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Experimental Study and Piezoresistive Mechanism of Electrostatic Self-Assembly of Carbon Nanotubes– Carbon Black/Epoxy Nanocomposites for Structural Health Monitoring

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Abstract  
Since electrostatic self-assembly carbon nanotubes–carbon black (CNTs–CB) composite fillers have good electrical conductivity and an easy dispersion process, they were incorporated into epoxy matrix to fabricate the CNTs–CB/epoxy nanocomposites. The mechanical properties, electrical conductivity, piezoresistivity, as well as microstructure characterization of the CNTs–CB/epoxy nanocomposites were experimentally investigated. The piezoresistive mechanisms of the CNTs–CB/epoxy nanocomposites were also established based on tunneling conduction and contacting conduction. The significant improvements in compressive yielding strength and elastic modulus of the nanocomposites could be attributed to the stable interfacial interaction. A moderate percolation threshold was obtained at 0.41 vol% for the CNTs–CB/epoxy nanocomposites in comparison with that of the nanocomposites filled with only CNTs or CB particles. In monotonic compressive loading, the CNTs-CB/epoxy nanocomposites exhibited highly sensitive and remarkably non-monotonic piezoresistive responses, which could be divided into three stages and showed dependence on the distinct evolution of hybrid conductive networks under compression. Moreover, the experimental results of cyclic loadings confirmed the stability, repeatability, and recoverability in piezoresistive responses of the nanocomposites. Overall, the highly sensitive and steadily piezoresistive responses demonstrate that the CNTs–CB/epoxy nanocomposites are attractive to be used as compressive strain sensors for structural health monitoring in civil infrastructures.

Introduction  
Civil infrastructures (e.g., buildings, bridges, and highways) often suffer from fatigue load, impact load, and various extreme natural disasters (e.g., earthquakes and hurricanes), which threaten the safety and service life of these infrastructures. Therefore, it is extremely important to install strain sensors at critical locations of structures for accurately diagnosing their real-time status. Currently, commonly used strain sensors for structural health monitoring (SHM) include metal foil resistance strain gauge, piezoelectric ceramic, optical fiber, and piezoresistive strain sensors.

Due to their characteristics of easy fabrication, excellent durability, low cost, and remarkable piezoresistive properties, electrically conductive composites have attracted great interest from researchers in the past few decades. Electrically conductive composites, which are used as piezoresistive strain sensors, are generally consisted of insulating or semiconducting matrix materials (e.g., polymer, cement, and ceramic) and conductive reinforcements (e.g., carbon black, nickel powder, carbon nanotubes, carbon nanofibers, and graphene platelet). The piezoresistive responses of these piezoresistive strain sensors generally stem from the relative change in electrical resistivity when they are subjected to external effects (e.g., stress, strain, and...
temperature. This variation in electrical resistivity is considered to be related to the change in the number of the contact point and the distance between conductive particles. Therefore, they have great potential applications for SHM in civil infrastructures. The cement-based conductive composites have attracted tremendous attention in the applications of piezoresistive strain sensors embedded in concrete due to their similar composition and natural compatibility with concrete structures. Dong et al. investigated the piezoresistive sensitivity and repeatability of cementitious composites reinforced with carbon nanotubes (CNTs) exposed to cyclic compression and impact. The results showed that the cementitious composites filled with CNTs could obtain stable and uniform fractional changes of resistivity during cyclic compression. Sun et al. investigated the piezoresistive cementitious composites with nanographite platelets (NGPs) under different cyclic compressive loading conditions and their capability in measuring dynamic loading rate. They concluded that the cement-based composites with NGPs not only possessed sensitive piezoresistive behavior but also showed stable repeatability to different loading conditions. However, cement-based conductive composites exhibit a strong moisture dependence, thus causing a serious adverse impact on the precision of such sensors, especially when exposed to harsh environments. Moreover, the ultimate compressive strain of confined concrete structures (e.g., steel tube or fiber-reinforced polymers confined concrete structures) is generally up to 1–3%, which is much higher than the strain sensing range of cement-based sensors which is limited to less than 0.5%. Therefore, it is necessary to fabricate piezoresistive strain sensors with excellent waterproofness and broad strain monitoring range for SHM in civil engineering.

Epoxy resins, which have dense and stable cross-linked structure and large deformation range, have been studied as the substitute for cement materials for the matrix in manufacturing piezoresistive strain sensors with favorable mechanical properties and wide strain sensing range, as well as excellent water resistance. Shen et al. explored the changes in electrical resistance of epoxy composites filled with CNTs under compression. It was revealed that the CNTs/epoxy composites showed a significant decrease in electrical resistance under compression. The test results reported by Tian et al. showed that the CNTs/epoxy composites had good piezoresistive properties when exposed to different loading levels. The aforementioned studies have confirmed that epoxy-based composites with low CNTs concentrations could exhibit excellent electrical conductivity and piezoresistivity. Nevertheless, the piezoresistive sensitivity of most nanocomposites with CNTs is usually limited between 0.5 and 5 at the bulk scale, especially under compression. In addition, the composites filled with CNTs are prone to exhibit poor repeatability under cyclic loading due to the large aspect ratio. Research has shown that carbon black (CB) nanoparticles could also endow composites with good electrical and piezoresistive properties. For instance, Wichmann et al. demonstrated that the epoxy-based nanocomposites with CB exhibited pronounced piezoresistive responses with higher strain sensitivity and good repeatability. However, the percolation threshold of nanocomposites filled with CB is generally much higher than that in nanocomposites containing CNTs. This leads to the large filler dosage of CB in the matrix, which may introduce local defects in the nanocomposites and significantly impair their mechanical properties. Synthetically considering these factors, blending CNTs and CB conductive nanoparticles into a polymer matrix might be a new alteration to balance the requirements of high strain sensitivity, good constancy, and appropriate filler concentration.
At present, there are many researches on conducting nanocomposites by simply mixing CNTs and CB nanoparticles into the polymer matrix\textsuperscript{41, 44, 47–49}. The researchers verified that the combination of CNTs and CB conductive fillers into a polymer matrix could effectively improve the electrical conductivity and piezoresistivity of nanocomposites via constructing the distinct conductive network in the matrix on account of the synergistic effect. However, the characteristics of easy agglomeration make it difficult to realize the uniform dispersion of CNTs and CB nanoparticles into a polymer matrix. Generally, ultrasonic treatment is often used to assist in dispersing CNTs and CB hybrid nanoparticles during the simple mix process\textsuperscript{5, 8, 10–12}. This dispersion process with ultrasonic treatment is usually sophisticated and time-consuming, which will increase the difficulty in fabricating nanocomposites. What is more, the synergistic effect between CNTs and CB relies heavily on their dispersion states in the polymer matrix\textsuperscript{49}. Electrostatic self-assembly CNTs–CB composite fillers with special grape clusters structures can be prefabricated before they are incorporated into the polymer matrix. Specifically, the surfaces of CNTs and CB are modified by cationic and anionic surfactants, respectively; and then, the CB particles are attached to the surrounding of CNTs by electrostatic force\textsuperscript{50}. This assembly process enables the CNTs–CB composite fillers to have a larger volume compared to the individual CNTs or CB particles, which not only simplifies the dispersion process without additional ultrasonic treatment, but also ensures the realization of a good synergistic effect between the hybrid nanoparticles\textsuperscript{4}. A few studies showed that the electrostatic self-assembly CNTs–CB composite fillers-reinforced silicon rubber (SR) matrix\textsuperscript{50} and cement matrix\textsuperscript{8} could achieve not only the enhancement in electrical conductivity and piezoresistivity with low filler concentration, but also the simplification in the filler dispersion method. However, there is no investigation of the effects of electrostatic self-assembly CNTs–CB composite fillers on the piezoresistive behavior of epoxy-based nanocomposites under compression.

