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A new kind of electro-active polymer composite composed of silicone elastomer and polyethylene glycol

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Abstract
In this work, a new kind of electro-active polymer composite composed of silicone and polyethylene glycol (PEG) was prepared by solution blending. Two types of PEG with average molecular weights of 600(PEG600) and 1500(PEG1500) were first blended with liquid silicone (DC3481) separately. Then, the dielectric, electrical, mechanical and electromechanical properties of pure silicone and the silicone/PEG composites were investigated. The silicone/PEG600 composite shows an increased dielectric constant and a decreased Young's modulus, resulting in an improved figure of merit (FOM) and an actuation strain of 11.5% at 40 V μm^{-1}, which is 64% higher than pure silicone. This indicates that the actuation properties of silicone are largely improved by the addition of PEG600. However, for the silicone/PEG1500 composite, a much higher dielectric constant and an increased modulus due to its semi-crystalline structure are obtained, which lead to a decreased FOM and a smaller actuation strain than that of pure silicone. In addition, a comparison between theoretical strain and the corresponding experiment value was made. And the results indicate that not only the FOM, but the dielectric loss and mechanical loss play an important role in the actuation properties.

(Some figures may appear in colour only in the online journal)

1. Introduction

Electro-active polymers (EAPs) inherently capable of converting electrical energy to mechanical work via large voltage-controlled changes in size or shape have been studied for many years [1–3]. One class of EAP known as dielectric elastomers (DEs) combines the best set of advantages for any soft actuator such as low modulus, high electromechanical coupling efficiency, high actuation strain, high response speed and facile process ability [4–7]. Several applications have already been demonstrated, including artificial muscles [8–10], optical lenses [11], robotics [12] and haptic displays [13]. Their application as electrical power generators is also of high interest [14]. Although there are many types of applications, there is still a severe lack of materials with a favourable profile of mechanical and dielectric properties [15].

Currently, for enhancing the dielectric permittivity of a polymer, two main routes are being followed—composites of polymer matrices with high permittivity ceramic fillers [16–18], i.e. sub-percolation conductive particles such as carbon nanotube, carbon black and graphene [19–22], and highly polarizable conjugated polymers such as poly(hexylthiophene) [23], polyaniline [24] and copper phthalocyanine oligomers [25]. Adding a large amount of ceramic fillers to a polymer matrix always results in an increase

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in modulus, loss of flexibility and inhomogeneous composites because the filler provides good reinforcement [26], which is an undesired side effect as the materials need to be very soft [27]. At the same time, the filler particles are difficult to disperse uniformly and the filler particles would fall off in the process of actuation if the interface of the filler particles and the polymer is not combined well. In the other route, i.e. adding conductive fillers or highly polarizable conjugated polymers, the dielectric loss is very high and difficult to control, because the particles can easily form a conductive path and produce local current in the composite as the filler concentration approaches the percolation threshold [27]. The dielectric loss would transform to thermal energy, which makes the breakdown of the material easy.

A dielectric elastomer actuator (DEA) is to some extent a compliant capacitor, in which actuation is due to the coupling of electrostatic stress with elastic stress. It has been well known for many years that the electric field pressure from free charges on the surface of all insulating materials induces stresses (Maxwell's stress) that make the material deform. For low strains (e.g. $<20\%$), the thickness strain of a DEA can be approximated by

$$S_z = -\frac{P}{Y} = -\frac{\varepsilon_0 \varepsilon_r E^2}{Y}. \qquad (1)$$

For strains greater than about 20%, equation (1) is unsatisfactory, and it is replaced by a more complicated nonlinear hyper-elastic equivalent, because Y generally depends on the strain itself [6]. Here P is Maxwell's stress on the material, ε_r is the relative permittivity, ε_0 is the permittivity of free space, E is the applied electric field and Y is Young's modulus. In addition to increased relative permittivity, DEA materials also need to be soft to improve performance. Therefore, the factor $\beta = \varepsilon_r / Y$ can be defined as the figure of merit (FOM) for a given electric field ratio, ε_r / Y, and quantifies the improvement in material properties for a DEA and it is clear that the ideal material for a DEA would have a high dielectric constant, a high electric breakdown and a low Young's modulus [1].

According to Mali's study [28], the dielectric constant of polyethylene glycol (PEG) with an average molecular weight of 600 is 12 at 10 MHz and room temperature. It is a good soluble in many kinds of solvents and compatible with many kinds of polymers. In order to prepare a composite combining the merits of high dielectric constant and low modulus, two types of PEGs with different molecular weights were blended with the silicone elastomer. The dielectric, mechanical and actuation properties of the silicone/PEG composites were studied, and a comparison between the theoretical strain and experimental value was made to discuss the relationship between the actuation strain and the intrinsic properties of the materials.

