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## Negative Magnetoresistance and Magnetic Susceptibility of Boronated Graphite

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Boronated graphite specimens were prepared from a grafoil heat-treated at 3000°C for 30 min. The transport properties, such as the electrical resistivity, Hall coefficient and transverse magnetoresistance, and the total magnetic susceptibility were measured for the pristine and boronated specimens. For the specimen with high boron concentration, a negative transverse magnetoresistance at liquid nitrogen temperature and weakly temperature-dependent resistivity were observed. Assuming that the negative magnetoresistance is due to a three-dimensional weak localization, a calculation was carried out for the temperature dependence of the resistivity and transverse magnetoresistance at low temperatures in terms of Slonczewski-Weiss-McClure band model and a weak localization theory obtained by extending Kawabata's theory. The experimental finding agreed well with that the calculation predicted.

**Keywords:** boronated graphite; magnetic susceptibility; negative magnetoresistance

### INTRODUCTION

Boron atoms dissolve graphite lattice substitutionally, of which maximum solid solubility for Natural Madagascar Graphite is 2.35 at. % at 2350°C<sup>[1]</sup>. Soule and Hishiyama et al. reported independently negative magnetoresistance at liquid nitrogen temperature for boronated graphite, when a sufficient amount of boron was substituted into natural graphite and/or kish graphite<sup>[2,3]</sup>. Soule observed small values of diamagnetic total susceptibility at room temperature for the specimens which showed the negative magnetoresistance<sup>[2]</sup>. Recently,

Hishiyama *et al.* prepared boronated natural graphite compacts and found that at liquid nitrogen temperature the boronated graphite compacts except slightly boronated ones show the negative transverse magnetoresistance and the resistivity almost independent of temperature<sup>[4]</sup>.

In the present paper, the transport properties such as the electrical resistivity, Hall coefficient and transverse magnetoresistance, and the total magnetic susceptibility were measured for boronated graphite, using a heat-treated grafoil as the pristine material. The goal of the study is to investigate the negative magnetoresistance of the sufficiently boronated graphite.

## EXPERIMENTAL

Commercially available grafoil with 100  $\mu\text{m}$  in thickness was heat-treated at 3000°C for 30 min to use as the pristine material for boronation. Boronation was carried out by heating a crucible at 2500°C for 30 min, which contained pristine specimens of proper size and a mixture of powders of  $\text{B}_4\text{C}$  and graphite (3000°C-treated petroleum coke), the specimens being embedded in the mixture.

For the pristine and boronated specimens, the interlayer spacing  $d_{002}$  and mosaic spread were obtained, using  $\text{Cu K}\alpha$  radiation. Each of pristine and boronated specimens was mounted on a specially designed sample holder for foils and the 004 and 006 diffraction lines were measured to determine  $d_{002}$ . The mosaic spread gives an information on the degree of orientation of the  $\text{c}$ -axes of crystallites normal to the specimen surface and defined by the full width at the half-maximum of the peak intensity recording of 002 diffraction plotted as a function of rotation angle of the specimen.

The electrical resistivity  $\rho$  was measured by a dc method at room and liquid nitrogen temperatures. The Hall coefficient  $R_H$  and magnetoresistance were also measured by a dc method at liquid nitrogen temperature with the magnetic field perpendicular to the specimen surface, in fields up to 1 T. The magnetoresistance measured was the transverse magnetoresistance  $\Delta\rho/\rho_0$ .

For the selected specimens of sufficiently boronated ones, the temperature dependence of the resistivity in a temperature range between 1.7 and 273 K and also the field dependence of  $\Delta\rho/\rho_0$  in fields up to 6.5 T at liquid helium temperature were measured.

The magnetic susceptibility was measured at room temperature for the pristine specimen and selected specimens of boronated ones, using a squid magnetometer. The magnetic field was applied either parallel (//) or perpendicular ( $\perp$ ) to the specimen surface up to 5T. Thus the total susceptibility  $\chi_T \equiv 2\chi_{||} + \chi_{\perp}$  was determined.

