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# Fractal structure and temperature-dependent electrical study of carbon nanotubes/epoxy polymer composites

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**ABSTRACT**

The main objective of this work was to study the electric response of the carbon nanotubes/epoxy composites upon varying the concentrations of the nanotubes, in order to obtain a comprehensive understanding of the influence of filler's percentage and temperature on the impedance parameters. Dielectric spectra of the carbon nanotubes–epoxy resin composites were recorded in the frequency range of 1 Hz–10 MHz and over the temperature range of 25°C–105°C. In a first part, the thermal properties of the composites were analyzed by differential scanning calorimetry, which reveals a decrease in the glass transition temperatures with the concentration of the nanotubes. The phenomenon of the positive temperature coefficient in resistivity that is recognizable for the carbon nanotube concentrations above the percolation threshold has been the interest of the second part of the work. The results reveal that gradually, as the filler concentration approaches the percolation threshold of the composite, the positive temperature coefficient in the resistivity effect becomes even more pronounced. Finally, the structure of the filled polymer samples is characterized using a small angle neutron scattering technique.

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Electrical properties; fractal structure; impedance spectroscopy; positive temperature coefficient in resistivity; small angle neutron scattering; thermal properties

## Introduction

The great interest in exploiting the exciting properties of carbon nanotubes (CNTs) by incorporating them into some form of polymer matrix leads us to study the electric properties in order to extend the applications of polymer materials, making the nanocomposites ideal for electromagnetic shielding (EMS), electrostatic discharge protection (EDP) in packaging or manufacturing sensitive electronic devices.[1–3] Metals can very well serve the previously mentioned applications due to their high electrical conductivity; indeed, they have been being used for years. But, their heavy weight, physical rigidity, and corrosion restricts them from being appropriate candidates to be employed. The most effective combinations that could overcome these shortcomings are the CNT–polymer composites.[4] The electrical and electromagnetic shielding performance of polymeric materials filled with multi-walled CNTs still attracts enormous interest; effects of sonochemical modification of CNTs on conductivity and EM shielding properties have been studied, according to the authors' conclusions, the two different CNT surfaces have different effects on the dispersion state, electrical and electromagnetic shielding properties.[5] Hattenhauer et al.[6] have studied the behavior of the CNT based nanocomposites by the application of an electric field during curing, which resulted in a further increase in the conductivity, later, in another publication of the same first author, epoxy resin/carbon sphere nanocomposite were successfully produced which revealed to be excellent alternative materials for the development of organic worm memory devices.[7]

In this paper, 10 samples containing different percentages of carbon nanotubes from 0.0% to 5.0% were studied. The first part of this work describes the analysis of the thermal properties of the composites by differential scanning calorimetry (DSC), to obtain and notice the effect of various filler concentrations on the glass transition.[8] Secondly, in addition to the influence of the variation of nanotube's concentrations at room temperature, which was reported in our previous work,[9] in this paper, the effect of temperature on the electrical properties of the carbon nanotubes/polymer composites, and the positive temperature coefficient of resistivity (PTCR) effect,[7,8] below and above the conduction threshold (in our case the percolation threshold was found at 2.7%) are discussed. The electrical properties of the materials have been investigated in the frequency range of 1 Hz–10 MHz and over the temperature range of 25–105°C using the complex impedance spectroscopy technique. In a third part, the structure of filled polymer samples is characterized using small angle neutron scattering (SANS). During evaluation of SANS data, a simple model that describes the whole Q range of the measurements has been used, and is expected to give results comparable to the separate models used in our previous work.[9] Both results indicate a fractal behavior of the CNT

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filled polymer samples[9,10] with no observable structural change upon varying the temperature in the studied range.

## Experimental

### Materials

The samples investigated in this study were CNTs (Cheap-Tubes, Cambridgeport Vermont, USA) dispersed in an insulating epoxy matrix diglycidylic ether of bisphenol A (DGEBA). The diameter of the primary CNTs was about 50 nm, their length was in the range of 10–20 μm and their purity was higher than 95 wt%. DGEBA had a density of 1.19 g/cm$^3$, a conductivity of the order of $\sigma_{DC} = 1.4 \times 10^{-16}$ S/cm, and a glass transition temperature of about $T_g \approx 60$°C. The CNTs were added to the polymer matrix in different concentrations, stirred thoroughly at room temperature, then 1% of hardener was added to make each mixture cohesive. Each CNT/DGEBA composite was mixed at room temperature for 5 min, promoting the gelation and after it was poured into the mold. After 2 h, the samples were unmolded. Complete polymerization took 24 h. In this work, 10 different samples were prepared with concentrations $\phi$ between 0.0% and 5.0%.