Simultaneously, the epoxy resin possesses the features of easy fabrication, good creep resistance, excellent water resistance, and broad strain sensing range, compared to silicon rubber matrix and cement matrix\textsuperscript{8, 19}. Therefore, it is very meaningful to investigate the various properties (i.e., mechanical properties, electrical conductivity, and piezoresistivity) of epoxy-based nanocomposites integrated with electrostatic self-assembly CNTs–CB composite fillers, and to explore their potential applications as a piezoresistive strain sensor.

The objectives of this study include two aspects: (1) analyzing the influence of the concentrations of electrostatic self-assembly CNTs–CB composite fillers on the mechanical properties, electrical conductivity, and piezoresistivity of epoxy-based nanocomposites under compression; and (2) verifying the piezoresistive mechanisms of electrostatic self-assembly CNTs–CB composite fillers-reinforced epoxy-based nanocomposites subjected to different levels of compressive load. To achieve these goals, epoxy-based nanocomposites filled with different volume percentages of electrostatic self-assembly CNTs–CB composite fillers ranging from 0.5 to 2.0 vol% were fabricated by a simple and facile mechanical stirring method. The mechanical and electrical properties of CNTs–CB/epoxy nanocomposites embedded with different electrostatic self-assembly CNTs–CB composite fillers' concentrations were investigated. The mechanisms of the piezoresistive response of the CNTs–CB/epoxy nanocomposites corresponding to different strain levels were then theoretically demonstrated. Furthermore, the piezoresistive responses of the nanocomposites at different loading conditions were experimentally studied to reveal their stability, repeatability, and feasible strain sensing range.
Experimental program

Raw materials and specimen fabrications

A commercially available epoxy resin (produced by Tianjin Swancor Wind Power Materials Co., Ltd., China), which is composed of base component (model 2511-1A) and hardener (model 2511-1BS) with a weight ratio of 100:30, was selected for the fabrication of the specimens in this study, because it exhibits the characteristics of low viscosity, moderate gel time, and good infiltration to carbon nanoparticles. The electrostatic self-assembly CNTs–CB composite fillers were acquired from the Chengdu Institute of Organic Chemistry Co., Ltd, China. Their resistivity is less than 0.01 Ω·cm and their specific surface area is 540–560 m²/g. They are composed of CNTs and CB with a weight ratio of 40:60. The average diameter of CB particles is 35 nm, and the specific surface area is 934 m²/g. The primary outside diameter of CNTs particles is 10–30 nm, and the average length is 10–30 μm. Figure 1 shows their specific grape clusters structure microscopically. Stainless steel mesh was used in specimens as the conductive electrodes with a size of 20 mm × 30 mm.

![Figure 1 SEM photographs of CNTs-CB composite fillers: a 50000X and b 100000X](image)

A simple and facile mechanical stirring method without additional ultrasonic treatments was used to fabricate the epoxy-based nanocomposites containing different concentrations of electrostatic self-assembly CNTs–CB composite fillers in this study. The detailed fabrication and molding procedures of CNTs–CB/epoxy nanocomposites are exhibited schematically in Fig. 2. Firstly, all different volume fractions of CNTs–CB composite fillers were weighed by a high-precision electronic balance and dried in a vacuum oven at 60 °C for 30 min before the dispersion process to eliminate possible absorbed moisture during storage. Then, the specific quantity of dry CNTs–CB composite fillers was incorporated into the preheated epoxy base component, which was heated in a water bath at 60 °C to further lower the viscosity. The mixture was first stirred at low speed (500 rpm) at 60 °C for 5 min and then at high speed (2000 rpm) at 60 °C for 25 min. Subsequently, the uniformly dispersed mixture was placed in a vacuum oven at 60 °C for 30 min to eliminate the entrapped air bubbles. After that, the mixture was cooled to room temperature and the required quantity of the hardener was added into the mixture with a gentle stirring process at room temperature to reduce the introduction of extra air bubbles. Next, the CNTs–CB/epoxy mixture was vacuumed at room temperature again to remove entrapped air bubbles and then poured into the silicone rubber molds with the dimension of 20 mm × 20 mm × 40 mm, which was pre-coated with silicone oil release agent (model BD-TM001, produced by Xi’an Beiduo Environmental Protection Technology Co., Ltd., China) to easily remove the specimens out of the molds after they were cured. Two stainless steel mesh electrodes with 10 mm of separation were embedded in the molds in advance. The specimens for mechanical property tests had no embedded electrodes. The casted specimens were first cured at room temperature for 24 h and then cured at 60 °C for another 24 h. It should be noted that after adding the hardener component, the operations such as the
stirring and the vacuuming should be performed quickly to prevent the epoxy resin from the curing before the pouring, which could tremendously damage the mechanical properties of the nanocomposites. For the epoxy-based nanocomposites, the selected CNTs-CB composite fillers concentrations in volume were fixed at 0.5 vol%, 0.75 vol%, 1.0 vol%, 1.25 vol%, 1.5 vol%, and 2.0 vol%, respectively.

