

## ARTICLE

# High dielectric constant epoxy nanocomposites containing ZnO quantum dots decorated carbon nanotube

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## Abstract

Zinc oxide (ZnO) quantum dot (QD) decorated multi-walled carbon nanotube (MWCNT) hybrid was utilized in the fabrication of high dielectric constant epoxy nanocomposites. Because of the shielding effect of ZnO QD, the well-dispersed epoxy hybrid nanocomposites exhibit frequency insensitive high dielectric constant as well as greatly reduced dielectric loss. With only 1.5 wt% of MWCNT addition, the epoxy/MWCNT-ZnO nanocomposite possesses dielectric constant as high as 31 and dielectric loss as low as 0.01 at 1 kHz. In addition, the epoxy nanocomposite exhibits greatly enhanced tensile properties. The role of ZnO QD decorated MWCNT in the preparation and property improvement of multi-functional polymer nanocomposites is discussed.

## KEYWORDS

dielectric properties, films, graphene and fullerene, resins, surfaces and interfaces

## 1 | INTRODUCTION

High dielectric constant (high-k) oxides and high-k polymer nanocomposites have been extensively studied and evaluated because of their promising applications in electronics.<sup>1–3</sup> Some potential fields of application include organic field-effect transistor,<sup>4,5</sup> invertors,<sup>6</sup> electro-optics,<sup>7</sup> sensors,<sup>8</sup> and energy storage devices.<sup>9,10</sup> Dielectric polymer nanocomposites are generally composed of a dielectric polymer as the matrix and nanoscale inorganic/organic fillers for enhancement.<sup>11,12</sup> Polymers have been demonstrated to exhibit high breakdown strengths along with easy processing, while the nanofillers are usually composed of high-k ceramic or conductive nanoparticles.<sup>13,14</sup> The combination of both usually provide superior dielectric properties.<sup>15</sup>

Conventional high-k polymer nanocomposites use ceramic materials as fillers.<sup>16</sup> These ceramic-based materials possess excellent dielectric permittivity and low dielectric loss, as well as excellent thermal stability.<sup>16</sup> However, high loadings (over 30 wt%) of the ceramic

fillers would usually be needed to achieve permittivity greater than 30, making it difficult for processing and resulting in poor mechanical properties.<sup>17</sup> An alternative strategy is to incorporate conductive fillers to the polymer matrix to prepare percolative insulator/conductor polymer nanocomposites, which can have high permittivity when the content of the conductive filler is slightly below its percolation threshold.<sup>18,19</sup> For polymer composites containing conductive fillers, the polarization characteristics of the matrix molecule, conductive filler, and their interface collectively play a pivotal role on the dielectric properties.<sup>20,21</sup> According to the percolation theory<sup>22</sup> and the Maxwell–Wagner–Sillars interfacial polarization effect,<sup>15</sup> the charge carriers in the composites would transport from the polymer matrix to the conductive fillers, resulting in the accumulation of space charge polarization at their interface. Among the conductive fillers, multi-walled carbon nanotube (MWCNT) is often used to prepare high-k polymer nanocomposites due to their excellent electrical properties and high aspect ratio, which reduces the concentration needed to reach the

desired percolation threshold.<sup>23</sup> The MWCNT-based polymers can possess a high dielectric constant at low MWCNT loadings.<sup>24,25</sup> However, the insulator/conductor pairs often lead to high dielectric loss as a result of high leakage currents due to possible direct contact between conductive fillers in the polymer matrix near the percolation threshold, which hinders their commercial applications.<sup>26</sup>

In order to address the aforementioned drawbacks, a core-shell structure has been proposed and prepared by coating CNT with a layer of insulating polymer.<sup>27,28</sup> MWCNT coated with phosphaphenanthrene<sup>29</sup> or multi-branched polyaniline<sup>30</sup> has been utilized as fillers in high-k polymer nanocomposites. The nanocomposites with core-shell hybridized CNTs not only exhibit high-k but also maintain relatively low dielectric loss, suggesting such a strategy is a promising approach to prepare high-k nanocomposites.<sup>31</sup> However, surface modification of CNTs with low molecular weight surfactants or polymers usually leads to an adverse effect on glass transition temperature ( $T_g$ ) and coefficient of thermal expansion of the hosting polymer, making it undesirable for microelectronic applications.<sup>19,32</sup> To overcome the disadvantages of organic coatings on MWCNT, inorganic materials, such as  $\text{SiO}_2$ ,<sup>33</sup>  $\text{CuO}$ ,<sup>34</sup> and  $\text{TiO}_2$ ,<sup>35</sup> were used to modify the MWCNT surfaces to maintain the physical and mechanical properties of the hosting matrix. However, the poor dispersion of the inorganic particle-coated CNTs in their polymer matrices limited their effectiveness for property improvement. MWCNT decorated by zinc oxide quantum dot (ZnO QD) (MWCNT-ZnO) was previously shown to exhibit excellent dispersion in epoxy matrix.<sup>4</sup> Unfortunately, the dielectric properties of epoxy/MWCNT-ZnO nanocomposite have not been studied in the past.

