



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



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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.

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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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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].

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

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. ε^{\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

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

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.

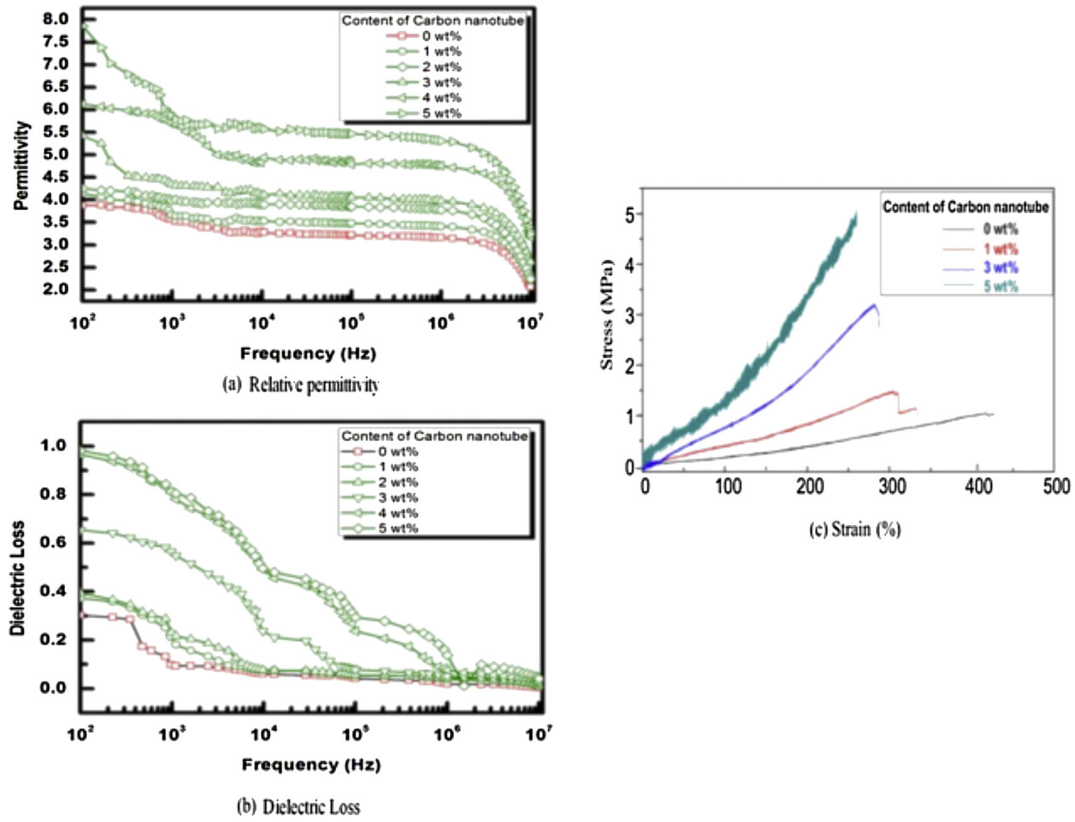


Fig. 1. Relative permittivity (a), dissipation factor (b) and stress-strain relation (c) for the silicone/MWCNTs composites at varying MWCNTs concentrations.

We know that a typical dielectric elastomer is a cross-linked polymer. Symmetry and cross-linking of dielectric elastomer molecular structure will affect its permittivity. The higher the symmetry is, the lower the permittivity is. The cross-linking shows a similar effect. The dielectric elastomer molecules are forced by pre-stretch to be arranged in order. The intermolecular forces increase simultaneously due to the increase of distances between molecules. As a result, the permittivity reduces due to the declination of the activity of polarity groups. And if the cross-links of the dielectric elastomer are at a relatively low degree and the pre-stretch is well below the extension limit, the molecular units of the polymers can be polarized as freely as in a polymeric liquid. In this case the corresponding permittivity is unaffected by the stretch [27].

