Received: 15 June 2009 Revised: 21 October 2009 Accepted: 16 November 2009 Published online in Wiley Interscience: 3 February 2010

(www.interscience.wiley.com) DOI 10.1002/pi.2786

Electromechanical stability of a Mooney–Rivlin-type dielectric elastomer with nonlinear variable permittivity

Yanju Liu,[a*] Liwu Liu,[a] Shouhua Sun[a] and Jinsong Leng[b*]

Abstract

The electromechanical stability of a Mooney–Rivlin-type dielectric elastomer undergoing large deformation with nonlinear permittivity is investigated. The stability is analyzed by applying a new kind of free energy model, which couples Ogden elastic strain energy and electric field energy density with nonlinear permittivity. Then, nominal electric field and nominal electric displacement of the dielectric elastomer are introduced. Based on this, the electromechanical stability of the Mooney–Rivlin-type dielectric elastomer is analyzed by simplifying the Ogden elastic strain energy. The critical breakdown electric fields under the conditions of two stretching ratios and various material constant ratios k ($n = km$, where m and n are material constants in the Ogden model, determined experimentally) are also obtained. According to the simulation results, for a larger dimensionless constant k of the dielectric material, the critical nominal electric field is higher, the corresponding dielectric elastomer or structure is more stable and the electromechanical stability of the dielectric elastomer is proved to be markedly enhanced by a pre-stretching process. These results agree well with experimental data and can be used as guidance in the design and fabrication of dielectric elastomer actuators.

Keywords: Mooney–Rivlin-type dielectric elastomer; electromechanical stability; Ogden elastic strain energy; nonlinear permittivity

INTRODUCTION

A dielectric elastomer (DE) film sandwiched between two compliant electrodes, when subjected to a high electric field, will expand in-plane and contract in the thickness direction[1–11] due to the electrostatic force between the two compliant electrodes. The thickness contraction leads to a higher value of the electric field; when the electric field reaches the critical breakdown level, the DE becomes electromechanically unstable.

In recent years, the analysis of the electromechanical stability of DEs has become increasingly thorough and concrete, after Suo and Zhao and others proposed the electromechanical stability theory of DEs.[12–27]

Zhao and Suo[12] and Zhao *et al.*[13] proposed that a discretionary free energy function can be used to analyze the electromechanical stability of a DE. As an example, the elastic strain energy function with one material constant was used to analyze the electromechanical stability of an ideal elastic elastomer subjected to both equal biaxial stresses and unequal biaxial stresses. The results illustrated the relation between the nominal electric displacement and the nominal electric field. It was the first time that the theoretical predictions concerning the fact that pre-stretching could enhance a DE's stability were observed experimentally. Meanwhile, the critical breakdown electric field strength was predicted using this method.

Norrisa used the Ogden elastic strain energy model to analyze elastic elastomer stability.[16] The relations among critical actual electric field, nominal strain and the pre-stretching ratio of DEs were obtained accurately. Simultaneously, as a particular case, a model named neo-Hookean which was a simplified version of the Ogden model was introduced to give more accurate results.

Liu *et al*. studied the stability of DEs using an elastic strain energy function with two material constants and found the ratio of the two constants could be used to represent the stability of different types of DE.[17] The relation between the nominal electric displacement and nominal electric field of different DEs was derived directly.

Further research was done on the stability of neo-Hookean silicone-based elastomers by Díaz-Calleja's group.[18] The Hessian matrix of DEs under two special conditions was deduced. Furthermore, the stable and unstable domains of DEs were determined. These results can help understand the stability performance of neo-Hookean silicone more thoroughly. Recently, we studied the stable domain of Mooney–Rivlin silicone, which provides useful theoretical guidance for the research on this kind of material.[19]

In much research work on the electromechanical stability analysis of DE actuators, the DE permittivity is assumed to be a constant. This is true if the DE undergoes only limited deformation.

* Correspondence to: Yanju Liu PO Box 301, Department of Astronautical Science and Mechanics, No. 92 West Dazhi Street, Harbin Institute of Technology (HIT), Harbin 150001, PR China. E-mail: yj_liu@hit.edu.cn

Jinsong Leng, PO Box 3011, Centre for Composite Materials, No. 2 YiKuang Street, Science Park of Harbin Institute of Technology (HIT), Harbin 150001, PR China. E-mail: lengjs@hit.edu.cn

a PO Box 301, Department of Astronautical Science and Mechanics, No. 92 West Dazhi Street, Harbin Institute of Technology (HIT), Harbin 150001, PR China

b PO Box 3011, Centre for Composite Materials, No. 2 YiKuang Street, Science Park of Harbin Institute of Technology (HIT), Harbin 150001, PR China

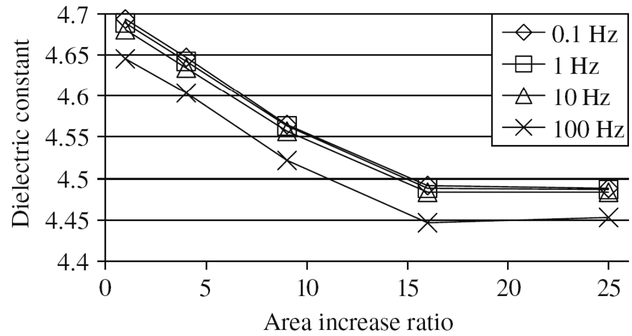

Figure 1. Relative dielectric constant of VHB 4910-type acrylic as it is stretched.

In their recent paper, Zhao and Suo used a permittivity which was a linear fitted function of experimental data in analyzing the mechanical behavior and stability of a DE undergoing large deformation.[22]

Actually, a typical DE is a kind of crosslinked polymer. The structural symmetry of the macromolecule, the degree of crosslinking, along with the tensile deformation can affect the dielectric permittivity enormously. For DEs with higher degree of crosslinking, or higher degree of molecular structural symmetry, the permittivity is relatively low. In addition, stretching can guide the macromolecule to be arranged in order, which can increase the intermolecular forces and reduce the activities of polar groups; as a result, the permittivity will decrease. However, if the degree of crosslinking is low and the deformation is well below the extension limit, the molecular units in the polymers can be polarized as freely as in a polymeric liquid. In this case the corresponding permittivity is unaffected by the deformation.

Based on all the research mentioned above, the Ogden elastic strain energy function with two material constants are used to analyze the electromechanical stability of Mooney–Rivlin-type silicone with nonlinear variable permittivity under two special loading conditions.

THEORETICAL

Recent experimental research results have shown that the dielectric permittivity of a DE changes while undergoing large deformation.[5,23] Kofod *et al*. measured the dielectric constant at various frequencies as a function of area increase ratio, as shown in Fig. 1,[23] which shows the decrease of the dielectric constant of the acrylic polymer VHB 4910, from 3M Company (capable of large strains, high energy densities, high efficiency, high responsive speed as well as good reliability and durability; VHB 4910 has been widely explored for use in actuators of different configurations), approaching a limit value as the frequency and area ratio increase. According to Kofod *et al*.,[23] the permittivity of the DE is variable and it is a function of the area increase ratio which depends on the stretch ratio. In this research, we suppose that the volume of DEs is incompressible.

