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EFFECT OF ANNEALING TEMPERATURE ON MAGNESIUM DEFICIENT IN (MgB_2) SUPERCONDUCTOR

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ABSTRACT

The Mg-deficient $Mg_{0.8}B_2$ superconductors were prepared using the solid-state reaction method at different annealing temperature between 650 °C and 800 °C. The x-ray diffraction patterns indicated that magnesium diboride (MgB_2) is a major phase and magnesium oxide (MgO) is the secondary phase. However, no unreacted Mg was detected by XRD at all annealing temperatures. The SEM images showed hexagonal grain structures with nano thickness distributions. The highest critical current density (J_c) at 5 K and 20 K was found in sample annealed at 700 °C. At 5 K and 6 T, the highest J_c achieved was 1.2×10^4 A/cm^2. At 20 K, the highest J_c achieved by 3.6×10^3 A/cm^2. The values of the onset superconducting transition temperature, T_c for all $Mg_{0.8}B_2$ were 37.0 K. The samples annealed at 700° C showed a sharper diamagnetic drop with $\Delta T_c = 3.0$ K compared to all samples.

Keywords: superconductors; MgB_2; Mg-deficient; solid-state reaction method;

INTRODUCTION

The discovery of superconductivity in MgB_2 with a transition temperature T_c of 39 K by Akimitsu has renewed the interest in metal borides [1]. Unlike cuprates, MgB_2 is an intermetallic compound with low contact resistance between the grain boundaries, eliminating the weak-link problem [2]. The primary advantage of MgB_2 over high temperature yttrium and bismuth based materials is due to its large coherence length (~50 Å) compared to 3-5 Å for high temperature superconductors (HTSC) [3]. MgB_2 crystal consists of hexagonal (AlB2 type, space group P6/mmm) honey-combed planes of boron atom separated by planes of magnesium atom. In spite of the chemical and structural simplicity, Fundamental studies of MgB_2 have proven to be very challenging [4-5]. Large difference between the melting point of B (2076 °C) and Mg (650 °C), B has limited solubility in melted Mg [5]. Although B is more soluble in melted Mg at higher temperature. Evaporation rate of Mg is very high at this temperature. In this work, $Mg_{0.8}B_2$ were prepared and their superconducting properties were compered in a wide range of temperature (650 °C – 800 °C).

EXPERIMENTAL

Polycrystalline samples with the nominal composition $Mg_{0.8}B_2$ were prepared via the conventional solid state reaction technique. The starting powders are Magnesium 99% (<10μm) from TangShan Weihao Magnesium Co Ltd. and amorphous boron powder (<1μm) from Pfaltz and Bauer. Appropriate amount of Mg and B powders were mixed and ground using a agate mortar. Pellets with 13 mm diameter were made using 5 tonnes of pressure, sealed in an iron tube and annealed between 650 and 800 °C for 1 hour, in flowing high purity Argon gas to minimize oxidation. The structural and phase analysis of the samples were done using X-ray diffractometer (Philips PW 3040/60 X'pert Pro) with CuKα radiation (wavelength of 1.5405 Å). Phase identification of the samples was performed using the X'Pert Highscore software with the support of ICDD-PDF-2 database. Lattice parameters were calculated using the X'pert Plus software. The superconducting transition temperature was determined by AC susceptibility measurement (Quantum Design, Physical Property Measurement System (PPMS)). Microstructure analysis was done using the JOEL: JSM-6400 Scanning Electron Microscope (SEM)

RESULT AND DISCUSSION

Figure 1 shows the SEM images of the $Mg_{0.8}B_2$ samples prepared at 650°C, 700°C, 750°C and 800°C, respectively. The samples exhibited some hexagonal-shaped crystals. Most of the crystals were found with typical edge angle of 120° and flat surfaces. The larger crystal is about 600 nm in diameter (700 °C). The sample annealed at 750 °C shows smaller hexagonal crystals of about 300 nm diameter. The crystals in $Mg_{0.8}B_2$ sample annealed at 750 °C also looks more compact compared to others. The smooth surface and sharp edges confirmed that samples are of high crystallinity [6].

