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# Electrostriction study for single-walled carbon nanotubes-based composite

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This paper proposes a 3D finite element method procedure to study the electrostrictive property of the single-walled carbon nanotube (SWCNT)-based composite. The numerical model in nanoscale is developed to investigate the electrostrictive behavior of the SWCNT and the polyvinylidene fluoride and trifluoroethylene P(VDF-TrFE) composite. A bond electrical contact model is adopted to reproduce the coupled electromechanical effects of the interface between SWCNT and P(VDP-TrFE). According to the model proposed, the mechanism of the enhanced electrostriction in SWCNT/P(VDP-TrFE) composite is intuitively demonstrated. Numerical results show that aside from the variation of electric field, the electrostriction of the SWCNT/P(VDP-TrFE) composite is greatly depend on the volume fraction of SWCNT and the difference of dielectric constant between SWCNT and P(VDP-TrFE) copolymer. While the dielectric constant of SWCNT is mainly depend on the chirality of SWCNT, which means that the electrostriction of the SWCNT/P (VDP-TrFE) composite is also largely depend on the chirality of SWCNT.

**Keywords:** nanocomposite; SWCNT; electrostriction; nanotechnology

## 1. Introduction

Since the discovery of the piezoelectricity by the Curie brothers in 1880–1881,[1] piezoelectric materials have been extensively adopted as sensors, actuators, and transducers in a variety of smart structure applications. Polyvinylidene fluoride and trifluoroethylene P(VDF-TrFE) copolymer, which has the properties of ferroelectricity, piezoelectricity, and pyroelectric to convert the electrical excitation to mechanical motion directly, shows great prospection in practical application. It is demonstrated that the P(VDF-TrFE) copolymer under a proper irradiation treatment can produce an electrostrictive strain of 5%.[2] The giant electrostrictive property makes P(VDF-TrFE) copolymer to be a preferred piezoelectric material.

Piezoelectricity arised from the small changes in the internal polarization of a dielectric material under applied mechanical changes. Alternative methods of increasing dielectric constant to improve piezoelectricity have been proposed. Li and Rao [3,4] presented a microstructure analysis for the electrostriction of the P(VDF-TrFE) polymer-based composite and elaborated the mechanism of the electrostriction enhancement in the composite. Their reports demonstrated that using the concept of composite, the

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effective electrostriction of the material can indeed be enhanced by incorporating a second phase with much larger dielectric constant than that of the ferroelectric polymer, even if the electrostriction of the second phase is zero. The enhanced dielectric constant for polymer matrix composite materials has also been reported in both analytical and experimental works.[5–9]

Recent developments in nanotechnology show that the carbon nanotube (CNT) rolled single (SWCNT) or multiple (MWCNT) sheets of graphite with diameter in nanometer size have unique electronical, mechanical, and chemical properties,[10] which make it a preferred material in the applications of nanocomposite and nanodevices. Guo demonstrated an exceptionally large axial electrostrictive deformation in SWCNTs using density functional theory.[11,12] El Hami and K. Matsushige used the AFM technique to apply a voltage across an aligned bundle of SWCNTs; the electrostriction strain of about 2.6% was detected under an applied electric field of 160 V/μm.[13] Deshmukh and Ounais investigated the coupled electromechanical response in a nonpolar polymer composite,[14] they discovered that the neat polyimide does not show any action response under an applied electric field, whereas the SWCNT-PI composites above the percolation threshold exhibit an electrostrictive behavior, and the strain is highly dependent on SWCNTs' content. Those exciting results indicate that carbon nanotube could be a prior reinforcement for electrostrictive composite.

Motivated by those efforts, this paper intends to investigate the electrostrictive effect of the P(VDF-TrFE) copolymer and single-walled carbon nanotube (SWCNT)-based composites. Here, we propose a 3D finite element method to predict the electrostrictive deformation of SWCNT-based P(VDF-TrFE) copolymer (SWCNT/P (VDF-TrFE)) composite under electrostatic field. Using finite element method, a bond electrical contact model is developed. Based on this model, the intrinsic electrostrictive properties of dielectric composite are intuitively demonstrated, which provides a numerical guideline for the design and optimization of the electrostriction for dielectric composites. The finite element model of SWCNT/P(VDF-TrFE) composite is demonstrated in Section 2.1. The interfacial properties of SWCNT and P(VDP-TrFE) are described in Section 2.2. Section 3 analyses the effects for the electrostriction of SWCNT/P(VDF-TrFE) composite. The conclusion is given in Section 4.

