Investigation of magnetic relaxation in MgB₂ wires

M Reissner¹, S Mohammad¹, P Kovac², I Husek² and T Melisek²

¹Institute of Solid State Physics, Vienna University of Technology, Wiedner Hauptstraße 8-10, A-1040, Wien, Austria
²Institute of Electrical Engineering, Centre of Excellence CENG, Slovak Academy of Science, Bratislava, Slovakia

E-mail: reissner@ifp.tuwien.ac.at

Abstract. MgB₂ wires prepared by combining ex-situ and in-situ routes in different ratios were investigated by magnetic relaxation measurements. The obtained mean effective activation energies are much larger than in high temperature superconductors. Differences in critical current densities are mainly due to increased number of pinning centers and not to changes in pinning energy.

1. Introduction

There are two main routes to prepare MgB₂ wires: in-situ and ex-situ [1, 2]. Whereas in the in-situ route powders of Mg and B are filled into metallic sheaths, in the ex-situ route MgB₂ powder is used for filling. One advantage of the in-situ process is that lower reaction temperatures (640 to 850°C) [3, 4] can be used for annealing in contrast to the ex-situ process for which annealing at around 950°C is necessary to obtain good grain connectivity. The disadvantage of the in-situ process is the low core density, which is due to the fact that the volume of MgB₂ is approximately 30% lower than the volume of the Mg+B powder mixture, whereas for the ex-situ process densities up to 90% of the theoretical one can be reached.

Substitution of boron by carbon was shown to increase critical current density dramatically. This substitution is best performed by addition of SiC [5].

A combination of both methods by preparation of wires from mixtures of MgB₂ with Mg and B in different ratios was shown to improve critical current density [6].

2. Experimental

Mg and B powder in ratio 1:2 mixed with different amount of MgB₂ (0, 23, 50, and 70%) plus 10 wt% SiC were mixed for 40 minutes in a planetary ball mill under Ar atmosphere. For comparison a pure ex-situ sample (100%) with 10 wt% SiC was prepared. The powders were filled into Ti tubes (5.8 mm out and 4.1 mm inner diameter) and deformed by rotary swaging to 3.5 mm followed by two-axial rolling to rectangular wires (2 x 2 mm²). In the next step the wires were inserted into a Cu tube (3 mm diameter) and two axially rolled to a cross section of 1.2 x 1.2 mm². Ti was used as barrier between superconducting core and Cu sheath. The obtained average core size was 0.53 x 0.53 mm². After rolling the samples were heat treated at 850°C for 30 min.
Dc magnetic measurements were carried out in a vibrating sample magnetometer from Quantum Design (PPMS-9T) in the temperature range 4.2 K to $T_c$ in fields up to 9 T. Samples with 3 mm length were measured with applied field parallel to the wire axis. Hysteresis loops were recorded at different temperatures with a field sweep rate of 60 Oe/s. At 1, 3 and 5 T both on the increasing and decreasing branch of the hysteresis the field sweep was interrupted and the change of magnetic moment monitored for up to 30 min.

3. Results

3.1. Relaxation measurements
Within the chosen time interval and the accuracy of the measurements the recorded decay of magnetization is in first approximation logarithmic for all samples and fields investigated (figure 1). This allows the determination of a creep rate $dM/d\ln t$. From the normalized creep rate $S = (dM/d\ln t)/M_{irr}$ with the irreversible part of the magnetization $M_{irr} = M - M_{rev}$ and the reversible one determined as the middle of the hysteresis loop $M_{rev} = (M_{up} - M_{down})/2$ a mean effective activation energy $<E>$ can be determined within the flux creep theory of Anderson [7, 8]. In this theory a linear relation between $<E>$ and the true pinning potential height $E_0$ is assumed according to $<E> = E_0 - |F/VX$, with $V$ the flux bundle volume and $X$ the hopping distance. The driving force $|F|$ is determined by the applied field and the field gradient in the sample and is proportional to the current density. A rather flat temperature dependence of $<E>$ is obtained at all three fields, with a small maximum shifting to higher temperatures for higher fields. Within the measuring accuracy no difference in the value of the mean effective activation energy is obtained for the different samples (figure 2). As expected $<E>$ decreases with increasing field.

