Influence of gamma irradiation on electrophysical properties of onion-like carbon

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Temperature dependence of the conductivity of onion-like carbon (OLC) was investigated before and after gamma irradiation. It is found that the temperature dependence of the conductivity is described by the mechanism of variable range hopping conductivity (VRHC). The density of states on Fermi level $N(E_F)$ is estimated on slope angle of curves $\sigma(T)$ in coordinates $\ln[\sigma(T)/\sigma_{293}] - T^{-1/2}$. The gamma irradiation leads to the increase $N(E_F)$ for OLC prepared by vacuum annealing of nanodiamond (ND) particles at temperatures 1400 K, 1650 K and to the decrease $N(E_F)$ for OLC produced by the annealing ND at temperatures 1850 K.

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1. Introduction

Recently the modifications of properties of the nanostructures by various irradiations attract the significant interest [1, 2]. It is related both with the scientific interest to mechanisms of influence of gamma irradiation, neutron irradiation, proton irradiation and electron irradiation on properties of materials and with the practical point of view. The gamma irradiation causes additional interest because its deeply gets into a material and can modify the volumetric properties of material. In this paper we give the results of influence of gamma irradiation with energies few eV on electrophysical properties of onion-like carbon.

2. Experimental and samples

Onion-like carbon (OLC) was synthesized by vacuum annealing of explosive nanodiamond with d~4.7 μm: the sample Dc-1 - at 1400 K, Dc-2 – at 1650 K, Dc-3 - at 1850 K [3, 4]. The straight dark contrast lines in micrographs (Fig. 1) correspond to the (111) crystallographic diamond planes of untransformers diamonds core. The distance between these lines is 2.06 A. Where the curved dark lines in samples correspond to the (0002) crystallographic graphite planes. The distance between these lines is 3.4-3.5 A. ND annealing at high temperature lead to transformation of each ND particle into onion-like carbon species. While ND primary particles form agglomerates OLC particles are combined into agglomerates consist of OLC primary cores joined by onion grapheme shells.

For electrical measurements the powder of OLC was pressed in a glass open tube. The electrical contacts were made by 0.1 mm silver wires. The temperature dependences of the conductivity $\sigma(T)$ were measured by four-point-probe technique in the temperature range 4.2-300 K. Our previous studies of powder carbon nanostructures were carried out by this method [4-6]

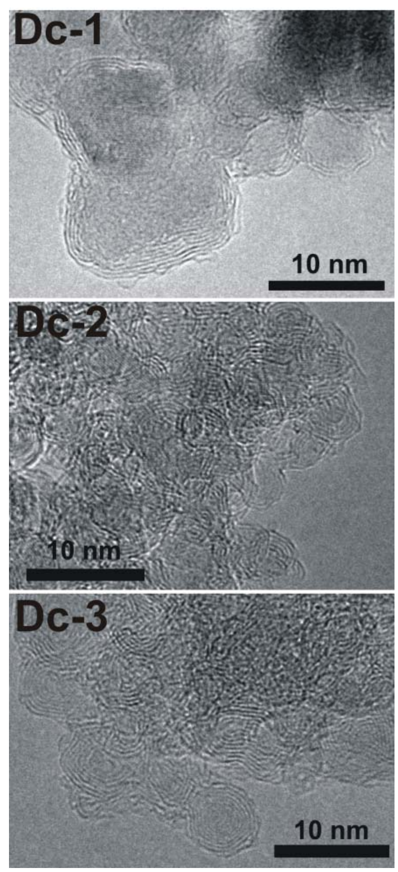

Fig. 1. HRTEM micrographs of OLC produced by annealing ND samples under a vacuum of 10^{-6} Torr for 1 h at 1400 K (Dc-1), 1650 K (Dc-2), 1850 K (Dc-3).

showed stability and reproducibility of results of the conductivity measurements.

3. Results and discussion

$\sigma(T)/\sigma_{293}$ before gamma irradiation

In corresponding with our previous study the concentration of current carriers of OLC grows with increase of the heat treatment temperature and conductivity is increased. Before measurements the sample was treated in vacuum (10^{-2} Torr) at 200^{o}C during 12 hours. After that the sample was placed on the holder of sample. We have carried out the degassing of measuring volume in vacuum (10^{-2} Torr) at room temperature during 1 hour and then the measuring volume was filled by gaseous helium. Fig. 2 shows the curves of temperature dependences of conductivity $\sigma(T)$ of OLC.

