Electromechanical stability of electro-active silicone filled with high permittivity particles undergoing large deformation

2010 Smart Mater. Struct. 19 115025

(http://iopscience.iop.org/0964-1726/19/11/115025)

Smart Mater. Struct. 19 (2010) 115025 (11pp) doi:10.1088/0964-1726/19/11/115025

Electromechanical stability of electro-active silicone filled with high permittivity particles undergoing large deformation

Liwu Liu[1], Yanju Liu[1,4], Zhen Zhang[2], Bo Li[3] and Jinsong Leng[2,4]

[1] Department of Astronautical Science and Mechanics, Harbin Institute of Technology (HIT), PO Box 301, No. 92 West Dazhi Street, Harbin 150001, People's Republic of China
[2] Centre for Composite Materials, Science Park of Harbin Institute of Technology (HIT), PO Box 3011, No. 2 YiKuang Street, Harbin 150080, People's Republic of China
[3] School of Mechanical Engineering, Xi'an Jiaotong University, No. 29 Xianning West Road, Xi'an 710049, People's Republic of China

E-mail: yj_liu@hit.edu.cn and lengjs@hit.edu.cn

Received 14 July 2010, in final form 6 September 2010
Published 5 October 2010
Online at stacks.iop.org/SMS/19/115025

Abstract
In this paper, an expression for the permittivity of electro-active silicone undergoing large deformation with high permittivity particles filled uniformly has been proposed. Two expressions are proposed for the permittivity, one based on experimental tests and the other based on the theory of composite material. By applying the thermodynamic model incorporating linear dielectric permittivity and nonlinear hyperelastic performance, the mechanical performance and electromechanical stability of the coupling system constituted by silicone filled with PMN–PT have been studied. The results show that the critical electric field decreases, namely the stability performance of the system declines when the content of PMN–PT $c(v)$ increases and the electrostrictive coefficients increase. The results are beneficial for us to understand deeply the influence of the filled particle on the stability performance of silicone and to guide the design and manufacture of actuators and sensors based on dielectric elastomers.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Due to its excellent properties such as large deformation, high elastic energy density, high efficiency, high responsive speed and long fatigue lifespan, silicone dielectric elastomer is widely used in fabricating actuators, sensors and energy harvesters [1, 2]. To gain large deformation, high voltages should be exerted on silicone, however, such a voltage may approach the elastomer's breakdown level. When doped with particles with high dielectric permittivity, the elastomer's permittivity would be increased [3, 36]. At the same time, the filled particles can also improve the mechanical performance of the elastomer, which implies that the driving voltage can be decreased while the Young's modulus can be increased. Therefore, actuators based on this type of composite elastomer will have advantages of lower driven voltage and high actuating force, as compared to the regular silicone dielectric elastomer material.

The electromechanical stability of dielectric elastomer is one of the most popular issues in the field of large electrostrictive deformation [4–13, 15–34, 37–40]. In [4, 5], Zhao and Suo proposed that any free energy functions could be applied to analyze the stability of the dielectric elastomer electromechanical coupling system. In our previous study, the electromechanical stability of dielectric elastomers was analyzed by applying the elastic strain energy with two material constants, which is great progress in analyzing

[4] Authors to whom any correspondence should be addressed.

the stability of a wide variety of dielectric elastomers by introducing the material constant ratio k [9]. Díaz-Calleja's group [10] and our group [11] have carried out further research on the prediction of the stability domain of neo-Hookean type silicone and Mooney–Rivlin type silicone. Recently, we employed analytical methods to study its stability and analytical expressions of the stability parameters were derived by applying different free energy functions [12]. According to the definition of stability parameters, we provided a kind of coherent criterion to evaluate the stability performance. However, all the research mentioned above regards dielectric elastomer as an idealized material.

The symmetry, crosslinking of dielectric elastomer molecular structure and pre-stress do affect its permittivity. The higher the symmetry is, the lower the permittivity is, equivalently the crosslinking shows a similar effect. In addition pre-stress induced the dielectric elastomer molecules to be arranged in order, hence the acting forces between molecules increase, but the permittivity declines due to the declining of the activity of polarity groups.

In this paper we propose an expression for the permittivity of silicone filled with high permittivity particles undergoing large electrostrictive deformation, including the expression of the permittivity based on experimental test and the permittivity based on the theory of composite material. By applying a model composed of complex linear dielectric behavior and nonlinear hyperelastic behavior, the electromechanical stability of the silicone filled with PMN–PT has been studied.

2. The permittivity of dielectric elastomer composite

2.1. The permittivity based on experimental test

The permittivity of the silicone is a function of the stretch, which has been proved experimentally and theoretically [13, 14]. At the same time, the permittivity of the silicone filled with high permittivity powder also depends on the electrostrictive deformation. Considering the influence of the uniformly filled high permittivity particles on the permittivity of silicone, the expression for the permittivity $\varepsilon^*(\lambda_i, v)$ can be written as

$$\varepsilon^*(\lambda_i, v) = \varepsilon^{\sim} + \sum_{i=1}^{3} m_i(\lambda_i - 1)\varepsilon^{\sim} + c(v)\varepsilon^{\sim} \tag{1}$$

where $\varepsilon^{\sim}$ is the permittivity of dielectric elastomer without deformation, v denotes the weight percentage of filled particles, $c(v)$ is the coefficient based on the particle content, λ_i $(i = 1–3)$ are the three principal stretches, m_i $(i = 1–3)$ are the electrostrictive coefficients. In the above expression of the permittivity, three terms are listed. The first term represents the permittivity of the ideal silicone without deformation, the second term reflects the influence of electrostrictive deformation, and the third term relates to the influence of the amount of filled particles. When $m_1 = a, m_2 = b, m_3 = a+b$, and $c(v)$ bears its special value respectively, then equation (1) can be predigested as the model proposed by Suo *et al* [13].

According to the experimental data obtained by Carpi [3], the permittivity shows nonlinear increases with increasing weight percentage of particles. Therefore, the generic expression of the coefficient $c(v)$ in equation (1) can be written as the following:

$$c(v) = dv^2 + ev + f \tag{2}$$

where d, e, f are material constants which can be determined by experiments. Fitting equation (2) with Carpi's data, the material constants d, e, f are determined as $d = 12.5$, $e = 3.75$, $f = 0$.

2.2. The permittivity based on the theory of composite material

Based on the series–parallel connection stiffness model in composite material theory, the upper and lower limits of the permittivity of electro-active silicone uniformly filled with high permittivity particles are expressed as follows:

$$\varepsilon^*_{\max}(\lambda_1, \lambda_2, \lambda_3, h_1, h_2) = \varepsilon_1^{\sim} h_1 + \varepsilon_2^{\sim} h_2 + \sum_{i=1}^{3} m_i(\lambda_i - 1)\varepsilon_1^{\sim} h_1 \tag{3a}$$

$$\varepsilon^*_{\min}(\lambda_1, \lambda_2, \lambda_3, h_1, h_2) = \frac{1}{\frac{1}{\varepsilon_1^{\sim} h_1} + \frac{1}{\varepsilon_2^{\sim} h_2} + \frac{1}{\sum_{i=1}^{3} m_i(\lambda_i - 1)\varepsilon_1^{\sim} h_1}} \tag{3b}$$

where h_1, h_2 are the volume fractions of silicone and particles, respectively and $h_1 + h_2 = 1$. The permittivity of composite silicone satisfies the following equation:

$$\varepsilon^*_{\min} \leqslant \varepsilon^*_{\mathrm{real}}(\lambda_1, \lambda_2, \lambda_3, h_1, h_2) \leqslant \varepsilon^*_{\max}. \tag{3c}$$

According to the permittivity model proposed by Jayasunder [3], the permittivity of composite silicone filled with particles is constructed as follows:

$$\begin{aligned} &\varepsilon^*_{\mathrm{real}}(\lambda_1, \lambda_2, \lambda_3, h_1, h_2) \\ &= \left\{ \frac{\varepsilon_1 h_1 + \varepsilon_2 h_2 \frac{3\varepsilon_1}{(2\varepsilon_1+\varepsilon_2)}\left[1 + 3h_2 \frac{(\varepsilon_2-\varepsilon_1)}{(2\varepsilon_1+\varepsilon_2)}\right]}{h_1 + h_2 \frac{3\varepsilon_1}{(2\varepsilon_1+\varepsilon_2)}\left[1 + 3h_2 \frac{(\varepsilon_2-\varepsilon_1)}{(2\varepsilon_1+\varepsilon_2)}\right]} \right\} + \sum_{i=1}^{3} m_i(\lambda_i - 1) \\ &\quad \times \left\{ \frac{\varepsilon_1 h_1 + \varepsilon_2 h_2 \frac{3\varepsilon_1}{(2\varepsilon_1+\varepsilon_2)}\left[1 + 3h_2 \frac{(\varepsilon_2-\varepsilon_1)}{(2\varepsilon_1+\varepsilon_2)}\right]}{h_1 + h_2 \frac{3\varepsilon_1}{(2\varepsilon_1+\varepsilon_2)}\left[1 + 3h_2 \frac{(\varepsilon_2-\varepsilon_1)}{(2\varepsilon_1+\varepsilon_2)}\right]} \right\}. \end{aligned} \tag{3d}$$

In the following the mechanical and electromechanical stability performance of composite dielectric elastomer filled particles is researched by applying the permittivity model obtained by experimental test.

3. Constitutive relation of dielectric elastomer composite

3.1. Constitutive relation of dielectric elastomer composite based on experimental test

The free energy function for the electromechanical coupling system of a dielectric elastomer can then be expressed as

$$W(\lambda_1, \lambda_2, \lambda_3, D^{\sim}) = U(\lambda_1, \lambda_2, \lambda_3) + V(\lambda_1, \lambda_2, \lambda_3, D^{\sim}) \tag{4}$$

where $U(\lambda_1, \lambda_2, \lambda_3)$ and $V(\lambda_1, \lambda_2, \lambda_3, D^{\sim})$ are the elastic strain energy and electric field energy density functions, respectively.

