Solid State Science and Technology, Vol. 17, No 1 (2009) 173-181
ISSN 0128-7389

COMPARATIVE STUDY ON THE CRITICAL CURRENT DENSITY OF MgB_2 PREPARED BY MIXED BORON POWDERS

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ABSTRACT

Polycrystalline MgB_2 was prepared from Mg and boron precursors consisting of boron powders with varying purity and form. T_c does not change largely for all samples. By replacing 10 wt.% of high purity amorphous boron with impure crystalline boron, comparable J_c to that of samples prepared from high purity amorphous boron powder alone can be obtained. High J_c can also be retained by replacing 20 wt.% of the high purity amorphous boron with impure amorphous boron. However, J_c decreases more rapidly with field by increasing the proportion of impure amorphous boron. By mixing both impure amorphous and crystalline boron powders even up to the proportion of 50 – 50 wt.%, the obtained MgB_2 exhibit enhanced J_c compared to samples prepared from the respective boron powder alone. The enhancement in J_c at 6K and 20K is more pronounced for applied field ≤ 3T.

INTRODUCTION

The discovery of much higher transition temperature, T_c (~ 39K) in MgB_2 [1] has drawn much attention from the scientific community as enormous effort has been put forward in investigating its superconducting properties. Apart from being a simple binary compound, the lack of weakly linked grain boundary promises higher current transport in MgB_2 compared to high temperature superconductors (HTS) [2]. Therefore, it offers some advantages over the latter for various electrical and electronic applications. It has been established that the critical current density, J_c of MgB_2 could be greatly increased by using simple chemical doping methods [3-7]. Very often, the residual resistivity ratio (*RRR*) in those samples is reduced owing to the increase of defect scattering [8]. While the research is going on intensively, there is a growing concern over the supply and the cost of high purity amorphous boron powder which is expensive and they are essential for synthesizing good quality samples with high T_c [9] and less impurities [10]. The availability of high purity amorphous boron is also limited in the market. Although the impure boron powder is commercially available but the synthesized samples have a lower critical current density, J_c [10]. Moreover, they are in semicrystalline form although being labeled as “amorphous” [10]. On the other hand, Mg powder can be obtained easily.

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Hence, there is a need to resolve the aforementioned issues of high purity amorphous boron powder. In this work, the extend of degradation in J_c by mixing impure and high purity boron powders of crystalline and amorphous forms is studied in order to establish the possibility of using impure boron powder as part of the entire precursor instead of solely relying on expensive high purity amorphous boron for obtaining comparable J_c.

EXPERIMENTAL

Samples were prepared by *in situ* reaction of Mg and boron precursors. High purity Mg powder (Alfa Aesar, 99.98%, 325 mesh) and three types of boron powders were used, i.e. crystalline boron 99%, amorphous boron 95 – 97% and amorphous boron 99.99% (denoted as B-C99, B-A9597 and B-A9999, respectively, hereafter) as described earlier in [10]. Three different boron precursors were prepared by mixing varying weight percent of (i) B-A9597 and B-A9999 (ii) B-C99 and B-A9999 (iii) B-C99 and B-A9597 (Table 1). They were then well mixed with Mg according to the stoichiometric ratio of 1:2 (Mg:B precursor) by hand grinding before pressing into pellets of 5 mm in diameter and about 2 mm thickness. The pellets were wrapped by Ta foil and loaded into a furnace for annealing at 900^{o}C for 15 min in 2%H_2-Ar flow environment with the heating and cooling rate of 15^{o}C/min. X-ray powder diffraction (XRD) spectra in the step-scanning mode θ - 2θ with 0.05^o increment were recorded using a Philips PW1050 diffractometer with a Cu-K_α radiation source. Commercial Quantum Design DC Magnetic Properties Measurement System (MPMS-XL) was used to obtain the superconducting transition temperature (T_c). Magnetisation hysteresis loops were performed on bar shaped samples. Magnetic J_c was estimated based on the critical state model [11].

RESULTS AND DISCUSSION

Samples prepared from boron precursor of mixed B-A9597 and B-A9999

Results from XRD show that the samples are dominated by MgB_2 phase with some identifiable MgO peaks (Figure 1). As shown in Figure 1, the unlabelled peaks can be indexed to MgB_2 phase.

As shown in Table 1, $T_{c\text{-onset}}$ of all samples varies only by 0.5K. T_c onset is defined as the point that deviates from linearity that signifies the transition from the normal to superconducting state in the magnetization versus temperature plot (not shown here). By mixing larger amount of B-A9597 with B-A9999, T_c decreases from 38K to 37.5K for samples prepared from 20 wt.% and 50 wt% of B-A9597. MgB_2 samples prepared from B-A9597 and B-A9999 alone, respectively, have the same T_c of 38.2K [10]. It should be noted that the step size used for measuring T_c in this study is 0.5 K compared to 0.1 K in reference [10].

