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Mg STOICHIOMETRY STUDY ON MgB_2 AT LOW ANNEALING TEMPERATURES

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ABSTRACT

An in-situ reaction of Magnesium (Mg) and Boron (B) at 650°C annealing temperature was used to compare the phase formation of Magnesium Diboride (MgB_2) by varying the nominal Mg. The x-ray diffraction pattern indicates that Magnesium Oxide (MgO) is the major secondary phase. Some of the unreacted Mg phase was found in the nominal MgB_2 stoichiometry and the Mg-excessed samples annealed at 650°C. However, no unreacted Mg was detected by XRD for Mg-deficient sample annealed at 650°C. Highest enhancement of critical current density (J_c) at 5 K and 20 K is found in Mg-deficient samples treated at both annealing temperatures. J_c is increased for the over-added Mg samples as compared to that of the nominal samples. The SEM image show a hexagonal grain structures with nano thickness distributions.

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 boride [1]. MgB_2 is made of very light and cheap elements. 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 material is derived from 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, MgB_2 required fundamental study which has been proven to be very difficult [4-5]. The presence of oxygen leads to reaction with Mg at high temperature producing MgO as impurity which degrades the superconducting properties. The influence of temperature is critical as to maximise the phase formation of MgB_2. Also, there is a large difference in melting point between B (2076°C) and Mg (650°C) [5]. At high temperature, however, Mg tends to evaporate severely. In this paper, synthesis of Mg_xB_2 with varying x in an iron tube at different growth conditions is analyzed and compared.

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EXPERIMENTAL

Polycrystalline bulk samples 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. Samples were prepared according to Mg_xB_2 with x = 0.8, 1.0 and 1.2. Pellets with 13 mm diameter were made with 5 tonnes of pressure, sealed in an iron tube and annealed at 650°C for 1 hour, in a flowing high purity Argon gas to minimize the contamination from oxygen. The structural and phase analysis of the samples were performed 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 X'Pert Highscore software with the support of ICDD-PDF-2 database. Lattice parameter was calculated using X'pert Plus. Critical temperature (*Tc*) of the superconducting transition is determine using ac susceptibility measurement (Quantum Design Physical Property Measurement System (PPMS)). Microstructure analysis was done using the Scanning Electron Microscope (SEM) model JOEL: JSM-6400.

RESULT AND DISCUSSION

Figure 1 shows the critical current density of Mg stoichiometry in MgB_2 at 650 °C. $Mg_{0.8}B_2$ show highest J_c, followed by $Mg_{1.2}B_2$ and MgB_2 for 20 K and 5 K. Based on XRD results shown in Figure 3, there are large amount of MgO and Mg in the second phase of the sample. Impurities in the sample can act as pinning centres that cause higher J_c. At 5 K and 6 T, the highest Jc achived by $Mg_{0.8}B_2$ sample is 8.5×10^3 A/cm^2, while $Mg_{1.2}B_2$ and MgB_2 J_c are 6.2×10^3 A/cm^2 and 3.2×10^3 A/cm^2 respectively. At 20 K, the trend for the J_c improvement is similar as 5 K and at 4T the highest J_c achived by $Mg_{0.8}B_2$ sample is 2.6×10^3 A/cm^2, while the measured J_c for $Mg_{1.2}B_2$ and MgB_2 J_c are 1.3×10^3 A/cm^2 and 6.6×10^2 A/cm^2. No obvious differences were found between these three samples.

Table 1: Comparison of $T_{c(offset)}$, $T_{c(onset)}$, ΔT_c and J_c at 5 K and 20 K for Stoichiometric MgB_2 sintered at 650°C.

Sample	$T_{c(offset)}$	$T_{c(onset)}$	ΔT_c	J_c (A/cm^2) (5K)		J_c (A/cm^2) (20K)	
$Mg_{0.8}B_2$ sintered at 650° C	35.0	37.0	2.0	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
MgB_2 sintered at 650° C	36.8	37.8	1.0	3 T	4.8×10^4	2 T	-
				5 T	9.5×10^3	3 T	6.5×10^3
				6 T	3.2×10^3	4 T	6.6×10^2
$Mg_{1.2}B_2$ sintered at 650° C	32.0	36.6	4.6	3 T	6.5×10^4	2 T	3.6×10^4
				5 T	1.4×10^4	3 T	8.5×10^3
				6 T	6.2×10^3	4 T	1.3×10^3

