



Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
<http://www.tandfonline.com/loi/gmcl19>

Effect of Bromine Intercalation on Thermomagnetic Properties of Pyrolytic Graphite "Nikafilm"

Takafumi Ono ^a, Tomohiko Ishii ^b, Ikushi Yoshida ^a & Seiichi Tanuma ^c

^a Department of Materials Science, Iwaki Meisei University, Iwaki, Fukushima, 970-8551, Japan

^b Department of Chemistry, Tokyo Metropolitan University, Hachioji, Tokyo, 192-0397, Japan

^c 2100-174, Horiuchi, Hayama, Kanagawa, 240-0112, Japan

Version of record first published: 24 Sep 2006

To cite this article: Takafumi Ono, Tomohiko Ishii, Ikushi Yoshida & Seiichi Tanuma (2000): Effect of Bromine Intercalation on Thermomagnetic Properties of Pyrolytic Graphite "Nikafilm", Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 340:1, 179-184

To link to this article: <http://dx.doi.org/10.1080/10587250008025463>

Full terms and conditions of use: <http://www.tandfonline.com/page/terms-and-conditions>

This article may be used for research, teaching, and private study purposes. Any substantial or systematic reproduction, redistribution, reselling, loan, sub-licensing, systematic supply, or distribution in any form to anyone is expressly forbidden.

The publisher does not give any warranty express or implied or make any representation that the contents will be complete or accurate or up to date. The accuracy of any instructions, formulae, and drug doses should be independently verified with primary sources. The publisher shall not be liable for any loss, actions, claims, proceedings, demand, or costs or damages whatsoever or howsoever caused arising directly or indirectly in connection with or arising out of the use of this material.

Effect of Bromine Intercalation on Thermomagnetic Properties of Pyrolytic Graphite "Nikafilm"

TAKAFUMI ONO^a, TOMOHIKO ISHII^b, IKUSHI YOSHIDA^a and
SEIICHI TANUMA^c

^a*Department of Materials Science, Iwaki Meisei University, Iwaki, Fukushima, 970-8551, Japan,* ^b*Department of Chemistry, Tokyo Metropolitan University, Hachioji Tokyo 192-0397 Japan and* ^c*2100-174, Horiuchi, Hayama, Kanagawa, 240-0112 Japan*

Nernst-Ettingshausen effect and longitudinal Nernst-Ettingshausen effect of the "NIKA-FILM" and bromine-graphite compound were studied at applied magnetic field strength 1.4 (T). The Nernst-Ettingshausen coefficient of the bromine-graphite compound decreased 23% in comparison with that of pristine "NIKAFILM". Similarly the longitudinal Nernst-Ettingshausen effect was 54% smaller for the bromine-graphite compound in comparison with the "NIKAFILM".

Keywords: NIKAFILM; Thermomagnetic effect; Nernst-Ettingshausen effect; longitudinal Nernst-Ettingshausen effect; Graphite-bromine compound

INTRODUCTION

Graphite-bromine compound is formed with the bromine gas by the reaction of the graphite at the room temperature. When the bromine vapor pressure increases, the graphite begins to absorb the bromine gas at around

10% of the partial pressure at which $\text{CBr}_{0.125}$ is formed, and then $\text{CBr}_{0.05}$, $\text{CBr}_{0.063}$ and $\text{CBr}_{0.083}$ are formed successively as the partial pressure increases, and finally, $\text{CBr}_{0.125}$ is formed. It easily decomposes to the chemical compound CBr_x when bromine vapor pressure decreases, x depending on the vapor pressure. However, residual chemical compounds exist even when the bromine vapor pressure is reduced to zero. It is reported that this residual chemical compound is near to $\text{CBr}_{0.036}$ [1]. The compound $\text{CBr}_{0.125}$ is known as an acceptor type Graphite Intercalation Compound (GIC) [2]. The electronic structure of CBr_x was studied by means of cyclotron resonance and de Hass-van Alphen effect [3]. According to them, three hole sheets were observed for $x=0.027$ and $x=0.032$. The effective mass for $x=0.027$ is $m_1=0.058$, $m_2=0.080$, $m_3=0.110$, and for $x=0.032$ it is $m_1=0.063$, $m_2=0.088$, $m_3=0.110$. In this paper, the study of thermomagnetic effect in graphite-bromine compound was conducted. The thermomagnetic effect is strongly influenced by the scattering mechanism of conduction electrons [4]. The study of the effects is, therefore, expected to afford a crucial information on the type of scattering mechanism at the staging phase transition of GICs.