It was also shown that the epoxy/MWCNT-ZnO exhibits significant improvements in Young's modulus (50%) and tensile strength (20%), without significantly compromising the  $T_g$  of the epoxy matrix. These improvements can be attributed to the increased interfacial interaction between MWCNT-ZnO and epoxy matrix, which benefits the load transfer. Regarding the dielectric properties, as the diameter of a particle decreases to QD size scale,<sup>36</sup> quantum confinement and Coulomb blockade effects will become dominant. Xie et al took advantage of this nanoscale phenomenon of ultra-small metallic nanoparticles to decrease dielectric loss and enhance the breakdown strength of high-k polymer nanocomposites.<sup>37</sup> Therefore, epoxy/MWCNT-ZnO is a good candidate as a high-k dielectric material while maintaining good physical and mechanical properties.

In this study, electrically insulative ZnO QD is chosen as a shield to prevent direct contact and charge transfer among the neighboring MWCNTs in the epoxy matrix.

It is highly possible that MWCNT-ZnO can be highly effective both as a high-k nanofiller to enhance dielectric properties greatly and as a reinforcement agent to improve the mechanical properties of the epoxy matrix. The effect of the loading of MWCNT-ZnO fillers on dielectric and tensile properties is studied. The usefulness of the MWCNT-ZnO hybrid for the preparation of multifunctional polymer nanocomposites is also discussed.

## 2 | EXPERIMENTAL SECTION

### 2.1 | Pretreatment of MWCNTs

To partially oxidize MWCNT, 100 mg of MWCNTs were added in a 30 ml mixture of sulfuric acid and nitric acid at a 3:1 vol/vol ratio. Then, the solution was ultrasonicated for 30 min at room temperature and for another 1 h after the addition of 190 ml of deionized water ( $\text{DI-H}_2\text{O}$ ). The oxidized MWCNTs were collected by filtration, washed four times with  $\text{DI-H}_2\text{O}$ , and additional four times with methanol ( $\text{MeOH}$ ). The partially oxidized MWCNTs were dispersed in  $\text{MeOH}$  at a concentration of 1 mg/ml for further preparation of ZnO decorated MWCNT.

### 2.2 | Synthesis of MWCNT-ZnO

The preparation of ZnO-decorated MWCNTs was conducted as described previously.<sup>38</sup>  $\text{KOH}$  (1 g) was added into oxidized MWCNTs (160 mg) suspended in  $\text{MeOH}$ , and sonicated for 5 min. Next, zinc acetate dihydrate (1.96 g) solution in  $\text{MeOH}$  (10 ml) was added. The system was heated to  $60^\circ\text{C}$  and refluxed for 2 h. After the reaction, the MWCNT-ZnO was filtered and rinsed four times with  $\text{MeOH}$  and then resuspended in  $\text{MeOH}$  for further preparation of epoxy nanocomposites.

### 2.3 | Preparation of epoxy/MWCNT-ZnO

The epoxy monomer used in this study is a diglycidyl ether of bisphenol F (DGEF) epoxy resin (D.E.R. 354 epoxy resin, The Dow Chemical Company). A diethyl toluene diamine (DETDA) (EPIKURE™ W, Momentive Specialty Chemicals) was used as a curing agent. The fabrication of epoxy nanocomposite thin films is described as follows. Initially, DGEF and DETDA in the weight ratio of 168:90 were thoroughly mixed in acetone with the assistance of ultrasonication. MWCNT-ZnO suspension in  $\text{MeOH}$  was added subsequently, and the mixture was then sonicated for 15 min. The excessive solvent was

removed by rotary evaporation at 60°C for 15 min. Afterward, the viscous mixture was degassed in vacuum at 80°C for 2 h to ensure the complete evaporation of the residual solvent. Then, the mixture was heated at 121°C for 45 min to afford low viscosity for thin film preparation. Subsequently, the Elcometer 4340 Motorized Film Applicator was used to prepare thin films with a thickness of 100  $\mu\text{m}$ . The film was then cured at 120°C for 2 h and post-cured at 170°C for an additional 3 h.

