



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



Nanying Ning^{a, b}, Chunmeng Miao^b, Hua Zou^{a, b}, Qian Shao^b, Sishu Wang^b,
Liqun Zhang^{a, b}, Ming Tian^{a, b, *}

^a State Key Laboratory for Organic-Inorganic Composites, Beijing University of Chemical Technology, Beijing, 100029, PR China

^b Key Laboratory of Carbon Fiber and Functional Polymers, Ministry of Education, Beijing University of Chemical Technology, Beijing, 100029, PR China

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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

* Corresponding author. State Key Laboratory for Organic-Inorganic Composites, Beijing University of Chemical Technology, Beijing, 100029, PR China.

E-mail address: tianm@mail.buct.edu.cn (M. Tian).

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 15 min 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

conductive network is closely related to the filler network [19]. RPA is commonly used to characterize the filler network formed by filler dispersion in the polymer [14,18]. Here we use RPA to study the filler network of SG/PMVS composites with different filler contents. As shown in Fig. 1, the storage modulus G' decreases rapidly with the increase in shear strain for all the samples. This phenomenon is known as Payne effect and it represents the strength of the interactions among SG in the PMVS matrix. For composites with filler contents of 0 vol%, 16.7 vol% and 28.6 vol%, the storage modulus G' is almost the same and Payne effect is weak, indicating that the strength of interaction among conductive fillers is weak. There is an obvious increase in Payne effect after the content of SG reaches 37.6 vol%. The Payne effect become stronger with the increase in filler content, suggesting that more and more SG contact with one another. As a result, the conductive network become stronger. The difference between the maximum and minimum of G' , named as the modulus attenuation ($\Delta G'$), was calculated according to RPA curves to reflect the strength of the filler network in matrix more clearly, as shown in Fig. 2. A larger $\Delta G'$ indicates a stronger filler network. $\Delta G'$ is very low as the filler content is low, and it increases with the filler content increasing to the percolation threshold. In this case, the conductive path is gradually formed, leading to the sharp increase in conductivity. $\Delta G'$ increases further as the content of fillers increasing to much larger than the percolation threshold, indicating that the filler network become stronger.

As shown in Fig. 2, the conductivity of the CECs with different contents of SG was also investigated, and the differential curve of $d(\lg\sigma)/dV_f$ is shown in the inset. The conductivity of SG/PMVS composites increases with the increase of filler content, and it increases by 18 times as the content of SG reaches the percolation threshold (31 vol%). The results of the conductivity of composites are consistent well with the filler (or conductive) network.

As shown in the differential curve in the inset of Fig. 2, the turning points of the conductivity and $\Delta G'$ are 31 vol% and 43 vol% respectively, illustrating the construction of conductive network preceded that of filler network. When the filler content is low (below 31 vol%), the filler network structure is not formed and the conductivity is poor. When the filler content is between 31 vol% to 43 vol%, the filler network increases slightly while the conductivity increases sharply, indicating the contribution of tunneling current to the conductivity. When the filler content is high (over 43 vol%), the filler forms a highly developed network structure, but the conductivity is not significantly improved, indicating that the late formed filler network only strengthens the previously formed three-dimensional conductive network but it does not form new

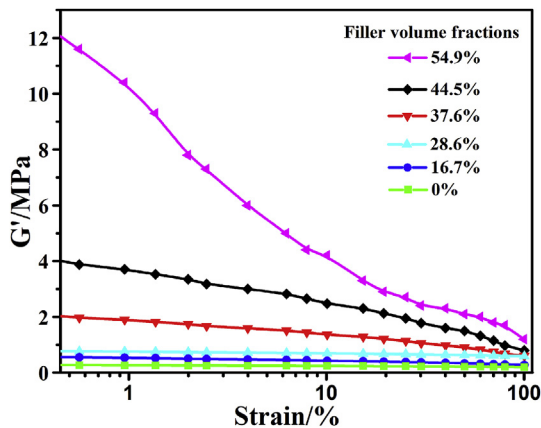


Fig. 1. Storage modulus as a function of shear strain for silver-coated glass particles/PMVS composites with different filler volume fractions.