## 2. Constitutive relationship of SWCNT-P(VDF-TrFE) composite

This numerical analysis is encouraged by the experimental model of El Hami [13,15]. In El Hami's work, they prepared the SWCNT/P(VDF–co–TrFE) composite with good physical property, the representation of the SWCNTs base composite is shown in Figure 1, which the SWCNTs are incorporated in the P(VDF–co–TrFE) copolymer in bundles state.

In order to study the effect of different structures of SWCNTs on the electrostriction of composite, here we assume that the individual SWCNT is aligned in the P(VDF-TrFE) copolymer matrix, as illustrated in Figure 2. Generally, the stiffness and electric filed distribution of composites are dependent on the contact media. In order to evaluate the whole deformation of the composites under electric field, the behavior of the contact region should be studied.

### *2.1. Finite element model of SWCNT/P(VDF-TrFE) composite*

Micromechanics technique is commonly used by many researchers to study the properties of composites at the macroscopic level. The micromechanics method aims at

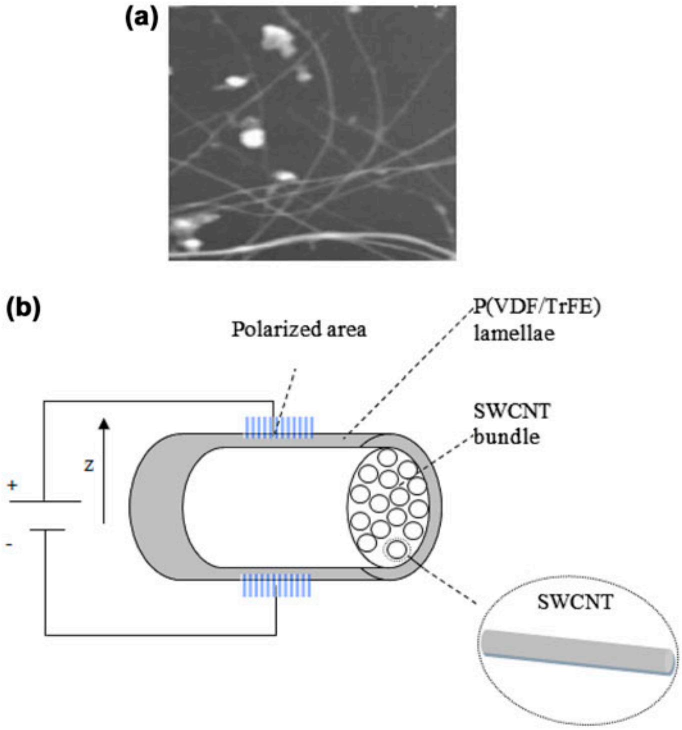


Figure 1. (a) Topography image of SWCNT bundles, scan area: $1000 \times 1000\,nm^2$[15] and (b) schematic illustration of SWCNT bundle covered by P(VDF-TrFE) lamellae with visible polarization domains.

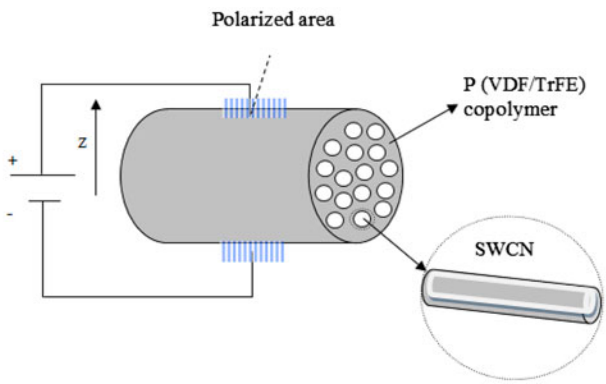


Figure 2. Schematic illustration of individual SWCNT aligned in the P(VDF-TrFE) copolymer matrix.