## 2.4 | Characterization

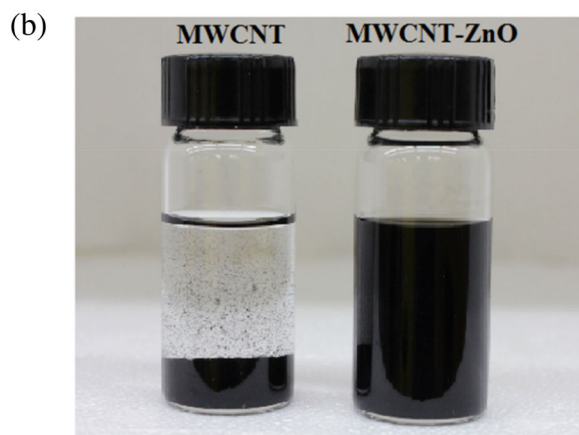
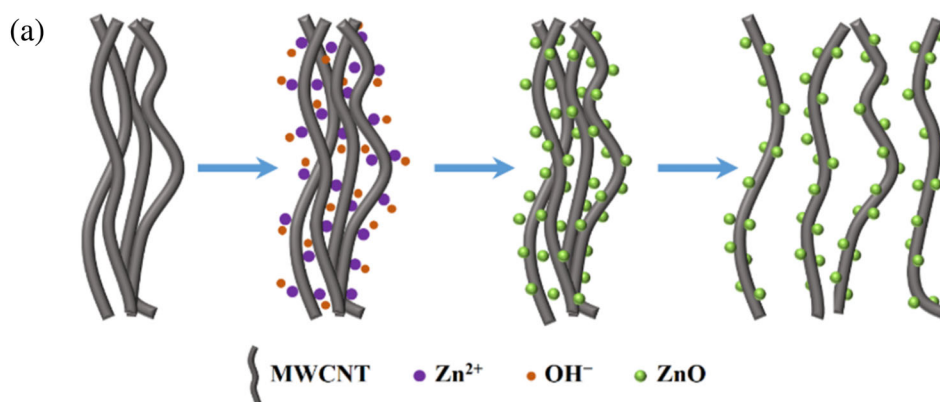
Thermogravimetric analysis (TGA) was carried out on a TGA Q500 (TA Instruments) in an oxygen atmosphere (40 ml/min) from ambient to 800°C at a constant heating rate of 10°C/min. X-ray diffraction (XRD) was performed on a Bruker-AXS D8 Advanced Bragg–Brentano X-ray Powder Diffractometer (Bruker AXS Inc, Madison, WI) with Cu-K $\alpha$  incident radiation ( $\lambda = 1.5418 \text{ \AA}$ ). Transmission electron microscopy (TEM) images were taken on a JEOL JEM-2010 TEM. Dielectric constant and loss tangents were measured on a Novocontrol broadband dielectric spectrometer with a frequency sweep between 10 Hz and 1 MHz. Tensile tests and dynamic mechanical analysis (DMA) were conducted using an RSA-G2 (TA Instruments) with a

tensile fixture. Tensile tests were performed at 25 °C on rectangular samples (3 mm  $\times$  0.01 mm cross section and 5 mm in length) with a crosshead speed of 2 mm/min. DMA was conducted in tensile mode at a strain amplitude of 0.05%, frequency of 1 Hz, and a heating rate of 3°C/min from room temperature to 180°C.

## 3 | RESULTS AND DISCUSSION

### 3.1 | Material characterization

The MWCNT-ZnO was obtained following the procedure described in our earlier report.<sup>4</sup> MWCNT was first treated with oxidizing acids to generate carboxylic groups on the surfaces, which serve as the nucleation sites for ZnO QD growth. Then, as shown in Figure 1, the acid-treated MWCNT was homogeneously dispersed in methanol in the presence of zinc acetate and KOH through ultrasonication. Zn<sup>2+</sup> ions are adsorbed onto the surface of MWCNT due to electrostatic interactions with RCOO<sup>−</sup> ions to form ZnO QD under basic conditions. The carboxylate groups are expected to lower the nucleation energy, which facilitates the growth of ZnO.<sup>39</sup> The reaction time was controlled to obtain a homogeneous coating of a monolayer of ZnO on MWCNT. After decoration with



**FIGURE 1** (a) Schematic of preparation of MWCNT-ZnO hybrids and (b) photo of MWCNT and MWCNT-ZnO suspension in methanol (1 mg/ml). MWCNT, multi-walled carbon nanotube; ZnO, zinc oxide [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

