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Review:

Constitutive models of artificial muscles: a review*

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Abstract: Artificial muscles are materials which possess muscle-like characteristics; they have many promising applications and many materials have been exploited as artificial muscles. In this review, the artificial muscles discussed are confined to dielectric elastomers and responsive gels. We focus on their constitutive models based on free energy function theory. For dielectric elastomers, both hyperelastic and visco-hyperelastic models are involved. For responsive gels, we consider different kinds of gels, such as hydrogel, pH-sensitive gel, temperature-sensitive gel, polyelectrolyte gel, reactive gel, etc. With an accurate, reliable, and powerful constitutive model, exact theoretical analysis can be achieved and the important intrinsic characteristics of artificial muscle based systems can be revealed.

Key words: Constitutive model, Artificial muscle, Dielectric elastomer, Responsive gel, Free energy function
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1 Introduction

Living animals can perform many kinds of moving activities, such as standing, walking, running, climbing, jumping, kicking, catching, rotating, etc. All these motions are accomplished with the aid of muscles. Muscles have the ability to generate forces and are the main internal motors of the movement of living animals (Knudson, 2007). Natural muscles are elastic and flexible and have high mechanical strength. They take various forms. Materials or devices which have muscle-like characteristics are described as artificial muscles (Otake, 2010). The two main noticeable properties of artificial muscles are "soft" and "active". In modern techniques, many materials have been exploited as artificial muscles in practical engineering applications, for example, dielectric elastomer electroactive polymers, ionic electroactive polymers, ionic polymeric gels, polyelectrolyte hydrogels, conductive polymers, liquid crystal elastomer materials, electrorheological materials, magnetorheological materials, shape memory alloys, small memory polymers, etc. (Shahinpoor *et al.*, 1998; 2007; Shahinpoor, 2003; Bassil *et al.*, 2008; O'Halloran *et al.*, 2008; Wallace *et al.*, 2009; Carpi *et al.*, 2011a; Aw and McDaid, 2014; Kwon *et al.*, 2014; Mutlu *et al.*, 2014).

As is well known, theoretical modeling and simulation play an important role in understanding the functionality of artificial muscles. To improve the performance of biomimetic robots (Kim *et al.*, 2013), artificial hands (Balasubramanian and Santos, 2014), tunable lenses (Carpi *et al.*, 2011b), etc., it is important to carry out qualitative or quantitative analysis. In the theoretical analysis, it is crucial to develop and use accurate, reliable, and powerful constitutive models.

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In this review, we will focus on constitutive models for dielectric elastomers and responsive gels. The models are based on free energy function theory.

2 Constitutive models of a dielectric elastomer

2.1 Hyperelastic model of a dielectric elastomer

Dielectric elastomers belong to one class of soft active materials. They deform largely through expanding in area and reducing in thickness in response to an applied voltage or external load. The large strain justifies dielectric elastomers being called artificial muscle. Various transducers for a dielectric elastomer membrane sandwiched between two soft electrodes (usually carbon grease) have been developed.

A decade ago, Pelrine *et al.* (2000) discovered that a dielectric elastomer can be actuated at a strain larger than 100% by applying a voltage. This discovery inspired many scientists and engineers to investigate and develop dielectric elastomers and great improvements have been made. The principle of operation of a dielectric elastomer membrane is shown in Fig. 1.

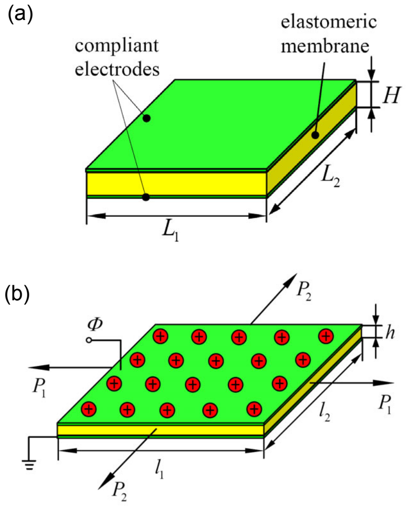

Fig. 1 Schematics of a dielectric elastomer in different states

In the reference state (a), an elastomeric membrane of sides L_1 and L_2 and thickness H is sandwiched between two compliant electrodes. In the deformed state (b), the dielectric elastomer deforms to sides l_1 and l_2 and thickness h when subjected to the mechanical loadings P_1 and P_2 and a high voltage Φ

