Chapter 1
A Review of Hyperelastic Constitutive Models for Dielectric Elastomers

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Abstract Dielectric elastomers are smart materials that are essential components in soft systems and structures. The core element of a dielectric elastomer is soft matter, which is mainly rubber-like and elastomeric. These soft materials show a nonlinear behaviour and have a nonlinear strain–stress curve. The best candidates for modelling the nonlinear behaviour of such materials are hyperelastic strain energy functions. Hyperelastic functions have been extensively used for modelling dielectric elastomer smart structures. This review paper introduces hyperelastic constitutive laws for modelling dielectric elastomers. For this purpose, first, a general scheme of hyperelastic models is expressed. Then, some well-known hyperelastic models are introduced. Finally, we review in detail the utilized hyperelastic models for different configurations of dielectric elastomers. Possible future works in this field are outlined eventually.

Keywords Dielectric elastomers · Hyperelastic models · Nonlinear elasticity · Smart polymers

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1.1 Introduction

For decades, research on various mechanical structures has been a major topic among scientists [1–5]. The literature in this field has mainly focused on conventional materials possessing a physically linear behaviour. However, many other materials in the world have a nonlinear behaviour. Many human tissues and bodies of animals and plants show a nonlinear behaviour and are modelled as soft structures. These soft structures have been the inspiration of new fields, specifically soft robotics [6, 7]. The main components in this field are soft materials [8, 9]. Using soft materials, we can fabricate soft systems and structures. Dielectric elastomers (DEs) [10, 11] have emerged as powerful smart materials that have been widely employed as soft actuators and soft sensors for developing soft materials and structures. The simplest form of a DE includes a soft membrane that is covered with compliant electrodes [12]. The soft membrane is a major part of a DE. Rubber-like and elastomeric materials with material nonlinearity are used as soft membranes in DEs. To induce deformation in DEs, it is necessary to apply electromechanical loading to them [13, 14]. Generally, a potential difference (voltage) and tensile mechanical load are utilized to induce deformation in DEs. For this reason, DEs are considered smart electromechanical systems.

In response to electromechanical loading, DEs deform nonlinearly and encounter nonlinear oscillation and vibration [15]. From mechanical and vibrational points of view, their potential energy should be calculated for modelling the deformation of DEs. Because DEs consist of nonlinear materials as the core element, linear elasticity cannot be employed for calculating their potential energy. The best alternative to linear elasticity is nonlinear elasticity. Usually, the nonlinear elasticity of DE is captured using hyperelastic constitutive laws [16, 17]. Hyperelastic models have been extensively employed for modelling static and dynamic responses of DEs. There is a large volume of published studies describing the nonlinear response of different types of DEs based on hyperelastic models [18–21]. As observed from prior studies, the knowledge of hyperelastic models is essential for the accurate modelling of DEs. This paper attempts to provide a more detailed investigation of hyperelastic models for DEs.

This review paper is organized as follows. First, in Sect. 1.2, the theory of DEs is described in brief. Then, in Sect. 1.3, the general formulation of hyperelasticity is expressed. Next, in Sect. 1.4, the available hyperelastic models for diverse geometries of DEs are reviewed. Finally, in Sect. 1.5, the main conclusions and perspectives for hyperelastic models and nonlinear elasticity in DEs are expressed.

1.2 Theory of Dielectric Elastomers

The basic theory of dielectric elastomers (DEs) originates from the theory of *electro-elasticity* [22]. Many researchers have tried to develop and review the *electro-elasticity* theory. For instance, in [23, 24], authors developed and reviewed some boundary-value problems of *electro-elasticity* by using the continuum mechanics notation and finite strain theory. A full discussion of *electro-elasticity* lies beyond the scope of this study, but the above-stated literature was introduced for more information.

In line with electro-elasticity, Suo [25] introduced the *theory* of dielectric elastomers. The basic equations for electromechanical large deformations of DEs have been formulated and extended. In another paper, Zhao and Suo [26] considered the electro-elasticity of DEs and discussed the electrical and mechanical equations.