2. Experimental

2.1. Materials

The elastomer used as a dielectric matrix is commercial silicone based on a polydimethylsiloxane mixture, available as a two-component product from Dow Corning (SILASTIC 3481 and hardener 81-F) in liquid form with a density of $1.213\,\mathrm{g\,cm^{-3}}$. PEG with different molecular weights was obtained from Longxi chemical Co., LTD (Guangdong Province, China). PEG600 with an average molecular weight of 600 and PEG1500 with an average molecular weight of 1500 have softening points of 22 °C and 46 °C, respectively. Tetrahydrofuran (AR) as a solvent is from Beijing Eastern Chemical Works (Beijing, China).

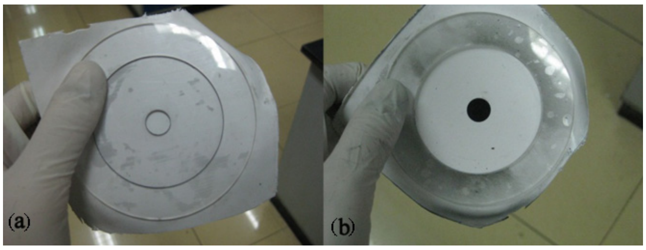

Figure 1. Dielectric elastomer films in the frame: (*a*) before spraying electrodes, (*b*) after spraying electrodes.

2.2. Processing

For film preparation, silicone fluid and PEG were separately dissolved in THF and stirred for several minutes, then the PEG in THF was heated to 50 °C to dissolve completely. After that, the PEG solvent and silicone hardener were added into the silicone. The well-mixed solvent was poured into a Teflon mould in a fume cupboard for 6 h at room temperature for curing. The thickness of the films was controlled by changing the amount of the suspension poured into the mould. For electromechanical tests the thickness of the film was around 0.35 mm, for Young's modulus and dielectric property tests, the thickness was around 1 mm.

2, 5 and 8 phr (phr refers to per 100 g rubber) PEG600 and PEG1500 were added to silicone separately to fabricate the composites.

2.3. Measuring methods

The dielectric properties were measured by an impedance analyser (HP4294A, Agilent, USA) with quadrate samples over the frequency range from 100 to 10^6 Hz at room temperature. Young's modulus of the pure and blended silicone elastomers was determined by the slope of the stress–strain curves (within 5% strain) obtained by a tensile apparatus (CMT4104, Shenzhen SANS Testing Machine Co., Ltd, China) at a strain rate of $50\,\mathrm{mm\,min^{-1}}$ whereby the samples were rectangular and of 25 mm length. The dynamic mechanical properties were tested through a strain sweep experiment by DMTA (PE7, Perkin-Elmer, Inc) at 1 Hz and room temperature. The compliant electrodes were fabricated by spraying graphite suspension on the two main surfaces of the film using an airbrush. The actuated strain measurements were carried out by a circular strain test. The film was fixed on a circle frame shown in figure 1. The strain was defined as the change in the pixel of the electrodes' area divided by the original pixel. The temperature of the elastomer films