Figure 2 Fabrication and molding procedures of CNTs–CB/epoxy nanocomposite specimens

Measurements
Microstructure characterization
The morphology and distribution of electrostatic self-assembly CNTs–CB composite fillers in the epoxy matrix were observed by a field emission scanning electron microscopy (FESEM). The samples used for inspection were cut from the fractured CNTs–CB/epoxy nanocomposites after their mechanical tests. Before the FESEM observation, a thin layer of platinum was sputtered on the samples' surface to enhance the electroconductibility of the samples for better imaging.

Mechanical measurement
The uniaxial compression test was conducted using an electronic universal testing machine (model WDE-200E, produced by Jinan Gold Testing Machines Inc., China). A displacement-controlled loading method was applied to the specimens with a fixed loading rate of 0.4 mm/min. The axial compression strain was measured by a pair of strain gauges symmetrically bonded to the center of both sides of the specimen along the length direction, as shown in Fig. 3. A static strain acquisition system (model DH3820, produced by Donghua Testing Technology Co., Ltd., China) was used to record real-time compressive strain data, and the sampling frequency was fixed at 1 Hz during the entire test process. Three specimens for each composite filler concentration were tested.

Figure 3 Arrangement of strain gauges on the specimen
Electrical measurement
Three specimens of each type of CNTs–CB/epoxy nanocomposites with different electrostatic self-assembly CNTs-CB concentrations (0 vol%, 0.5 vol%, 0.75 vol%, 1.0 vol%, 1.25 vol%, 1.5 vol%, and 2.0 vol%) were tested for the electrical properties. Compared with four-electrode measurement method, two-electrode measurement method has the advantages of easy operation and stable electrical signal\(^{17,18}\). Therefore, the two-electrode measurement method was adopted in the study to obtain the electrical resistance. A Keithley 2100 digital multimeter was used to measure direct current (DC) electrical resistance. The volumetric electrical resistivity of specimens, \(\rho\), was calculated by\(^{20}\):

\[\rho = \frac{R \times S}{L}\]

(1)

where \(R\), \(S\), and \(L\) are the bulk resistance of specimens, the cross-sectional area of specimens, and the distance between the two embedded electrodes, respectively.

Piezoresistive measurement
A synthetical data collection system, combining WDE-200E electronic universal testing machine, Keithley 2100 digital multimeter, and DH3820 static strain acquisition system, was used for inspecting the piezoresistive properties. The three devices were all connected with a computer to synchronously record the real-time force data, bulk electrical resistance, and compressive strain during the compression, respectively. It should be noted that the piezoresistivity is highly dependent on the electrical conductivity of the nanocomposites\(^{41,54}\). The nanocomposites containing 0.5 vol% and 0.75 vol% electrostatic self-assembly CNTs–CB composite fillers were unable to output stable electrical signals due to their poor electrical conductivity. Moreover, the nanocomposites containing 2.0 vol% electrostatic self-assembled CNTs–CB composite fillers lead to obvious agglomeration of the fillers due to excessive filler concentration. Therefore, its economy, long-term working life, and strain sensitivity are inevitably reduced. Thus, the nanocomposites with volume fractions of CNTs–CB composite fillers of 1.0 vol%, 1.25 vol%, and 1.5 vol%, respectively, were selected to investigate the piezoresistive responses of CNTs–CB/epoxy nanocomposites under monotonic and cyclic loading. As shown in Fig. 4, two different cyclic compressive loading modes were adopted including a constant amplitude cyclic and an incremental amplitude cyclic loading. During the constant amplitude cyclic loading, every specimen was subjected to six loading–unloading cycles with the maximum compressive stress level at 20 MPa, which kept the nanocomposites in the linear elastic stage. A stepwise increase compressive loading was applied from 5 to 45 MPa with the incremental amplitude of 5 MPa, and every level of compressive stress was conducted for three cycles. Both monotonic and cyclic loadings were performed at a constant speed of 0.4 mm/min. To obtain the stable conductive network before the piezoresistive properties testing, each specimen was pre-compressed by applying several cyclic compressive loading under a low-stress level (5 MPa). The test setup is shown in Fig. 5.

Figure 4 Cyclic compressive loading modes: a constant amplitude; b incremental amplitude
The fractional change in volumetric resistivity (FCR), $f$ (unit:%), was approximately determined according to Eq. (2) by ignoring the small deformation between two electrodes under small deformation of the entire specimen 20.

$$ f = \frac{R - R_0}{R_0} = \frac{\Delta R}{R_0} $$

(2)

where $R$ (unit:Ω) is the isochronous electrical resistance during the compression; $R_0$ (unit:Ω) is the initial electrical resistance of the specimens without compression; and $\Delta R$ (unit:Ω) is the relative change in bulk electrical resistance during the compression.