Considering the coupling influence of the polarization effect of conductive particles and elastomer's deformation, we put forward the permittivity expression of the silicone dielectric elastomer composites filled with MWCNT conductive particles as follows

$$\varepsilon(\lambda_1, \lambda_2, P) = \varepsilon^{\sim} \left(\alpha + \beta \lambda_1 \lambda_2 - 1 + \frac{2P + 1}{1 - P} \right) \quad (3)$$

in which $\varepsilon^{\sim} = 3.91\varepsilon_0$ according to the aforementioned experiment, $\varepsilon_0 = 8.85 \times 10^{-12}$ F/m is the vacuum permittivity. When the polarization effect is neglected, $P=0$, Equation (3) can be reduced to Equation (2); When the dielectric elastomer remains undeformed, $\lambda_1 \lambda_2 = 1$, Equation (3) can be reduced to Equation (1), where $\alpha = 1$ and $\beta = 0$.

Fig. 1(c) shows the stress-strain curve of silicon rubber dielectric elastomer/MWCNT composites under stretching at room temperature. The mechanics performance testing of dielectric elastomer composite materials is conducted by the Zwick/roell Universal Material Testing Machine. From Fig. 1(c) we can see, as the increase

of the content of carbon nanotube, the modulus of elasticity of dielectric elastomer composite materials increase too. The stretching of pure silicon rubber dielectric elastomer is beyond 400%, while the stretching of composite material within 5 wt% carbon nanotube is beyond 250%. This experiment also proved that the dielectric elastomer composite material doping MWCNT still maintain good superelasticity.

To illustrate an electromechanical coupling system, the elastic strain energy function and the electric energy density function should be coupled in a free energy function. Assume the dielectric elastomer to be incompressible, the free energy function can be written as follows [7–21]:

$$W(\lambda_1, \lambda_2, \nu, D^{\sim}) = \xi(\lambda_1, \lambda_2, \nu) + \psi(\lambda_1, \lambda_2, \nu, D^{\sim}) \quad (4)$$

where $W(\lambda_1, \lambda_2, \nu, D^{\sim})$ denotes the free energy function of dielectric elastomer electromechanical coupling system, $\xi(\lambda_1, \lambda_2, \nu)$ represents the elastic strain energy function, $\psi(\lambda_1, \lambda_2, \nu, D^{\sim})$ reveals the electric energy density function, ν denotes the weight percentage of filled particles, and D^{\sim} denotes the nominal electric displacement.

It is assumed by Mooney and Rivlin that the elastomer was isotropic and incompressible. In their model, therefore, elastic strain energy is only related to stretch at both directions. It has been experimentally validated that this model is effective in predicting the stress-strain behavior for strain less than 200% [29]. The present paper chooses Mooney-Rivlin type elastic strain energy model with two material constant, as the elastic strain energy, $\xi(\lambda_1, \lambda_2, \nu)$.

The elastomer is regarded as a cross-linked network of long and flexible polymer, which follows the Gaussian statistics. Each polymer chain is composed of numerous monomers. Here researchers assume liquid-like dielectric behavior of the elastomer, which means it is unaffected by the deformation. The dielectric energy per

unit volume can be expressed as $\frac{D^{\sim 2}}{2\epsilon^{\sim}}\lambda_1^{-2}\lambda_2^{-2}$, while the permittivity, ϵ^{\sim} , can be a constant.

The coupling effect of polarization and deformation is considered in the present work. In accordance with Equations (3) and (4), the free energy of the thermodynamic system of dielectric elastomers filled with MWCNT conductive particles can be written as

$$W(\lambda_1, \lambda_2, \nu, D^{\sim}) = \frac{C_1(\nu)}{2}(\lambda_1^2 + \lambda_2^2 + \lambda_1^{-2}\lambda_2^{-2} - 3) + \frac{C_2(\nu)}{2}(\lambda_1^{-2} + \lambda_2^{-2} + \lambda_1^2\lambda_2^2 - 3) + \frac{D^{\sim 2}}{2\left(\alpha + \beta\lambda_1\lambda_2 + \frac{2P+1}{1-P}\right)\epsilon^{\sim}}\lambda_1^{-2}\lambda_2^{-2} \quad (5)$$