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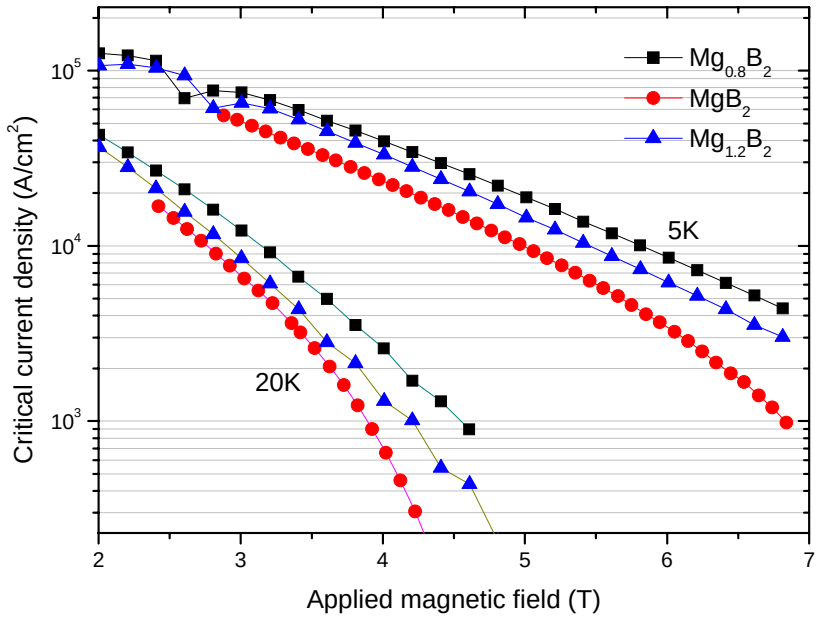

Figure 1: Critical current density as a function of applied magnetic field at 5 K and 20 K for Stoichiometric MgB_2 sintered at 650° C.

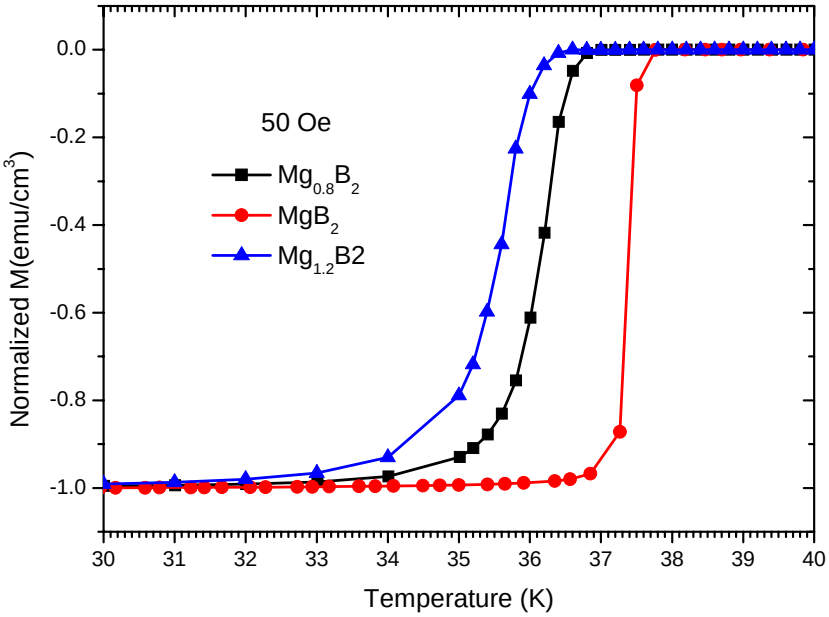

Figure 2: AC Susceptibility measurement on Stoichiometric MgB_2 superconductor sintered at 650°C

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Figure 2 shows the normalised temperature dependence of the zero-field-cooled (ZFC) magnetization M(T) measured under 50 Oe for stoichiometric MgB_2. The M(T) curve for all samples show one step superconducting transition below the onset of the demagnetization. In addition, the large diamagnetic signal shown in the M(T) curve indicates the quality of the samples and their superconducting property as well. The superconducting transition temperature $T_{c(0)}$ deduced from the figure for Mg_xB_2 (x=0.8, 1.0 and 1.2) samples spans between 37.5 K and 34.0 K. The value of the onset T_c for stoichiometric MgB_2 samples is 37.8 K. For $Mg_{0.8}B_2$ and $Mg_{1.2}B_2$ samples, $T_{c(onset)}$ value were recorded at 37.0 K and 36.6 K, respectively. MgB_2 shows a sharper drop as 1.0 ΔT_c compared to $Mg_{0.8}B_2$ at 2.0K ΔT_c and $Mg_{1.2}B_2$ at 4.6 ΔT_c. Excessive Mg decreased the T_c of MgB_2 [7].