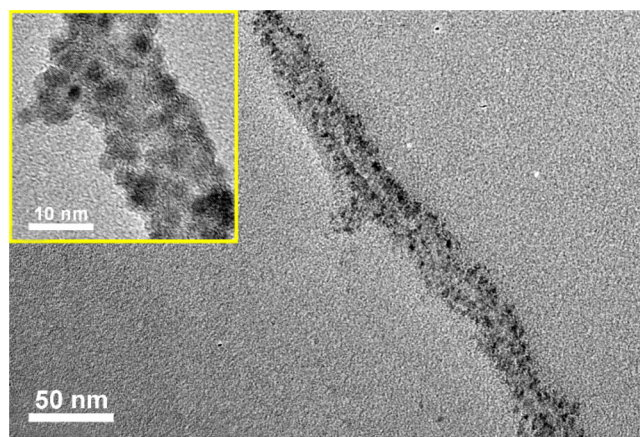
ZnO, the zeta potential changes from  $-21$  mV for MWCNT to  $+38$  mV for MWCNT-ZnO, demonstrating the successful functionalization of MWCNTs and enhanced dispersion stability of MWCNT-ZnO hybrids in methanol.

The structure of MWCNT-ZnO was examined by powder XRD. As shown in Figure S1, all diffraction peaks match well with that of ZnO QD, indicating the formation of highly crystalline ZnO on MWCNTs. The characteristic peaks for MWCNTs can also be observed. The representative TEM images of MWCNT-ZnO are shown in Figure 2. ZnO QD with a uniform size of  $5$  nm are well distributed on the surface of MWCNT.

In addition, the composition of MWCNT-ZnO was investigated using TGA (Figure S2). TGA results show that MWCNT-ZnO decomposes in two stages. The first stage can be attributed to the solvent remaining in the sample, acetate bound to the surface defects of MWCNT-ZnO nanohybrids,<sup>38</sup> and the amorphous carbon. The second stage corresponds to the degradation of the MWCNT. The composition of MWCNT-ZnO can be calculated by analyzing TGA data.<sup>4</sup> The weight fraction of ZnO is 60%. The epoxy nanocomposites containing MWCNT-ZnO exhibit high thermal stability up to  $300^{\circ}\text{C}$ . The TGA results indicate that the decoration of ZnO does not compromise the thermal stability of MWCNT-based epoxy nanocomposites, which is also one of the advantages of using inorganic nanoparticles to modify the interface in polymer nanocomposites.

### 3.2 | Morphological characterization

Epoxy/MWCNT-ZnO thin films were prepared by thermal curing using a hot press. TEM was then utilized to



**FIGURE 2** TEM images of MWCNT-ZnO hybrids. MWCNT, multi-walled carbon nanotube; TEM, transmission electron microscopy; ZnO, zinc oxide [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

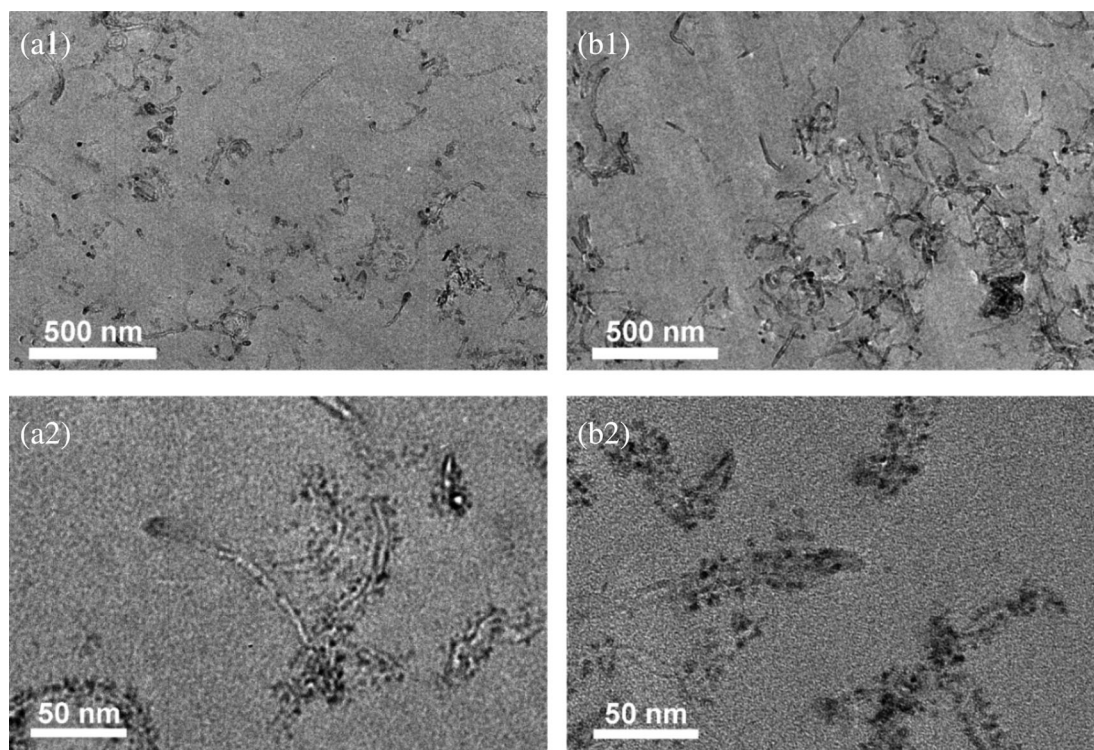
determine the dispersion of MWCNT-ZnO in epoxy. Figure 3 shows the TEM images of epoxy/MWCNT-ZnO with 1.5 and 3.0 wt% loading of MWCNT in epoxy. The nanocomposite containing 1.5 wt% of MWCNT are well dispersed in the epoxy matrix, which shows the effectiveness of ZnO QD decoration on MWCNT in assisting MWCNT dispersion in epoxy. However, by increasing the MWCNT loading to 3 wt%, partial MWCNT aggregation appears to be present. Without the decoration of ZnO nanoparticles on MWCNT, poor dispersion of MWCNT in epoxy is observed (Figure S3).