Under dead load P=25.5g and at voltage ramp rate 500 V/s, Huang *et al.* (2012a) achieved the areal strain of 488%. Not long after, this record was broken by Li *et al.* (2013). They obtained a giant voltage-induced area expansion at 1692% by inflating a dielectric membrane which was mounted on a chamber. Zou *et al.* (2014) performed experiments and simulations on the active shape control of dielectric elastomer structures with various 2D patterned electrodes by means of an applied voltage. In their experiment, the dielectric elastomer membrane with a pattern of 2D electrodes was mounted on an air chamber. They observed a large voltage-induced deformation and the coexistence of two stable states in the dielectric elastomer. Qu *et al.* (2012) observed stretching rate dependent behaviors of dielectric elastomer membranes (VHB™9473) subjected to pure shear-like loading and electric loading simultaneously. Their experimental observations were compared with theoretical predictions by a viscoelastic model. It is known that there are multiple large-scale failure modes in dielectric elastomers, such as material strength failure, electric breakdown, loss of tension, electromechanical instability, etc. (Plante and Dubowsky, 2006; Zhao and Suo, 2007; Liu *et al.*, 2008; 2009a; Leng *et al.*, 2009; Zhou *et al.*, 2013; Joglekar, 2014; Li *et al.*, 2014; Liu *et al.*, 2014a; Mao *et al.*, 2014; Zhu, 2015). To prevent failure and to harness the large deformation of dielectric elastomers, it is necessary to construct a theoretical framework. Based on parameter analysis and loading design, many sensors (Jung *et al.*, 2008; Son and Goulbourne, 2009; Gisby *et al.*, 2013; Liu J.J. *et al.*, 2015), actuators (Carpi and Rossi, 2004; Carpi *et al.*, 2005; 2007; Aschwanden and Stemmer, 2006; Plante and Dubowsky, 2007; Shankar *et al.*, 2007; Wissler and Mazza, 2007; Biddiss and Chau, 2008; Moscardo *et al.*, 2008; Kovacs *et al.*, 2009; Liu *et al.*, 2009b; Carpi *et al.*, 2010; Fang *et al.*, 2010; Akbari and Shea, 2012; Son *et al.*, 2012; Giousouf and Kovacs, 2013; Haus *et al.*, 2013; La and Lau, 2013; Shian *et al.*, 2013; Hunt *et al.*, 2014; Lu *et al.*, 2014; Nguyen C.H. *et al.*, 2014; Nguyen C.H. *et al.*, 2014; Mao *et al.*, 2015), and energy harvesters (Koh *et al.*, 2009; 2011; Liu *et al.*, 2010; McKay *et al.*, 2010; 2011; Ahnert *et al.*, 2011; Kaltseis *et al.*, 2011; Wang *et al.*, 2012a; Chiba *et al.*, 2013; Huang *et al.*, 2013; Graf *et al.*, 2014) with high performance have been achieved.

The mechanical deformation behavior of a dielectric elastomer can be well predicted by one or some of the above models under specific conditions. In order to study the electromechanical behavior of dielectric elastomers, Suo (2010) presented a nonlinear field theory of dielectric elastomers. The condition of thermodynamic equilibrium of a dielectric elastomer demands (Suo *et al.*, 2008):

$$\delta W(\boldsymbol{F},\tilde{\boldsymbol{D}})=\boldsymbol{s}\delta\boldsymbol{F}+\tilde{\boldsymbol{E}}\delta\tilde{\boldsymbol{D}}, \tag{1}$$

