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https://doi.org/10.1016/j.actamat.2008.02.030

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Title: Tunneling Effect in a Polymer/Carbon Nanotube Nanocomposite Strain Sensor

Article Type: Full Length Article

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Tunneling Effect in a Polymer/Carbon Nanotube Nanocomposite Strain Sensor

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Abstract:

A strain sensor has been fabricated from a polymer nanocomposite with multi-walled carbon nanotube (MWNT) fillers. The piezoresistivity of this nanocomposite strain sensor has been investigated based on an improved three-dimensional (3D) statistical resistor network model incorporating the tunneling effect between the neighbouring carbon nanotubes (CNTs), and a fiber reorientation model. The numerical results agree very well with the experimental measurements. As compared to the traditional strain gauges, much higher sensitivity can be obtained in the nanocomposite sensors when the volume fraction of CNT is close to the percolation threshold. Corresponding to a small CNT volume fraction, weak nonlinear piezoresistivity is observed in the experimental measurement and numerical simulation. The tunneling effect is considered as the major working mechanism of the sensor under small strains.
1. Introduction

Carbon nanotubes (CNTs) based nanocomposites are increasingly being reviewed as a realistic alternative to conventional smart materials, offering higher sensitivity and superior electrical properties. It has been confirmed that the conductance of a CNT could be dramatically changed by introduction of strain using atomic force microscopy (AFM), as a consequence of the structural change under the effect of mechanical strain, such as the change of chirality in a single-walled carbon nanotube (SWNT) [1]. Due to this piezoresistance property of CNTs, it was predicted that integrating CNTs into polymers would open up a whole range of smart structure applications [2-3]. In particular, great interest has recently been aroused in building strain sensors with CNTs [4-13]. Generally, CNTs are Raman active, and are able to be blended with a polymer to make a strain sensor provided a relationship between mechanical strain and Raman band shift can be calibrated [4]. Obviously, implementation of complex equipment in this technique remains a technical challenge, especially for a field application. Alternatively, resistance-type strain sensors have been increasingly used to measure the strains on the surfaces of a structure. To this end, two types of strain sensors have been developed, i.e., SWNT buckypaper sensors [5-7] and sensors made from polymer CNTs nanocomposites [7-13]. As compared to conventional sensors, higher sensitivity has been observed in these novel sensors, at least at a macro-scale [7, 10-13]. Linear piezoresistivity has been identified in these sensors [7, 9, 12], whereas the nonlinear piezoresistivity has also been reported [8, 10, 11]. In spite of these promising results, the fundamental understanding of conductivity in a CNT polymer nanocomposite is still lacking, largely due to the less effort being put into theoretical and numerical...
investigations on the piezoresistance behavior in these materials.

In this work, to investigate piezoresistive behavior which underpins the working principle of these sensors, we propose an improved 3D statistical resistor network model by which the tunneling effect among randomly distributed CNTs in a polymer matrix can be evaluated. The change of CNT networks in the polymer under a given strain is predicted using a fiber reorientation model. Then, the resistance change of the nanocomposites caused by the applied strain is estimated by using the 3D resistor network and the fiber reorientation models in an iterative way. To verify the numerical simulation, experiment has also been conducted on the sensors made from polymer/multi-walled carbon nanotube (MWNT) nanocomposites.

2. Computational procedure

To predict the electrical conductivity of the nanocomposite, a 3D resistor network that contains randomly distributed CNTs in the polymer has been constructed. So far, the resistor network model has been well documented [14-16]. However, because it is a troublesome and large-scale computational work, numerical studies based on a fully 3D statistical resistor network model, even for predicting the electrical conductivity of conventional electronic composites with filler materials such as short fibers, have been very limited. Most of these models have been based on the RC (resistor-capacitor) from a simulated microstructure. For convenience, in this research, the electrical conductive paths in the matrix phase are completely neglected. Also, as shown in Fig. 1(a) for a fractured surface of a sample with a 2.0 wt% MWNT loading [11], there is no obvious aggregation in our various specimens using different fabrication processes for this nanocomposite. Therefore, aggregation of CNTs is neglected in this study. The CNTs
are considered as ‘soft-core’ cylinders of length $L$ and diameter $D$, and are allowed to penetrate each other [14]. This assumption can lead to the tremendous reduction in the computational cost, which results in a proper solution method suitable for the Monte-Carlo procedure used here. For the ideal state of uniformly dispersed straight CNTs in matrix as shown in Fig. 1(b), the simulations are carried out using the following procedure:

(a) First, a 3D unit cell is constructed. The size of the unit cell is varied in order to achieve a stable and converged electrical conductivity of nanocomposites.

