Synthesis of $Mg(B_{1-x}C_x)_2$ Powders

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Abstract

We have synthesized bulk $Mg(B_{1-x}C_x)_2$ from a mixture of elemental Mg, B, and the binary compound B_4C . Carbon incorporation was dramatically improved by a two step reaction process at an elevated temperature of 1200 °C. This reaction process results in a solubility limit near $x\sim0.07$. We found that impurities in the starting B cause an additive suppression of T_c . We combine these data with T_c and $H_{c2}(T=0)$ data from CVD wires as well as plasma spray synthesized powders and present a unifying H_{c2} and T_c versus x plot.

Key words: MgB₂, carbon doping, upper critical field

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1 Introduction

Superconductivity in MgB₂ near 40 K [1] has attracted much interest due to its potential for applications in the 20-30 K range. The low upper critical field [2] and high anisotropy ratio, $\gamma = H_{c2}^{\parallel ab}/H_{c2}^{\perp ab}$ [3,4,5], of pure MgB₂, limit its potential usefulness [6]. Carbon doping of MgB₂ has been shown to be an effective method for enhancing H_{c2} [7,8,9] while simultaneously decreasing the anisotropy ratio by increasing $H_{c2}^{\perp ab}$ more rapidly than $H_{c2}^{\parallel ab}$ [10,11]. Carbon doped MgB₂ filaments require high reaction temperatures which lead to large grain sizes and poor J_c values, in spite of the enhancements in H_{c2} [12]. Reports of successful fabrication of superconducting wire using powder-in-tube processing [13,14], coupled with the fact that J_c values in powder samples can readily be increased by the addition of various particles [15,16,17,18] motivate

a desire for synthesizing single phase carbon doped MgB₂ powder. Carbon doped bulk polycrystalline MgB₂ with approximately 10% carbon incorporation has previously been synthesized by mixing elemental Mg and the binary B₄C [19,20]. Systematic carbon doping of Mg(B_{1-x}C_x)₂ with x<0.10 has been achieved in single crystals [8,21] and polycrystalline wires fabricated by chemical vapor deposition (CVD) [7,12]. In this paper we explore the possibility of preparing Mg(B_{1-x}C_x)₂ with x<0.10 using B₄C as the carbon source.

2 Experimental Methods

Powder samples of $Mg(B_{1-x}C_x)_2$ were prepared in a two step process. First, stoichiometric mixtures of distilled Mg, elemental B, and the binary compound B₄C were reacted for 48 hours at 1200 °C. The resultant sample was then reground in acetone, pressed into a pellet, and re-sintered for an additional 48 hours at 1200 °C. Two different batches were made. The first with 0.995 purity B (metals basis) from Alpha Aesar, and the second with 0.9997 purity isotopically enriched ¹¹B from Eagle Picher. The three main impurities in the 0.995 purity B are C, Si, and Fe, which have relative atomic abundances of 0.25% and 0.20%, and 0.10% respectively. The isotopically enriched $^{11}\mathrm{B}$ contained 0.02% Ta, 0.001% Cu, and 0.001% Fe. Samples made with isotopically enriched ¹¹B were shown to have a higher residual resistivity ratio (RRR) than samples made with nominal 0.9999 purity B, indicating the isotopic enrichment process yields perhaps the purest boron available [22]. B₄C from Alpha Aesar was used as the carbon source in both runs. The B₄C had a nominal purity of 0.994 metals basis with the two primary impurities being Si and Fe, which occur in relative abundances of 0.37% and 0.074% respectively, values similar to those in the 0.995 purity B. Since impurities in the boron have been shown to suppress T_c [22], two differing boron purities were used to examine the effects of the starting boron purity on the T_c and $H_{c2}(T=0)$ values in carbon doped MgB₂. We found it necessary to use multiple reaction steps to incorporate the carbon as uniformly as possible. To avoid confusion regarding the meaning of x in $Mg(B_{1-x}C_x)_2$, we will henceforth refer to the nominal carbon content as x_n and the inferred carbon content after the m-th reaction step as x_{im} .

Powder x-ray diffraction (XRD) measurements were made at room temperature using $CuK\alpha$ radiation in a Rigaku Miniflex Diffractometer. A silicon standard was used to calibrate each pattern. Lattice parameters were determined from the position of the MgB_2 (002) and (110) peaks. DC magnetization measurements were performed in a Quantum Design MPMS-5 SQUID magnetometer. Transport measurements were done using a four probe technique, with platinum wires attached to the samples with Epotek H20E silver epoxy. Resistance versus temperature in applied fields up to 14 T were carried out in

a Quantum Design PPMS-14 system and resistance versus field was measured up to 32.5 T using a lock-in amplifier technique in a resistive DC magnet at the National High Magnetic Field Laboratory in Tallahassee, Florida.