where W is the free energy density function. $\boldsymbol{s}$ is the nominal stress tensor and $\boldsymbol{F}$ is the deformation gradient tensor. $\boldsymbol{s}$ is work conjugate to $\boldsymbol{F}$. $\tilde{\boldsymbol{E}}$ is the nominal electric field vector and $\tilde{\boldsymbol{D}}$ is the nominal electric displacement vector. Also, $\tilde{\boldsymbol{E}}$ is work conjugate to $\tilde{\boldsymbol{D}}$ (Suo *et al.*, 2008). Then mathematically, the constitutive equations are obtained as:

$$\boldsymbol{s}=\frac{\partial W(\boldsymbol{F},\tilde{\boldsymbol{D}})}{\partial \boldsymbol{F}},\quad \tilde{\boldsymbol{E}}=\frac{\partial W(\boldsymbol{F},\tilde{\boldsymbol{D}})}{\partial \tilde{\boldsymbol{D}}}. \tag{2}$$

Eq. (1) or (2) means that the form of $W(\boldsymbol{F},\tilde{\boldsymbol{D}})$ determines the constitutive law of the dielectric elastomer.

A soft dielectric elastomer consists of long and flexible polymer chains with covalently bonded links. It is usually assumed that the elastomer can polarize nearly as freely as can liquids and so can be treated as an ideal dielectric elastomer (Zhao *et al.*, 2007). That is, the electrical polarization of soft dielectrics is independent of their deformation. Then the free energy density of an ideal dielectric elastomer can be written as

$$W(\boldsymbol{F},\tilde{\boldsymbol{D}})=W_s(\boldsymbol{F})+W_E(\boldsymbol{F},\tilde{\boldsymbol{D}}), \tag{3}$$

where $W_E(\boldsymbol{F},\tilde{\boldsymbol{D}})$ is expressed as (Zhao *et al.*, 2007; Zhao and Wang, 2014):

$$W_E(\boldsymbol{F},\tilde{\boldsymbol{D}})=\frac{(\boldsymbol{F}\tilde{\boldsymbol{D}})\cdot(\boldsymbol{F}\tilde{\boldsymbol{D}})}{2\varepsilon\det(\boldsymbol{F})}=\frac{F_{iK}F_{iL}}{2\varepsilon\det(\boldsymbol{F})}\tilde{D}_K\tilde{D}_L. \tag{4}$$

In Eq. (4), ε is the permittivity of the dielectric, which is taken to be a constant independent of deformation (Suo, 2010). $W_S(\boldsymbol{F})$ is the free energy function for rubber-like (hyperelastic) materials. Generally, for isotropic incompressible rubber-like materials, $W_s(\boldsymbol{F})$ is a function of I_1 and I_2, which are the first and second invariants of the right Cauchy-Green deformation tensor $\boldsymbol{C}=\boldsymbol{F}^{\mathrm{T}}\boldsymbol{F}$, respectively.

For rubber-like materials such as a dielectric elastomer, constitutive models of rubber elasticity in the frameworks have been approached either through statistical mechanics or through continuum mechanics. The statistical mechanics approach is based on Gaussian and non-Gaussian statistics and long-chain molecular network theory. In a Gaussian treatment of a single chain, the Gaussian distribution is used to describe the end-to-end of the chain. The results are valid only when the stretch of the chain is limited, i.e., not approaching the limiting stretch. When the chain is highly extended, a non-Gaussian statistical treatment should be applied to obtain the properties of the network using Langevin chain statistics, including the three-chain model (Wang and Guth, 1952), the four-chain model (Flory and Rehner, 1943), the eight-chain model (Arruda and Boyce, 1993), and the full chain model (Wu and van der Giessen, 1993). The continuum mechanics approach takes the rubber as a hyperelastic material with the deformation energy function dependent on the invariants of the stretch tensor. Thereafter, the Mooney-Rivilin model (Mooney, 1940), the Ogden model (Ogden, 1972), the Gent model (Gent, 1996), etc., have been developed to capture the stretch behavior of these kinds of materials. Treloar (1975) delivered a comprehensive review of the statistical description of rubber elasticity. Boyce and Arruda (2000) reviewed several constitutive models of incompressible rubber-like materials based on classic statistical mechanics and phenomenological theory (continuum mechanics). Marckmann and Verron (2006) compared 20 hyperelastic models of rubber-like materials. Both the material parameters and the stretch range of validity of each model were addressed. Horgan and Saccomandi (2006) reviewed the phenomenological hyperelastic constitutive models in respect of their limiting chain extensibility. Beda (2014) summarized the model-building strategies of hyperelastic constitutive models.