(b) Next, CNTs are randomly put (one at a time) in the 3D cube and their orientations in space are chosen randomly as follows:

The coordinates of two ends of a randomly dispersed CNT, i.e., $(x_1, y_1, z_1)$ and $(x_2, y_2, z_2)$ can be set as follows

\[x_1 = \text{rand} \times L_x, \quad y_1 = \text{rand} \times L_y, \quad z_1 = \text{rand} \times L_z \quad (1a)\]

\[x_2 = x_1 + L \cdot v_1 \cdot \cos(w_1), \quad y_2 = y_1 + L \cdot v_1 \cdot \sin(w_1), \quad z_2 = z_1 + L \cdot u_1 \quad (1b)\]

where $L_x$, $L_y$ and $L_z$ are the lengths of the 3D element along $x$, $y$ and $z$ axes, respectively, as shown in Fig. 1(b), \text{rand} is a random number located in $[0,1]$, which is uniformly generated. Also, the parameters representing alignment directions of CNTs, i.e., $u_1$, $v_1$ and $w_1$ are expressed as follows:

\[u_1 = 1.0 - 2.0 \times \text{rand}, \quad v_1 = \sqrt{1.0 - u_1^2}, \quad w_1 = 2\pi \times \text{rand} \quad (2)\]

Some generated CNTs may be partially located outside of the 3D representative cube. In this case, by finding the interactions of these CNTs with the 6 boundary planes of 3D cube, the portions on these CNTs, which are located outside of the 3D cube, are removed automatically and the intersections on the 6 boundary planes are
numbered as the ends of these CNTs.

(c) Each time after a new CNT is added into the unit cell, it is checked if it is in contact with one or more of CNTs already present in the unit cell. This is done by determining the minimum distance between the axes of the CNT in question and the axes of the remaining CNTs. In general, the shortest distance \( d \) between two skew CNTs can be calculated from the length of common perpendicular to the two axis lines of CNTs. If such distance for two CNTs is smaller than the nanotube diameter \( D \), the CNTs are considered to be in contact.

(d) When two CNTs are found to be in contact, the intersection is numbered. Until the amount of added CNTs reaches to the required volume fraction of CNTs in the 3D cube and all intersections among CNTs are numbered sequentially to form a global conductive network.

To construct the 3D resistor network model as shown in Fig. 2 (only a 2D model is shown), for a CNT with two contacting points \( i \) and \( j \) with neighbouring CNTs, the conductance \( g_{ij} \) between \( i \) and \( j \) (the inverse of resistance \( R_{ij} \)) can be evaluated as:

\[
g_{ij} = \sigma_{CNT} \frac{S_{CNT}}{l_{ij}}
\]

where \( l_{ij} \) is the length between the points \( i \) and \( j \), and \( \sigma_{CNT} \) and \( S_{CNT} \) are electrical conductivity and cross section area of the CNT, respectively.

The tube-tube contacts among CNTs are assumed to be perfect here with zero resistance. Based on the well-known matrix representation for a resistor network [14-16] and Kirchhoff’s current law, the total current \( I \) under an applied voltage can be estimated. This is a large-scale linear system, because the number of CNTs involved in the numerical model is very large, and ranges from several thousands to several tens of
thousands depending on the aspect ratio of the CNTs. An iterative equation solver, i.e.,
the incomplete Cholesky conjugate gradient method (ICCG) has been employed to
solve these linear equations for obtaining the total current $I$. Then the macroscopic
electrical conductivity of nanocomposites can be evaluated by the Ohm’s law.

Due to the short inter-distance between adjacent CNTs, it is necessary to
investigate the possible tunneling effect among the CNTs and its effects on the electrical
conductivity. The random distribution and possible tunneling paths between adjacent
CNTs have been examined using scanning electron microscopy (SEM), as shown in Fig.
3(a). This physical picture is simplified as a model for evaluating the resistance, as
schematically shown in Fig. 3(b). The tunneling resistance between two neighbouring
CNTs can be approximately estimated as [17],

$$ R_{tunnel} = \frac{V}{AJ} = \frac{h^2 d}{Ae^2 \sqrt{2m\lambda}} \exp \left( \frac{4\pi d}{h} \sqrt{2m\lambda} \right) $$

(4)

where $J$ is tunneling current density, $V$ the electrical potential difference, $e$ the quantum
of electricity, $m$ the mass of electron, $h$ Planck’s constant, $d$ the distance between CNTs,
$\lambda$ the height of barrier (for epoxy, 0.5 eV~2.5 eV), and $A$ the cross sectional area of
tunnel (the cross sectional area of CNT is approximately used here).