Based on the studies mentioned above, in general, a DE consists of a membrane sandwiched between two compliant electrodes. The membrane may take different geometries such as square, rectangular, spherical, tubular, cylindrical, plate-like, and beam-like [27–33]. Tensile mechanical loads are applied to the membrane, and a potential difference (voltage) is applied to the electrodes. The electrodes are located on the top and bottom surfaces of the soft membrane. When the electrodes are connected to the potential difference, one surface gains the positive electric charge, and another surface gains the negative electric charge. These opposite electric charges attract each other and thereby induce large deformation in the DE such that in the thickness direction, the membrane shrinks, and in the in-plane direction, it stretches and expands. During this process, depending on the type of loading, DEs experience different responses and behaviours. The potential difference and tensile load can be time-varying or static. When electrical or mechanical loadings depend upon time, the response of DE becomes complicated, and, in this sense, they undergo nonlinear vibrations (Fig. 1.1).

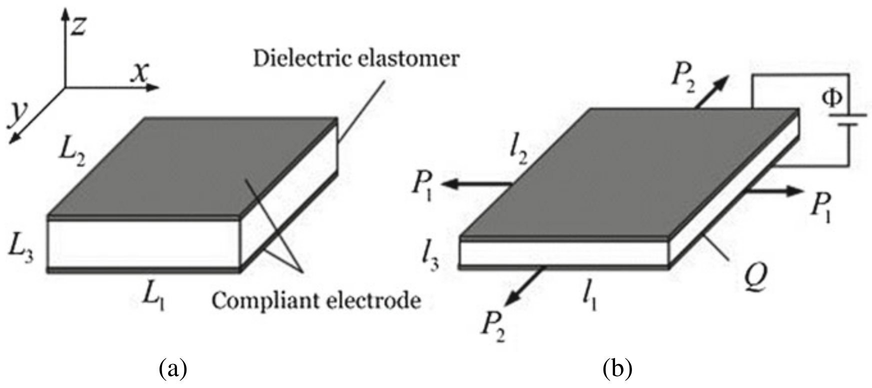

Fig. 1.1 The schematic view of a dielectric elastomer membrane with voltage $\boldsymbol{\Phi}$ and pressures $\boldsymbol{P}_1$, $\boldsymbol{P}_2$. **a** reference configuration, **b** current configuration [34]

1.3 Hyperelasticity

Dielectric elastomers are soft structures whose structural materials are mainly rubbery and elastomeric. For instance, polydimethylsiloxane, silicone, rubber, and VHB-based elastomers have been extensively used for structural materials of DEs [35]. However, these materials possess an inherent nonlinear trend in their stress–strain curves. Thus, they do not follow linear elasticity and Hooke's law. Therefore, the best candidates to describe the nonlinear elasticity of DE structures are hyperelastic material models. Up to now, diverse hyperelastic models have been employed for DE systems [36, 37], e.g., the neo-Hookean model, Mooney–Rivlin model, Gent model and modified versions of the Gent model, and Ogden model.

1.3.1 General Form of Hyperelasticity

Hyperelastic models are expressed in a functional form called the strain energy function and are mainly denoted by W. The hyperelasticity function may take numerous mathematical forms, such as polynomial, exponential, and logarithmic. The strain energy functions are mainly formulated in terms of material parameters and principal invariants of right or left Cauchy–Green deformation tensors. The material parameters are obtained from empirical tests, and principal invariants are expressed based on the field of deformation and the corresponding deformation gradient. One important difference between hyperelasticity and linear elasticity is that the former requires two configurations to describe the deformation. They are reference configuration (nominal quantity) and current configuration (true quantity). Generally, hyperelasticity is a subfield of finite strain theory, and in this field, tensors play a crucial role in the description of hyperelastic models. Therefore, it seems that a good knowledge of tensors may help researchers in understanding hyperelasticity. Before mathematically speaking of hyperelasticity, we introduce some very important tensors.