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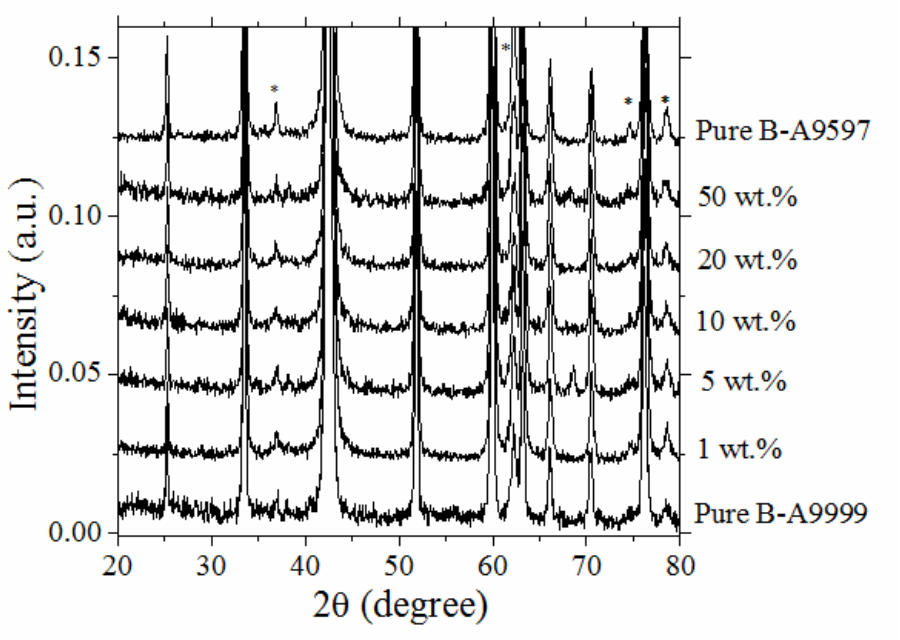

Figure 1 XRD spectra of MgB_2 samples prepared from boron mixture of B-A9597 and B-A9999.
∗ : MgO.

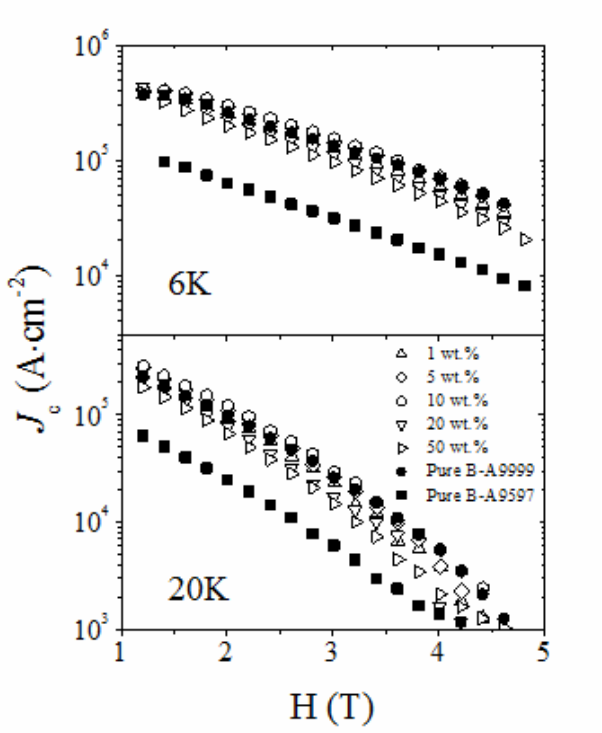

Figure 2: J_c versus field at 6K and 20K.

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Table 1: The values of critical transition temperature, T_c for MgB_2 samples prepared from various boron mixtures.

