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Development of MgB₂-based, bulk supermagnets

M. R. Koblishka, A. Wiederhold, M. Muralidhar, K. Inoue, T. Hauet, B. Douine, K. Berger, M. Murakami and U. Hartmann

Abstract—A series of disk-shaped, bulk MgB₂ superconductors (sample diameter up to 4 cm, reaction temperatures ranging between 750 °C and 950 °C) was prepared in order to improve the performance for superconducting supermagnets. These samples were characterized by magnetic and electric measurements in fields up to 7 T and at various temperatures 10 K < T < 35 K. The irreversibility lines, the current densities, the resistance and the achievable trapped fields were determined. To analyze the data, a scaling of the flux pinning forces, $F_p = j_c \times B$, was performed. The different scaling behavior of the samples prepared at low and high reaction temperatures is discussed considering the achieved microstructures.

Index Terms—MgB₂, supermagnets, trapped fields, flux pinning forces.

I. INTRODUCTION

SUPERCONDUCTING MgB₂ trapped-field magnets ("supermagnets") may find a large variety of commercial and industrial applications at temperatures around 20 K, especially in the medical field, e.g., for MRI instruments similar to the 123-cuprate high- T_c superconductors operating at liquid nitrogen temperature (77 K) [1,2]. The main reason for this is the character of the grain boundaries in MgB₂, which do not show a weak-link behavior like in the case of the cuprates [3-10]. The fabrication of bulk MgB₂ superconducting materials is even more attractive due to low fabrication cost and simpler processing since this material does not require a strong texture to reach a high enough critical current density, and eventually, high trapped field values around 20 K. Further, the smaller weight and a higher mechanical

strength are other important factors for possible applications. On the other hand, problems arise due to percolative current flow in the polycrystalline samples, and due to the porous character of the bulk samples. Therefore, we prepared a series of samples with diameters up to 4 cm at various reaction temperatures in order to determine the optimum preparation conditions for bulk MgB₂ samples.

II. EXPERIMENTAL PROCEDURE

Several samples were fabricated using a solid state reaction in pure Ar atmosphere from 750 to 950 °C in order to determine the optimum processing parameters to obtain the highest critical current density as well as large trapped field values. The X-ray analysis of these samples is presented elsewhere [11]. The magnetic characterization measurements were performed using SQUID magnetometry (Quantum Design MPMS) and the magneto-resistance and I/V -characteristics were recorded using an Oxford Instruments 8 T Teslatron system. The critical current densities were calculated from the magnetization loops using the extended Bean model for rectangular samples. The trapped magnetic field was studied by magnetizing the bulk sample in a 10 T superconducting magnet over a wide temperature range between 20 K and 40 K. The transverse Hall sensor, which was directly glued at the center of the top and bottom surfaces of the bulk, has then only 0.5 mm distance between the probe and the surface of the sample. For SQUID measurements, small samples ($2 \times 2 \times 1.5 \text{ mm}^3$) were cut from the big pellets. For the resistance measurements, small bars ($10 \times 1 \times 1 \text{ mm}^3$) were prepared, and the contacts were fixed with silver paint. The AFM topography images were recorded using a Veeco/DI Nanoscope IV AFM employing micromachined Si-cantilevers.



Fig. 1. Overview of the prepared bulk MgB₂ samples for the trapped field

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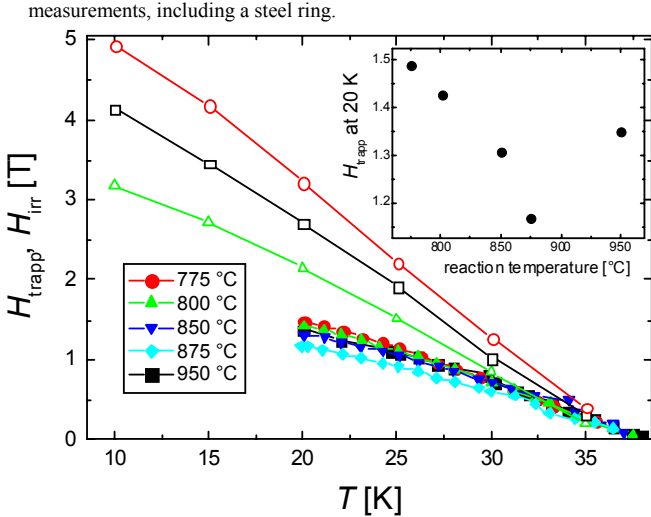


Fig. 2. The irreversibility lines (open symbols) and the measured trapped fields (full symbols) for several selected samples. The inset illustrates the recorded trapped fields at 20 K as a function of the reaction temperature.

