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SINTERING EFFECT ON THE SUPERCONDUCTING PROPERTIES OF NANO-SiC ADDED MgB_2

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ABSTRACT

In this work, MgB_2 samples added with different weight percentages (wt%) of nano-SiC additions were synthesized by the conventional solid state method. They were sintered at 650 ºC and 850 ºC respectively, in order to study the effect of sintering temperature on their phase formation and superconducting properties. XRD spectra show relatively higher intensity of Mg_2Si at lower sintering temperature while Rietveld Refinement shows severe lattice contraction at *a*-axis but the *c*-axis remains unchanged with increasing SiC additions and sintering temperatures. The transition temperature degrades with the additions level but higher sintering temperature led to more gradual decrease in T_c. At 5K, samples sintered at lower temperature shows enhanced flux pinning properties with higher J_c. However, at 20K, samples sintered at higher temperature showed improved J_c at high field (>3T) when the addition level exceeds 5 wt%.

Keywords: MgB_2; superconductor; sintering temperature; silicon carbide;

INTRODUCTION

MgB_2 is found for its superconductivity in the year of 2001 [1]. With its unique characters such as the two band gap superconductivity and long coherence length, it has aroused the interest of scientific community to further understand its behaviors [2]. Due to its higher transition temperature (T_c~39K) and lower costs of production relative to Nb-based superconductors, it became the potential replacement for Nb-based superconductor and has been made into wires [3], tapes [4], thin films [5, 6] in order to study its performance in different applications. However, for application-wise, the pristine MgB_2 shows rapid degradations in critical current density (J_c) with increasing magnetic field. Fortunately, it has been shown that through chemical doping, the J_c can be greatly enhanced. Among the dopants, nano-SiC is a promising one that leads to weaker dependence of J_c on field [7]. There are reports [8, 9] that heat treatments have a

crucial influence on the phase formation of MgB_2. Hence, this study is aimed to optimize the phase formation and superconducting properties of MgB_2 after adding nano-SiC in varying sintering temperature.

EXPERIMENTAL DETAILS

Conventional solid state method was used to synthesize the samples. The molar ratio for the raw powder used is Mg:B:SiC = 1:2:*x* where *x* = 0 wt%, 3 wt% 5 wt% and 10 wt% relative to the weight of MgB_2 prepared. The powders were weighed according to the ratio and hand mixed thoroughly. The mixed powder was then pressed into pellets and sealed inside stainless steel tubes. They were sintered at 2 different temperatures which are 650 ºC and 850 ºC, respectively, for 1 hour in Argon flow atmosphere. The sintered pellets were ground into fine powders for X-ray Diffractions (XRD) using Panalytical X'Pert Pro PW 3040 MPD X-ray diffractometer with Cu anode to investigate the phase formations. The θ-2θ scanning mode was carried out in 0.02º. The superconducting properties were measured by using a Quantum Design Magnetic Property Measurement System (MPMS-XL) with the bar shape sample placed parallel to the applied field and zero field cooled. The superconducting transition temperature (T_c as T_c onset) was measured by heating up the samples from 20K to 40K under 20 Oe of applied field. Similarly, the critical current density (J_c) was measured by varying the magnetic field at 5K and 20K, respectively after zero fields cooling. Table 1 shows the sample identity.

Table 1: Sample identity of different weight percentage of SiC added MgB_2 sintered at 650 ºC and 850 ºC, respectively

Sintering Temperature (°C)	SiC Addition Level (wt%)	Sample Identity
650	0	6500
	3	6503
	5	6505
	10	6510
850	0	8500
	3	8503
	5	8505
	10	8510

RESULTS AND DISCUSSIONS

Figure 1 shows the phase formation of nano-SiC added MgB_2 samples sintered at 650 ºC and 850 ºC, respectively. Pure samples were also made at both temperatures to be used as a reference. It is observed that MgB_2 appeared as primary phase at both sintering temperatures regardless of SiC addition level. Although the samples were sealed inside the stainless steel tube and sintered in Argon gas atmosphere, detectable amount of MgO is found as one of the secondary phases in all samples. The formation