3 Results and Discussion

Using the lower purity boron, we reacted a sample with nominal carbon content of $x_n=0.05$ for 48 hours at 1200 °C. The carbon content can be estimated by the shift of the x-ray (110) peak position relative to that of a nominally pure MgB₂ sample made under the same conditions [7,20]. Although the B₄C is a different B source than that used for the reference sample, the 0.994 purity B₄C and 0.995 purity B contain similar concentrations of impurity phases, and we cautiously proceed with estimates of the carbon content ignoring minor differences between boron sources. Indexing of the (110) peak for the pure sample and that containing a nominal carbon content of $x_n=0.05$ yielded an inferred carbon level after this first reaction step of approximately $x_{i1}=0.031$. In order to ensure the carbon was fully incorporated and uniformly distributed within this sample, it was reground in acetone, pressed into a pellet, and sintered for an addition 48 hours at 1200 °C. The subsequent sample showed a further increase in the (110) peak position (Figure 1) which yielded an inferred carbon content of $x_{i2}=0.069$. This sample also showed decrease in T_c (Figure 2), which is consistent with more carbon being incorporated in the structure. In addition to shifting to higher 2θ , the (110) peak became sharper. The fullwidth-at-half-maximum (FWHM) decreased from 0.221° to 0.157° after the second sintering step. Therefore the second sintering step not only incorporated more carbon but it appears to have resulted in a more uniform carbon distribution. It should be noted that after the two sintering steps the FWHM values of the MgB₂ peaks were comparable to those of the Si standard, indicating we have achieved a fairly high level of homogeneity. It is also worth noting that whereas the (110) peak shifts and sharpens as a result of a second reaction step, the (002) peak does neither, having FWHM values comparable to, but slightly larger than, the neighboring Si (311) peak after both reaction steps. This indicates that the c-axis spacing and periodicity are particularly insensitive to this degree of carbon doping and/or disorder.

Whereas the x_{i2} value exceeds the nominal value of x_n =0.05 in the starting material, the presence of MgB₄, as evidenced by strong peaks in the x-ray spectrum (Figure 3) may account for the discrepancy if we assume no carbon enters the MgB₄ structure. Comparison of the x-ray spectra for the single and two step reactions shows an increase in the intensity of the MgB₄ peaks after the second reaction step. This step was done without the addition of any extra Mg to compensate for potential Mg loss. It is possible that while sintering at 1200 °C, some Mg is driven out of the MgB₂ structure and this loss

results in conversion of MgB₂ to MgB₄, with the excess Mg forming MgO and possibly condensing on the walls of the tantalum reaction vessel during the quench. To determine whether or not Mg loss is responsible for the apparent increase in the carbon content, the second sintering step for a sample with nominal concentration x_n =0.05 was carried out in an atmosphere of excess Mg vapor. This sample exhibited a T_c of 29.8 K and a shift in the (110) x-ray peak yielding an inferred carbon concentration of x_{i2} =0.050 (Figure 4), consistent with the nominal concentration. Thus the apparent difference in carbon content for samples which undergo a second sintering step without the presence of excess Mg to compensate for Mg loss relative to those which undergo the second sintering step with excess Mg is presumably the result of a fixed amount of carbon being incorporated into a decreased amount of MgB₂. To avoid the potential creation of percolation networks of Mg within the samples we chose to perform the second sintering step without any additional Mg.

Using 0.995 purity boron and 0.994 purity B_4C , an entire series with nominal carbon levels of $x_n=0$, 0.0125, 0.025, 0.035, 0.05, and 0.075 was prepared using the two step reaction profile. The (002) and (110) x-ray peak positions for the entire series are plotted in figure 5. The (002) peak position is roughly constant for all carbon levels, consistent with the results found by Wilke et al. [7] and Avdeev et al. [20], which showed only a slight expansion along the c-axis for carbon doping levels up to $10\pm2\%$. The (110) peak position shifts towards higher 2θ values as x is increased up to $x_n=0.05$, at which point it appears to be saturated. Using the (110) peak position for the nominally pure sample as our standard, the inferred carbon concentrations for the entire series are $x_{i2}=0$, 0.01, 0.034, 0.044, 0.069, and 0.067. The samples with carbon concentrations saturating near $x_{i2}\sim0.07$ show an increase in the MgB₂C₂ phase as a function of the nominal carbon content (Figure 5b). Thus the excess carbon is precipitating out as MgB₂C₂.

Normalized magnetization measurements (Figure 6) confirm the highest two doping levels have incorporated roughly the same amount of carbon. Their transition temperatures all lie slightly below 28 K. Defining T_c using a 2% screening criteria the x_{i2} =0.069 and 0.067 levels have T_c values of 27.5 K and 27.8 K respectively. For these higher doping levels, the nominal concentrations did not yield systematic increases in carbon level, but the change in the alattice parameter and T_c are consistent with one another; i.e. samples which apparently incorporated more carbon had smaller a-lattice parameters and lower T_c values.