III. RESULTS AND DISCUSSION

Figure 2 shows that the MgB₂ sample processed at 775 °C (3 h) showed the highest critical current density, j_c , of 176 kAcm⁻² and 55 kAcm⁻² at 20 K in self-field and a trapped field of 1.49 T, respectively. Furthermore, the critical current density values are increasing further to 250 kA cm⁻² and 150 kAcm⁻² on decreasing the temperature further down to 10 K [10]. Accordingly, this sample exhibits the highest $H_{irr}(T)$ -line, and also the highest trapped fields (measured only down to 20 K). $H_{irr}(T)$ and the trapped fields are then found to decrease when increasing the reaction temperature, however, the data of the sample processed at 950 °C show again an increase of $H_{irr}(T)$ and the trapped field (see the inset to Fig. 2). In order to understand this behavior, a more detailed investigation of the magnetic properties is required.

The obtained magnetization data were evaluated further and transferred into a pinning force analysis, where the pinning force $F_p = j_c \times B$ is plotted versus a scaling field $h = H_a/H_{irr}$, with H_{irr} denoting the irreversibility field [Figs. 3 (a,b)]. This analysis revealed for the sample prepared at 950 °C a well developed scaling of the normalized pinning force data at low and intermediate temperatures with the scaling parameters $p = 0.65$, $q = 1.35$ and a peak located at $h_0 = 0.32$. Such a peak position would indicate pinning at normal conducting precipitates according to the model of Dew-Hughes (DH) [12]. Similar values were obtained in the literature for small, pure MgB₂ samples. For a dominating flux pinning at the grain boundaries, $h_0 \sim 0.22$ would be expected, which is not the case here. In contrast to this behavior, the samples prepared at 775 and 800 °C reveal a clear non-scaling. As indicated in Fig. 2 (b), the resulting fit to all data cannot describe the situation. Further, there is a clear shape change of the F_p -curve itself, which was previously observed in thin film samples [13]. At higher T , the peak position shifts to $h_0 = 0.4$, and at low T , a peak position $h_0 < 0.2$ is reached. Such small peak positions cannot be explained by the common scaling model, except

that the anisotropy γ plays a dominating role. This effect was discussed previously in Ref. [14]. The calculation of j_c using the parameters $\gamma = 4$ and a percolation, $p_c = 0.25$, produces a shift of the peak position from 0.33 (isotropic case) to about 0.18. This further implies that to deduce the acting pinning mechanism from the peak position of the pinning force scaling, the anisotropy and percolation should be known.