Samples		Mixture of boron powders			T_c (K)
		Wt.% of B-C99	Wt.% of B-A9597	Wt.% of B-A9999	
Set I	1	-	0	100	38.2
	2	-	1	99	38.0
	3	-	5	95	38.0
	4	-	10	90	38.0
	5	-	20	80	37.5
	6	-	50	50	37.5
	7	-	100	0	38.2
Set II	1	0	-	100	38.2
	2	1	-	99	37.5
	3	5	-	95	37.5
	4	10	-	90	38.0
	5	20	-	80	37.5
	6	50	-	50	37.5
	7	100	-	0	37.9
Set III	1	0	100	-	38.2
	2	1	99	-	38.0
	3	5	95	-	38.0
	4	10	90	-	38.0
	5	50	50	-	38.0
	6	100	0	-	37.9

Figure 2 shows the $J_c(H)$ at 6K and 20K for samples prepared from B precursor consisting of different weight percent of B-A9597 and B-A9999. Increasing B-A9597 up to 10 wt.% gives comparable J_c to that of pure B-A9999. Although J_c(6K) at 2T is close to sample prepared from pure B-A9999 by increasing the amount of B-A9597 up to 20 wt.%, its field dependence becomes stronger and J_c decreases with field more rapidly above 3T. Its J_c at 6K, 4T is 5.0×10^4 A/cm^2 compared to 6.9×10^4 A/cm^2 for pure B-A9999, i.e. a decrease of 27.5 % for the former. At 20K, 4T, J_c is 1.6×10^3 A/cm^2 compared to 5.6×10^3 A/cm^2 for sample prepare from pure B-A9999.

Set II: Samples prepared from boron precursor of mixed B-C99 and B-A9999
From the XRD spectra shown in Figure 3, minority peaks which can be indexed to MgO and MgB_4 are seen in samples prepared with 50 - 50 wt.% of B-C99 and B-A9999. Similar to sample prepared from pure B-C99, an unknown peak at $2\theta \sim 44^o$ is also noticeable. Other impurities such as B_2O and $Mg_3(BO_3)_2$ also start to appear.

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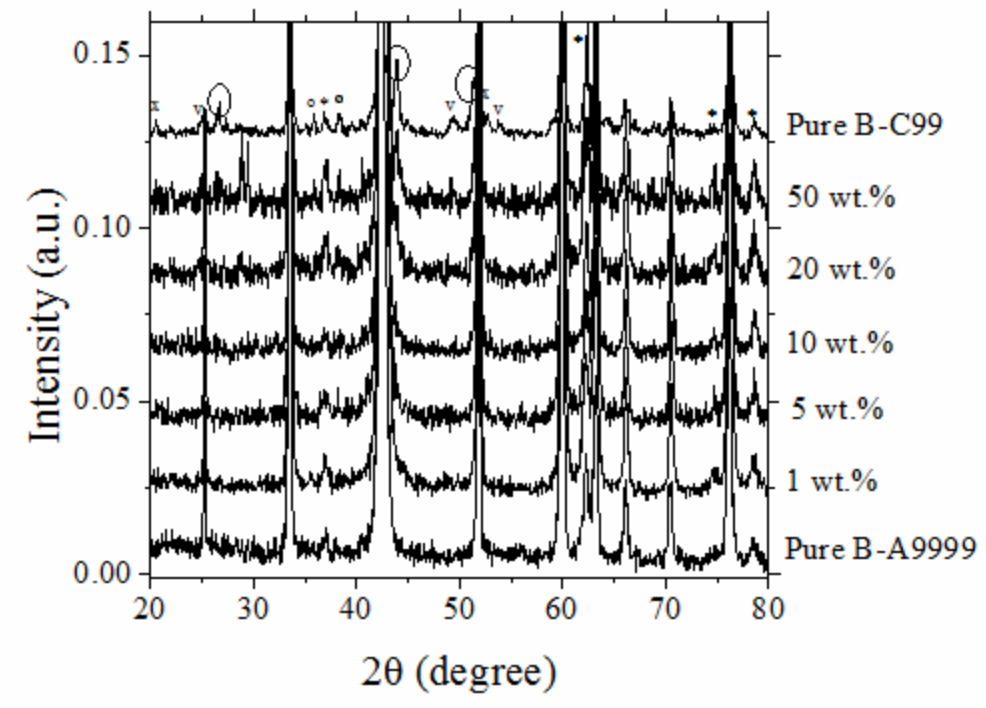

Figure 3: XRD spectra of MgB_2 samples prepared from boron mixture of B-C99 and B-A9999. The circled peaks correspond to unidentified phases. X: $B_{13}O_2$; V:$Mg_3(BO_3)_2$; o: B_2O; ∗ : MgO.

For MgB_2 samples prepared from the mixed boron powders of B-C99 and B-A9999, magnetic measurements also show a variation of 0.5K in T_c between 37.5K and 38K as indicated in Table 1. The measured T_c for C99 is 37.9K [10].