This saturation near $x_{i2}\sim0.07$ is not entirely unexpected given the results of $10\pm2\%$ carbon incorporation using B₄C reported by Ribeiro et al. [19] and Avdeev et al. [20]. In optimizing the reaction, Ribeiro found that under certain conditions, T_c values below the near 22 K reported for the optimal

24 hours at 1100 °C reaction could be attained, suggesting higher carbon content phases may be metastable. To test whether the saturation we observed was an effect due to the use of a two step reaction, as opposed to the single step employed by Ribeiro et al., we repeated their work, making a sample of nominal concentration $Mg(B_{0.8}C_{0.2})_2$ using only Mg plus B_4C . This sample underwent an initial 48 hour reaction at 1200 °C to form the superconducting phase and a second sintering for 48 hours at 1200 °C. After the first 48 hours at 1200 °C we find a superconducting phase with T_c near 22 K, and a lattice parameter shift which yielded inferred values of $x_{i1}=0.092$ slightly less than the $x_i=0.10$ obtained by Ribeiro and coworkers using isotopically enriched $^{11}B_4C$ as the carbon source [20]. After the second sintering step, T_c rises to 27.9 K. The (110) x-ray peaks shifted to lower 2θ , yielding an inferred $x_{i2}=0.065$ (Figure 7). Although two reaction steps were used, no change in the FWHM of the (110) peak was observed. As in the case of the $x_n=0.075$ sample, the decrease in carbon content could be due to carbon precipitating out in the form of MgB₂C₂. The relative intensity of the most prominent MgB₂C₂ peak to that of MgB₂ approximately doubles going from the single step reaction to the two step reaction. In order to check if more carbon would be precipitated out in the form of MgB₂C₂ by simply adding more sintering steps to the growth process an additional sample underwent a three step reaction: an initial 48 hours at 1200 °C to form the superconducting phase followed by two additional sintering steps of 48 hours at 1200 °C. After this third reaction step, the sample exhibited a T_c of 27.9 K and the (110) peak position yielded an inferred carbon content of $x_{i3}=0.064$ (Figure 7). These values are comparable to our previous results with only two sintering steps. Thus the carbon content appears to saturate in the vicinity of an inferred carbon content of $x_i=0.065$. The fact that saturation near $x_{i2}=0.065$ occurred for samples which had x_{i1} both above and below this level indicates that in equilibrium the solubility limit for 1200 °C reactions near 1 atm is in the range $0.065 < x_i < 0.07$.

Transport measurements were made in order to determine the upper critical field. An onset criteria was used in both resistance versus temperature and resistance versus field. Figure 8a plots resistance versus temperature in applied fields up to 14 T and figure 8b plots resistance versus field at temperatures down to 1.4 K for the sample with a carbon level of x_{i2} =0.069 (x_n =0.05). The zero field resistive transition has a width of less than 1 K, but this significantly broadens as the strength of the applied field increases. The R vs. H measurements show a related broadening. For example, the width of the transition at 1.4 K is nearly 20 T wide. This should be compared to the 10 T wide, approximately linear transitions reported for 5.2% carbon doped filaments [12]. Optical images taken under polarized light show the superconducting grains in the powder sample are 5-10 μ m in size. In the case of the wire sample a majority of the grains are in the 1-5 μ m range [12]. We therefore ascribe the increased width of this transition to a combination of poor flux pinning due to the large grain size associated with the high reaction temperature as well

as to possible remaining inhomogeneities in carbon incorporation within the sample.

 H_{c2} curves for x_{i2} =0.034 and x_{i2} =0.069 along with a pure wire [2] and a carbon doped wire with an inferred carbon content of x_i =0.052 [7] are plotted in figure 9. The powder sample with x_{i2} =0.034 has a T_c slightly less than that of the carbon doped wire with x_i =0.052 and an $H_{c2}(T=0)$ more than 5 T lower. This marked difference shows that for carbon doped samples made with differing nominal boron purities, T_c alone is not a good caliper of $H_{c2}(T=0)$. The sample with inferred carbon content of x_{i2} =0.069 has a T_c nearly 7 K below the aforementioned wire and an $H_{c2}(T=0)$ just above 30 T. Comparing the two powder samples to one another, we see an increase in the slope of H_{c2} near T_c for the higher doping level, which results in a higher $H_{c2}(T=0)$, consistent with our earlier findings [7,12].

Carbon has been shown to suppress T_c at a rate of roughly 1 K/%C for up to 5% carbon substitution [12]. The magnetization and transport measurements indicate T_c of the powder samples made with the 0.995 purity Alpha Aesar boron is also being suppressed at a rate of roughly 1 K/%C, but relative to the suppressed, near 37 K, transition temperature of the nominally pure sample. The suppressed transition temperature of the nominally pure MgB₂ sample lies approximately 2 K below results obtained using high purity natural boron wires [23]. MgB₂ made from lower purity boron has been shown to have lower transition temperatures [22]. In figure 10, a comparison of T_c versus $|\Delta a|$ for carbon doped samples prepared with lower purity boron to carbon doped wires made with high purity boron shows the manifold associated with the impure boron powder is shifted downward by approximately 2 K for all carbon levels. To confirm that this difference is a result of the purity of the starting boron, a second set of two step process samples made with isotopically enriched ¹¹B were measured. The results are included in figure 10. Also included is a set of carbon doped powders made by a plasma spray process [24]. The agreement between the CVD wires, plasma spray powders, and ¹¹B samples shows that high purity boron in a variety of forms responds to carbon doping in a similar manner. These data also seem to indicate that there is some additional impurity in the 0.995 pure Alpha Aesar boron that is systematically suppressing T_c .

