



A new insight on the variation of the electric conductivity and conductive network of silver-coated glass particles/silicone elastomer composites under tensile strain



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ABSTRACT

In this study, we introduced silver-coated glass particles (SG) into methyl vinyl silicone rubber (PMVS) matrix to prepare conductive elastomeric composites (CECs) with high electrical conductivity. The effect of the content of SG, the tensile strain and the stretch-recovery cycle times on the conductivity of CECs were studied. Interestingly, the conductivity of all the composites obviously increases with the increase in the applied tensile strain or the stretch-recovery cycle times to certain degree, different from that reported in many previous studies. The reason is that the dispersion of SG in PMVS matrix becomes more nonuniform with the increase in tensile strain, and the redistribution of SG in matrix results in the formation of much more new conductive network. On the other hand, the change in conductivity of the composites with the content of SG just exceeding the percolation threshold is much more significant than that with the content of SG far exceeding the percolation threshold. This is attributed to the more perfect conductive network in the composites with higher content of SG. The relationship between the conductivity and conductive filler network of SG/PMVS composites were clarified. This study provides guidance for the preparation and application of high performance CECs with excellent conductivity and stability of conductivity.

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1. Introduction

Conductive elastomeric composites (CECs) combine the good flexibility, high elasticity and sealability of elastomers as well as the excellent electrical conductivity of conductive fillers [1,2], and thus CECs have been widely used in industry such as electromagnetic interference (EMI) shielding, electrostatic charge dissipation, touch control switches and sensor [3–5]. As the content of conductive filler far exceeds the percolation threshold, the conductive network is fully formed and the conductivity of CECs is closer to that of the conductive filler. The commonly used metal powders such as silver (Ag) and copper (Cu), have high electrical conductivity, but they are quite expensive and have high densities. In recent years, much attention has been paid to reduce the percolation threshold of CECs

and obtain high conductivity and mechanical properties simultaneously by the introduction of high aspect ratio conductive filler such as conductive carbon fiber (CF), carbon nanotubes (CNTs) and graphene [6–8]. Although CNTs and graphene theoretically have high conductivity, the electrical conductivity of the CECs is still not high due to structural defects and low purity of these carbon fillers. On the other hand, the nano-scale dispersion of these carbon nanofillers in elastomer matrix is quite difficult to be obtained on a large scale [9]. Compared with metal powders and conductive carbon fillers, metal-coated inorganic fillers such as silver coated-glass particles, silver coated-aluminum, and nickel-coated graphite etc., have the advantages of high electrical conductivity, low density and low cost, thus are widely used as conductive filler in EMI shielding materials [10].

CECs are subjected to tensile stress or compress stress during storage, installation and application, leading to the destruction and reconstruction of conductive network and thus inevitably affect the electrical conductivity of the composites. Therefore, in addition to

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obtaining high conductivity, the stability of electrical conductivity during deformation is the most important performance [11]. Many previous studies show that the conductivity of these CECs decreases with increasing the tensile strain. For example, it has been reported that the conductivity of carbon black (CB)/rubber composite decrease with the increase in tensile strain because of the rapid destruction of conductive path under deformation [12]. In addition, Sun et al. [13] found that the conductivity of nickel-coated graphite (NCG)/silicone rubber composites decrease and stability of conductivity become worse with the increase of tensile strain and cycles of tensile deformation. In our previous studies, we also [14] found that the conductivity and stability of conductivity of nickel-coated carbon fiber (NCF)/polymethylvinylsiloxane (PMVS) composites decrease with the increase in external strain. On the other hand, several microscopic characterization techniques such as Scanning Electron Microscope (SEM) and Transmission Electron Microscopy (TEM), were used to characterize the conductive filler network. However, the evolution of conductive network of CECs under tensile strain and the relationship between conductive network and conductivity is still not clear.

Silicone elastomer has good flexibility, high elasticity, excellent resistance to high and low temperatures, low viscosity, and low hardness, and thus widely used as CECs matrix. Herein, we used methyl vinyl silicon rubber (PMVS) as matrix and silver-coated glass particles (SG) as conductive filler. We selected two filler contents (one just exceeded the percolation threshold and the other far exceeded the percolation threshold), and studied the effect of the SG content, the tensile strain and the stretch-recovery cycles on the conductivity and the stability of conductivity of the SG/PMVS composites. We aim to clarify the relationship between the conductivity and conductive filler network of SG/PMVS composites. We also aim to provide guidance for the preparation high performance CECs with excellent conductivity and stability of conductivity.