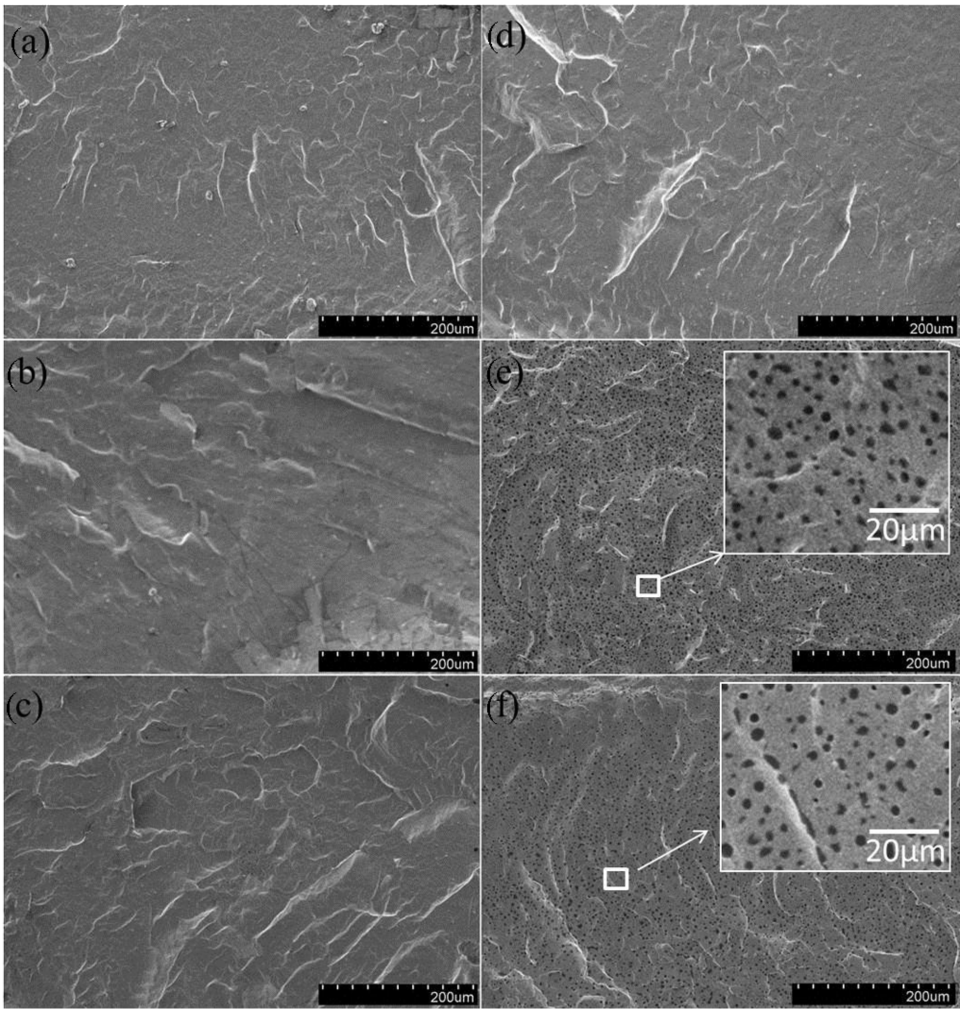

Figure 2. SEM images of (*a*) pure silicone, (*b*) 5 phr PEG600/silicone composite, (*c*) 5 phr PEG1500/silicone composite, (*d*) pure silicone etched by ethanol, (*e*) 5 phr PEG600/silicone composite etched by ethanol and (*f*) 5 phr PEG1500/silicone composite etched by ethanol.

after the actuation test was measured by a laser sight infrared thermometer (Optris LS). The voltage was supplied by a high-voltage direct current generator (DTZG-60, Wuhan Dotek Electric Co., Ltd). In order to obtain the actuated strain, a video camera was fixed to capture the actuator plane before and after applying the voltage with the same focal length, and then the captured video pictures were processed with Photoshop software. The morphology of the DE films was analysed by scanning electron microscopy (SEM, S-4700, Hitachi). First, the samples were obtained from liquid nitrogen brittle fracture, then one of the brittle fracture surfaces was etched by ethanol for 40 min at room temperature. The etched surface and untreated surface were both observed by SEM.

3. Results and discussion

3.1. PEG dispersion in the silicone matrix

In order to verify the dispersion state of PEG in the silicone matrix, composite films with 5 phr PEG600 and PEG1500 were prepared, and they were observed by SEM. Figures 2(*a*), (*b*) and (*c*) show the untreated brittle fracture surface of pure silicone, 5 phr PEG600/silicone and 5 phr PEG1500/silicone, and figures 2(*d*), (*e*) and (*f*) show the etched brittle fracture surface of pure silicone, 5 phr PEG600/silicone and 5 phr PEG1500/silicone, respectively. The micro-appearance had no difference between the untreated surface and the etched surface of pure silicone, indicating that ethanol could not dissolve the silicone matrix. The surfaces of both the etched 5 phr PEG600/silicone and PEG1500/silicone composites exhibit several uniformly dispersed micro-sized etching points, indicating that the dispersion of PEG in silicone is relatively uniform, but it is not dispersed in the form of individual molecules but PEG particles. This is because PEG dispersed in the silicone THF solution in the form of molecules, but as the THF evaporated they aggregated and phase separation occurred because of the difference in polarization between silicone and PEG. The dispersed phase was PEG, and the continuous phase was silicone.

3.2. Effect of PEG on the dielectric and electrical properties of silicone elastomers

The dielectric properties of the PEG600/silicone and PEG1500/silicone composites were characterized. Figure 3 shows the dielectric spectra of composites with 2, 5 and 8 phr

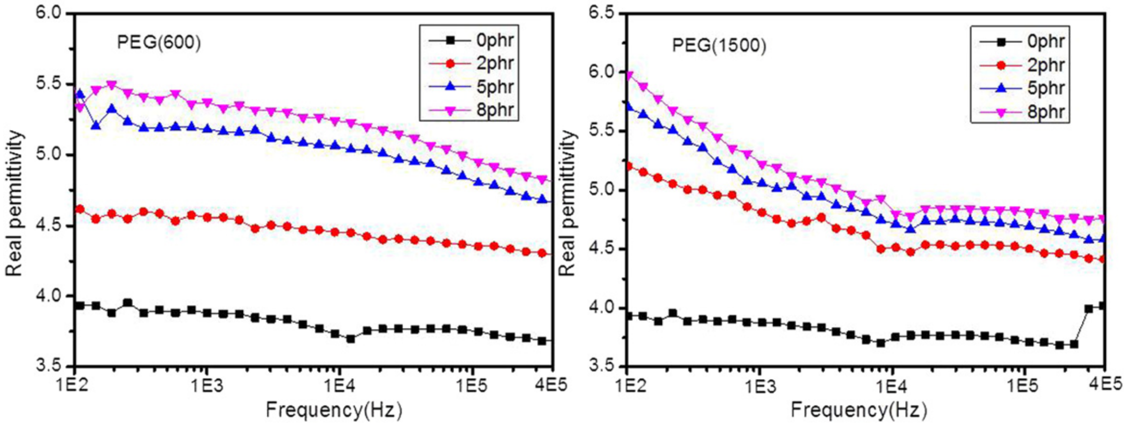

Figure 3. Dielectric constant of pure silicone and silicone/PEG composites.