transforming a body of a heterogeneous material into a constitutively equivalent body of homogeneous material. Using micromechanics method, Ray and Batra determined the effective piezoelectric coefficient of armchair single-walled carbon nanotubes and piezoelectric fibers-based composite.[16] However, under the action of external electric field, the interfacial particle of SWCNT dielectric fiber and its surrounding piezoelectric polymer matrix are not free to deform because of the elastic clamping imposed by the second-phase material. Besides, the electrical and mechanical properties in composite media are different in matrix and reinforcement, which makes the electrostrictive strain not uniform. The traditional technique of micromechanics modeling could not take those factors into consideration. Therefore, it is crucial to define a rational interfacial relationship between SWCNT and copolymer matrix to predict the overall electrostrictive behavior of composite.

In this paper, one of the individual SWCNT and its surrounded copolymer matrix are taken from the composite as the analysis target. Figure 3 is the configuration of the model employed with the omitting mesh pattern. The P(VDF-TrFE) copolymer matrix is regarded as isotropic continuum medium that wrap outside the carbon nanotube. The inner radius of the cylindrical medium is equal to the radius of nanotube. The nanotube is viewed as a transversely isotropic column. As the piezoelectric effect of CNT is very small,[16] in the model, a comparable small value of piezoelectricity constant for SWCNTS is set. The piezoelectric constitutive equations for the P(VDF-TrFE) and SWCNT are

$$\begin{aligned} T^P &= C^P S^P - e^P E \\ D^P &= e^P S^P + \varepsilon^P E \\ T^N &= C^N S^N - e^N E \\ D^N &= e^N S^N + \varepsilon^N E \end{aligned} \tag{1}$$

where $T^P$, $C^P$, $S^P$, $D^P$, $\varepsilon^P$, $e^P$, and $T^N$, $C^N$, $S^N$, $D^N$, $\varepsilon^N$, $e^N$ are the stress vector, elasticity matrix, strain vector, electric displacement, dielectric permittivity, and the piezoelectric stress matrix of the P(VDF-TrFE) copolymer and SWCNT, respectively, and $E$ is the electric field intensity vector.

According to the Laplace's equation for the electric potential ($\varnothing$), the governing equation for $\varnothing$ in each media is [17]:

$$\nabla^2 \varnothing = \frac{\partial^2 \varnothing}{\partial^2 x} + \frac{\partial^2 \varnothing}{\partial^2 y} + \frac{\partial^2 \varnothing}{\partial^2 z} = 0 \tag{2}$$

The boundary conditions at the interface of SWCNT and P(VDP-TrFE) are as follows.

(1) The continuous tangential field strength is defined as

$$\varnothing_N = \varnothing_P \tag{3}$$

or

$$E_{tN} = E_{tP} \tag{4}$$

(2) The continuity of normal electric flux density $D_n$ is expressed as

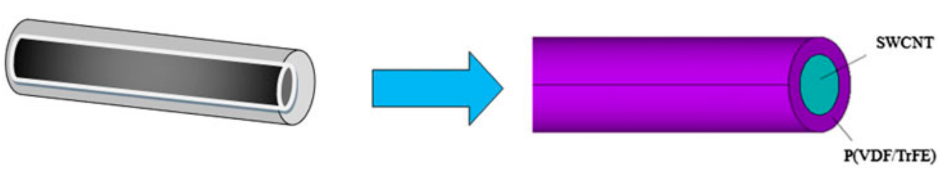


Figure 3. The configuration of the SWCNT/P(VDP-TrFE) composite.

$$D_{nN} = D_{nP} \tag{5}$$

which can be also written as

$$\varepsilon^N E_{nN} = \varepsilon^P E_{nP} \tag{6}$$

In Equations (3)–(6), $\varnothing_N$, $D_{nN}$ and $\varnothing_p$, $D_{nP}$ are the electric potential and normal electric flux density at the interface of SWCNT and P(VDP-TrFE).

