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Article

Experimental Study on the Phase Formation of MgxB2 (x=0.8, 1.0, 1.2)

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ABSTRACT

Phase formation of Magnesium diboride Mg_xB_2 (x=0.8, 1.0, 1.2) by *in situ* reaction of Magnesium (Mg) and Boron (B) at different annealing temperature by varying the nominal Mg composition was compared. The X-ray difffraction pattern indicates that Magnesium Oxide (MgO) is the major secondary phase. Some unreacted Mg was found for nominally stoichiometric and Mg excess samples annealed at or below 750°C. However, no unreacted Mg was detected by XRD for Mg deficient samples. Scanning Electron microscopy images show the porous nature of synthesised samples.

| Magnesium diboride | Phase formation | Porous |

1. 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]. 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 [3-4]. The presence of oxygen leads to reaction with Mg at high temperature producing MgO as impurities and 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 evaporates severely. In this paper, synthesis of Mg_xB_2 with varying x in iron tube at different growth condition was compared.

2. Material and Method

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 13mm diameter were made with 5 tonnes of pressure, sealed in an iron tube and annealed at 650°C and 800°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. *Tc* of the superconducting transition was determined 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.

3. Results and Discussion

The X-ray diffraction patterns of four MgB_2 powders prepared at different temperatures are presented in figure 1. It is clearly observed that MgB_2 phase dominates all samples with MgO as impurity phase. Also, some unreacted Mg presents in samples annealed at 650°C and 700°C. The results show the sintering temperature at 750°C and above is suitable for the preparation of pure MgB_2 by eliminating unreacted Mg.

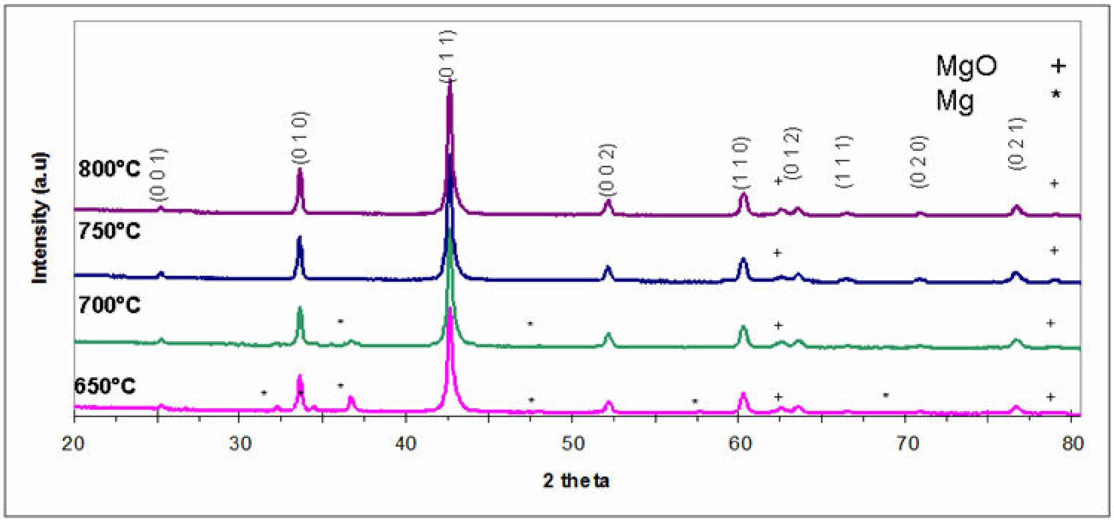

Figure 1: X-ray diffraction pattern on MgB_2 powder subjected to different sintering temperatures.

Table 1 shows that unreacted Mg volume fraction reduces with increasing annealing temperature and vanishes at 750°C. At this temperature, volume fraction of MgB_2 is the highest achieved among other samples.

Table 1: Volume fraction of MgB_2 superconductor with different annealing temperature.

	MgB2	MgO	Mg
MgB_2 650°C	80.8(5)%	13.64(1)%	5.511(4)%
MgB_2 700°C	85.8(5)%	12.369(9)%	1.879(1)%
MgB_2 750°C	88.4(5)%	11.556(8)%	-
MgB_2 800°C	87.8(5)%	12.161(9)%	-

The X-ray diffraction patterns of Mg_xB_2 with different nominal Mg content annealed at 750^{o}C is presented in figure 2. Again, MgB_2 is the dominant phase in all the samples with MgO as impurity. However, unreacted Mg presents in $Mg_{1.2}B_2$. This may be due to amount of Mg which is more than enough for reaction formation at this annealing temperature.