In 1977, Knowles proposed the Knowles model [19]. To study the mechanical performance and electromechanical stability of the silicone dielectric elastomer composite, a developed Knowles model is given as follows:

$$U(\lambda_1,\lambda_2,\lambda_3,v) = \frac{\mu(v)}{2b(v)}\left\{\left[1+\frac{b(v)}{n(v)}(\lambda_1^2+\lambda_2^2+\lambda_3^2-3)\right]^{n(v)}-1\right\} \tag{5}$$

where $\lambda_1^2+\lambda_2^2+\lambda_3^2>3$, $\mu(v)$ is the infinitesimal shear modulus, $n(v)$ is a 'hardening' parameter, $b(v)$ is a third material constant. All of these are related to the content of filled particles, and can be determined from experiments. It will be assumed throughout that $\mu(v)>0$, $n(v)>0$ and $b(v)>0$.

According to [13], the electric field energy density function can be expressed as $V(\lambda_1,\lambda_2,\lambda_3,D^{\sim}) = D^{\sim 2}\lambda_1^{-1}\lambda_2^{-2}\lambda_3/2\varepsilon^*(\lambda_1,\lambda_2,\lambda_3)$, and $D^{\sim}$ is the nominal electric displacement. The Knowles model is introduced to evaluate the nominal stress and the true stress of the silicone filled with particles in the following content. The nominal stress s_1, s_2, s_3 and the nominal electric field $E^{\sim}$ depend on the following functions respectively:

$$s_i = \frac{\partial W(\lambda_1,\lambda_2,\lambda_3,D^{\sim},v)}{\partial\lambda_i} \tag{6a}$$

$$E^{\sim} = \frac{\partial W(\lambda_1,\lambda_2,\lambda_3,D^{\sim},v)}{\partial D^{\sim}}. \tag{6b}$$

Substituting equations (5), (1), and (4) into (6), the nominal stress and the nominal electric field can be expressed respectively as follows:

$$\begin{aligned} s_i &= \mu(v)\lambda_i\left[1+\frac{b(v)}{n(v)}(I_1-3)\right]^{n(v)-1} \\ &\quad + (-1)^{\frac{1}{\sqrt{5}}\left[\left(\frac{\sqrt{5}+1}{2}\right)^{i+3}-\left(\frac{1-\sqrt{5}}{2}\right)^{i+3}\right]} \\ &\quad \times \frac{\lambda_i^{-1}\lambda_1^{-1}\lambda_2^{-1}\lambda_3 D^{\sim 2}}{2\left[1+\sum_{i=1}^{3} m_i(\lambda_i-1)+c(v)\right]\varepsilon^{\sim}} \\ &\quad - \frac{m_i\lambda_1^{-1}\lambda_2^{-1}\lambda_3 D^{\sim 2}}{2\left[1+\sum_{i=1}^{3} m_i(\lambda_i-1)+c(v)\right]^2\varepsilon^{\sim}} \end{aligned} \tag{7a}$$

$$E^{\sim} = \frac{\lambda_1^{-1}\lambda_2^{-1}\lambda_3 D^{\sim}}{\left[1+\sum_{i=1}^{3} m_i(\lambda_i-1)+c(v)\right]\varepsilon^{\sim}} \tag{7b}$$

where $I_1=\lambda_1^2+\lambda_2^2+\lambda_3^2$.

The true stresses can be written as: $\sigma_1 = s_1/\lambda_2\lambda_3$, $\sigma_2 = s_2/\lambda_1\lambda_3$, $\sigma_3 = s_3/\lambda_1\lambda_2$. By substituting them into equation (7), we then have:

$$\begin{aligned} \sigma_i &= \mu(v)\lambda_i^2\lambda_1^{-1}\lambda_2^{-1}\lambda_3^{-1}\left[1+\frac{b(v)}{n(v)}(I_1-3)\right]^{n(v)-1} \\ &\quad + (-1)^{\frac{1}{\sqrt{5}}\left[\left(\frac{\sqrt{5}+1}{2}\right)^{i+3}-\left(\frac{1-\sqrt{5}}{2}\right)^{i+3}\right]} \\ &\quad \times \frac{\lambda_1^{-2}\lambda_2^{-2}D^{\sim 2}}{2\left[1+\sum_{i=1}^{3} m_i(\lambda_i-1)+c(v)\right]\varepsilon^{\sim}} \\ &\quad - \frac{m_i\lambda_i\lambda_1^{-2}\lambda_2^{-2}D^{\sim 2}}{2\left[1+\sum_{i=1}^{3} m_i(\lambda_i-1)+c(v)\right]^2\varepsilon^{\sim}} \end{aligned} \tag{8a}$$

$$E = \frac{D}{[1+\sum_{i=1}^{3} m_i(\lambda_i-1)+c(v)]\varepsilon^{\sim}} \tag{8b}$$

where $D = D^{\sim}\lambda_1^{-1}\lambda_2^{-1}$ is the true electric displacement, $E = E^{\sim}\lambda_3^{-1}$ is the true electric field. Substituting equation (8*b*) into (8*a*), the item $\lambda_1^{-1}\lambda_2^{-2}D^{\sim 2}/2[1+\sum_{i=1}^{3} m_i(\lambda_i-1)+c(v)]\varepsilon^{\sim}$ in equation (8*a*) is replaced by $\varepsilon^* E^2/2$, and the item $m_i\lambda_i\lambda_1^{-2}\lambda_2^{-2}D^{\sim 2}/2[1+\sum_{i=1}^{3} m_i(\lambda_i-1)+c(v)]^2\varepsilon^{\sim}$ in equation (8*a*) by $\frac{\partial\varepsilon^*}{2\partial\lambda_i}\lambda_i E^2$. The true stress of the composite silicone is related to the four factors including hyperelasticity, Maxwell stress, electrostrictive deformation, particle content.

3.2. *Constitutive relation of dielectric elastomer composite based on the theory of composite material*

Substituting equations (3*d*), (4), and (5) into (6), the nominal stress and the nominal electric field can be expressed respectively as follows:

$$\begin{aligned} s_i &= \mu(v)\lambda_i\left[1+\frac{b(v)}{n(v)}(I_1-3)\right]^{n(v)-1} \\ &\quad + (-1)^{\frac{1}{\sqrt{5}}\left[\left(\frac{\sqrt{5}+1}{2}\right)^{i+3}-\left(\frac{1-\sqrt{5}}{2}\right)^{i+3}\right]}\frac{\lambda_1^{-1}\lambda_2^{-1}\lambda_3 D^{\sim 2}\lambda_i^{-1}}{2\varepsilon^*} \\ &\quad - \frac{m_i\left\{\dfrac{\varepsilon_1 h_1+\varepsilon_2 h_2\frac{3\varepsilon_1}{(2\varepsilon_1+\varepsilon_2)}\left[1+3h_2\frac{(\varepsilon_2-\varepsilon_1)}{(2\varepsilon_1+\varepsilon_2)}\right]}{h_1+h_2\frac{3\varepsilon_1}{(2\varepsilon_1+\varepsilon_2)}\left[1+3h_2\frac{(\varepsilon_2-\varepsilon_1)}{(2\varepsilon_1+\varepsilon_2)}\right]}\right\}\lambda_1^{-1}\lambda_2^{-1}\lambda_3 D^{\sim 2}}{2\varepsilon^{*2}} \end{aligned} \tag{9a}$$

$$E^{\sim} = \frac{D^{\sim}}{\varepsilon^*}\lambda_1^{-1}\lambda_2^{-1}\lambda_3. \tag{9b}$$

The corresponding true stresses and the true electric field are

$$\begin{aligned} \sigma_i &= \mu(v)\lambda_i^2\lambda_1^{-1}\lambda_2^{-1}\lambda_3^{-1}\left[1+\frac{b(v)}{n(v)}(I_1-3)\right]^{n(v)-1} \\ &\quad + (-1)^{\frac{1}{\sqrt{5}}\left[\left(\frac{\sqrt{5}+1}{2}\right)^{i+3}-\left(\frac{1-\sqrt{5}}{2}\right)^{i+3}\right]}\frac{\lambda_1^{-1}\lambda_2^{-1}\lambda_3 D^{\sim 2}}{2\varepsilon^*} \\ &\quad - \frac{m_i\lambda_i\left\{\dfrac{\varepsilon_1 h_1+\varepsilon_2 h_2\frac{3\varepsilon_1}{(2\varepsilon_1+\varepsilon_2)}\left[1+3h_2\frac{(\varepsilon_2-\varepsilon_1)}{(2\varepsilon_1+\varepsilon_2)}\right]}{h_1+h_2\frac{3\varepsilon_1}{(2\varepsilon_1+\varepsilon_2)}\left[1+3h_2\frac{(\varepsilon_2-\varepsilon_1)}{(2\varepsilon_1+\varepsilon_2)}\right]}\right\}\lambda_1^{-1}\lambda_2^{-1}\lambda_3 D^{\sim 2}}{2\varepsilon^{*2}} \end{aligned} \tag{10a}$$

$$E = \frac{D}{\varepsilon^*}. \tag{10b}$$

4. Electromechanical stability of dielectric elastomer composite

4.1. *Electromechanical stability of dielectric elastomer composite based on experimental test*