of MgO is inevitable as oxygen is entrapped inside the stainless steel tubes well before the sintering. Moreover, the oxygen might be contributed by the oxidized raw materials. As the addition of SiC increases, Mg_2Si phases become more significant and the crystallinity degrades as shown by the peak broadening. This indicates that the dissociation of nano-SiC and the reaction between Si and Mg to form Mg_2Si phase which is common as reported elsewhere [7, 10, 11]. Reaction of nano-SiC with Mg forms Mg_2Si phase and causes the relative peak intensity of MgB_2 to be reduced with the increasing addition. However, there is no higher boride phase such as MgB_4 or MgB_7 found in any of the samples, even for samples sintered at higher temperature in which the Mg loss is more severe. One of the secondary phases revealed with the increasing SiC addition is the unreacted nano-SiC as shown in 6510 and 8510 (Figure 1). However, the relative peak intensity of unreacted nano-SiC is found to be higher at higher sintering temperature. This is probably because Mg-SiC reaction is unfavorable at higher sintering temperature [12]. The relative intensity fraction of MgO and Mg_2Si was calculated according to [13] and shown in Table 2.

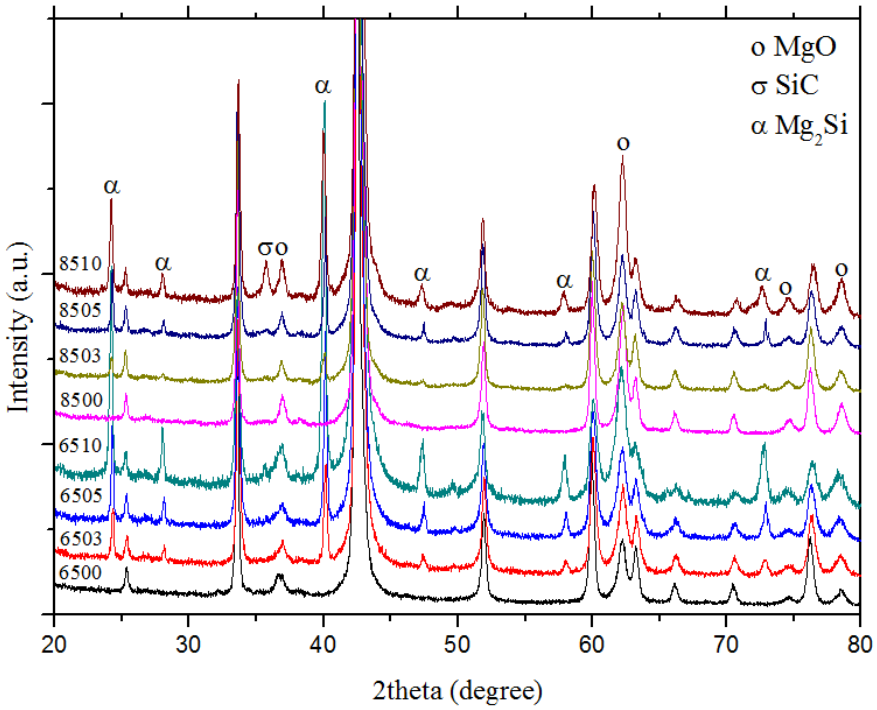

Figure 1: XRD θ-2θ scan of the nano-SiC added MgB_2 sintered at 650 ºC and 850 ºC. All unlabelled peaks are belonged to MgB_2

Table 2: The estimated relative intensity fractions of MgO and Mg_2Si

Sample	Relative Intensity Fraction of Phase (%)	
	MgO	Mg_2Si
6500	4.67	0.00
6503	5.20	6.49
6505	5.58	12.07
6510	8.84	19.97
8500	8.50	0.00
8503	5.29	1.79
8505	5.72	8.10
8510	8.64	10.96

As shown in Table 2, 6500 has the least secondary phase with no Mg_2Si (undetectable from x-ray pattern) and the least MgO relative intensity fraction. As the addition level increases, it was found that both the relative intensity fraction of MgO and Mg_2Si increase in each series of samples. The relative intensity fraction of Mg_2Si is much more reduced in 8510 (10.96%) compared to 6510 (19.97%). Generally, the relative intensity fraction of Mg_2Si is found to be much lower for sample sintered at higher temperature while the slight difference in relative intensity fractions of MgO implies that the oxidation is mainly originated from the raw material. The reduction in the relative intensity fraction of Mg_2Si at higher sintering temperature is in agreement with the observation of the XRD spectra in Figure 1 showing the lower relative intensities for those peaks. Therefore, this points to the fact that less nano-SiC was used for Mg-SiC reaction at higher sintering temperature.