### 3.3 | Dielectric properties

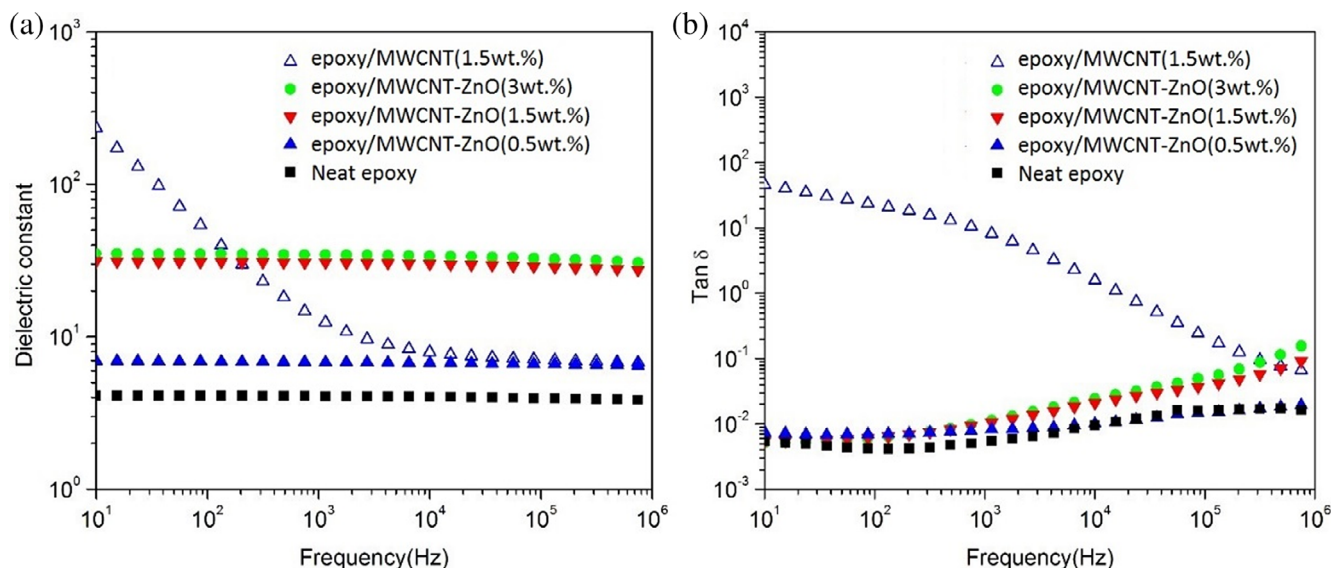
The dielectric properties of epoxy nanocomposites were measured in the frequency range of  $10$ – $10^6$  Hz at room temperature. Figure 4 shows the dependence of dielectric constant and dielectric loss on frequency for epoxy/MWCNT-ZnO nanocomposites at different levels of MWCNT-ZnO content. The data of neat epoxy and epoxy/MWCNT nanocomposite are also included for comparison. It can be observed that for epoxy/MWCNT nanocomposites at 1.5 wt% MWCNT loading, its dielectric constant and dielectric loss are closely related to the frequency and decrease with increasing frequency. As frequency varies from  $10$  Hz to  $1$  MHz, the dielectric constant decreases from 234 to 7, and the dielectric loss decreases from 46 to 0.065. This behavior is characteristic of a percolative polymer composite that exhibits conductive pathways in the matrix. For a percolative system, its dielectric loss mainly consists of the conduction loss (dominant at below  $10^2$  Hz) and the polarization loss of space charges (dominant at  $10^2$ – $10^6$  Hz). Generally, the former leads to more dielectric loss than the latter.<sup>16</sup> A large amount of charges stored by MWCNTs are not stable under high-frequency conditions. Hence, the dielectric loss decreases significantly as the frequency increases because of the conduction relaxation.<sup>40</sup>