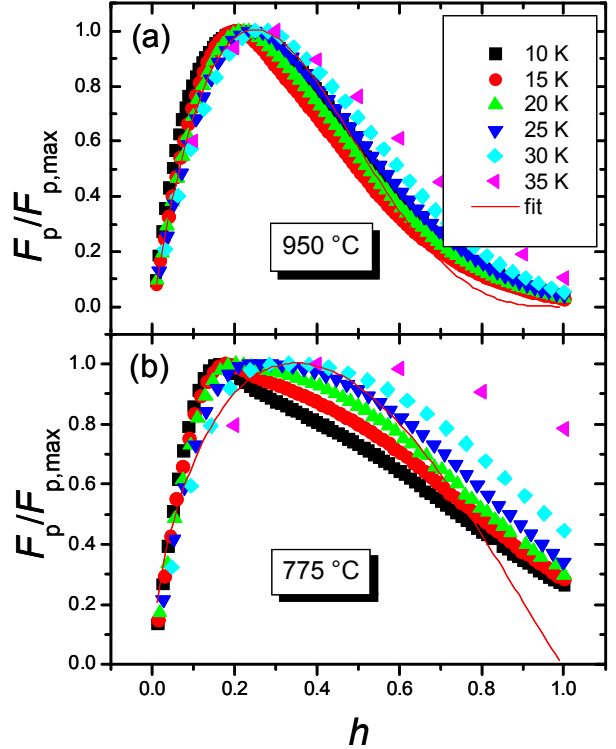


Fig. 3. (a,b) Pinning force scaling for the sample prepared at 950 °C (a) and 775 °C (b). The shown fits are fits to all data. The strong non-scaling of (b) is clearly visible.

In Fig. 4, the resulting peak positions h_0 are shown as a function of temperature. Only the data of the sample prepared at 950 °C exhibits a plateau which is a prerequisite for the scaling, whereas the data of the other samples are continuously increasing with increasing temperature.

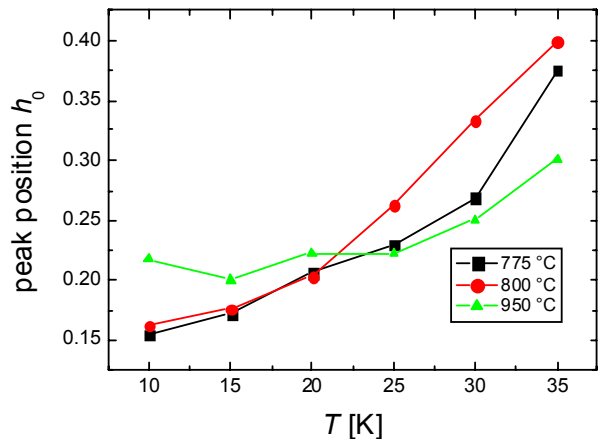


Fig. 4. The peak positions of the pinning force scaling, h_0 , as a function of

temperature.

The data of Fig. 4 indicate that the grain boundary pinning is significant only at low temperatures, whereas at higher temperatures, the flux pinning at normal-conducting precipitates plays the dominating role. As this conclusion also holds after applying the modified scaling procedure by employing the scaling field, H_n , (see Fig. 5 below), this finding is an important one for the further development of MgB₂ bulk magnets. A different scaling procedure was proposed in Ref. [14], where a field H_n is employed instead of H_{irr} . H_n is defined as the field where the pinning force F_p drops to 50% of its value above the peak position. The advantage of using this field for scaling is that an area just below $h = 1$ is not so sensitive to the effective values of γ and p_c . Therefore, Fig. 5 presents the results of this modified scaling procedure.

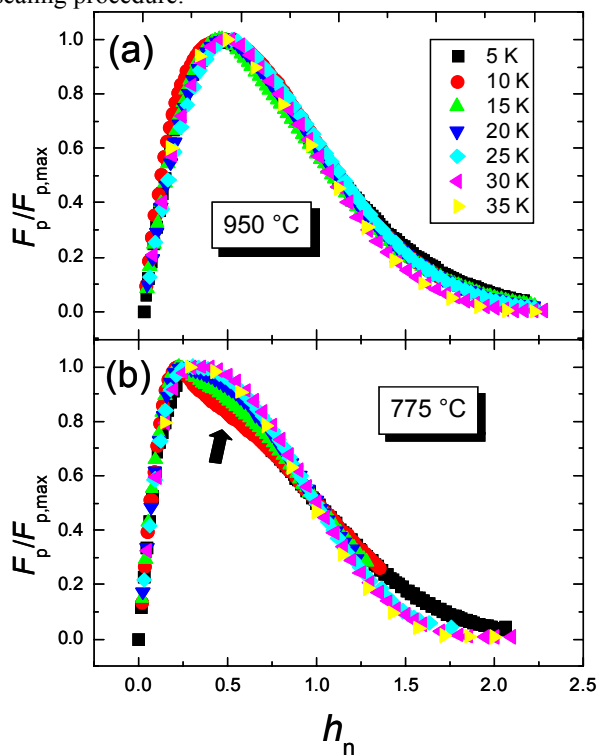


Fig. 5. Modified scaling procedure using a scaling field, h_n , [12] for the sample prepared at 950 °C (a) and at 775 °C (b). The arrow in (b) indicates the shape change at low temperatures.