## RESULTS AND DISCUSSION

The boronated specimens could be classified into two classes according to the sign of  $\Delta\rho/\rho_0$  measured at liquid nitrogen temperature, i.e. a class of slightly boronated specimens and that of sufficiently boronated specimens. The slightly boronated specimens showed positive values of  $\Delta\rho/\rho_0$  in the entire field range investigated and were characterized by a temperature dependent resistivity and temperature and field dependent  $R_H$ . For the sufficiently boronated specimens negative values of  $\Delta\rho/\rho_0$  were measured at liquid nitrogen temperature. Almost temperature independent resistivity and almost temperature and field independent  $R_H$  were characteristics of the sufficiently boronated specimens.

For the slightly boronated specimens, the  $\Delta\rho/\rho_0$  measured in the field of 1 T ( $[\Delta\rho/\rho_0]_{77K,1T}$ ) was used for a measure of the substituted boron concentration, because  $\Delta\rho/\rho_0$  decreases with increasing substituted boron concentration<sup>[2]</sup>. For the sufficiently boronated specimens, the number of holes per carbon atom  $h/C$  determined from  $R_H$  was employed for a measure of boron atom ratio B/C, the number of boron atoms per carbon atom, because the conduction carriers are holes only.

All boronated specimens were well crystallized, because the specimens

showed splitting of each of the 004 and 006 diffraction lines due to  $K\alpha_1$  and  $K\alpha_2$ . For the slightly boronated specimens, the same values of  $d_{002}$  as that of the pristine grafoil, 0.3356 nm, were obtained. On the other hand, the sufficiently boronated specimens had  $d_{002}$  values lower than that of the pristine specimen, which decreased gradually with increasing  $h/C$ . The  $d_{002}$  value of the specimen B-N-0.7 ( $h/C = 0.0028$ ) in Fig. 1, one of the sufficiently boronated specimens, was 0.3349 for example. The values of the mosaic spread for the pristine and all boronated specimens were measured as the same values of  $20^\circ$ , indicating well-oriented constituent crystallites for each specimen.

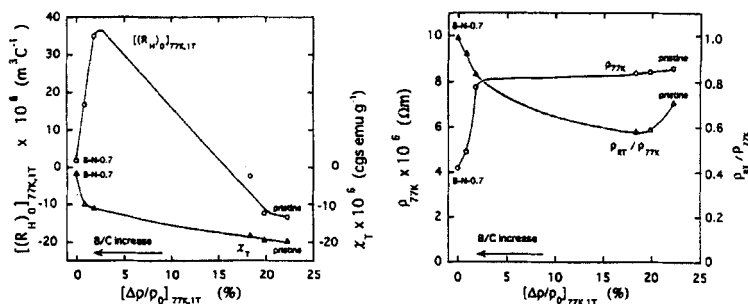


FIGURE 1  $[(R_H)_0]_{77K}$ ,  $\chi_T$  at room temperature,  $\rho_{77K}$  and  $\rho_{RT}/\rho_{77K}$  as a function of  $[\Delta\rho/\rho_0]_{77K,1T}$  for the pristine and boronated specimens.

Results of  $R_H$  extrapolated to zero field  $[(R_H)_0]_{77K}$ ,  $\rho_{77K}$ , the resistivity ratio  $\rho_{RT}/\rho_{77K}$ , where  $\rho_{RT}$  and  $\rho_{77K}$  are the resistivities at room and liquid nitrogen temperatures, and  $\chi_T$  for the pristine and boronated ones are shown in Fig. 1 plotted as a function of  $[\Delta\rho/\rho_0]_{77K,1T}$ . Notable changes were observed for the values of  $[(R_H)_0]_{77K}$ ,  $\chi_T$  and  $\rho_{77K}$  at the same boron concentration. The negative values of  $\Delta\rho/\rho_0$  at liquid nitrogen temperature were observed for the specimens with  $h/C$  higher than 0.00026. The value of  $[\Delta\rho/\rho_0]_{77K,1T}$  for the specimen B-N-0.7 in Fig. 1 was  $-0.0717\%$ .

$[\Delta\rho/\rho_0]_{T_{K,1T}}$  for the sufficiently boronated specimen was found to increase gradually with increasing  $h/C$ .

