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Effect of temperature on the electrical property of epoxy composites with carbon nanotube

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ABSTRACT

Eliminating the influence of environment temperature is critical for high-accuracy carbon nanotube polymer nanocomposites sensors. In this work, the temperature effects on the nanocomposite are studied by both experiment investigation and simulation calculation. Nearly constant resistance values were found at a CNT loading around 3 wt%. By considering the temperature effect and CNT structural distortion in the developed percolation network model, simulation results agree well with experimental data. On this basis, results show that the thermally assisted tunneling on CNT junctions and thermal expansion of polymer matrix are the two core mechanisms, relaxed CNT junctions in CNT/polymer nanocomposite depressed the temperature effect, and the influence of environment temperature could be significantly reduced by adjusting CNT loadings and choosing a matching polymer matrix. All of these findings will benefit for the design of high-accuracy sensors.

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Carbon nanotubes (CNTs) enhanced polymer composites are expected to become more and more integral for multifunctional nanoelectromechanical sensors, due to their lightweight, high sensitivity, mechanically durable and easily conformable for desired shapes and applications [1–8]. For instance, the CNT/polymer composite sensors can be directly embedded in structural composites to provide real-time internal strain sensing for structural health monitoring [9]. They also can be used as highly functional e-skin, which could sense multiple stimuli caused by temperature and strain simultaneously [10]. However, in practice, the performances of these composite are sensitive to environmental thermal condition. Especially in the field of high precision sensor, the influence of environment temperature cannot be ignored [11–13]. Considering the temperature difference between day and night and heat up by direct sunlight, the outdoor working temperature range for these devices could be from 233 K to 383 K. This temperature effect on electrical properties of CNT/polymer composite is not well understood yet.

The electrical conductivity of CNT/polymer composites depends heavily on the morphology of CNT networks and the properties of CNTs and polymers that form the resistance networks. The CNT intrinsic conductance and the tunneling or contact conductance between CNTs are the two basic types of conductance in CNT/polymer composite [14–16]. Experimental observation shows the interactions of tube-tube or tube-matrix could lead to structural distortions of CNTs [17–19]. It could substantially affect local electrical structure and act as a strong scatter to reduce the intrinsic conductance. And it could also result in a large overlap of wave functions to improve the electron tunneling and increase the contact conductance [20]. Thus, neglect of structural distortion of CNTs will result in gross overestimation of electrical conductivity in CNT/polymer composite. Most of the CNT distortions in polymer matrix cannot be observed directly, since the CNT distortions are very sensitive to the stress state and they also have good elastic recovery. It is difficult to keep the original topography during the processes of sample preparation and topography observation (SEM, TEM or AFM). Here, we introduced a percolating network model to consider the CNT distortion on the resulting electrical performance.

In this study, the temperature effects on electrical conductivity of CNT/polymer composites are systematically studied by

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experiment and simulation. The thermally assisted tunneling on crossed CNT junctions and structural distortion of CNTs are modeled based on the percolation theory, which has been widely used in simulations of CNT network systems [21–25]. The thermally assisted tunneling transport through the crossed CNT junctions is calculated by the Landauer–Büttiker formula with Wentzel–Kramers–Brillouin approximation [16]. The structural distortion of CNTs is determined self-consistently by minimizing the pseudo-potential energy at CNT crossed junctions based on the Lennard–Jones potential and the simulation of coarse grained molecular dynamics [1]. Both experiment measurements and Monte Carlo simulations are carried out to obtain the electrical conductivity of CNT/polymer composites within the mentioned temperature range. Based on the experiment and simulation results, the following questions are studied and discussed in order to improve the accuracy of sensors. Such as, in what case the CNT/polymer composite are more sensitive to temperature? What are the core mechanisms in this system? And how to reduce the influence of environment temperature or how to separate the effect of the temperature through the system?