Figure 11 plots a comparison of the (002) and (110) x-ray peaks of pure MgB_2 using the three different purity levels of boron. The sample made with the nominal 0.995 purity boron shows a shift of the (110) peak to higher 2θ by 0.09°, which, if it were to be associated with carbon doping, would be consistent with carbon doping of approximately 1.8%. This level far exceeds the stated carbon impurity level of 0.25% in the 0.995 purity B as claimed in the certificate of analysis provided by Alpha Aesar. To check whether by using lower purity boron we have inadvertently doped with carbon to such a high

level, we measured the resistive onset of superconductivity in an externally applied 14 T field for the nominal pure MgB₂ using the 0.995 purity boron and compared the temperature with those attained for carbon doped fibers reported in reference [12]. MgB₂ fibers reacted at 1200 °C for 48 hours showed an onset of superconductivity in an externally applied magnetic field of 14 T at 10.2 K, 14.8 K, and 18.5 K for pure, 0.6%, and 2.1% carbon doping [12]. If the shift of the (110) peak in the nominally pure MgB₂ made from 0.995 purity boron were a result of inadvertent carbon doping, we would expect an onset of superconductivity in an applied 14 T field at a temperature between 15 K and 18 K. However, such a measurement yielded an onset near 13K indicating if carbon is present as an impurity, it is less than 0.6%, which consistent with the estimate provided by Alpha Aesar. Therefore the manifold of T_c versus $|\Delta a|$ for the lower purity boron is shifted downward by some as of yet unidentified impurity associated with the Alpha Aesar boron.

 H_{c2} values were determined using an onset criteria in resistivity versus temperature and resistance versus field measurements. Pellets made using the isotopically enriched ¹¹B lacked structural integrity and were unsuitable for transport measurements. Therefore we could only attain $H_{c2}(T=0)$ values only for samples made from the CVD wires, plasma spray powders, and the 0.995 purity boron (Figure 12). For the dirtier, 0.995 purity powder, at a doping level of $x_{i2}=0.034$ the upper critical field agrees with the results of a carbon doped wire with an inferred carbon content of $x_i=0.038$ from reference [7]. At doping levels near $x_{i2}=0.065$, H_{c2} values fall several Tesla below the manifold for "clean" carbon doped samples. In carbon doped MgB_2 , enhancement of H_{c2} due to scattering effects [11,26] competes with the suppression of T_c caused by electron doping [11]. By further suppressing T_c by introducing additional impurities in the system, we may have limited the maximum H_{c2} attainable through carbon doping.

4 Conclusions

We have established a method for synthesizing $Mg(B_{1-x}C_x)_2$ using a mixture of distilled magnesium, boron and the binary compound B_4C . Impurities in the starting boron effect T_c and the magnitude of the a-lattice parameter. By tracking $|\Delta a|$ and T_c we were able to show that different boron purities lead to differing $T_c(|\Delta a|)$ manifolds. There appears to be a solubility limit in the carbon content for samples synthesized at 1200 °C and 1 atm near $x\sim0.07$. Lower purity boron in the starting material results in lower transition temperatures and appears to limit the maximum achievable upper critical field.

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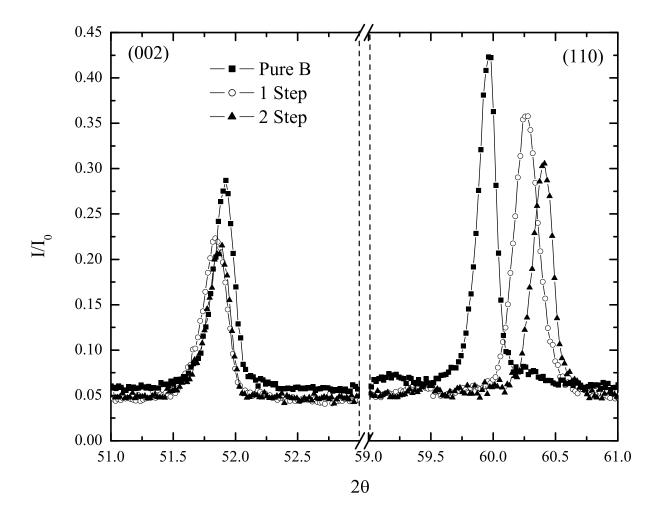


Fig. 1. Powder x-ray diffraction (002) and (110) peaks for pure and nominal $Mg(B_{.95}C_{.05})_2$ samples made using the 0.995 purity boron as the starting material. The pure sample was reacted using two steps. For the carbon doped sample, the second sintering step shifts the (110) peak position to higher 2θ indicating the incorporation of a higher carbon concentration.

