

# AC Magnetic Flux Profile and Critical Current Density Obtained by Campbell's Method in Polycrystalline MgB<sub>2</sub>

Baorong Ni, Yoshihiro Morita, Zhiyong Liu, Cuifang Liu, Keizo Himeki,  
Edmund Soji Otabe, Masaru Kiuchi and Teruo Matsushita

**Abstract**— In this study, we focused on the estimation of the global critical current density, super-current path, grain connectivity and their relationships with the faults volume fraction in the MgB<sub>2</sub> bulks prepared by a modified PIT (powder in tube) method. Campbell's method was applied for the purpose of acquiring the penetrating AC magnetic flux profile and the characteristic of AC magnetic field vs. penetration depth from the specimen's surface. A computer simulation on the penetrating AC flux profile in MgB<sub>2</sub> bulks with certain amount of randomly distributed voids, oxidized grains and other faults was also carried out. The global critical current density obtained by Campbell's method turned out to be smaller than that obtained from the SQUID measurement, implying that the global super-current was reduced by the existence of various kinds of faults and the lack of the electrical connectivity between grains. It was verified that the relationship between the global critical current characteristics and the faults contained in MgB<sub>2</sub> specimens can be quantitatively clarified by comparing the simulated critical current densities and other factors with the experimental results.

**Index Terms**— Polycrystalline MgB<sub>2</sub> bulk, Campbell's method, AC flux profile, grain connectivity.

## I. INTRODUCTION

THE highest transition temperature in conventional metallic superconductors, the low cost of the raw materials, the low anisotropy and less weak links at grain boundaries and other unique characteristics of MgB<sub>2</sub> bring us a great potential for practical applications of MgB<sub>2</sub>. Since the discovery of superconductivity in MgB<sub>2</sub> [1] in 2001, extensive researches have been performed and great progress has been made in clarifying the superconducting properties, optimizing the synthesis methods, enhancing the critical current characteristics, understanding the flux pinning mechanisms and so on.

Manuscript received 19 August 2008. This work was supported in part by Electronics Research Laboratory of Fukuoka Institute of Technology, Japan.

Baorong Ni and Yoshihiro Morita are with the Department of Life, Environment and Material Science, Fukuoka Institute of Technology, Japan (corresponding author: Baorong Ni, phone: ++81-92-606-3789; fax: ++81-92-606-3789; e-mail: nee@fit.ac.jp).

Zhiyong Liu and Cuifang Liu are with the Key Lab of New Processing Technology for Nonferrous Metals and Materials, Guilin University of Technology, China.

Keizo Himeki, Edmund Soji Otabe, Masaru Kiuchi and Teruo Matsushita are with the Department of Computer Science and Electronics, Kyushu Institute of Technology, Japan.

As the results of these researches, many reports maintain that the grain boundaries are one of the dominant pinning centers in polycrystalline MgB<sub>2</sub>. Several results succeeded in showing an apparent correlation between the grain size and the critical current density in their MgB<sub>2</sub> specimens [2]. However, it is pointed out [3] that the critical current densities in polycrystalline MgB<sub>2</sub> are far below the values observed in Nb<sub>3</sub>Sn under the same conditions, although it is considered that the flux pinning mechanisms originating in grain boundaries in these two materials resemble each other. Rowell [4] suggested that the reduction of the effective current-carrying cross-sectional area by the existence of voids, oxide phases present in the grain boundaries and other impurity factors in polycrystalline MgB<sub>2</sub>, which directly lead to a reduction of super-current path, is one of the causes that largely suppress the critical current characteristic. Yamamoto [5] explained the relationship between the normal-state conductivity and the electrical connectivity in superconducting MgB<sub>2</sub> by using a site-percolation model, and concluded that the low packing factor and poor connectivity are the fatal reasons restricting the critical current density in MgB<sub>2</sub>. However, quantitative estimation of the critical current characteristic from the measured electromagnetic properties remains as an unsolved problem.