Results and discussions

Morphology

Figures 6 and 7 exhibit the morphology and distribution of electrostatic self-assembly CNTs-CB composite fillers in the epoxy matrix. As shown in Fig. 6, the CNTs–CB composite fillers were completely incorporated in the epoxy matrix and could remain their original grape clusters structure in the matrix, which was essential to realize the synergetic effect. In addition, Fig. 7 demonstrates the distribution of CNTs–CB composite fillers with different filler volume fractions. From the SEM images of the fracture surfaces from the failed specimens in mechanical tests, it can be concluded that the CNTs–CB composite fillers could achieve a relatively well-distributed state in the epoxy matrix via the facile and simple direct mixing method used in this study due to their special grape clusters structure. However, the agglomeration of CNTs began to occur when the filler concentration reached 2.0 vol% as shown in Fig. 7f. This behavior is believed to associate with the relatively high viscosity of the mixture with high CNTs–CB composite fillers concentration. The uniform distribution of the conductive fillers within a certain concentration range contributes to the favorable establishment of conductive networks in the epoxy matrix, which conduces to the considerable enhancements in the mechanical properties, electrical properties as well as piezoresistivity of the epoxy-based nanocomposites as described in the following sections.
Mechanical properties

The uniaxial compressive loading was conducted to investigate the mechanical properties of the CNTs–CB/epoxy nanocomposites. Figure 8 illustrates the compressive mechanical properties of nanocomposites with different concentrations of electrostatic self-assembly CNTs-CB composite fillers. Figure 8a shows the representative compressive stress–strain curves for the pure epoxy and its nanocomposites, in which all typical stress–strain curves exhibited a similar initial linear variation trend and then followed with a large scale of plasticity\(^{14}\). Additionally, Fig. 8a shows that the linear elastic limits of CNTs–CB/epoxy nanocomposites were around 35–40 MPa. It is well recognized that the nanocomposites should stay within the range of linear elastic limit when they are used as piezoresistive strain sensors to avoid permanent deformation that has adverse effects on the precision of the compressive sensors\(^{55}\). Figure 8b and c indicate that the introduction of CNTs–CB composite fillers into epoxy resin had significant improvements in the mechanical properties of the nanocomposites in comparison with those of pure epoxy resin. Compared with that of pure epoxy resin (i.e., compressive yield strength: 66.2 MPa), the compressive yield strengths of the nanocomposites filled with 0.5 vol%, 1.0 vol%, 1.25 vol%, 1.5 vol%, and 2.0 vol% CNTs-CB composite fillers were increased by 11.5%, 19.2%, 23.1%, 25.5%, and 30.8%, respectively. At the same time, the corresponding elastic moduli of the nanocomposites filled with 0.5 vol%, 1.0 vol%, 1.25 vol%, 1.5 vol%, and 2.0 vol% CNTs–CB composite fillers were also increased by 6.1%, 15.7%, 18.3%, 25.4%, and 30.0%, respectively, compared with that of pure epoxy resin (i.e., elastic modulus: 2.46 GPa).

Figure 8 Compressive mechanical properties of CNTs-CB/epoxy nanocomposites with different concentrations of CNTs–CB composite fillers: a typical stress–strain curves; b compressive yield strength; and c elastic modulus
These improvements in mechanical properties could be ascribed to the reinforcing effect of the CNTs–CB composite fillers. Generally, favorable interfacial interaction is the foundation of this reinforcing effect on mechanical properties. When the CNTs–CB composite fillers are evenly distributed into the epoxy, excellent interfacial interaction will be generated between the CNTs–CB composite fillers and the epoxy matrix, thereby effectively transferring stresses from the epoxy matrix to the composite fillers. Thus, CNTs–CB composite fillers possessing excellent mechanical properties can support a portion of the compressive loading, and also significantly enhance the compressive yield strength and the elastic modulus of the composites. When the CNTs–CB composite fillers concentration is increased, the number of solid interfaces used for stress transfer will also be increased, resulting in better stress transfer efficiency and thus sustaining more stress. Meanwhile, due to the constraint of CNTs–CB composite fillers (e.g., bridging effect), the lateral deformation of epoxy is restricted and the extension of microcracks in the matrix is subsequently limited, thus improving the compressive yield strength and elastic modulus of nanocomposites. The fractured CNTs under high-stress level and the bridging effect are shown in Fig. 9.

Figure 9 SEM images of CNTs–CB/epoxy nanocomposites: fractured CNTs in a and b; and bridging effect in c and d
Electrically conductive properties

The relationship between the DC electrical resistivity and the concentration of electrostatic self-assembly CNTs–CB composite fillers without applying external compressive stress is shown in Fig. 10. It can be seen that the concentration of CNTs–CB composite fillers had a prominent effect on the DC electrical resistivity. It is obvious that increasing the concentration of CNTs–CB composite fillers significantly decreased the DC electrical resistivity of CNTs–CB/epoxy nanocomposites. The electrical resistivity of the specimens with 1.0 vol% CNTs–CB composite fillers was around 6 orders of magnitude lower than that of pure epoxy resin. This sudden decrease in electrical resistivity implies the occurrence of a significant percolation phenomenon (i.e., the conductive paths begin to form in the epoxy matrix).

Figure 10 Electrical resistivity of CNTs–CB/epoxy nanocomposites as a function of CNTs–CB concentration. (The inset is the plot of the logarithm of conductivity vs. the logarithm of reduced volume fraction.)

To better understand the percolation characteristic of CNTs–CB/epoxy nanocomposites, the general effective media (GEM) theory was applied to estimate their percolation threshold and critical coefficient:

$$\rho = \rho_0 (\varphi - \varphi_c)^{-t}$$  \hspace{1cm} (3)

where $\rho_0$ is a constant value; $\varphi$ is the volume fraction of CNTs–CB composite fillers; $\varphi_c$ is the critical volume fraction of CNTs–CB composite fillers (i.e., percolation threshold); and $t$ is the critical coefficient (which is used to judge the dimensionality of the conductive networks in the nanocomposites). The resistance of conductive composite fillers is negligible compared to the extremely high resistance of the epoxy matrix. The values of $\varphi_c$ and $t$ of nanocomposites were obtained by fitting the experimental data. And the estimated results according to GEM were shown in the insert in Fig. 10. According to the fitting result, the percolation threshold value of CNTs–CB/epoxy nanocomposites was estimated as 0.41 vol%, which was between the value of nanocomposites comprising only CNTs and that of nanocomposites with only CB particles. Based on the researches on epoxy-based nanocomposites containing only CNTs or CB, the filler dimension has significant effects on the electrical percolation threshold. For example, Cao et al. demonstrated that the ultralow percolation threshold of 0.0125 vol% for CNTs/epoxy composites was obtained due to their
large aspect ratio. However, Buketov et al.\textsuperscript{43} found that the composites filled with CB particles possessed a much higher percolation threshold around 8 wt\%. Thus, the moderate percolation threshold of nanocomposites containing CNTs–CB composite fillers in this study indicates that blending CNTs and CB particles into polymer matrix could well tune the percolation threshold of nanocomposites. This phenomenon could be ascribed to the unique hybrid conductive networks due to the unique synergistic effect between CNTs and CB particles. Chen et al.\textsuperscript{41} also confirmed that introducing a certain amount of CNTs into CB/isoprene rubber composite could significantly reduce the percolation threshold due to the construction of hybrid CNTs–CB networks. Moreover, the value of $t$ fitted with experimental data according to GEM was estimated as 2.88, which was higher than the theoretical numerical value for a three-dimensional random network $t \approx 2$\textsuperscript{42}. This much higher $t$ value may be attributed to a three-dimensional electrically conductive network accompanied by a relatively wide distribution of inner conductive particles distance (i.e., tunneling resistance) around the percolation thresholds\textsuperscript{42,47,56–58}. Such hybrid three-dimensional conductive networks were derived from the numerous inner gaps between CNTs and CB particles, e.g., CNTs–CNTs, CNTs–CB, CB–CB, as shown in Fig. 11.