We express the Hessian matrix of the silicone dielectric elastomer composite electromechanical coupling system as

$$H = \begin{bmatrix} \frac{\partial s_1}{\partial\lambda_1} & \frac{\partial s_1}{\partial\lambda_2} & \frac{\partial s_1}{\partial\lambda_3} & \frac{\partial s_1}{\partial D^{\sim}} \\ \frac{\partial s_2}{\partial\lambda_1} & \frac{\partial s_2}{\partial\lambda_2} & \frac{\partial s_2}{\partial\lambda_3} & \frac{\partial s_2}{\partial D^{\sim}} \\ \frac{\partial s_3}{\partial\lambda_1} & \frac{\partial s_3}{\partial\lambda_2} & \frac{\partial s_3}{\partial\lambda_3} & \frac{\partial s_3}{\partial D^{\sim}} \\ \frac{\partial E^{\sim}}{\partial\lambda_1} & \frac{\partial E^{\sim}}{\partial\lambda_2} & \frac{\partial E^{\sim}}{\partial\lambda_3} & \frac{\partial E^{\sim}}{\partial D^{\sim}} \end{bmatrix} \tag{11}$$

where

$$
\begin{aligned}
\frac{\partial s_j}{\partial \lambda_j} &= \mu(v)\left[1+\frac{b(v)}{n(v)}(I_1-3)\right]^{n(v)-2} \\
&\quad\times\left\{1+\frac{b(v)}{n(v)}(I_1-3)+2\lambda_j^2\frac{[n(v)-1]}{n(v)}b(v)\right\} \\
&\quad+\frac{\lambda_j^{-2}\lambda_1^{-1}\lambda_2^{-1}\lambda_3 D^{\sim 2}}{\varepsilon^*}+\frac{m_j\lambda_j^{-1}\lambda_1^{-1}\lambda_2^{-1}\lambda_3 D^{\sim 2}\varepsilon^{\sim}}{\varepsilon^{*2}} \\
&\quad+\frac{m_j^2\lambda_1^{-1}\lambda_2^{-1}\lambda_3 D^{\sim 2}\varepsilon^{\sim 2}}{\varepsilon^{*3}} \\
\frac{\partial s_1}{\partial \lambda_{j+1}} &= \frac{\partial s_{j+1}}{\partial \lambda_1} = 2\mu(v)b(v)\lambda_1\lambda_{j+1} \\
&\quad\times\frac{[n(v)-1]}{n(v)}\left[1+\frac{b(v)}{n(v)}(I_1-3)\right]^{n(v)-2} \\
&\quad-(-1)^{j+2}\frac{\lambda_{j+1}^{-1}\lambda_1^{-2}\lambda_2^{-1}\lambda_3 D^{\sim 2}}{2\varepsilon^*} \\
&\quad+\frac{\lambda_1^{-1}\lambda_2^{-1}\lambda_3 D^{\sim 2}\varepsilon^{\sim}(m_{j+1}\lambda_1^{-1}-(-1)^{j+2}m_1\lambda_{j+1}^{-1})}{2\varepsilon^{*2}} \\
&\quad+\frac{m_1 m_{j+1}\lambda_1^{-1}\lambda_2^{-1}\lambda_3 D^{\sim 2}\varepsilon^{\sim 2}}{\varepsilon^{*3}} \\
\frac{\partial s_i}{\partial D^{\sim}} &= \frac{\partial E^{\sim}}{\partial \lambda_i} = (-1)^{\frac{1}{\sqrt{5}}\left[\left(\frac{\sqrt{5}+1}{2}\right)^{i+3}-\left(\frac{1-\sqrt{5}}{2}\right)^{i+3}\right]}\frac{\lambda_i^{-1}\lambda_1^{-1}\lambda_2^{-1}\lambda_3 D^{\sim}}{\varepsilon^*} \\
&\quad-\frac{m_i\lambda_1^{-1}\lambda_2^{-1}\lambda_3 D^{\sim}\varepsilon^{\sim}}{\varepsilon^{*2}} \\
\frac{\partial s_3}{\partial \lambda_3} &= \mu(v)\left[1+\frac{b(v)}{n(v)}(I_1-3)\right]^{n(v)-2} \\
&\quad\times\left\{1+\frac{b(v)}{n(v)}(I_1-3)+2\lambda_3^2\frac{[n(v)-1]}{n(v)}b(v)\right\} \\
&\quad-\frac{\lambda_1^{-1}\lambda_2^{-1}D^{\sim 2}\varepsilon^{\sim}(1+m_3^2)}{2m_3\varepsilon^{*2}}+\frac{m_3^2\lambda_1^{-1}\lambda_2^{-1}\lambda_3 D^{\sim 2}\varepsilon^{\sim 2}}{\varepsilon^{*3}} \\
\frac{\partial s_2}{\partial \lambda_3} &= \frac{\partial s_3}{\partial \lambda_2} = 2\mu(v)b(v)\lambda_2\lambda_3 \\
&\quad\times\frac{[n(v)-1]}{n(v)}\left[1+\frac{b(v)}{n(v)}(I_1-3)\right]^{n(v)-2}-\frac{\lambda_1^{-1}\lambda_2^{-2}D^{\sim 2}}{2\varepsilon^*} \\
&\quad+\frac{\lambda_1^{-1}\lambda_2^{-1}D^{\sim 2}\varepsilon^{\sim}(m_3\lambda_3-m_2)}{2\varepsilon^{*2}} \\
&\quad+\frac{m_2 m_3\lambda_1^{-1}\lambda_2^{-1}\lambda_3 D^{\sim 2}\varepsilon^{\sim 2}}{\varepsilon^{*3}} \\
\frac{\partial E^{\sim}}{\partial D^{\sim}} &= \frac{\lambda_1^{-1}\lambda_2^{-1}\lambda_3}{\varepsilon^*}, \qquad j=1,2.
\end{aligned}
$$

To obtain a stable state for the silicone composite, the determinant of the Hessian matrix should be positive. When the dielectric elastomer electromechanical coupling system reaches the critical state, $\det(H_C)=0$. Solving equation (11), we can get the critical electromechanical stability parameters of silicone composite, such as the critical nominal electric field $E^{\sim}_{\max}$, the critical true electric field $E_{\max}$, the critical nominal stress s_C, and the critical true stress σ_C.

Now, considering the in-plane deformation of dielectric elastomer, $\lambda_3\approx 1$, let $m_1=m_2=r$, equation (1) can be simplified as

$$\varepsilon^*(\lambda_1,\lambda_2,v)=\varepsilon^{\sim}+r(\lambda_1+\lambda_2-2)\varepsilon^{\sim}+c(v)\varepsilon^{\sim}. \tag{12}$$

The special case $n(v)\equiv 1$ in equation (5) reduces to the neo-Hookean strain energy model equation (13) with one material constant:

$$U(\lambda_1,\lambda_2,\lambda_3)=\frac{\mu(v)}{2}(\lambda_1^2+\lambda_2^2+\lambda_3^2-3). \tag{13}$$

$\mu(v)$ is the shear modulus related to the content of filled particles, which can be determined in experiments. In order to investigate the influence of the permittivity changing, which results from the deformation, on the silicone electromechanical stability, we take $r=-0.03, 0, 1, 2, 3$ respectively in the following content [17].

Applying pre-stress on the silicone dielectric composite materials, let $\lambda_1=\lambda_2=\lambda$; this means $\lambda_3=\lambda^{-2}$ due to the volume incompressibility property. In this case, the free energy function can be written as

$$
\begin{aligned}
W(\lambda,D^{\sim}) &= \frac{\mu(v)}{2}(2\lambda^2+\lambda^{-4}-3) \\
&\quad+\frac{\lambda^{-4}D^{\sim\,2}}{2[1+2r(\lambda-1)+c(v)]\varepsilon^{\sim}}.
\end{aligned} \tag{14}
$$

To get the relation between nominal electric displacement and nominal electric field, stretch and nominal electric field, $\partial W(\lambda,D^{\sim})/\partial\lambda=0$ is used, substituting equation (14) into it, we have:

$$\frac{D^{\sim}}{\sqrt{\varepsilon^{\sim}\mu(v)}}=\sqrt{\frac{2(\lambda^6-1)[1+2r(\lambda-1)+c(v)]}{2+r\lambda[1+2r(\lambda-1)+c(v)]^{-1}}} \tag{15a}$$

$$\frac{E^{\sim}}{\sqrt{\mu(v)/\varepsilon^{\sim}}}=\sqrt{\frac{2(\lambda^{-2}-\lambda^{-8})[1+2r(\lambda-1)+c(v)]^{-1}}{2+r\lambda[1+2r(\lambda-1)+c(v)]^{-1}}}. \tag{15b}$$

Based on equation (15), taking the stretch λ as a variable, altering r and $c(v)$, then the changing trends of the nominal electric field and the nominal electric displacement are obtained. In the following we discuss the sign of the square roots in equation (13). The equal biaxial stretch ratio changes $\lambda\in[1,5]$ undergoing large deformation, hence we set $\lambda^6-1\geqslant 0$ in equation (15a). Then the signs of $1+2r(\lambda-1)+c(v)$ and $2+r\lambda[1+2r(\lambda-1)+c(v)]^{-1}$ are discussed. Based on Capri's experimental data [3], we have $0\leqslant c(v)\leqslant 3$. If $1+2r(\lambda-1)\geqslant 0$, the minimum value of the electrostrictive coefficient r is -0.125 under the equal biaxial stretch ratio. If $1+2r(\lambda-1)+3\geqslant 0$, the minimum value of the electrostrictive coefficient r is -0.5. To get $2+r\lambda[1+2r(\lambda-1)+c(v)]^{-1}\geqslant 0$, if $2+r\lambda[1+2r(\lambda-1)]^{-1}\geqslant 0$, we have $r\geqslant -0.08$ and if $2+r\lambda[1+2r(\lambda-1)+3]^{-1}\geqslant 0$, $r\geqslant -0.42$. In all, to get $D^{\sim}/\sqrt{\varepsilon^{\sim}\mu(v)}\geqslant 0$, the electrostrictive coefficient must be $r\geqslant -0.08$. In a similar way, the electrostrictive coefficient should be $r\geqslant -0.08$ when $E^{\sim}/\sqrt{\mu(v)/\varepsilon^{\sim}}\geqslant 0$.