On the other hand, it is known that the AC inductive methods, such as Campbell's method [6], are useful for the estimation of the intricate critical current characteristics in polycrystalline superconducting materials. We once succeeded in distinguishing the inter-grain critical current density from the intra-grain critical current density in polycrystalline YBaCuO bulk by Campbell's method [7]. In this study, we applied Campbell's method as an effective mean for the discussion and estimation on the critical current density, super-current path and their relationships with the faults volume fraction in the polycrystalline MgB<sub>2</sub> bulks. We also carried out a computer simulation on the penetrating AC flux profile in MgB<sub>2</sub> bulks with randomly distributed voids, oxidized grains and other faults.

## II. EXPERIMENTAL

The specimens of MgB<sub>2</sub> bulk were prepared by a modified PIT (Powder-In-Tube) method. Powders of magnesium (99.99%, 125 – 300 μm), amorphous boron (99%, 300 mesh) and commercial MgB<sub>2</sub> powder were used for the starting

materials, while the molar ratio of these three materials was 1.2:2:0.5. The details of the specimen preparation were given in our previous paper [8]. The heat treatment temperature was fixed at 900 °C, while the times of the heat-treatment were varied from 12 h to 300 h, in order to bring variations in the average grain size and the extent of oxidation in MgB<sub>2</sub>. The densities of most of the specimens, estimated by Archimedes method, were larger than the value of 90% of the theoretical density. SEM images of those specimens with high densities show few voids observed, denoting that the packing factor was quite high. However, XRD results show some small peaks corresponding to MgO, implying that certain extent of oxidation occurred during the process of specimen preparation. Fig. 1 shows a heat treatment time dependence of average grain size in the bulk. By analyzing the SEM images and EPMA data, we believe that the grains can be regarded as the single crystals.

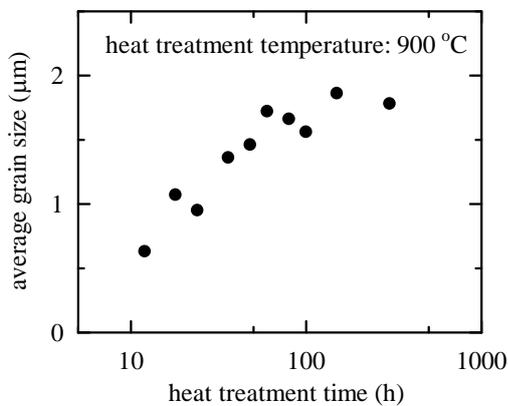


Fig. 1 Heat treatment time dependence of the average grain size measured from the SEM images of the specimens. The heat treatment temperature was fixed at 900 °C.

The critical temperature  $T_c$ 's of specimens were measured by a SQUID magnetometer. All specimens show the  $T_c$  values closing to 38.5 K, with the deviations smaller than 1 K. The critical current density,  $J_c$ , was measured and estimated by DC magnetization (SQUID magnetometer) and Campbell's method, respectively. The temperature of specimen was 4.2 K – 35 K, while the magnetic field was up to 6 T for SQUID measurement and up to 0.4 T for Campbell's method measurement. The frequency of the AC magnetic field in Campbell's method was fixed at 37 Hz.

### III. RESULTS AND DISCUSSION

The magnetic field dependence of  $J_c$  estimated from measured DC magnetization at  $T = 20$  K is shown in Fig. 2. As a pure MgB<sub>2</sub> bulk without any carbide doped, the specimen with 12 hours heat-treatment shows a tolerable  $J_c$  value, whereas the specimens experienced a longer heat-treatment time show an apparent decrease in  $J_c$  value. The fact could be considered as the result of the grain's coarsening in MgB<sub>2</sub> due to the heat-treatment, since the grain boundaries are one of the dominant pinning centers, and the measured grain size was increased according as the heat-treatment time lengthened as shown in Fig. 1. Another reason that reduces  $J_c$  value is probably the oxidizing progress of the surface of MgB<sub>2</sub> grains, since it is reasonable to consider that the longer heat-treatment