An electric energy density function with variable permittivity is introduced as follows:

$$W_1(\lambda_1,\lambda_2,\lambda_3,D^{\sim}) = \frac{D^2}{2\varepsilon(\lambda_1,\lambda_2,\lambda_3)} \tag{1}$$

where $D = \lambda_3 D^{\sim}$ and D is the true electric displacement. Further, the electric energy density function $W_1(\lambda_1,\lambda_2,\lambda_3,D^{\sim})$ together with the elastic strain energy function $W_0(\lambda_1,\lambda_2,\lambda_3)$ constitute the system's free energy:

$$W(\lambda_1,\lambda_2,\lambda_3,D^{\sim}) = W_0(\lambda_1,\lambda_2,\lambda_3) + W_1(\lambda_1,\lambda_2,\lambda_3,D^{\sim}) \tag{2}$$

where λ_i, $i = 1, 2, 3$, represent the principal stretch ratio after deformation and $D^{\sim}$ is the nominal electric displacement.

Based on the data of Kofod *et al.*[23] for VHB 4910 from 3M company is about 3MPa. And for single acrylic acid mon (The elastic modulus for the omer, its molecular formula is $C_3H_4O_2$ with a molecular weight of 72), the following relation holds approximately:

$$\varepsilon(\lambda_1,\lambda_2,\lambda_3) = \begin{cases} (-0.016S^{\sim} + 4.716)\varepsilon_0 = (C_1 S^{\sim} + C_2)\varepsilon_0, & S^{\sim} \le 16 \\ 4.48\varepsilon_0, & S^{\sim} > 16 \end{cases} \tag{3}$$

where ε_0 is the permittivity of free space, and $C_1 = -0.016$ and $C_2 = 4.716$ for VHB 4910. Note here $S^{\sim}$ is the area increase ratio and has a critical value of 16, $S^{\sim} = (1+\lambda_1)(1+\lambda_2)$. By considering the incompressibility of DEs, i.e. $\lambda_1\lambda_2\lambda_3 = 1$, we get $\lambda_3 = 1/\lambda_1\lambda_2$.

The Ogden model was proposed by Ogden in 1972. According to this model, the elastic strain energy function can be written as

$$W_0(\lambda_1,\lambda_2) = \sum_{p=1}^{N} \frac{\mu_p}{\alpha_p}\left(\lambda_1^{\alpha_p} + \lambda_2^{\alpha_p} + \lambda_1^{-\alpha_p}\lambda_2^{-\alpha_p} - 3\right) \tag{4}$$

where μ_p is a material constant determined by experiments and α_p is a constant (positive or negative real number). For $S^{\sim} \le 16$, the corresponding system's free energy function is

$$W(\lambda_1,\lambda_2,D^{\sim}) = \sum_{p=1}^{N} \frac{\mu_p}{\alpha_p}\left(\lambda_1^{\alpha_p} + \lambda_2^{\alpha_p} + \lambda_1^{-\alpha_p}\lambda_2^{-\alpha_p} - 3\right) + \frac{D^{\sim 2}}{2\lambda_1^{2}\lambda_2^{2}(C_1 S^{\sim} + C_2)\varepsilon_0} \tag{5}$$

Hence the nominal stress and nominal electric field can be derived as

$$s_1 = \frac{\partial W}{\partial \lambda_1} = \sum_{p=1}^{N} \mu_p\left(\lambda_1^{\alpha_p - 1} - \lambda_1^{-\alpha_p - 1}\lambda_2^{-\alpha_p}\right) - \frac{D^{\sim 2}}{2\varepsilon_0}\,\frac{2(C_1 S^{\sim} + C_2)\lambda_1^{-1} + C_1(\lambda_2 - 1)}{\lambda_1^{2}\lambda_2^{2}(C_1 S^{\sim} + C_2)^2} \tag{6}$$

$$s_2 = \frac{\partial W}{\partial \lambda_2} = \sum_{p=1}^{N} \mu_p\left(\lambda_2^{\alpha_p - 1} - \lambda_2^{-\alpha_p - 1}\lambda_1^{-\alpha_p}\right) - \frac{D^{\sim 2}}{2\varepsilon_0}\,\frac{2(C_1 S^{\sim} + C_2)\lambda_2^{-1} + C_1(\lambda_1 - 1)}{\lambda_1^{2}\lambda_2^{2}(C_1 S^{\sim} + C_2)^2} \tag{7}$$

$$E^{\sim} = \frac{\partial W}{\partial D^{\sim}} = \frac{D^{\sim}}{\varepsilon_0(C_1 S^{\sim} + C_2)}\lambda_1^{-2}\lambda_2^{-2} \tag{8}$$

The Hessian matrix is as follows:

