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Electrical Properties of Single- and Multi-Walled Carbon Nanotubes Composites at Low Temperatures

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Abstract – *Electrical properties of single- and multi-walled carbon nanotubes composites (PEDOT:PSS/CNTs) were investigated in the range of 50 to 200 K. It is established that composite films with multi-walled nanotubes loading show lower resistances as compared to their single-walled counterpart.*

Keywords – nanotube, electrical properties, low temperature, composite, polymer.

I. Introduction

Carbon nanotubes, or CNT, are cylindrical objects with a high aspect ratio that results from wrapping a graphene layer onto itself. In case there is a single graphene layer, the structures obtained are referenced to as single-walled carbon nanotubes (SWNTs). If a few layers of graphene are wrapped, the resulting formations are called multi-walled carbon nanotubes (MWNTs). Since their discovery back in 1991 [1], enormous research efforts have been put into the investigations of CNTs properties which are in many ways unique. Composites that have a good homogeneity and dispersion of nanofiller are of considerable interest for a vast variety of applications – from simple electromagnetic interference shielding to sophisticated optical devices [2-4]. Speaking of polymer-based nanocomposites, increased attention is recently paid to poly(3,4-ethylenedioxythiophene) (PEDOT) host matrices doped by poly(styrenesulfonate) (PSS) [5] and reinforced with carbon nanotubes.

A number of reports focus on the interaction of external fields with PEDOT:PSS/CNT structures [6,7] and the behaviour of electrical properties of such nanocomposites as a function of frequency was also extensively studied recently [8,9]. Not so much, however, is known about the variation of conductivity/dielectric properties of these composites with temperature, especially in the low-temperature region. On the other hand, the knowledge about the influence of nanofiller on such parameters of the composite as loss tangent or dielectric permittivity is crucial when material engineering for functional applications is considered.

Here we present an attempt of a systematic study of electrical properties of PEDOT:PSS polymer composites reinforced with single-walled and multi-walled carbon nanotubes, specifically focusing on the temperature range of 50-200 K.

II. Experimental

Hybrid composite films were prepared starting from 1% water suspension of poly-3,4-ethyldioxiophen from Sigma Aldrich (USA) stabilized with surface active anion substance (polystyrene sulfonic acid). Two types of nanofiller were used: purified (90 wt%) single-walled carbon nanotubes (SWCNTs) with average diameter of 1 nm and lengths within the range of 5 to 30 μm and purified multiwalled carbon nanotubes (95 wt%) with average outside diameter of 65 nm, average inside diameter of 10 nm and lengths within 10-20 μm (MWCNTs). In order to unbundle nanotubes agglomerates efficiently, 30 minutes ultrasonication at 42 kHz frequency was applied. Then, deionized water cleaning process took place, upon completion of which water suspension containing 0.5 mg of nanotubes per 1ml of water was obtained. This suspension was mixed with PEDOT:PSS polymer solution and ultrasonically processed again during 4 hours. After 48 hours of proper drying at room temperature, composite films of PEDOT:PSS/nanotubes were formed on the glass. By varying the ratio between PEDOT:PSS solution and suspension of nanotubes films with different concentration of nanotubes (12 wt% and 16 wt%) were fabricated.

Electrical contacts were deposited on the film surface with conductive paint at the opposite side of the sample (coplanar, or lateral geometry), the distance between the contacts being set to 3 mm.

III. Results and Discussion

Prepared samples of PEDOT:PSS/CNTs composite films were subjected to low-temperature impedance measurements. Fig. 1 shows temperature dependencies of the measured sheet resistance of the SWCNT and MWCNT reinforced composite films measured at 100 kHz frequency in the range of 50 to 200 K. Reasons for selecting such temperature range limits were i) relatively small variation of resistance from 200 K up room temperature (total resistance at higher temperatures is determined by random network of nanotubes with tunneling barriers between individual tubes [10]) and ii) rapid increase of resistance for SCWNT composites below 50 K, so that the values of R were beyond the range of measurements for RLC meter.

Generally, lateral resistance of PEDOT:PSS/CNTs composite films increase non-linearly upon cooling. The dependencies in Fig. 1 are split in two sub-ranges, since, as shown below, there are possibly different mechanisms involved below and above 90 K. As far as different loadings of nanofiller are considered, sheet resistances decrease with nanotube concentration.

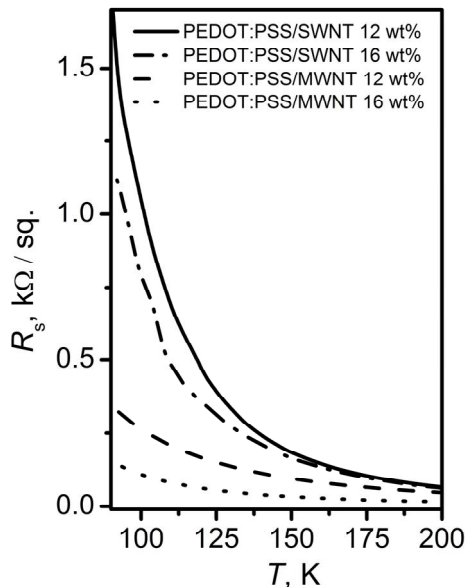
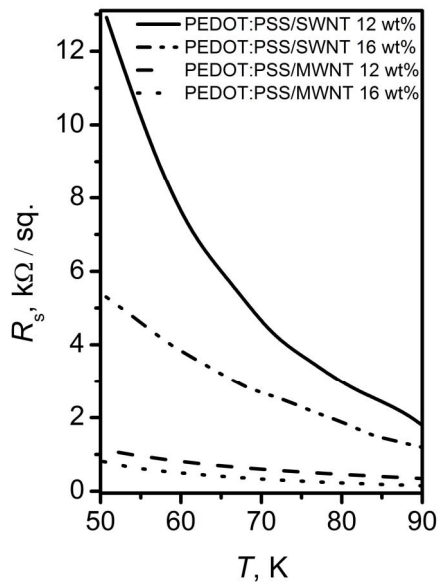


Fig. 1. Sheet resistances of CNT PEDOT:PSS composite films vs. temperature (measured in cooling regime)

Assuming that low-temperature conduction mechanisms follow activation-type relation, sheet resistance can be expressed as

$$R_s \approx T \cdot \exp\left(\frac{eE_a}{kT}\right), \quad (1)$$

where e is an elementary charge and k denotes Boltzmann constant. Plotting $\ln(R_s/T)$ as a function of reciprocal temperature and applying least squares fitting procedure one can estimate activation energy for conductivity.

Conclusion

Resistance of PEDOT:PSS composite films with the addition of single- and multi-walled NTs was studied by conductivity measurements in the lateral direction with respect to the substrate surface. Composites with multi-

walled NT loading show lower resistances at same T as compared to their single-walled counterpart.

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