Figure 11 Three different potential inner gaps in CNTs–CB composite fillers in the nanocomposites

Generally, tunneling conduction and contacting conduction are the mainly conductive mechanisms in the aforementioned hybrid conductive networks. Furthermore, the sophisticated inner gaps (Fig. 11) generated different electron conduction modes in the epoxy matrix, including short-range conduction between CB and remote-range conduction between CNTs\textsuperscript{44,59}. These multiple modes of electron conduction produce the complex hybrid conductive networks and the moderate percolation threshold based on the synergistic effect between CNTs and CB particles. As shown in Fig. 7, adding more CNTs–CB composite fillers led to a more noticeable reduction in the inner distance between fillers. In this case, the composite fillers gradually transform from the isolated state to partial mutual overlap with the increase in the filler concentration\textsuperscript{13}. Thus, the dominant conduction mechanism transforms from tunneling conduction to contacting conduction with the increase in the CNTs–CB concentrations. This is very beneficial for the construction of continuous and abundant conductive networks, increasing the electrical conductivity of nanocomposites.

**Piezoresistive properties**

*Piezoresistive properties under monotonic loading*

To investigate the piezoresistive properties of CNTs–CB/epoxy nanocomposites and explore their potential application as piezoresistive sensors, both strain and electrical resistivity variations of specimens were tested simultaneously when they were subjected to monotonic compressive loading. Figure 12 shows the changes of FCR of specimens filled with 1.0 vol\%, 1.25 vol\%, and 1.5 vol\%
electrostatic self-assembly CNTs–CB composite fillers, respectively, under compression. The FCR values of CNTs–CB/epoxy nanocomposites changed synchronously with the external compressive strain, manifesting a timely resistance response. In addition, the FCR–strain curves of all nanocomposites filled with different CNTs–CB composite filler concentrations had obvious non-monotonic changes during the entire compression process. The FCR vs. strain curves of CNTs–CB/epoxy nanocomposites started with a linear decline initially, followed by an abrupt decline, and then reversed the trend to increase. This phenomenon is highly dependent on the evolution of the conductive networks in the matrix, which is related to the construction and breakdown of electrically conductive pathways with the increasing compressive strain, causing the decrease and increase in the electrical resistivity. The FCR–strain curves of CNTs-CB/epoxy nanocomposites can be segmented into three different stages: (1) linear elastic stage, (2) viscoelastic stage, and (3) plastic stage. In the first two stages, all FCR values decreased linearly with the increasing compressive strain until the electrical resistivity plateau was reached as shown in Fig. 12. This result is related to the decreasing tunneling resistivity and the optimization of the conductive network. The transition point denoted as $\varepsilon_2$ in Fig. 12 is closely corresponding to the yielding points of nanocomposites in stress–strain curves according to the 0.2% offset rule. In the third stage (i.e., beyond the yielding points), the FCR values exhibited an increasing trend until the nanocomposites failed. This is mainly due to the generation of microcracks and the destruction of conductive pathways, which leads to the violent separation between CNTs–CB composite fillers and then hinders electron conduction. In summary, the evolution of the conductive network structures governs the piezoresistivity of CNTs–CB/epoxy nanocomposites.

Figure 12 FCR and compressive stress vs. strain for nanocomposites loaded with a 1.0 vol%; b 1.25 vol%; and c 1.5 vol% of CNTs–CB composite fillers