Interestingly, the above phenomenon is not observed for the epoxy/MWCNT-ZnO nanocomposites. As the frequency increases from  $10$  Hz to  $1$  MHz, the dielectric constant remains essentially the same, with only 15% reduction. There are two main reasons for this observed phenomenon. First, as MWCNT-ZnO are well dispersed throughout the epoxy matrix, conductive pathways cannot form at either 1.5 wt% or 3.0 wt% of MWCNT loading in epoxy/MWCNT-ZnO systems. Second, considering the quantum confinement and coulomb blocking effect of ZnO QD, ZnO will likely act as an electrically insulating layer between MWCNT.<sup>37</sup> As a result, the transfer of electrons from an individual MWCNT to its adjacent MWCNT will be hindered. Therefore, the substantial





**FIGURE 3** TEM images of epoxy/MWCNT-ZnO (1.5 wt%) at (a1) low magnification and (a2) high magnification, and epoxy/MWCNT-ZnO (3 wt%) at (b1) low magnification and (b2) high magnification. MWCNT, multi-walled carbon nanotube; TEM, transmission electron microscopy; ZnO, zinc oxide



**FIGURE 4** Dielectric spectra of neat epoxy, epoxy/MWCNT (1.5 wt%), and epoxy/MWCNT-ZnO: (a) dielectric constant versus frequency and (b) Tan  $\delta$  versus frequency. MWCNT, multi-walled carbon nanotube; ZnO, zinc oxide [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

increase of dielectric constant is mainly caused by interfacial polarization, that is, a large difference of permittivity between MWCNT-ZnO and epoxy matrix, which is insensitive to frequency. Table 1 lists the specific dielectric

properties of epoxy and epoxy nanocomposites at 10 Hz, 1 kHz, and 1 MHz.

For epoxy nanocomposite containing ZnO decorated MWCNT, the increased capacitance is mainly responsible

Samples	10 Hz		1 kHz		1 MHz	
	K	K loss	K	K loss	K	K loss
Neat epoxy	4.13	0.0054	4.09	0.0055	3.84	0.016
1.5 wt% MWCNT	234.39	45.64	12.42	8.21	6.73	0.065
0.5 wt% MWCNT-ZnO	6.97	0.0074	6.83	0.0084	6.46	0.021
1.5 wt% MWCNT-ZnO	31.28	0.0055	30.69	0.0105	27.98	0.12
3.0 wt% MWCNT-ZnO	35.05	0.0052	34.50	0.0113	30.55	0.21

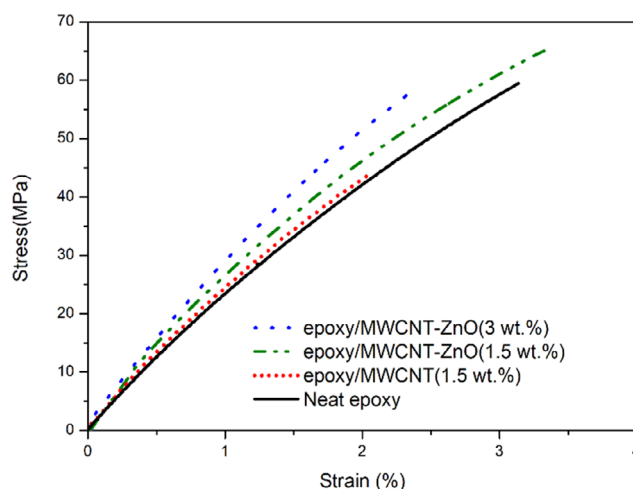
Abbreviations: MWCNT, multi-walled carbon nanotube; ZnO, zinc oxide.

for the increase of dielectric constant. The composite can be simulated by the numbers of equivalent elemental capacitors where MWCNTs are insulated by ZnO QD and epoxy to serve as electrodes when an external electric field is applied to the nanocomposites. As the ZnO decorated MWCNT content increases in the composites, the distance between MWCNT decreases, capacitive coupling increases, which leads to higher permittivity. When the concentration of MWCNT increase from 0.5 to 1.5 wt%, interfacial polarization becomes greatly enhanced. However, when the loading is further increased to 3 wt%, slight agglomeration of MWCNT occurs. In that case, the interface polarization that contributes to the dielectric properties of the system does not increase much. Therefore, the dielectric constant is comparable to that of 1.5 wt% MWCNT system. Moreover, the dielectric loss of epoxy nanocomposites containing 3 wt% hybrids increases as the frequency rises, and value is higher than that of the epoxy system contains 1.5 wt% of MWCNT, corresponding to the increased interface polarization relaxation caused by the slight aggregation of MWCNT-ZnO hybrids. Therefore, the dispersion state of MWCNT plays an essential role in the dielectric behavior of polymer nanocomposites. Merely increasing the content of conductive fillers without considering their dispersity will not only cause a reduction in dielectric constant but also results in enhanced polarization relaxation and a higher dielectric loss.

