Composites Part B 78 (2015) 35–41

Contents lists available at ScienceDirect

Composites Part B

journal homepage: www.elsevier.com/locate/compositesb

Stability of dielectric elastomer/carbon nanotube composites coupling electrostriction and polarization

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ARTICLE INFO

Article history:
Received 25 June 2014
Received in revised form
27 February 2015
Accepted 21 March 2015
Available online 28 March 2015

Keywords:
C. Analytical modelling
C. Numerical analysis
B. Electrical properties
Electromechanical stability

ABSTRACT

Experiments are conducted to test the permittivity of dielectric elastomer composites adulterated with multi-walled carbon nanotube (MWCNT). The results show that the permittivity of dielectric elastomer composites can be significantly improved by adding MWCNT conductive particles. A thermodynamic model is presented to investigate the stability of MWCNT particle-doped dielectric elastomer composites. The theoretical investigation proves that the polarization of MWCNT, the electrostriction deformation and material constants of the elastomer significantly affect the stability of the thermodynamic system. The numerical analysis shows that comparing to ideal dielectric elastomer, the stability of dielectric elastomer composites filled with MWCNT coupling between electrostriction and polarization can be significantly enhanced.

1. Introduction

Dielectric elastomers have been intensely studied in recent years [1–5]. Dielectric elastomer film can be sandwiched between two compliant electrodes to form a capacitor [3,4]. When a voltage is applied through the electrodes, the induced charge causes an electrostatic attraction between two electrodes. The resulting compressive force leads to a reduction in the film thickness, and an elongation in the film plane. The potential applications include medical devices, soft robots, energy harvesters, optical devices, etc [3,4].

Dielectric elastomers usually fail because of the onset of electromechanical instability, snap-through instability and electrical breakdown [4–6]. Electromechanical instability behaviors are resulted from exorbitant driving voltage imposed on dielectric elastomers, relatively low shear modulus and so on [7–21]. To suppress and eliminate electromechanical instability, researchers have been applying various approaches to lower the driving voltage and improve the stability of dielectric elastomer [22–25].

The driving voltage of dielectric elastomers can be reduced by preparing dielectric elastomer composites with high permittivity [22,23]. There are generally two ways of achieving such material modification. The first is to fill dielectric elastomers with ferroelectric ceramic particles, such as barium titanate and lead magnesium niobate–lead titanate (PMN-PT), which possess high permittivity. As increase in the content of filler particles, both permittivity and elastic modulus of the composites increases. Such composites require relatively low driving voltage and exhibit higher driving force than conventional dielectric elastomers under the same driving voltage [22]. The second way is to blend dielectric elastomers with conjugated polymers with high polarity, such as PHT [poly(3-hexylthiophene)] [23]. Comparing to the particle-doped composites, conjugated polymers blended composites possess improved permittivity but lower elastic modulus.

For enhancing the stability of dielectric elastomers, researchers mainly adopt the following methods in their researches: employing dielectric elastomers with high modulus and low permittivity [7], pre-stretching dielectric elastomers [7,9,12], producing interpenetrating polymer network dielectric elastomers [24], charge

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http://dx.doi.org/10.1016/j.compositesb.2015.03.069
1359-8368/

control dielectric elastomers [25], taking the advantage of polarization saturation effect [19,21,26] and so on.

The experiments in this paper prove that the increasing content of multi-walled carbon nanotubes in dielectric elastomers can improve the composites' permittivity notably. An expression of permittivity is proposed for the dielectric elastomers filled with MWCNT particles. The stability of the dielectric elastomer composites coupling system is investigated through a thermodynamic model considering the deformation of elastomers and the polarization of conductive MWCNT particles. A noticeable improvement of stability compared to ideal dielectric elastomer is proved for dielectric elastomer composites filled with MWCNT coupling between electrostriction and polarization.

2. Experiment

The morphology of conductive particles has a great impact on the permittivity of the composite material. For example, the aspect ratio of the conductive particles could affect the permittivity of the composite material. The greater the aspect ratio is, the higher the permittivity is. Consequently, in this paper, we choose the carbon nanotube as conductive nano fillers to increase the permittivity of the dielectric elastomer composites.

The preparation process of the dielectric elastomer composites are described as follows. Multi-walled carbon nanotubes were purchased from Carbon Solutions Inc with average particle diameter of 40 nm and length of 0.5 μm. A commercial, three component, poly-dimethyl-siloxane (PDMS) based formulation by BJB Enterprises Inc., U.S.A. (manufacturer type TC-5005 A/B–C) was used as the matrix. Both the silicone and catalyst were degassed in a vacuum oven for one hour to remove trapped air bubbles and moisture. The multi-walled carbon nanotubes were well dispersed in the silicone base. The mixture was stirred at a rotational speed of 700 Revolutions Per Minute (RPM) for 15 min. Then the MWCNT-doped mixture was dispersed by ultrasonic for 10 min. Silicone dielectric elastomer/MWCNT films were put over aluminum substrates and were left drying in air at room temperature for 24 h. Then samples were prepared for experimental tests.

