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Variations in the capacitance and dielectric constant of multi-wall carbon nanotube filled silicone rubber composite during compressive creep

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The changes in the capacitance and the dielectric constant of multi-wall carbon nanotube filled silicone rubber composite during the compressive creep are studied. At the moment of the sudden compression, the capacitance and the dielectric constant decrease instantaneously, and the absolute values of the instantaneous decrements increase with the increase of the sudden compressive pressure. When the compressive pressure holds constant, the capacitance and the dielectric constant increase gradually and tend to be stable over time. Based on the viscoelastic theory, the mathematical model for the variations in the dielectric constant during the compressive creep is established. With the increase of the sudden compressive pressure, the dielectric constant retardation times hold constant and the stable normalized dielectric constant increases. With the increase of the carbon nanotube volume percent, the dielectric constant retardation times and the stable normalized dielectric constant decreases.

1. Introduction

Conductive powders can be dispersed into the polymer matrix to fabricate conductive polymer composite which is a popular functional material [1–8]. The conductive powders constitute a three dimensional conductive network [9–15], which can be changed under the external pressure [16–23]. Therefore, the composite has the potential to be used as a pressure sensitive material. Many researchers studied the piezoresistive effect of this composite [24–31], and developed the piezoresistive element based on the composite [32–39]. The composite can also be considered as a dielectric network composed of the conductive powders and the polymer segments. Capacitance, which is one of the macroscopic electrical characteristics of the composite, should change with the variations in the dielectric network of the composite caused by the external pressure. Therefore, the capacitance of the composite has the potential to be used to measure the applied pressure. Although there are some researches on the dielectric performances of the composite [40–44], there are seldom research on the relation between the compressive pressure and the capacitance of the composite during the compressive creep.

In this paper, we study the changes in the capacitance and the dielectric constant of the composite during the sudden compression and the compressive creep. As the composite is a viscoelastic material, the pressure sensitive properties (e.g. piezoresistivity) of the composite are time dependent [45–51]. Based on the viscoelastic theory and the experimental results, we know that the creep occurs when a constant pressure is applied on the composite. Some researchers have found that the resistance of the composite attenuates over time during the creep. They defined the attenuation of the composite resistance during the creep as the “resistance creep” [49–51]. Similarly, we define the changes in the capacitance and the dielectric constant during the compressive creep as the “compressive capacitance creep” and the “compressive dielectric constant creep”, respectively. In this paper, the solution mixing method is used to fabricate the carbon nanotube filled silicone rubber composite. The mathematical model for the variations in the dielectric constant during the compressive creep is established, and the effects of the compressive pressure and the carbon nanotube content on the key parameters of the model are also researched.

2. Experimental details

Multi-wall carbon nanotube (SZ Nanotech Port Co. Ltd, China) is used as the conductive phase. The average diameter ranges from

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40 nm to 60 nm. The average length ranges from 5 μm to 15 μm. Silicone rubber (Shenyang Silicone Plant, China) is used as the insulating phase. The relative dielectric constant is 3. The dielectric strength is 15 kV/mm. Ethyl silicate is used as crosslinker. Hexane is used as a solvent. Mechanical stirring along with ultrasonic vibration is used for filler dispersion (Type of the digital ultrasonic cleaner: KQ-100DB, working frequency: 40 kHz; Type of the electromotive stirrer: JJ-1100, power: 100 W). After 3 h of vigorous mixing, the solvent is evaporated. The mixture is molded into a rectangular sheet (30 mm × 30 mm × 1 mm). After 60 h of the vulcanization, the sandwich element based on the composite is completed. SEM (scanning electron microscope) micrograph of the composite fracture is shown in Fig. 1, indicating that the carbon nanotubes are dispersed in the matrix to constitute a conductive network.

The schematic for the sandwich element and the measurement method is shown in Fig. 2. The structure of the sandwich element ensures a constant cross-sectional area of the composite during the compression. The materials of the insulating layer and the rigid mold are the phenolic paper laminate which is rigid and insulating. The top electrode and the bottom electrode are made of copper, and the thickness of each electrode is 1 mm. The aforementioned design can ensure that the top electrode moves as a whole unit under the compression. Therefore, there is no local deformation of the top electrode at the edges of the insulating plate when the compressive pressure is applied. The composite is compressed instantaneously from zero-pressure to a certain pressure by exerting weights on the rigid head covered on the top electrode. Then, the compressive pressure is kept invariant. The compressive pressure ranges from 0 to 0.07 MPa. The values for the capacitance of the sandwich element based on the composite during compression are acquired by using a digital meter (UA9205A). All experiments are done under the condition of room temperature.