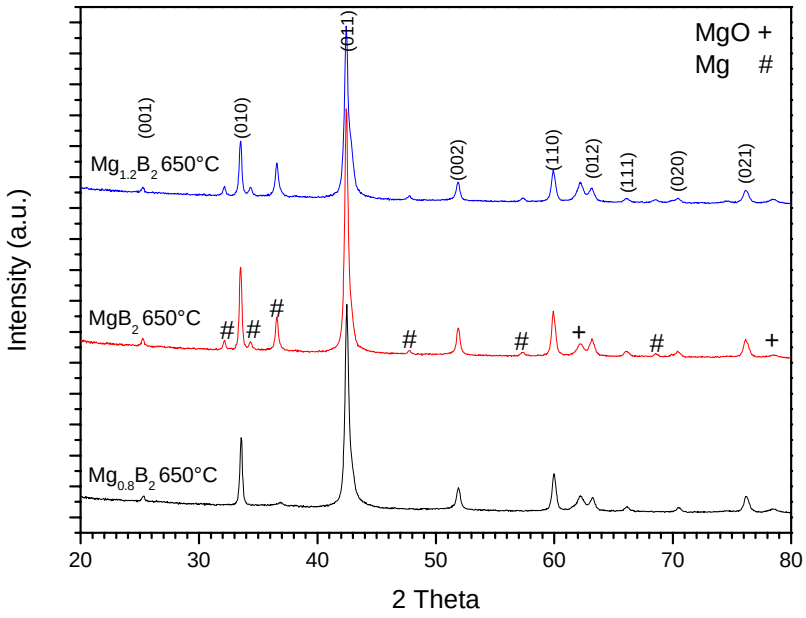

Figure 3: XRD pattern of Stoichiometry MgB_2 superconductor sintered at 650°C

Table 2: Weight fraction comparison of Stoichiometric MgB_2 superconductor sintered at 650°C.

	MgB_2 (%)	MgO (%)	Mg (%)
$Mg_{0.8}B_2$ 650°C	75.8(6)	24.2(4)	
MgB_2 650°C	78.9(5)	15.2(3)	5.9(2)
$Mg_{1.2}B_2$ 650°C	67.9(5)	24.3(3)	7.8(2)

Figure 3 shows a well developed MgB_2 which exhibit MgO phases in all samples, Table 2 tabulated. The figure also shows the presence of Mg (#) in MgB_2 and $Mg_{1.2}B_2$. Some impurity phases are present because of excess elements. It might be those impurity

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phases responsible in decreasing the superconducting properties. The existence of Mg as impurities phases due to the low sintering temperature and excessive amount of Mg in the stoichiometry resulted in complete reaction of Mg phase. Mg phase is however not present in lower $Mg_{0.8}B_2$ due to lower Mg content.