In order to limit the expected decrease of the dielectric strength of the composites with respect to that of the pure silicone, the following low percentages of carbon nanotube are explored: 1, 2, 3, 4, and 5 wt%. Fig. 1 presents room-temperature dielectric spectra for both the pure silicone and the composites in the range of 10^2–10^7 Hz. Both the relative permittivity and dielectric loss spectra monotonically decrease with frequency over the whole range explored. As expected, by increasing the carbon nanotube content a progressive increase of permittivity of the composites is achieved at all frequencies compared to pure silicone. In particular, at 10^2 Hz frequency the relative permittivity increases from 3.91 to 7.83. However, as a counterpart, this also corresponds to an analogous increase of the dielectric loss, up to a maximum value of 0.97 for the composite with 5 wt% of carbon nanotube. Experimental data showed that the permittivity of dielectric elastomer composites could be significantly improved by adding MWCNT particles.

3. Theory

Traditional percolation theory may not be adequate to explain this phenomenon. When the content of conductive particles is approaching the threshold, there is a drastically increase in the permittivity of the composites. However, in a dielectric elastomer carbon nanotube composite, this phenomenon does not occur. The reason is that the content of the carbon nanotube conductive particles in the composite is much lower than threshold.

We use polarization theory of the dielectrics to illustrate this mechanism. The polarization of dielectric elastomers can be attributable to two contributions: electric dipoles of each polymer chain and electric dipoles of conductive particles. An elastomer is a three-dimensional network of long and flexible polymer chains, held together by cross-links. In an elastomer, each polymer chain may consist of monomers of electric dipoles. Besides, numerous electric dipoles also exist in the conductive particles.

Polar distance of polarity molecule with permanent dipole, in the absence of external electric field, tends to be oriented randomly when experiencing thermal fluctuation. The average polar distance of the total molecule is zero. When the dielectrics are subjected to an electric field, the dipoles tend to turn as a result of the application of torques. In fact, with the increase of external electric field, the corresponding increased torques aligns the dipoles along the same direction as the external electric field. Eventually the dipoles are completely polarized under a sufficiently high electric field. The polarization charge induces an additional electric field, which is opposite to the external electric field. The rotation of the dipoles affects the polarization and consequently influences the permittivity of the material. Since the filled conductive particles can promote the rotation of the dipoles, they can enhance the polarization of the dielectrics. When the polarity conductive particles are introduced to the dielectric elastomer, the permittivity of the composites will increase.

To present the permittivity of the dielectric composite filled with conductive nano particles, the Clausius-Mossotti equation is applied considering the influence of polarity [22].

$$\varepsilon(P) = \frac{2P+1}{1-P}\varepsilon^{\sim} \tag{1}$$

Ideally uniform doping is assumed in the present paper. In Equation (1), $P = N\alpha/3\varepsilon_0$ is the polarizability of the composite medium sphere, which may describe the degree of polarization of the dielectric elastomer composites. N represents the number of molecules per unit volume of the dielectric elastomer composites. α denotes the polarizability of the particles and ε_0 is the vacuum permittivity. $\varepsilon^{\sim}$ is the relative permittivity of dielectric elastomer. It is appropriate to describe the influence of polarization on the permittivity of dielectric elastomer composites using the Clausius-Mossotti equation. In fact, Equation (1) predicts reinforcement of composites well when the uniformly filled particles have relative small volumes. As noticed in Equation (1), the permittivity of the composites increases with the content of the conductive particles. Of course, in the light of specific experimental features, other models, such as the Clausius-Mossotti-Doyle-Jacobs equation, can also be applied to characterize the effect of polarization on the permittivity of dielectric elastomer composites.

When the degree of cross-link is not low, or the deformation approaches the extension limit, the permittivity of the elastomer will be affected by the deformation. The dielectric elastomer composites will experience large deformation when subject to electric field [27]. The permittivity of the silicone dielectric elastomer composites is a function of the stretch, which has been proved experimentally and theoretically [26–28]. In recent experiments, Jean-Mistral and Sylvestre stretched the dielectric polymer film (VHB 4910, 3 M, USA) by a equal biaxial tension in the planar directions, they found that the permittivity is a function of the pre-stretch [28]. We fit their experimental data with the following function

$$\varepsilon(\lambda_1, \lambda_2) = \varepsilon^{\sim}(\alpha + \beta\lambda_1\lambda_2) \tag{2}$$

Where β is the coefficient of electrostriction and α is a phenomenological parameter, λ_1 and λ_2 are stretches of the dielectric elastomer in principal planar directions.