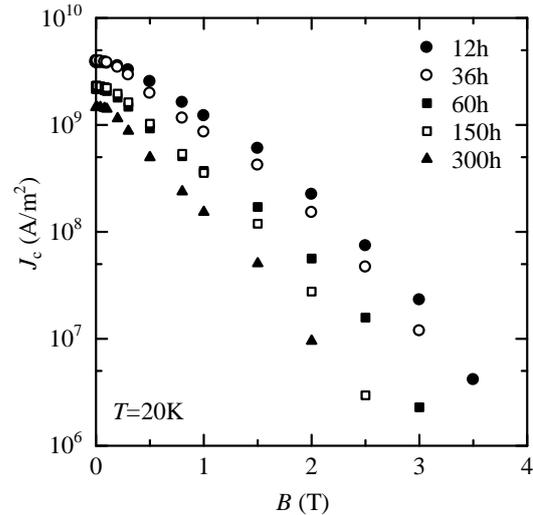


Fig. 2 Magnetic field dependence of the critical current densities obtained from the SQUID measurement at  $T = 20$  K. The MgB<sub>2</sub> bulks experienced different heat-treatment time from 12 hours to 300 hours.

time lasts, the more oxidizing proceeds.

As one of the measurements dealing with the electromagnetic properties in superconductor, Campbell's method measures the penetrating AC flux corresponding to the applied DC and variable-amplitude AC magnetic fields. By analyzing the measured AC flux profile and AC magnetic field  $b_{ac}$  vs. penetration depth from the surface of the specimen,  $\lambda'$ , one can derive not only the critical current density but also the relationship between the force and the displacement of the flux lines. In this study, we focused our attention on the AC flux profile itself and the estimation of global  $J_c$  in MgB<sub>2</sub> bulk.

Taking the factor of electric field criterion, which defines the critical current density in superconductors, into account,  $J_c$  value from Campbell's method is usually overestimated, since the induced electric field under the applied AC magnetic field with several tens Hz or more should be larger than those in DC magnetization method or resistive method. If the  $n$  value in the current-voltage curve ( $V \propto I^n$ ) is sufficient large, then, the error caused by the overestimation should be negligible; otherwise the error should be a considerable difference. Fig. 3

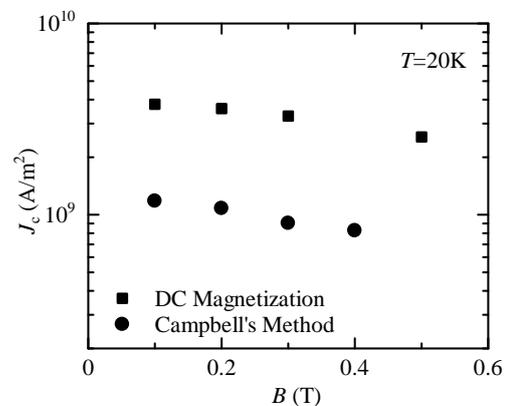


Fig. 3 Comparison of the critical current densities at  $T = 20$  K from DC magnetization method (square symbols) and Campbell's method (circle symbols), respectively. The specimen experienced a 12 hours heat-treatment at 900 °C.