Table 1 lists some frequently-used stretching energy functions for analyzing incompressible dielectric elastomers. Fig. 2 shows the comparison of the nominal stress-stretch behavior of four frequently-used models (the neo-Hookean model,

Table 1 Some frequently-used stretching energy functions for incompressible dielectric elastomers

Model	Expression of W_s	WLCE	NMP	Parameter	Type
neo-Hookean (Treloar, 1943)	$W_s=\frac{\mu}{2}(I_1-3)$	No	1	μ	A
Mooney-Rivlin (Mooney, 1940; Rivlin, 1948)	$W_s=C_1(I_1-3)+C_2(I_2-3)$	No	2	C_1, C_2	B
Ogden (Ogden, 1972)	$W_s=\sum_{i=1}^{N}\frac{\mu_i}{\alpha_i}\left(\lambda_1^{\alpha_i}+\lambda_2^{\alpha_i}+\lambda_3^{\alpha_i}-3\right)$	Yes	2N	μ_i, α_i (i=1, 2, …, N)	B
Yeoh (Yeoh, 1990)	$W_s=\sum_{i=1}^{3}C_i\left(I_1-3\right)^i$	No	3	C_1, C_2, C_3	B
Arruda-Boyce (Arruda and Boyce, 1993)	$W_s=\mu n\left[\frac{\beta_{ch}\lambda_{ch}}{\sqrt{n}}+\ln\left(\frac{\beta_{ch}}{\sinh\beta_{ch}}\right)\right]$, $\lambda_{ch}=\sqrt{\frac{I_1}{3}}=\sqrt{n}\left(\frac{1}{\tanh\beta_{ch}}-\frac{1}{\beta_{ch}}\right)$	Yes	2	μ, n	A
Gent (Gent, 1996)	$W_s=-\frac{\mu}{2}J_{\lim}\ln\left(1-\frac{I_1-3}{J_{\lim}}\right)$	Yes	2	μ, $J_{\lim}$	B

* Note: type A: classic statistical mechanics model; type B: phenomenological model; NMP: number of material parameters; WLCE: with limiting chain extensibility; I_1 and I_2 are the first and second invariants of the right Cauchy-Green deformation tensor

the Gent model, the Arruda-Boyce model, and the Ogden model with N=3) when subjected to uniaxial tension (Fig. 2a) and equibiaxial tension (Fig. 2b). The computational parameters are set as follows: in the Gent model, $J_{\lim}$ is set as $J_{\lim}$=101; in the Arruda-Boyce model, n is set as n=35; in the Ogden model, the parameters are set as N=3, α_1=1.3, α_2=5.0, α_3=−2.0, μ_1=1.491μ, μ_2=0.003μ, and μ_3=−0.023μ (Liang and Cai, 2015).

There are also some other forms of $W_s(\boldsymbol{F})$ for incompressible rubber-like materials. For instance, Horgan and Saccomandi (2002) presented another constitutive model considering limiting chain extensibility. Horgan and Saccomandi (2005) obtained constitutive models for fiber-reinforced incompressible nonlinearly elastic solids. Guo *et al.* (2006) developed a composites-based hyperelastic constitutive model for soft materials. Hyperelastic models for incompressible fiber-reinforced elastomers were also proposed (Agoras *et al.*, 2009; Fereidoonnezhad *et al.*, 2013). A large-strain continuum mechanics framework for the development of constitutive models was presented by Danielsson *et al.* (2004). Huang (2014) presented a novel constitutive model for rubber-like materials with a deformation energy function including effects of both compressibility and temperature changes.

