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Observation of power-law behavior in temperature dependent conductivity of multiwall carbon nanotube/polystyrene composites

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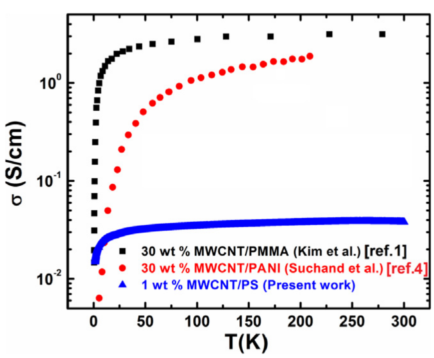
HIGHLIGHTS

- Temperature dependence of conductivity (300–1.4 K) of MWCNT/PS composites is studied.
- Conductivity of MWCNT/PS composites exhibit weak temperature dependence.
- Conductivity ratio (σ_r) is smaller than 3 for all samples.
- Power-law explains the conduction mechanism in the low temperature regime.
- Previous similar measurements resulted a pronounced temperature dependence with σ_r~500.

GRAPHICAL ABSTRACT



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ABSTRACT

For last two decades, a pronounced temperature dependence of electrical conductivity (σ) in the multiwall carbon nanotube (MWCNT) - based polymer composite (even above the percolation threshold) has been widely reported; the conduction mechanism has been understood by employing conventional models, namely variable range hopping and fluctuation induced tunneling. Herein, we report on the observation of a weak temperature dependence of σ in the MWCNT/polystyrene composites above percolation threshold (0.7–7 wt %) at temperatures down to 1.4 K, with a conductivity ratio smaller than 3. The low temperature conductivity data follow power-law exhibiting two slope behavior, with exponent values β_1 ~0.10–0.14 (at T > 5 K) and β_2 ~0.23–0.36 (at T < 5 K). The observation of weak temperature dependence of σ is attributed to high aspect ratio (more than 4000), achievement of high degree of dispersion, and excellent electrical properties of MWCNT as well as optimized composite processing. Further, all the samples exhibit negative magnetoresistance which can be explained within the framework of weak localization model.

1. Introduction

Low temperature electrical transport properties of both single- and multiwall carbon nanotubes (SWCNT & MWCNT) - based polymer composites have been widely investigated, mostly

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resulting in the observation of exponential suppression of electrical conductivity (σ) with temperature in the low temperature regime (i.e. 300–1.4 K) [1–7]. Most often, $\sigma(T)$ of low weight (wt) fraction polymer composite samples ($p \leq p_c$, p is filler loading and p_c is percolation threshold) exhibits a pronounced temperature dependence, and σ decreases exponentially as a function of temperature from room temperature to below the liquid helium temperature; the exponentially decreasing conductivity with temperature indicates the dominance of hopping transport [1–7]. In such samples, conventional models like variable range hopping (VRH) and fluctuation induced tunneling (FIT) or a combination of these models have been used to explain the low temperature charge transport [1–8]. However in the high weight fraction polymer composite samples ($p > p_c$), a weak temperature dependence of conductivity should be expected, which has not been observed so far. Further, charge transport in such samples cannot be understood by employing the exponential models but a power-law model should be used in corroboration with magnetotransport (MR) data; the MR data is generally explained in the framework of weak localization and/or electron-electron interaction models (for disordered systems like carbon nanotubes).

In this work, the charge transport properties of MWCNT/polystyrene (PS) composite films of 0.7–7 wt % are investigated in the temperature range from 300 K (RT) down to 1.4 K, and fields up to 11 T. The temperature dependence of conductivity is observed to be rather weak for all these samples, and seems to follow power-law behavior. Notably, a weak temperature dependence of conductivity is yet to be reported in such composite systems.