For a dielectric elastomer, experiments have suggested $1\leqslant\lambda\leqslant 6$ for equal biaxial stretch, we choose the stretch range as [1, 6] [23, 35], and calculate the range of nominal electric displacement $D^{\sim}$ as [0.0006, 6], using representative

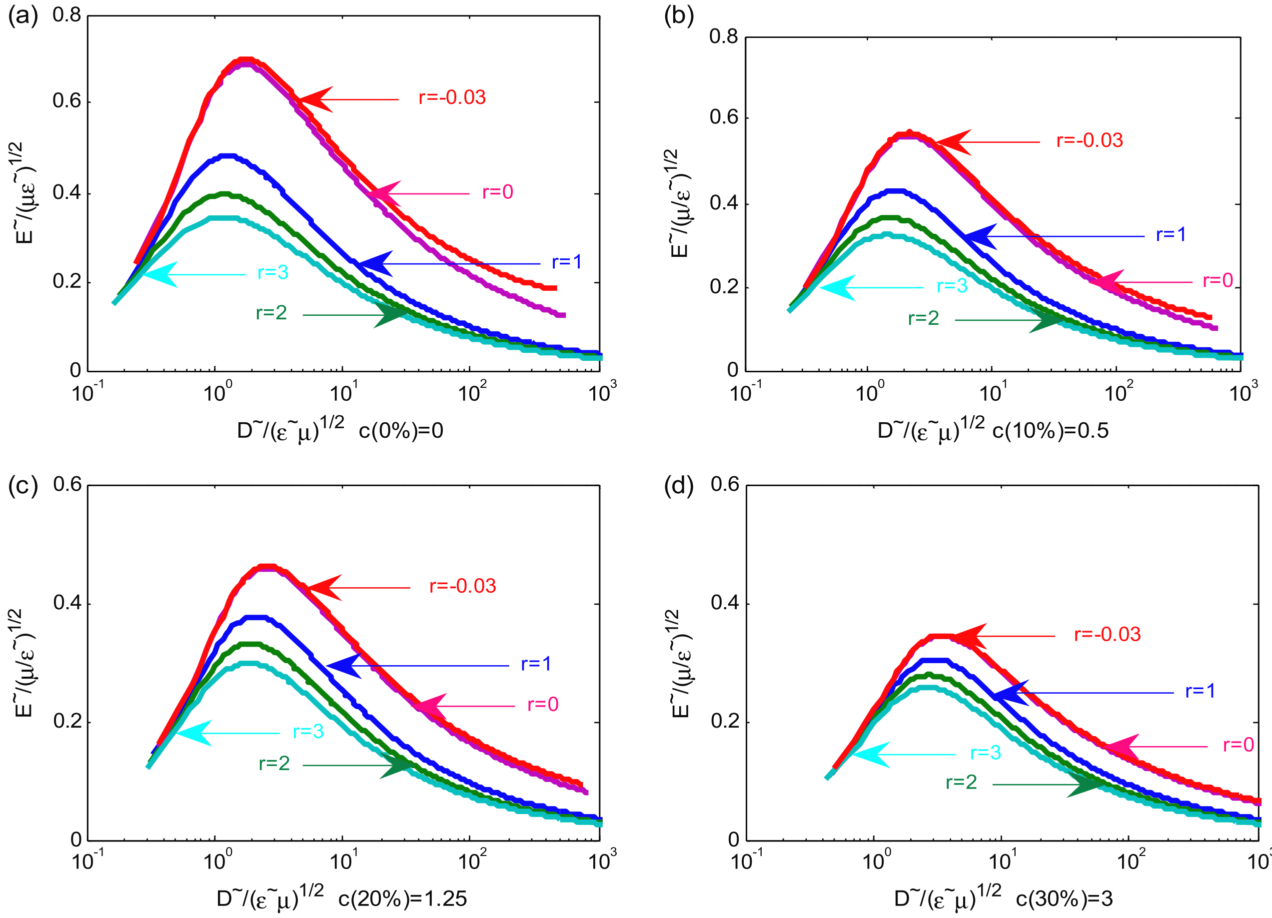

Figure 1. Influence of uniformly filled PMN–PT on the nominal electric field and nominal electric field of silicone under the specific load condition $\lambda_1 = \lambda_2 = \lambda$. (a) $c(0\%) = 0$, (b) $c(10\%) = 0.5$, (c) $c(20\%) = 1.25$, (d) $c(30\%) = 3$.

values $\mu = 1 \times 10^6$ N m^{-2}, $\varepsilon = 4 \times 10^{-11}$ F m^{-1}, $10^{-1} \leqslant \frac{D^{\sim}}{\sqrt{\mu\varepsilon}} \leqslant 10^3$.

Figure 1 shows the coupling influence of electrostrictive deformation and weight percentage of filled PMN–PT on the electromechanical stability performance of the silicone. The PMN–PT is lead magnesium niobate–lead titanate ferroelectric powder. According to Carpi's paper, the PMN–PT particles appear almost perfectly spherical with diameters ranging from less than 1 μm to nearly 100 μm [3]. Clearly, the critical nominal electric field of the electromechanical coupling system decreases along with increasing content of PMN–PT due to the particles of PMN–PT destroying the relatively regular microstructure of the silicone. Simultaneously the critical nominal electric field decreases along with the increasing electrostriction coefficient r. In a special case, when $r = 0$, it indicates that the permittivity only depends on the weight percentage of PMN–PT content variation. In the case when $c(0\%) = 0$, $E^{\sim}_{\max} = 0.6877\sqrt{\mu(v)/\varepsilon^{\sim}}$, which is consistent with the conclusion obtained by Zhao and Suo [4]. For example, if $c(v)$ takes different values of 0.5, 1.25 and 3 respectively, the corresponding nominal electric field peaks are $0.5600\sqrt{\mu(v)/\varepsilon^{\sim}}$, $0.4582\sqrt{\mu(v)/\varepsilon^{\sim}}$, $0.3437\sqrt{\mu(v)/\varepsilon^{\sim}}$, respectively. For representative values $\varepsilon^{\sim} = 7 \times 10^{-11}$ F m^{-1}, $\mu(0\%) = 6.3 \times 10^4$ Pa, $\mu(10\%) = 9.4 \times 10^4$ Pa, $\mu(20\%) = 1.3 \times 10^5$ Pa, $\mu(30\%) = 1.35 \times 10^5$ Pa, the corresponding critical nominal electric field is 2.06×10^7 V m^{-1}, 2.05×10^7 V m^{-1}, 1.97×10^7 V m^{-1}, 1.51×10^7 V m^{-1} respectively. The nominal electric field decreases by an amount of 26.7% when the particle content reaches 30% under the same electrostrictive coefficient. This means that the stability performance of silicone composite declines sharply due to the great influence of filled PMN–PT on the uniformity and compactability of the electro-active silicone composite.

Figure 2 shows the relation between the nominal electric field and the stretch of the composite dielectric elastomer. Clearly, the nominal electric field increases along with the increasing stretch, until it reaches to the critical value, when the electric field decreases and tends to stability. When $r = 0$ and $c(0\%) = 0$, the critical stretch can be obtained as $\lambda^{C} = 1.26$. This result agrees well with the conclusion obtained by Zhao and Suo [4]. By comparison we observe that the filled PMN–PT has no influence on the critical nominal stretch at all, though it may affect the critical electric field. Clearly, the critical stretch corresponding to the critical nominal electric field increases along with the decreasing of r. As a special example, various values of r are selected, namely $r = 3, 2, 1, 0, -0.03$. When $c(10\%) = 0.5$, the critical nominal electric field reaches $0.3230\sqrt{\mu(v)/\varepsilon^{\sim}}$, $0.3648\sqrt{\mu(v)/\varepsilon^{\sim}}$, $0.4299\sqrt{\mu(v)/\varepsilon^{\sim}}$, $0.5612\sqrt{\mu(v)/\varepsilon^{\sim}}$, $0.5678\sqrt{\mu(v)/\varepsilon^{\sim}}$, the corresponding critical stretches are 1.16, 1.17, 1.19, 1.26, 1.26, respectively. The corresponding critical nominal electric field is 1.18×10^7, 1.34×10^7, 1.57×10^7, 2.06×10^7,