The finite element model developed in this study is based on the finite code ANSYS [18] and the finite element theoretical studies.[19–21] A three-dimensional twenty-node element Solid 226 and a three-dimensional ten-node element Solid 98 in ANSYS are adopted to model the P(VDF-TrFE) copolymer and SWCNT material, respectively. The electrostatic analysis is performed. The structure is symmetric about the *Z*-axis. The finite element model and the boundary conditions of the composite are shown in Figure 4.

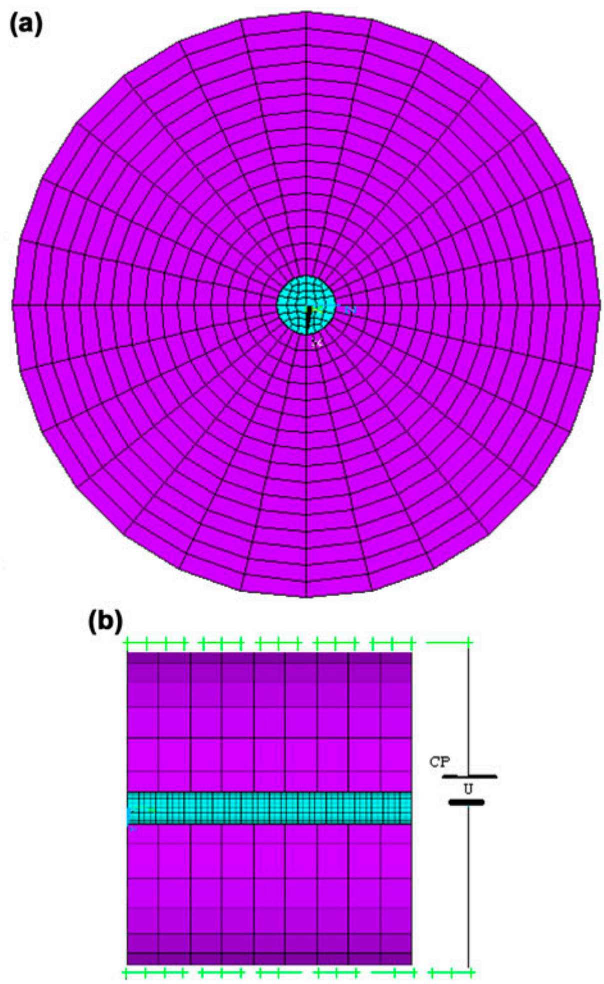


Figure 4. The finite element model of the SWCNT/P(VDP-TrFE) composite.

### 2.2. SWCNT/P(VDP-TrFE) interfacial properties

At the interfacial of SWCNT and its surrounding P(VDF-TrFE) copolymer, the effect of increased dielectric constant in composite is usually restricted by the highly increased stiffness, which causes a lower strain. Besides, the dielectric composite media composed of multiple dielectrics, the electric field near the contact region will significantly increase, which causes a higher force. However, the caused higher force may bring about local debonding, which will weaken the electric field of interfacial. In order to correctly illustrate the interaction between SWCNT and P(VDF-TrFE) copolymer, the electrical/mechanical behavior of SWCNT/P (VDF-TrFE) interface is characterized through the contact/target (conta174/targe170) element pairs in ANSYS,[22] and the surface-to-surface contact model is applied in this work, which illustrated in Figure 5. The interfacial bond strength between SWCNT and the surrounding matrix is assumed to be average along the length of composite, the bond stiffness is supposed to be strong enough that there is no decohesion under the electric excitation. At the interface, since the stiffness of SWCNT is much larger than the matrix, the SWCNT is selected as the target, which is overlaid by the contact element of P(VDF-TrFE) copolymer. The initial stiffness of the interface depends on the normal and tangential stiffness, which is determined by the relative stiffness of the two materials and the initial contact surface

