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06

The effect of temperature on the electrophysical properties of elastomers containing metallized carbon nanotubes

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The time dependences of the current on the voltage and temperature of a polymer composite containing carbon nanotubes, obtained by ultra-high frequency electromagnetic radiation, have been studied. The current consumption of the composite is nonlinearly dependent on the applied voltage, taking into account the inrush current, which initiates heating, and the temperature in the range from -60 to $+60\,^\circ$C. The inrush current in the nanocomposite is associated with the charge transfer process in the microstructure formed by carbon nanotube bundles due to the polarization of the polymer matrix. The scientific novelty of this study lies in the analysis of the electrophysical properties of the nanocomposite, considering the positive temperature coefficient of resistance effect. It was revealed that a stable current regime forms in the voltage range of 32–36 V, allowing for stable heat emission up to $-60\,^\circ$C.

Keywords: polymer composites, temperature self-regulation, metallized carbon nanotubes, electrical conductivity.

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Polymer composites (PCs) used as heating elements offer a wide range of electrical and thermal properties, since an extensive set of polymer matrices and various conductive additives are available [1,2]. The functional properties and self-regulation capability of a PC are governed by the dispersed filler content [3–5]. The effect of temperature self-regulation is associated with a positive temperature coefficient of resistance (PTCR) of a composite [6].

Low-density polyethylene (LDPE) and carbon black (CB) are used to form composites with a PTCR [7]. Phase transitions in the polymer matrix [8] induce a PTCR in conductive composites based on high-density polyethylene (HDPE) and CB. Phase transitions are affected by the rheological properties of a polymer matrix [9]. Hybrid fillers are used to improve the electrophysical properties of a binary polymer matrix [10]. Mixtures of LDPE/HDPE with graphite powders [10,11] constitute another class of conductive composites with a PTCR. Metallic particles may be used to improve the electrical conductivity of composites with a PTCR [12]. The surface of CB may be modified with Ag, which enhances adhesion and electrical conductivity [13]. Elastomers are used as a polymer matrix to produce flexible heating elements [14,15].

The use of carbon nanotubes (CNTs) in a PC allows one to improve its electrophysical properties, since higher electrical conductivity and thermal stability levels may be achieved at lower (compared to other carbon materials, such as CB and graphite) CNT concentrations [16,17]. CNT synthesis still remains a complex and costly process. CNTs obtained with the use of microwave (ultrahigh-frequency) electromagnetic radiation are of interest in this regard. This approach to production of CNTs appears to be more accessible and allows for significant improvement of their properties through structural optimization (metallization) during synthesis [15]. Metallization should enhance the thermal and mechanical stability of CNTs.

The aim of the present study is to examine the dependence of electrophysical characteristics of the obtained polymer composite with metallized CNTs on the time of current consumption in different temperature conditions.

CNTs were synthesized by microwave irradiation from a mixture of ferrocene ($C_{10}H_{10}Fe$) and graphite (GSM-1, GOST 17022–81). The microwave emitter power was 700 W, and the synthesis time was 10 s. Prior to synthesis, ferrocene and graphite mixed in a ratio of 5 : 1 (by weight) were subjected to mechanical activation in a vortex layer device (AVS-100) for 20 s. The „Silagerm 8030“ (organosilicon and two-component with phases A and B; tensile strength, 3 MPa; Shore hardness, 27A) elastomer was used as a polymer matrix. Component A and multi-walled CNTs were mixed using a mechanical stirrer. The second component (B), which initiates polymerization, was added to the resulting mixture with subsequent stirring for 2 min at a temperature of $22\,^\circ$C. A heating element in the form of a flat plate with upper and lower aluminum electrodes was formed in vacuum.

The morphology and EDX studies of CNTs and the polymer composite were performed using a JSM 7001F (JEOL, Japan) scanning electron microscope (SEM) with