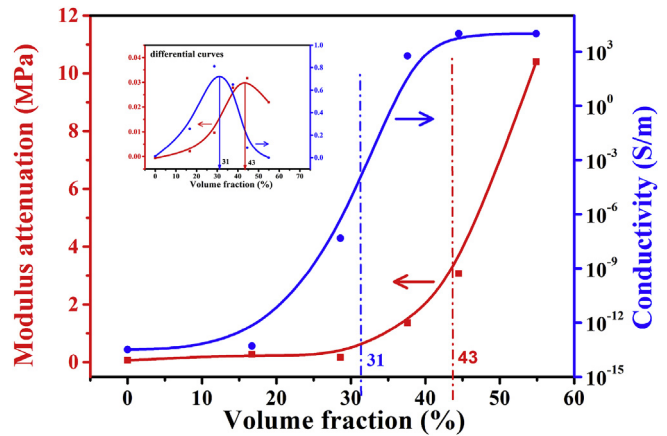


Fig. 2. Plots of the modulus attenuation ($\Delta G'$) and conductivity against the volume fraction V_f of filler in the composites. The inset in figure show the differential curves.

conductive network. It is simple and easy to establish the relationship between the conductivity and filler network (an indication of conductive network) by using RPA within certain range.

When the filler content is close to threshold percolation, the conductivity of conductive composites is unstable and could be easily affected by external forces. Therefore, the composite with filler content of 37.6 vol% corresponding to the point just exceeding the percolation threshold was studied for the following research. While the composite with filler content of 44.5 vol% corresponding to the point far exceeding the percolation threshold was studied as the control group.

3.2. Effect of tensile strain and strain cycle on conductivity

The study on the conductivity variation after being exposed to external forces is important to provide guidance for the achievement of a long lifetime of CECs. In this paper, the evolution of the conductivity of the composite under tensile strain is systematically studied. The change in conductivity is represented in terms of relative conductivity (σ/σ_0), where σ and σ_0 refers to the conductivity of the samples after and before being exerted tensile strain respectively. The closer the σ/σ_0 is to 1.0, the more stable of the conductivity.

3.2.1. Effect of single tensile strain on conductivity

The σ/σ_0 against tensile strain after stretching for 10 min and then relaxing for 30 min for composites with filler contents of 37.6 vol% and 44.5 vol% are shown in Fig. 3. Interestingly, the conductivity of both the two composites continuously increases with the tensile strain increasing to 50% and then the conductivity is almost unchanged when the tensile strain is larger than 50%. The σ/σ_0 of all the composites is higher than 1.0, indicating that the conductivity of all the composites increases after stretching, different from that reported in previous studies [20,21]. This indicates that single tensile strain benefits for the formation of new conductive network, and thus resulting in better conductivity. The σ/σ_0 of the composite with filler content of 37.6 vol% increases from 1.06 to 1.89 when the tensile strain increases from 10% to 50%, and it further slightly increases to 1.92 when the tensile strain reaches 100%. The σ/σ_0 of the composite with filler content of 44.5 vol% are 0.95 and 1.08 at the tensile strain of 10% and 30%, respectively. When the tensile strain increases to 50%, the σ/σ_0 increases to 1.15, and there is no significant change as the tensile strain reaches 100%. The σ/σ_0 at the tensile strain of 100% of the composites with filler contents of 37.6 vol% and 44.5 vol% are about 2.0 and 1.1,

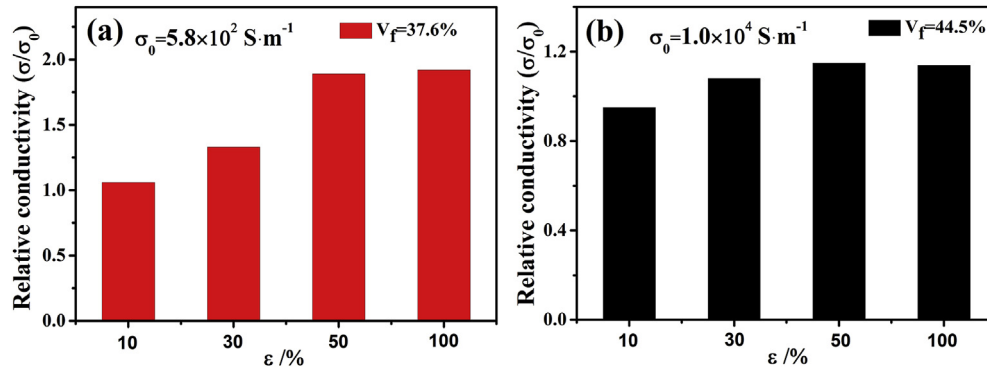


Fig. 3. Bar graphs of relative conductivity (σ/σ_0) against tensile strain for the composites with different filler volume fractions.

respectively, indicating the conductivity of the composite with higher filler content is more stable. These results also indicate that the maximum tensile strain in practical application shouldn't exceed 30% and 50% for composites with filler contents of 37.6 vol% and 44.5 vol%, respectively.