The first one and maybe the most important tensor is the deformation gradient tensor. When the solids deform, we should explore how two elements dx and dX change and find their relation. We formulate the deformation gradient tensor as [38]

$$d\vec{x} = \mathbf{F} d\vec{X}, \tag{1.1}$$

where $\mathbf{F}$ is the deformation gradient tensor (material deformation gradient). Depending upon the coordinate system, $\mathbf{F}$ may take different forms.

Based on the deformation gradient tensor, two other important tensors are introduced, and the hyperelastic models are formulated according to these tensors. They are the right and left Cauchy–Green deformation tensors. We formulate these tensors as [39]

$$\mathbf{C} = \mathbf{F}^T\mathbf{F}, \tag{1.2}$$

$$\mathbf{b} = \mathbf{F}\mathbf{F}^T. \tag{1.3}$$

In the above equation, $\mathbf{C}$ is the right Cauchy–Green deformation tensor, and $\mathbf{b}$ stands for the left Cauchy–Green deformation tensor; the letter T is the symbol of the *transpose operation.*

Using the *eigenvalue problem*, we can derive the principal invariants of the right and left Cauchy–Green deformation tensor, which are the main elements of hyperelastic models. The invariants of the right Cauchy–Green deformation tensor are written as

$$\begin{aligned} I_1 &= \mathrm{tr}(\mathbf{C}), \\ I_2 &= \frac{1}{2}\left(\mathrm{tr}\mathbf{C}^2 - \mathrm{tr}\left(\mathbf{C}^2\right)\right), \\ I_3 &= \det(\mathbf{C}) = J^2. \end{aligned} \tag{1.4}$$

In the above equation, I_1 and I_2 are respectively the first and second invariants; "tr" stands for *trace*; "det" shows the *determinant*; J represents the determinant of $\mathbf{F}$, i.e., $J = \det(\mathbf{F})$.

The invariants of the left Cauchy–Green deformation tensor are expressed as

$$\begin{aligned} I_1 &= \mathrm{tr}\ (\mathbf{b}), \\ I_2 &= \frac{1}{2}\left(\mathrm{tr}\ \mathbf{b}^2 - \mathrm{tr}\left(\mathbf{b}^2\right)\right), \\ I_3 &= \det(\mathbf{b}) = J^2. \end{aligned} \tag{1.5}$$

The general form of a strain energy function takes the following form:

$$W = W_{\mathrm{iso}} + W_{\mathrm{aniso}}, \tag{1.6}$$

in which W_{iso} is the isotropic part; W_{aniso} stands for the anisotropic part. It is noted that the majority of materials for DEs are considered to be incompressible. The isotropic part itself is formulated as $W_{\mathrm{iso}} = W_{\mathrm{dev}} + W_{\mathrm{vol}}$ in which W_{dev} stands for the isochoric deformation and W_{vol} shows volume change. Due to the incompressibility, the volumetric part (W_{vol}) becomes zero. In the next section, the common hyperelastic models for DEs are introduced and formulated.

1.3.2 Neo-Hookean Model

Neo-Hookean constitutive law is the simplest hyperelastic model that has been used for DEs. This model has been developed for both compressibility and incompressibility states. The neo-Hookean materials model for the compressibility state is expressed as

$$\mathrm{W}_{\text{Neo-Hookean}} = c_1(I_1 - 3 - 2\ln J) + D_1(J-1)^2, \tag{1.7}$$

where c_1 and D_1 are material constants. Considering the incompressibility, the neo-Hookean model is formulated as

$$\psi_{\text{Neo-Hookean}} = c_1(I_1 - 3). \tag{1.8}$$

1.3.3 Gent Model

Some constituent materials for DEs, such as the VHB-based elastomers, have revealed the strain-stiffening effect in response to external loading. The strain-stiffening effect defines a specified value of the stretch in elastomers. The Gent model for a compressible nonlinear elastic material is written as [40]