### Experimental procedures

#### Differential scanning calorimetry

Differential Scanning Calorimetry (DSC) was carried out using a DSCQ100 V9.9 Build 303 (TA Instrument) programmed between 20°C and 110°C, at a heating rate of 5°C/min. The results were used to determine the glass transition temperature for the composite for four selected nanotube concentrations besides the neat, undoped polymer.

#### Impedance spectroscopy

For the electrical measurements, the samples were prepared as discs of thickness about 1 mm, with aluminum electrodes of 10 mm diameter on the opposite sides of the sample. The electrical contacts were formed by silver paint. The temperature dependent AC impedance spectra were recorded using a Novocontrol Alpha-A Analyzer combined with the ZG4 impedance interface, in a 4-wire arrangement in the frequency range of 1 Hz–10 MHz. Measurements and data recording were controlled with the software WinDeta.[11] The bulk AC conductivity was determined by nonlinear mean-square-deviation curve fitting of the impedance spectrum using the software WinFit.[11]

#### Small-angle neutron scattering

Small angle neutron scattering (SANS) is a technique for characterizing the structure of the matter at the scale of nanometers. With its aid the shape, size, size-distribution, and surface of the so-called scattering objects can be studied. The obtained data are evaluated by model fitting, the parameters of an adequate model are characteristic to the structure of the sample. The scattering intensity is obtained as a function of the momentum transfer $Q$:

$$Q = \frac{4\pi}{\lambda} \sin 2\theta \qquad (1)$$

where $2\theta$ is the scattering angle and $\lambda$ is the used wavelength. The same samples, used for the electrical measurements, were measured by SANS.

The measurements were carried out at the Yellow Submarine (YS) and FSANS instruments of the Budapest Neutron Centre (the detailed description of the experiments were given elsewhere[9]). YS is a pin-hole type SANS machine, while FSANS is a time-of-flight type instrument. The covered $Q$ range was 0.003–0.020 Å$^{-1}$ for FSANS and 0.010–0.300 Å$^{-1}$ for YS. The raw data treatment included correction for beam attenuation (according to the measured sample thickness), sample transmission, background noise, and detector pixel sensitivity.[9] Four samples were measured by FSANS and SANS, containing 0.0%, 0.2%, 1.0%, and 5.0% CNTs; the data measured by the two-dimensional detector was radially averaged and plotted versus the scattering vector. The scattered intensity $I$ for the whole $Q$ range – measured on the two different instruments – was interpreted by a single model:

$$I(Q) = A \exp\left(\frac{-Q^2 R_g^2}{3}\right) + B\left(\frac{Q}{erf\left(\frac{QR_g}{\sqrt{6}}\right)^3}\right)^{-p} \qquad (2)$$

where $A$ and $B$ are proportional to the contrast, to the volume of the irradiated sample, and to the particle number density. $R_g$ is the radius of gyration, $p$ is the exponent related to the fractal behavior of the scattering surfaces.

## Results and discussion

### Thermal results

According to the method of the tangents,[12] a small transition indicating a glass transition in the vicinity of 60°C is shown in Fig. 1, it is noticeable that the glass transition temperature $T_g$ slightly decreases from 63°C (0.0% of CNTs) to 58°C (5.0% of CNTs) (Table 1). The decrease of $T_g$ with increasing CNT concentration can be explained by the acceleration of the local dynamics of the structural relaxation of the polymer molecules. It is due to the presence of non-negligible amount

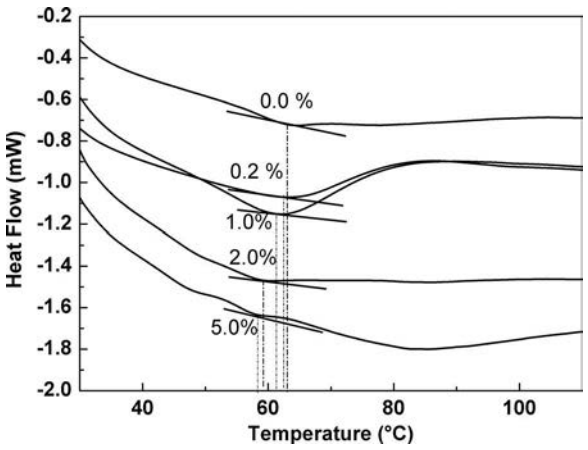


Figure 1. Differential scanning calorimetry thermograms of carbon nanotubes/epoxy polymer composite samples containing different percentages of carbon nanotubes from 0.0% to 5.0%. AC – alternating current; Ω.m – ohm.meter; Hz – hertz; $\phi$ – concentration of carbon nanotubes; $\phi_C$ – conduction threshold concentration of carbon nanotubes.