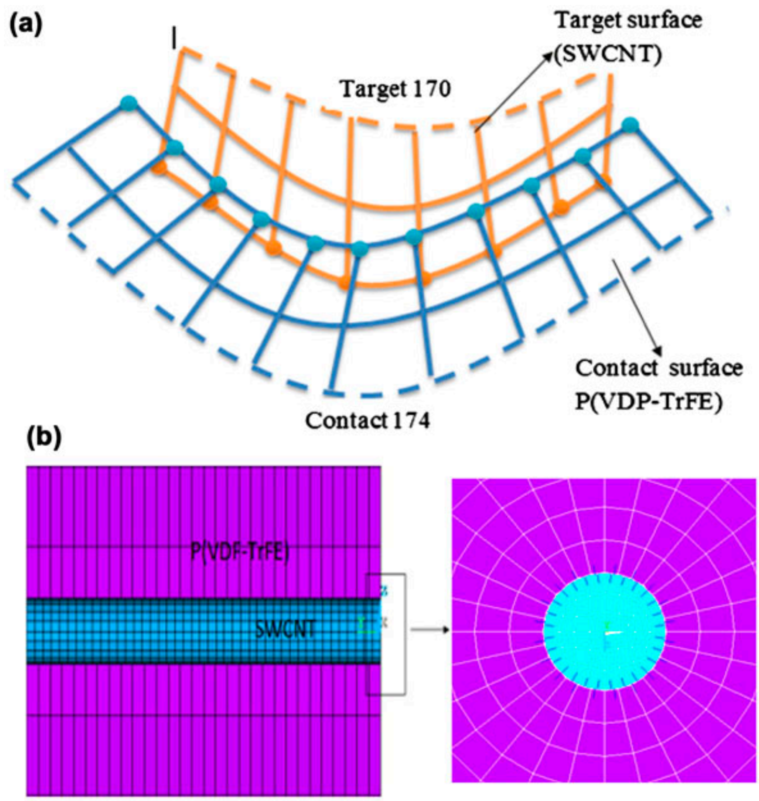


Figure 5. (a) The configuration of the SWCNT/P(VDP-TrFE) interfacial and (b) the finite element model of SWCNT/P(VDP-TrFE)interfacial.

condition setting in ANSYS. The contact stiffness is updated automatically after each iteration based on the initial condition of interface. The interaction between the SWCNT and the P(VDFTrFE) copolymer matrix is determined by the properties assigned to the contact element/target element pairs.

## 3. Results and discussion

SWCNTs can be metallic or semiconductor, which depends on the diameter and helicity of the carbon atoms. Band structure calculations have predicted that armchair SWCNTs with (*n*, *n*) indices exhibit metallic conduction with finite density of states at the Fermi level, whereas Zigzag SWCNTs with indices (*m*, 0) and chiral SWCNTs with indices (*m*, *n*) are predicted to be metallic when $m–n = 3 \times$ integer and semi metallic when $m - n \neq 3 \times$ integer.[23] Paul L. McEuen and his colleagues found that metallic tubes have conductivities and current densities that meet or exceed the best metals, and semiconducting tubes have mobilities and transconductances that meet or exceed the best semiconductors.[24] The electrical resistivity of individual SWCNTs has been measured under ballistic conduction of around $10^{-6}$Ωcm.[25] In this paper, the conductivity of SWCNTs is adopted as the commercial carbon of 285 $\mathrm{Scm}^{-1}$.[26] The elastic moduli and the dielectric constants of SWCNTs estimated from [25,27–29]. The elastic constant and electrical properties of P(VDF-TrFE) copolymer are obtained from.[30,31] The capacitance of SWCNT/P (VDF-TrFE) interface is obtained from Loh et al. [32].

### 3.1. *The effect of volume fraction of SWCNTs*

In this paper, firstly, the effect of volume fraction of SWCNTs for the electrostriction of composite is studied. In order to avert the radius' impact, we studied the variance of electrostriction to the corresponding volume fraction of armchair (5, 5), Zigzag (10, 0), and chiral (6, 4) SWCNTs-based composite. The SWCNT/P (VDF-TrFE) composites have the approximate diameters at the same SWCNT fraction.

A constant DC electric field of 0.1 MV/m is applied to the series of SWCNT/P(VDF-TrFE) composites. In order to reduce the edge effect and save the computational time, the length of the composite is selected according to the length to radius ratio ($h/r \geq 10$); the thickness strain is calculated by the measured thickness deformation $\Delta D$.

$$S_{33} = \frac{\Delta D}{D} \tag{7}$$

where $S_{33}$ is the thickness strain, and $D$ is the diameter of composite.