$$\mathrm{W}_{\text{Gent}} = -\frac{\mu J_{lim}}{2}\ln\left(1 - \frac{I_1 - 3}{J_{lim}}\right) + \frac{\kappa}{2}\left(\frac{J^2 - 1}{2} - \ln J\right)^4, \tag{1.9}$$

in which μ and κ stand for material parameters; J_{lim} is a dimensionless parameter, the so-called stiffening parameter (Gent parameters) that measures the strength of the strain-stiffening; as J_{lim} is decreased, the strain-stiffening effect increases. The Gent model for the incompressibility condition takes the following form [41]:

$$\psi_{\text{Gent}} = -\frac{\mu J_{lim}}{2}\ln\left(1 - \frac{I_1 - 3}{J_{lim}}\right). \tag{1.10}$$

1.3.4 Mooney–Rivlin Model

Another hyperelastic model that has been utilized for DEs is the Mooney–Rivlin model. This model is a good candidate for deformations with large strains. The absence of the strain-stiffening effect is, however, the model's most significant flaw. The Mooney–Rivlin model for the compressibility condition is expressed as

$$W_{\text{Mooney-Rivlin}} = C_{10}\left(\overline{I}_1 - 3\right) + C_{01}\left(\overline{I}_2 - 3\right) + \frac{1}{D_1}(J - 1)^2, \tag{1.11}$$

where C_{10} and C_{01} represent material constants. The Mooney–Rivlin constitutive law for an incompressible material is expressed as

$$\psi_{Mooney-Rivlin} = C_1\left(\overline{I}_1 - 3\right) + C_2\left(\overline{I}_2 - 3\right), \tag{1.12}$$

where C_1 and C_2 stand for material constants. In Eqs. (1.11) and (1.12), $\overline{I}_1 = J^{-2/3} I_1$ and $\overline{I}_1 = J^{-4/3} I_2$.

1.3.5 *Gent–Gent Model*

The Gent–Gent model is a modified version of the Gent model. The existence of the second principal invariant differentiates between the original Gent model and the Gent–Gent model. The Gent model with the incompressibility condition is formulated as [42, 43]

$$W_{\text{Gent-Gent}} = -c_1 J_{lim} \ln\left(1 - \frac{I_1 - 3}{J_{lim}}\right) + c_2 \ln\left(\frac{I_2}{3}\right), \tag{1.13}$$

where c_1 and c_2 are material parameters.

1.4 Studies on Dielectric Elastomers Based on the Type of Hyperelastic Model

This section reviews the published literature on dielectric elastomers (DEs) based on hyperelastic models. First, the literature based on the Gent strain energy function is reviewed. After that, the studies based on the neo-Hookean model, Mooney–Rivlin model, and Ogden model are reviewed. Finally, we discuss modified versions of the Gent model for DEs.

1.4.1 *Studies Based on Gent Model*

A large amount of literature on DEs based on the Gent model is available. For instance, electromechanical instability in dynamical modes in a DE balloon was identified by Chen et al. [44], who utilized the Gent hyperelastic model. Furthermore, by plotting the voltage-stretch curve and pressure-stretch curve, the electromechanical instabilities of an interconnected DE spherical shell were studied by Sun et al. [45], who utilized the Gent hyperelastic material model.

By depicting the time-stretch response and voltage-stretch curve, the oscillations and instability of a balloon made of DE were analysed by Chen and Wang [46] with the aid of the incompressible Gent constitutive model. Furthermore, nucleation and Propagation of Wrinkles were investigated in [47] by using the Gent model to consider the strain-stiffening.

The response of interconnected DE balloons was studied and simulated by Chen and Wang [48], who utilized the Gent model. Mao et al. [49] addressed the wrinkling phenomenon experimentally in DE balloons by using the Gent model. They used the stretch limit in the Gent model as $J_m = 220$. Snap-through instability and electrical breakdown were also explored in their study.