### 3.4 | Tensile behavior

The tensile engineering stress-strain curves of the neat epoxy and the nanocomposites are presented in Figure 5, and the results are summarized in Table 2. Both at 1.5 wt% of MWCNT loading, the epoxy/MWCNT-ZnO exhibits 20% higher Young's modulus than that of epoxy/MWCNT, which should be due to a better stress transfer between the MWCNT-ZnO and epoxy matrix. Also, since the epoxy/MWCNT-ZnO at 1.5 wt% of MWCNT exhibits the best dispersion in epoxy, this system shows the best

**TABLE 1** Dielectric properties of neat epoxy and epoxy nanocomposites



**FIGURE 5** Tensile engineering stress-strain curves obtained at 25°C for neat epoxy and epoxy nanocomposites [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

tensile performance with significantly improved modulus and tensile strength, while retaining its ductility. The above findings reveal the importance of the nanoparticle dispersion and strong interfacial interaction between nanoparticles and the polymer matrices in enhancing the tensile properties of the nanocomposites.

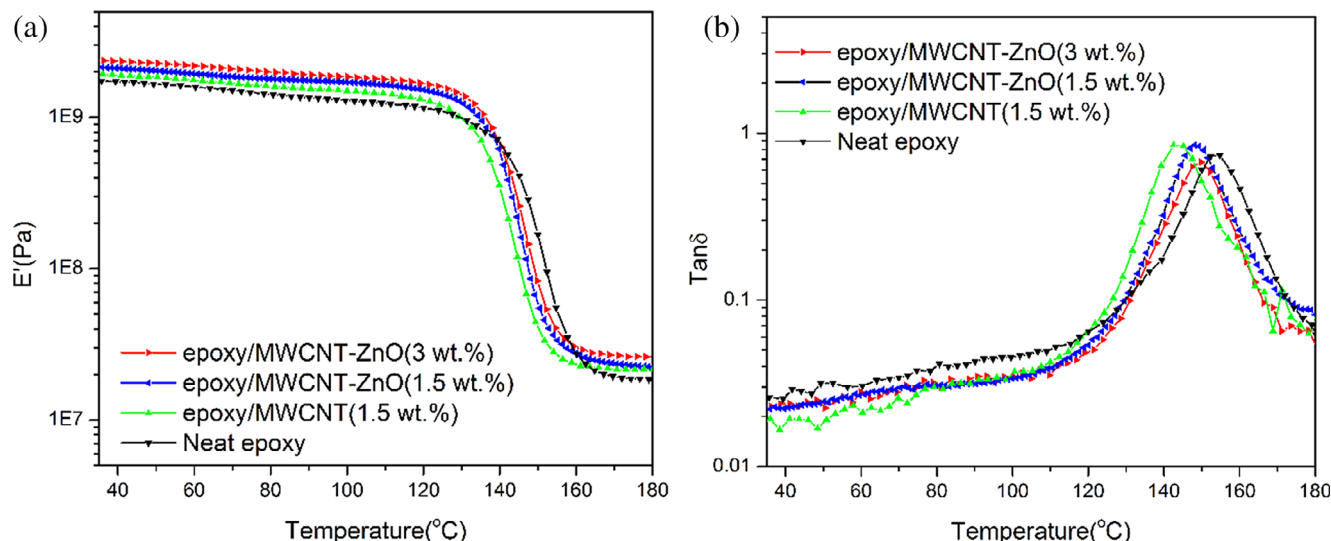
### 3.5 | DMA study

DMA was performed in tensile mode to determine the effect of MWCNT-ZnO on the dynamic mechanical behavior of the epoxy nanocomposites. As shown in Figure 6, all epoxy nanocomposites show an increase in storage moduli, with a slight reduction in  $T_g$ . In addition, the epoxy/MWCNT composite exhibits about 10°C lower in  $T_g$  than neat epoxy, suggesting the composites possesses lower crosslinking density as the curing agent is absorbed by carboxyl groups on the surface of MWCNT during the curing process.<sup>4</sup> With the decoration of ZnO QD on the MWCNT surface, the curing reaction is less hindered and leads to a relatively higher  $T_g$  (Table 3).