EXPERIMENTAL PROCEDURES

The pristine graphite sample which we used, "NIKAFILM", is produced by Nippon Carbon Co.. For the preparation of bromine-graphite compound, vapor pressure was controlled by varying the concentration of bromine solution diluted with CCl_4 . The thermomagnetic effect was measured as a function of reaction time during the intercalation of Br_2 into graphite. The study of the Nernst-Ettingshausen effect or longitudinal Nernst-Ettingshausen effect in the "NIKAFILM" and graphite-bromine compound were carried out near the

room temperature. The magnetic field was applied by a 1.4 (T) split-type electromagnet. There is a temperature gradient along the length of a specimen and the magnetic field is applied normal to the temperature gradient, therefore, a potential difference is brought forth in the direction perpendicular both to the temperature gradient and to the magnetic field. The Nernst-Ettingshausen coefficient Q_{NE} is defined as

$$V_y = Q_{NE} \cdot B \cdot w \cdot \Delta T / \Delta L, \quad (1)$$

where V_y is the Nernst-Ettingshausen voltage, $\Delta T / \Delta L$ is the temperature gradient, B is the magnetic field strength, and w is the width of specimen. The longitudinal Nernst-Ettingshausen effect is the change of the thermoelectric power on applying a magnetic field. The longitudinal Nernst-Ettingshausen coefficient A_{LNE} is defined as

$$E_x(B) - E_x(0) \equiv \{S(B) - S(0)\} (\Delta T / \Delta L) = A_{LNE} (\Delta T / \Delta L), \quad (2)$$

where E_x is the electric field produced by the magnetic field B ; $E_x(B)$ and $E_x(0)$ are electric field along the length of specimen in the presence and in the absence of a magnetic field, respectively; $S(B)$ and $S(0)$ are the thermoelectric powers when $B \neq 0$ and $B=0$.

EXPERIMENTAL RESULTS AND DISCUSSION

The magnetic field dependence of the Nernst-Ettingshausen voltage divided by the temperature difference of "NIKAFILM" measured at 310K is shown in Fig. 1. In the range from 0 (T) to +0.8 (T), $V_y / \Delta T$ increased proportionally to the magnetic field strength B . In the range from 0.8(T) to 1.28 (T), it increased non-linearly against the applied magnetic field strength. The field dependence described above remained the same when the magnetic field direction was reversed. Naturally, the sign of V_y changed from positive

to negative on reversing the direction of magnetic field. The Nernst-Ettingshausen coefficient in "NIKAFILM" is estimated from equation (1), *i.e.*, it is 1.98×10^{-5} ($\text{m}^2/\text{s} \cdot \text{K}$) at 310K. Two other samples were prepared from the identical sheet of "NIKAFILM", and the measurement was repeated. The result is: Q_{NE} is $1.73 \times 10^{-5}(\text{m}^2/\text{s} \cdot \text{K})$ at 317K for the sample #2 and $2.09 \times 10^{-5}(\text{m}^2/\text{s} \cdot \text{K})$ at 307K for the sample #3.

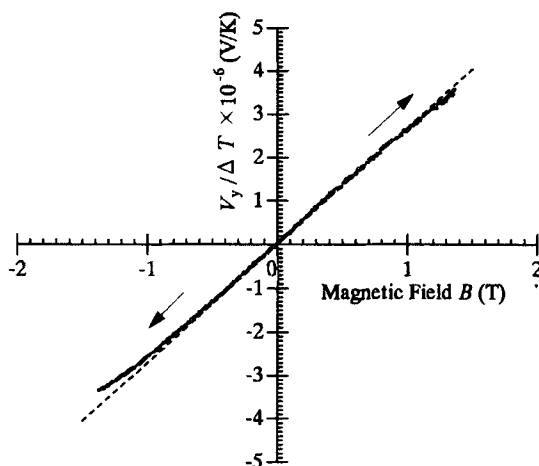


Fig. 1. Relationship between the Nernst-Ettingshausen voltage divided by the temperature difference, $V_y / \Delta T$, and the magnetic field B at 310K. The broken line is the fitting line by least squares method.