2. Experimental

The chemical vapor deposition (CVD)-grown MWCNT, with high aspect ratio (>4000) and average diameter of ~50 nm, were dispersed in polystyrene (PS) by ultrasonication for the preparation of free standing MWCNT/PS composite films. The estimation of aspect ratio and diameter was based on statistical analysis of micrographs obtained from scanning and transmission electron microscopes (SEM & TEM). The insets of Fig. 1 showed the SEM (right inset) and TEM (left inset) micrographs of MWCNT sample that revealed the morphology and physical dimensions of as-grown MWCNT. The films were prepared with different wt % of MWCNT having an average thickness of ~50 μm. The details of the fabrication and characterization of these composite films are reported elsewhere [9]. A standard linear four probe technique was employed to carry out the transport measurements of the composite films (sample dimensions ~3 mm × 10 mm). A constant current in the range of 0.1–10 μA was applied to the outer leads of the samples using Keithley 220 programmable current source, and the voltage in the inner two probes was measured through Keithley 2000 multimeter. For carrying out the temperature dependent conductivity measurement, four collinear electrical contacts (with contact separation ~1.5 mm) are made using fine enameled copper wire of 38 AWG (0.1 mm diameter) on the sample with conductive silver paste. For thermal contact with copper sample holder of the cryostat probe, vacuum grease with high thermal conductivity was used. The sample was immersed directly in liquid helium bath in a Janis variable-temperature system, equipped with a superconducting magnet. The resistance of the sample was recorded as a function of temperature in range of 300 to 1.4 K. Lakeshore 330 auto tuning temperature controller was used for temperature measurement and control.

3. Results and discussion

Fig. 1 presents the variation of the room temperature conductivity (σ_{300K}) of composite films as a function of MWCNT concentration, displaying the improvement of σ_{300K} by a factor of ~40 as MWCNT loading increases from 0.7 to 7 wt %. This conductivity enhancement is obviously due to the improvement in the connecting pathways of MWCNT networks within the polymer which

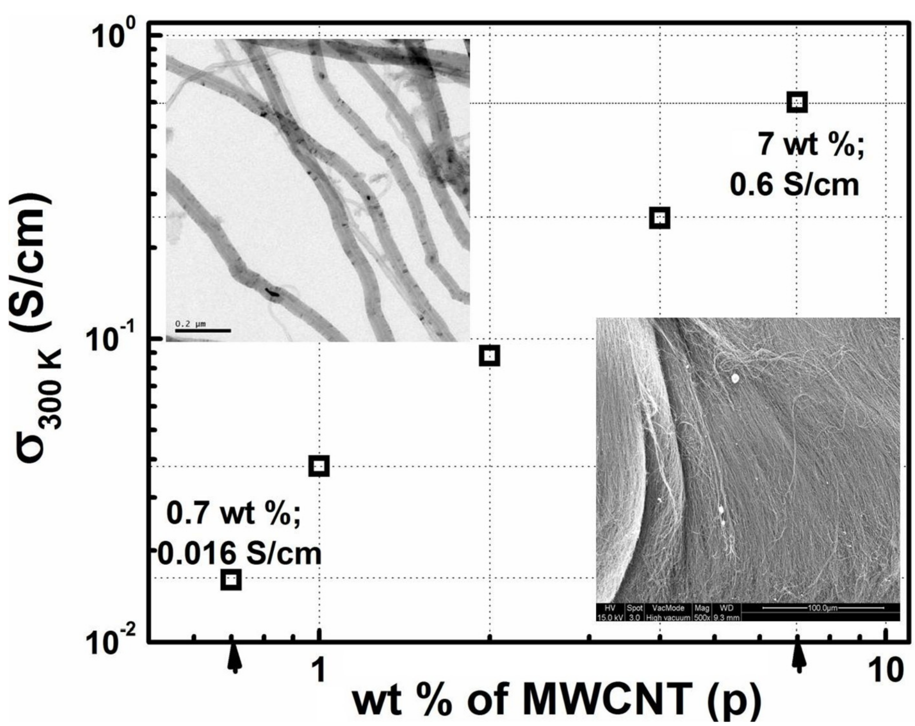

Fig. 1. Room temperature conductivity of MWCNT/PS composites as a function of MWCNT loading. Left Inset. TEM micrograph of MWCNT (Scale bar is 0.2 μm). Right Inset. SEM micrograph of MWCNT sample (Scale bar is 100 μm).