**TABLE 2** Tensile properties of neat epoxy and epoxy nanocomposites

Samples	Young's modulus (GPa)	Tensile strength (MPa)	Elongation (%)
Neat epoxy	$3.0 \pm 0.3$	$58 \pm 8$	$3.0 \pm 1.3$
1.5 wt% MWCNT	$3.3 \pm 0.3$	$47 \pm 3$	$2.1 \pm 0.7$
1.5 wt% MWCNT-ZnO	$4.0 \pm 0.4$	$69 \pm 6$	$3.5 \pm 0.8$
3 wt% MWCNT-ZnO	$4.2 \pm 0.3$	$61 \pm 4$	$2.4 \pm 1.1$

Abbreviations: MWCNT, multi-walled carbon nanotube; ZnO, zinc oxide.



**FIGURE 6** DMA plots of neat epoxy, epoxy nanocomposites containing 1.5 wt% MWCNT, 1.5 wt% and 3 wt% MWCNT-ZnO (a) storage modulus versus temperature (b)  $\tan \delta$  versus temperature. DMA, dynamic mechanical analysis; MWCNT, multi-walled carbon nanotube; ZnO, zinc oxide [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

**TABLE 3** Storage modulus and  $T_g$  of neat epoxy and epoxy nanocomposites

Samples	$E'$ at 30°C (GPa)	$T_g$ (°C)
Neat epoxy	1.89	154
1.5 wt% MWCNT	2.01	144
1.5 wt% MWCNT-ZnO	2.35	150
3 wt% MWCNT-ZnO	2.48	149

Abbreviations: MWCNT, multi-walled carbon nanotube; ZnO, zinc oxide.

Based on the experimental findings above, to achieve significant increase in dielectric while maintaining tensile properties of MWCNT-based polymer nanocomposites, two main critical factors should be considered. One is that MWCNTs tend to aggregate in a polymer matrix. The aggregation of MWCNT limits the enhancement in dielectric and mechanical properties of epoxy. Furthermore, poor dispersion of MWCNT can form local conductive pathways, leading to high dielectric loss and low breakdown voltage. The other factor is that

the poor interfacial interaction between the MWCNT and polymer matrix leads to interfacial debonding when mechanically loaded, which hinders stress transfer between the polymer matrix and MWCNT. The utilization of ZnO QDs to decorate MWCNT can improve the dispersion of MWCNT, enhancing the dielectric and tensile properties of the epoxy nanocomposites without compromising  $T_g$  and thermal stability. Therefore, the strategy presented here is applicable in modifying the properties of polymer nanocomposites used in many applications, such as microelectronics, energy storage, and structural components.

Combined with the previous study,<sup>4</sup> our findings show that the surface treatment of MWCNT also plays a vital role in controlling the morphology and properties of epoxy/MWCNT-ZnO. Previously, we found that ZnO QD can grow on the surface of pristine MWCNT (P-MWCNT). But when the P-MWCNT-ZnO were introduced in epoxy, most of the ZnO became detached from MWCNT (Figure S4) due to the weak interaction between ZnO and P-MWCNT. Therefore, without the insulation of ZnO, MWCNT can be in physical contact



with each other and form conductive pathways. On the other hand, if the MWCNT is acid-treated mildly to introduce sufficient amount of  $\text{RCOO}^-$  on MWCNT to form stronger electrostatic interaction with ZnO QDs, no detachment of ZnO is observed. Consequently, the tensile and dielectric properties can be maximized in epoxy/ZnO-MWCNT nanocomposites by the ZnO decoration on MWCNT. The epoxy nanocomposites containing ZnO-decorated MWCNT with mild acid treatment are more suitable for dielectric device applications. Therefore, through the manipulation of the surface chemistry of MWCNT and utilization of ZnO QDs, a variety of multifunctional polymer nanocomposites using the similar concept can be designed for microelectronic and structural applications.

## 4 | CONCLUSION

In this study, high- $k$  epoxy nanocomposites were prepared by using ZnO decorated MWCNT hybrids as a functional nanofiller. The dielectric properties of these nanocomposites show superior frequency independent properties over the range of 10 Hz–1 MHz. With MWCNT-ZnO content at 1.5 wt%, the nanocomposite exhibits dielectric constant value as high as 31 and dielectric loss value as low as 0.01 at 1 kHz. The well-distributed ZnO on MWCNT surface acts as electrical barriers between the nearby MWCNT, which leads to reduced dielectric loss. The decoration of ZnO on MWCNT surface improves the dispersion of MWCNT and form strong bonding with the epoxy matrix effectively. Therefore, compared with the epoxy/MWCNT, the epoxy/MWCNT-ZnO nanocomposites exhibit significant increase in dielectric constant as well as tensile properties, which make them attractive for applications in flexible high- $k$  components, such as flexible energy storage devices.

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Additional supporting information may be found online in the Supporting Information section at the end of this article.

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