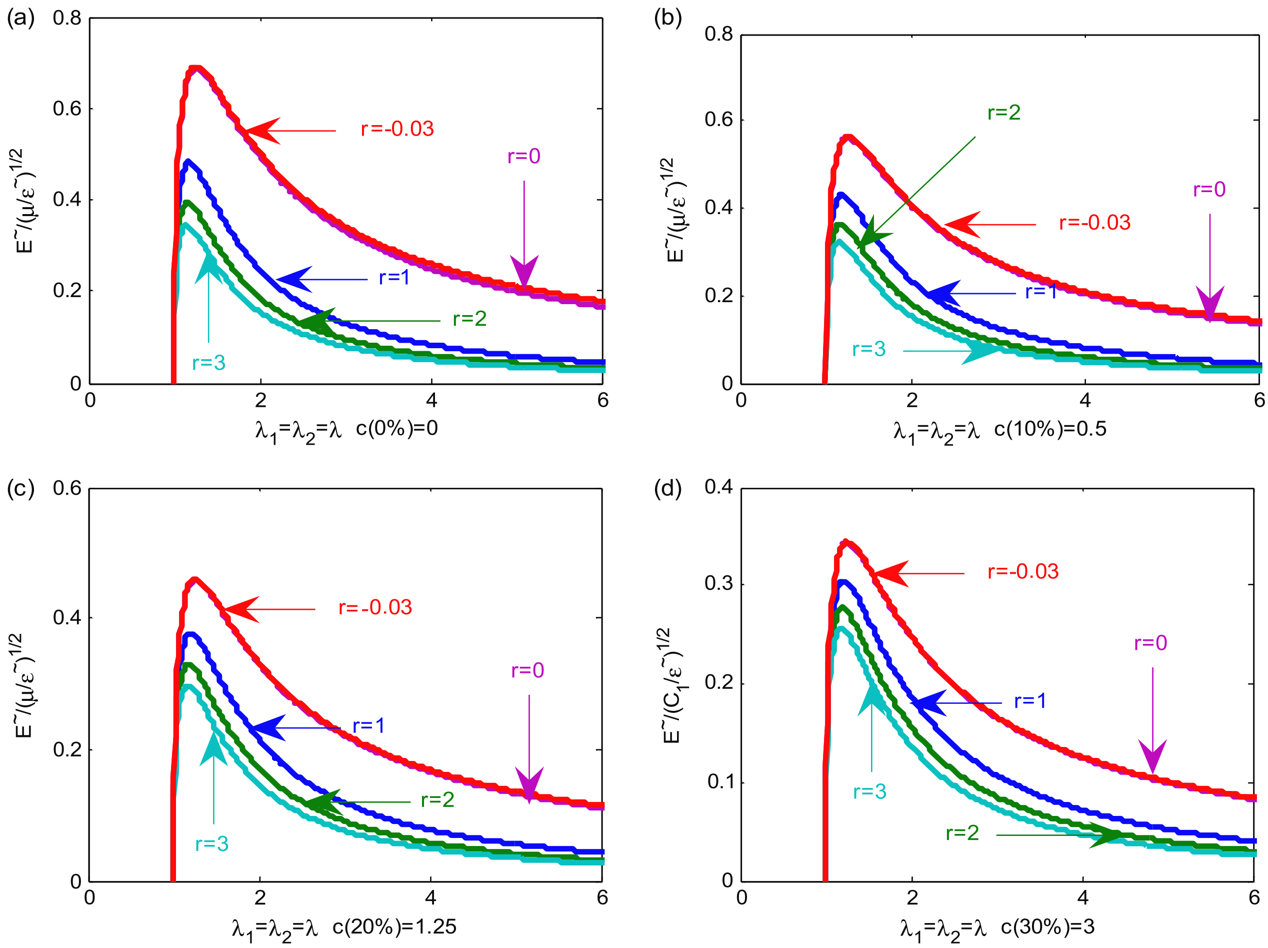

Figure 2. The relation between nominal electric field and the stretch of silicon with uniformly mixed PMN–PT under the specific load condition $\lambda_1 = \lambda_2 = \lambda$. (a) $c(0\%) = 0$, (b) $c(10\%) = 0.5$, (c) $c(20\%) = 1.25$, (d) $c(30\%) = 3$.

2.08×10^7 V m^{-1}. The nominal electric field declines by 43.2% when the electrostrictive coefficient r reaches 3 from -0.03 under the same particle content. Clearly, the electromechanical stability performance of composite silicone is lower for the original structure being damaged by the large deformation.

Next, for another case, assuming $\lambda_3 = \lambda$, i.e. $\lambda_1 = \lambda_2 = \lambda^{-1/2}$, applying a similar method of investigation, we express the nominal electric displacement and the nominal electric field as

$$\frac{D^{\sim}}{\sqrt{\mu(v)/\varepsilon^{\sim}}} = \left\{ \frac{2(\lambda^{-2} - \lambda)}{2\lambda + r\lambda^{1/2}[1 + 2r(\lambda^{-1/2} - 1) + c(v)]^{-1}} \times [1 + 2r(\lambda^{-1/2} - 1) + c(v)] \right\}^{1/2} \quad (16a)$$

$$\frac{E^{\sim}}{\sqrt{\mu(v)/\varepsilon^{\sim}}} = \left\{ \frac{2(\lambda^{2} - \lambda^5)}{2\lambda + r\lambda^{1/2}[1 + 2r(\lambda^{-1/2} - 1) + c(v)]^{-1}} \times [1 + 2r(\lambda^{-1/2} - 1) + c(v)]^{-1} \right\}^{1/2}. \quad (16b)$$

Here $\lambda \in [0.04, 1]$ in equations (16), hence $\lambda^{-2} - \lambda \geqslant 0$. Similarly to the investigation of equal biaxial condition, to get $D^{\sim}/\sqrt{\varepsilon^{\sim}\mu(v)} \geqslant 0$ and $E^{\sim}/\sqrt{\mu(v)/\varepsilon^{\sim}} \geqslant 0$, the electrostrictive coefficient must be $r \geqslant -0.08$.

Figure 3 shows the relation between nominal electric displacement and nominal electric field after the silicon was uniformly mixed with different weight percentages of PMN–PT powder under the load condition $\lambda_3 = \lambda$. Owing to the fact that the mechanical performances in the planes expanded along the stretch direction and perpendicular to the stretch direction are essentially the same, hence the change trend shown by figure 3 is similar to that shown in figure 1.

Figure 4 shows the relation between the nominal electric field and the stretch of silicone filled with PMN–PT under the load condition $\lambda_3 = \lambda$. Clearly, the nominal electric field increases along with the increasing stretch. When the stretch approaches its critical value, the electric field decreases and eventually tends to stabilize at zero. The systematic critical electric field decreases with increasing PMN–PT content and the increasing electrostriction coefficient r. If $r = 0$, i.e. neglecting the influence of deformation, the critical stretch $\lambda^C = 0.37$. This result agrees well with the conclusion obtained by Zhao and Suo [4]. In a special example ($c(20\%) = 1.25$), if r takes different values (3, 2, 1, 0, -0.03), when the nominal electric field reaches its peak ($0.2978\sqrt{\mu(v)/\varepsilon^{\sim}}$, $0.3301\sqrt{\mu(v)/\varepsilon^{\sim}}$, $0.3770\sqrt{\mu(v)/\varepsilon^{\sim}}$, $0.4582\sqrt{\mu(v)/\varepsilon^{\sim}}$, $0.4618\sqrt{\mu(v)/\varepsilon^{\sim}}$), the corresponding critical stretches are 0.72, 0.71, 0.69, 0.63, 0.63, respectively. The corresponding critical nominal electric

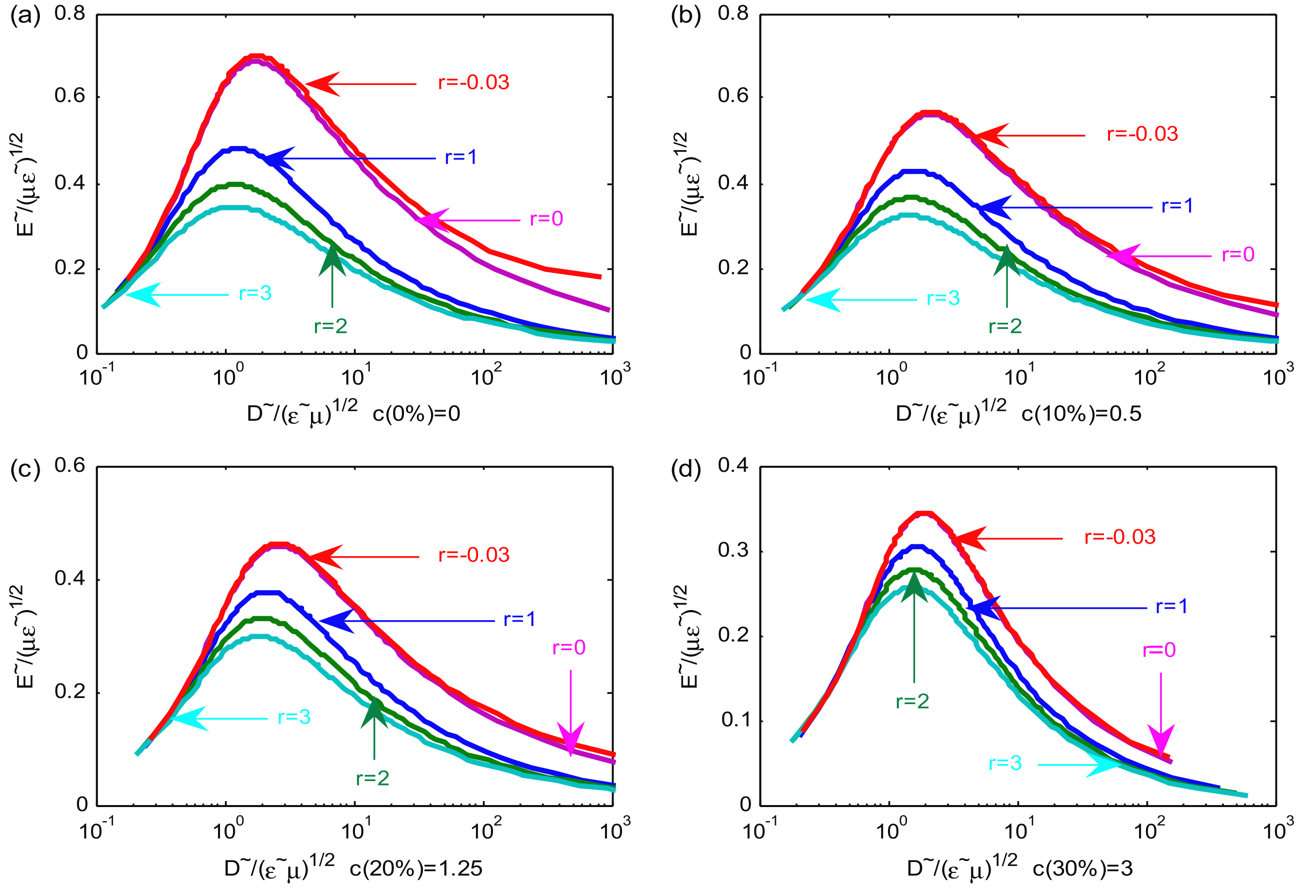

Figure 3. The relation between the nominal electric field and nominal electric displacement of silicon with uniformly mixed PMN–PT under the specific load condition $\lambda_3 = \lambda$. (a) $c(0\%) = 0$, (b) $c(10\%) = 0.5$, (c) $c(20\%) = 1.25$, (d) $c(30\%) = 3$.

fields are 1.28×10^7, 1.42×10^7, 1.62×10^7, 1.97×10^7, 1.99×10^7 V m^{-1}.

According to the comparison of the two stretch processes, all of the critical nominal electric fields and the critical stretches increase with decreasing of the electrostrictive coefficient r or particle content $c(v)$, which indicates that the composite silicone or structure is more stable.

