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Thermopower of Pregraphitic Carbons

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The paper presents the results of investigations in thermoelectric power of pregraphitic carbons in the temperature range from 20 to 200K. The thermoelectric power of pregraphitic carbon has been shown to follow the model of sequential connection of islets with metallic conductivity, islets with hop conductivity of variable hop length and islets with hop conductivity of constant hop length. The description of thermoelectric power in islets of metallic conductivity requires the dependences of major mechanisms of charge carrier scattering on the temperature to be taken into account. The parameters of pregraphitic carbon electronic structure have been calculated from the experimental results on temperature dependence of electrical resistivity.

Keywords: pregraphitic carbon; thermoelectric power; hop conductivity; mobility edge

INTRODUCTION

Pregraphitic carbons are a large class of carbon-graphite materials produced by annealing of raw carbon at the temperatures up to 2000^0C. The structure of pregraphitic carbon represents a pack of two-dimensional band-shaped crystallites randomly oriented in space in relation to each other. They are deformed by various defects of their layers, but the graphite layers have two-dimensional symmetry however. The crystallite sizes L in pregraphitic carbon are no more than 100Å, the distance d_{002} between the layers is ~3.44Å. Such pregraphitic carbon structure differs essentially from the structure of highly-oriented pyrolytic graphite and this results in essential difference between

electro-physical characteristics of pregraphitic carbon and other carbon-graphite materials.

The aim of this work was to investigate the mechanism of thermoelectric power in pregraphitic carbon.

RESULTS AND DISCUSSION

In our work we investigated the mechanism of thermoelectric power behavior in pregraphitic carbons: ВПР-1 and ВПР-2 fibers produced from polyacrylonitrile, BMH fiber and AB1 and AB2 pregraphitic carbons. Thermoelectric power was studied in the temperature range from 20K to 200K by the technique described in [1]. Fig. 1 presents the typical temperature dependence of thermoelectric power for carbon materials with pregraphitic structure. As it is clear from Fig. 1 for all materials under study there is a minimum on S(T) dependence in the temperature range 30-40K. The thermoelectric power increases in all the samples with increase of temperature but the rate of increase is different for each sample. It has been found previously[1] that the electrical conductivity in the samples under study is proportional to $\exp(-T_o/T)^{1/4}$, (where T_o is const) in a rather wide temperature range. This corresponds to disordered systems with Anderson localization, i.e. with variable-length hopping conductivity. In ref. [2] the description of thermoelectric power in pregraphitic carbons took into account the contribution of regions exhibiting metallic conduction and variable-length hopping conduction connected in series. Each region is characterized by its own thermopower. The total thermopower can be presented as:

$$S=S_1+S_2 \qquad (1)$$

The thermoelectric power S_1 in the region with metallic conductivity can be presented as follows[3]:

$$S_1 = \frac{k_b}{e}\cdot\frac{\pi^2}{3}\cdot\frac{k_b T}{\Delta}\cdot(1+p), \qquad (2)$$

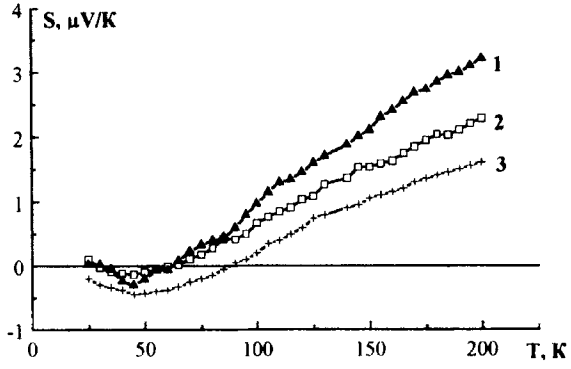

FIGURE 1. Temperature dependence of thermoelectric power S(T) in pregraphitic carbons and fibers: 1 - BMH fiber, 2 - AB2 pregraphitic carbon, 3 - AB1 pregraphitic carbon.

where k_b is Boltzmann constant, e is an electron charge, Δ is Fermi level shift, p is the parameter which is determined by the prevailing mechanism of charge carrier scattering. The thermoelectric power S_1 is positive since the regions with metallic conductivity represent a degenerate hole conductor. The electrical conductivity σ_2 and thermoelectric power S_2 of region with variable-length hopping can be presented as [2]:

$$\sigma_2 = \sigma_0 \exp\left[-\left(\frac{T_0}{T}\right)^{1/4}\right], \qquad S_2 = \frac{k_b^2}{2e}\cdot(T_0\cdot T)^{1/2}\cdot\left.\frac{d\ln N(E)}{dE}\right|_{E=E_F} \qquad (3)$$