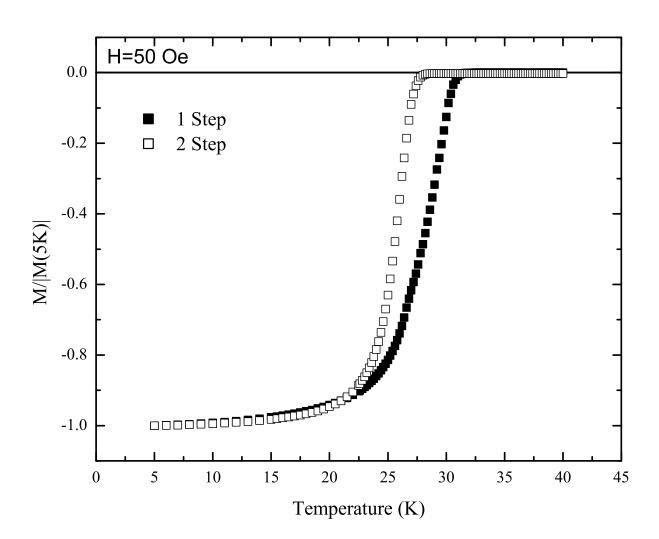


Fig. 2. Normalized magnetization curves for nominal $Mg(B_{.95}C_{.05})_2$ samples. The second sintering step lowers T_c , consistent with the incorporation of a higher carbon concentration.

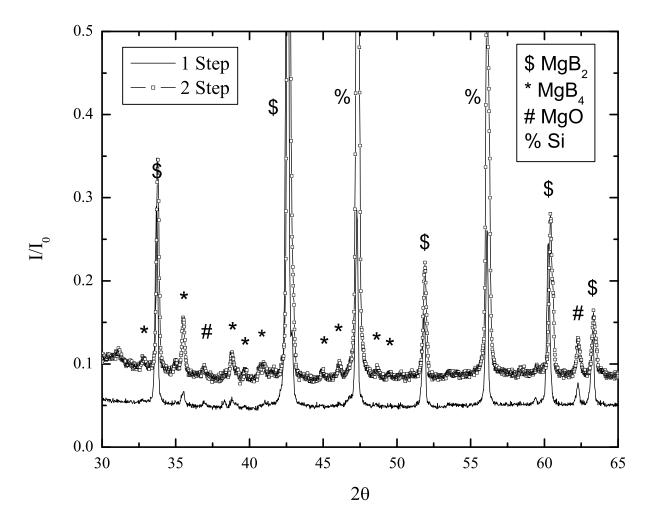
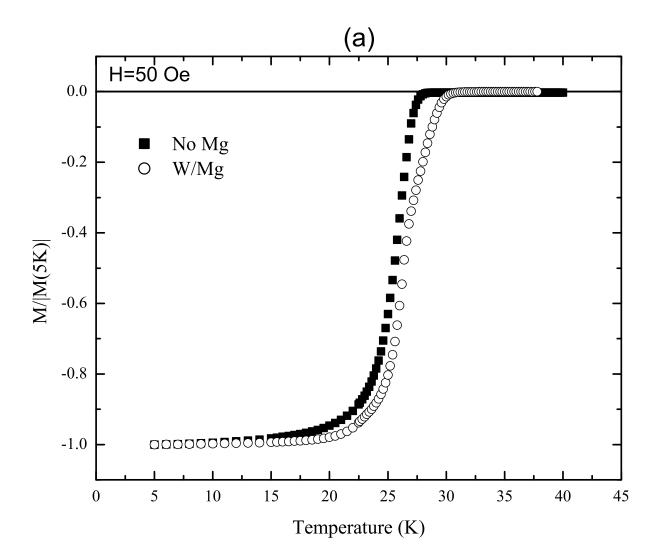


Fig. 3. Normalized powder x-ray pattern for a sample with $x_n=0.05$ synthesized using a one step and a two step reaction. The two step sample clearly contains enhanced amounts of MgB₄.



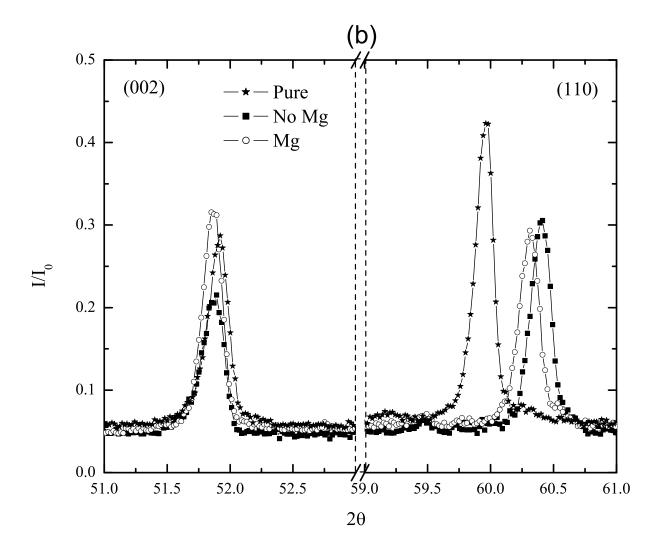
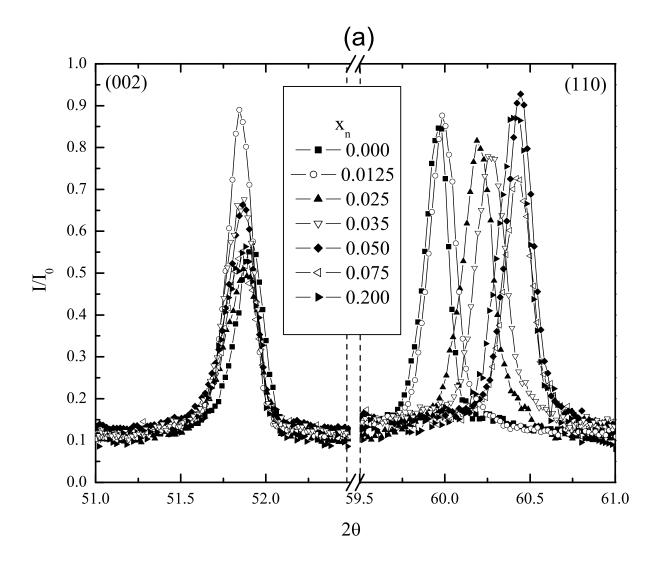