The thickness strains $S_{33}$ for composite with the volume fraction of SWCNT from 0.5 to 10 vol% are plotted in Figure 6. The results show that the strains increase non-linearly with the volume fraction of SWCNT. This result is in agreement with Kim et al.'s study [33] that SWCNT/P(VDP-TrFE) composites demonstrate higher dielectric constants and ferroelectricity than P(VDP-TrFE) copolymer depends on the weight fraction of CNT.

### 3.2. *The effect of electric field*

The variation of electrostrictions for armchair (5, 5), Zigzag (10, 0), and chiral (6, 4) SWCNTs-based composite with the respected electric field is then studied. As we assumed that there is no decohesion between SWCNT and P(VDP-TrFE) copolymer

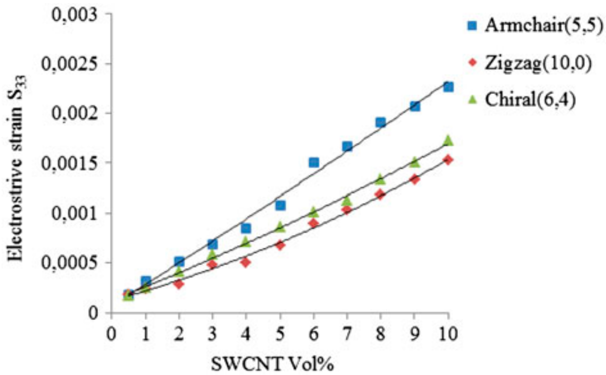


Figure 6. The electrostrictive deformation of the SWCNT/P(VDP-TrFE) composite with SWCNT fraction. Solid lines are quadratic fit.

under electric force, we applied a serious of relatively small electric field to the composite. Figure 7 shows the electrostrictive strains of SWCNT/P(VDF-TrFE) composite with 1 vol% of the SWCNT under different electric field, which demonstrates that the strains increase approximate linearly with the electric field.

From Figures 6 and 7, we find that the volume fraction of SWCNT has more influence to the electrostriction of the SWCNT/P(VDP-TrFE) composite than electric field. This could be due to (a) variation of dielectric constant in composite caused by different structures of SWCNT and (b) electric field intension in composite because of the second-phase reinforcement.

Figure 8 illustrates the electric field distribution in 0.5 vol% armchair (5, 5) SWCNT/P(VDP-TrFE) composite. Figure 8(a) and (b) shows that the electric fields strength at the interface is more than 100-fold the edge of composite.

The results obtained in Figure 8 can be explained by the work of Takuma and Techaumnat's study [17]. The contact of a cross-sectionally straight dielectric interface as represented in Figure 9(a). At some points on the boundary of two dielectrics, the

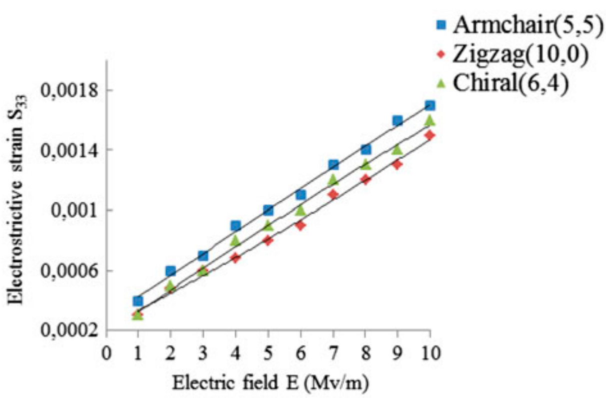


Figure 7. The electrostrictive deformation of the SWCNT/P(VDP-TrFE) composite with electric field. Solid lines are linear fit.