Also in these graphs, the temperature dependence of the peak position is still visible. This implies that either γ and/or p_c are changing with increasing the reaction temperature. On the other hand, the anisotropy is normally changing with temperature [15], which implies that a given temperature, the value of p_c must be different. This effect is also observed in the resistance measurements (measured during field-cooled warming) as presented in Fig. 6. The magnetoresistance data reveal a much sharper superconducting transition for the sample prepared at 950 °C, whereas the transitions broaden up on increasing the magnetic field for the sample prepared at 775 °C. This reflects the non-scaling of the pinning forces, as

the $R(T,B)$ curves correspond to the $M(T,B)$ behavior as discussed in Refs. [16,17].

Microstructural observations obtained from scanning electron microscopy (SEM) and atomic force microscopy (AFM) indicated that the grain size is the crucial parameter to improve the critical currents as well as the trapped field values. This confirms the results of previous works [18-20]. Figure 7 presents typical AFM topography images of the samples recorded in the tapping mode. The images clearly reveal the polycrystalline nature of the sample. Individual MgB₂ grains show a specific crystallite shape and the dimensions of the grains are up to 400 nm. Obviously, the grain size and grain arrangement of (a) represents an optimal configuration, thus leading to the high trapped field values. The increase of the trapped fields for the sample prepared at 950 °C is attributed to the formation of nano-sized MgB₄ particles at the higher reaction temperature, which can provide additional, strong flux pinning by $\Delta\kappa$ -pinning [21], but also may affect the current flow through the sample.

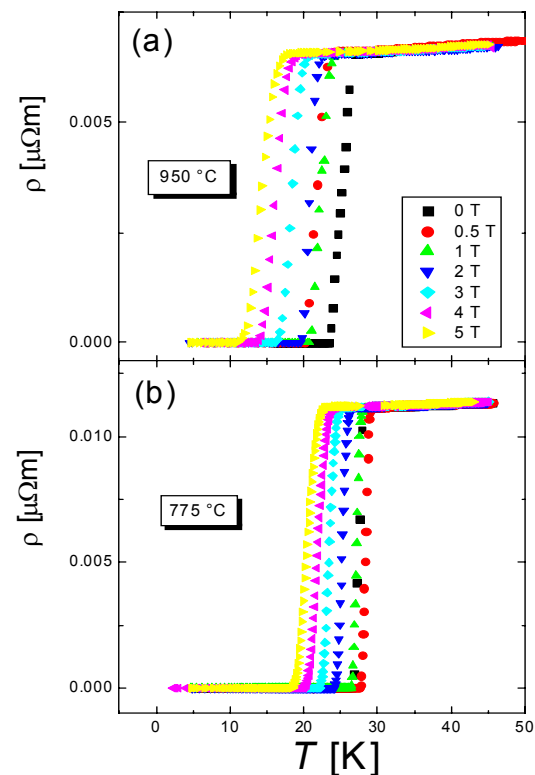


Fig. 6. Specific resistance as function of temperature for various applied fields for the sample prepared at 775 °C (a) and at 950 °C (b).

Another important aspect for the development of bulk, MgB₂-based supermagnets is the energizing procedure applied. MgB₂ suffers – in contrast to the bulk RE-123 samples – from flux jumps and the specific flux penetration in form of dendrites, especially at low temperatures [22,23]. The microstructures shown in Fig. 7 directly confirm such a complicated current flow. The magnetization loops measured at 5 and 10 K with a sweep rate of 10 mT/s have clearly revealed this behavior. Therefore, for energizing MgB₂

supermagnets, it will be an essential task to develop a suited procedure, which may also include countermeasures like the application of a conductive thin-film cover onto the samples. A first attempt of such a procedure can be found already in Ref. [24], which indeed helps to reduce the number of flux jumps occurring in MgB₂ thin film samples.