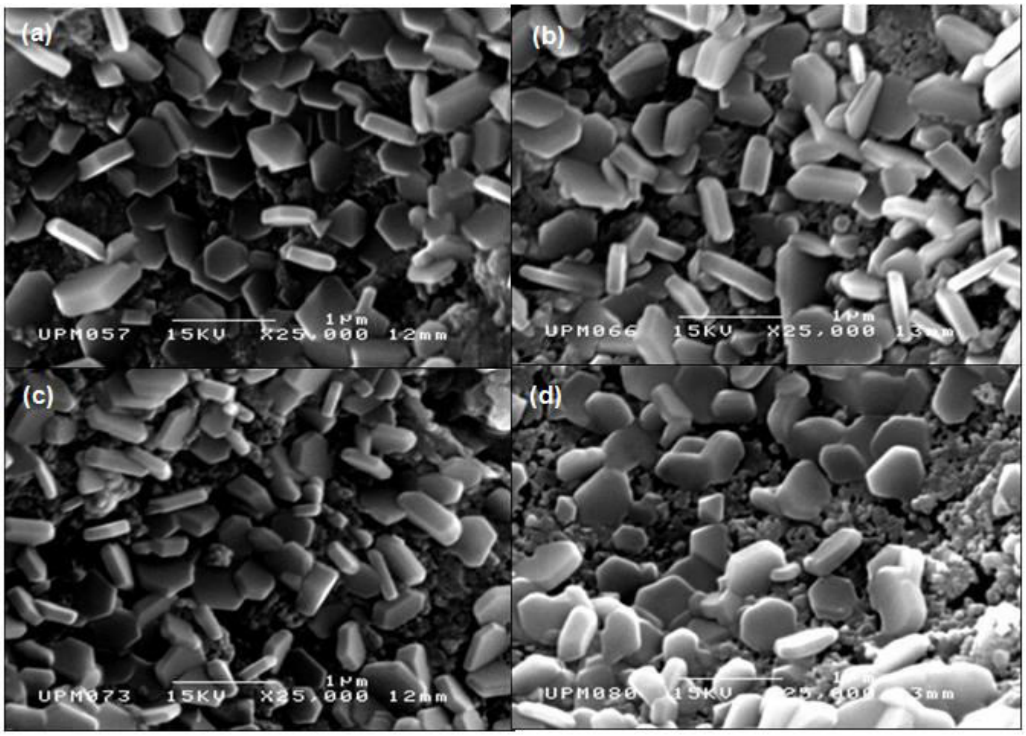

Figure 1: SEM images of $Mg_{0.8}B_2$ annealed at (a) 650 °C (b) 700 °C (c) 750 °C (d) 800 °C

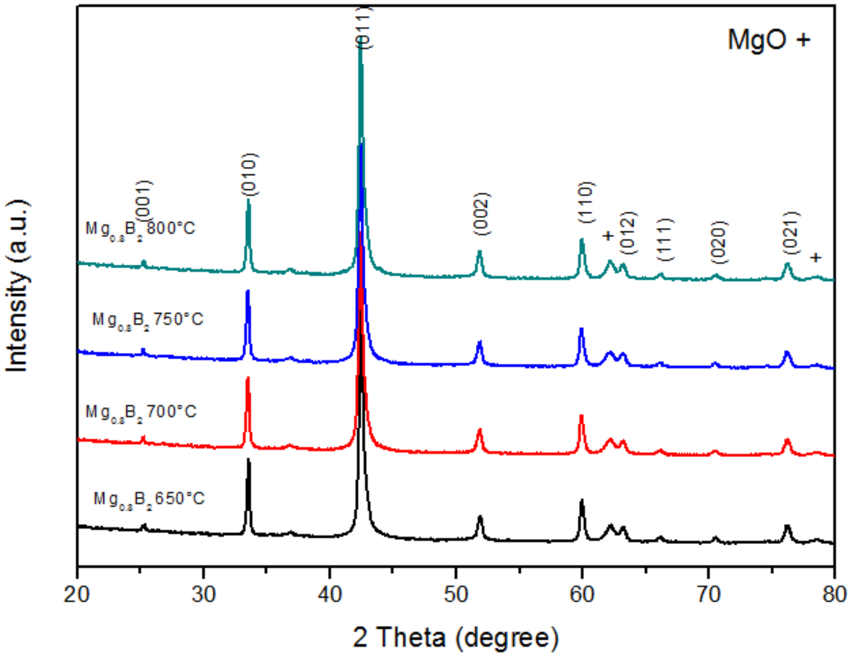

Figure 2: XRD patterns of $Mg_{0.8}B_2$ annealed at various temperature

When the temperature exceeded the melting point of pure Mg, the residual Mg powders began to melt and parts of them evaporated to generate Mg vapour. At this stage, the molten Mg not only reacts with the residual B to form MgB_2 unceasingly, but it also react trace with oxygen to form MgO. It is well known that the starting powders may contain a moderate amount of oxygen which cannot be completely degassed. Other sources of oxygen for oxidation can be: (1) adsorption of oxygen physically on to the surface of Mg and B powders during the heat treatment (2) the leakage of air into the reaction system through the joint of the furnace [7]. Figure 2 shows the XRD patterns of the $Mg_{0.8}B_2$ annealed between 650 °C to 800 °C. Table 1 shows the volume fraction of MgB_2 and MgO for the samples annealed between 650 °C to 800 °C. It can be seen that MgO increases when the annealing temperature is reduce. It has been shown before that Mg-deficient causes lattice distortion on MgB_2 and creates pinning centres from the structural defects leading to enhanced J_c [8].