where σ_0 and T_0 are constants, $N(E_F)$- is the density of states on Fermi level. The sign of S_2 is determined by the sign of the derivative of the density of states $N(E)$ with respect to energy dN/dE. However this approximation does not allow the low temperature anomalies and availability of positive "tail" on S(T) curves to be accounted for. For more precise descriptions of S(T) behavior we propose also to take into account the regions with hop conductivity of constant hop length in which thermoelectric power S_3 arises. The total thermoelectric power in this case can be presented as follows:

$$S = aS_1 + bS_2 + cS_3 \qquad (4)$$

a,b,c coefficients determine the fraction of each of the regions in the sample. Thermoelectric power in the regions with hop conductivity with constant hop length can be described by the following expression:

$$S_3 = \frac{k_b}{2e}\cdot\left(\frac{\varepsilon}{k_b T} + C\right), \qquad \varepsilon = E_C - E_F \qquad (5)$$

where E_C is the mobility edge, $\varepsilon > 0$, C is a constant that may be both negative and positive[4].

From the experimental data on temperature dependence of thermoelectric power at low temperature in AB1 and AB2 samples we have calculated the parameters: Δ - Fermi level shift in the regions with metallic conductivity, E_F - the value of Fermi energy in the regions with hop conductivity of variable hop length, ε - the difference between the mobility edge and Fermi level and C - the constant for the regions with hop conductivity of constant hop length. The results of the calculations are given in Table 1.

The calculated parameters for AB1 and AB2 pregraphitic carbons.

Sample	σ_0, $(\text{Ohm}\cdot\text{m})^{-1}$	T_0, K	E_F, eV	Δ, eV	ε, eV	C
AB1	169	1967	0.4	0.45	$3.8\ 10^{-5}$	0.01
AB2	436	552	0.22	0.35	$4.4\ 10^{-5}$	0.012

As it is seen from the table the calculated parameters Δ, E_F, ε, C and T_0 have quite reasonable values. By using the obtained values for Δ, E_F, ε, C and T_0 parameters the temperature dependence of thermoelectric power was calculated for AB1 sample in the temperature range up to 200K. It has been found that the calculated thermoelectric power S(T) satisfactorily correlates with that experimentally obtained at low temperatures but at higher temperatures there is a considerable (about two times) deviation. As it was

shown by K.Sugihara[5,6] the description of temperature dependence of diffusion thermoelectric power (eq.2) cannot assume p to be constant for a wide temperature range but should take into account all major mechanisms of charge carrier scattering resulting in complicated dependences of p on temperature. As it is clear from Fig.2 the account of temperature dependence of mechanisms of charge carrier scattering provides a good agreement of the results obtained by calculation and experiment.

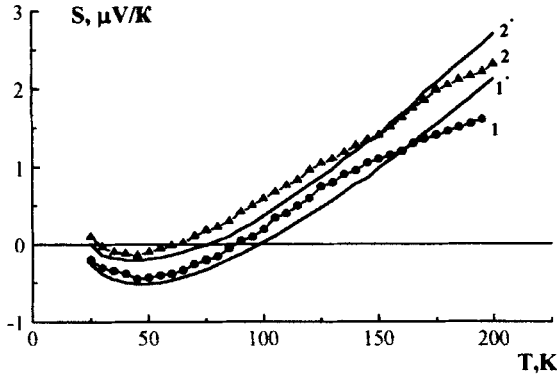

FIGURE 2 Temperature dependences of thermoelectric power of pregraphitic carbons: 1-AB1 carbon, 2 - AB2 carbon (experiment); 1*, 2* - the calculations in the framework of the model of sequential connection of islets with different types of conductivity by taking into account dependences of p on T.

The temperature dependences of thermoelectric power calculated with taking into account different fractions of islets with different types of conductivity showed that as the number of regions with metallic conductivity is growing and the number of regions with hop conductivity is decreasing with increase of the temperature of annealing the values of thermoelectric power at the same temperatures become larger, the rate of thermoelectric power

growth increases, the temperature of minimum S(T) is shifted to low temperature range and this is consistent with the experimental data of the samples under study. The increase in the fraction of regions of hop conductivity, i.e. the increase in fraction of amorphous phase results in thermoelectric power reduction to negligibly small values.

Thus, the performed investigations show that the thermoelectric power of pregraphitic carbons may be described by means of the model of sequential connection of islets with metallic and hop conductivity with variable and constant hop length. The model requires the extended description of diffusion thermoelectric power which takes into account different mechanisms of charge carrier scattering.

The presented results of investigations in thermoelectric power in pregraphitic carbons show that the behavior of temperature dependence of thermoelectric power and its value are very sensitive to the parameters of crystalline structure of carbon materials determined by the method and conditions of these materials production.

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