Fig. 4. (a) Normalized magnetization curves and (b) x-ray (002) and (110) peaks for a sample of x_n =0.05 reacted using a two step process. If the second sintering step is performed without any excess Mg to compensate for potential losses, the resultant carbon content within the MgB₂ phase is increased.



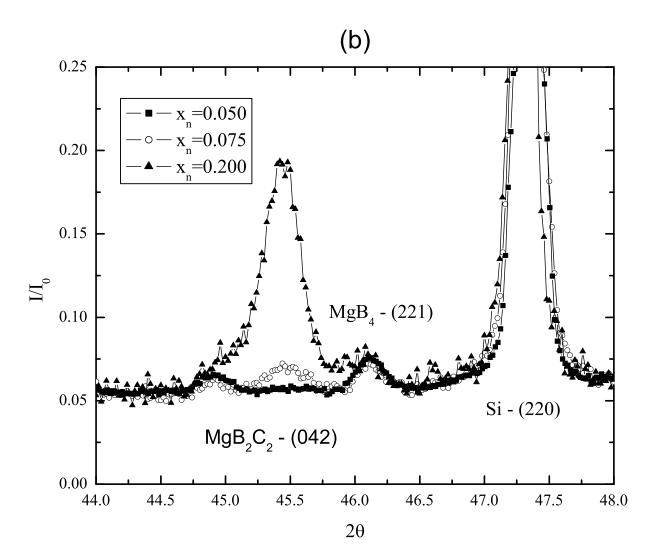


Fig. 5. (a) Evolution of the (002) and (110) x-ray peaks for $Mg(B_{1-x}C_x)_2$ samples with nominal $x_n=0$, 0.0125, 0.025, 0.035, 0.05, 0.075, and 0.20 synthesized using a two step reaction. The shift of the (110) peak relative to that of the un-doped yields inferred carbon concentrations of $x_{i2}=0.01$, 0.034, 0.044, 0.069, 0.067, 0.065. (b) For samples saturating near $x_{i2}=0.07$ the excess carbon precipitates out in the form of MgB_2C_2 as can be seen by the emergence of the MgB_2C_2 (042) peak as a function of nominal carbon content.

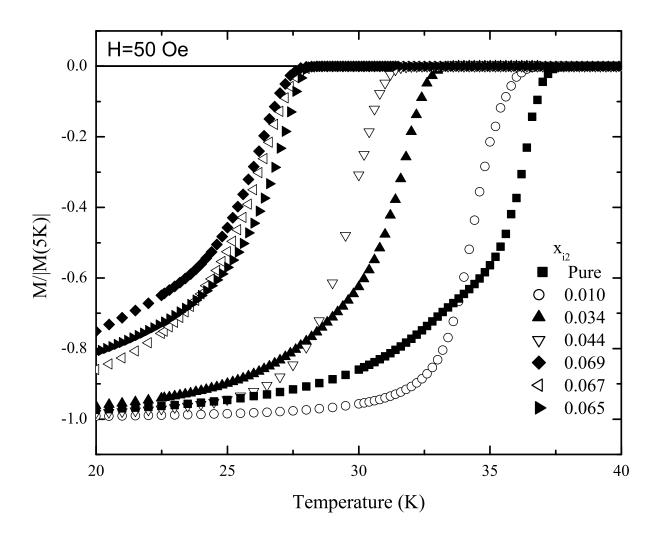
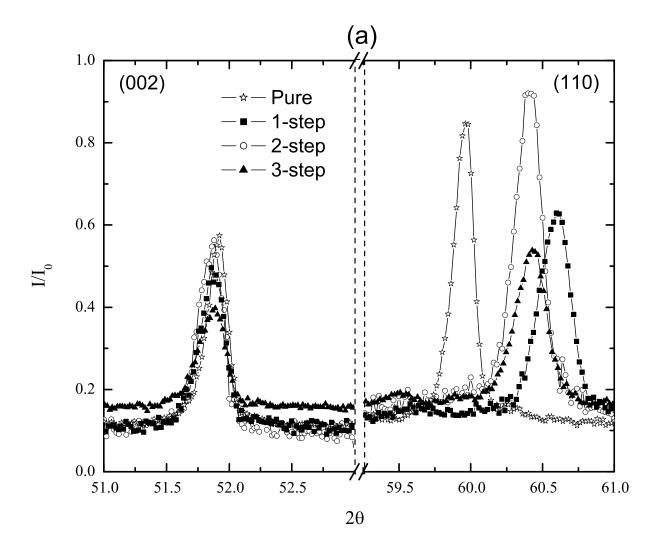


Fig. 6. Normalized magnetic transitions for the series of $Mg(B_{1-x}C_x)_2$ with x_{i2} =0.01, 0.034, 0.044, 0.069, 0.067, 0.065, synthesized with 0.995 purity B and reacted using a two step process.