The gauge factor (GF) of strain sensors, which is defined as the ratio of $\Delta R/R_0$ to concurrent compressive strain $\varepsilon$ (i.e., FCR/$\varepsilon$), is generally used to evaluate the strain sensitivity of nanocomposites and provides a more accurate description of the piezoresistive response. The linear regression analysis results for the first two stages of FCR–strain curves are listed in Table 1. The GFs in stage I of the CNTs–CB/epoxy nanocomposites with the filler concentrations of 1.0 vol%, 1.25 vol%, and 1.5 vol% were calculated as 6.94, 4.66, and 4.60, respectively, and the GF values in stage II were fitted as 30.10, 26.39, and 18.53, respectively. A larger compressive strain amplitude led to a higher strain sensitivity. This increasing strain sensitivity could be ascribed to that denser conductive networks were established when specimens were exposed to higher strain levels, leading to more decrease in the electrical resistivity. In addition, the GF values in Table 1 for CNTs–CB/epoxy nanocomposites are markedly higher than those in many other researches studying thermosetting matrix
nanocomposites\textsuperscript{36–39}. For example, Ku-Herrera et al.\textsuperscript{38} investigated the piezoresistivity of the CNTs/vinyl ester composites filled with 0.3, 0.5, and 1.0 wt\% CNTs in compression and found that the strain sensitivities of them before matrix yielding were 0.91, 2.39, and 4.55, respectively, which are much lower than those in CNTs–CB/epoxy nanocomposites (i.e., 30.10 for the nanocomposites with 1.0 vol\% CNTs–CB composite fillers before yielding). This distinct phenomenon indicates that the introduction of CB particles makes the conductive network more susceptible to external forces, resulting in higher strain sensitivities. However, these GF values are lower than those of thermoplastic\textsuperscript{12, 54} and elastomeric matrix composites\textsuperscript{41, 60}. This manifests that the strain sensitivity of the nanocomposites is also relevant to the flexibility of matrix materials\textsuperscript{12}. In addition, the research of Yang et al.\textsuperscript{61} showed that when the concentration of graphite nanoplatelets(GNP) reached 50 wt\%, the GF value for strain sensing of freestanding CNT/GNP buckypapers was up to 8.18, and the corresponding $R^2$ was 0.999. Another research of Yang et al.\textsuperscript{62} showed that hybrid CNT/Graphene (GN) film sensors were highly sensitive to the initiation and evolution of shear damages. This manifested that the film sensors fabricated by composite fillers free from flexible substrates might have unique advantages in the sensitivity and the linearity. Furthermore, Table 1 shows that the GF values also show high dependency on the CNTs–CB composite fillers concentrations. There is a gradually decreasing strain sensitivity for the nanocomposites with the increase in the CNTs–CB composite filler concentration. The reduction in the inter-particle gaps and the increase in the conductive network compactness account for this decreasing sensitivity. Considering that the linearity of the FCR–strain curves of the nanocomposites with three different filler concentrations in this paper is not much different, the nanocomposite with 1.0 vol\% CNTs–CB composite fillers, due to its high strain sensitivity and the minimum filler concentration, show greater potential in making compressive strain sensor. Other researches\textsuperscript{40, 51} showed that higher electrical resistivity manifests higher strain sensitivity at lower conductive filler concentration, and the dominance of tunneling effects could be used to explain this phenomenon. The CNTs–CB concentrations of specimens in this research were beyond the percolation threshold. Therefore, the dominance of tunneling effects on piezoresistive responses gradually reduced and the contact conduction became dominant, leading to a more compact conductive network and lower strain sensitivity.

### Table 1 Gauge factors of CNTs–CB/epoxy nanocomposites

<table>
<thead>
<tr>
<th>CNTs–CB concentrations (vol%)</th>
<th>Gauge factors in stage I (GF\textsubscript{1})</th>
<th>Coefficient of determination ($R^2$)</th>
<th>Gauge factors in stage II (GF\textsubscript{2})</th>
<th>Coefficient of determination ($R^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.0</td>
<td>6.94</td>
<td>0.993</td>
<td>30.10</td>
<td>0.994</td>
</tr>
<tr>
<td>1.25</td>
<td>4.66</td>
<td>0.994</td>
<td>26.39</td>
<td>0.996</td>
</tr>
<tr>
<td>1.5</td>
<td>4.60</td>
<td>0.995</td>
<td>18.53</td>
<td>0.996</td>
</tr>
</tbody>
</table>

**Piezoresistive mechanisms under monotonic loading**

According to the previous discussion, the evolution of the conductive networks in the matrix may result in different piezoresistive mechanisms under different compressive strain amplitudes. The evolution of conductive networks in schematic form and the expansion of microcracks in SEM images of nanocomposite with 1.0 vol\% electrostatic self-assembly CNTs–CB composite fillers are illustrated in Figs. 13 and 14, respectively.
As shown in Fig. 13a, the CNTs-CB composite fillers were uniformly distributed and there was no damage in the matrix in the original state without applied compressive strain. In the initial state, the distribution of conductive junctions (red circle in Fig. 13a) was limited, resulting in less conductive paths and showing high electrical resistivity. As the compressive strain was in stage I (i.e., linear elastic stage in Fig. 12), the nanocomposites generated only a minor volumetric deformation in the direction of compression, and the morphology of conductive networks could hold their original structure form. When the larger compressive strain was applied to the nanocomposites, the inter-particle gaps decreased and the partial contacts between neighboring fillers increased, leading to the reduction in electrical resistivity. Under this circumstance, the number of conductive junctions increased (Fig. 13b). Therefore, more conductive paths could be formed and the electrical resistivity gradually decreased. In this stage, tunneling conduction plays a dominant role and accounts for the decreasing electrical resistivity. Moreover, Fig. 14a shows that the structure of the epoxy matrix also could remain unchanged without any microcracks in the linear elastic stage. This phenomenon indicates that such a low strain level corresponding to the linear elastic deformation would not damage the matrix structure. Therefore, more conductive networks could be generated in this stage, mainly owing to the reduction in the distance between neighboring CNTs–CB composite fillers.

For the larger compressive strains in stage II (i.e., viscoelastic stage in Fig. 12), more conductive paths were generated, and thus, the electrical resistivity of nanocomposites decreased along with the compression. Therefore, a higher strain sensitivity was obtained. In addition, under larger strain amplitudes, the deformation of nanocomposites gradually increased and gave rise to the generation of damage in the epoxy matrix. These microcracks in the matrix shown in SEM images (Fig. 14b) demonstrate that the higher strain level caused the formation of localized microcracks. However, the number of microcracks was still limited due to the excellent mechanical properties of the epoxy matrix. Therefore, the construction of new conductive paths is much faster than the destruction within a certain high strain range, and the decline in electrical resistivity still dominates. According to "Piezoresistive properties under monotonic loading" section, all FCR versus strain curves had an abrupt
decreasing trend and the corresponding GF values showed a significant increase in this stage. The
dramatic improvements in strain sensitivity are considered to be associated with not only the further
reduction in the adjacent gaps between composite fillers, but also the rearrangements of CNTs–CB
nanoparticles along with the deformation of the epoxy matrix under larger strains. On the one hand,
the gaps between adjacent CNTs-CB composite fillers further decrease and the number of conductive
junctions also continuously increases (Fig. 13c), which significantly reduces the electrical resistivity of
the nanocomposites. On the other hand, the spherical CB aggregates attached around CNTs in the
CNTs–CB composite fillers are more prone to move and slip along with the motion of epoxy molecular
segments. Thus, the gaps between CNTs are effectively bridged and the mutual contacts are further
strengthened, contributing to the formation of new conductive paths and the enhancement of existing
conductive networks in the nanocomposites47, 49. Under these circumstances, contact conduction
gradually becomes dominant. Therefore, the aforementioned two superposed positive effects
generate denser and more thorough conductive networks, resulting in the lower electrical resistivity
and the higher strain sensitivity of nanocomposites.