$$H=\begin{bmatrix}\frac{\partial^2 W}{\partial\lambda_1^2} & \frac{\partial^2 W}{\partial\lambda_1\partial\lambda_2} & \frac{\partial^2 W}{\partial\lambda_1\partial\tilde{D}}\\ \frac{\partial^2 W}{\partial\lambda_1\partial\lambda_2} & \frac{\partial^2 W}{\partial\lambda_2^2} & \frac{\partial^2 W}{\partial\lambda_2\partial\tilde{D}}\\ \frac{\partial^2 W}{\partial\lambda_1\partial\tilde{D}} & \frac{\partial^2 W}{\partial\lambda_2\partial\tilde{D}} & \frac{\partial^2 W}{\partial\tilde{D}^2}\end{bmatrix}=\begin{bmatrix} \begin{array}{l}\sum_{p=1}^{N}\mu_p[(\alpha_p-1)\lambda_1^{\alpha_p-2}+(\alpha_p+1)\lambda_1^{-\alpha_p-2}\lambda_2^{-\alpha_p}]+\\ \frac{\tilde{D}^2}{\varepsilon_0}[\frac{6\lambda_1^{-1}(C_1\tilde{S}+C_2)+2C_1(\lambda_2+1)}{(C_1\tilde{S}+C_2)^2\lambda_1^3\lambda_2^2}+\\ \frac{2C_1(C_1\tilde{S}+C_2)(\lambda_1^{-1}(C_1\tilde{S}+C_2)+C_1(\lambda_2+1)^2)}{(C_1\tilde{S}+C_2)^4\lambda_1^2\lambda_2^2}]\end{array} & \begin{array}{l}\sum_{p=1}^{N}\mu_p\alpha_p\lambda_1^{-\alpha_p-1}\lambda_2^{-\alpha_p-1}+\\ \frac{\tilde{D}^2}{2\varepsilon_0}[\frac{4\lambda_2^{-1}(C_1\tilde{S}+C_2)+2C_1(\lambda_1+1)}{(C_1\tilde{S}+C_2)^2\lambda_1^3\lambda_2^2}+\\ \frac{C_1(C_1\tilde{S}+C_2)((1+2\lambda_2^{-1})(C_1\tilde{S}+C_2)+2C_1\tilde{S})}{(C_1\tilde{S}+C_2)^4\lambda_1^2\lambda_2^2}]\end{array} & -\frac{\tilde{D}}{\varepsilon_0}\frac{2(C_1\tilde{S}+C_2)\lambda_1^{-1}+C_1(\lambda_2+1)}{\lambda_1^2\lambda_2^2(C_1\tilde{S}+C_2)^2}\\ \begin{array}{l}\sum_{p=1}^{N}\mu_p\alpha_p\lambda_1^{-\alpha_p-1}\lambda_2^{-\alpha_p-1}+\\ \frac{\tilde{D}^2}{2\varepsilon_0}[\frac{4\lambda_2^{-1}(C_1\tilde{S}+C_2)+2C_1(\lambda_1+1)}{(C_1\tilde{S}+C_2)^2\lambda_1^3\lambda_2^2}+\\ \frac{C_1(C_1\tilde{S}+C_2)((1+2\lambda_2^{-1})(C_1\tilde{S}+C_2)+2C_1\tilde{S})}{(C_1\tilde{S}+C_2)^4\lambda_1^2\lambda_2^2}]\end{array} & \begin{array}{l}\sum_{p=1}^{N}\mu_p[(\alpha_p-1)\lambda_2^{\alpha_p-2}+(\alpha_p+1)\lambda_2^{-\alpha_p-2}\lambda_1^{-\alpha_p}]+\\ \frac{\tilde{D}^2}{\varepsilon_0}[\frac{6\lambda_2^{-1}(C_1\tilde{S}+C_2)+2C_1(\lambda_1+1)}{(C_1\tilde{S}+C_2)^2\lambda_1^2\lambda_2^3}+\\ \frac{2C_1(C_1\tilde{S}+C_2)(\lambda_2^{-1}(C_1\tilde{S}+C_2)+C_1(\lambda_1+1)^2)}{(C_1\tilde{S}+C_2)^4\lambda_1^2\lambda_2^2}]\end{array} & -\frac{\tilde{D}}{\varepsilon_0}\frac{2(C_1\tilde{S}+C_2)\lambda_2^{-1}+C_1(\lambda_1+1)}{\lambda_1^2\lambda_2^2(C_1\tilde{S}+C_2)^2}\\ -\frac{\tilde{D}}{\varepsilon_0}\frac{2(C_1\tilde{S}+C_2)\lambda_1^{-1}+C_1(\lambda_2+1)}{\lambda_1^2\lambda_2^2(C_1\tilde{S}+C_2)^2} & -\frac{\tilde{D}}{\varepsilon_0}\frac{2(C_1\tilde{S}+C_2)\lambda_2^{-1}+C_1(\lambda_1+1)}{\lambda_1^2\lambda_2^2(C_1\tilde{S}+C_2)^2} & \frac{1}{\varepsilon_0(C_1\tilde{S}+C_2)}\lambda_1^{-2}\lambda_2^{-2}\end{bmatrix} \quad (9)$$

When $\tilde{S} > 16$, the free energy, nominal stress and nominal electric field of the system can be expressed as

$$W(\lambda_1,\lambda_2,\tilde{D})=\sum_{p=1}^{N}\frac{\mu_p}{\alpha_p}({\lambda_1}^{\alpha_p}+{\lambda_2}^{\alpha_p}+{\lambda_1}^{-\alpha_p}{\lambda_2}^{-\alpha_p}-3)+\frac{\tilde{D}^2}{8.96\varepsilon_0}{\lambda_1}^{-2}{\lambda_2}^{-2} \quad (10)$$

$$s_1=\frac{\partial W}{\partial\lambda_1}=\sum_{p=1}^{N}\mu_p({\lambda_1}^{\alpha_p-1}-{\lambda_1}^{-\alpha_p-1}{\lambda_2}^{-\alpha_p})-\frac{\tilde{D}^2}{4.48\varepsilon_0}{\lambda_1}^{-3}{\lambda_2}^{-2} \quad (11)$$

$$s_2=\frac{\partial W}{\partial\lambda_2}=\sum_{p=1}^{N}\mu_p({\lambda_2}^{\alpha_p-1}-{\lambda_2}^{-\alpha_p-1}{\lambda_1}^{-\alpha_p})-\frac{\tilde{D}^2}{4.48\varepsilon_0}{\lambda_1}^{-2}{\lambda_2}^{-3} \quad (12)$$

$$\tilde{E}=\frac{\partial W}{\partial\tilde{D}}=\frac{\tilde{D}}{4.48\varepsilon_0}{\lambda_1}^{-2}{\lambda_2}^{-2} \quad (13)$$

The Hessian matrix is

We postulate that $\mu_1 = k_2\mu_2 = k_3\mu_3 = \cdots k_N\mu_N$, $k_2, k_3, \ldots, k_N$ are material constants. By substituting them into Eqns (6)–(8) and (11)–(13), the nominal electric field and the nominal electrical displacement can be evaluated:

where $N = [2C_1(1+\lambda)^2 + 2C_2 + C_1(\lambda^2+\lambda)]/[C_1(1+\lambda)^2 + C_2]^2$, $P = [2C_1(1+\lambda)^2 + 2C_2 + C_1(\lambda^2+\lambda)]\lambda^3$ and $c = 4.48$.

Equation (15) illustrates the electromechanical stability analysis method by applying Ogden elastic strain energy when the DE undergoes large deformation under the condition of two kinds of stretching ratios. Evidently they are functions taking the stretch ratio λ as the variable parameter. This means that the relationship between the nominal electric field and the nominal electrical displacement can be derived by changing the value of s/μ_2.

Here we consider the condition that the Ogden elastic strain energy formulation is limited to that with two material constants. Let $N = 2$, $\alpha_2 = \alpha_1 = -2$, $\mu_1 = m$ and $\mu_2 = n$. The elastic strain energy functional with two material constants can be written as follows:[17,19,21]