1. Experiment and simulation

1.1. Experimental investigation

The CNT/polymer composites were fabricated by mixing CNTs into epoxy resin. The multi-wall carbon nanotubes were NC7000 of NANOCYL with an average diameter and length of 9.5 nm and 1.5 μm respectively. The epoxy resin and curing agent were Bisphenol-A PT-2712 and PT-2712 Part B3 from Aeropoxy. These two parts were mixed at the room temperature with a resin/hardener mixing weight ratio of 100/22. This epoxy resin was chosen as following. Firstly, the glass transition temperature of polymer matrix should be higher than 383 K, since the studied environmental thermal condition ranging from 233 K to 383 K. Then the influence of glass transition is ruled out. Secondly, the viscosity of polymer matrix should be low enough to improve the dispersibility of CNTs and avoid CNT aggregation. CNT aggregations are assumed not exist in our model. Thirdly, the thermal expansion coefficient of polymer matrix should be smaller than 70, then the nearly constant resistance can be found in the whole range from 233 K to 383 K according to our calculation results. As shown in Table 1, few polymer matrixes meet the requirements and the epoxy resin in our work is the best candidate.

The CNTs were treated and then mixed into the part-A of epoxy resin by mechanical stirring for 30 min and sonicating bath (Sonicator SC-40) for 6 h at the room temperature. The CNT/resin mixture

was then mixed with the amine hardener by mechanical stirring for 30 min and sonicating bath for 1 h with iced water to prevent the premature solidification of epoxy. After the mixing process, the CNT/polymer mixture was degassed in a vacuum oven (Fisher Scientific Model 281A) at the room temperature for 30 min before it was injected into moulds. Four electrical probes were moulded into samples with equal spacing of 1.5 mm between probes in order to measure the volumetric electrical conductivity by the four-point probe method. The CNT/polymer nanocomposites were cured at the room temperature for 24 h and followed by several heat-cool-heat cycles ranging from room temperature to 200 °C to remove possible thermal history. The electrical conductivity of the CNT/polymer nanocomposite samples was measured by the four-point probe method as per ASTM F1529-97.

The constant DC current was supplied by a Keithley 6221 power supply and the voltage drop over two inner probes was measured by a Keithley 6517B high resistance meter. The electrical conductivity of the CNT/polymer nanocomposites was estimated by the Ohm's Law as the probes are evenly spaced,

$$\sigma = \frac{1}{2\pi d} \frac{I}{V} \quad (1)$$

where V is the voltage drop measured between the two inner probes, I is the current from the power supply, d is the distance between probes.

1.2. Simulation procedure

It is well established that the electrical conduction path in CNT/polymer composites is formed by percolating networks of inter-contacting CNTs [3,16,25]. The CNT percolation network theory assuming the resistivity of the percolating CNT networks can be represented by an effective resistor network. Consider a random distribution of CNTs in a representative volume cuboid of CNT/polymer composites. The distance between each CNT pair is evaluated to check if the crossed-tube junction is formed. By using the formula for calculating the shortest distance between two straight lines in space, the shortest distance between each pair of the line segments (CNT segments) can be calculated. As the distances within the tunneling range (about 1.4 nm), CNT tunneling junctions are formed. If the CNTs are too close to keep the Van der Waals distance (0.34 nm), the CNT distortions are considered to be exist on the junction. The geometric configuration at the junction is determined by minimizing a constrained total energy. In that process, the position of the atoms near the junction is fully relaxed while the center-to-center tube distance is fixed. The total pseudo potential energy (E) of the junction can be expressed by

Table 1
Properties of different polymer matrixes [4–6,26,27].

Materials	T_g (K)	TEC (10^{-6} m/m K^{-1})	Viscosity (Pa s)
Polystyrene (PS)	353–378	70–80	1000–4000 (513 K)
Poly(vinyl alcohol) (PVA)	348–358	30	3–70 (298 K)
Poly(vinyl chloride) (PVC)	333–363	50.4	6200 (423 K)/310 (463 K)
Polyamide (PA)	320–333	110	1000–10000 (483 K)
Polysulfone (PSO)	458	55.8	300–3000 (644 K)
Poly(carbonate) (PC)	408–422	60–70.2	250 (553 K)
Polytetrafluoroethylene (PTFE)	388	112–125	1000 (653 K)
Acrylonitrile butadiene styrene (ABS)	378	70–130	255 (513 K)
Polypropylene (PP)	253	58–100	0.1–2 (298 K)
Polyetheretherketone (PEEK)	416	55	250 (633)/100 (673 K)
Low density polyethylene (LDPE)	153	100–200	350 (483 K)
Polybutylene terephthalate (PBT)	293	130–160	120 (538 K)
High Density Polyethylene (HDPE)	385–390	110–130	32–200 (463 K)
Polymethylmethacrylate (PMMA)	378	70	100–1000 (493 K)
Epoxy resins (This work)	373–393	45–65	0.32 (298 K)

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