Here the free energy density is the function of stretches and nominal electric displacement. The nominal stresses and nominal electric field can be written as the partial differential coefficients

$$S_1 = C_1(\nu)\left(\lambda_1 - \lambda_1^{-3}\lambda_2^{-2}\right) + C_2(\nu)\left(\lambda_2\lambda_1^2 - \lambda_1^{-3}\right) - \frac{D^{\sim 2}\lambda_1^{-3}\lambda_2^{-2}\left\{\epsilon^{\sim}\left[3\beta\lambda_1\lambda_2 + 2(\alpha - 1) + \frac{2P+1}{1-P}\right]\right\}}{2\left[\epsilon^{\sim}\left(\alpha + \beta\lambda_1\lambda_2 - 1 + \frac{2P+1}{1-P}\right)\right]^2} \quad (6)$$

$$S_1 = C_1(\nu)\left(\lambda_2 - \lambda_1^{-2}\lambda_2^{-3}\right) + C_2(\nu)\left(\lambda_2\lambda_1^2 - \lambda_2^{-3}\right) - \frac{D^{\sim 2}\lambda_1^{-2}\lambda_2^{-3}\left\{\epsilon^{\sim}\left[3\beta\lambda_1\lambda_2 + 2(\alpha - 1) + \frac{2P+1}{1-P}\right]\right\}}{2\left[\epsilon^{\sim}\left(\alpha + \beta\lambda_1\lambda_2 - 1 + \frac{2P+1}{1-P}\right)\right]^2} \quad (7)$$

$$E^{\sim} = \frac{D^{\sim}}{\epsilon^{\sim}\left(\alpha + \beta\lambda_1\lambda_2 - 1 + \frac{2P+1}{1-P}\right)}\lambda_1^{-2}\lambda_2^{-2} \quad (8)$$

When the dielectric elastomer composites is subjected to the voltage and mechanical load, the dielectric elastomer composites film would decrease in thickness and expand in area. The decrease in thickness will induce a higher electric field. This positive feedback may cause the dielectric elastomer composites film thins down drastically. When the electric field exceeds the critical value, the dielectric elastomer composites breaks down and the actuator becomes invalid. This process is referred to as electromechanical instability.

In order to investigate the electromechanical instability of dielectric elastomer composites, we consider a special loading case. It is assumed that the dielectric elastomer film is uniformly stretched $S_1 = S_2 = S$ and $\lambda_1 = \lambda_2 = \lambda$. We postulate that $C_1(\nu) = kC_2(\nu)$ where k is material constant ratio [9,11].

According to Equations (6)–(8), the nominal electric field and nominal electric displacement can be written as follows:

$$\frac{D^{\sim}}{\sqrt{C_1(\nu)\epsilon^{\sim}}} = \sqrt{\left[\left(\lambda^6 - 1\right) + \frac{1}{k}\left(\lambda^8 - \lambda^2\right) - \frac{S\lambda^5}{C_1(\nu)}\right]M} \quad (9)$$

$$\frac{E^{\sim}}{\sqrt{C_1(\nu)/\epsilon^{\sim}}} = \sqrt{\frac{2\left[\left(\lambda^{-2} - \lambda^{-8}\right) + \frac{1}{k}\left(1 - \lambda^{-6}\right) - \frac{S\lambda^{-3}}{C_1(\nu)}\right]}{3\beta\lambda^2 + 2\left[(\alpha - 1) + \frac{2P+1}{(1-P)}\right]}} \quad (10)$$

$$\text{where } M = \frac{2\left[\epsilon^{\sim}\left(\alpha + \beta\lambda^2 - 1 + \frac{2P+1}{1-P}\right)\right]^2}{3\epsilon^{\sim 2}\beta\lambda^2 + 2\epsilon^{\sim 2}(\alpha - 1) + \frac{(2P+1)\epsilon^{\sim 2}}{1-P}}$$

4. Results and discussion

As shown in Fig. 1(a), the mass fraction of the MWCNT have values of 0, 1, 3, and 5 wt% and the permittivity of the dielectric elastomer composite is 3.91, 4.18, 5.48 and 7.83. According to Equation (1), $P = 0, 0.023, 0.118, \text{ and } 0.251$.