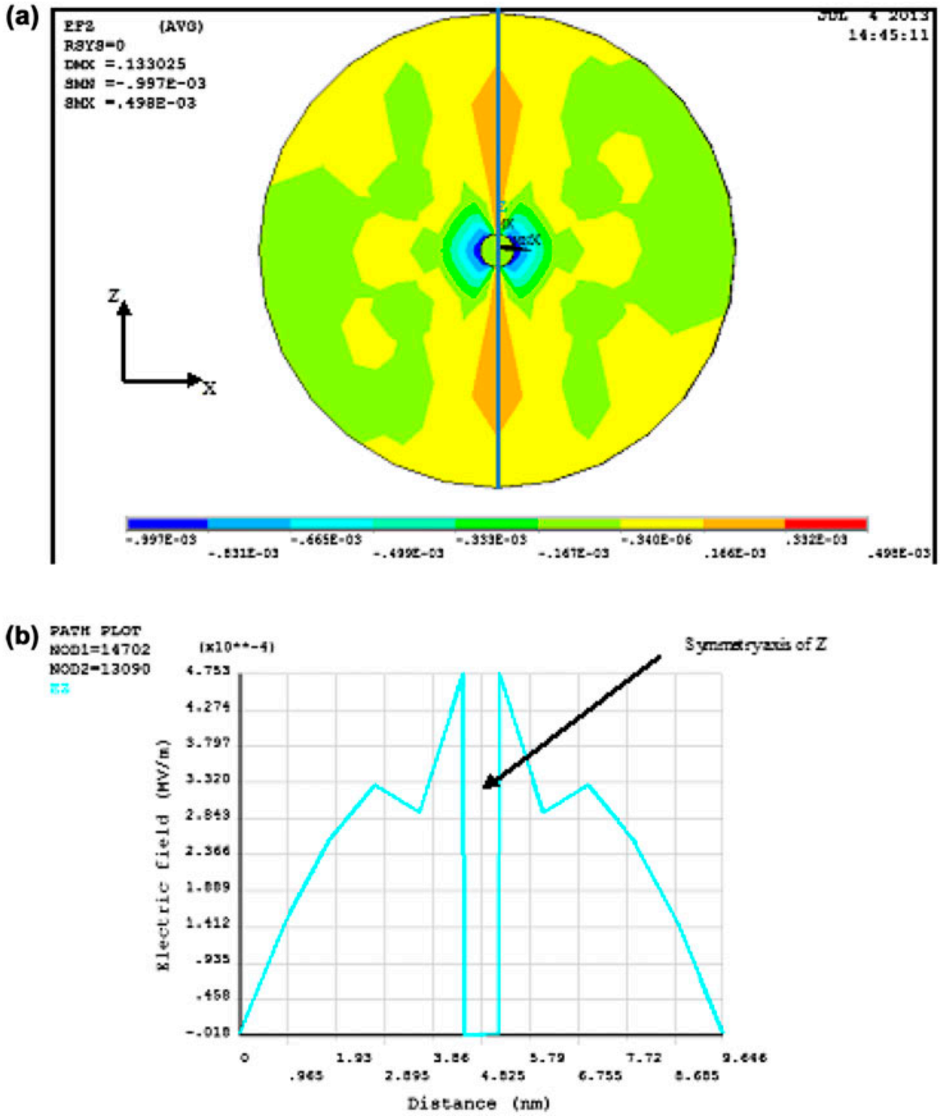


Figure 8. The electric field distribution in 0.5 vol% armchair (5, 5) SWCNT-based composite. (a) The *z*-component of electric field distribution and (b) electric field distribution along the symmetry axis *z*.

electric field strength $E$ may change as a function of $k \cdot l^m$. $l$ is the distance from the contact point, and $K$ is a constant depending on the overall physical configuration and the ratio of the two dielectric constants $\varepsilon_1/\varepsilon_2$. The exponent m is negative or positive according to whether the ratio of the two dielectric constants is greater or less than unity. This means the electric field distribution of composite is greatly dependent on the ratio of two dielectric constants. Figure 9(b) illustrates the electric field strength on the A and B sides along the interface with different ratio of two dielectric constants. Figure 9(b) also indicates that the more the dielectrics near the interface, the larger of