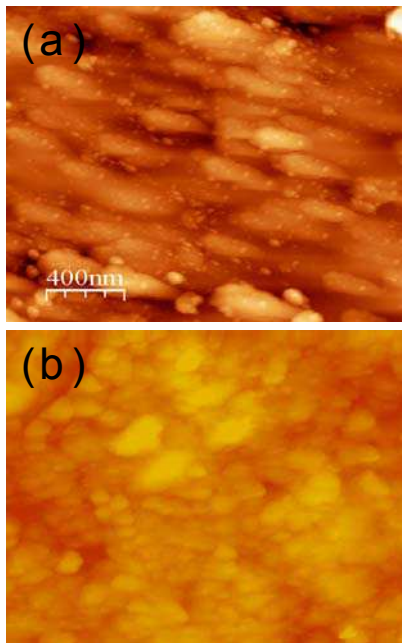


Fig. 7. AFM topography images (tapping mode) of the MgB₂ sample prepared at 775 °C (a) and of the sample prepared at 850 °C (b).

IV. CONCLUSION

To summarize, MgB₂ samples of 20 mm in diameter and 7 mm thickness, prepared with optimized processing temperature, exhibited a trapped field of ~1.5 T at 20 K. The characterization of small samples showed a change of flux pinning mechanisms as seen in the pinning force scaling, depending on the preparation temperature. 775 °C was found to be the optimum temperature to reach high trapped fields, while the sample prepared at 950 °C exhibits additional flux pinning provided by MgB₄ particles.