Table 1: Volume fraction of $Mg_{0.8}B_2$ annealed at various temperature

Anneling Temperature	MgB_2 (%)	MgO (%)
650°C	75.8(6)	24.2(4)
700°C	76.9(6)	23.1(5)
750°C	77.7(6)	22.3(4)
800°C	77.3(6)	22.7(4)

Figure 3 shows a summary of Rietveld refinement data for $Mg_{0.8}B_2$ annealed between 650 °C to 800 °C. Overall, *a*-axis decreases and and *c*-axis increases. The unit cell volume of the sample decreases slightly overall. The very small change in the lattice parameters may be due to the lattice strain induced by the annealing temperature.

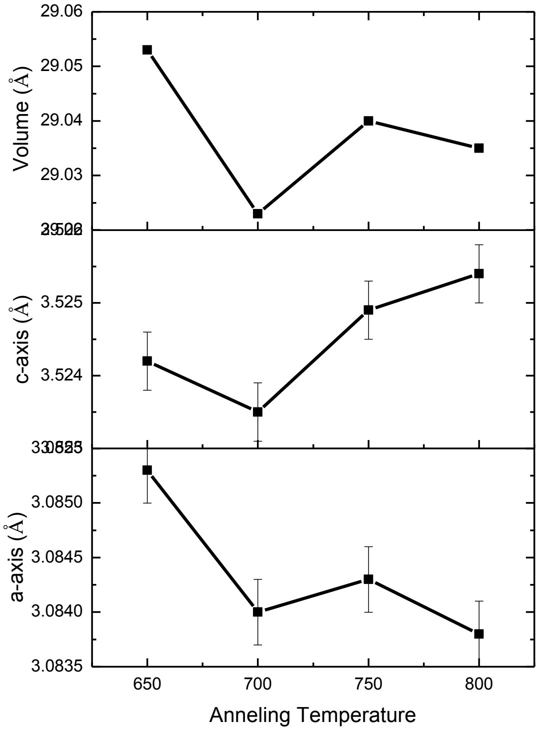

Figure 3: Lattice parameters of *a*-axis and *c*-axis of $Mg_{0.8}B_2$ annealed at different temperatures

Figure 4 and Table 2 shows the effect of annealing temperature on the J_c of $Mg_{0.8}B_2$ at 5 K and 20 K. At 5 K and 6 T, the highest J_c was achieved in $Mg_{0.8}B_2$ annealed at 700°C (1.2×10^4 A/cm^2), at 20 K, the trend for the J_c improvement is similar to that to that at 5 K. At 20K, 4T, the highest J_c achieved in the sample annealed at 700 °C is 3.6×10^3 A/cm^2, followed by the samples annealed at 800 °C, 750 °C and 650 °C with J_c of 2.8×10^3 A/cm^2, 2.7×10^3A/cm^2 and 2.6×10^3 A/cm^2, respectively.

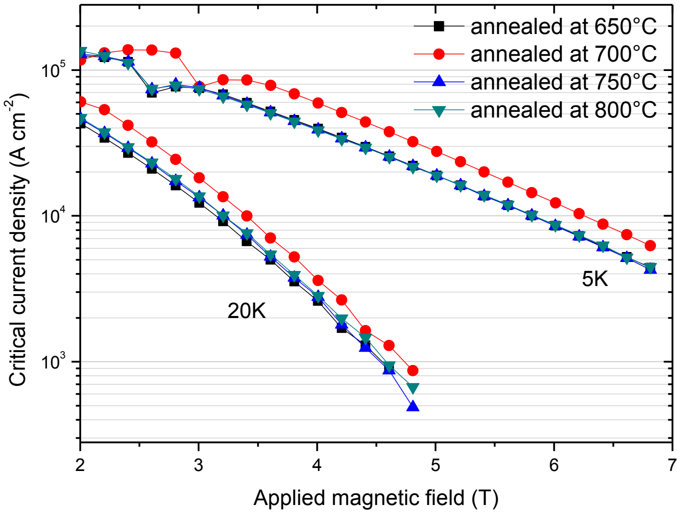

Figure 4: Critical current density as a function of applied magnetic field at 5 K and 20 K for $Mg_{0.8}B_2$ annealed at various temperatures