PEG and pure silicone films as a function of frequency at room temperature. By comparing the spectra of the PEG600 and PEG1500 composites, some observations promptly arise: (1) the dielectric constant increases with the addition of either PEG600 or PEG1500, (2) the dielectric constant of the composites with PEG1500 is slightly higher than PEG600 and varies with loading fraction and test frequency. Increased permittivity of up to 6.0 at 100 Hz, 50% higher than pure silicone, was observed for the composite films with 8 phr PEG1500. The reason for the observation (1) is obvious and it is easy to explain. According to some classical models such as the logarithmic mixing rule,

$$\ln(\varepsilon_c) = v_1 \ln(\varepsilon_1) + v_2 \ln(\varepsilon_2), \tag{2}$$

the dielectric constant of composites depends on the volume fraction and the dielectric constant of constituents, and it will increase with the addition of a high dielectric constant filler. According to Ingo Aligs's research, PEG1500 has a semi-crystalline structure. And the semi-crystalline structure could provide an internal surface between the amorphous and the crystalline phases, which is sufficient for Maxwell–Wagner–Sillars-type polarization [29]. Since the softening point of PEG1500 (46 ± 2 °C) is much higher than the experimental temperature (room temperature), it would always maintain the semi-crystalline structure. PEG600 is a liquid at room temperature. So there are more interfaces in the PEG1500/silicone composites than in the PEG600/silicone composites and there is more localization of charge carriers at the interface of the PEG1500/silicone composites. The higher dielectric constant of PEG1500/silicone than PEG600/silicone can be related to the interfacial polarization of Maxwell–Wagner–Sillars type caused by trapping of free charges at the boundaries between the crystalline structure and the amorphous regions [29].

For the PEG1500/silicone composites, a strong frequency dispersion of permittivity was observed in the low-frequency region followed by a nearly frequency-independent behaviour above 10 kHz. The coexistence of amorphous and crystalline regions in the polymer is the main cause of frequency dependence of conductivity and permittivity, as has been demonstrated by Dygas *et al* [30]. As the frequency increases, the time available for dipole polarization at the phase boundary is reduced, which is the main reason for the decrease in the dielectric constant with the increase in frequency. In addition, the movement of PEG chain unit (CH_2CH_2O) is restricted at room temperature, which can also lead to a decrease in the dielectric constant with the frequency increasing. On the other hand, the dielectric spectra of the PEG600/silicone composites just show slight frequency dependence. This can be for the following two reasons. First, PEG600 is amorphous, thus it is homogeneous in the composite. Second, PEG600 is of high chain mobility at room temperature due to the lower glass transition temperature (about 20 °C). Therefore, the dielectric constant of PEG600 is less frequency dependent.

Figure 4 shows the loss tangent of the PEG/silicone composite films versus frequency for different loading fractions at room temperature. The reason for the increase in loss tangent of both PEG600/silicone and PEG1500/silicone composites is that local current occurs in the process of dipole polarization, and the local current increases with increasing PEG content. Figure 5 shows that the ac resistivity of the PEG/silicone composites decreases from 10^{11} to 10^9 Ω m. The decrease in resistivity is in accordance with the variation of the loss tangent. The loss tangent of PEG600/silicone is frequency independent, and this is because the dipole polarization rate is fast and the time available for dielectric relaxation is enough. For the PEG1500/silicone composite, the variation trend of loss tangent is similar to that of the dielectric constant and the mechanism is the same.

3.3. *Effect of PEG on the mechanical properties of silicone elastomers*

In order to evaluate the effect of blending on the actual mechanical properties of the resulting elastomers, the materials were mechanically tested, by applying increasing uniaxial elongations up to sample breaking. The variation of Young's modulus observed can give a better insight into the filler–polymer matrix interaction and the properties are also important for the DEA application. Figure 7 shows the Young's modulus of composites with varying PEG contents. Two opposite trends of the Young's modulus versus PEG contents were obtained, the modulus of PEG600/silicone