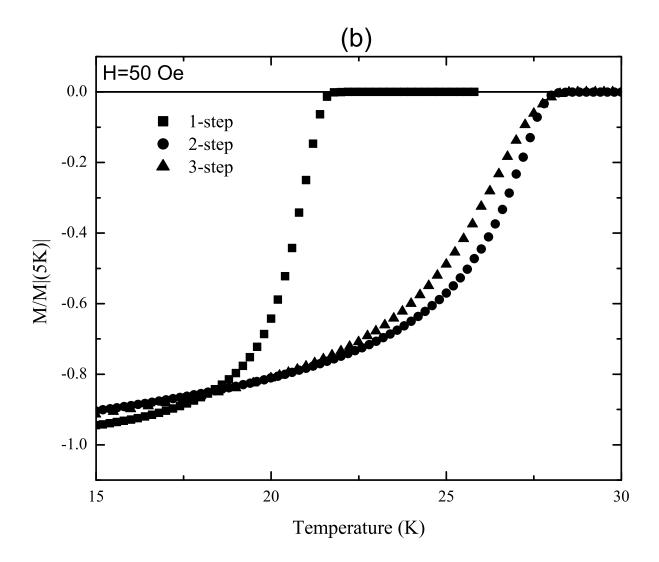
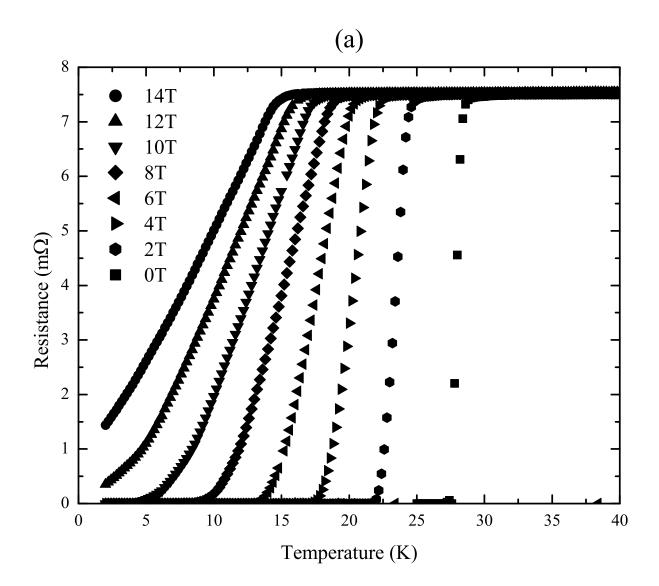


Fig. 7. (a) (002) and (110) x-ray peaks for nominal $Mg(B_{0.8}C_{0.2})_2$ using B_4C as the boron and carbon source and reacted using 1, 2, and 3 step reaction processes. (b) Normalized magnetic transitions for these samples.



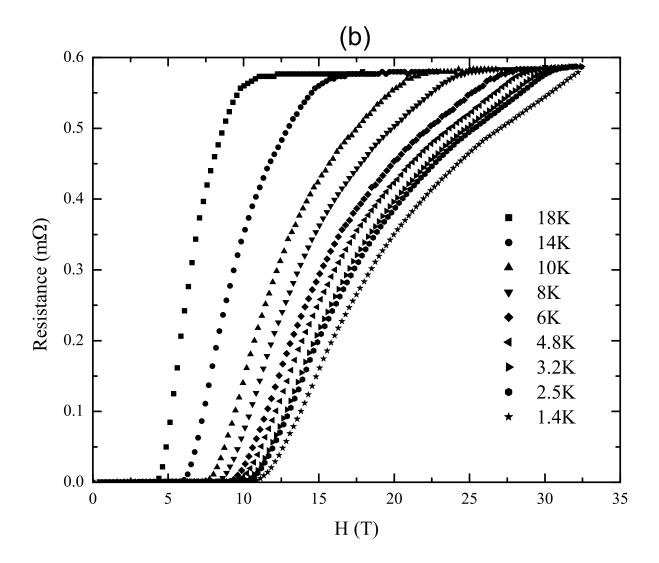


Fig. 8. (a) Resistance versus temperature and (b) resistance versus field for a sample with x_{i2} =0.069.