4.2. Electromechanical stability of dielectric elastomer composite based on the theory of composite material

Applying pre-stress on the silicone dielectric composite materials, let $\lambda_1 = \lambda_2 = \lambda$; this means $\lambda_3 = \lambda^{-2}$ due to the volume incompressibility property. In this case, the free energy function can be written as

$$W(\lambda, D^{\sim}) = \frac{\mu(h_1, h_2)}{2}(2\lambda^2 + \lambda^{-4} - 3) + \frac{\lambda^{-4} D^{\sim 2}}{2[1 + (m_1 + m_2)(\lambda - 1) + m_3(\lambda^{-2} - 1)]\varepsilon^*} \quad (17)$$

where

$$\varepsilon^* = \left\{\frac{\varepsilon_1 h_1 + \varepsilon_2 h_2 \frac{3\varepsilon_1}{(2\varepsilon_1+\varepsilon_2)}\left[1 + 3h_2 \frac{(\varepsilon_2-\varepsilon_1)}{(2\varepsilon_1+\varepsilon_2)}\right]}{h_1 + h_2 \frac{3\varepsilon_1}{(2\varepsilon_1+\varepsilon_2)}\left[1 + 3h_2 \frac{(\varepsilon_2-\varepsilon_1)}{(2\varepsilon_1+\varepsilon_2)}\right]}\right\} + \sum_{i=1}^{3} m_i(\lambda_i - 1)\left\{\frac{\varepsilon_1 h_1 + \varepsilon_2 h_2 \frac{3\varepsilon_1}{(2\varepsilon_1+\varepsilon_2)}\left[1 + 3h_2 \frac{(\varepsilon_2-\varepsilon_1)}{(2\varepsilon_1+\varepsilon_2)}\right]}{h_1 + h_2 \frac{3\varepsilon_1}{(2\varepsilon_1+\varepsilon_2)}\left[1 + 3h_2 \frac{(\varepsilon_2-\varepsilon_1)}{(2\varepsilon_1+\varepsilon_2)}\right]}\right\},$$

let $m_1 = \beta_2 m_2 = \beta_3 m_3 = m = r$, β_2 and β_3 are defined as the electrical stretching coefficient ratios, which are related with both the dielectric elastomer composite and the load of the coupled field. To get the relations between the nominal electric displacement and the nominal electric field, and the stretch and the nominal electric field, $\partial W(\lambda, D^{\sim})/\partial\lambda = 0$ is used, substituting equation (17) into it, we have:

$$\frac{D^{\sim}}{\sqrt{\varepsilon^* \mu(h_1, h_2)}} = \sqrt{\frac{2(\lambda - \lambda^{-5})}{\Lambda} - \frac{s}{\Lambda\mu(h_1, h_2)}} \quad (18a)$$

$$\frac{E^{\sim}}{\sqrt{\mu(h_1, h_2)/\varepsilon^*}} = \frac{\lambda^{-4}}{\{1 + r[(1 + \frac{1}{\beta_2})(\lambda - 1) + \frac{1}{\beta_3}(\lambda^{-2} - 1)]\}} \times \sqrt{\frac{2(\lambda - \lambda^{-5})}{\Lambda} - \frac{s}{\Lambda\mu(h_1, h_2)}} \quad (18b)$$

where $\Lambda = \frac{2\{1+r[(1+\frac{1}{\beta_2})(\lambda-1)+\frac{1}{\beta_3}(\lambda^{-2}-1)]\}+r[\frac{1}{2}(1+\frac{1}{\beta_2})\lambda-\frac{1}{\beta_3}\lambda^{-2}]}{\lambda^5\{1+r(1+\frac{1}{\beta_2})(\lambda-1)+\frac{r}{\beta_3}(\lambda^{-2}-1)\}^2}$.

Figure 5 plots the relationship between the nominal electric displacement and the nominal electric field of the composite dielectric elastomer with different values of r ($-0.03, -0.01$) and different values of β_2 (1, 1.2) and β_3 (1, 1.2), respectively. Clearly, in this case, the critical nominal electric field of the composite increases along with the increasing electrostriction coefficient r, the comparative stability performance of the material is even

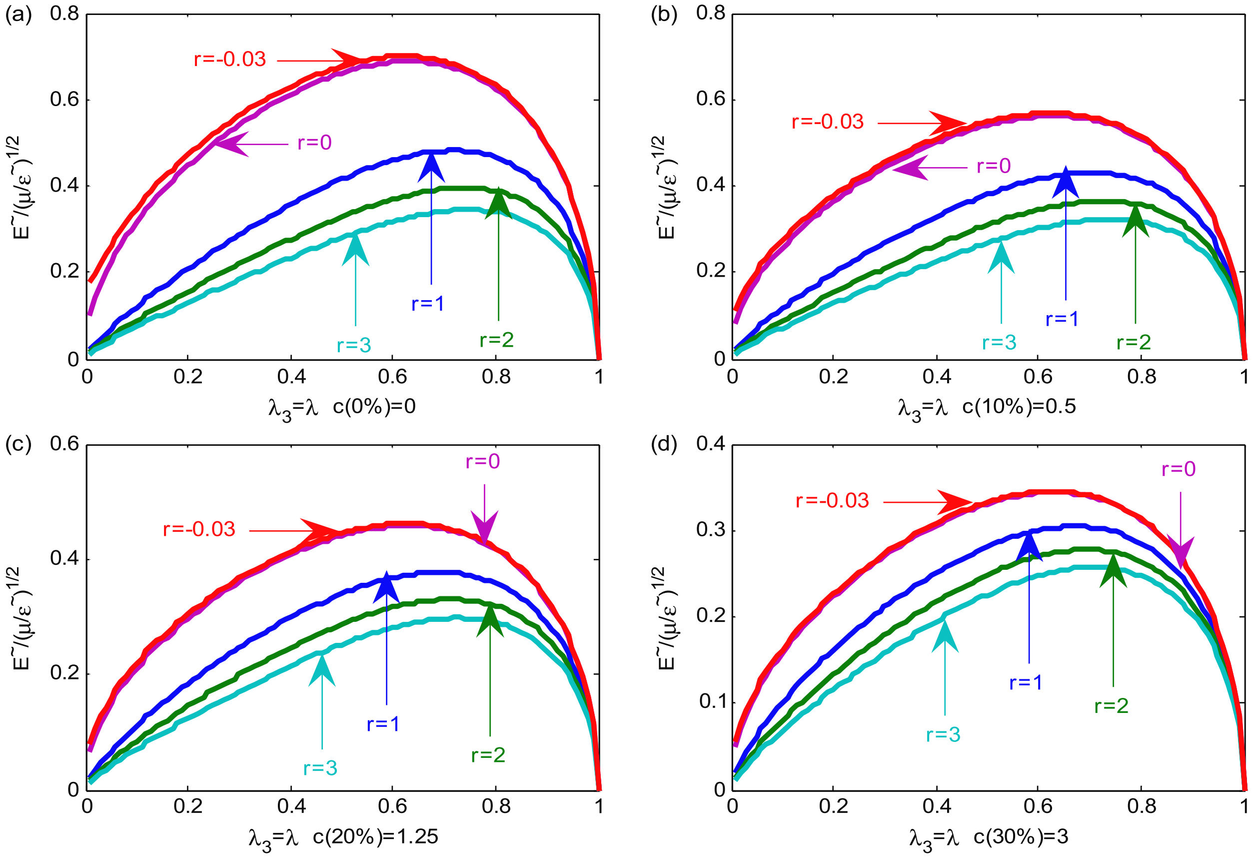

Figure 4. The relation between nominal electric field and stretch of silicon with uniformly mixed PMN–PT under the specific load condition $\lambda_3 = \lambda$. (a) $c(0\%) = 0$, (b) $c(10\%) = 0.5$, (c) $c(20\%) = 1.25$, (d) $c(30\%) = 3$.

higher. Along with the decrease of β_2 or increase of β_3, the peaks of the nominal electric field increase and the electromechanical stability of the composite material is improved. When $\beta_2 = 1$, $\beta_3 = 1$, with r of -0.03, -0.01, the corresponding critical nominal electric fields are $0.5875\sqrt{\mu(h_1, h_2)/\varepsilon^*(h_1, h_2)}$, $0.6303\sqrt{\mu(h_1, h_2)/\varepsilon^*(h_1, h_2)}$, the corresponding critical stretches are 1.51, 1.40. And if $\beta_2 = 1.2$ or $\beta_3 = 1.2$, the corresponding critical nominal electric fields are $0.5770\sqrt{\mu(h_1, h_2)/\varepsilon^*(h_1, h_2)}$, $0.6050\sqrt{\mu(h_1, h_2)/\varepsilon^*(h_1, h_2)}$, and the corresponding critical stretches are 1.56, 1.45.

According to Capri's research [3], $\varepsilon_{\mathrm{PMN-PT}} = 28\,000$, $\varepsilon_{\mathrm{Silicone}} = 8$, considering o Jayasunder's permittivity model in equation (3), we have $\varepsilon_J^*(0, 1) = 8$, $\varepsilon_J^*(0.1, 0.9) = 12.64$, $\varepsilon_J^*(0.2, 0.8) = 17.58$, $\varepsilon_J^*(0.3, 0.7) = 27.50$. Therefore, the corresponding critical nominal electric fields are 1.76×10^7 V m^{-1}, 1.70×10^7 V m^{-1}, 1.69×10^7 V m^{-1}, 1.38×10^7 V m^{-1}, respectively. The nominal electric field decreases by an amount of 21.6% when the particle content reaches 30%.

Comparing the critical nominal electric field obtained by experiments with that obtained by the theory of composite materials, we have that the latter critical nominal electric field is smaller than that obtained by experiments. However, the falling range of the critical nominal electric fields obtained by the two methods is consistent along with the particle content increasing.