$$\begin{bmatrix} \sum_{p=1}^{N}\mu_p[(\alpha_p-1)\lambda_1^{\alpha_p-2}+(\alpha_p+1)\lambda_1^{-\alpha_p-2}\lambda_2^{-\alpha_p}]+\frac{3\tilde{D}^2}{4.48\varepsilon_0}\lambda_1^{-4}\lambda_2^{-2} & \sum_{p=1}^{N}\mu_p\alpha_p\lambda_1^{-\alpha_p-1}\lambda_2^{-\alpha_p-1}+\frac{\tilde{D}^2}{2.24\varepsilon_0}\lambda_1^{-3}\lambda_2^{-3} & -\frac{\tilde{D}}{2.24\varepsilon_0}\lambda_1^{-3}\lambda_2^{-2}\\ \sum_{p=1}^{N}\mu_p\alpha_p\lambda_1^{-\alpha_p-1}\lambda_2^{-\alpha_p-1}+\frac{\tilde{D}^2}{2.24\varepsilon_0}\lambda_1^{-3}\lambda_2^{-3} & \sum_{p=1}^{N}\mu_p[(\alpha_p-1)\lambda_2^{\alpha_p-2}+(\alpha_p+1)\lambda_2^{-\alpha_p-2}\lambda_1^{-\alpha_p}]+\frac{3\tilde{D}^2}{4.48\varepsilon_0}\lambda_2^{-4}\lambda_1^{-2} & -\frac{\tilde{D}}{2.24\varepsilon_0}\lambda_2^{-3}\lambda_1^{-2}\\ -\frac{\tilde{D}}{2.24\varepsilon_0}\lambda_1^{-3}\lambda_2^{-2} & -\frac{\tilde{D}}{2.24\varepsilon_0}\lambda_2^{-3}\lambda_1^{-2} & \frac{1}{4.48\varepsilon_0}\lambda_1^{-2}\lambda_2^{-2}\end{bmatrix} \quad (14)$$

$$\begin{cases} \left.\begin{array}{l} \frac{\tilde{D}}{\sqrt{\varepsilon_0\mu_2}}=\sqrt{\frac{2k_2}{N}(\lambda^{\alpha_1-1}-\lambda^{-2\alpha_1-1})+\frac{2(\lambda^{\alpha_2-1}-\lambda^{-2\alpha_2-1})}{N}+\frac{2k_2}{Nk_3}(\lambda^{\alpha_3-1}-\lambda^{-2\alpha_3-1})+\cdots+\frac{2k_2}{Nk_N}(\lambda^{\alpha_N-1}-\lambda^{-2\alpha_N-1})-\frac{2s}{N\mu_2}}\\ \frac{\tilde{E}}{\sqrt{\varepsilon_0\mu_2}}=\sqrt{\frac{2k_2}{P}(\lambda^{\alpha_1-1}-\lambda^{-2\alpha_1-1})+\frac{2(\lambda^{\alpha_2-1}-\lambda^{-2\alpha_2-1})}{P}+\frac{2k_2}{Pk_3}(\lambda^{\alpha_3-1}-\lambda^{-2\alpha_3-1})+\cdots+\frac{2k_2}{Pk_N}(\lambda^{\alpha_N-1}-\lambda^{-2\alpha_N-1})-\frac{2s}{P\mu_2}} \end{array}\right\}\lambda_1\lambda_2\le\chi \\ \left.\begin{array}{l} \frac{\tilde{D}}{\sqrt{\varepsilon_0\mu_2}}=\sqrt{k_2c(\lambda^{\alpha_1+4}-\lambda^{-2\alpha_1+4})+c(\lambda^{\alpha_2+4}-\lambda^{-2\alpha_2+4})+\frac{ck_2}{k_3}(\lambda^{\alpha_3+4}-\lambda^{-2\alpha_3+4})+\cdots+\frac{ck_2}{k_N}(\lambda^{\alpha_N+4}-\lambda^{-2\alpha_N+4})-\frac{sc}{\mu_2}\lambda^5}\\ \frac{\tilde{E}}{\sqrt{\varepsilon_0\mu_2}}=\sqrt{\frac{k_2}{c}(\lambda^{\alpha_1-4}-\lambda^{-2\alpha_1-4})+\frac{(\lambda^{\alpha_2-4}-\lambda^{-2\alpha_2-4})}{c}+\frac{k_2}{ck_3}(\lambda^{\alpha_3-4}-\lambda^{-2\alpha_3-4})+\cdots+\frac{k_2}{ck_N}(\lambda^{\alpha_N-4}-\lambda^{-2\alpha_N-4})-\frac{s}{c\mu_2}\lambda^{-3}} \end{array}\right\}\lambda_1\lambda_2>\chi \end{cases} \quad (15)$$

$$W_0(\lambda_1, \lambda_2, \lambda_1^{-1}\lambda_2^{-1}) = \frac{m}{2}(\lambda_1{}^2 + \lambda_2{}^2 + \lambda_1{}^{-2}\lambda_2{}^{-2} - 3) + \frac{n}{2}(\lambda_1{}^{-2} + \lambda_2{}^{-2} + \lambda_1{}^2\lambda_2{}^2 - 3) \quad (16)$$

where λ_1, λ_2 denote the in-plane principal stretching ratios, m, n are material constants, which can be determined by experiments, $\tilde{D}$ is the nominal electric displacement and ε denotes the permittivity of the DE. According to Suo's theory,[12–14] $s_1 = \partial W/\partial\lambda_1$, $s_2 = \partial W/\partial\lambda_2$, $\tilde{E} = \partial W/\partial\tilde{D}$ are obtained, where $\tilde{E}$ is the nominal electric field.

For $\tilde{S} \leq 16$, the electric energy density function becomes

$$W_1(\lambda_1, \lambda_2, \lambda_3, \tilde{D}) = W_1(\lambda_1, \lambda_2, \tilde{D}) = \frac{\tilde{D}^2}{2\lambda_1{}^2\lambda_2{}^2(C_1\tilde{S} + C_2)\varepsilon_0} \quad (17)$$

and the corresponding system's free energy function is

$$W(\lambda_1, \lambda_2, \tilde{D}) = \frac{m}{2}(\lambda_1{}^2 + \lambda_2{}^2 + \lambda_1{}^{-2}\lambda_2{}^{-2} - 3) + \frac{n}{2}(\lambda_1{}^{-2} + \lambda_2{}^{-2} + \lambda_1{}^2\lambda_2^2 - 3) + \frac{\tilde{D}}{2\varepsilon_0(C_1\tilde{S} + C_2)}\lambda_1{}^{-2}\lambda_2{}^{-2} \quad (18)$$

Hence the nominal stress and nominal electric field can be derived as

$$s_1 = \frac{\partial W}{\partial\lambda_1} = m(\lambda_1 - \lambda_1^{-3}\lambda_2^{-2}) + n(-\lambda_1^{-3} + \lambda_1\lambda_2^2) - \frac{\tilde{D}^2}{2\varepsilon_0}\frac{2(C_1\tilde{S} + C_2)\lambda_1^{-1} + C_1(\lambda_2 + 1)}{\lambda_1^2\lambda_2^2(C_1\tilde{S} + C_2)^2} \quad (19)$$

$$s_2 = \frac{\partial W}{\partial\lambda_2} = m(\lambda_2 - \lambda_2^{-3}\lambda_1^{-2}) + n(-\lambda_2^{-3} + \lambda_2\lambda_1^2) - \frac{\tilde{D}^2}{2\varepsilon_0}\frac{2(C_1\tilde{S} + C_2)\lambda_2^{-1} + C_1(\lambda_1 + 1)}{\lambda_1^2\lambda_2^2(C_1\tilde{S} + C_2)^2} \quad (20)$$

$$\tilde{E} = \frac{\partial W}{\partial\tilde{D}} = \frac{\tilde{D}}{\varepsilon_0(C_1\tilde{S} + C_2)}\lambda_1{}^{-2}\lambda_2{}^{-2} \quad (21)$$