**Table 1.** Glass-transition temperatures ($T_g$) for different concentrations ($\phi$) of carbon nanotubes, obtained from the differential scanning calorimetry spectra.

| $\phi$ (%) | 0.0 | 0.2 | 1.0 | 2.0 | 5.0 |
|---|---|---|---|---|---|
| $T_g$ (°C) | 63 | 62 | 61 | 59 | 58 |

of nanoparticles, since during the preparation of the nanocomposites, the filler hinders the formation of cross-linking between polymer chains. Similar behavior was observed on various nanocomposites on several studies.[13–15]

## Dielectric and electric results

Former measurements have shown that the CNT/DGEBA composite exhibits a percolation transition at the concentration $\phi_C \approx 2.7\%$ of the CNTs, which has consequences on the electrical properties.[9] Figure 2a and 2b shows the AC conductivity $\sigma_{AC}$ of the composites for two selected concentrations, $\phi_C = 2.5\% < \phi_C$ and $\phi = 3\% > \phi_C$, respectively, in the frequency range from 1 Hz to 10 MHz for temperatures between 25°C and 105°C. It can be seen that $\sigma_{AC}$ increases monotonically with the frequency $f$, for all samples at any temperatures. For $\phi < \phi_C$(Fig. 2a), the particles are not physically in contact, and the conduction process occurs by tunneling. This is a thermally activated process, that is why conductivity increases with the increase of the temperature, showing a negative temperature coefficient in resistivity (NTCR) effect. For $\phi > \phi_C$ (Fig. 2b), the particles are physically in contact, as a percolating path is formed. In this case, for $T < T_g$, the increasing in volume due to swelling is predominant, and then the conductivity decreases as the temperature increases, showing a positive temperature coefficient in resistivity (PTCR) effect. This is not observed for $T > T_g$, where the AC conductivity increases with increasing temperature.

This behavior can even better be demonstrated via plotting the temperature dependence of the DC resistivity $\rho_{DC} = 1/\sigma_{DC}$. The DC conductivity, $\sigma_{DC}$, is obtained from the low frequency plateau of $\sigma_{AC}(f)$, by extrapolating it to $f = 0$. Figure 3a and 3b exhibits $\rho_{DC}(T)$ for CNT concentrations below and above the percolation threshold, respectively. It can be seen that for $\phi < \phi_C$, the temperature coefficient of $\rho_{DC}(T)$ is negative (Fig. 3a), but for $\phi > \phi_C$, the resistivity becomes a non-monotonic function of $T$ with a sign inversion of the temperature coefficient at around $T_g$; thus exhibiting PTCR effect at low temperatures, but having a negative temperature coefficient in resistivity (NTCR) at high $T$, as shown in Fig. 3b. The PTCR effect is observed for temperatures under $T_g$, where the conductive filler particles are not in physical contact. As the temperature increases, the gap between particles also widens, so the electrons should tunnel through the widening insulating gap between them. This explains the increase in resistivity with rise in temperature. The NTCR effect is observed for temperatures