Figure 2 shows the change of lattice parameters for the nano-SiC added MgB_2 sintered at 650 ºC and 850 ºC evaluated by using Rietveld Refinement. It is found that the lattice is distorted significantly and systematically with the addition of nano-SiC compared to the pure samples. The *a*-axis shrinks steadily while *c*-axis remains unchanged with SiC addition as reported in most of the previous findings [7, 11]. From Figure 2, it is obvious that samples sintered at 850 °C showed shorter *a*-axis for addition level less than 10 wt% suggesting that more C atoms have been substituted into B sites at higher temperature. Various reports have shown that C substitution is more favorable at higher temperature from pure C source alone due to its sufficient activation energy [14, 15]. For our case, C is incorporated into the MgB_2 matrix which probably due to the active C freed from Mg-SiC reaction. This can be observed by higher shrinkage in *a*-axis for sample sintered at higher temperature.

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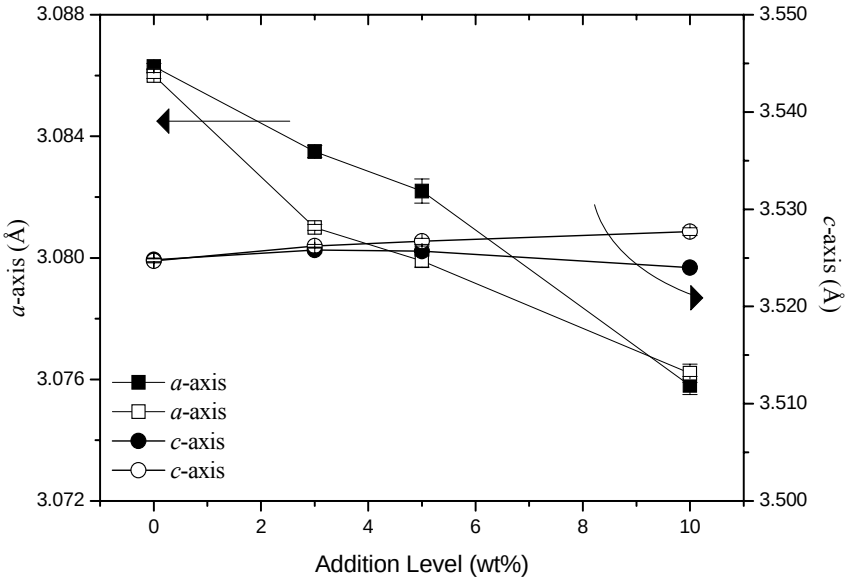

Figure 2: Evolution of lattice parameters with nano-SiC addition for samples sintered at 650 ºC (closed symbol) and 850 ºC (open symbol), respectively

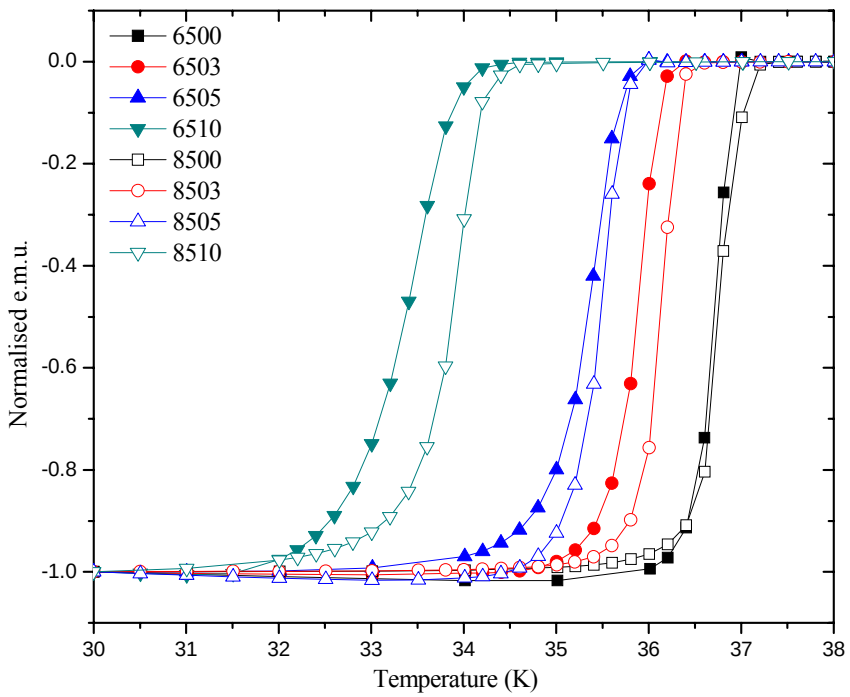

Figure 3: Normalized magnetic moment (emu) versus temperature for both series of samples