In a deep analysis conducted by Lv et al. [50], dynamic characteristics and instabilities of an electromechanically actuated hyperelastic balloon were assessed, where a Gent model was used to capture the strain-stiffening, and the damping effect was also considered.

In [51], by implementing the Gent model with stretch limits as $J_m = 270$ and 97.2, new electromechanical instabilities in DE spherical shells were identified and discussed. Wang et al. [52] by employing the Gent model analysed the anomalous bulging behaviour in a DE spherical balloon.

In [53], using a visco-hyperelastic Gent model, the delayed electromechanical characteristic of a spherical balloon was explored. In that study, a rheological model with two springs and one dashpot was employed. In addition, the Gent energy function in conjunction with the viscoelastic effect was employed for investigating the wrinkling behaviour of a DE balloon (see [54]).

Zhang et al. [55] harnessed the dielectric breakdown and instabilities in a DE with the Gent hyperelastic material model. Zhou et al. [56] considered the Gent model to study the nonlinear behaviour of a DE membrane. They utilized Equi-biaxial loading and incorporated the viscoelasticity for the system.

Zhu et al. [57] investigated the response of DE encountering the wrinkling phenomenon, employed the Gent constitutive law, and plotted the voltage-stretch curves to provide a profound analysis. Finally, with the aid of the finite strain theory and the Gent model, Garnell et al. [58] explored the sound radiation and vibrations of a DE membrane.

A systematic investigation was conducted by Wang et al. [59] to assess the vibrational behaviour of circular DEs. They reported the chaos and quasi-periodic motion in DEs when the Gent model is employed. The influence of geometrical sizes on the nonlinear vibration of a DE membrane was assessed in [60], who employed the Gent model and assumed that the elastomer is incompressible. Alibakhshi and Heidari [61] investigated nonlinear vibrations of a microbeam made of DEs employing the Gent model, and different phenomena were identified in that paper. In a series of papers [62–67], the thermoelasticity of DEs has been investigated, where the Gent material law was considered to calculate the elastic energy part.

1.4.2 Studies Based on Neo-Hookean, Mooney–Rivlin, and Ogden Models

In this subsection, we concentrate on the neo-Hookean model, Mooney–Rivlin, and Ogden models. Zhu et al. [68] analysed the nonlinear vibrations of a DE spherical shell by modelling the elastomer based on the neo-Hookean strain energy function. They analytically and numerically solved the problem and discussed the equilibrium stretches of the elastomers.

The performance of a DE in electromechanical situations was addressed by Zhang and Chen [69], who adopted the neo-Hookean model in an incompressible condition. The bifurcation phenomenon in a spherical balloon was analysed by Liang and Cai [70], who employed the Ogden hyperelastic model and applied the pressure and voltage to the balloon.

In [71], random vibrations of a spherical balloon were analysed with the help of the neo-Hookean strain energy function. The author of that paper used the method of stochastic averaging to solve the random problem analytically. In another paper [72], the random response of a spherical balloon made of DE was investigated by using the Mooney–Rivlin strain energy function. In that paper, stochastic averaging and Monte Carlo simulation were implemented to help the author to identify different aspects of stochastic problems.

With the application of inflation pressure and potential difference, the bifurcation of a DE balloon was analysed by Xie et al. [73]. They utilized the Ogden, neo-Hookean, and Mooney–Rivlin strain energy functions and compared the results with the Gent model. In another paper published [74], the free vibration of a DE spherical balloon was analytically and numerically solved using the neo-Hookean model, Runge–Kutta method, and Newton-harmonic balance.

Static pull-in and snap-through instabilities and DC static instability in a DE balloon with the aid of the neo-Hookean, Mooney–Rivlin, and Ogden models were analysed by Sharma et al. [75]. In [76, 77], the nonlinear vibration and resonance of DE balloons were explored numerically and analytically using multiple timescales and incremental harmonic balance methods. In those papers, the neo-Hookean strain energy function was applied. The parametric excitation of a DE was analysed using the neo-Hookean model (see [78]), where the equilibrium points and primary and secondary resonances were addressed. In [79, 80], by utilizing the Ogden and Mooney–Rivlin strain energy functions, the dynamic response of DEs was experimentally and analytically captured.