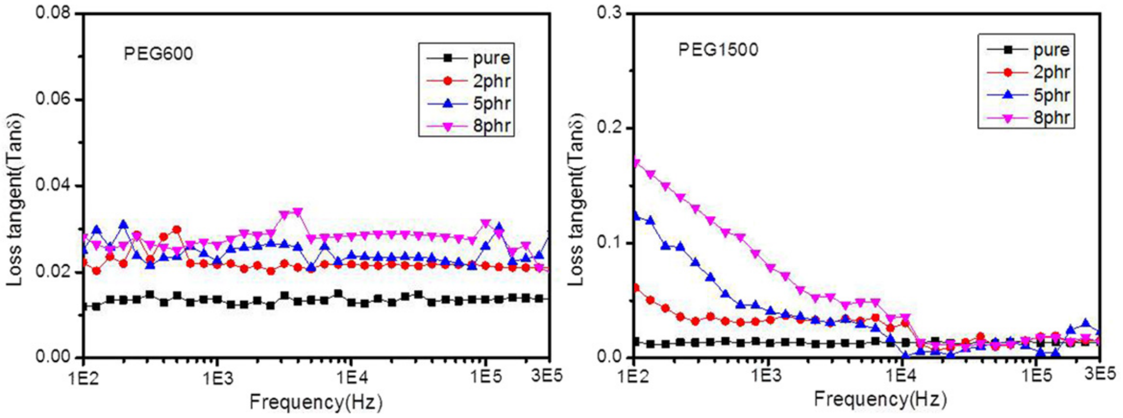

Figure 4. Loss tangent of pure silicone and silicone/PEG composites.

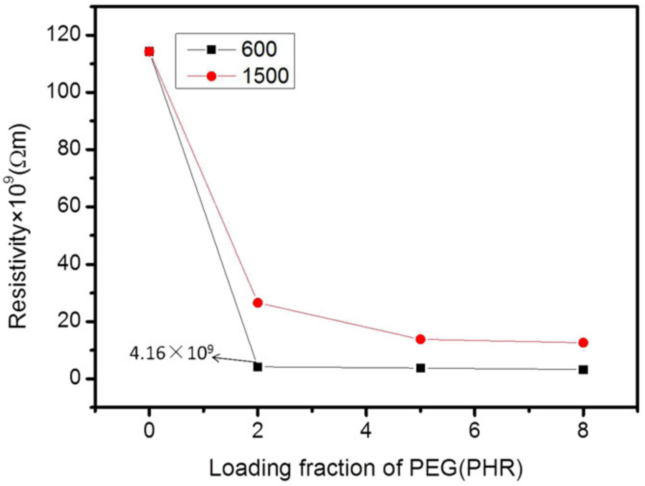

Figure 5. Ac resistivity of pure silicone and silicone/PEG composites.

composite decreases with loading fraction increase, while modulus of PEG1500/silicone composite shows an increasing trend. Figure 6 shows the crosslink density of the PEG/silicone composites. The crosslinking of silicone was affected by PEG to some extent, so its crosslinking density was lowered [31]. At the same time, PEG with low molecular weight is usually used as a plasticizer [32]; it would introduce a weak van der Waals force and increase the molecular chain spacing, resulting in lower Young's modulus. For the PEG1500/silicone composites, though its crosslinking density was lowered, the semi-crystalline structure would reinforce the silicone matrix strongly, and the increased modulus was due to the compromise between the lowered crosslinking density and the effect of reinforcement.

Figure 8 shows the dynamic mechanical properties of silicone and PEG/silicone composites. The dynamic modulus of the composites owes to the interactions between fillers, the effect of the filler–polymer interface, and the effect of polymer chain entanglement. In particular, the filler network contributes to the dynamic modulus more than the latter two factors [33]. The dynamic modulus begins to decrease obviously where the filler network is damaged as the strain increases, which is called the Payne effect [33].

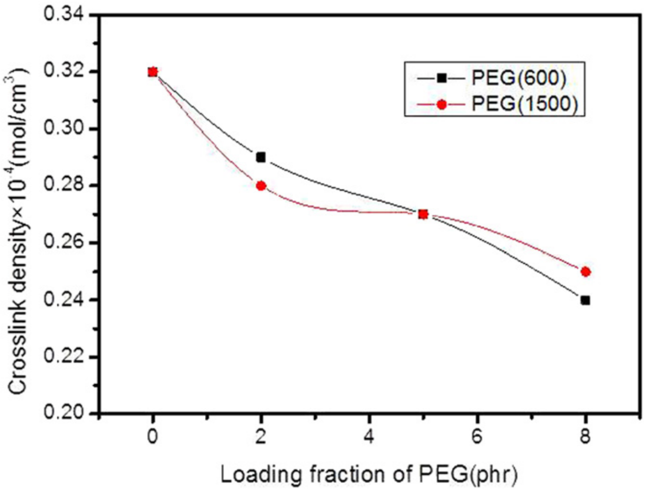

Figure 6. Crosslinking density of pure silicone and silicone/PEG composites.