When the strain entered into stage III (i.e., plastic stage in Fig. 12), all FCR curves firstly experienced a
platform and then followed by an increase. This platform could be attributed to the coexistence of the
establishment and destruction of conductive networks. During this stage, the nanocomposites were
further compressed, leading to the occurrence and progress of large deformation in nanocomposites
as well as the rapid accumulation and development of microcracks in the matrix (Fig. 13c). These
expanding microcracks intensify the enlargement of gaps and the reduction in contacts between CNTs–
CB composite fillers, which excessively destroy the electrically conductive paths and seriously block the
transmission of electrons. Given this dominance, the electrical resistivities of nanocomposites
continuously increase. Thus, the establishment and destruction of conductive paths gradually begin to
reach a balance along with the increase in the strain amplitude. With further increasing the applied
strain, this balance is gradually destroyed, and the destructive process of conductive networks
increasingly plays a key role. In addition, the Poisson’s effect also could enhance the transverse
expansion along with the compression deformation of nanocomposites, leading to the transformation
from microcracks to macrocracks and the further increase in electrical resistivity. Hence, both of these
two factors contribute to the increase in the electrical resistivity of nanocomposites in stage III.

Piezoresistive properties under constant amplitude cyclic loading
The aforementioned investigations have shown that the CNTs–CB/epoxy nanocomposites had obvious
piezoresistive responses and broad strain sensing ranges. In addition to the strain sensitivity, the
stability and repeatability of the nanocomposites also play essential roles in the application of
piezoresistivity strain sensors47. To study their stability and repeatability, the CNTs–CB/epoxy
nanocomposites were subjected to six constant amplitude loading–unloading compressive cycles
within the linear elastic stage of the nanocomposites to verify their remarkable piezoresistivity in real
service conditions.

The values of stress, strain, and FCR as a function of time during the loading–unloading cyclic process
are shown in Fig. 15. It is obvious that the FCR value closely followed the variation of the external
compressive stress/strain, indicating the instantaneous piezoresistive response. When the specimen
was compressed, the movement of the molecular network dragged the bound CNTs–CB composite
fillers through a strong interface bond, resulting in the decline in the inter-particle gaps, therefore decreasing the electrical resistivity of the nanocomposites. On the contrary, when the compression was released, the electrical resistivity of the nanocomposites gradually increased due to the increase in the inter-particle gaps. Additionally, the minimum FCR value did not change significantly after each loading–unloading cycle compared with the initial state, indicating that the electrically conductive network could be basically reset. In addition, the maximum value of FCR remained almost unchanged without slight fluctuations during the six compression loading and unloading cycles, which indicates that the nanocomposites possess considerable repeatability. Figure 16 shows the FCR–strain curves of composites subjected to compressive cycles. As illustrated in the figure, the fitting results of the three sets of data showed a strong linear relationship without fluctuation which was consistent with the results under monotonic compression loading. Among them, the nanocomposites containing 1.0 vol% CNTs–CB composite fillers still showed the greatest strain sensitivity, which was 36.8% higher than that of 1.5 vol% fillers. This phenomenon further verifies the significantly stable and repeatable piezoresistive response of the nanocomposites. In general, all aforementioned results exhibit that CNTs-CB/epoxy nanocomposites have good stability, and repeatability without hysteresis effect, which is quite beneficial for the applications of piezoresistive sensors in SHM.

Figure 15 Compressive stress, strain, and FCR value vs. time under constant cyclic compressive loading for nanocomposites loaded with: a 1.0 vol%; b 1.25 vol%; and c 1.5 vol% CNTs–CB composite fillers

Figure 16 FCR value of CNTs–CB/epoxy nanocomposites as a function of strain under constant cyclic compressive loading for nanocomposites loaded with: a 1.0 vol%; b 1.25 vol%; and c 1.5 vol% CNTs–CB composite fillers
Piezoresistive properties under incremental amplitude cyclic loading

Piezoresistive strain sensors will be inevitably used for the entire service life monitoring of infrastructures under different loading conditions. Therefore, the strain sensing properties of CNTs–CB/epoxy nanocomposites under incremental amplitude cyclic compressive testing were investigated in this study to simulate the real service circumstance.

The typical piezoresistive responses of CNTs–CB/epoxy nanocomposites as a function of loading time and interrelated compressive strain and stress during the loading–unloading compressive cycles are shown in Fig. 17. It is observed that FCR values monotonously increased and decreased as the variation of applied stress/strain regardless of the composite filler concentration, which verifies their excellent piezoresistive response under different loading amplitudes. Moreover, the absolute value of the FCR displayed an increasing trend with the increase in the compressive strain. This phenomenon could be ascribed to the decrease in distance between the adjacent CNTs–CB particles and the compact distribution of the conductive networks under high strain amplitudes. In other words, the increasing stress/strain amplitude gradually reduces the gaps of adjacent fillers and increases the number of conductive paths in composites, resulting in a more compact conductive network and a significant increase in piezoresistive responses. Therefore, the piezoresistive properties of the nanocomposite are significantly increased with the increase in the compressive strain/stress amplitude20.

Figure 17 Relationships of incremental cyclic compressive stress/strain and FCR versus time of nanocomposites filled with a 1.0 vol%; b 1.25 vol%; and c 1.5 vol% CNTs–CB composite fillers

Figure 17 shows that the maximum FCR values for the strain amplitudes below 1.2%, which was in the linear elastic stage of the nanocomposites, remained unchanged within each strain level. In this stage, the conductive networks deformed with the compression of the nanocomposites and reverted to the original morphology without residual deformation or irreversible damage after compression was removed. For the strain amplitudes beyond 1.2%, the maximum FCR values not only showed a clear increase but also exhibited a continuous upward tendency in the next two cycles. The distinct conductive networks generated by the synergistic effect in CNTs–CB composite fillers govern this particular phenomenon. This might presumably stem from the construction of some additional conductive pathways due to the rearrangement of the CNTs–CB composite fillers (mainly the movement and slippage of CB particles) at a higher strain level, thus causing the higher sensitivity41, 47–49. Moreover, the FCR values can not return to their initial values and obtain small positive values at the end of releasing, showing that irreversible destruction is formed in the conductive networks. The accumulation and expansion of microcracks in the matrix account for this phenomenon. Therefore, this result reveals that electrical resistivities of the CNTs–CB/epoxy nanocomposites are very sensitive to
the irreversible damage occurring within the matrix, which could be used to distinguish the linear elastic and the viscoelastic deformations.