![Figure 1. Typical relaxation curves for the investigated samples.](image)

3.2. Hysteresis measurements
Within the critical state model [9] critical current densities were determined by using the Bean formula for rectangular shaped samples [10] $J_c(A/cm^2) = 40|M_{irr}|/[b(1-b/3a)]$ with $b$ the width and $a$ the length of the sample. In the analysis the full core dimension ($a = b = 0.53$ mm) was used as the characteristic length scale for the supercurrent flow, because it is well known that MgB$_2$ behaves in a non-granular way [11]. Typical curves for the field dependence of $J_c$ are shown in figure 3 for 4.2 K. In contrast to
the results of the relaxation measurements a clear difference in the field dependence of the critical current density for the different samples is found. Up to 8 T the highest values are found for the sample with 23% ex-situ contribution. At higher fields the pure in-situ compound has the higher $J_c$ values. The sample with 70% ex-situ contribution has high $J_c$ values only up to 3 T. The pure ex-situ compound (100%) shows very bad performance ($J_c$ outside of scale in figure 3). In figure 4 the temperature dependence of the critical current density for all samples measured at 1.5 and 5.5 T is shown. Up to 20 K best values are obtained for the 23% sample, followed by the 0% and 70% composition. Above 20 K the series changes and $J_c$ for the 70% sample has slightly higher values than the other two. $J_c$ for the pure ex-situ wire is extremely low. Similar results are obtained for the irreversibility line (inset figure 4), which is practically the same for the 0% and 23% sample. For the 70% wire $B_{irr}$ is lower at low temperatures and higher above 25 K.

Figure 2. Temperature dependence of the mean effective activation energy at 1, 3, and 5 T for the investigated samples.

4. Discussion

A strong influence of different ex-situ admixture on the critical current density is obtained. Whereas addition of 23% (50%) of MgB$_2$ shows a strong (weak) improvement compared to the pure in-situ compound, 70% admixture gives only at high fields slightly larger $J_c$ values. The 100% sample has very low current density. This is due to the fact that the chosen annealing temperature of 850°C is by about 100°C too low for a pure ex-situ process. Recrystallization and formation of good grain connectivity is not sufficient [12, 13]. Higher annealing temperatures are not possible, because of the low lying eutectic reaction temperature for Ti-Cu (885°C). A change of $J_c$ can be caused by an increase in strength of the active pinning centers or by an increase in their number. As Eisterer [14] has pointed out care has to be taken in interpretation of the field dependence of $J_c$, because at higher fields the upper critical field plays a more important role than the pinning. With the exception of the pure ex-situ compound for the other samples the irreversibility line is very similar. The samples with 0 and 23% have identical irreversibility line. For the 70% sample the irreversibility line is slightly more flat. From this it can be concluded that the upper critical field will not change drastically. In contrast to the behaviour of $J_c$ the mean effective activation energy is within the measuring accuracy the same for the samples with 0, 23 and 70% ex-situ admixture. The values are much higher than the typical ones found for high temperature superconductors in the same field and temperature range [15].
In conclusion, with the exception of the pure ex-situ compound the pinning centers responsible for the current carrying properties of the samples with in- and ex-situ mixture are nearly identical. Changes in critical current density seem to be mainly caused by an increase in number of pinning centers and not by an improvement of the pinning strength.

References
[1] Glowacki B, Majoros M, Vickers M, Evetts J E, Shi Y and McDougall I 2001 Supcond. Sci. Technol. 14 193
[2] Grasso G, Malagoli A, Ferdeghini C, Roncallo S, Braccini V, Cimberle M R and Siri A S 2001 Appl. Phys. Lett. 79 230
[3] Dou X, Horvath J, Soltanian S, Wang X L, Qin M J, Zhou S H, Liu H K and Munroe P G 2003 IEEE Trans. on Appl. Supercond. 13 3199
[4] Fu B Q, Feng Y, Yang G, Liu C F, Zhou L, Cao L Z, Ruan K Q and Li X G 2003 Physica C 392-396 1035
[5] Dou S X, Pan A V, Qin M J and Silver T 2005 Frontiers in Supconducting Materials (Springer)
[6] Kov P, Husek I, Melisek T, Reissner M and Mohammad S 2008 in preparation
[7] Anderson P W 1962 Phys. Rev. Lett. 9 309
[8] Anderson P W and Kim Y B 1964 Mod. Phys. 36 39
[9] Bean C P 1962 Phys. Rev. Lett. 8 250
[10] Chen D X and Goldfarb B 1989 J. Appl. Phys. 66 2489
[11] Larbalestier D C et al 2001 Nature (London) 410 186
[12] Suo H L, Beneduce C, Dhallé M, Musolino N, Genoud J E and Flükiger R 2001 Appl. Phys. Lett. 79 3116
[13] Kovac P, Husek I and Melisek T 2002 Supercond. Sci. Technol. 15 1340
[14] Eisterer M 2007 Supercond. Sci. Technol. 20 R47-R73
[15] Reissner M, Ambrosch R and Steiner W 1991 Supercond. Sci. Technol. 4 S436