The Hessian matrix is then

$$\begin{bmatrix} m(1+3\lambda_1^{-4}\lambda_2^{-2})+n(3\lambda_1^{-4}+\lambda_2^{-2})+\frac{\tilde{D}^2}{\varepsilon_0}\left[\frac{6\lambda_1^{-1}(C_1\tilde{S}+C_2)+2C_1(\lambda_2+1)}{(C_1\tilde{S}+C_2)^2\lambda_1^3\lambda_2^2} + \frac{2C_1(C_1\tilde{S}+C_2)(\lambda_1^{-1}(C_1\tilde{S}+C_2)C_1(\lambda_2+1)^2}{(C_1\tilde{S}+C_2)^4\lambda_1^2\lambda_2^2}\right] & 2m\lambda_1^{-3}\lambda_2^{-3}+2n\lambda_1\lambda_2+\frac{\tilde{D}^2}{2\varepsilon_0}\left[\frac{4\lambda_2^{-1}(C_1\tilde{S}+C_2)+2C_1(\lambda_1+1)}{(C_1\tilde{S}+C_2)^2\lambda_1^3\lambda_2^2} + \frac{C_1(C_1\tilde{S}+C_2)((1+2\lambda_2^{-1})(C_1\tilde{S}+C_2)2C_1\tilde{S})}{(C_1\tilde{S}+C_2)^4\lambda_1^2\lambda_2^2}\right] & -\frac{\tilde{D}}{\varepsilon_0}\frac{2(C_1\tilde{S}+C_2)\lambda_1^{-1}+C_1(\lambda_2+1)}{\lambda_1^2\lambda_2^2(C_1\tilde{S}+C_2)^2} \\ 2m\lambda_1^{-3}\lambda_2^{-3}+2n\lambda_1\lambda_2+\frac{\tilde{D}^2}{2\varepsilon_0}\left[\frac{4\lambda_2^{-1}(C_1\tilde{S}+C_2)+2C_1(\lambda_1+1)}{(C_1\tilde{S}+C_2)^2\lambda_1^3\lambda_2^2} + \frac{C_1(C_1\tilde{S}+C_2)((1+2\lambda_2^{-1})(C_1\tilde{S}+C_2)2C_1\tilde{S})}{(C_1\tilde{S}+C_2)^4\lambda_1^2\lambda_2^2}\right] & m(1+3\lambda_2^{-4}\lambda_1^{-2})+n(3\lambda_2^{-4}+\lambda_1^2)+\frac{\tilde{D}^2}{\varepsilon_0}\left[\frac{6\lambda_2^{-1}(C_1\tilde{S}+C_2)+2C_1(\lambda_1+1)}{(C_1\tilde{S}+C_2)^2\lambda_1^2\lambda_2^3} + \frac{2C_1(C_1\tilde{S}+C_2)(\lambda_2^{-1}(C_1\tilde{S}+C_2)C_1(\lambda_1+1)^2}{(C_1\tilde{S}+C_2)^4\lambda_1^2\lambda_2^2}\right] & -\frac{\tilde{D}}{\varepsilon_0}\frac{2(C_1\tilde{S}+C_2)\lambda_2^{-1}+C_1(\lambda_1+1)}{\lambda_1^2\lambda_2^2(C_1\tilde{S}+C_2)^2} \\ -\frac{\tilde{D}}{\varepsilon_0}\frac{2(C_1\tilde{S}+C_2)\lambda_1^{-1}+C_1(\lambda_2+1)}{\lambda_1^2\lambda_2^2(C_1\tilde{S}+C_2)^2} & -\frac{\tilde{D}}{\varepsilon_0}\frac{2(C_1\tilde{S}+C_2)\lambda_2^{-1}+C_1(\lambda_1+1)}{\lambda_1^2\lambda_2^2(C_1\tilde{S}+C_2)^2} & \frac{1}{\varepsilon_0(C_1\tilde{S}+C_2)}\lambda_1^{-2}\lambda_2^{-2} \end{bmatrix} \quad (22)$$

Now consider a special loading case. It is assumed that the DE film is uniformly pre-stretched, $s_1 = s_2 = s$, and $\tilde{S} \leq 16$. We postulate that $n = km$, where k is constant, and factorization of the resulting equation gives

$$(\lambda_1 - \lambda_2)\left\{(1 + \lambda_1{}^3\lambda_2{}^3) + k(\lambda_1{}^2 + \lambda_1\lambda_2 + \lambda_2{}^2 - \lambda_1{}^4\lambda_2{}^4) + \frac{\tilde{D}^2}{2m\varepsilon_0}\left[\frac{2(C_1\tilde{S} + C_2)\lambda_1\lambda_2 + C_1\lambda_1{}^3\lambda_2{}^3}{(C_1\tilde{S} + C_2)^2}\right]\right\} = 0 \quad (23)$$

The solution of the above equation is $\lambda_1 = \lambda_2 = \lambda$, which means that the stretch ratios in the two major directions of the plane are the same when exerting equal-axis pre-stress over the DE.

The equation of nominal stress can be rewritten in another form as

$$\frac{\tilde{D}}{\sqrt{m\varepsilon_0}} = \sqrt{\frac{2(\lambda^6 - 1)}{N} + \frac{2k(\lambda^8 - \lambda^2)}{N} - \frac{2s\lambda^5}{Nm}} \quad (24)$$

where

$$N = \frac{[2C_1(1 + \lambda)^2 + 2C_2 + C_1(\lambda^2 + \lambda)]}{[C_1(1 + \lambda)^2 + C_2]^2}$$

Hence the nominal electric field is

$$\frac{\tilde{E}}{\sqrt{m/\varepsilon_0}} = \sqrt{\frac{2(\lambda - \lambda^{-5}) + 2k(\lambda^3 - \lambda^{-3})}{[2C_1(1 + \lambda)^2 + 2C_2 + C_1(\lambda^2 + \lambda)]\lambda^3} - \frac{2s}{m[2C_1(1 + \lambda)^2 + 2C_2 + C_1(\lambda^2 + \lambda)]\lambda^3}} \quad (25)$$

RESULTS

When k is assigned different values, for the variables in the stretch ratio λ, we can analyze the electromechanical stability of various DEs undergoing large deformation processes. Figure 2 shows the relationship between $\tilde{D}/\sqrt{m\varepsilon_0}$ and $\tilde{E}/\sqrt{m/\varepsilon_0}$ when $k = 1$, 1/2, 1/4 and 1/8. In each case, s/m is set different values (0, 0.5, 1, 1.5, 2, 2.5) so that $\tilde{E}$ will reach peak values. The curves to the left-hand side of these peaks make the Hessian matrix positive, and, conversely, to the right-hand side make the Hessian matrix negative; however, the peaks make the Hessian matrix det (H) = 0. With s/m increasing, the nominal electric field decreases for the various values of the constant k. This shows that pre-stretch can be enforced to improve the stability of DEs.