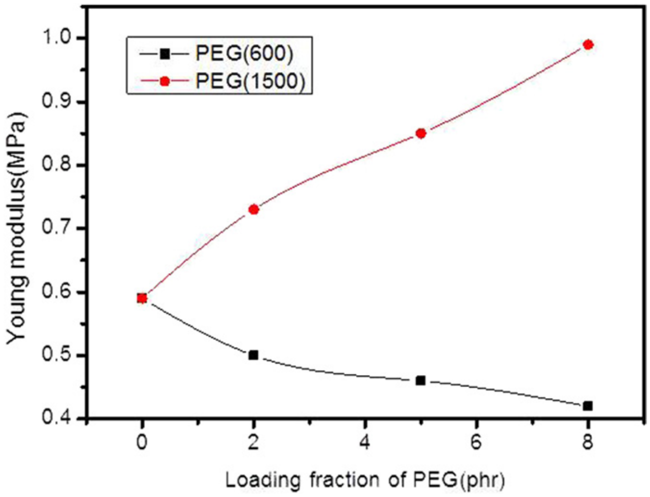

Figure 7. Young's modulus of pure silicone and silicone/PEG composites.

We can see from figure 8 that both the storage modulus and the loss modulus of the PEG1500/silicone composite are the highest among the three types of silicone films, while the storage modulus and the loss modulus of the PEG600/silicone

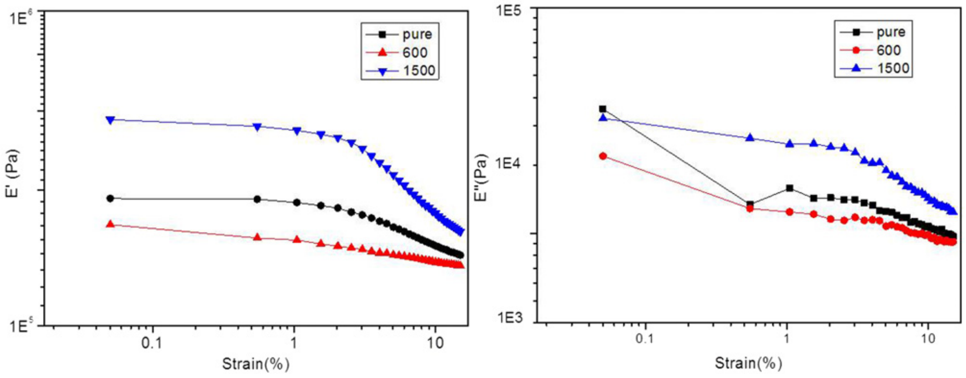

Figure 8. Dynamic mechanical properties of silicone and PEG/silicone composites.

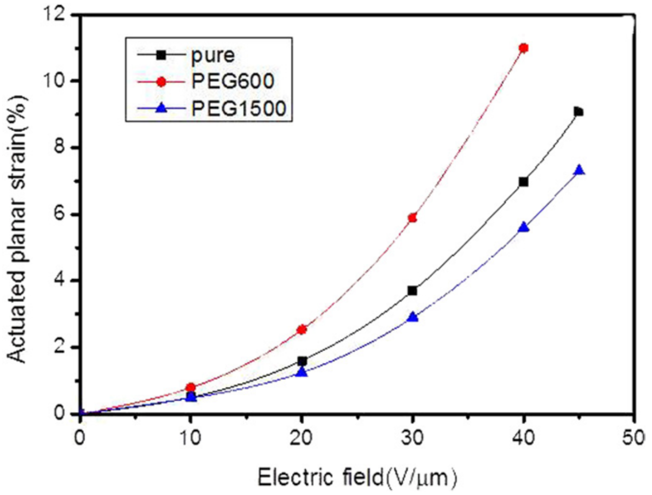

Figure 9. Actuated strain of pure silicone and silicone/PEG composites with 5 phr PEG.

composite are the lowest. It is suggested that PEG1500 was not dispersed in single molecule form and formed a filler network because of the direct contact of PEG particles, which will be completely damaged until the dynamic modulus decreases sharply at a strain of 3%. The other reason for the sharp decrease may be the damage of the semi-crystalline structure. PEG600 is a liquid in the experimental temperature range, and it has good plasticization, resulting in the decrease in both the storage modulus and the loss modulus.

Mechanical loss is the resultant of viscoelastic hysteresis. The mechanical loss, originated from the friction of polymer chains, filler–filler and filler–polymer chains, would be transformed into thermal energy finally, resulting in the breakdown of the DE films.

3.4. *Effect of PEG on the actuation properties of silicone elastomers*

In order to identify whether PEG can provide good improvement of actuation properties, a circular strain test on silicone dispersed with 5 phr PEG600 and PEG1500 was performed separately. The materials' actuation performance as DEA was measured demonstrating their suitability for dielectric applications, and the results are plotted in figure 9.

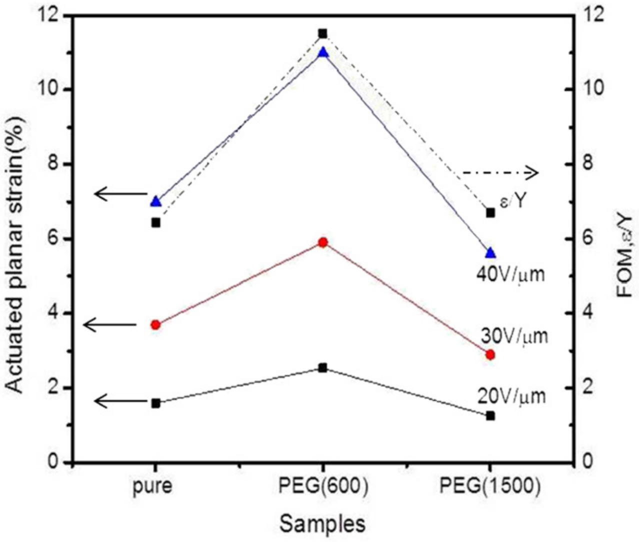

Figure 10. Comparison between the variation trend of the FOM (ε/Y) and the actuated planar strain.