From Figure 4, it is obvious that samples prepared from boron precursor consisting of B-C99 and B-A9999 show the same field dependence of J_c at 6K and 20K. As expected, J_c decreases with increasing weight percent of B-C99 due to the blockage of current carrying paths by impurities [10]. Samples prepared from 10 wt.% of B-C99 do not show degradation in J_c at 6K and 20K. However, J_c decreased drastically by increasing B-C99 to 20 wt.%. J_c (20K) at 3T is 6.8×10^3 A/cm^2 which is about 74% lower compared to 2.6×10^4 A/cm^2 for pure B-A9999. Therefore, the detrimental effect on J_c is more severe in MgB_2 samples prepared by mixing B-C99 over B-A9597 into B-A9999.

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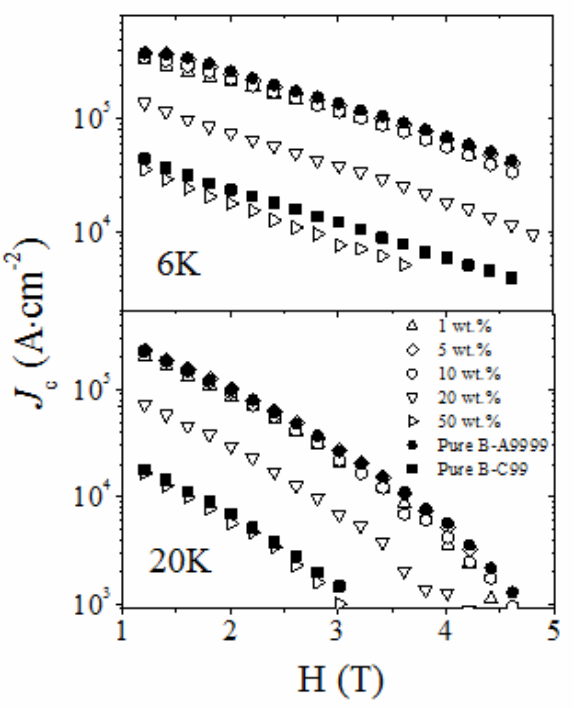

Figure 4 J_c versus field at 6K and 20K.

Set III: Samples prepared from B precursor by mixed B-C99 and B-A9597
With MgB_2 remains as a dominant phase, XRD pattern shows that peaks of MgO, B_2O and MgB_4 can be found in addition to the unknown peak at $2\theta \sim 44^o$ (Figure 5).

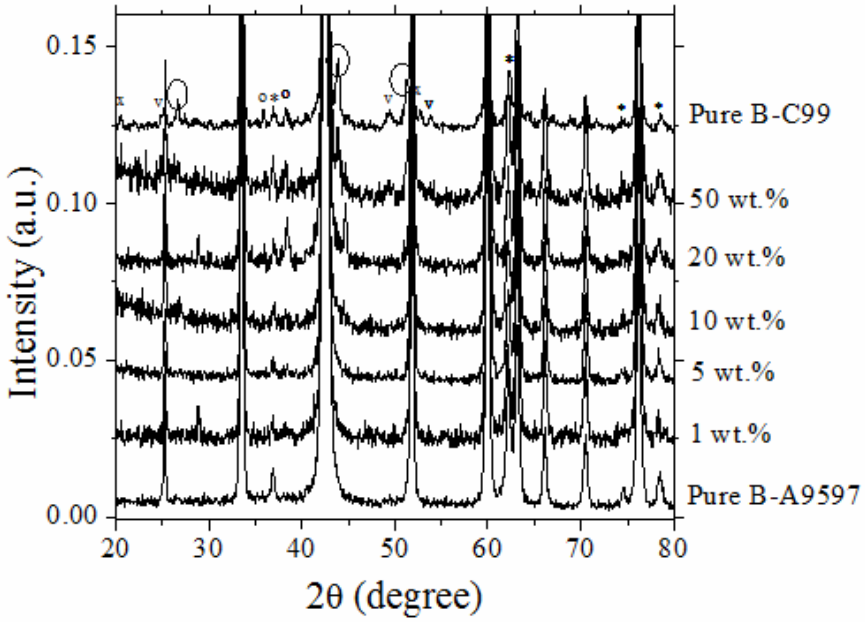

Figure 5: XRD spectra of MgB_2 samples prepared from boron mixture of B-C99 and B-A9597. The circled peaks correspond to unidentified phases. X: $B_{13}O_2$; V:$Mg_3(BO_3)_2$; o: B_2O; ∗ : MgO [11].