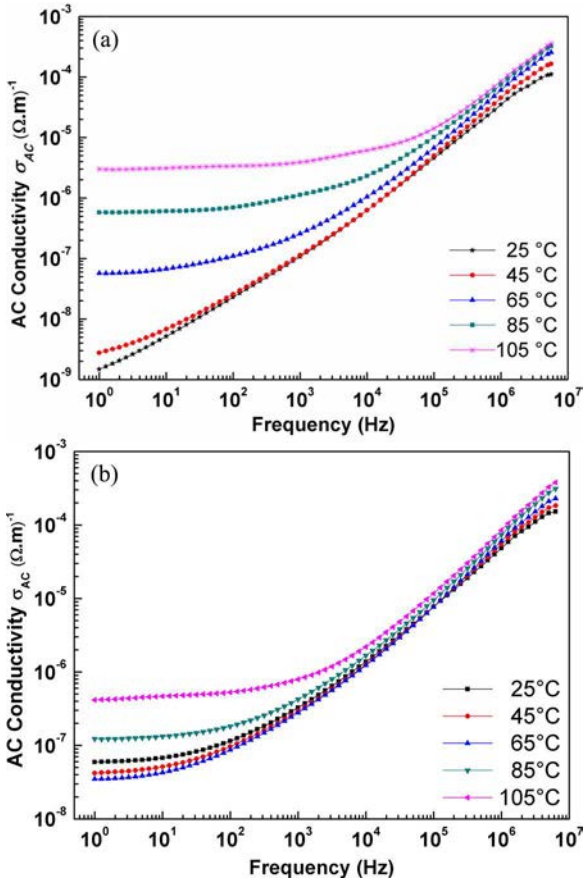


**Figure 2.** Dependence of the AC conductivity ($\sigma_{AC}$) on the frequency at various temperatures for carbon nanotubes/epoxy polymer composite samples: (a) for 2.5% of carbon nanotubes ($\phi < \phi_C$) and (b) for 3.0% ($\phi > \phi_C$) of carbon nanotubes. DC – direct current; Ω.m – ohm.meter;°C – temperature (Celsius); $\phi$ – concentration of carbon nanotubes; $\phi_C$ – conduction threshold concentration of carbon nanotubes.

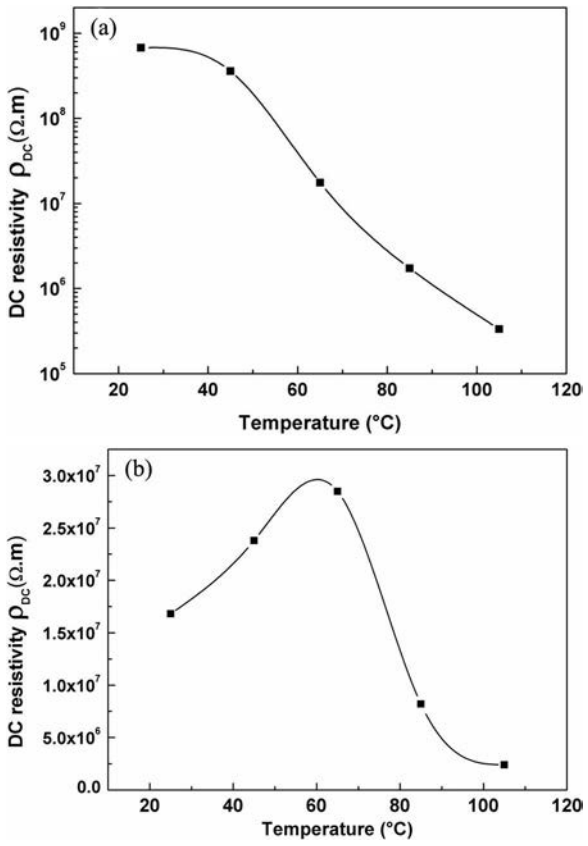


**Figure 3.** Temperature dependence of the DC resistivity ($\rho_{DC}$) for carbon nanotubes/epoxy polymer composite samples: (a) for 2.5% of carbon nanotubes ($\phi < \phi_C$), and (b) for 3.0% of carbon nanotubes ($\phi > \phi_C$). DC – direct current; Ω.m – ohm.meter; $\phi$ – concentration of carbon nanotubes.

higher than $T_g$; in this case continuous chains of conducting nanoparticles are developed, which counteract the reduction in the ease of tunneling; this is a possible explanation to the decrease of resistivity with increasing temperature. It can be concluded that the mechanism responsible for the change in resistivity at this stage is predominantly tunneling.[10]

The DC conductivity $\sigma_{DC}$ is shown in Fig. 4, as the function of the concentration $\phi$ of carbon nanotubes, at various temperatures. For low temperatures (below $T_g \approx 60°C$), the conductivity of the composite increases with increasing amount of CNTs. For $T > T_g$, however, $\sigma_{DC}(\phi)$ becomes non-monotonic. Nevertheless, one can observe that at the highest CNT concentration ($\phi = 5.0\%$), $\sigma_{DC}$ reaches similar maximum values of about $10^{-3}(\Omega.m)^{-1}$, for all temperatures, as it is expected for a percolation phenomenon. By increasing the concentration of CNTs from 0.2% to 5.0%, an increase of six orders of magnitude was observed in the conductivity at the temperature of 25°C, while for the temperature of 105°C, the change of $\sigma_{DC}$ was just only about two orders of magnitude. Under the conditions where the NTCR behavior is found, the DC conductivity increases exponentially with the temperature, indicating that electrical conduction is a thermally activated process. This can be expressed mathematically by the well-known Arrhenius relation:[16]