For materials with a disordered carrier system, weak localization is known to occur occasionally. The weak localization is a low temperature effect and causes enhancement of the resistivity  $\delta\rho$ , which superposes on the Boltzmann type resistivity as demonstrated in Fig. 2 (a). The absolute values of  $[\Delta\rho/\rho_0]_{T_{K,1T}}$  for the sufficiently boronated specimens were so small that the decrease of the resistivity by application of magnetic field can be assumed to be due to suppression of weak localization by application of magnetic field, because the carrier system is disordered by randomly distributed boron atoms. However, the present boronated specimens are well crystallized, so that the three-dimensional weak localization can be assumed at low temperatures for the carrier system.  $\delta\rho$  and the dependence of  $\Delta\rho/\rho_0$  on magnetic field  $B$  in low fields at low temperatures due to the three-dimensional weak localization were calculated in terms of Slonczewski-Weiss-McClure band model and a weak localization theory obtained by extending Kawabata's theory<sup>[5-7,8]</sup>. The calculation showed that  $\delta\rho \propto T^{1/2}$  and  $|\Delta\rho/\rho_0| \propto B^{1/2}$ .

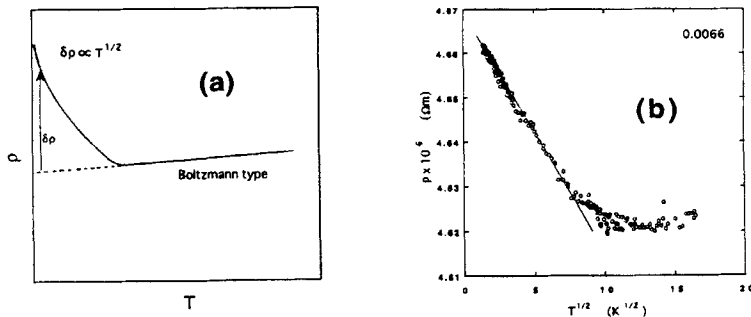


FIGURE 2 Schematic illustration of temperature dependence of resistivity  $\rho$  for sufficiently boronated specimen (a) and experimental result of  $\rho$  for specimen with  $h/C$  of 0.0066 plotted as a function of  $T^{1/2}$  (b).

As an example of the experimental results of the temperature dependence of the resistivity, the result for the sufficiently boronated specimen with  $h/C$  of

0.0066 is shown in Fig. 2 (b). Similar results were obtained for other sufficiently boronated specimens. The results agreed well with the calculation on the temperature dependence of the resistivity. The dependence of  $\Delta\rho/\rho_0$  on  $B^{1/2}$  measured at liquid helium temperature for the specimens with  $h/C$  of 0.0012, 0.0037 and 0.0066 is shown in Fig. 3. The  $B^{1/2}$  dependence of  $\Delta\rho/\rho_0$  can be seen partly in low fields, as the calculation predicted.

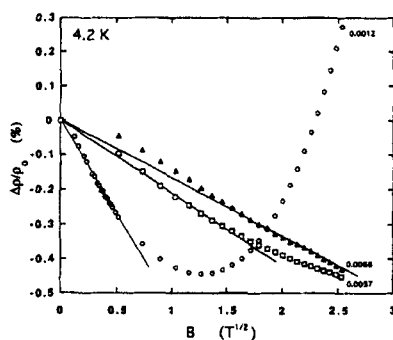


FIGURE 3  $\Delta\rho/\rho_0$  vs.  $B^{1/2}$  plots at 4.2 K for specimens with  $h/C$  of 0.0012, 0.0037 and 0.0066.

### Acknowledgements

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### References

- [1] C. E. Lowel, *J. Amer. Ceram. Soc.*, **50**, 142 (1967).
- [2] D. E. Soule, *Proceedings of the Fifth Conference on Carbon*, Vol. 1, Pergamon Press, Elmsford, N. Y., 1962, p. 13.
- [3] Y. Hishiyama, S. Mrozowski, and A. S. Vagh, *Carbon*, **9**, 367 (1971).
- [4] Y. Hishiyama, Y. Kaburgi, K. Kobayashi, and M. Inagaki, *Molecular Crystals & Liquid Crystals*, **310**, 279 (1998).
- [5] J. C. Slonczewski, and P. R. Weiss, *Phys. Rev.*, **109**, 272 (1958).
- [6] J. W. McClure, *Phys. Rev.*, **108**, 272 (1957).
- [7] J. W. McClure, *Phys. Rev.*, **119**, 606 (1960).
- [8] A. Kawabata, *J. Phys. Soc. Jpn.*, **49**, 628 (1980).