Table 2: Comparison of $T_{c(zero)}$, $T_{c(onset),}$ ΔT_c and J_c at 5K and 20K for $Mg_{0.8}B_2$ annealed at between 650° C - 800° C

Annealing temperature	$T_{c\ (zero)}$ (K)	$T_{c\ (onset)}$ (K)	ΔT_c (K)	J_c (A/cm^2) (5K)		J_c (A/cm^2) (20K)	
650°C	34.0	37.0	3.0	3 T	7.5×10^4	2 T	4.2×10^4
				5 T	1.8×10^4	3 T	1.2×10^4
				6 T	8.5×10^3	4 T	2.6×10^3
700°C	34.0	37.0	3.0	3 T	7.6×10^4	2 T	6.0×10^4
				5 T	2.7×10^4	3 T	1.9×10^4
				6 T	1.2×10^4	4 T	3.6×10^3
750°C	32.0	36.8	4.8	3 T	7.5×10^4	2 T	4.6×10^4
				5 T	1.8×10^4	3 T	1.3×10^4
				6 T	8.5×10^3	4 T	2.7×10^3
800°C	34.0	37.0	3.0	3 T	7.4×10^4	2 T	4.6×10^4
				5 T	1.8×10^4	3 T	1.3×10^4
				6 T	8.5×10^3	4 T	2.8×10^3

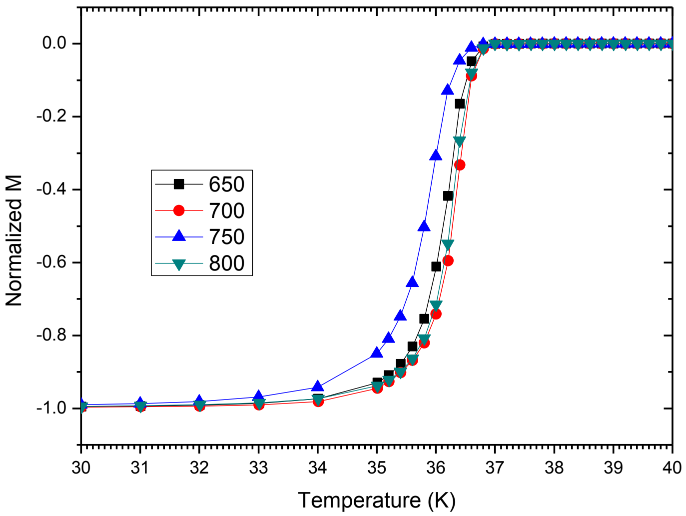

Figure 5: AC susceptibility measurement of $Mg_{0.8}B_2$ annealed at various temperatures

Figure 5 shows the normalised temperature dependence of the magnetic moment measured under 0.1 Oe for $Mg_{0.8}B_2$ annealed at various temperatures. The *M(T)* curves show one step superconducting transition below the onset of diamagnetism. The superconducting transition temperature $T_{c(0)}$, for the samples annealed at 650 °C, 700 °C and 800 °C at 37.0 K, and 750° C is 36.8 K. A sharp drop, in T_c indicates a homogeneous superconducting phase in a sample [9].

CONCLUSIONS

At 5 K and 20 K, J_c was enhanced when the $Mg_{0.8}B_2$ sample was annealed at 700 °C. All samples showed a sharp diamagnetic transition at around 37.0 K. The XRD results showed that there was no unreacted Mg for all the samples. MgO exists as impurity phase due to oxidation. The SEM images showed a hexagonal grain structures with nano thickness distributions. Larger crystals of 600 nm in diameter were observed in the samples anneled at 700 °C. The $Mg_{0.8}B_2$ sample annealed at 750 °C showed smaller hexagonal crystals of about 300 nm in diameter. The hexagonal crystals in $Mg_{0.8}B_2$ annealed at 750 °C also look more compact compared to other samples. At 5 K and 6 T, the highest J_c was achieved in $Mg_{0.8}B_2$ annealed at 700°C sample with the value of 1.2×10^4 A/cm^2, At 20 K, the trend for the J_c improvement is similar to that at 5 K. At 20K, 4T, the highest J_c achieved in MgB_2 annealed at 700 °C is 3.6×10^3 A/cm^2, followed by the sample annealed at 800 °C, 750 °C and 650 °C with J_c of 2.8×10^3 A/cm^2, 2.7×10^3A/cm^2 and 2.6×10^3 A/cm^2, respectively.

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