The response time to applied stress/strain is also a crucial element in evaluating the piezoresistivity of nanocomposites for the applications of strain sensors\textsuperscript{59, 63}. Currently, the peak shift is used to evaluate the response time of sensors, and the lower peak shift indicates the smaller hysteresis, implying the accurately synchronous response between the variation of electrical resistivity and applied load\textsuperscript{63}. The schematic of the peak shift is shown in Fig. 18a and its definition is determined as follows:

$$\text{Peak shift} = \frac{\Delta t}{t} \times 100\%$$

where $\Delta t$ is the difference in time between peak stress and corresponding peak FCR value (Fig. 18a); and $t$ represents the time interval in one loading–unloading compression cycle (Fig. 18a).

Figure 18 Peak shifts of the CNTs–CB/epoxy nanocomposites: a the definition of the peak shift, and b the calculated peak shifts of the nanocomposites with different CNTs–CB under different compressive strain levels

In Fig. 18b, the peak shift values of CNTs–CB/epoxy nanocomposites with different concentrations of composite filler under different stress are displayed. Figure 18b shows that the peak shift values of nanocomposites maintained a relatively low level compared to some other research\textsuperscript{59, 63}. It should be noted that the testing equipment might have an unknown delay to catch and record data. If this was taken into account, the actual peak shift values for the nanocomposites would be lower. The result showed that the increasing CNTs–CB amount and compressive loading amplitudes could both reduce the peak shift values. The reduced distance between the adjacent CNTs–CB particles could be used to illustrate this phenomenon. In this case, the electron mobility between the composite fillers is highly strengthened based on the tunneling effect, thus improving the response time\textsuperscript{19, 59}.

Conclusions
The epoxy-based nanocomposites containing different concentrations of electrostatic self-assembly CNTs–CB composite fillers were fabricated in this study. The mechanical and electrical properties as well as the piezoresistive response of the CNTs–CB/epoxy nanocomposites were experimentally investigated. The mechanisms of the piezoresistive response of the CNTs–CB/epoxy nanocomposites at different strain levels were also explored. The main conclusions could be obtained from this study:

1. FESEM images displayed that the electrostatic self-assembly CNTs–CB composite fillers could achieve relatively uniform distribution in the epoxy matrix via a facile and simple mechanical stirring method without ultrasonic treatment on account of the synergistic effects. This is
because the grape clusters structure of the electrostatic self-assembly CNTs–CB composite fillers can prevent CNTs from approaching each other and forming agglomeration.

2. Compared with epoxy-based nanocomposites filled with only CNTs or CB particles, a moderate percolation threshold was obtained at 0.41 vol% for CNTs–CB/epoxy nanocomposites. This is because a more significant hybrid conductive network including short-range conduction and remote-range conduction was constituted by CNTs and CB particles due to the synergistic effect between CNTs and CB particles.

3. A highly sensitive and remarkably non-monotonic piezoresistive response of CNTs-CB/epoxy nanocomposites under monotonic compressive loading was achieved, which could be divided into three stages corresponding to different mechanisms. The strain sensitivities of the CNTs-CB/epoxy nanocomposites were much higher than that of the nanocomposites comprising CNT only, especially when they were subjected to larger strains. This is because the conductive networks tuned by CB particles were more vulnerable, leading to higher sensitivity.

4. Schematic models were proposed to illustrate the different mechanisms of the piezoresistive responses induced by the tunneling conduction and contacting conduction. The strain sensitivities of nanocomposites exhibited a linear increase followed by an abrupt rise and then a decrease. This could be interpreted by the distinct evolution of hybrid conductive networks constituted by the electrostatic self-assembly CNTs–CB composite fillers. The reduction in tunneling resistance dominated at the beginning; then, the CB particles could move into the gaps to strengthen the conductive networks, and ultimately, the conductive networks were gradually destroyed.

5. The electrical resistivities of CNTs–CB/epoxy nanocomposites synchronously and closely varied with the applied compressive strain (around 0.70%) under constant amplitude cycle loading, demonstrating excellent stability, recoverability, and repeatability in piezoresistive responses. These regular changes in electrical resistivities are attributed to the variation of nanoscale conductive networks structure in the matrix, i.e., the stochastic approaching and separation of inter-particle gaps induced by the compressive strains.

6. The piezoresistive responses of CNTs–CB/epoxy nanocomposites under incremental amplitude cyclic compressive loading also demonstrated their stable recoverability within a relatively broad strain range (around 1.20%). Beyond this strain range, slight irreversible resistivities were observed under high compressive strains. This phenomenon demonstrates that electrical resistivities are very sensitive to the irreversible damage in the matrix. This result could be used to distinguish the linear elastic and the viscoelastic deformations of nanocomposites and qualitatively identify the occurrence of irreversible deformation in the nanocomposites.

Due to the special grape cluster structure of electrostatic self-assembly CNTs–CB composite fillers, the distribution process of composite fillers could be greatly simplified. The introduction of electrostatic self-assembly CNTs–CB composite fillers into epoxy could generate complicated conductive networks and endow the CNTs–CB/epoxy nanocomposites with excellent mechanical properties, moderate percolation threshold, highly sensitive and repeatable piezoresistive response, which well balance the shortcomings of nanocomposites filled with only CNTs or CB particles. Among them, the nanocomposites containing 1.0 vol% CNTs-CB composite fillers had significantly higher strain sensitivity
and the minimum filler concentration, which showed attractive potentials for the fabrication of compressive strain sensors for structural health monitoring in civil infrastructures.

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Declarations
Conflict of interest
The authors declare that they have no conflict of interest.

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