3.2.2. Effect of stretch-recovery cycle on conductivity

The σ/σ_0 against the stretch-recovery cycle of composites with filler content of 37.6 vol% and 44.5 vol% are shown in Fig. 4, the composites were stretching under a fixed tensile strain (20% and 50%) and then relaxing for 30 min. From these bar graphs we could see similar phenomenon occurred in single stretch-recovery experiment that the conductivity of both two composites increases with increasing the cycle times. The formation of new conductive networks benefits from tensile strain. Under the fixed tensile strain of 20%, the σ/σ_0 of the composite with filler content of 37.6 vol% increases from 2.13 to 4.76 with the stretch-recovery cycle increasing and that of the composite with filler content of 44.5 vol% increases from 1.25 to 1.54. Under the fixed tensile strain of 50%, with the stretch-recovery cycles increasing, the σ/σ_0 of composites with filler content of 37.6 vol% and 44.5 vol% increase from 2.13 to 1.25 to 2.86 and 1.41, respectively. Obviously, the variation of conductivity of the composites is higher under tensile strain 20% than that under 50%, indicating that a smaller tensile strain can more strongly affect the conductivity of the composites. On the other hand, the variation of conductivity of the composites with lower filler content is higher than that of the composites with higher filler content. Meanwhile, for the composites with filler content of 37.6 vol%, the conductivity become better and the variation of conductivity increases with the increase of stretch-recovery cycles under either 20% or 50% strain, and the change is bigger as stretch-

recovery cycle exceeds 10^3 . Compared with single stretch-recovery experiment, the σ/σ_0 of the composites in repeated stretch-recovery experiment is higher. These results suggest that the increase in stretch-recovery cycles lead to better conductivity but worse stability of conductivity of the composites.

3.3. The mechanism of strain effect on conductivity and conductive network

We used SEM to characterize conductive network of composites after tensile strain. The micrographs of the composite with filler content of 37.6 vol% after single stretch-recovery cycle with different tensile strain are shown in Fig. 5 a–d. SG particles are evenly distributed in PMVS matrix in un-stretched samples and there are almost no aggregation of SG particles, as shown in Fig. 5a. Compared with the un-stretched samples, the dispersion of SG in PMVS matrix becomes more nonuniform after being applied to tensile strain owing to the weak interaction between the SG and PMVS. SG in some regions become denser while SG in some other regions become sparser, leading to the breakdown of previously formed conductive network and the formation of new conductive network. The redistribution of conductive filler in the matrix result in the increase in conductivity of the composites owing to the formation of many additional new conductive network. For the composite with filler content just exceeding the percolation threshold, moderate dispersion of the conductive filler is beneficial for the formation of conductive network and the achievement of high conductivity, which is different from that reported in many previous studies that a better dispersion of conductive filler leads to better conductivity [22]. The similar phenomenon occurred in the composites with filler content far exceeding the percolation

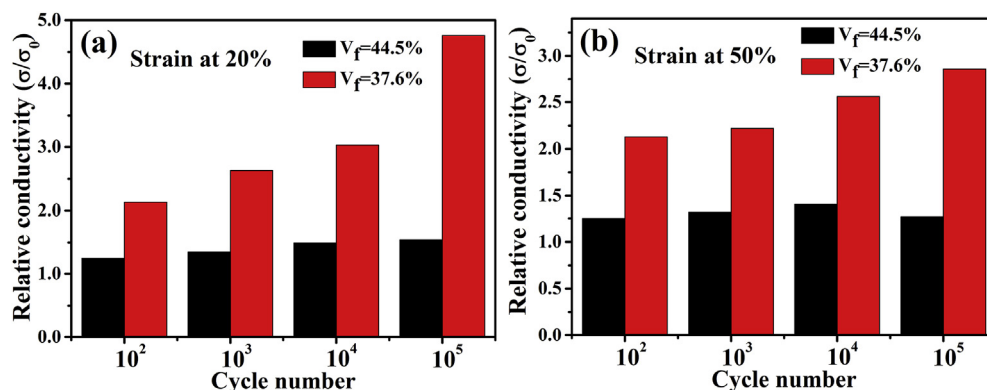


Fig. 4. Bar graphs of relative conductivity (σ/σ_0) against stretch-recovery cycle number at different tensile strain.