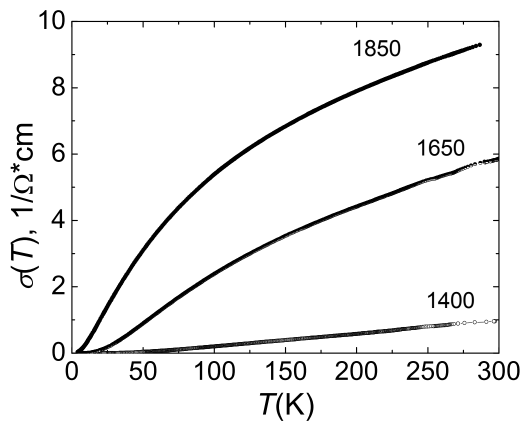

Fig. 2. Temperature dependence of conductivity of OLC. Bottom curve – OLC received by vacuum annealing of ND at 1400K, middle curve –at 1650 K, top curve – at 1850 K.

According our previous investigations [4-6] for all samples annealed at temperature higher 1170 K the electrical conductivity shows the temperature dependence typical for the systems with variable range hopping conductivity (VRHC) [7]. This dependence takes place in strong disordered materials with a length of local disorder approximately a few interatomic distances [8-10]. For these systems, the temperature dependence of conductivity σ(T) may be described by the equation:

$$\sigma(T) = A^{*}\exp[-(T_0/T)^{1/n}] \qquad (1)$$

where A - is constant, the value of n is determined by dimensionality d of the motion of current carriers as n=1/(1+d) [7], $T_0=C_T\alpha/k_BN(E_F)$, C_T~1 for one-dimensional VRHC [11, 12], α is the inverse length on which the amplitude of wave function fall down (α~8-10 Å), $N(E_F)$ - density of localized state at the Fermi level, k_B is the Boltzmann constant. On the Fig. 3 we present data for OLC in the $\ln[\sigma(T)/\sigma_{293}]$ – $T^{-1/2}$ axis, where σ_{293} - conductivity at room temperature.

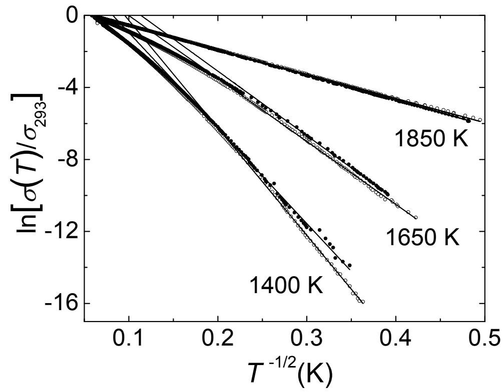

Fig. 3. Temperature dependence logarithm of relative conductivity $\ln[\sigma(T)/\sigma_{293}]$ from $T^{1/2}$ of OLC. Bottom curve – OLC received by vacuum annealing of ND at 1400K, middle curve –at 1650 K, top curve – at 1850 K.

In order to find out the influence of gamma irradiation on behavior of conductivity of OLC we investigated the temperature dependence of conductivity after irradiation.

$\sigma(T)/\sigma_{293}$ after γ - irradiation

In the literature the data on the influence of gamma irradiation on conducted properties of nanostructure conductors are practically absent. According to our preliminary investigation the graphitization nanodiamond goes by formation the quasi-one-dimensional graphite-like strips which lead to the quasi one-dimensionality of current carriers (Eq. (1)). The concentration of current carriers grows with increase of the heat treatment temperature and conductivity is increased. Gamma irradiation leads to both the increase of defects in these strips and to the increase of strips quantity (as well as at the increase of the heat treatment temperature). In this work we used data of temperature dependences of the relative conductivity for the estimation of density of states at Fermi level $N(E_F)$. After that the investigated samples of OLC were exposed to gamma irradiation on the electron accelerator in the Institute of Nuclear Physics of the Siberian Branch of the Russian Academy of Science. The total dose of the gamma irradiation was 0.2 J/kg (0.2 Sv). After an irradiation the measurements of temperature dependences of conductivity were again carried out and $N(E_F)$ was again estimated. Fig. 3 shows the $\sigma(T)/\sigma_{293}$ for OLC in the $\ln[\sigma(T)/\sigma_{293}]$ – $T^{-1/2}$ axis.