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$J_c(H)$ at 6K and 20K are shown in Figure 6. Surprisingly, samples with 50 wt.% of B-C99 shows better J_c than those prepared from B-A9597 alone at 6K for field below 4T. Although there is an increment in J_c, but its field dependence is stronger than both pure B-C99 and pure B-A9597. J_c at 6K, 2T for samples with 10 wt.% of B-C99 is 1.4×10^5 A/cm^2 compared to 6.3×10^4 A/cm^2 of pure B-A9597, i.e. about 22% of increment in J_c for the former. Its J_c approaches that of pure B-A9597 at about 4T. At 20K, the discrepancy in the enhanced J_c over pure B-A9597 is smaller. J_c at 20K, 2T for samples with 10 wt.% of B-C99 and pure B-A9597 is 4.0×10^4 A/cm^2 and 2.5×10^4 A/cm^2, respectively. Its J_c is comparable or lower than pure B-A9597 itself at field above 3T. As expected, the decrease in J_c by increasing weight percent of B-C99 is due to the blockage of current paths by impurities [10].

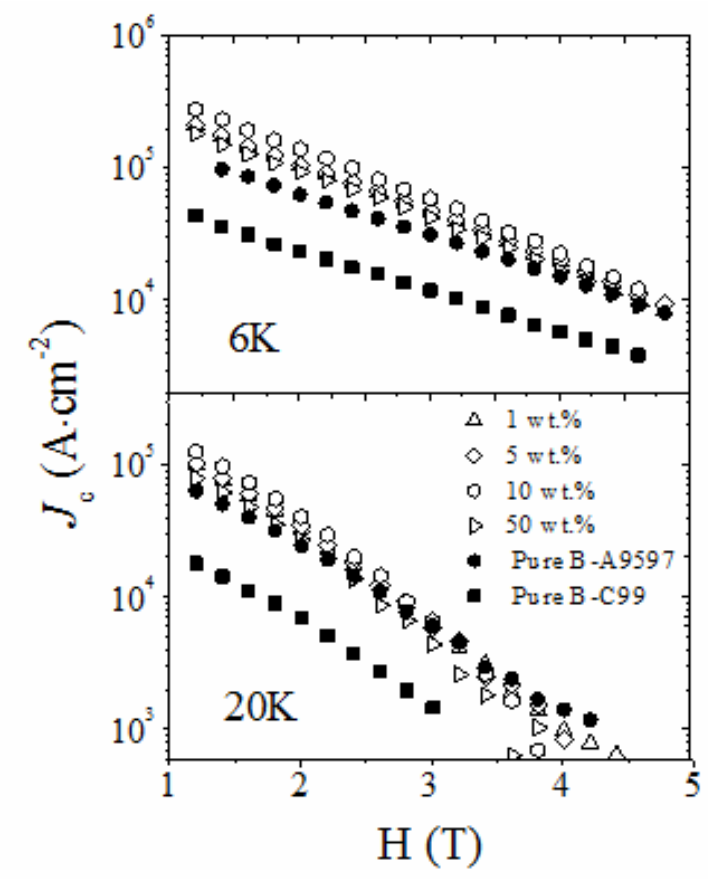

Figure 6: J_c versus field at 6K and 20K.

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We have seen that about 10 wt.% of the boron precursor can be replaced by impure amorphous or crystalline boron to obtain comparable J_c to that of samples prepared from high purity amorphous boron powder alone. Increasing the weight percent of impure crystalline boron led to more severe degradation in J_c (more than 50%) compared to impure amorphous boron. However, the field dependence of J_c for the latter is stronger. By mixing both impure amorphous and crystalline boron to serve as precursor, the reacted MgB_2 samples show enhanced J_c at 6K and 20K. Due to the different chemical reactivity of each boron powder, the mixing between them could give rise to structural distortion in the final products. This effect may be enhanced in samples prepared from precursor consisting of B-C99 and B-A9597 as both of them have bimodal particle size distribution [10] leading to higher J_c. Thus, J_c is increased without recourse to chemical doping or additions. Finally, it should be noted that the obtained results are based on the three types of boron powders used in this work. Therefore, results could be different for other boron powders.

CONCLUSIONS

By mixing boron powders of various purity and forms to serve as precursor to react with Mg, T_c of the resultant MgB_2 samples does not degrade significantly. By compromising the desired J_c and cost, appropriate amount of impure boron powder can be used as substituent without solely depending on the expensive high purity amorphous boron. Also, samples synthesized from boron precursor consisting of both impure amorphous and crystalline borons were shown to exhibit enhanced J_c at 6K and 20K at low field ($\leq$ 3T).

ACKNOWLEDGEMENT

Funding from Ministry of Science, Technology and Innovation Malaysia (MOSTI) under project no.: 03-01-04-SF0920 is gratefully acknowledged. We would like to thank J. L. MacManus-Driscoll for the research facilities.

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