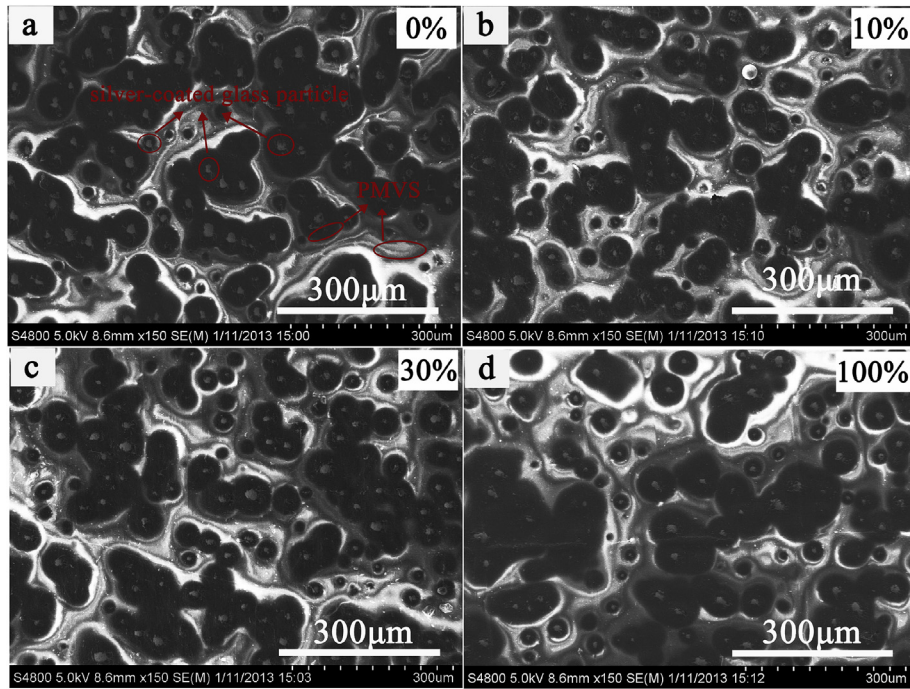


Fig. 5. SEM micrographs of composite with filler content of 37.6 vol% after single stretch-recovery cycle with different tensile strain (unstrained state 0%, 10%, 30%, 100%).

threshold, but the variation of conductivity is smaller as the conductive filler increases, attributed to the more perfect conductive network in conductive region as shown in Fig. S2 a–d. In addition, the filler dispersion in PMVS matrix becomes more nonuniform with the increase of tensile strain, leading to the formation of more new conductive network and thus the increase in the conductivity.

The micrographs of the composite with filler content of 37.6 vol%

% after different repeated stretch-recovery cycles (0, 10^2 , 10^4 , and 10^5) under fixed tensile strain 50% are shown in Fig. 6 a–d. The redistribution of SG in PMVS matrix in repeated stretch-recovery experiment is even more obvious than that in single stretch-recovery experiment. More interestingly, we can even distinguish conductive paths newly formed by conductive fillers in the composite after being applied to repeated stretch-recovery cycle in Fig. 6 (c) and (d). A schematic representation of the enrichment of