3. Results and discussion

Fig. 3 shows the relation between the carbon nanotube volume percent and the relative dielectric constant of the composite (i.e., the ratio of the dielectric constant of the composite to the vacuum dielectric constant). The results indicate that there is an occurrence of percolation in the composite. The relative dielectric constant increases slightly with the increase of the carbon nanotube volume percent φ when φ is lower than 4 vol%. When the carbon nanotube volume percent is higher than 6 vol%, the relative dielectric constant is improved substantially. As the low carbon nanotube volume percent cannot improve the dielectric constant substantially and the high carbon nanotube volume percent deteriorates the mechanical properties of the composite (e.g. elastic modulus), the range of the carbon nanotube volume percent which has the engineering application values is between 6 vol% and 12 vol%. Therefore, the composites with the aforementioned range (6 vol % < φ < 12 vol%) are studied in this paper. Based on our investigation, the conclusions regarding the effects of the compression on the capacitance and dielectric constant for the composite with the carbon nanotube volume percent of 8 vol % are the same as those for the composites with other carbon nanotube volume percents. Therefore, for the conciseness of discussion, we take the sandwich element based on the composite with the carbon nanotube volume percent of 8 vol% as a representative to study the changes in the capacitance and the dielectric constant during the compression in this paper.

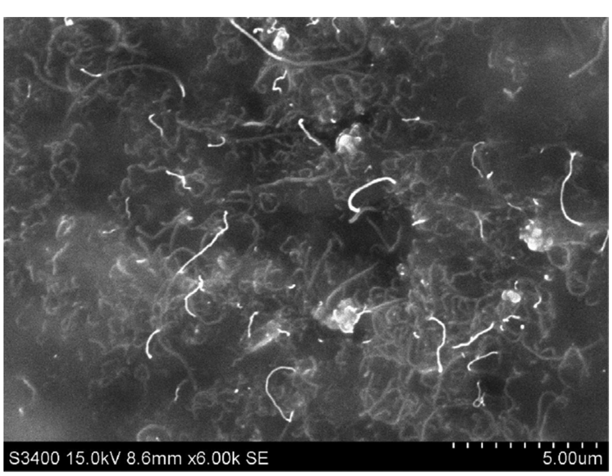

Fig. 1. SEM micrograph of the fractured surface for the composite.

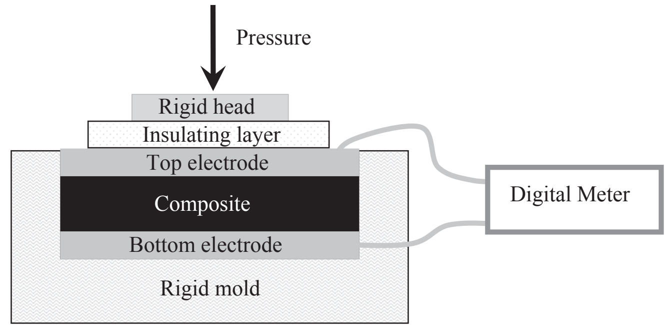

Fig. 2. Schematics for the structure of the sandwich element and the experimental set up.

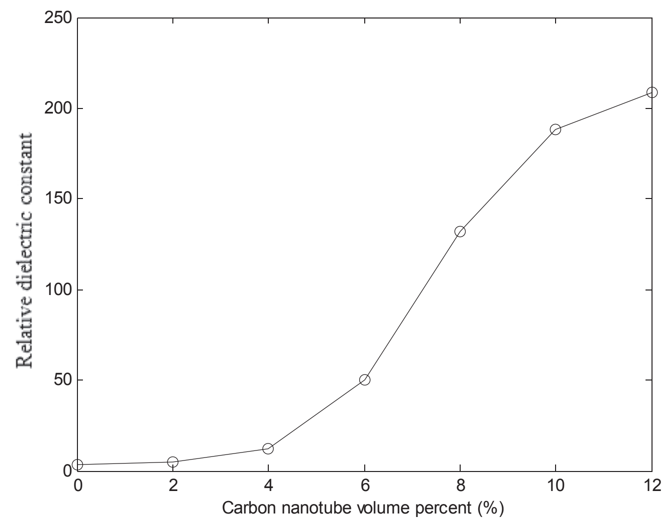

Fig. 3. Relation between the carbon nanotube volume percent and the relative dielectric constant of the composite.

As shown in Fig. 4(1), there are two stages for the changes in the strain of the composite during the compression. At the first stage, there is an instantaneous increase in the absolute value of the strain under the sudden compression. At the second stage, when the compressive pressure is kept constant, the absolute value of the strain increases gradually and tends to be stable over time (i.e., the compressive creep).

The changes in the capacitance of the composite are shown in Fig. 4(2). There are also two stages. At the first stage, there is an instantaneous decrease in the capacitance of the composite under