To consider the tunneling effect among CNTs, there is a small modification in Step
(c) stated above for judging the contacting state among CNTs. If the shortest distance $d$
between two CNTs is: $D < d \leq D + d_t$, where $d_t$ is the cutoff distance of tunneling
effect, two new nodes are added as shown in Fig. 3(b). By using Eq. (4), we can
evaluate the tunneling conductivity corresponding to various distances between two
CNTs and various $\lambda$ as shown in Fig. 3(c). From this figure, the above cutoff distance $d_t$,
is set to be 1.0 nm in this study, which relates to very low tunneling conductivity (lower than $10^2$ S/m in Fig. 3(c)) compared with that of CNTs, i.e. $10^4$ S/m used later. Note that in our numerical model, the ‘soft-core’ CNTs are used, therefore there may be overlapping among CNTs to a certain extent in our numerical model. Actually, this un-physical overlapping can be avoided by translating the newly added CNT in a randomly selected direction until the minimum distance between the two CNTs becomes equal to $D$. However, to avoid multiple penetrations of the new CNT with many other pre-existing CNTs is a very time-consuming task as the CNT loading increases in polymer, therefore this will not be further pursued here. This overlapping may result in possible errors in the following stage where we evaluate the pizoresistivity of sensors, since the possibility of breakup of CNT network may be slightly underestimated for serious penetration states among CNTs. However, when the volume fraction of CNTs is low and the aspect ratio of CNTs is large, the error caused by this overlapping is negligible if we explore this problem qualitatively.

3. Results and discussion

3.1 Electrical conductivity of nanocomposites

First, we compare the experimental electrical conductivity of nanocomposites with that predicted using the above 3D resistor network model. Here, we briefly describe the fabrication process for making nanocomposites [11]. In experimentation, the polymer/CNT nanocomposite was fabricated by in situ polymerization. MWNTs (060125-01K) were obtained from the Nano Carbon Technologies Co. (NCTC) in Japan. They were made via chemical vapor deposition, with a purity of higher than 99.5%. The average diameter and length of the MWNTs were 50 nm and 5 µm, respectively. The
aspect ratio was approximately 100. An insulating bisphenol-F epoxy resin (jER806, Japan Epoxy Resins Co., Ltd.) and an amine hardener (Tomaido 245-LP, Fuji Kasei Kogyo Co., Ltd.) were used. The nanocomposite was prepared by mixing the epoxy and the hardener using a planetary mixer at 2000 rpm for 20 seconds. Then, the MWNTs were added into the mixture and mixed again at 2000 rpm for 1 minute. The final mixture was poured into a silicon mold, and cured in a vacuum oven at 80°C for 3 hours. The experimental specimens with a length of 70 mm, a width of 20 mm, and a thickness of 2 mm were prepared from the cured epoxy/CNT mixture. Silver paste was placed on the two sides of the specimens to maintain good contact between the sample surfaces and the electrodes. The electrical conductivity of this nanocomposite was evaluated using a four-probe resistance method in dry air at ambient temperature. LCR meter (HIOKI 3522-50) with Cu electrodes was used. Five specimens were measured to obtain the average values of electrical conductivity of the current nanocomposite. The isotropy of electrical conductivity along length, width and thickness directions of the specimens and the stable distribution of electrical conductivity in several sub-segments along the length direction of the specimens have been experimentally checked and guaranteed.

In principle, the electrical conductivity should be independent to unit cell sizes if there are sufficient CNTs in the matrix as a stable conductive network is expected to form. To reduce the computation cost, the numerical simulation are conducted in a volume cell with dimensions of 25 µm (length) × 25 µm (width) × 25 µm (thickness) containing MWNTs of the length of 5 µm and the diameter of 50 nm, which has been tested to be large enough to achieve an isotropic behavior and numerical convergence.
Generally, $\sigma_{\text{CNT}}$ of MWNTs in Eq. (3) ranges from $5 \times 10^3$ to $5 \times 10^6$ S/m [18, 19]. Considering a situation where the aspect ratio ($L/D$) of a CNT is 100 and $\sigma_{\text{CNT}}=10^4$ S/m, the average conductivity predicted from 50 Monte-Carlo numerical simulations without consideration of tunneling effects among CNTs is compared with the experimental measurement, as shown in Fig. 4(a). Two other experimental results [20, 21], obtained with the same MWNTs but different fabrication processes, are also included. It is clear that the numerical prediction is in good agreement with the experimental data, confirming the capability of the numerical model in evaluating the conductivity of these nanocomposites. Moreover, as shown in Fig. 4(b), the tunneling effect can be identified by the increase of electrical conductivity when the volume fractions of CNT are near the percolation threshold of the composite. The percolation threshold is around 0.6165 vol% obtained from the statistical percolation model for CNTs of the aspect ratio of 100 [11]. The tunneling effect disappears gradually with increasing the amount of added CNTs. This result implies that high sensitivity in strain measurement can be achieved in this nanocomposite if the CNT loading is managed to be close to the percolation threshold. A similar result was reported in some experimental investigations, e.g. [7].