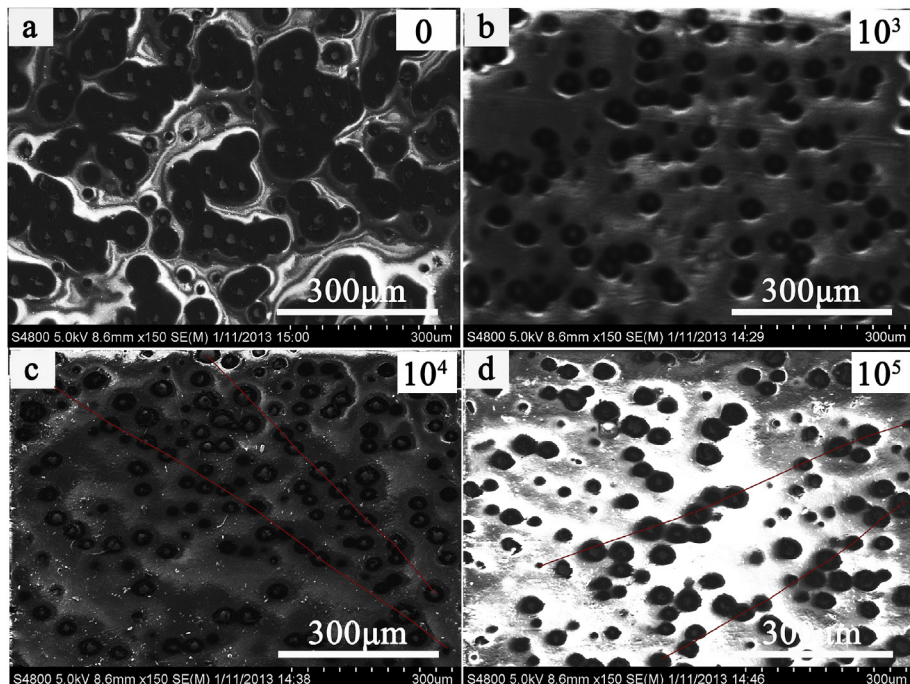


Fig. 6. SEM micrographs of composite with filler content of 37.6 vol% after different repeated stretch-recovery cycles (0, 10^2 , 10^4 , 10^5) under fixed tensile strain 50%. 300 μm.

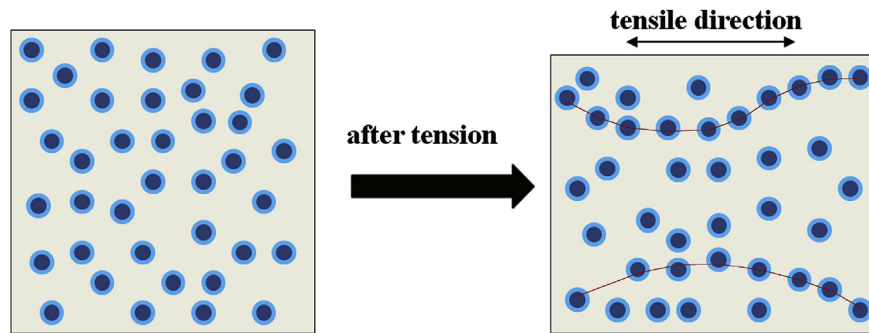


Fig. 7. Schematic representation of the enrichment of conductive fillers along the tensile direction under tensile strain.

conductive fillers along the tensile direction under tensile strain is illustrated in Fig. 7. The nonuniformity of filler dispersion increases and breakdown of original conductive network occurs, facilitating the reconstruction of conductive network along the tensile direction. As a result, the conductivity in the tensile plane increases, consistent well with our previous simulation calculations [23]. It explained why the σ/σ_0 in repeated stretch-recovery experiment is higher than that in single stretch-recovery experiment. In addition, we can also explain that smaller tensile strain have more obvious influence on the increase in relative conductivity than large tensile strain in repeated stretch-recovery experiment. At higher tension strain (50%), conductive particles gradually redistributed along tensile direction with the increase in repeated stretch-recovery cycles. The orientation degree of conductive particles in matrix in repeat stretch-recovery experiment under tensile strain of 50% is higher than that under smaller tensile strain (20%). A lower orientation degree has more obvious influence on the increase in the number of conductive path. Thus, smaller tension strain (20%) has more obvious influence on the increase in relative conductivity.

4. Conclusions

We introduced SG into PMVS matrix to prepare CECs with high electrical conductivity and studied the effect of the content of SG, the tensile strain and the stretch-recovery cycle times on the conductivity of CECs. The results show that 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. The dispersion of SG in the PMVS matrix become more nonuniform after being applied to tensile strain owing to the weak interaction between SG and PMVS. The redistribution of SG in the matrix results in the formation of much more new conductive network and thus results in the increase in conductivity of the composites. The enrichment of conductive filler along the tensile direction occurs and the conductivity in the tensile plane increases for composites after repeated stretch-recovery cycle. In addition, the change in conductivity of the composites with the content of SG just exceeding the percolation threshold is much more than that with the content of SG far exceeding the percolation threshold because of the more perfect conductive network in the composites with higher content of SG. This study provides guidance for the preparation and application of high performance CECs with excellent conductivity and stability of conductivity.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.compscitech.2016.10.001>.

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