The estimation of $N(E_F)$ from data of $\sigma(T)/\sigma_{293}$

Using the received experimental data (Fig. 3) we have estimated density of states on Fermi level. Fig. 4 shows the result of this estimation: the open points -before an irradiation and the closed points - after the gamma irradiation by the dose 0.2 Sv. The $N(E_F)$ of the OLC sample annealed at 1400 K is increased after the irradiation. But $N(E_F)$ of the OLC samples annealed at 1650 K and 1850 K is decreased after the irradiation.

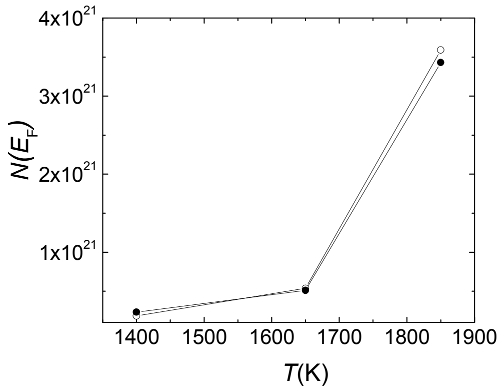

Fig. 4. Dipendence of density of localized state at the Fermi level $N(E_F)$ from the temperature of vacuum annealing of ND before irradiated (open circles) and after irradiation (filled circles).

4 Conclusion

In summary, the gamma irradiation causes different effect on the concentration of a current carriers on Fermi level $N(E_F)$ in OLC samples produced at different temperature from ND.

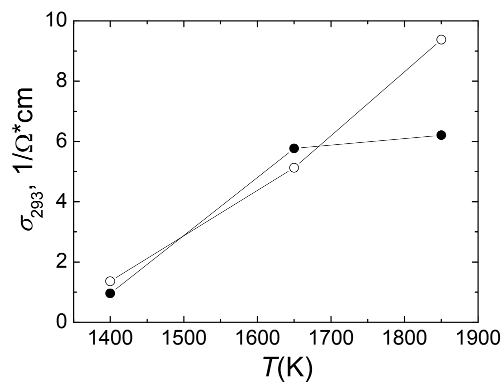

Fig. 5. Dependences of conductivity at room temperature σ_{293} from the temperature of vacuum annealing of ND before irradiated (open circles) and after irradiation (filled circles).

The conductivity of OLC at a room temperature practically linearly grows with growth of heat treatment temperature from 1400 K to 1850 K (the open circles on Fig. 5) because of decrease of diamond core content with increasing annealing temperature. The conductivity of the irradiated samples Dc-1 and Dc-2 closely to the data before irradiation. But the conductivity of the sample Dc-3 after irradiation is smaller by 30% of the conductivity before irradiation. Density of states on Fermi level $N(E_F)$ of OLC after irradiation is smaller than density of states before irradiation for sample Dc-3 (Fig. 4). Such change $\sigma(T)$ and $N(E_F)$ of OLC after gamma irradiation can be related to decrease of concentration of defects in the graphite-like conducting layers in comparison with the initial sample. As is well known [13-16], in graphite-like defective structures the charge carrier concentration is basically determined by concentration of defects in the graphite-like layers. Earlier we have found [4-6], that the maximum of concentration of defects is observed in samples annealed at temperature ~1800 K. Gamma irradiation of these samples can lead to reduction of defects and, consequently, to the decrease of charge carrier concentration. The concentration of current carriers in conductors is increased when the density of state on Fermi's level is increased. The similar reasoning can be used for $N(E_F)$.

Thus, it is founded, that the gamma irradiation with energy of few eV leads to reduction of defects in the graphite-like conducting layers in OLC that directly affects the conductivity of samples.

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