The Nernst-Ettingshausen effect in the bromine-graphite compound was measured. $\text{Br}-\text{CCl}_4$ solution for this measurement was $\text{Br}_2:\text{CCl}_4=1:30$ (in volume). The measurement was carried out on the bromine-graphite compound in a fixed bromine vapor pressure, because the compound decomposes when vapor pressure decreases. The Seebeck coefficient was measured as the reaction proceeds, and determined as a function of reaction time. The Seebeck coefficient took a saturated value for the reaction time longer than 60,000 seconds, so that the compound is considered to be fully

stabilized. The thermomagnetic effect was measured with varying magnetic field strength in this stabilized condition of compound.

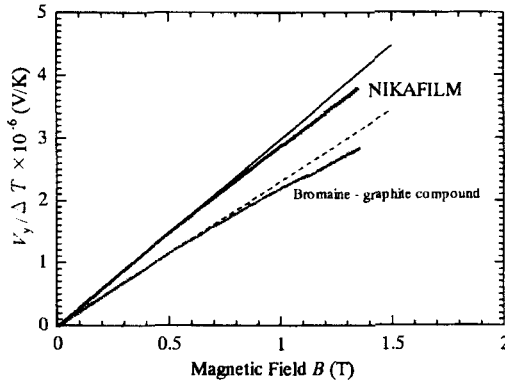


Fig. 2. Magnetic field dependence of the $V_y / \Delta T$ in NIKAFILM and bromine-graphite compound at 317K. The straight and broken line is the fitting line by least squares method.

The fitting in the weak magnetic field region is carried out using the least squares method, and the Nernst-Ettingshausen coefficient in “NIKAFILM” and bromine-graphite compound are estimated. The coefficient of “NIKAFILM” and bromine-graphite compound were estimated as $1.73 \times 10^{-5} (\text{m}^2/\text{s} \cdot \text{K})$ and $1.33 \times 10^{-5} (\text{m}^2/\text{s} \cdot \text{K})$, respectively. As a result, the Nernst-Ettingshausen coefficient of the bromine-graphite compound decreased 23% in comparison with that of “NIKAFILM” (Fig. 2). However, it deviated from the straight line for a weak field approximation from 0.6(T) to 1.36(T) for both samples. A similar result to this finding was observed for the InSb compound[5]. According to I.M.Tsidil'kovskii[6], two terms, one proportional to B and the other proportional to B^{-1} , contribute to the Nernst-Ettingshausen coefficient with a different sign with each other, which at least qualitatively elucidates the above mentioned behaviors. Next, the longitudinal Nernst-Ettingshausen effect was measured. The magnetic field

dependence of the longitudinal Nernst-Ettingshausen effect, $\{S(B)-S(0)\}/S(0)$, was B^2 in the weak field region, while a tendency to approach B^1 dependence was observed for higher magnetic field strength. For the bromine-graphite it decreased 76% in comparison with that of "NIKAFILM".

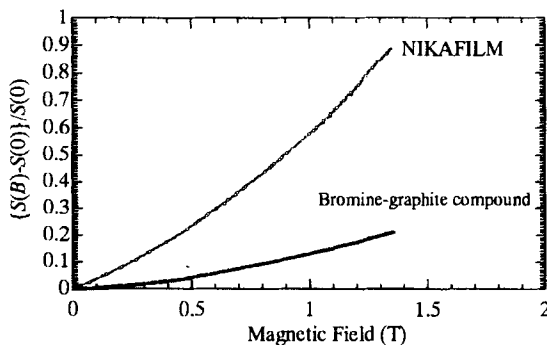


Fig.3 Magnetic field dependence of the $\{S(B)-S(0)\}/S(0)$ in NIKAFILM and bromine-graphite compound at 317K.

Acknowledgment

This work was financially supported in part by a grant-in-aid from Japan Ministry of Education. The authors express their thanks to Messrs. Naoyuki Hobara and Hiroshi Honma for their assistance in the experiment.

References

- [1] W.T. Eeles, J.A. Turnbull, *Proc. R. Soc.*, **283A**, 179(1965).
- [2] T. Sasa, Y. Takahashi, and T. Mukaibo, *Carbon*, **9**, 407(1971).
- [3] H. Suematu, S. Tanuma and K. Higuti, *Physica*, **99B**, 420(1980).
- [4] R. A. Smith: *Semiconductors*, (Cambridge University Press Cambridge, 1959), p.183.
- [5] V. P. Zhuze and I. M. Tsidil'kovskii, *Zhur. Tekh. Fiz.*, **28**, 2372(1958).
- [6] I. M. Tsidil'kovskii: *Thermomagnetic effects in Semiconductors*, (Inforsearch Limited London, 1960), p.234.