According to the tests of acrylic acid's permittivity at temperatures of 273, 293 and 313 K which was conducted by Jean-Mistral *et al* [28], the corresponding phenomenological parameters, α s, and electrostriction coefficient, β s, under the three conditions are calculated as $\alpha = 1.02, \beta = -0.01$; $\alpha = 1, \beta = -0.0097$; $\alpha = 0.95, \beta = -0.0094$.

Fig. 2 depicts the influence of electrostriction on the stability of dielectric elastomer composites. In Fig. 2(a), $\alpha = \beta = 0$ and $P = 1$. Equations (9) and (10) degenerate into governing equations of stability of ideal Mooney-Rivlin-type pure dielectric elastomers. A conclusion can be reached through comparing Fig. 2(a)–(d). After experiencing their summits, the curves in Fig. 2(a) descend at first and then tend to be steady, which means the elastomers can undergo the electromechanical instability. The curves in Fig. 2(b)–(d), by contrast, exhibit snap-through (represented by the green line in Fig. 2(b)) and continue ascending after passing their acmes. This phenomenon results from electrostriction and polarization and indicates that the elastomers can bear snap-through instability. The comparison above reveals that compared to ideal dielectric elastomers, dielectric elastomer composites with coupling between deformation and polarization can exhibit noticeably improved stability.

Fig. 3 reveals the effect of polarization of MWCNT on the stability of the dielectric elastomer composites for $\alpha = 1$ and $\beta = -0.0097$. The critical nominal electric field decreases monotonically as the polarization increases. The so-called critical nominal electric field denotes the nominal electric field under which the dielectric elastomer composites are in the critical state switching from electromechanically stable state to instable state. Obviously, addition of conductive particles can decrease dielectric strength of dielectric elastomers. It is also indicated that adding multiple walled carbon nanotubes can lower the breakdown electric field of the electric elastomer composites.

As shown in Fig. 1(c), based on Mooney-Rivlin model (the first two items of Equation (5)) the calculated material parameters of dielectric composite materials with different content of MWCNT are as follows: $C_1(0) = 0.049$ MPa, $C_2(0) = 0.129$ MPa; $C_1(1\%) = 0.045$ MPa, $C_2(1\%) = 0.41$ MPa; $C_1(3\%) = 0.057$ MPa, $C_2(3\%) = 0.96$ MPa; $C_1(5\%) = 0.45$ MPa, $C_2(5\%) = 0.74$ MPa. Therefore, we obtain the parameter k of 0.059, 0.11, 0.38, 0.61 to do the stability analysis of dielectric elastomer composite materials.

In Fig. 4, the relations between the nominal electric displacement and the nominal electric field in the dielectric elastomer composites with various values of k and S/C_1 are plotted. Simultaneously illustrate the stability performance of the dielectric elastomer composites under equal biaxial stretch, $\lambda_1 = \lambda_2 = \lambda$. Evidently, with k or S/C_1 decreasing, the peaks of the nominal electric field increase and the electromechanical stability is improved. Furthermore, when k is relatively small, the local maximal nominal electric field of the dielectric elastomer composite materials will disappear and the curve will rise monotonously. If k is relatively large, the dielectric elastomer composite material will undergo snap-through instability.