Kim et al. analyzed vibration frequencies of a DE membrane by implementing the neo-Hookean, Ogden, and Mooney–Rivlin models [81]. Dai and Wang [82] carried out a dynamic analysis of the in-plane oscillations of neo-Hookean DEs. They applied the incompressibility conditions for DE and depicted time-stretch response and phase portraits in nonlinear vibration analysis.

The nonlinear response of a DE-based smart system was studied by Srivastava and Basu [83], who utilized the single-term Ogden strain energy function in their work. A neo-Hookean-based viscoelastic model was implemented in [84] to assess the performance of a circular DE. Random oscillation of a DE balloon was reported in [85], in which the neo-Hookean model was adopted. Some researchers have tried to develop DE-based systems incorporating neo-Hookean and Mooney–Rivlin models in a thermoelectricity theory [86–89].

1.4.3 Studies Based on Gent–Gent Model

Researchers decided to discard this limitation because the Gent model cannot accurately capture the deformation of hyperelastic materials at large strains. Some papers extended the Gent model and by which its new versions were introduced. However, the literature on modelling DEs based on the modified version of DEs is very limited.

In recent years, some researchers utilized the Gent–Gent model for DEs, which is a modified version of the Gent model. For instance, Alibakhshi and Heidari investigated the nonlinear vibration and chaos in DE balloons based on the Gent–Gent model [90] and concluded that this hyperelastic model is influential in controlling instabilities and chaos in such systems. Alibakhshi et al. [91] analysed the nonlinear resonance of a DE membrane employing the Gent–Gent model. They also considered a new version of the Gent model in that paper introduced by Bien-aimé et al. [92]. Chen et al. using a compressible Gent–Gent model, researched elastic waves of a DE laminate [93].

1.5 Future Works on Nonlinear Elasticity of Dielectric Elastomers

Researchers have been working on developing the responsiveness of DEs based on anisotropy in recent years. They are considering fibre-reinforced hyperelastic materials for such smart structures. The hyperelastic models incorporating fibre-reinforcement are utilized for this kind of system [94–99]. The Holzapfel–Gasser–Ogden hyperelastic model has been used in the majority of investigations in this field [100, 101]. Thus, future works developing fibre-reinforcement of DEs seem to be of interest.

Another emerging topic in this field of study is employing hyperelastic models with the inclusion of humidity. It has been reported that humidity might affect the performance of DEs in real-world applications [102–104]. Thus, this topic may also be interesting to researchers and can be a major topic of study in future works.

1.6 Conclusion

This paper reviewed the hyperelastic strain energy functions utilized for dielectric elastomers. First, the mathematical formulations for nonlinear electro-elasticity and finite strain theory were explained. Then, the studies for DEs based on the type of hyperelastic model were reviewed. The following are some of the findings of this research:

- The most widely used hyperelastic model for dielectric elastomers is the Gent model.
- After the Gent model, the neo-Hookean model has been a candidate for capturing the nonlinear elasticity of dielectric elastomers. This model has been used for dielectric elastomers but not as much as the Gent model.
- The modified versions of the Gent model, such as the Gent–Gent model, are new hyperelastic materials for analysing dielectric elastomers.
- The role of the second invariant of the Cauchy–Green deformation tensor is prominent for dielectric hyperelastic smart structures.
- In the majority of the literature on DEs, incompressibility condition has been assumed.
- The type of hyperelastic model defines the range of chaos, nonlinear vibrations, and electromechanical instabilities.

For further reading on nonlinear elasticity, we refer to classic books [105–107] as well as corresponding chapters [108–111]. In particular, in [110, 111] other useful models of elastomers could be found.

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