Consider the unequal biaxial condition of dielectric elastomer composite, $\lambda_2 = q\lambda_1 = q\lambda$, and $\lambda_3 = \lambda^{-2}/p$, the free energy function is

$$W(\lambda, D^{\sim}) = \frac{\mu(h_1, h_2)}{2}\left[(1+q^2)\lambda^2 + \frac{\lambda^{-4}}{q^2} - 3\right] + \frac{\lambda^{-4}D^{\sim 2}}{2[1 + m_1(\lambda-1) + m_2(q\lambda - 1) + m_3(\lambda^{-2}/q - 1)]\varepsilon^*}. \quad (19)$$

Then, the nominal electric field and the nominal electric displacement, the nominal electric field and the stretch can be expressed as follows

$$\frac{D^{\sim}}{\sqrt{\varepsilon^*\mu(h_1, h_2)}} = \sqrt{\frac{q^{-2}(1+q^{-2})\lambda - 2\lambda^{-5}}{q^{-2}\Lambda'} - \frac{s}{\Lambda'\mu(h_1, h_2)}} \quad (20a)$$

$$\frac{E^{\sim}}{\sqrt{\mu(h_1, h_2)/\varepsilon^*}} = \frac{\lambda^{-4}}{\left\{1 + r(\lambda-1) + \frac{r}{\beta_2}(q\lambda - 1) + \frac{r}{\beta_3}\left(\frac{\lambda^{-2}}{q} - 1\right)\right\}} \times \sqrt{\frac{q^{-2}(1+q^{-2})\lambda - 2\lambda^{-5}}{q^{-2}\Lambda'} - \frac{s}{\Lambda'\mu(h_1, h_2)}} \quad (20b)$$

where $\Lambda' = \frac{2\{1+r[(\lambda-1)+\frac{1}{\beta_2}(q\lambda-1)+\frac{1}{\beta_3}(\frac{\lambda^{-2}}{q}-1)]\}+r[\frac{1}{2}(1+\frac{q}{\beta_2})\lambda-\frac{1}{q\beta_3}\lambda^{-2}]}{\lambda^5\{1+r[(\lambda-1)+\frac{1}{\beta_2}(q\lambda-1)+\frac{1}{\beta_3}(\frac{\lambda^{-2}}{q}-1)]\}^2}$.

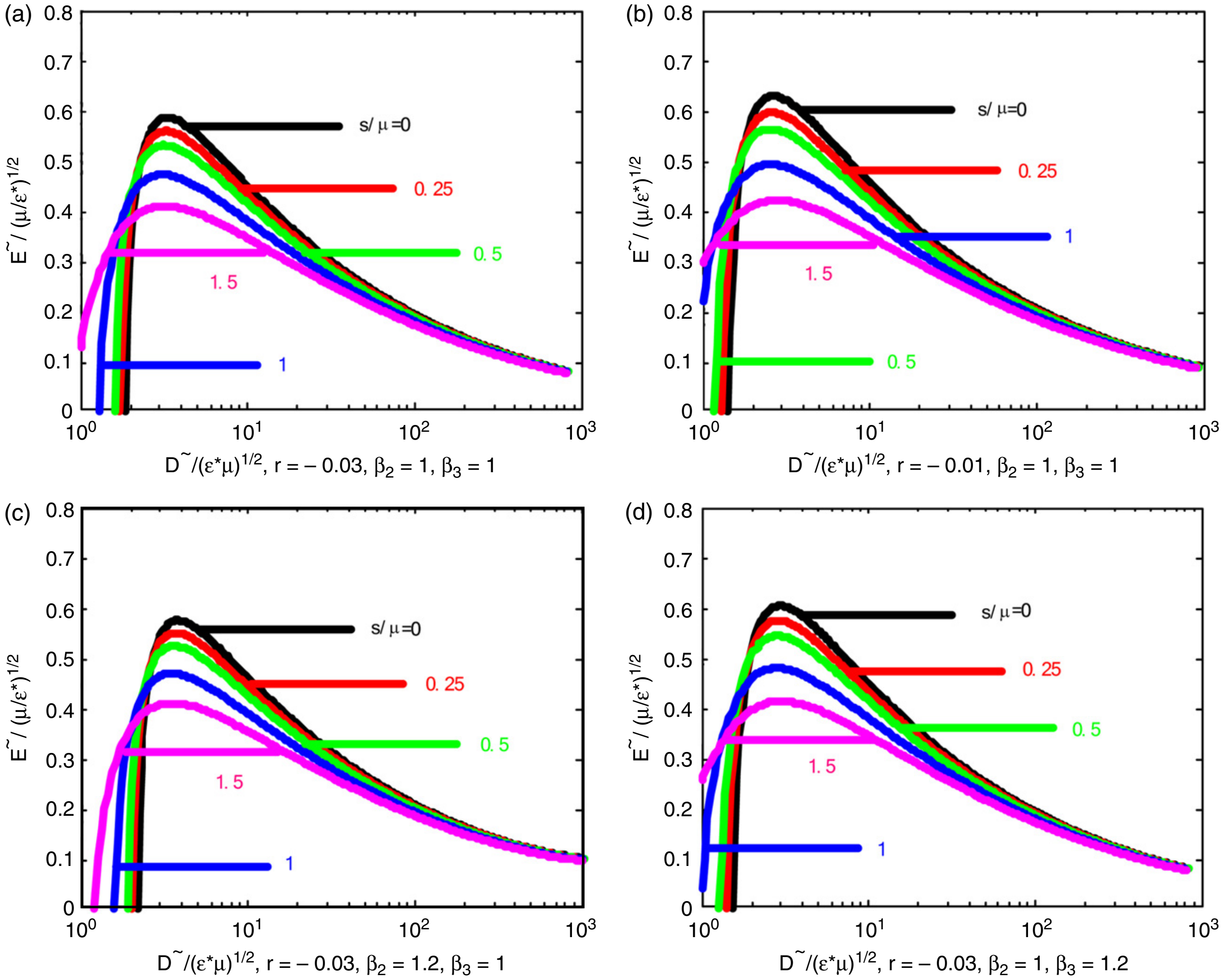

Figure 5. The relationship between the nominal electric displacement and the nominal electric field of silicon with uniformly mixed PMN–PT for various values of $\frac{s}{\mu}$, r, β_2 and β_3, when the stretches are equal biaxial $\lambda_1 = \lambda_2 = \lambda$.

Figure 6 illustrates the electromechanical stability performance of different compressible dielectric elastomer materials or structures under the unequal biaxial condition. The critical nominal electric field of the composite increases along with the decreasing ratio between principal planar stretches, q, the comparative stability performance of the material is even higher. Let $q = 0.9$ and 1.1, the corresponding critical nominal electric fields are $0.6025\sqrt{\mu(h_1, h_2)/\varepsilon^*(h_1, h_2)}$, $0.5451\sqrt{\mu(h_1, h_2)/\varepsilon^*(h_1, h_2)}$, the corresponding critical stretches are 1.58, 1.57. For representative values above, when $q = 0.9$ and 1.1, the corresponding critical nominal electric fields are 1.79×10^7 V m^{-1}, 1.75×10^7 V m^{-1}, 1.74×10^7 V m^{-1}, 1.41×10^7 V m^{-1}; 1.62×10^7 V m^{-1}, 1.58×10^7 V m^{-1}, 1.57×10^7 V m^{-1}, 1.28×10^7 V m^{-1}, respectively.

The critical nominal electric field can be expressed as $\alpha_1\sqrt{\frac{\mu}{\varepsilon}}$ of ideal dielectric elastomers, α_1 is the material parameter. For the neo-Hookean model applied by Suo [4], $\alpha_1 = 0.345$, where $\mu = 1 \times 10^6$ N m^{-2}, $\varepsilon = 4 \times 10^{-11}$ F m^{-1}, for the Mooney–Rivlin model applied by Liu [4], $\alpha_1 = 1.324$, where $\mu = 0.25 \times 10^6$ N m^{-2}, the magnitude is 10^8 V m^{-1} [4, 9, 12]. In this study, For dielectric elastomer composite, the critical nominal electric field is $\alpha_2\sqrt{\frac{\mu(v \text{ or } h)}{\varepsilon(v \text{ or } h)}}$, where $\mu(v$ or $h)$ and $\varepsilon(v$ or $h)$ are functions of the particle content, the magnitude is 10^7 V m^{-1}. The results means that the critical electric field decreases when the PMN–PT content increases or the electrostrictive coefficients r increase, i.e. the stability of the PMN–PT filled silicone decreases.

5. Electro-active deformation test of silicone composite

The numerical results mentioned above are validated by the following electrostrictive deformation experiments on silicone composite filled with particles. In our previous research [16], silicone (BJB TC5005 A–B/C) and barium titanate were used to synthesize new silicone composite with high permittivity and high elastic modulus. The silicone composites are A + B + 30%C + different mass ratio of $BaTiO_3$ (from 0% to 10%, with a increase of 2%), and A, B and C represent silicone glue, catalyst, and plasticizer respectively. For a specific explanation, A + B + 30%C + 10%$BaTiO_3$ denotes