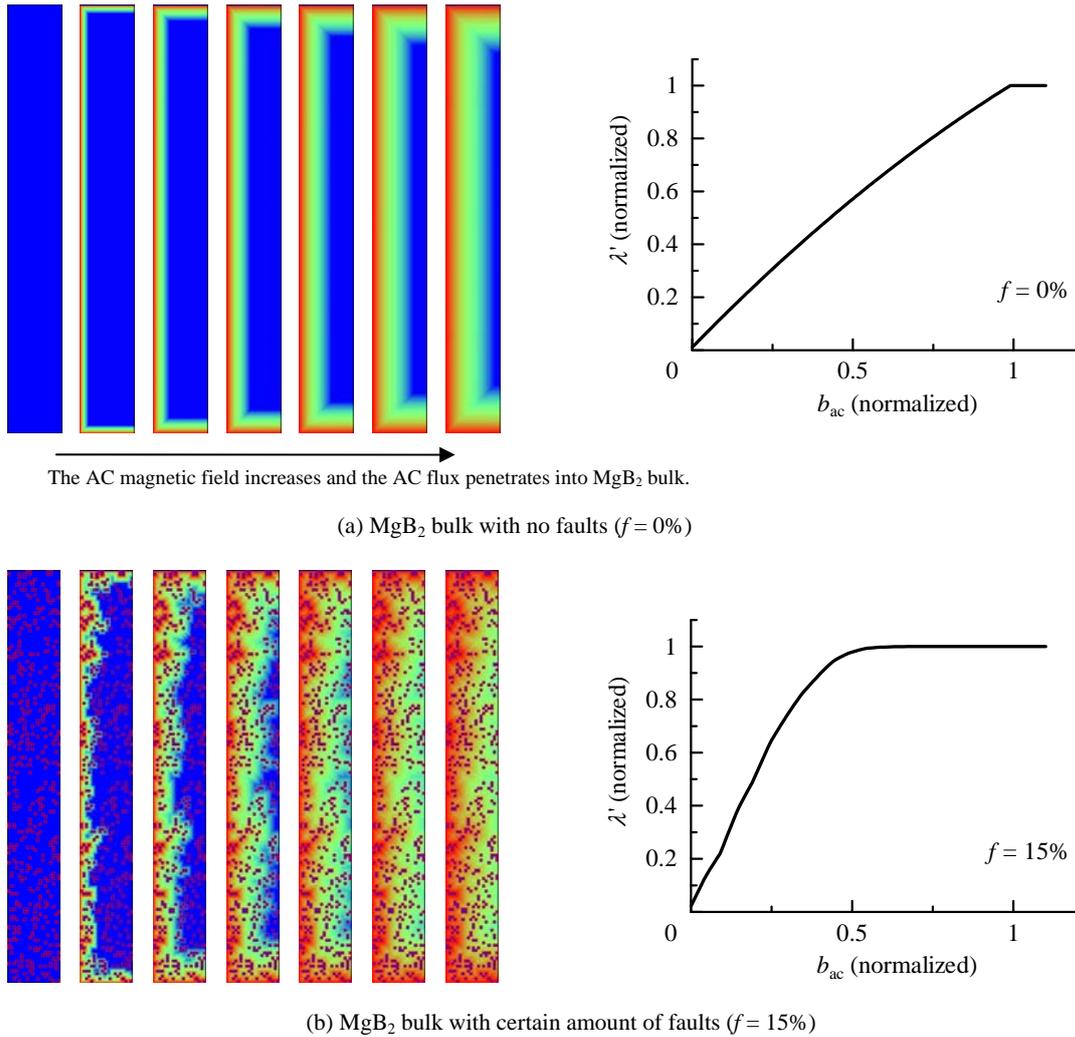


Fig. 4 Simulation of the AC flux profiles (left) and the corresponding  $b_{ac}$  vs.  $\lambda'$  curves (right) in polycrystalline MgB<sub>2</sub> bulk. (a) and (b) show the simulation results in MgB<sub>2</sub> bulks without and with certain amount of faults, respectively. The rectangles of the left denote the half of the rectangular cross section area of MgB<sub>2</sub> bulk, i.e., the right side of the rectangular shape is the center of the MgB<sub>2</sub> bulk, and the AC flux penetrates into the specimen from the rest of the three sides. When the applied AC magnetic field increases, the AC flux reaches deeper into the specimen, and the value of  $\lambda'$  increases as shown in the left and the right part of the figure, respectively.

shows a comparison of  $J_c$  values at  $T=20$  K obtained from DC magnetization and Campbell's method, respectively. Contrary to our expectations,  $J_c$  from Campbell's method shows a smaller value. Generally, it is understood that the global (inter-grain) critical current density is equal to or smaller than the local (intra-grain) one in a polycrystalline superconductor. Considering the fact that in Campbell's method  $J_c$  was derived from the initial part of the  $b_{ac}$  vs.  $\lambda'$  curve, meaning that the value is corresponding to the global current in MgB<sub>2</sub> bulk, while  $J_c$  from DC magnetization was estimated based on the average magnetization per unit volume, meaning that the value is just an average of the global and the local currents, the result strongly implies that the super current in MgB<sub>2</sub> bulk flows in a complicated path, bringing a reduction in current density value.