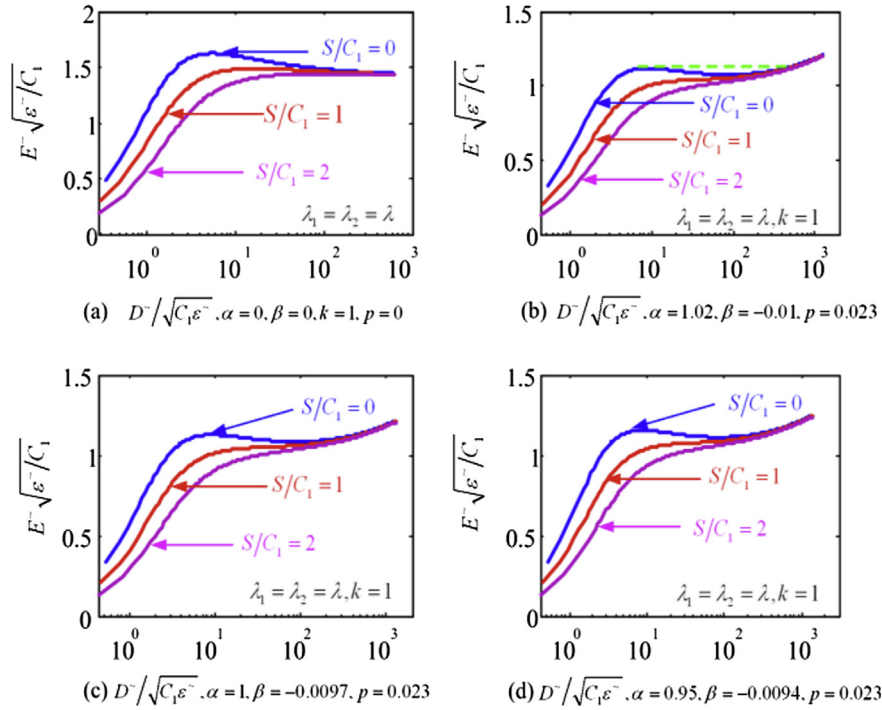


Fig. 2. Relationship between the nominal electric displacement and the nominal electric field of dielectric elastomers composites with various values of $\beta = 0, -0.094, -0.097, -0.01$, while $\lambda_1 = \lambda_2 = \lambda, k = 1$, and $P = 0, 0.023$.

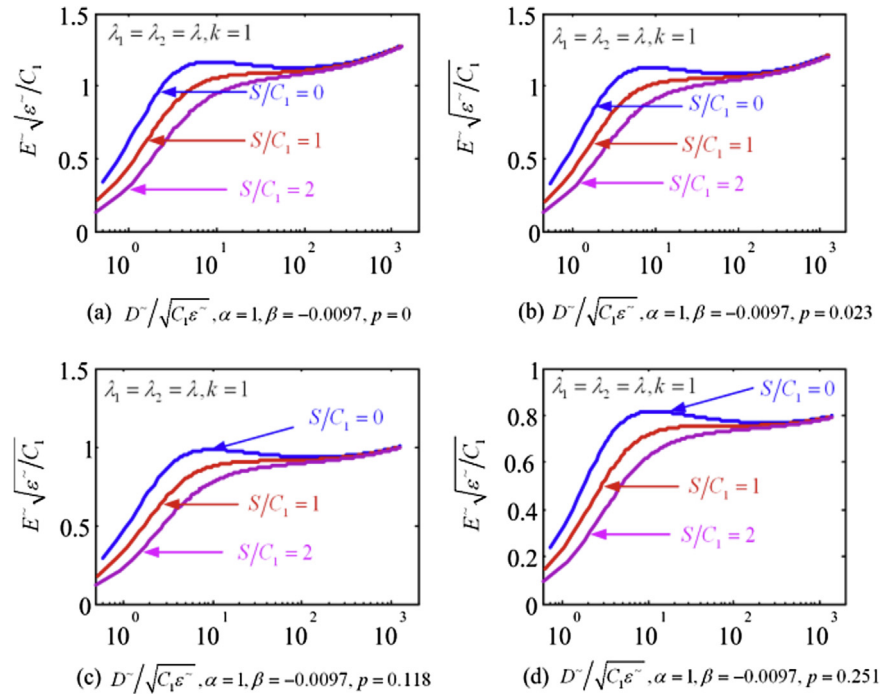


Fig. 3. Relationship between the nominal electric displacement and the nominal electric field of dielectric elastomers composites with various values of $P = 0, 0.023, 0.118, 0.251$, while $\lambda_1 = \lambda_2 = \lambda, k = 1$, and $\beta = -0.097$.

