Negative magnetoresistance in Boronated Graphite

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Introduction

Low temperature behaviors of the transport properties have been the subject of great interest in pregraphitic carbons and low-stage acceptor graphite intercalation (GIC) compounds as the two-dimensional (2D) weakly disordered electronic system[1,2]. Negative magnetoresistance is a typical phenomena due to the 2D weak localization of carriers. The weak localization occues when partial waves interfere constructively in the back-partial waves in the back-ward direction in a zero magnetic field. The application of a magnetic field perpendicular to the 2D carrier system produces a phase difference between the partial waves and back-partial waves, so the effect of weak localization is reduced, and a small negative magnetoresistance is observed[1]. In addition, an anomalous behavior ascribed to the weak localization and intercalation effects is observed in the low temperature electrical resistivity of the low-stage acceptor GICs; a logarithmic increase of the resistivity with decreasing temperature[2].

On the other hand, when boron was introduced by heating a kish graphite with boric acid at 2500℃, the resulting boronated graphite shows the negative magnetoresistance at 77 K[3]. Boron atoms are known to go into substitutional sites in the graphite lattice, and behave as acceptors, depressing the Fermi level of graphite[4]. In the present study, we investigate the electrical properties in boronated graphite compacts prepared from natural geaphite powder and boron carbide B_4C powder under pressure, focusing on the weak localization effect.

Experimental

Natural graphite powder was heat-treated at 2200℃ for 2 hrs with B_4C powder under a pressure of 19.6 MPa. The values of the interlayer spacing d_{002} of the boronated graphite compacts prepared are listed in Table 1. For the electrical measurements, a bridge-shaped specimen was cut along each compact surface perpendicular to the pressure direction. Each specimen obtained was designated by B_4C followed by the value of nominal content from 1 to 50wt%. Electrical resistivity at room temperature (RT) ρ_{RT}, resistivity ratios ρ_{RT}/ρ_{77K} and $\rho_{RT}/\rho_{4.2K}$, where ρ_{77K} and $\rho_{4.2K}$ are the resistivity at 77 and that at 4.2 K, transverse magnetoresistance $\Delta\rho/\rho$ and Hall coefficient R_H at RT and 77 K were measured by a dc method for the specimens and pristine natural graphite compact. For B_4C-2.5wt%, $\Delta\rho/\rho$ and R_H were measured at 4.2 K in the fields up to 6.5 T, and electrical resistivity was also measured in the temperature range between 1.4 and 290 K.

Results and Discussion

Values of ρ_{RT} and R_H at 77 K and 1 T for the specimens are shown in Table 1. For each boronated specimen, a constant positive R_H against magnetic field was observed, and the values at RT are the same as those at 77 K, and as that at 4.2 K in B_4C-2.5wt%. The results are consequence of the substitution of B atoms into graphite lattice. The values of d_{002} and ρ_{RT} decreased by the boronation in B_4C-1 and -2.5wt%, while they increased in B_4C-50wt%. This suggests that the boronation proceeds in two steps: substitution of B atoms into graphite lattice and then intercalation into interstitial sites. The concentration of B atoms substituted was estimated from R_H data, and the values for the boronated specimens are listed in Table 1. From these data, the maximum concentration of B in the graphite lattice is evaluated to be about 0.4 at%.

Large positive $\Delta\rho/\rho$ was observed for the pristine graphite compact at RT and 77 K as listed in Table 1. As shown in Fig. 1, small negative $\Delta\rho/\rho$ was observed at 77 K for all the boronated specimens, but no $\Delta\rho/\rho$ was observed at RT, exhibiting the weak localization effect at low temperatures. $\Delta\rho/\rho$ measured at 4.2 K for B_4C-2.5wt% is shown in Fig. 2. The absolute value of $\Delta\rho/\rho$ is much larger than that at 77 K, and $\Delta\rho/\rho$ decreases rapidly in the low field region, in contrast with that at 77 K. In the high field region it becomes to saturates. This behavior of $\Delta\rho/\rho$ is similar to that for the low-stage acceptor GICs[2].

Values of ρ_{RT}/ρ_{77K} and $\rho_{RT}/\rho_{4.2K}$ for the specimens are listed in Table 1. The values for the boronated graphite specimens indicate an existence of the resistivity minimum at a low temperature. Fig. 3 shows the electrical resistivity as a function of square root of temperature $T^{1/2}$ down to 1.4 K for B_4C-2.5wt%. The resistivity minimum was found at a temperature around 110～160 K, and the resistivity increases with decreasing temperature as proportional to $T^{1/2}$ at temperatures below about 64 K. The results may also be attributed to the weak localization effect. However, this behavior is different from that for the low-stage acceptor GICs; a logarithmic increase of the resistivity with decreasing temperature[2]. Theoretical consideration will be needed to account for the present phenomena.