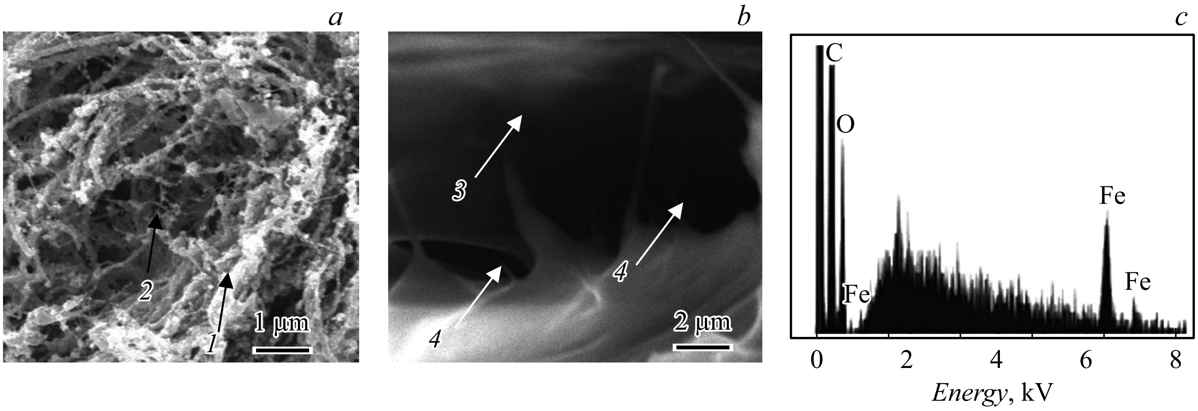

Figure 1. SEM images of CNTs (*a*) and the nanocomposite (*b*) metallized with iron; energy-dispersive spectrum of CNTs (*c*).

an Inca Penta FETx3 detector and a TESCAN LYRA 3 (Czech Republic) SEM, respectively.

Electrical resistance R was determined with an E6-13A (Estonia) teraohmmeter and a UT71E (UNI-T, China) multimeter. Specific bulk conductivity σ of the nanocomposite was determined using the percolation equation [15]:

$$\sigma = \sigma_c + (\sigma_m - \sigma_c)\left(\frac{\varphi - \varphi_c}{F - \varphi_c}\right)^t, \qquad (1)$$

where σ is the specific bulk conductivity [S/cm], σ_m is the specific bulk conductivity at the maximum mass content of CNTs [S/cm], σ_c is the specific bulk conductivity at the percolation threshold [S/cm], t is the critical conductivity index, F — is the CNT packing factor in the elastomer, φ is the volume fraction of CNTs, and φ_c is the volume fraction of CNTs at the percolation threshold.

To establish the required temperature conditions, the PCs were introduced into a special heat-insulated heat/cold chamber 0.01 m^3 in volume with liquid nitrogen cooling and electrical heating elements (tubular electric heaters) with a power of 1 kW.

Dependences of the current strength (I) on the ambient temperature (T) were measured in order to evaluate the parameters of temperature self-regulation of the composite with CNTs. Since the $I(T)$ dependence correlates with the variation of charge in the microstructure formed by individual CNT bundles after polarization of the polymer matrix, one needs to examine the time dependence of the inrush current of the nanocomposite.

Figure 1 presents the results of examination of CNTs and the nanocomposite.

It follows from the results of microstructure analysis that CNTs are intertwined with each other (label *1* in Fig. 1, *a*) and form dense bundles with diameters ranging from 30 to 60 nm. Iron nanoparticles (10−20 nm) (label *2* in Fig. 1, *a*) are seen on the surface of CNTs, which is also confirmed by the energy-dispersive spectrum of CNTs with Fe lines (Fig. 1, *c*). Figure 1, *b* shows that CNTs are coated with polymer (label *3*) and reveals the formation of CNT sections with dense packing in the polymer matrix structure (label *4*).

The parameters characterizing the electrical conductivity of composites, which were determined experimentally using the percolation dependence of the electrical conductivity of the nanocomposite on the concentration of CNTs, are listed in the table.

Figure 2 shows the measured time dependences of the inrush current at different supply voltage levels.

At the initial moment of time, the maximum inrush current varied from 0.3 to 1.1 A within the voltage range from 6 to 16 V (Figs. 2, *a*−*d*). Within the subsequent time interval, current got stabilized throughout the entire voltage range (6–16 V) within an average time of 100−120 s, which is indicative of an active-capacitive characteristic of the nanocomposite resistance.

Figure 3 presents the results of examination of the electrical current consumption as a function of voltage with the temperature varying from −60 to +60 °C.

According to the data from Fig. 3, the current consumption of the nanocomposite increases as the temperature in the heat/cold chamber decreases. This is consistent with the fact that CNTs have a PTCR and the obtained heating elements feature a temperature self-regulation effect. At a voltage of 32−36 V, a steady increase in current, which is sufficient to reach the required heat generation levels within the entire studied range of temperature conditions, is observed. A reduction in current at temperatures from 0 to −30 °C is seen in Fig. 3 at a voltage of approximately 32−36 V. With a voltage of 12−14 V, a reduction in current was observed at a temperature of −50 °C. At positive temperatures of 20–60 °C, the current consumption of the

Parameters characterizing the electrical conductivity of composites

Volume fraction of CNTs at the percolation threshold φ_c	CNT packing factor in the elastomer F	Critical conductivity index t
0.05	0.7	2.3