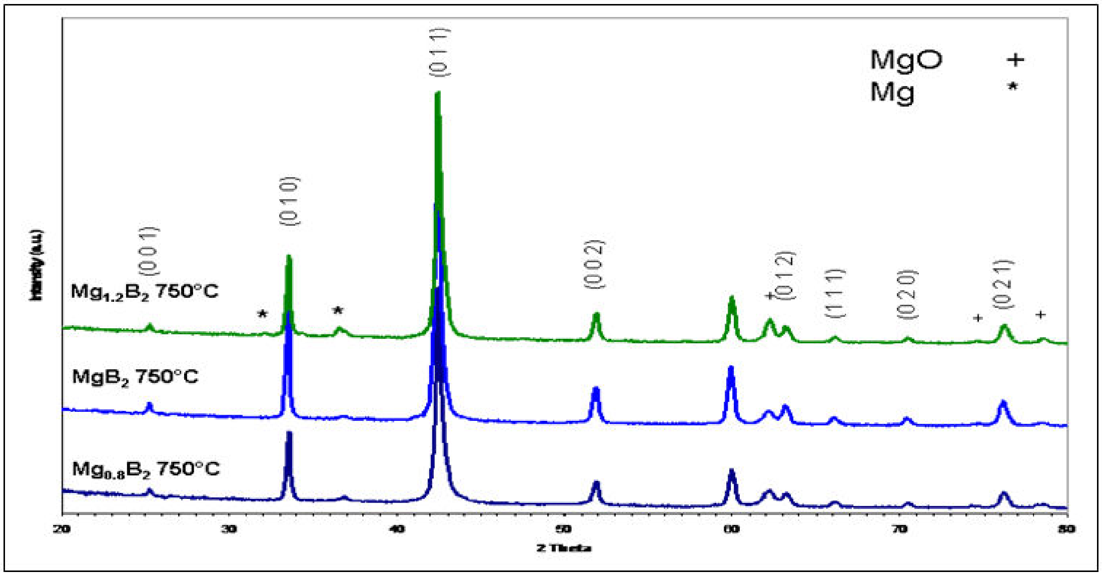

Figure 2: X-ray diffraction pattern on MgB_2 with different Mg content anneal at 750°C.

Table 2 shows that MgB_2 samples with the highest MgB_2 volume fraction and lowest MgO impurity. Mg impurities increase after excess Mg was added into MgB_2 sample showing that it is not the optimal level to react with B in producing MgB_2. Table 2 also show MgB_2 with 750°C annealing temperature is the best way to get highest MgB_2 phase fraction.

Table 2: Volume fraction of MgB_2 superconductor with different Mg content annealed at 750°C.

	MgB_2 (%)	MgO (%)	Mg (%)
$Mg_{0.8}B_2$ 750°C	77.7(6)	22.3(4)	-
MgB_2 750°C	88.4(5)	11.556(8)	-
$Mg_{1.2}B_2$ 750°C	79.6(5)	19.0(3)	1.5(1)

Figure 3 shows the normalised temperature dependence of the zero-field-cooled (ZFC) magnetization M(T) measured under 50Oe for all sample. The M(T) curve for all sample show nearly sharp superconducting transition below the onset of the demagnetization. In additional, 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$Tc_{(0)}$ deduce from the figure for Mg_xB_2 (x=0.8, 1.0 and 1.2) samples spans between 37.5K and 35.5K. The onset Tc for x=1.0 is about 38.25K while that for x=0.8 and 1.2 samples $Tc_{(onset)}$ is further suppress to around 36.10K. It is interesting to note that both $Tc_{(0)}$ and the diamagnetic onset temperature were suppressed to the effect of off-stoichiometry in Mg element in the system. Full diamagnetization is finally reached at around 32K for the samples with x=0.8 and 1.2, respectively. The suppression of diamagnetic shielding and respective $Tc_{(0)}$ and $Tc_{(onset)}$ in x=0.8 and x=1.2 samples does not severely affect the temperature-dependent of diamagnetism. The main cause of the superconducting properties suppression in these samples is however may relate to off stoichiometry. This evidenced from x-ray patterns and the data presented in Table 2, which indicate the presence of considerable amount of MgO impurity in these sample.

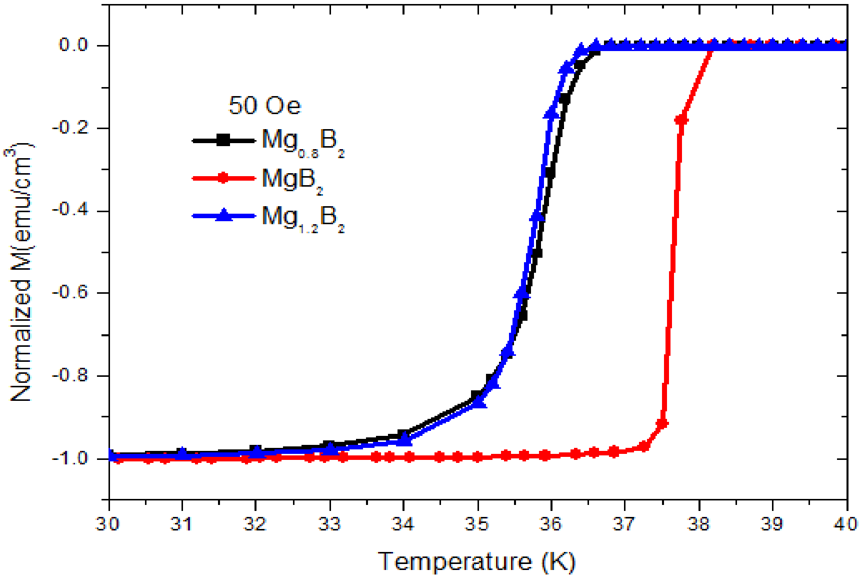

Figure 3: Normalise M-T plot for samples annealed at 750°C.