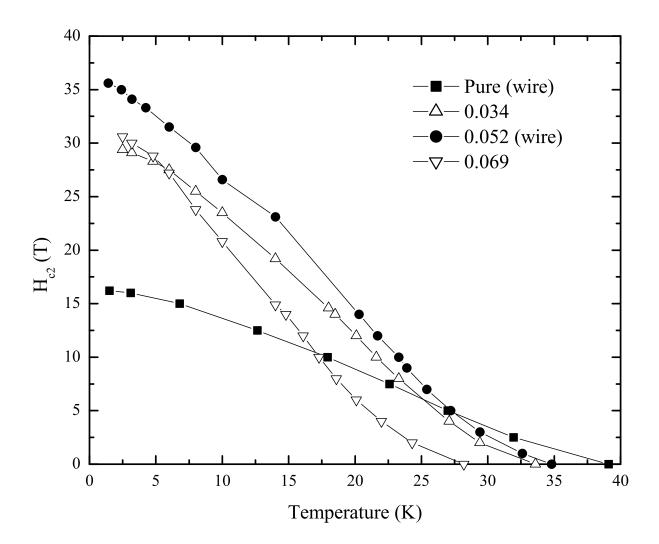


Fig. 9. Comparison of upper critical field curves for pellets with carbon doping levels of $x_{i2}=0.034$ and 0.069 with wires containing $x_i=0$ and 0.052. The $x_i=0$ and 0.052 samples were made by reacting Mg vapor with boron filaments, see reference [7].

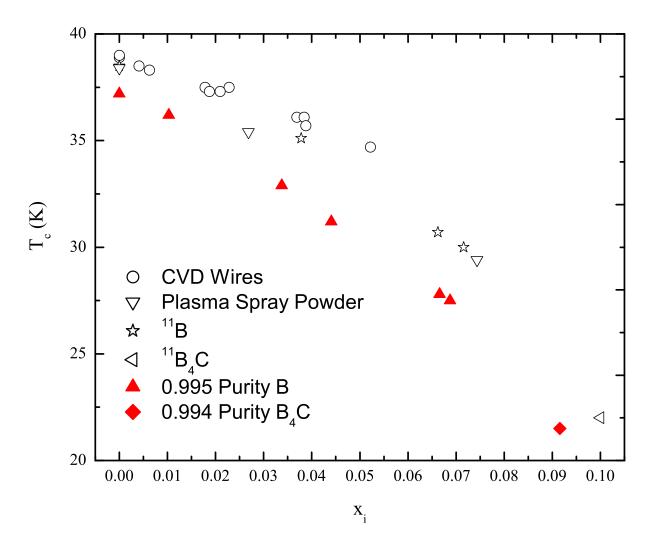


Fig. 10. Evolution of T_c as a function of x_i . CVD wires and powders are from references [12] and [24] respectively. Data on the sample prepared with isotopically enriched $^{11}B_4C$ is from reference [20].

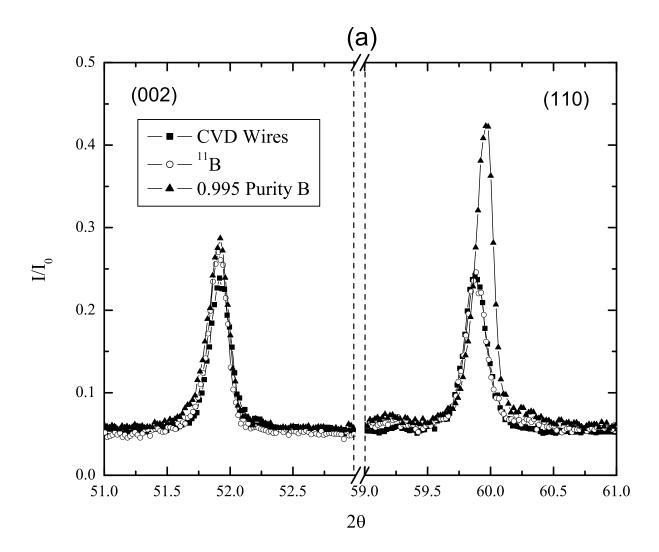


Fig. 11. Comparison of (002) and (110) x-ray diffraction peaks for pure MgB_2 made using different purity boron as the starting material. The 0.995 purity shows a shift in the (110) peak which presumably is not a result of inadvertent carbon doping.

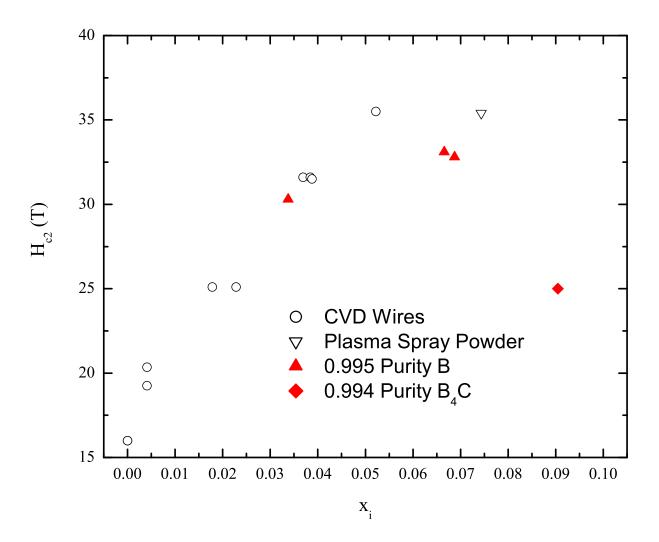


Fig. 12. $H_{c2}(T=0)$ curves for the samples with different purity in the starting boron. CVD wires and plasma spray powders are from references [12] and [24] respectively. H_{c2} data on the highest doping level is from reference [25].