Figure 3 shows the superconducting transition temperature of nano-SiC added MgB_2 sintered at 650ºC and 850ºC, respectively. It is shown that 6500 and 8500 share the same T_c of 37.0K with the transition width (ΔT) of 1K. With the addition of SiC increases to 10 wt%, the ΔT increases to 3.0K and 2.4K for 6510 and 8510, respectively. It can be observed from Figure 3 that samples sintered at higher temperature show higher T_c and lower ΔT than in samples sintered at lower temperature. This is consistent with the XRD spectra shown previously as more MgB_2 phase appeared in samples sintered at higher temperature because higher sintering temperature does not favor the formation of Mg_2Si. Moreover, higher sintering temperature produces samples with improved crystallinity and homogeneity which are crucial in reducing T_c suppression and ΔT broadening [16].

Figure 4 shows the critical current density as a function of applied field at 5K and 20K. It is clear that at 5K, samples sintered at lower temperature always give higher magnitude of J_c compared to samples sintered at higher temperature. Among the plots, 6505 has the highest magnitude at 5T, 5K. Furthermore, it is observed that the 6510 and 8510 showed crossover to the pure sample only at higher field (>6T). As the SiC addition increases, the J_c degrades more gradually. This shows that the field dependence of J_c has been improved for all SiC added samples regardless of sintering temperature. At 20K, both 6505 and 8505 showed similar improvement in J_c. However, they did not show any crossover to the pure sample even at high field. Hence, higher Jc at higher field is expected for SiC added samples compared to the pure one. Better J_c performance is obvious in sample sintered at lower temperature which is in agreement in XRD spectra. This is due to lower crystallinity with the formation of Mg_2Si at lower sintering temperature and higher degree of C substitution in MgB_2 matrix (lattice defect) contributed by Mg-SiC reaction which serve as effective pinning centers.

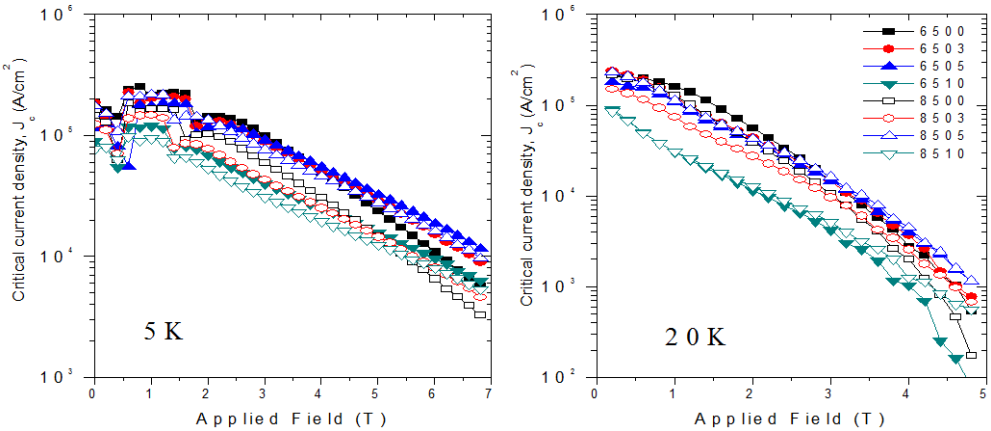

Figure 4: Field dependence of critical current density at 5k and 20k

CONCLUSION

This study shows that lower sintering temperature leads to lower shrinkage in *a*-axis for low addition of SiC (<10wt%). However, higher sintering temperature produced samples with higher T_c and lower ΔT. With degraded crystallinity and C substitution, samples sintered at lower temperature show higher magnitude of J_c compared to samples sintered at higher temperature at 5K. It is clear that the field dependence of J_c is enhanced with the SiC additions attributed to effective pinning centers existed in the samples.

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