Figure 4 shows MgB_2 bulk annealed at 750°C. It is porous with significant amount of voids of micron size. Figure 5 show all the samples randomly look like hexagonal shape in nanosize thickness. For x=1.0, the thickness is thinner compare then other. The homogeneity of the microstructure is greater than x=1.0 sample.

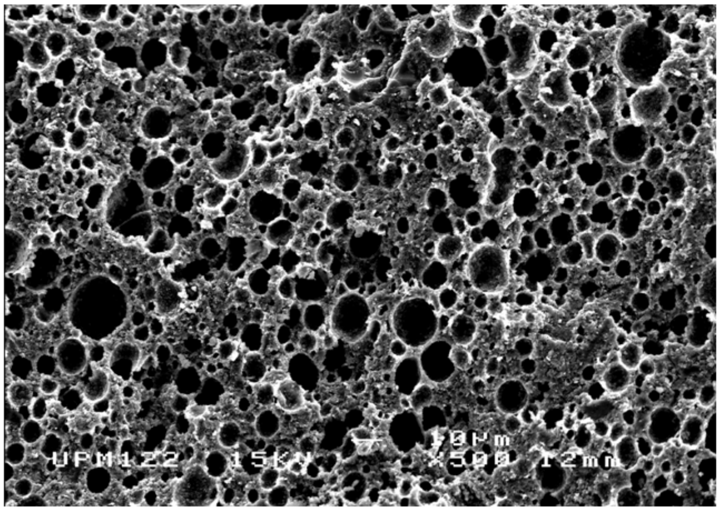

Figure 4: Scanning electron image of MgB_2 Superconductor annealed at 750°C.

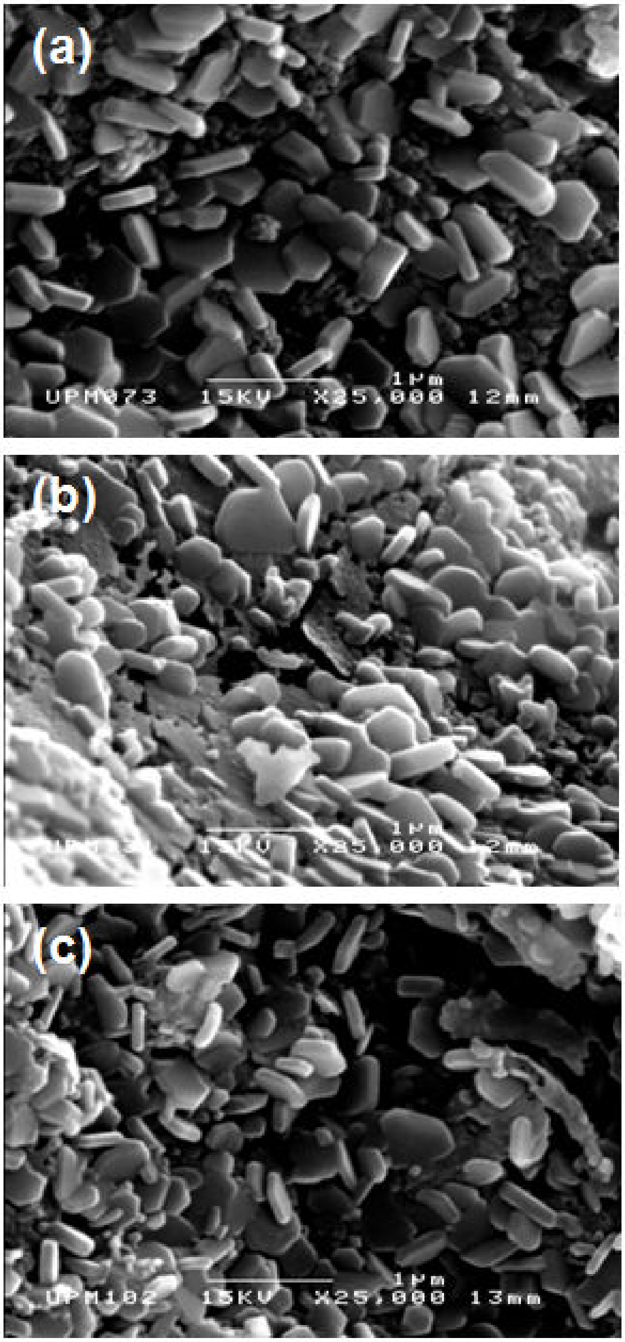

Figure 5: Mg_xB_2 stoichiometry annealed at 750°C (a) x=0.8, (b) x=1.0 and (c) x=1.2.

4. Conclusion

The XRD result indicated that the optimum MgB_2 phase formation temperature is at 750°C and 800°C. MgB_2 with 750°C anneal temperature is best way to get highest MgB_2 amount in the sample. The highest *Tc* value was observed in sample with x=1.0 indicates that the present of impurity is minimum.

5. Acknowledgements

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