References

1. Bayot, V., Piraux, L., Michenaud, J.-P. and Issi, J.-P., *Phys. Rev. B*, 1989, **40**, 3514.
2. Piraux, L., Bayot, V., Gonze, X., Michenaud, J.-P. and Issi, J.-P., *Phys. Rev. B*, 1987, **36**, 9045.
3. Hishiyama, Y., Mrozowski, S., and Vagh, A. S., *Carbon*, 1971, **9**, 367.
4. Klein, C. A., *J. Appl. Phys.*, 1962, **33**, 3338.

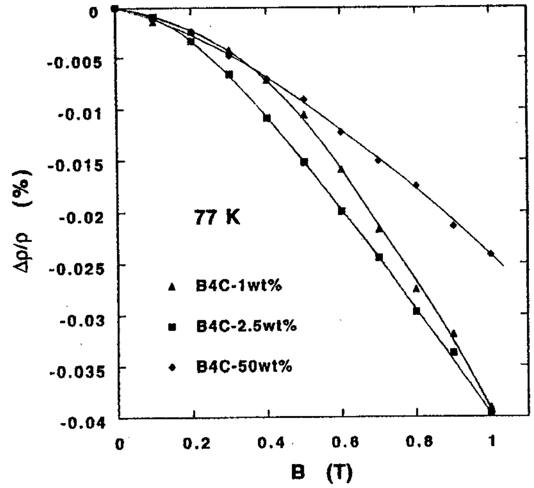

Fig. 1. Transverse magnrtoresistance as a function of magnertic field at 77 K for the boronated graphite specimens.

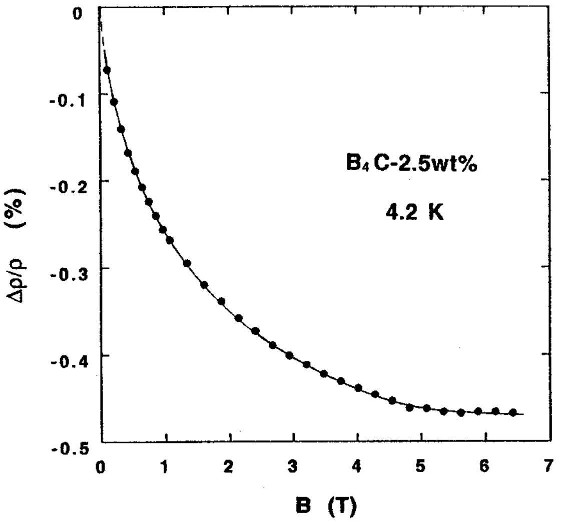

Fig. 2. Transverse magnrtoresistance as a function of magnetic field at 4.2 K for B_4C-2.5wt%.

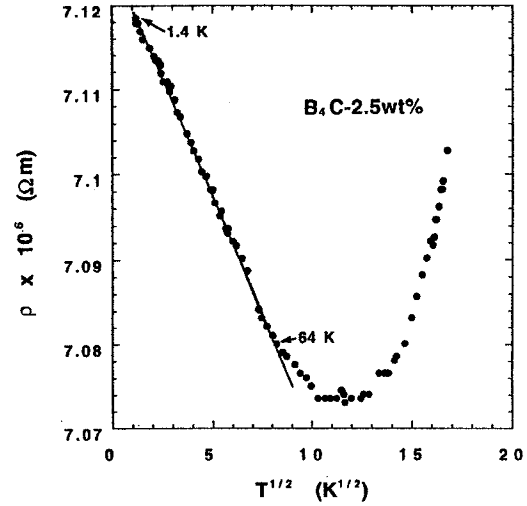

Fig. 3. Electrical resistivity as a function of square root of temperature for B_4C-2.5wt%.

Table 1. Parameters for the boronated and pristine graphite specimens.

sample code	d_{002} (nm)	ρ_{RT}x10^6 (Ωm)	ρ_{RT}/ρ_{77K}	$\rho_{RT}/\rho_{4.2K}$	$(\Delta\rho/\rho)_{77K,1T}$ (%)	$(R_H)_{77K,1T}$(x10^8m^3/C)	n_B (at%)
Pristine	0.3354	13.5	0.727	0.667	22.45	-7.38	-----
B_4C-1wt%	0.3351	6.38	1.007	-----	-0.0391	1.44	0.37
B_4C-2.5wt%	0.3349	8.5	1.004	0.9929	-0.0396	1.29	0.41
B_4C-50wt%	0.3361	18.6	1.005	0.9992	-0.0242	1.30	0.40