$$\sigma_{DC} \propto \frac{1}{T}\exp\left(\frac{-E_a}{K_b T}\right) \quad (3)$$

The natural logarithm of ($\sigma_{DC}T$) is plotted as a function of the reciprocal of the temperature for the five compositions 0.2%, 1.0%, 2.5%, 3.0%, and 5.0% in Fig. 5. These Arrhenius plots of the DC conductivities reveal the activation energies $E_{a(dc)}$, as listed in Table 2. It is found that the activation energy is only weakly sensitive to the presence of carbon nanotubes for all the concentrations ($1.17\ eV \leq E_a \leq 1.50\ eV$), except for $\phi = 3.0\%$ and $\phi = 5.0\%$ which is higher than $\phi_c = 2.7\%$, where the activation energy became much lower ($E_a = 0.71$ eV and $E_a = 0.06$ eV for 3.0% and 5.0% respectively). We observe that, for $\phi \leq \phi_c$, the activation energies calculated are almost the same and are insensitive to CNT volume fractions. This

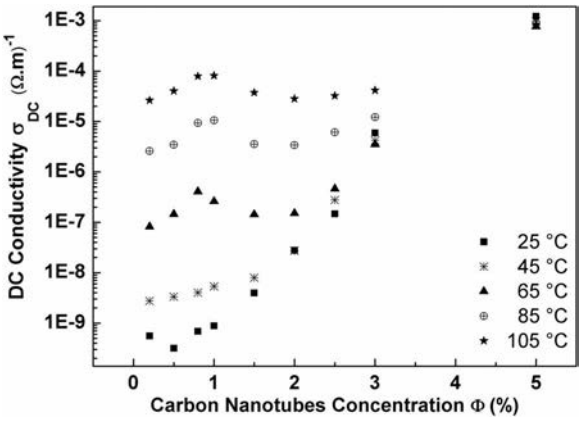


**Figure 4.** DC electrical conductivity ($\sigma_{DC}$) at various temperatures, as a function of the concentration of carbon nanotubes ($\phi$) for carbon nanotubes/epoxy polymer composite samples. $\sigma_{DC}$ – DC conductivity; DC – direct current; T – temperature; K – Kevin; Ω.m – ohm.meter.

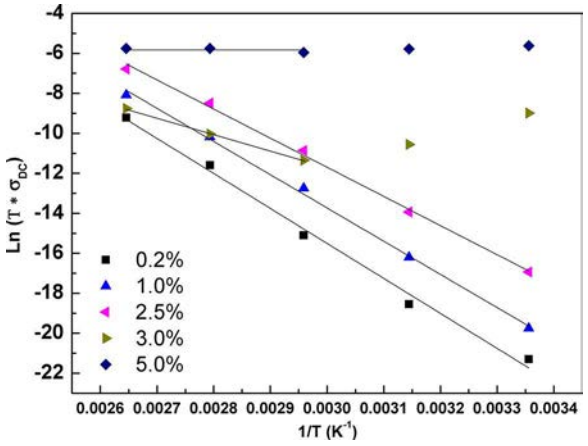


**Figure 5.** Arrhenius plot of the DC conductivity ($\sigma_{DC}$) versus 1/T for carbon nanotubes/epoxy polymer composite samples with different concentrations of carbon nanotubes. Å – Ångström; D – fractal dimension; p – fractal exponent.

result means that when CNT is present inside the epoxy polymer matrix, the conducting particles do not interact considerably with the chain segments of the macromolecules in the epoxy polymer. Similar results have been obtained by Macutkevic et al.[17] on the composite materials based on two types of CNT with average outer diameter ~9 and 12–14 nm in a polymethylmethacrylate polymer matrix. The results obtained on these both series of samples, shown that the activation energy, for different concentrations of CNT between 0.0% and 2.5%, decreased weakly from 1.28 to 1.07 eV, respectively. In another work established by the same authors studied the electrical transport in carbon black-epoxy resin composites[18] and shown that the obtained activation energy is lower in composites than in pure polymer matrix. Such decrease is related to the rearrangement of gaps between the carbon black aggregates.[19]