The silicone dispersed with 5 phr PEG600 composite shows much larger actuated strain than pure silicone, while the actuation strain of silicone with 5 phr PEG1500 composite is lower than the pure silicone. And it is in accordance with the FOM (ε/Y) shown in figure 10. The reason for the lower actuation strain of the PEG1500/silicone composite is that the FOM is a little lower than that of pure silicone, at the same time, the dielectric loss is almost 18 times larger than that of pure silicone and the mechanical loss is also much larger than that of pure silicone. Next, we discuss the influence of loading fraction of PEG600 on the actuation property of silicone composite films.

3.5. *Effect of filling contents of PEG600 on the properties of DEs*

Planar actuators were made from pure silicone and the composites with 2, 5 and 8 phr PEG600. In order to identify the influence of loading fraction of PEG600 on the actuation properties of silicone composite films, silicone films filled with 2, 5 and 8 phr PEG600 were tested. It is clearly shown in figure 11 that the actuated planar strain steadily increases with

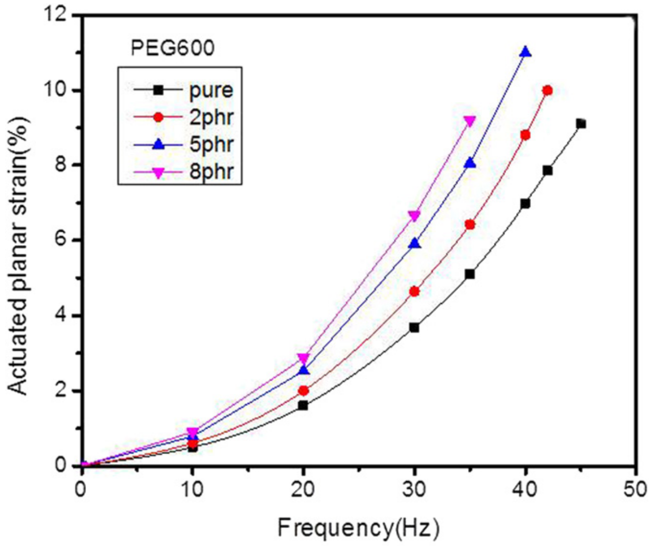

Figure 11. Actuated strain of pure silicone and silicone/PEG600 composites with 2, 5 and 8 phr PEG600.

the addition of PEG600 at the same electric field. This is because the modulus decreases with the addition of PEG600, while the dielectric constant increases, resulting in an increase in FOM with the addition of PEG600. A strain of 11.5%, which is 64% larger than that of pure silicone, was obtained from silicone filled with 5 phr PEG600 at 40 V μm^{-1}.

From figure 11 we can see that the electric breakdown strength decreases gradually with the addition of PEG600. The dielectric loss increases and the resistivity decreases with the addition of PEG600. The larger dielectric loss will be transformed to more thermal energy during the actuation test process; at the same time, the lower resistivity results in a stronger local current, during the actuation test process, which will also produce more thermal energy. So the electric breakdown strength decreases.

From the introduction we know that, for low strains (e.g. <20%), the thickness strain S_z can be approximated by

$$S_z = -\frac{P}{Y} = -\frac{\varepsilon_0 \varepsilon_r E^2}{Y}. \tag{3}$$

Since the polymer is typically nearly incompressible, the total volume is constant. Thus, the following equation can be obtained:

$$(1 + S_z)(1 + S_p) = 1, \tag{4}$$

where S_z is the thickness strain and S_p is the planar strain.

Therefore, based on equations (3) and (4), the planar strain S_p can be derived as follows:

$$S_p = \frac{1}{1 + S_z} - 1 = \frac{\varepsilon_0 \varepsilon_r E^2}{Y - \varepsilon_0 \varepsilon_r E^2}, \tag{5}$$

where P is the electrostatic pressure, and ε_0 and ε_r are the free-space permittivity and the relative dielectric constant of the elastomer, respectively. E denotes the electric field strength. The theoretical planar strain can be calculated according to equation (5).