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REFERENCES

- [1] K. Vinod, R. G. Abhilash Kumar and U. Syamaprasad, "Prospects for MgB₂ superconductors for magnet application", *Supercond. Sci. Technol.*, vol. 20, pp. R1-R13, 2007.
- [2] M. Tomsic, M. Rindfleisch, J. Yue, K. McFadden, J. Phillips, M.D. Sumption, M. Bhatia, S. Bohnenstiehl, E.W. Collings, "Overview of MgB₂ Superconductor Applications", *Int. J. Appl. Ceram. Technol.*, vol. 4, pp. 250–259, July 2007.
- [3] P. Mikheenko, E. Martinez, A. Bevan, J. S. Abell, and J. MacManus-Driscoll, "Grain boundaries and pinning in bulk MgB₂", *Supercond. Sci. Technol.*, vol. 20, pp. S264-S270, 2007.
- [4] E. Martinez, M. Martinez-Lopez, A. Millan, P. Mikheenko, A. Bevan and J. S. Abell, "Temperature and magnetic field dependence of the n-values of MgB₂ superconductors", *IEEE Trans. Appl. Supercond.*, vol. 17, pp. 2738-2741, Jun. 2007.
- [5] E. Martinez, P. Mikheenko, M. Martinez-Lopez, A. Millan, A. Bevan and J. S. Abell, "Flux pinning force in bulk MgB₂ with variable grain size", *Phys. Rev. B*, vol. 75, p. 134515, 2007.
- [6] S. X. Dou, S. Soltanian, J. Horvat, X. L. Wang, S. H. Zhou, M. Ionescu, H. K. Liu, P. Munroe and M. Tomsic, "Enhancement of the critical current density and flux pinning of MgB₂ superconductor by nanoparticle doping", *Appl. Phys. Lett.*, vol. 81, pp. 3419-3421, 2002.
- [7] A. Yamamoto, J. Shimoyama, S. Ueda, Y. Katsura, S. Horii and K. Kishio, "Improved critical current properties observed in MgB₂ bulks synthesized by low-temperature solid-state reaction", *Supercond. Sci. Technol.*, vol. 18, pp.116-121, 2005.
- [8] G. Giunchi, G. Ripamonti, T. Cavallin and E. Bassani, "The reactive liquid Mg infiltration process to produce large superconducting bulk MgB₂ manufactures", *Cryogenics*, vol. 46, pp. 237-242, 2006.
- [9] J.H. Durrell, C.E.J. Dancer, A. Dennis, Y. Shi, Z. Xu, A.M. Campbell, N. Hari Babu, R.I. Todd, C.R.M. Grovenor and D.A. Cardwell, "A trapped field of >3 T in bulk MgB₂ fabricated by uniaxial hot pressing", *Supercond. Sci. Technol.*, vol. 25, p. 112002, 2012.
- [10] T. Naito, T. Sasaki, and H. Fujishiro, "Trapped magnetic field and vortex pinning properties of MgB₂ superconducting bulk fabricated by a capsule method", *Supercond. Sci. Technol.*, vol. 25, p. 095012 (2012).
- [11] M. Muralidhar, K. Inoue, M.R. Koblischka, M. Tomita, and M. Murakami, "Optimization of processing conditions towards high trapped fields in MgB₂ bulks", submitted.
- [12] D. Dew-Hughes, "Flux pinning mechanisms in type II superconductors", *Philos. Mag.*, vol. 30, pp. 293-305, 1974.
- [13] S.G. Jung, W. K. Seong and W. N. Kang, "Flux pinning mechanism in single-crystalline MgB₂ thin films", *J. Phys. Soc. Jpn.*, vol. 82, p. 114712, 2013.
- [14] M. Eisterer, "Calculation of the volume pinning force in MgB₂ superconductors", *Phys. Rev. B*, vol. 77, p. 144524, 2008.
- [15] M. Eisterer, "Magnetic properties and critical currents of MgB₂", *Supercond. Sci. Technol.*, vol. 20, pp. R47-R74, 2007.
- [16] M.R. Koblischka, M. Muralidhar, T. Higuchi, K. Waki, N. Chikumoto and M. Murakami, "Superconducting transitions of Nd-based 123 superconductors in fields up to 7 T", *Supercond. Sci. Technol.*, vol. 12, pp. 288-292, 1992.
- [17] A.K. Pradhan, M. Muralidhar, M.R. Koblischka, M. Murakami, K. Nakao and N. Koshizuka, "Evidence of strong flux pinning in melt-processed ternary (Nd-Eu-Gd)Ba₂Cu₃O_y superconductors", *Appl. Phys. Lett.*, vol. 75, pp.253-255, July 1999.
- [18] M. Muralidhar, A. Ishihara, K. Suzuki, Y. Fukumoto, Y. Yamamoto and M. Tomita, "Optimization of the fabrication process for high trapped field MgB₂ bulks", *Physica C*, vol. 494, pp. 85-88, 2013.
- [19] M. Muralidhar, Y. Fukumoto, A. Ishihara, K. Suzuki, M. Tomita, M.R. Koblischka, A. Yamamoto and K. Kishio, "Recent developments in melt processed Gd-123 and MgB₂ materials at RTRI", *Physica C*, vol. 496, pp. 5-10, 2014.
- [20] G. Fuchs, W. Häbler, K. Nenkov, J. Scheiter, O. Perner, A. Handstein, T. Kanai, L. Schultz and B. Holzapfel, "High trapped fields in bulk MgB₂ prepared by hot-pressing of ball-milled precursor powder", *Supercond. Sci. Technol.*, vol. 26, p. 122002, 2012.
- [21] Q. Cai, Y. Liu, Z. Ma, H. Li and L. Yu, "Variation of pinning mechanism and enhancement of critical current density in MgB₂ bulk containing self-generated coherent MgB₄ impurity", *Appl. Phys. Lett.*, vol. 103, p. 132601, 2013.
- [22] J.I. Vestgård, Y.M. Galperin and T.H. Johansen, "Dendritic flux avalanches in rectangular superconducting films – Numerical simulations", *Physica C*, vol. 479, pp. 92-94, 2012.
- [23] D. V. Denisov, D. V. Shantsev, Y. M. Galperin, E.-M. Choi, H.-S. Lee, S.-I. Lee, A. V. Bobyl, P. E. Goa, A. A. F. Olsen, and T. H. Johansen, "Onset of dendritic flux avalanches in superconducting films", *Phys. Rev. B*, vol. 73, p. 014512, 2006.
- [24] C. Stahl, S. Treiber, G. Schütz and J. Albrecht, "The avalanche process in gold covered MgB₂ films", *Supercond. Sci. Technol.*, vol. 26, p. 015007, 2013.