When $k = 1$, 1/2, 1/4 and 1/8, respectively, the corresponding critical nominal values of electric field are $0.4536\sqrt{m/\varepsilon_0}$, $0.3760\sqrt{m/\varepsilon_0}$, $0.3303\sqrt{m/\varepsilon_0}$ and $0.3057\sqrt{m/\varepsilon_0}$. Let the representative parameters $m = 1 \times 10^6$ Pa, $\varepsilon_0 = 8.85 \times 10^{-12}$ F m^{-1}, then critical nominal values of electric field are 1.53×10^8, 1.26×10^8, 1.11×10^8 and 1.02×10^8 V m^{-1}. The critical nominal electric field of DEs not only corresponds to the results that Suo *et al.* calculated, but also corresponds to the experimental value of the breakdown electric field which has been imposed on the DE material.[12,13]

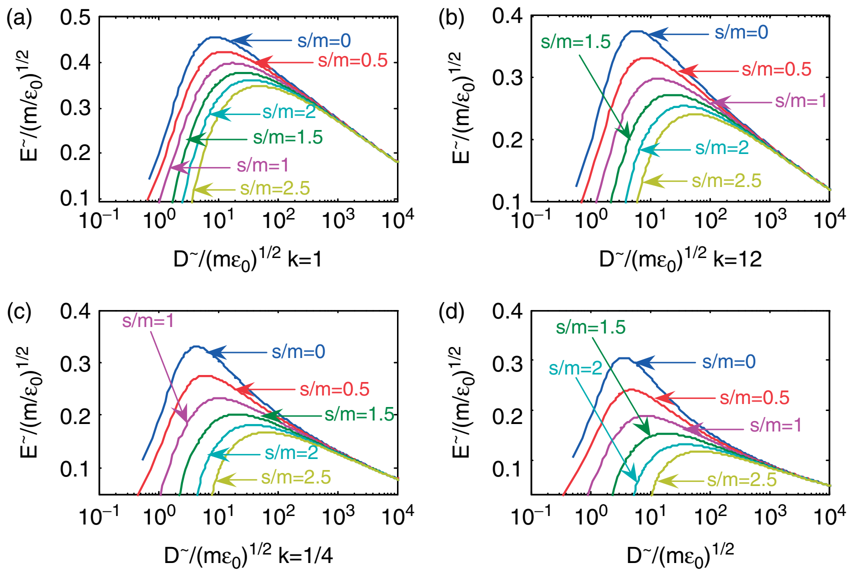

Figure 2. Nominal electric field *versus* nominal electric displacement when $\lambda_1 = \lambda_2 = \lambda, S^{\sim} \leq 16$.

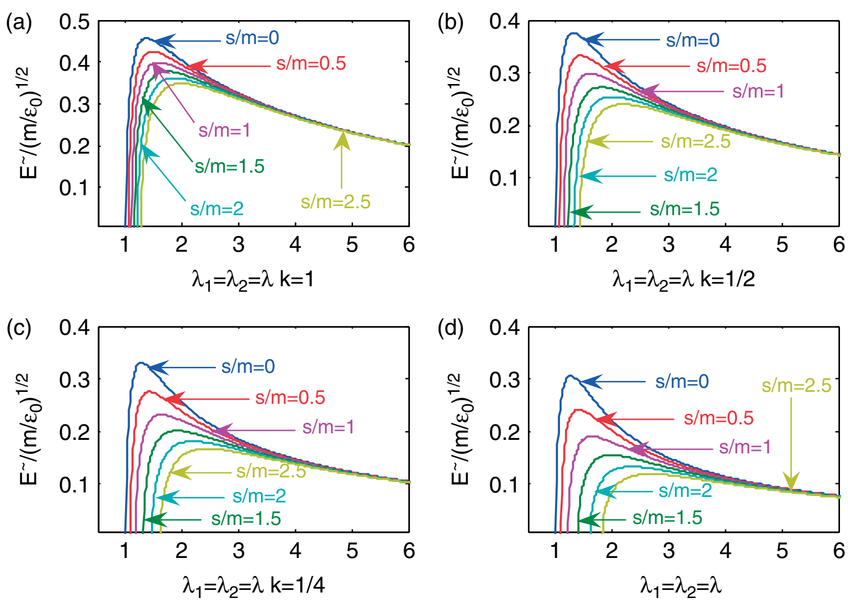

Figure 3. Nominal electric field *versus* stretch ratio when $\lambda_1 = \lambda_2 = \lambda, S^{\sim} \leq 16$.

This stretch ratio point can be obtained as 1.37, 1.32, 1.27 and 1.25, corresponding to thickness strains of 46, 42, 38 and 36%, respectively, which agrees with the fact that it cannot exceed 40% of the experimental value.[12]

As k increases, the critical electric field increases, indicating that a larger value of k for the DE material leads to higher electrical and mechanical stability; the corresponding threshold that can be achieved is higher and the thickness tensile strain rate is also higher.

Figure 3 shows the relation between stretching rate and nominal electric field for various values of k (1, 1/2, 1/4, 1/8). It shows that when s/m increases, $E^{\sim}/\sqrt{m/\varepsilon_0}$ becomes smaller; that is, as the nominal stress increases, the nominal electric field decreases. This shows that imposing pre-tension to DEs can significantly improve their electromechanical stability. The electrostriction experiments on the pre-stretching of DEs by Kofod's group showed that when the two in-plane pre-stretch ratios increase from 0 to 500%, the breakdown electric field increases from 18 to 218 MV m^{-1}, amounting to 1100%.[23] This means that the electromechanical stability is evidently enhanced. Our numeric model applying the Mooney–Rivlin elastic strain energy function produces the same result as obtained by Kofod's group,[23] as well as by Zhao and Suo.[12]

Similarly, for $S^{\sim} > 16$, we have

$$W_1(\lambda_1, \lambda_2, \lambda_3, D^{\sim}) = W_1(\lambda_1, \lambda_2, D^{\sim}) = \frac{D^{\sim 2}}{8.96\lambda_1^2\lambda_2^2\varepsilon_0} \tag{26}$$

Then the free energy, nominal stress and nominal electric field of the system can be expressed as

