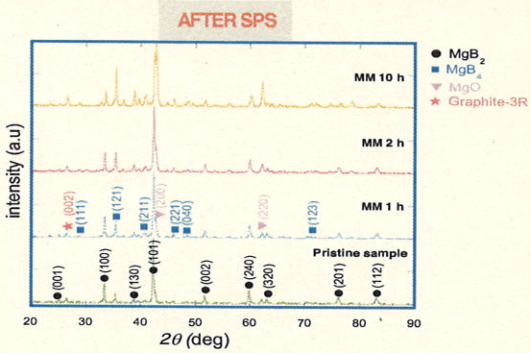


Fig.1 (c) XRD patterns of samples after SPS using pristine sample, and 1 h, 2 h,10 h mortar mixed samples.

fields compared to SPS bulk using mortar mixed powder and shows about the same trapped field as a HIP bulk. For example, at 25 K, SPS ball milled bulk and HIP bulk show the same B_T=1.71 T, instead SPS pristine bulk, SPS mortar mixed bulk and CAP bulk show B_T value 1.52 T, 1.39 T and 1.31 T respectively. We suggest that high trapped field of SPS bulk using ball milled powder was enhanced due to the enhancement of the grain growth and the grain boundaries effect of high energy impact of the ball milling.

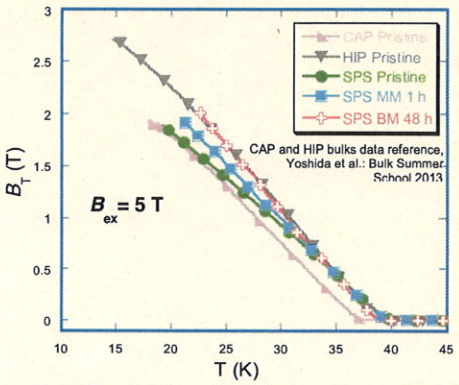
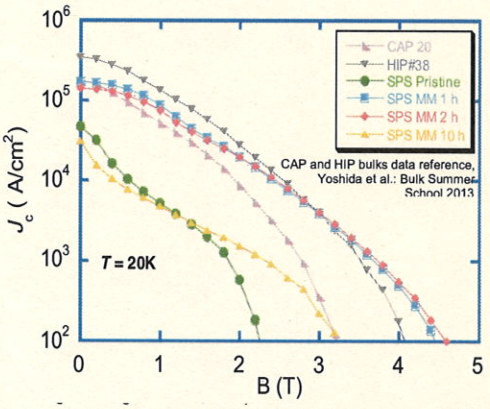


Fig. 2 Temperature dependence of trapped fields, B_T for MgB_2 bulks with CAP, HIP and SPS fabrication methods at B_{ex} = 5 T

Fig. 3 depicts the magnetic field dependence of the critical current density, J_c at 20 K for SPS bulk using pristine powder, SPS bulks using 1 h, 2 h and 10 h mortar mixed powders comparing with CAP and HIP bulks. At zero magnetic field (B=0 T), HIP bulks shows the highest J_c value 3.5×10^5 (A/cm^2). CAP bulk and SPS bulks using 1h and 2 h mortar mixed powders show J_c values in the range 1.4×10^5 ~1.7×10^5 (A/cm^2) and SPS bulk pristine powder shows a J_c value of 4.7×10^4 (A/cm^2). We realized that SPS bulks fabricated by mortar mixing powder and CAP method produced low J_c superconducting bulks compared to the HIP method. Furthermore, by increasing the mortar mixing time, J_c increases due to the coexistence of impurities constraining the magnetic flux lines in the bulk. However, for SPS bulk using 10 h mortar mixed powder shows the lowest J_c. We suggest even though many impurities exist, they do not act as the pinning centers for the bulk and just disturb the magnetic flux lines. In the future, we are planning to measure J_c for SPS bulks using ball milled powder. Related to the high B_T result of SPS bulk using ball milled powder, we expect it will show high J_c values too.



current density, J_c at 20 K for SPS bulk using pristine powder, SPS bulks using 1 h, 2 h and 10 h mortar mixed powders comparing with CAP and HIP bulks

Fig. 4 presents the temperature dependences of the electrical resistivity for SPS bulk using pristine powder, SPS bulk using mortar mixed 1 h, 2 h and 10 h powders. The inset shows resistivity, ρ at 40 K and critical temperature, T_c. Besides, the electrical connectivity, κ which is the effective cross section of the bulk supercurrent path, was evaluated from the following equation, κ=(Δρ crystal / Δρ exp) × 100%, where Δρ crystal=6.32 μΩcm and Δρ exp= ρ(300K) - ρ(40K), i.e., the ratio of the resistivity difference between the resistivity ideal of the ideal MgB_2 grains and that of the present bulks. The connectivity value of SPS pristine, SPS mortar mixed 1 h, 2 h and 10 h are 5.4%, 32.7%, 18.7% and 6.5% respectively. Note that as the mortar mixing time increases, the ρ increases, T_c decreases and κ decreases due to the low purity of the MgB_2 *ex-situ* powder after the mixing process. However, the SPS pristine sample shows the low T_c, highest ρ and the lowest κ. We expect that the bulk was not well-fabricated and some cracks may exist.

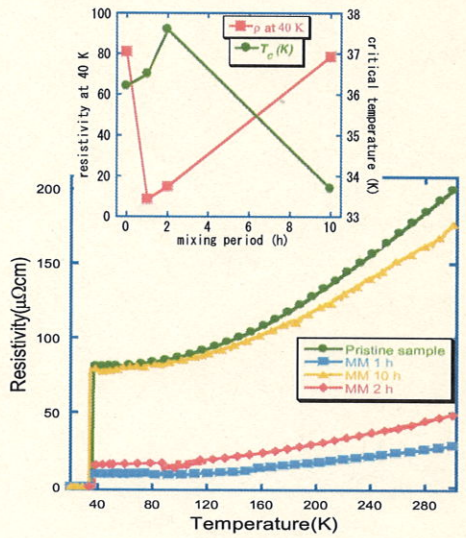


Fig. 4 Temperature dependences of the electrical resistivity for SPS pristine, SPS mortar mixed 1 h, 2 h and 10 h. The inset shows ρ at 40 K and T_c of the bulks.

4. CONCLUSION

In this study, to optimize the mixing and milling condition, we have investigated the trapped field and superconducting properties of MgB_2 bulks fabricated by SPS method using several kinds of MgB_2 *ex-situ* powder. Result of the X-ray diffraction, shows that the grain size decreases as the ball milling time increases. The optimal condition for ball-milling MgB_2 *ex-situ* is 12h at 250 rpm. The mortar mixing results show as the mixing time increases, MgO and MgB_4 peaks increase after SPS due to the reaction occurred during the mixing process. Using a SPS ball milling method, we are able to fabricate a bulk with high B_T which is similar to values attained in the bulk material fabricated by the HIP method, and this is due to an enhancement of the grain growth and the number of grain boundaries. Related to the high B_T result of SPS ball milled bulk, we expect it will show high J_c values too. Besides, mortar mixed bulks show as the mortar mixing time increases, J_c increases due to the coexistence of impurities acting as pinning centres in the bulk. Furthermore, the electrical resistivity result depicts the ρ increases, T_c decreases, κ decreases as the mortar mixing time increases due to low purity of the MgB_2 *ex-situ* powder after mixing process.