In order to give a more quantitative discussion, a computer simulation of penetrating AC flux profile was carried out. Supposing that MgB<sub>2</sub> bulk contains some certain amount of randomly distributed voids and grains which have an oxide layer on their surface, and the amplitude of the AC magnetic field is sufficient small so that the influence on critical current

density is negligible, then, the AC flux profile can be simulated and illustrated as the left of Fig. 4. In the simulation, the voids and the grains with oxide layer on the surface were approximated to a cubic normal phase with same size. The rectangles in the left part of Fig. 4 denote the half of the rectangular cross section area of MgB<sub>2</sub> bulk, i.e., the right side of the rectangular shapes are the centers of the MgB<sub>2</sub> bulk, and the AC flux penetrates from the rest of the three sides. When the applied AC magnetic field increases, the AC flux reaches deeper from the surface of MgB<sub>2</sub> bulk, and the calculated value of  $\lambda'$  (see below) increases as shown in the left and the right part of Fig. 4, respectively. Comparing (a), the AC flux profile in the specimen without any faults, with (b), that in the specimen with 15% faults, it is clear that if MgB<sub>2</sub> bulk contains some faults, then the AC flux penetrates into the specimen rapidly, which brings on a decrease of global shielding current density.

Based on the penetrating AC flux profile as shown in the left part of Fig. 4, the  $b_{ac}$  vs.  $\lambda'$  curve can be easily obtained according to the equation used in Campbell's method as

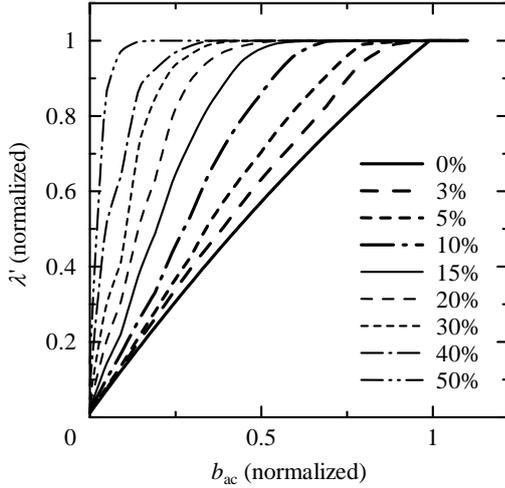


Fig. 5 Calculated  $b_{ac}$  vs.  $\lambda'$  curves of the MgB<sub>2</sub> bulks with different faults volume fractions.  $b_{ac}$  and  $\lambda'$  were normalized by  $b_p$  and  $d/2$ , respectively.

$$\lambda' = \frac{1}{4}(w+d) \left[ 1 - \left( 1 - \frac{4}{(w+d)^2} \frac{d\Phi}{db_{ac}} \right)^{\frac{1}{2}} \right] \quad (1)$$

where  $\Phi$ ,  $w$  and  $d$  are the penetrating AC flux, specimen's width and thickness, respectively. Fig. 5 shows the calculated  $b_{ac}$  vs.  $\lambda'$  curves of the specimens with different faults volume fractions,  $f$ , while  $b_{ac}$  and  $\lambda'$  were normalized by the penetration magnetic field  $b_p$ , at which the AC flux reaches the center of the specimen without any faults, and the half of the thickness of the specimen, respectively. Since the global critical current density is calculated from the gradient of the initial part of  $b_{ac}$  vs.  $\lambda'$  curve in Campbell's method, it is obvious that the increase of faults amount leads to a decrease of  $J_c$  value.

On the other hand, the DC magnetization, which is usually obtained from SQUID magnetometer or other inductive measurements, can also be simulated by the penetrating AC flux profile. In this case, the penetration magnetic field  $B_p$  was regarded as the AC magnetic field at which the AC flux reaches the center of the superconductor. The averaged value of the local and global  $J_c$  was calculated by applying the equation of the critical state model. Fig. 6 shows a comparison of  $f$  vs.  $J_c$  with the experimental data obtained from the DC magnetization of MgB<sub>2</sub> bulks with different packing factors [5]. In Fig. 6, the packing factor defined in [5] is identified with  $1 - f$  in our simulation and  $J_c$  was normalized by the value in the specimen without any faults. The good agreement between our simulation and the experimental result suggests that the assumptions of the faults existence and the tangled current path in MgB<sub>2</sub> bulk in our simulation are reasonable.