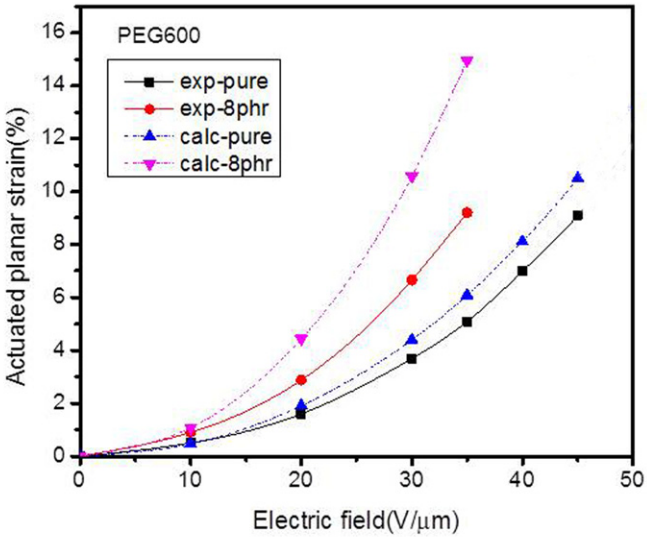

Figure 12. Comparison between the calculated strain and the experimental value.

Table 1. Temperature of pure silicone and its composites before and after the electromechanical test.

Sample surface temperature	T (°C) Before test	T (°C) 15 V μm^{-1}	T (°C) 30 V μm^{-1}
Pure silicone	27.6	28.9	31.2
8 phr PEG600/silicone	27.6	31.5	36.4

Figure 12 shows a comparison between theoretical and experimental actuated strains of pure silicone and silicone blended with 8 phr PEG600. We find some interesting phenomena. First, there is an obvious difference between the theoretical and experimental actuated strains for both pure silicone and PEG600/silicone composites. This could be ascribed to the fact that a small part of electric energy is converted to thermal energy through mechanical loss and dielectric loss during the process of actuation. In addition, wrinkling is induced with the increase in strain due to the in-plane compressive stresses caused by the constrainment of the DE on the whole edge to a rigid frame [34–36]. Second, the difference for the PEG600/silicone composite is higher than that of pure silicone. The reason is that the dielectric loss of the PEG600/silicone composite is three times higher than that of pure silicone, thus more electric energy was transformed to thermal energy in the PEG600/silicone composite during the actuation process. And this could be confirmed by the fact that the temperature of the PEG600/silicone composite film was several degrees higher than that of pure silicone after the actuation strain test, as shown in table 1. Third, the difference between theoretical and experimental strains increases with the enhancement of electric field. This is ascribed to the fact that the dielectric loss increases with the enhancement of electric field, and the mechanical loss increases with the actuated strain increasing, which leads to more energy transformation from electric energy to thermal energy. And this can also be demonstrated by the temperature test: the higher the electric field, the higher the temperature. In addition, wrinkling is another important factor for the increase in such kind of difference [34–36].

DEA is to some extent like a capacitor, so it can absorb polarization charge and store energy in the electric field, and the amount of polarization charge depends on the dielectric constant. Part of the electric energy is transformed to thermal energy through dielectric loss and mechanical loss, which would lead to a temperature rise in the elastomer and would eventually cause a breakdown. In the meantime, the DE expands in the planar direction because of the electrostatic stress, and the strain depends on the modulus of the polymer. As the strain increases, the DE begins to buckle and it makes the actuated strain less than the theoretical value. In addition, in-plane compressive stresses may arise, which could lead to wrinkling instabilities. On the other hand, the polymeric film could be prestretched, which could favour pull-in-type instabilities [34–36]. For the PEG600/silicone composite, the dielectric constant is relatively high, but the modulus and mechanical loss are the lowest among the three types of composites, therefore it has the highest actuated strain. Because of conservation of energy, the actuated strain of DE is related to the dielectric constant, elastic modulus, mechanical loss and dielectric loss. Under the condition of high dielectric constant and low modulus, mechanical loss and dielectric loss are the other two important factors that also have a significant effect on the actuated strain.

4. Conclusion

A silicone dielectric elastomer dispersed with polyethylene glycol with two different average molecular weights was successfully prepared. The major conclusions are as follows. (1) The dielectric constant and loss increase with the addition of PEG that has high polarity, while the volume resistivity and the breakdown strength decrease. (2) The dielectric and mechanical properties of PEG600/silicone and PEG1500/silicone composites are different. Compared with PEG1500/silicone composites, the addition of PEG600 into a silicone matrix resulted in a higher dielectric constant but lower Young's modulus and dielectric and mechanical loss, and thus can effectively improve the electromechanical properties of silicone.

By comparing the results between the theoretical strain and the experimental data, it is found that not only the ratio of dielectric to modulus but also the dielectric loss and mechanical loss have an obvious effect on the actuation strain. Compared with pure silicone, the actuated strain of the PEG600/silicone composite film is much larger, but the actuated strain of the PEG1500/silicone composite is much lower. It is suggested that, as for doped dielectric elastomer composites, the stronger interfacial polarization could improve the dielectric constant, but it could also lead to an increase in modulus. Therefore, the dispersion of PEG should be improved to obtain a composite with high dielectric constant but with low modulus.

Acknowledgments

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