3.2 Piezoresistivity of sensors made from nanocomposites

The working mechanisms of strain sensors made from conventional electronic composites, such as short carbon fiber fillers, have been widely investigated [17, 22-25]. In terms of piezoresistivity, it was mainly attributed to (1) breakup of network formed by conductive fillers or loss of contact between the fillers [22, 24]; (2) increase of the inter-filler distances promoting the tunneling effect [17, 23, 25]. Besides these two factors, another possible mechanism was considered to be the conductivity change of
the CNTs when subjected to stresses, confirmed in a single SWNT by experiment [1] and a theoretical approach [26] and a single MWNT [27] by experiment, and a nanocomposite [12] although these results were of high discrepancy, which still need further evidence. Until now, very limited theoretical analysis and numerical simulation have been conducted in the traditional conductive electronic composites. Taya et al. [24] proposed a method to explain the effect of microstructure on the conductivity in filler and short fiber modified composites under finite strain. In their work, the threshold volume fraction of fiber was evaluated using a fiber percolation model while the fiber movement and reorientation with strain were taken into account. The effective conductivity of the composites was then estimated by using the power-law conductivity law with consideration of the change of percolation threshold in this law, which was caused by the redistribution of fibers due to strain.

In this work, on the other hand, the piezoresistivity is completely evaluated via numerical simulation, incorporating the change of inter-filler distances and possible breakup of conductive networks when subjected to strains. The resistance change of CNTs under elastic strain is ignored since its contribution can be considered to be insignificant under a small strain, as confirmed in the study of SWNT buckypaper film [5]. Also, the experiment used the tip of an AFM to manipulate MWNTs, revealing that changes in the sample resistance existed, but were very small unless the MWNTs were fractured [27]. In fact, very limited deformation is expected in the CNTs due to the poor stress transfer from the polymer matrix to these tubes, caused not only by the large elastic mismatch between the CNTs and the polymer but also by the weak interface strength. The elastic modulus of a CNT (1.0 TPa) is about 300 times higher than the
epoxy (2.4 GPa). Figure 5(a) also shows the complete debonding of a CNT from the polymer matrix, indicating low interface strength in our nanocomposite. Considering the rigid-body movement of the CNTs, the change of position and orientation of the CNTs under the effects of strain and Poisson’s ratio are evaluated using the 3D fiber reorientation model [24] based on the an affine transformation and the assumption of the incompressibility of the nanocomposite, as schematically shown in Fig. 5 (b). Corresponding to an updated distribution of the CNTs under a prescribed strain, a new network of CNTs can be formed by re-calculting the possible intersections between CNTs and tunneling resistances between CNTs within the cutoff distance. The switch of the intersections of CNTs to possible tunneling effect due to the breakup of CNT contacts and the distance update of pre-existing tunneling effects are modeled. Then, the resistance of the nanocomposite can be re-evaluated using the 3D resistor network. In this iterative way, the electrical resistance change of the nanocomposites with different CNT loadings has been investigated, as shown in Fig. 6(a). The same unit cell sizes, data of CNTs (electrical conductivity and aspect ratio) and the Monte-Carlo procedure as stated in Section 3.2 are employed. It is interesting to note that no consistent resistance change can be observed in the simulation if only the effect of the breakup of CNT conductive network is taken into account. Although our ‘soft-core’ model may partially underestimate this effect as stated previously, even for a low CNT loading (2.0 wt%) and a higher strain (1.0%), the tendency of resistance change is still unclear by only modeling the breakup of CNT network. In other words, it may imply that the contribution of the network breakup is not significant for small strains (under 1.0 %), in contrast to the case under a much higher strain [24]. Once again, the tunneling effect
plays a very important role in determining the overall performance of the nanocomposites when the CNT loading is close to the percolation threshold. According to Eq. (4), a 1 Å increase of $d$ (the distance between two CNTs) can lead to 10 times lower tunneling current. From Eq. (4), the tunneling resistance increases exponentially with the average distance $d$. Approximately, $d$ is assumed to change proportionally to an applied strain. As a result, a nonlinear relationship between the resistance and an applied strain is expected, as shown in Fig. 6(a). Especially for the cases of low CNT loading and higher strains, this nonlinear behavior is more obvious which indicates that the tunneling effect plays a dominant role in these cases. For convenience, this nonlinear piezoresistivity can be calibrated in a log-log plot in an approximate linear form, as shown in Fig. 6(b).