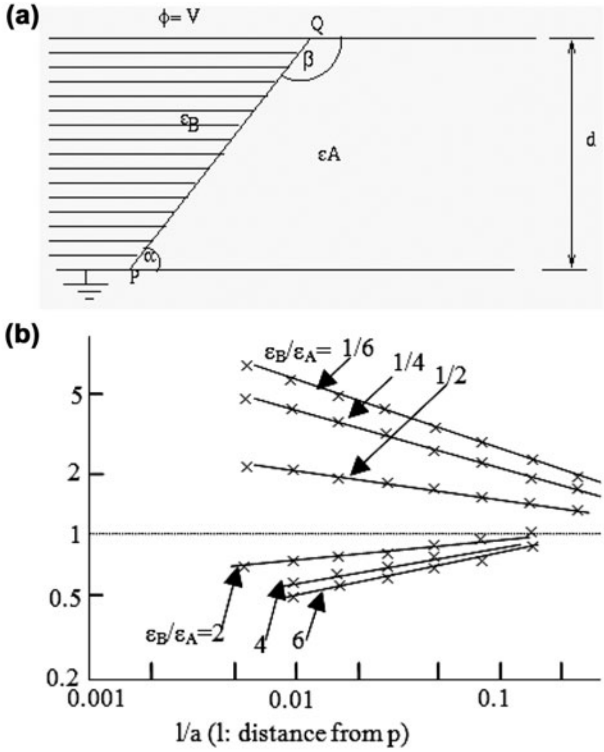


Figure 9. (a) The interface of two dielectrics and (b) electric field strength on the A and B sides along the interface.[17]

the field strength, the further the dielectrics away from the interface, the field strength trends to the initial field of $E_0$ ($E_0$ = U/D). Our numerically results (Figure 8(a) and (b)) agree well with this theoretical analysis.

### 3.3. *The effect of chirality*

From Figures 6 and 7, we can also conclude that with the approximate diameters but different chirality of SWCNT-based SWCNT/P(VDP-TrFE) composite, the armchair (5, 5)/P(VDP-TrFE) composite has the largest electrostrictive deformation while Zigzag (10, 0)/P(VDP-TrFE) composite has the smallest electrostrictive deformation at the same external electric field and volume fraction of SWCNT. This is mainly due to the difference in dielectric constants of SWCNT and P(VDP-TrFE) not the dielectric constants of SWCNT itself. Guo and Chu's study [25] shows that armchair (5, 5) SWCNT has a larger static dielectric constant than Zigzag (10, 0) SWCNT, and Zigzag (10, 0) SWCNT has a larger static dielectric constant than chiral (6, 4). While the tendency of the electrostrictive deformation for those three kinds of SWCNT/P (VDP-TrFE) composite is not with this rule but consistent with the ratio of dielectric constants for SWCNT to P(VDP-TrFE). The static dielectric constant for SWCNT and the ratio of two dielectric constants are listed in Table 1. Compare the ratio of dielectric constants for SWCNT to P(VDP-TrFE), our results are agree well with the theoretical analysis for dielectric composite in [17].

Table 1. Static dielectric constant for SWCNT from [25] and dielectric constant for P(VDP-TrFE) from [28].

| | $\varepsilon_{xx}$ ($\varepsilon_{zz}$) | $\varepsilon_{yy}$ | $\varepsilon_{xx\mathrm{CNT}}/\varepsilon_p$ |
|---|---|---|---|
| Armchair (5, 5) | 16 | 34.1 | 16/11 |
| Zigzag (10, 0) | 8.2 | 26.8 | 8.2/11 |
| Chiral (6, 4) | 7.3 | 22.1 | 7.3/11 |
| P(VDP-TrFE) | 11 | 11 | |



## 4. Conclusions

This paper proposes a 3D finite element method to study the electrostrictive property of SWCNT/P(VDP-TrFE) composite. The electrostriction of armchair (5, 5), Zigzag (10, 0), and chiral (6, 4) SWCNT-based composite is studied. The mechanism of the enhanced electrostriction in SWCNT/P(VDP-TrFE) composite is intuitively demonstrated in the model proposed. Numerical results show that the electrostriction of SWCNT/P(VDP-TrFE) composite is greatly depend on the volume fraction of SWCNT and the differences of dielectric constant between SWCNT and P(VDP-TrFE) copolymer. While the dielectric constant of SWCNT is depend on the chirality of SWCNT, which provide a numerical guideline for the design and optimization for SWCNTs-based nanocomposites and other electrostrictive composite.

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