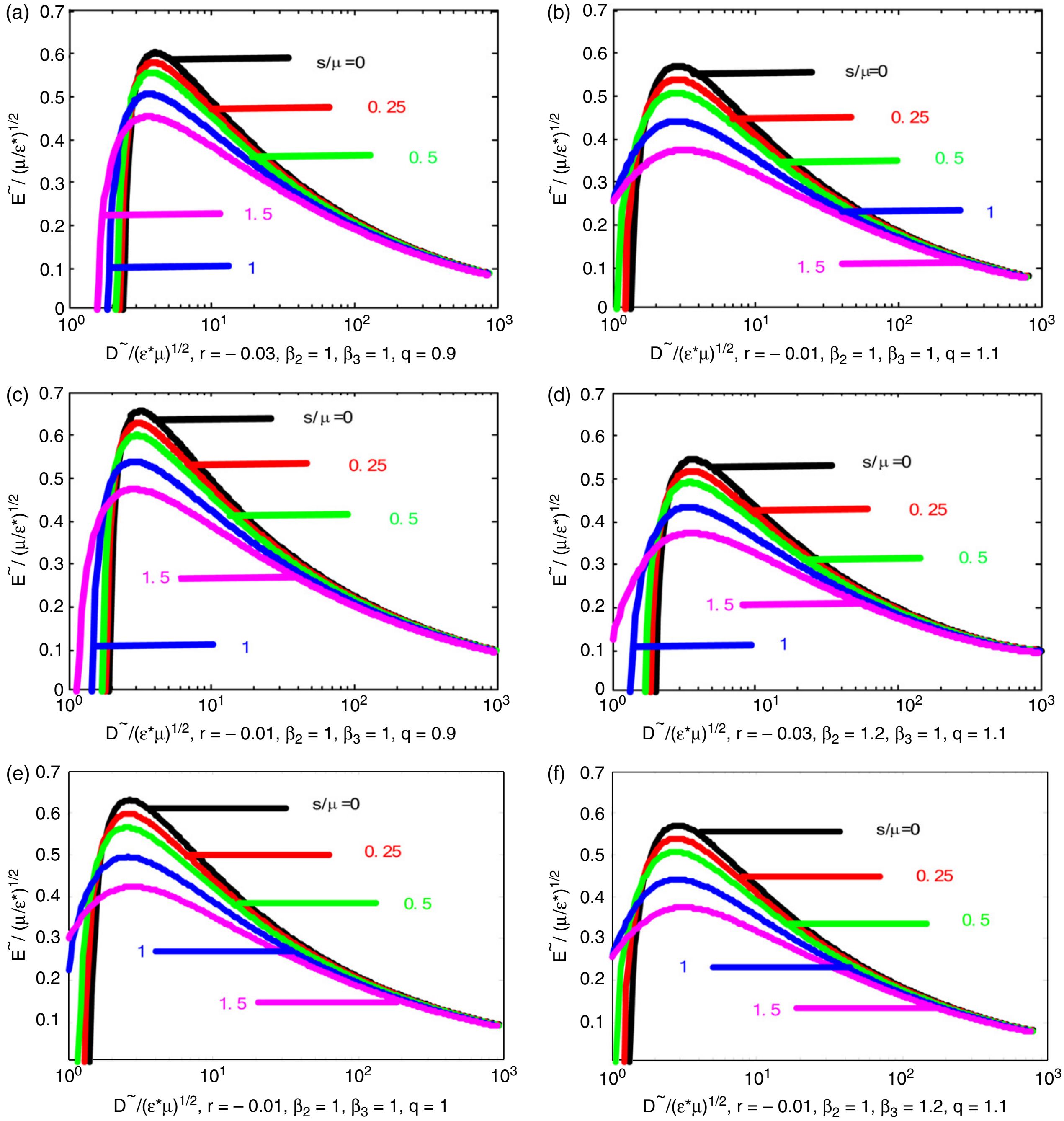

Figure 6. The relationship between the nominal electric displacement and the nominal electric field of silicon with uniformly mixed PMN–PT for various values of $\frac{s}{\mu}$, r, β_2 and β_3, when the stretches are equal biaxial $\lambda_1 = \lambda_2 = \lambda$.

that the mass ratio of $M_A/M_B = 10$, $M_C/M_{A+B} = 0.3$ and $M_{BaTiO_3}/M_{A+B+30\%C} = 0.1$.

The silicone composite film actuators obtained from the matrix with different percentages of the barium titanate all have a size of $100 \times 100 \times 0.5$ mm^3. The carbon grease is applied in a round area with a radius of 30 mm. The test method applied in the previous research is used to investigate the relation between the electric field and the electrostrictive deformation [16]. As shown in figure 7, the breakdown electric field declines along with barium titanate content increasing, which coincides with the theoretical results mentioned above. And the maximum area strain declining results from the declining hyperelasticity performance with elastic modulus increasing. But under the same electric field, the electrostrictive area strain of composite silicone increases with barium titanate content increasing, and this phenomenon is explained as the filled barium titanate enhances the dielectric performance and reduces the breakdown voltage.

6. Conclusions

In this paper, the linear expression of permittivity of silicone filled with PMN–PT particles, which depends on the PMN–PT content and electrostriction deformations, is proposed to

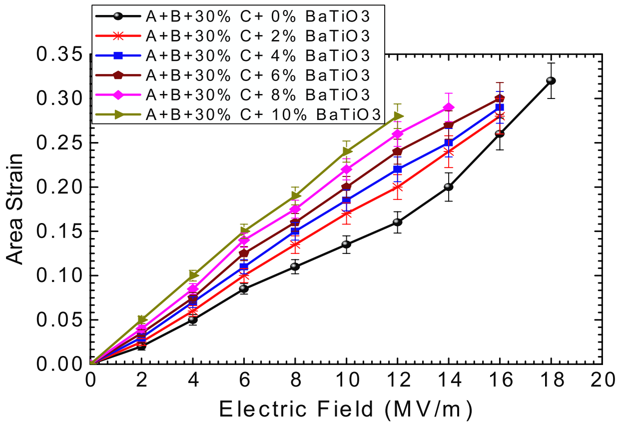

Figure 7. Electric field versus area strain at various barium titanate contents with 30% C.

construct the electric energy density function. The linear expression of permittivity includes: the expression of the permittivity based on experimental test and the permittivity based on the theory of composite material. The influence of PMN–PT on the stability performance of electro-active silicone is analyzed by applying the combination of the electric energy density function and the Knowles elastic strain energy function. The results indicate that the critical electric field decreases when the PMN–PT weight percentage content factor $c(v)$ increases or the electrostrictive coefficients r increase, i.e. the stability of the PMN–PT filled silicone decreases. Such results are meaningful for us to know more about the influence of the filled particles on the stability performance of silicone. Furthermore, these results are useful to guide the design and manufacturing of sensors and actuators based on composite dielectric elastomers.

References

[1] Pelrine R, Kornbluh R, Pei Q B and Joseph J 2000 *Science* **287** 836

[2] Liu L, Fan J, Zhang Z, Shi L, Liu Y and Leng J 2008 *Adv. Mater. Res.* **47–50** 298–301

[3] Gallone G, Carpi F, Rossi D D, Levita G and Marchetti A 2007 *Mater. Sci. Eng.* C **27** 110

[4] Zhao X and Suo Z 2007 *Appl. Phys. Lett.* **91** 061921

[5] Zhao X, Hong W and Suo Z 2007 *Phys. Rev.* B **76** 134113

[6] Suo Z, Zhao X and Greene W H 2008 *J. Mech. Phys. Solids* **56** 476

[7] Zhou J, Hong W, Zhao X, Zhang Z and Suo Z 2008 *Int. J. Solids Struct.* **45** 3739

[8] Norrisa A N 2007 *Appl. Phys. Lett.* **92** 026101

[9] Liu Y, Liu L, Zhang Z, Shi L and Leng J 2008 *Appl. Phys. Lett.* **93** 106101

[10] Díaz-Calleja R, Riande E and Sanchis M J 2008 *Appl. Phys. Lett.* **93** 101902

[11] Liu Y, Liu L, Sun S, Shi L and Leng J 2009 *Appl. Phys. Lett.* **94** 096101

[12] Leng J, Liu L, Liu Y, Yu K and Sun S 2009 *Appl. Phys. Lett.* **94** 211901

[13] Zhao X and Suo Z 2008 *J. Appl. Phys.* **101** 123530

[14] Kofod G, Sommer-Larsen P, Kronbluh R and Pelrine R 2003 *J. Intell. Mater. Syst. Struct.* **14** 787

[15] Liu Y, Liu L, Leng J, Yu K and Sun S 2009 *Appl. Phys. Lett.* **95** 019902

[16] Liu Y, Liu L, Zhang Z and Leng J 2009 *Smart Mater. Struct.* **18** 095024

[17] Liu Y, Liu L, Yu K, Sun S and Leng J 2009 *Smart Mater. Struct.* **18** 095040

[18] Liu Y, Liu L, Sun S, Zhang Z and Leng J 2009 *Sci. China* E **52** 2715–23

[19] Knowles J K 1977 *Int. J. Fract.* **13** 611–39

[20] Liu Y, Liu L, Sun S and Leng J 2010 *Polym. Int.* **59** 371–7

[21] Wang H M and Ding H J 2006 *Struct. Eng. Mech.* **23** 525–42

[22] Suo Z and Zhu J 2009 *Appl. Phys. Lett.* **95** 232909

[23] Koh S A, Zhao X and Suo Z 2009 *Appl. Phys. Lett.* **94** 262902

[24] Zhu J, Cai S and Suo Z 2010 *Polym. Int.* **59** 378–83

[25] Moscardo M, Zhao X, Suo Z and Lapusta Y 2008 *J. Appl. Phys.* **104** 093503

[26] Zhao X and Suo Z 2008 *J. Appl. Phys.* **104** 123530

[27] He T, Zhao X and Suo Z 2009 *J. Appl. Phys.* **106** 083522

[28] Zhao X and Suo Z 2008 *Appl. Phys. Lett.* **93** 251902

[29] Zhao X and Suo Z 2009 *Appl. Phys. Lett.* **95** 031904

[30] Hong W, Zhao X and Suo Z 2009 *Appl. Phys. Lett.* **95** 111901

[31] Wang H M, Ding H J and Chen Y M 2005 *Int. J. Solids Struct.* **42** 85–102

[32] Keplinger C, Kaltenbrunner M, Arnold N and Bauer S 2010 *Proc. Natl Acad. Sci.* **107** 4505

[33] Zhao X and Suo Z 2010 *Phys. Rev. Lett.* **104** 178302

[34] Liu Y, Liu L, Zhang Z, Jiao Y, Sun S and Leng J 2010 *Europhys. Lett.* **90** 36004

[35] Plante J S and Dubowsky S 2006 *Int. J. Solids Struct.* **43** 7727

[36] Gallone G, Galantini F and Carpi F 2010 *Polym. Int.* **59** 400–6

[37] Adrian Koh S J, Keplinger C, Li T, Bauer S and Suo Z 2010 Dielectric elastomer generators: how much energy can be converted, submitted

[38] Zhu J, Cai S and Suo Z 2010 Resonant behavior of a membrane of a dielectric elastomer *Int. J. Solids Struct.* at press

[39] Zhu J, Stoyanov H, Kofod G and Suo Z 2010 Electromechanical instability of a dielectric elastomer tube actuator *J. Appl. Phys.* at press

[40] Zhu J, Li T, Cai S and Suo Z 2010 Snap-through expansion of a gas bubble in an elastomer, submitted