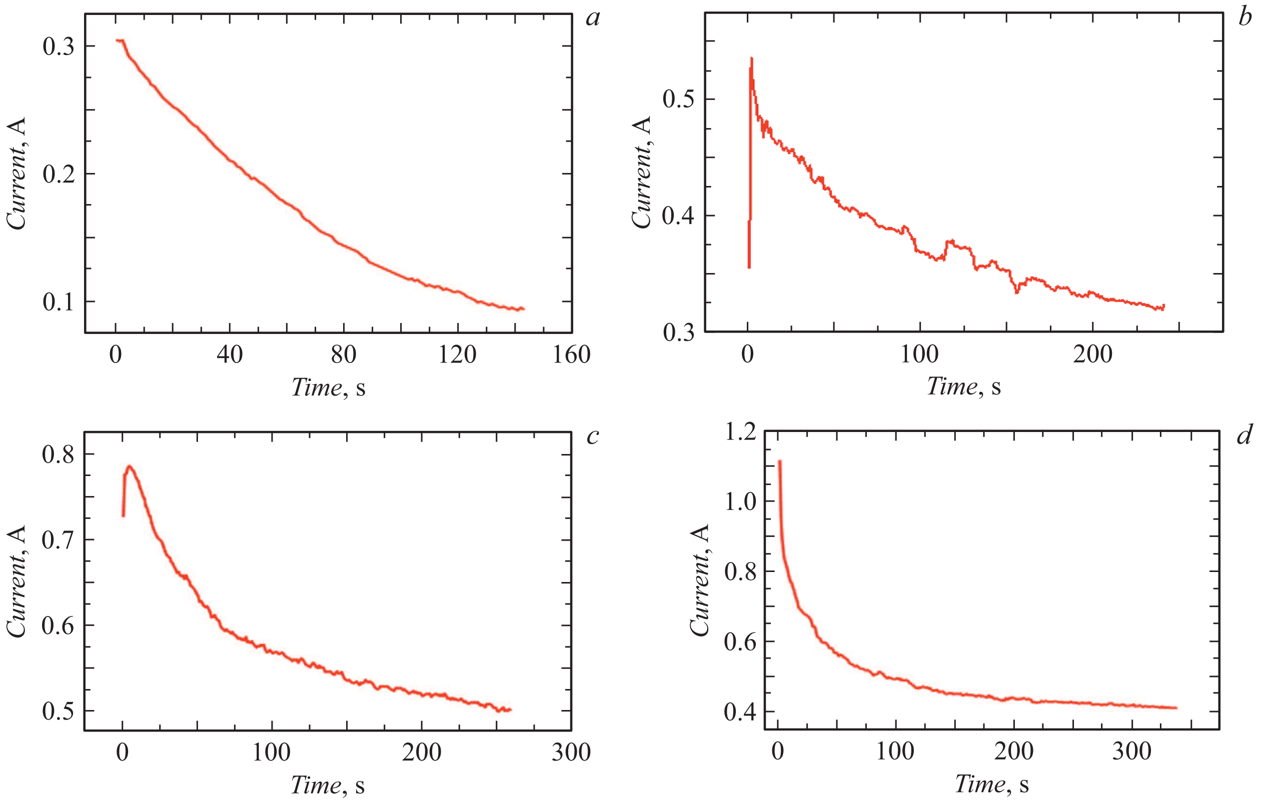

Figure 2. Time dependences of the inrush current of the nanocomposite at different voltage values: 6 (*a*), 10 (*b*), 14 (*c*), and 16 V (*d*).

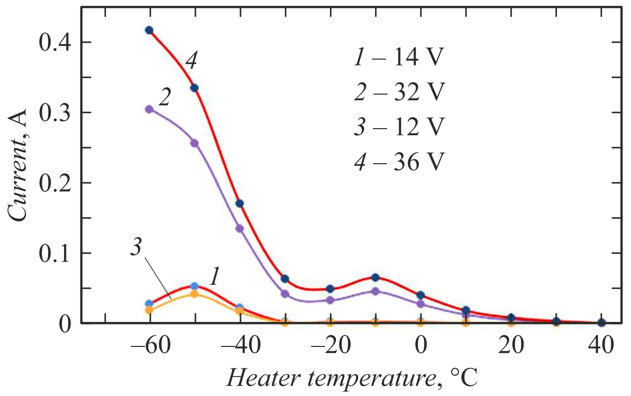

Figure 3. Dependences of the current consumption of the nanocomposite on voltage in various temperature conditions.

nanocomposite is virtually zero, which provides further evidence of temperature self-regulation.

It was established that

(1) the nanocomposite enters a stable current regime, which supports steady heat generation up to a temperature of −60 °C, within the voltage range of 32−36 V, and a stable increase in current up to a temperature of −30 °C is observed at a voltage of 12–14 V;

(2) the polymer composite containing metallized CNTs is characterized by a fast response (inrush current) to the applied voltage (due to the active-capacitive characteristic of the nanocomposite resistance) and is suitable for application within a wide temperature range from −60 to +60 °C.

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Conflict of interest

The authors declare that they have no conflict of interest.

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