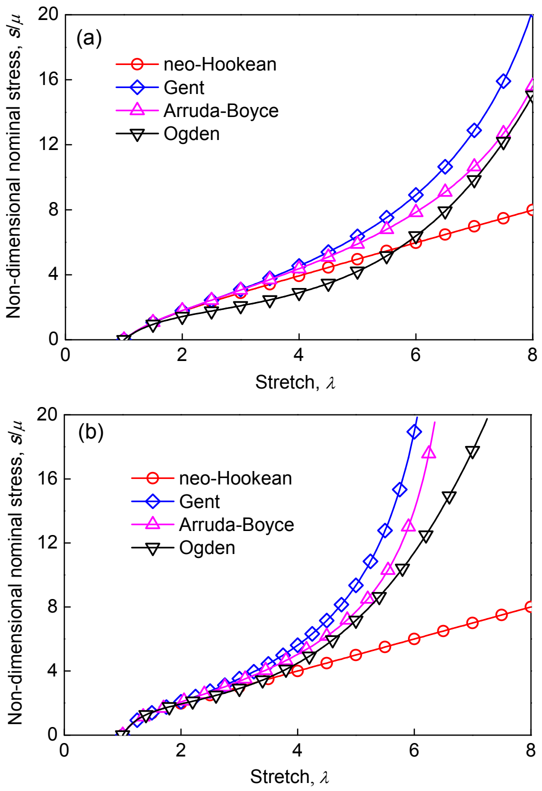

Fig. 2 Comparison of the nominal stress-stretch behavior of four frequently-used models, i.e., the neo-Hookean model, the Gent model, the Arruda-Boyce model, and the Ogden model with N=3

(a) Uniaxial tension; (b) Equibiaxial tension

To model the electromechanical behaviors of dielectric elastomers in the context of engineering practice, some practical factors have also been considered, as shown in Table 2, such as compressibility (Tagarielli *et al.*, 2012; Vertechy *et al.*, 2012), polarization effect (Li B. *et al.*, 2012; Liu *et al.*, 2012; 2014b), unidirectional constraint (Huang *et al.*, 2012b; Lu *et al.*, 2012), variation of permittivity (Zhao and Suo, 2008a), thermally coupled (Liu *et al.*, 2011; 2014c), hydrostatically coupled (Wang *et al.*, 2012b), air coupled (Keplinger *et al.*, 2012; Li *et al.*, 2013), etc.

Furthermore, Yong *et al.* (2012) investigated electromechanical instability in incompressible dielectric elastomers by considering the effect of anisotropy. The free energy form was assumed to be the same as in Eq. (3) while the stretching energy function $W_s(\boldsymbol{F})$ was assumed to be a function of I_1, I_2, I_4, and I_5. Here $I_4=C_{12}^2+C_{13}^2$ and $I_5=C_{11}$. Siboni and Castañeda (2014) studied the finite-strain response and stability of fiber-constrained dielectric elastomer composites (DECs). The DECs consist of very long fibers, embedded firmly in an ideal dielectric matrix. Liu *et al.* (2015) investigated the effect of mass fraction of multi-walled carbon nanotubes on the stability of dielectric elastomer composites.

Based on Suo's theory (Suo *et al.*, 2008; Suo 2010), the finite element method is able to study the dielectric elastomer structures with complicated configurations and/or loadings, and predict large