According to the procedure described in [7], the local or intragrain critical current density,  $J_g$ , can be obtained from fitting a calculated  $b_{ac}$  vs.  $\lambda'$  curve to the experimental result for a large  $b_{ac}$ , if the distribution of the grain size of MgB<sub>2</sub> is known and it can be assumed that  $J_g$  is the same in all grains. As an example, with a primitive estimate, a  $J_g$  value was obtained as to be  $8.14 \times 10^9$  A/m<sup>2</sup> at  $B = 0.1$  T and  $T = 30$  K, which is about one order of magnitude larger than the global  $J_c$  obtained from

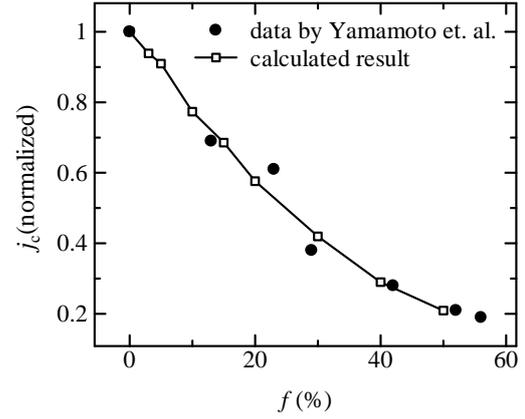


Fig. 6 Comparison of the calculated normalized  $J_c$  values and the experimental data obtained from the DC magnetization of MgB<sub>2</sub> bulks with different packing factors [5]. The packing factor defined in [5] is identified with  $1 - f$ .

SQUID ( $1.80 \times 10^9$  A/m<sup>2</sup>) and Campbell's method ( $8.78 \times 10^8$  A/m<sup>2</sup>). The result directly supports the understanding that the potential and essential critical current density is obstructed by various kinds of faults and the poor electrical connectivity between grains in MgB<sub>2</sub> bulk. Further investigation is in progress.

#### IV. CONCLUSION

Campbell's method was applied for the purpose of obtaining the penetrating AC flux profile and the AC magnetic field vs. penetration depth. A computer simulation on the penetrating AC flux profile in MgB<sub>2</sub> bulks with randomly distributed voids, oxidized grains and other faults was also carried out. We succeeded in estimating the global and local critical current densities based on the experimental data obtained by Campbell's method. The results strongly imply that the global super-current was reduced by the existence of various faults and the poor electrical connectivity in MgB<sub>2</sub> bulks.

#### REFERENCES

- [1] J. Nagamatsu, N. Nakagawa, T. Muranaka, Y. Zenitani, J. Akimitsu, "Superconductivity at 39 K in magnesium diboride," *Nature*, vol. 410, 2001, pp. 63 – 64.
- [2] Y. Katsura, A. Yamamoto, S. Ueda, I. Iwayama, S. Horii, J. Shimoyama and K. Kishio, "Flux pinning properties of undoped and C-doped MgB<sub>2</sub> bulks with controlled grain sizes," *Physica C*, vol. 460 – 462, 2007, pp. 572 – 573.
- [3] T. Matsushita, "Flux pinning in superconductors," *Springer-Verlag, Berlin Heidelberg*, 2007, p.416.
- [4] J. M. Rowell, "The widely variable resistivity of MgB<sub>2</sub> samples," *Supercond. Sci. Technol.* vol. 16, 2003, pp. R17 – R27.
- [5] A. Yamamoto, J. Shimoyama, K. Kishio, T. Matsushita, "Limiting factors of normal-state conductivity in superconducting MgB<sub>2</sub>: an application of mean-field theory for a site percolation problem," *Supercond. Sci. Technol.* vol. 20, 2007, pp. 658 – 666.
- [6] A. M. Campbell, "The response of pinned flux vortices to low-frequency fields," *J. Phys. C2*, 1969, pp. 1492 – 1501.
- [7] B. Ni, T. Munakata, T. Matsushita, M. Iwakuma, K. Funaki, M. Takeo, K. Yamafuji, "AC inductive measurement of intergrain and intragrain currents in high- $T_c$  oxide superconductors," *Jpn. J. Appl. Phys.* vol. 27, 1988, pp. 1658 – 1662.
- [8] B. Ni, Y. Morita, Z. Liu, C. Liu, K. Himeki, E. S. Otobe, M. Kiuchi and T. Matsushita, "Estimation of critical current density and grain connectivity in superconducting MgB<sub>2</sub> bulk using Campbell's method," *Physica C*, to be published.