To verify the reliability of the numerical simulation, the variation of resistance with mechanical strain has been investigated using a piece of the nanocomposite with a thickness of about 170 µm, as shown in Fig. 7(a). Note that, in experiments, this nanocomposite sheet was attached to the top surface of an insulating cantilevered beam of the thickness of 2.0 mm, and a traditional strain gauge was glued to the bottom surface of the beam, in a symmetrical position to the nanocomposite sheet. With this arrangement, the strain gauge was able to measure the strain close to the bottom surface of the beam. The position of two sensors was close to the clamped end of the beam. As mentioned before, the electrical resistance of the nanocomposite sensor was measured using a LCR meter and the results are shown in Fig. 7(b). Compared to the results in Fig. 6(a), it is clear the numerical simulation which incorporates the tunneling effect among the CNTs can qualitatively catch the variation of resistance with the strain although the
‘soft-core’ CNTs are modeled here. On the other hand, in comparison to a traditional strain gauges whose gauge factor (sensitivity) is 2, higher gauge factors can be observed in these polymer/CNT sensors with different CNT loadings. For instance, the gauge factor of the sensor with 1.0 wt% CNT loading is about 8 times higher than that of the traditional strain gauges. When the CNT loading approaches to the percolation threshold, the gauge factor of the polymer/CNT sensors increases remarkably. These experimental results also confirm our numerical results in Fig. 4(b). Weak nonlinear piezoresistivity is observed in these nanocomposite sensors for the cases of lower CNT loading (Fig. 7(b)). However, corresponding to a high CNT loading level, the piezoresistivity can be approximately regarded as linear under small strains. This experimental result can also be calibrated as shown in the log-log plot (Fig. 7(c)).

4. Conclusions

In summary, extensive numerical simulation and experimental measurement have been conducted to understand the piezoresistivity in the polymer/CNT nanocomposites for applications as strain sensors. In combination of a 3D resistor network model with a 3D fiber reorientation model, the resistance change and tunneling effect and their dependence with applied strains and the redistribution of CNTs are successfully simulated. There is a good agreement between the numerical simulation and experimental measurement. Compared to a traditional sensor of strain gauge, higher sensitivity is observed in these nanocomposite sensors, especially when the CNT loading is close to the percolation threshold. Under small strains, the resistance change is dominated by the tunneling effect between the neighbouring CNTs, instead of the breakup of the conductive network. The nonlinear piezoresistivity is numerically and
experimentally identified in these nanocomposites for the cases of low CNT loading, which can be explained qualitatively by tunneling effect. However, at a high CNT loading level, the piezoresistivity can be approximately regarded as linear. Further work is required to address the effect of strain on the resistance in the CNTs.

References


Captions of the figures

Fig. 1. Modelling of nanocomposites containing randomly distributed CNTs: (a) SEM image of fracture surface of the nanocomposite, (b) A 3D representative unit element

Fig. 2. Schematic of a resistor model with random distribution of CNTs

Fig. 3. Modelling of tunneling effect in the resistor network: (a) SEM image of possible tunneling effect among CNTs in the nanocomposites, (b) Modelling of tunneling resistance in the resistor network, (c) Tunneling conductivity for various distances

Fig. 4. Results of electrical conductivity of nanocomposites: (a) Comparison of experimental and numerical conductivities, (b) Effect of tunneling effect on conductivity

Fig. 5. Reconstruction of CNT network in polymer for predicting sensor piezoresistivity: (a) Evidence of weak interface between polymer and CNTs (SEM image), (b) Rigid-body movements of CNTs in polymer due to strain and Poisson’s ratio

Fig. 6. Numerical piezoresistivity of sensor made from nanocomposites: (a) Numerical piezoresistivity of sensor for various CNT loadings, (b) Logarithm plot of numerical piezoresistivity

Fig. 7. Experimental piezoresistivity of polymer/CNT nanocomposite strain sensor: (a) Polymer/CNT nanocomposite strain sensor, (b) Experimental piezoresistivity in the polymer with different CNT loadings, (c) Logarithm plot of experimental piezoresistivity
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(b) Logarithm plot of numerical piezoresistivity

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Fig. 7. Experimental piezoresistivity of polymer/CNT nanocomposite strain sensor

(a) Polymer/CNT nanocomposite strain sensor

(b) Experimental piezoresistivity in the polymer with different CNT loadings

(c) Logarithm plot of experimental piezoresistivity