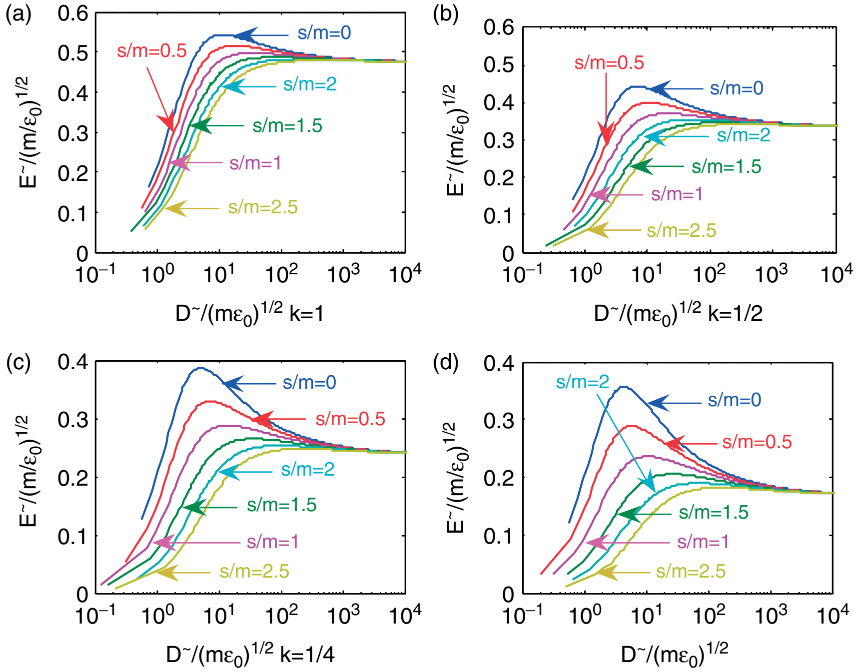

Figure 4. Nominal electric field *versus* nominal electric displacement when $\lambda_1 = \lambda_2 = \lambda, S^{\sim} > 16$.

$$W(\lambda_1,\lambda_2,D^{\sim}) = \frac{m}{2}({\lambda_1}^2+{\lambda_2}^2+{\lambda_1}^{-2}{\lambda_2}^{-2}-3) + \frac{n}{2}({\lambda_1}^{-2}+{\lambda_2}^{-2}+{\lambda_1}^{2}{\lambda_2}^{2}-3) + \frac{D^{\sim 2}}{8.96\varepsilon_0}{\lambda_1}^{-2}{\lambda_2}^{-2} \quad (27)$$

$$s_1 = \frac{\partial W}{\partial \lambda_1} = m(\lambda_1-{\lambda_1}^{-3}{\lambda_2}^{-2}) + n(-{\lambda_1}^{-3}+\lambda_1{\lambda_2}^{2}) - \frac{D^{\sim 2}}{4.48\varepsilon_0}{\lambda_1}^{-3}{\lambda_2}^{-2} \quad (28)$$

$$s_2 = \frac{\partial W}{\partial \lambda_2} = m(\lambda_2-{\lambda_2}^{-3}{\lambda_1}^{-2}) + n(-{\lambda_2}^{-3}+\lambda_2{\lambda_1}^{2}) - \frac{D^{\sim 2}}{4.48\varepsilon_0}{\lambda_1}^{-2}{\lambda_2}^{-3} \quad (29)$$

$$E^{\sim} = \frac{\partial W}{\partial D^{\sim}} = \frac{D^{\sim}}{4.48\varepsilon_0}{\lambda_1}^{-2}{\lambda_2}^{-2} \quad (30)$$

The corresponding Hessian matrix is

Similarly, we now consider an almost identical case except that $S^{\sim} > 16$. To ensure the stresses in the two major directions of the plane are the same, the DE film is pre-stretched uniformly, i.e. $s_1 = s_2 = s$.

Adopting similar processing methods to those mentioned above, we postulate that $n = km$, then process the factorization:

$$(\lambda_1-\lambda_2)\left[\begin{array}{l}(1+{\lambda_1}^3{\lambda_2}^3)+k({\lambda_1}^2+\lambda_1\lambda_2+{\lambda_2}^2-{\lambda_1}^4{\lambda_2}^4)\\ +\dfrac{D^{\sim 2}}{4.48m\varepsilon_0}\end{array}\right] = 0 \quad (32)$$

The corresponding solution is $\lambda_1 = \lambda_2 = \lambda$, which means that the ratios in the two major directions of the plane are the same when applying equal-axis pre-stress over the DE:

$$\frac{D^{\sim}}{\sqrt{m\varepsilon_0}} = \sqrt{4.48\left[(\lambda^6-1)+k(\lambda^8-\lambda^2)-\frac{s}{m}\lambda^5\right]} \quad (33)$$

$$\frac{E^{\sim}}{\sqrt{m/\varepsilon_0}} = \sqrt{\frac{(\lambda^{-2}-\lambda^{-8})}{4.48}+\frac{k(1-\lambda^{-6})}{4.48}-\frac{s}{4.48m}\lambda^{-3}} \quad (34)$$

The relationships between nominal electric field and nominal electric displacement and between nominal electric field and pre-stretch ratio are shown in Figs 4 and 5, respectively. Similar results have been obtained as previously when $S^{\sim} \le 16$. In this case, the values of λ^C are 1.47, 1.38, 1.32 and 1.29 and the corresponding values of strain in the thickness direction are 53, 47, 42 and 40%, and when the non-dimensional parameter is set as $k = 1$, 1/2, 1/4 and 1/8, the maximum values of the nominal electric field are $E^{\sim}_{max} = 0.5424\sqrt{m/\varepsilon_0}$, $0.4424\sqrt{m/\varepsilon_0}$, $0.3862\sqrt{m/\varepsilon_0}$ and $0.3562\sqrt{m/\varepsilon_0}$. Let $m = 1\times10^6$ Pa, $\varepsilon_0 = 8.85\times10^{-12}$ F m^{-1}, then critical nominal values of electric field are 1.82×10^8, 1.48×10^8, 1.29×10^8 and 1.19×10^8 V m^{-1}, respectively. These are consistent with experimental results[23] and the conclusions of Zhao and Suo.[12]

$$H = \begin{bmatrix} m(1+3\lambda_1^{-4}\lambda_2^{-2})+n(3\lambda_1^{-4}+\lambda_2^{2})+\dfrac{3D^{\sim 2}}{4.48\varepsilon_0}\lambda_1^{-4}\lambda_2^{-2} & 2m\lambda_1^{-3}\lambda_2^{-3}+2n\lambda_1\lambda_2+\dfrac{2D^{\sim 2}}{4.48\varepsilon_0}\lambda_1^{-3}\lambda_2^{-3} & -\dfrac{2D^{\sim}}{4.48\varepsilon_0}\lambda_1^{-3}\lambda_2^{-2} \\ 2m\lambda_1^{-3}\lambda_2^{-3}+2n\lambda_1\lambda_2+\dfrac{2D^{\sim 2}}{4.48\varepsilon_0}\lambda_1^{-3}\lambda_2^{-3} & m(1+3\lambda_2^{-4}\lambda_1^{-2})+n(3\lambda_2^{-4}+\lambda_1^{2})+\dfrac{3D^{\sim 2}}{4.48\varepsilon_0}\lambda_2^{-4}\lambda_1^{-2} & -\dfrac{2D^{\sim}}{4.48\varepsilon_0}\lambda_2^{-3}\lambda_1^{-2} \\ -\dfrac{2D^{\sim}}{4.48\varepsilon_0}\lambda_1^{-3}\lambda_2^{-2} & -\dfrac{2D^{\sim}}{4.48\varepsilon_0}\lambda_2^{-3}\lambda_1^{-2} & \dfrac{1}{4.48\varepsilon_0}\lambda_1^{-2}\lambda_2^{-2} \end{bmatrix} \quad (31)$$