Zhigang Suo and Jian Zhu proposed the dielectric elastomer critical breakdown electric field is on the order $\sqrt{\frac{\mu}{\epsilon}}$ where μ and ϵ is the shear modulus and permittivity of dielectric elastomer [20]. In this paper, the parameters of pure dielectric elastomer are $C_1(0\%) = 0.049$ MPa, $\epsilon(0\%) = 3.91\epsilon_0$, $\epsilon_0 = 8.85 \times 10^{-12}$ F/m and those

of dielectric elastomer with 5% multi-walled carbon nanotube are $C_1(5\%) = 0.45$ MPa, $\epsilon(5\%) = 7.83\epsilon_0$. We can obtain that $\sqrt{\frac{C_1(0\%)}{\epsilon}} = \sqrt{\frac{0.049 \times 10^6}{3.91 \times 8.85 \times 10^{-12}}} = 0.38 \times 10^8$ V/m for pure dielectric elastomer and $\sqrt{\frac{C_1(5\%)}{\epsilon}} = \sqrt{\frac{0.45 \times 10^6}{7.83 \times 8.85 \times 10^{-12}}} = 0.81 \times 10^8$ V/m for

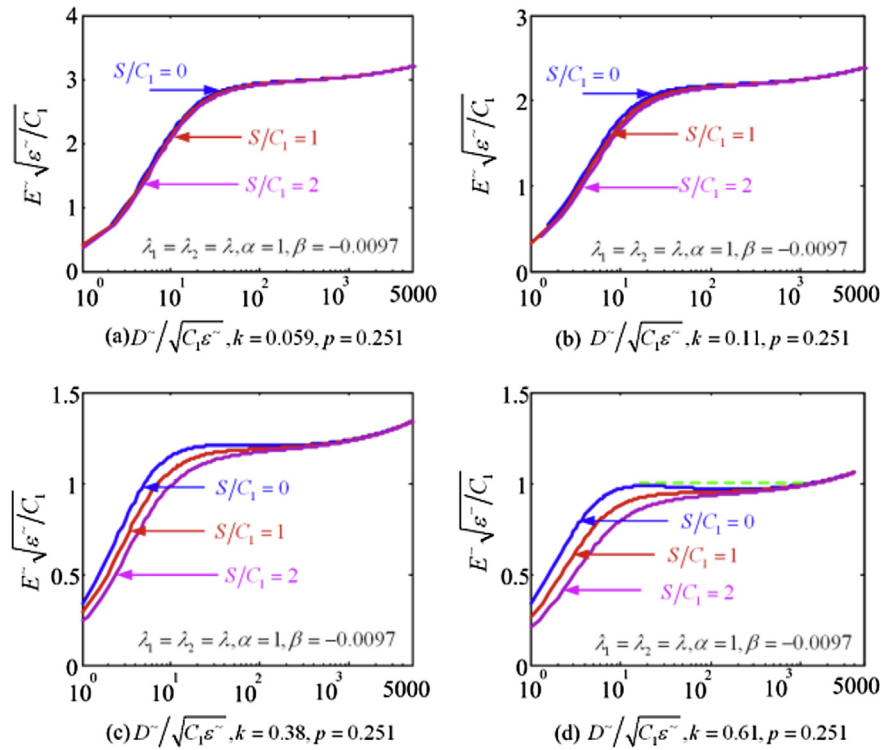


Fig. 4. Relationship between the nominal electric displacement and the nominal electric field of dielectric elastomers composites with various values of $k = 0.059, 0.11, 0.38, 0.61$, while $\lambda_1 = \lambda_2 = \lambda$, $P = 0.251$, and $\beta = -0.097$.

dielectric elastomer-MTCNW composites. It is obvious that the critical breakdown electric field of dielectric elastomer composite is higher than that of pure dielectric elastomer. 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.

5. Conclusions

In conclusion, we experimentally investigated the permittivity of dielectric elastomer composites doped with multiple walled carbon nanotube. The experiment results show that the permittivity of dielectric elastomer composites can be significantly improved by adding of MWCNT particles. Base on experiments, we developed a thermodynamic model coupling electrostriction with polarization to investigate the stability of MWCNT particle-doped dielectric elastomer composites. We prove that polarization of MWCNT, the electrostriction deformation and the material constants of elastomer significantly affect the stability of the dielectric elastomer composites. The numerical analysis shows that the coupling between deformation and polarization can significantly improve the stability of dielectric elastomer composites over that of conventional dielectric elastomers. The analyses are beneficial for further understanding the effects of conductive multiple walled carbon nanotube particles on the stability of dielectric elastomers. Experiment and numerical results may guide the design and manufacture of high-performance dielectric elastomer transducers.

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