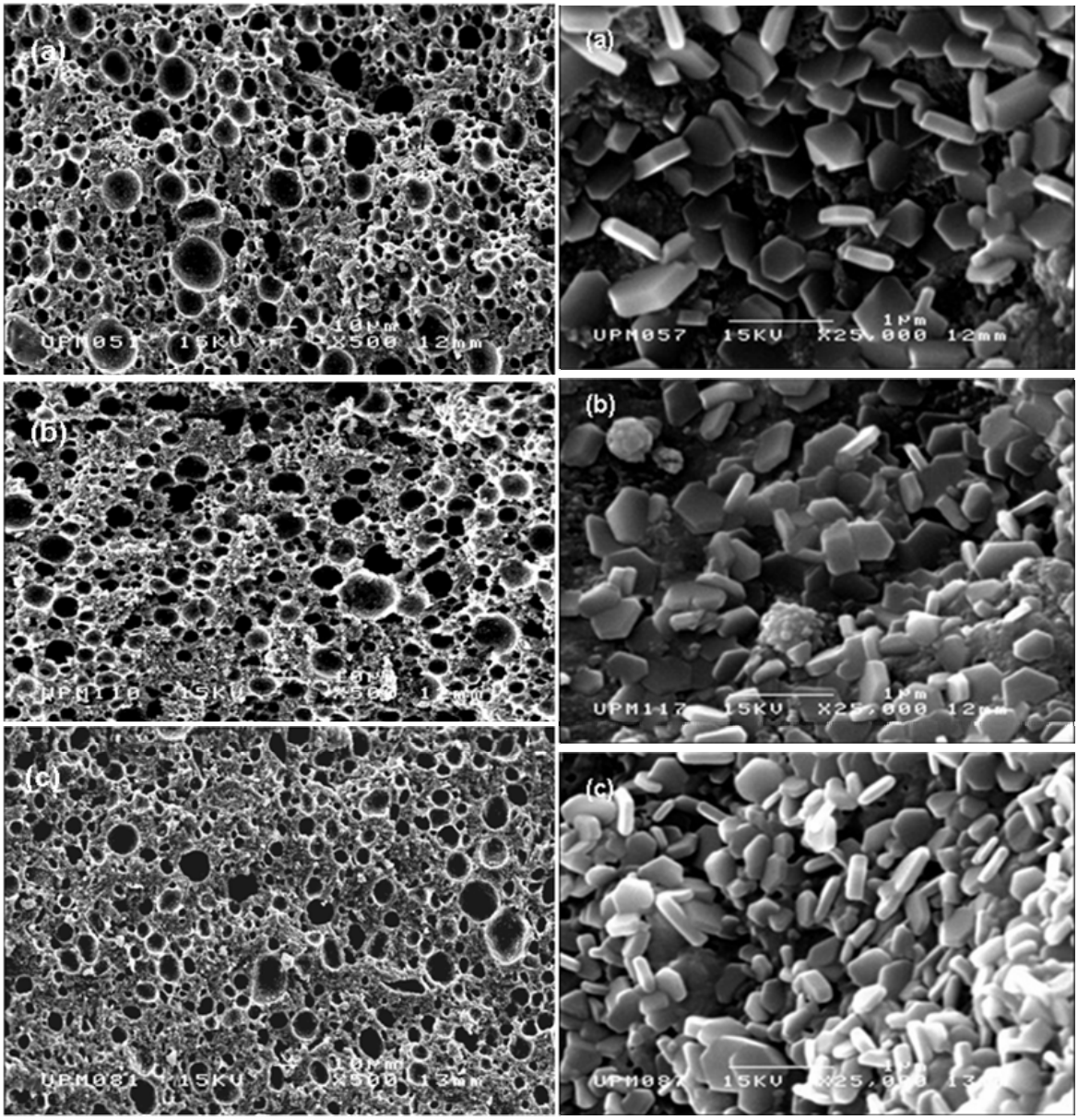

Figure 4: SEM Image of (a) $Mg_{0.8}B_2$ (b) MgB_2 (c) $Mg_{1.2}B_2$ superconductor sintered at 650°C view at X500 and X25000

The morphology of entire sample is very similar and has porous structure. The higher resolution image shows that single crystal exhibits well hexagonal grain structures with nano thickness destributions Typical hexagonal structure is documented on high-

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resolution images shown on the right panel of Figure 4. The stoney-shaped hexagonal grain distribute randomly along the surface plane in a more/less similar grain sizes for $Mg_{1.2}B_2$ sample, the grain size looks generally small compare to those for MgB_2 and $Mg_{0.8}B_2$ samples. The hexagonal surface, which was detected by X-ray energy dispersion (EDS), as shown in Figure 5 is composed of Mg, B and O.

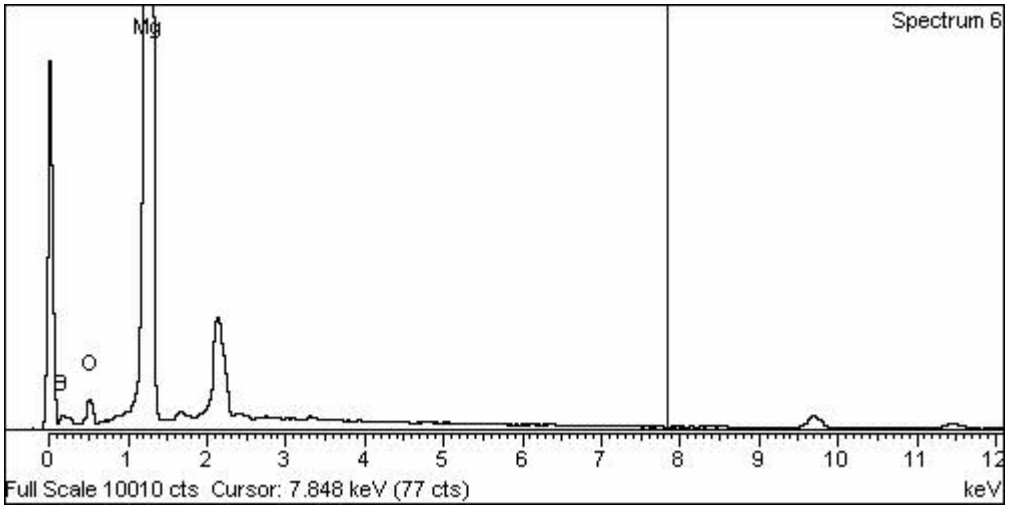

Figure 5: EDS of MgB_2 crystal surface.

CONCLUSION

We investigated Mg_xB_2 samples with $0.8<x<1.2$. These have shown that the most Mg deficient samples observe there is no evidence of MgB_4 and Mg. Sample with excessive Mg is unreacted Mg. All samples exhibits MgO. Based on the XRD results, there are a large amount of MgO and Mg in the second phase of the sample. Impurities in the sample can act as pinning centre that cause higher J_c. Highest enhancement of critical current density (J_c) at 5 K and 20 K is found in Mg-deficient samples treated at 650°C annealing temperatures.

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