## Structural results

Figure 6 presents the cumulated data measured for three composites (with 0.2%, 1.0%, and 5.0% of CNTs) on both, YS and FSANS, instruments. The scattering from the inhomogeneities of the neat epoxy resin matrix was subtracted from the scattering of the nanotube containing samples. It was found that the whole $Q$ range, measured by the two SANS instruments, can be described by a single model (Eq. [2]), containing less fitting parameters than the two separate fits for the different $Q$ ranges used before;[9] thus the fit can be considered simpler, and at the same time, more precise. The fitting parameters of Eq. [2] provided an average diameter of 66 nm for the nanotubes, while the exponent $p$ describing their surface was 3.52, 3.82, and 3.89 for the samples with 0.2%, 1.0%, and 5.0% of CNTs, respectively. These correspond to fractal exponents ($D = 6\text{-}p$) of 2.11, 2.18, and 2.48, respectively. The fractal exponents describe a surface fractal

**Table 2.** The values of the activation energy ($E_{a(dc)}$), for different concentrations ($\phi$) of the carbon nanotubes.

| $\phi$ (%) | 0.2 | 0.5 | 1.0 | 1.5 | 2.5 | 3.0 | 5.0 |
|---|---|---|---|---|---|---|---|
| $E_{a(dc)}$ (eV) | 1.47 | 1.5 | 1.42 | 1.20 | 1.17 | 0.71 | 0.06 |

mW – heat flow (milliwatt); °C – temperature (Celsius).

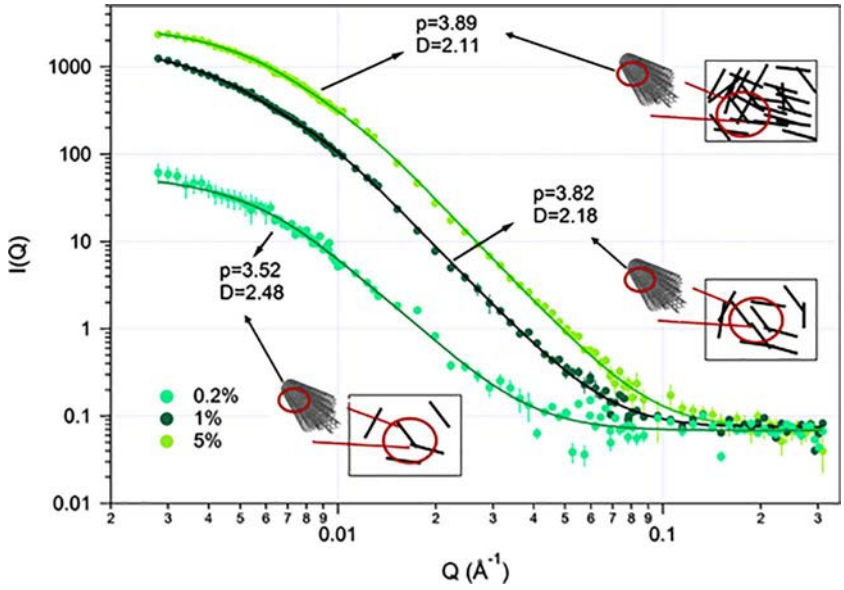


**Figure 6.** Small angle neutron scattering curves of the measured and fitted data for carbon nanotubes/epoxy polymer composite samples. Dots represent the measured values, and lines represent the fitted curves.

behavior[9] that can be attributed to the imperfections of the surface, the presence of the carbon nanoclusters, pores, or additives on the surface.

## Conclusion

The studied samples of carbon nanotubes based epoxy nanocomposites exhibit a positive temperature coefficient of resistivity in the vicinity of the glass transition temperature of the polymer matrix composites; their conductivity sharply decreased with the increase of temperature. It was concluded that the mechanism responsible for the change in resistivity is tunneling. Analysis of the DC conductivity allowed us also to observe the percolation phenomenon and to study the activation energy. Three samples, containing 0.2%, 1.0%, and 5.0% nanotubes, were analyzed with the aid of a neutron technique. It gives information about the bulk of the sample volume placed into the neutron beam without disturbing the structure. The obtained results present an average over the whole volume of the sample; that is an important advantage of this technique in comparison to microscopy. The diameter of the nanotubes was found as 66 nm. From the model fitting of the data, a fractal dimension on the scale of nanometers was obtained, characterizing the surface between the nanotubes and epoxy resin. The scattering from the epoxy–resin matrix was subtracted from the scattering of the nanotubes; therefore, it was possible to improve the data treatment procedure compared to previous results.

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