2. Experimental

2.1. Materials

Methyl vinyl silicone rubber masterbatch (PMVS, Grade 110-2, silica-containing, 1.06 g/cm³) was chosen as silicone rubber matrix. Silver-coated glass particles (SG, average particle size of them is 34 μm) were supplied by PQ Co., Inc (America). The coupling agent vinyltriacetoxysilane (A151) was supplied by Beijing Reagent Chemical Corporation (China). The vulcanizing agent 2,5-dimethyl-2,5-bis(*tert*-butyl peroxy) hexane (DBPMH, Grade 101-SC20) was purchased from Akzo Nobel. The co-curing agent triallyl isocyanurate (TAIC) was purchased from Hunan Liuyang Chemical Co., Ltd.

2.2. Preparation of composites

First of all, we mixed conductive filler SG with coupling agent A-151 for 3–5 min until there was no dry SG powders left. Then, SG/PMVS composites were prepared with two-roll open mill by introducing PMVS, the mixture of conductive filler and A-151 and vulcanizing agent DBPMH, in proper sequence. The composites were first vulcanized on a lab platen press at 170 °C under 10 MPa for 20 min and then were vulcanized in a draught drying cabinet under air atmosphere at 200 °C for 2 h.

The size of samples used to be tested was 10 mm × 100 mm × 2 mm. The samples needed to be kept for 24 h in the standard experimental condition (23 ± 2 °C, relative humidity 50 ± 10%) before testing.

2.3. Characterizations

The volume resistivity of the composites with a high resistivity ($\geq 10^4 \Omega$) was measured by using a numeric high resistance meter (EST 121, Beijing Huajinghui Science and Technology Co., Ltd., China). A DC Bridge (QJ84, Shanghai Zhengyang Instrument Factory, China) was used to measure the volume resistivity of composites with low resistivity according to Chinese standard GB/T 2439-2001. The volume resistivity ρ was calculated by:

$$\rho = R \times S/L \quad (1)$$

where R is the electrical resistance of the sample, S is the cross-sectional area, and L is the length of the sample. The conductivity (σ) is the reciprocal of the volume resistivity. Five samples were tested for each composite and the average value was reported.

The conductivity stability was characterized by the ratio of the conductivity of samples after and before tension. In the single stretch-recovery experiment, samples were stretched to a certain strain (10%, 30%, 50%, or 100%) by using a self-made tensile installation and kept for 10 min, the stretching speed is 50 mm/min. Repeated stretch-recovery experiments were carried out by a fatigue testing machine for rubber. Samples were stretched to a certain stretch-recovery cycle number under a fixed tensile strain (20% and 50%), the stretching speed is also 50 mm/min. The volume resistivity was measured and conductivity was calculated after the samples were unloaded and placed in the standard experimental conditions for 30 min. Three samples were tested for each composite and the average value was reported.

The network structure of conductive filler in the composites was characterized by using a RPA-Rubber Process Analyzer (RPA-2000, Alpha Technologies Co., Ltd., USA). Samples were placed at the temperature 170 °C for 15min for vulcanization and tested under a frequency of 1 Hz with the strain amplitude ranging from 0.28% to 100%. The morphology of conductive filler network structure was observed by using a Scanning Electron Microscope (SEM, XL-30, FEI Co., Ltd., USA). Before observation, samples were cryogenically fractured in liquid nitrogen and coated with gold.

3. Results and discussion

3.1. The relationship between conductive network and conductivity

The conducting mechanism of composite generally includes the penetration theory, tunnel effect theory, and electric field emission theory [15–17]. It is well known that the conduction of electricity is owing to the formation of conducting pathway or network when the conductive filler content reaches a critical volume fraction known as percolation threshold, which allows the movement of electrons. The distribution of SG in PMVS matrix was observed with SEM. The micrographs of composites with filler contents of 16.7 vol%, 28.6 vol%, 37.6 vol% and 44.5 vol% are shown in Fig. S1 a–d, representing different levels of conductive network. As shown in these micrographs, SG powers in composites with filler contents of 16.7 vol% and 28.6 vol% are almost isolated, while conductive fillers become partially contacted with each other in composites with filler content of 37.6 vol%. Fillers begin to gather and most particles contact with each other in composites with filler content of 44.5 vol%. Inter-particle contact leads to the formation of conductive paths in different directions to constitute 3D filler network, which we called conductive network.

Conductive filler form a filler network in the polymer matrix, which can also be indicated by the decreasing of the storage modulus (G') with shear strain by using rubber processing analysis (RPA), as reported in our previous study [18]. We found that the

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