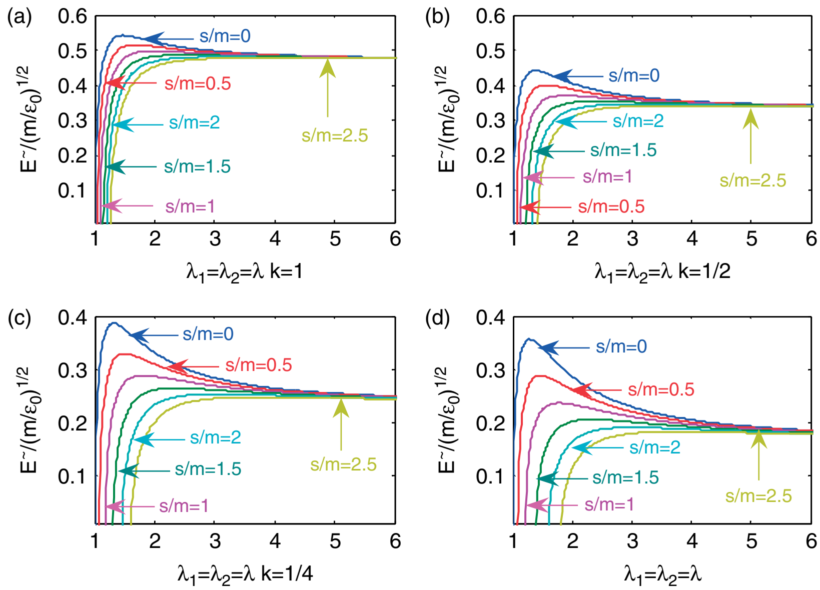

Figure 5. Nominal electric field *versus* stretch ratio when $\lambda_1 = \lambda_2 = \lambda$, $S^{\sim} > 16$.

DISCUSSION

From the above analysis, it is observed that when $S^{\sim}$ varies from 0 to a finite value, the permittivity of the DE film decreases linearly until it approaches a constant of $4.48\varepsilon_0$. For both of the two situations $S^{\sim} \leq 16$ and $S^{\sim} > 16$, it is clear that if the DE film is pre-stretched, its permittivity will decrease and the critical nominal electric field will increase. (For example, for $k = 1$, the critical nominal electric fields under the two kinds of stretch ratio are $0.4536\sqrt{m/\varepsilon_0}$ and $0.5424\sqrt{m/\varepsilon_0}$, respectively.) Hence, the DE treated by pre-stretching shows better electromechanical stability. This conclusion is consistent with experimental results and the theoretical results of Zhao and Suo.[12]

CONCLUSIONS

The electromechanical stability of a Mooney–Rivlin-type DE undergoing large deformation has been investigated. The nonlinear expression of permittivity as a function of the stretch ratio is also proposed. In our study, the electromechanical stability is analyzed using a free energy model consisting of Ogden elastic strain energy and electric field energy with nonlinear permittivity. Based on the model, the relations between nominal electric displacement and nominal electric field are evaluated. Further, the simple form of Ogden elastic strain energy, with only two material constants, is applied to investigate the stability performance of a Mooney–Rivlin-type DE and the critical breakdown electric fields of various DEs are evaluated. The results show that the proportionality constant k is beneficial for representing the stability of different types of DE. For a larger dimensionless constant k of the dielectric material, the nominal value of electric breakdown field is higher, the corresponding DE or structure is more stable and the pre-stretch deformation can notably improve the film's electromechanical stability. These results match the experimental data well and can be applied as guidance in the design and fabrication of DE actuators.

ACKNOWLEDGEMENTS

This work was supported by plans for the development of high technology (863) and supported by the Program for New Century Excellent Talents in University.

REFERENCES

1 Pelrine R, Kornbluh R, Pei QB and Joseph J, *Science* **287**:836 (2000).
2 Carpi F, De Rossi D, Kornbluh R, Pelrine R and Sommer-Larsen P (eds), *Dielectric Elastomers as Electromechanical Transducers*. Elsevier, Amsterdam (2008).
3 Plante JS and Dubowsky S, *Int J Solids Struct* **43**:7727 (2006).
4 Gallone G, Carpi F, De Rossi D, Levita G and Marchetti A, *Mater Sci Eng C* **27**:110 (2007).
5 Wissler M and Mazza E, *Sens Actuators A* **134**:494 (2007).
6 Kofod G, Wirges W, Paajanen M and Bauer S, *Appl Phys Lett* **90**:081916 (2007).
7 Keplinger C, Kaltenbrunner M, Arnold N and Bauer S, *Appl Phys Lett* **92**:192903 (2008).
8 Pelrine RE, Kornbluh RD and Joseph JP, *Sens Actuators A* **64**:77 (1998).
9 Plante JS and Dubowsky S, *Smart Mater Struct* **16**:S227 (2007).
10 Wissler M and Mazza E, *Sens Actuators A* **138**:185 (2007).
11 Kofod G, Paajanen M and Bauer S, *Appl Phys A* **85**:141 (2006).
12 Zhao X and Suo Z, *Appl Phys Lett* **91**:061921 (2007).
13 Zhao X, Hong W and Suo Z, *Phys Rev B* **76**:134113 (2007).
14 Suo Z, Zhao X and Greene WH, *J Mech Phys Solids* **56**:476 (2008).
15 Zhou J, Hong W, Zhao X, Zhang Z and Suo Z, *Int J Solids Struct* **45**:3739 (2008).
16 Norrisa AN, *Appl Phys Lett* **92**:026101 (2007).
17 Liu Y, Liu L, Zhang Z, Shi L and Leng J, *Appl Phys Lett* **93**:106101 (2008).
18 Díaz-Calleja R, Riande E and Sanchis MJ, *Appl Phys Lett* **93**:101902 (2008).
19 Liu Y, Liu L, Sun S, Shi L and Leng J, *Appl Phys Lett* **94**:096101 (2009).
20 Leng J, Liu L, Liu Y, Yu K and Sun S, *Appl Phys Lett* **94**:211901 (2009).
21 Liu Y, Liu L, Sun S and Leng J, *Sci China Ser E* **52**:2715 (2009).
22 Zhao X and Suo Z, *J Appl Phys* **101**:123530 (2008).
23 Kofod G, Sommer-Larsen P, Kronbluh R and Pelrine R, *J Intell Mater Syst Struct* **14**:787 (2000).
24 Liu Y, Liu L, Sun S and Leng J, *Smart Mater Struct* **18**:095040 (2009).
25 Liu Y, Liu L, Zhang Z and Leng J, *Smart Mater Struct* **18**:095024 (2009).
26 Suo Z and Zhu J, *Appl Phys Lett* **95**:232909 (2009).
27 Koh SJA, Zhao X and Suo Z, *Appl Phys Lett* **94**:262902 (2009).