Table 2 Some special effects in modeling dielectric elastomers

Factor	Key contribution or description
Compressibility	Another strain energy term $W_b(J)$ is appended to the existing strain energy form: $W(\boldsymbol{F},\tilde{\boldsymbol{D}})=W_s(\boldsymbol{F})+W_E(\tilde{\boldsymbol{D}})+W_b(J)$, $W_b=-p(J-1+0.5p/K)$ (Vertechy *et al.*, 2012) or $W_b=0.5K(J-1)^2$ (Tagarielli *et al.*, 2012), where K is the bulk modulus and p is the hydrostatic pressure
Conditional polarization	$W_E(\tilde{\boldsymbol{D}})=\dfrac{\lambda_1^{-2}\lambda_2^{-2}}{2\varepsilon(\lambda_1,\lambda_2)}\tilde{D}^2$, $\varepsilon=\varepsilon_0+\dfrac{N}{3kT}\left[\phi(1+\Lambda^{-1})\mu_{\mathrm{B}}^2+(1-\phi)\mu_{\mathrm{S}}^2\right]$, where ε_0 is the permittivity of the vacuum and N is the number of molecules per unit volume. ϕ is the volumetric fraction of the backbone dipoles over the total monomer dipoles in a single chain. Λ is a negative value representing the number of states that the dipoles may locate before polarization. μ_{B} and μ_{S} are the dipolar moments of monomers in the backbone and in the side chains (Li B. *et al.*, 2012)
Variation of permittivity	$W_E(\tilde{\boldsymbol{D}})=\dfrac{\lambda_1^{-1}\lambda_2^{-1}\lambda_3}{2\varepsilon(\lambda_1,\lambda_2,\lambda_3)}\tilde{D}^2$, $\varepsilon=\varepsilon_0[1+a(\lambda_3-1)+b(\lambda_1+\lambda_2+\lambda_3-3)]$, where ε_0 is the permittivity of the dielectric in the absence of deformation, and a and b are the coefficients of electrostriction (Zhao and Suo, 2008a)
Thermally coupled	$W(\boldsymbol{F},\tilde{\boldsymbol{D}},T)=W_s(\boldsymbol{F},T)+W_E(\tilde{\boldsymbol{D}})$, $W_s(\boldsymbol{F},T)=\dfrac{T}{2T_0}\left[C_1(I_1-3)+C_2(I_2-3)\right]+c_0\left[(T-T_0)-T\ln\left(\dfrac{T}{T_0}\right)\right]$, where T is the current temperature and T_0 is the reference temperature. c_0 is the specific heat of dielectric elastomers (Liu *et al.*, 2011)
Unidirectional constraint	The dielectric elastomeric membrane contracts in the directions normal to the fibers, but keeps its dimension in the direction along the fibers (Huang *et al.*, 2012b; Lu *et al.*, 2012)
Hydrostatically coupled	The fluid sealed between the active and passive membranes remains constant at a prescribed volume $2V_0$. That is, the confined fluid is taken to be incompressible. $V_{\mathrm{act}}+V_{\mathrm{pas}}=2V_0$ (Wang *et al.*, 2012b)
Air coupled	The air enclosed in the chamber and bubble obeys the ideal-gas law: $(p+p_{\mathrm{atm}})(V+V_{\mathrm{c}})=(p_0+p_{\mathrm{atm}})(V_0+V_{\mathrm{c}})$. Here $p+p_{\mathrm{atm}}$ and $V+V_{\mathrm{c}}$ are the pressure and volume in the current state; p_0+p_{atm} and V_0+V_{c} are those quantities in the reference state. p_{atm} is the atmosphere pressure and V_{c} is the volume of the chamber (Keplinger *et al.*, 2012; Li *et al.*, 2013)

deformation and electromechanical instability behaviors. Zhao and Suo (2008b) implemented a user subroutine (UMAT) in the commercial finite element software ABAQUS, using the neo-Hookean model. This UMAT is powerful for analyzing dielectric elastomer transducers undergoing inhomogeneous deformation. Qu and Suo (2012) adopted the same framework to develop a finite element method implemented in ABAQUS by defining two sets of elements on one set of nodes, with each set of elements used for either mechanical deformation or electric induced deformation. This superposition-like method can use any material mechanical model in ABAQUS. Klinkel *et al.* (2013) developed a solid shell finite element formulation to investigate the electromechanical behavior of thin dielectric elastomer structures. Recently, Wang *et al.* (2014) used a nonlinear, dynamic finite element model with a finite deformation viscoelastic constitutive law to study the inhomogeneous deformation and instabilities of dielectric elastomers subjected to a constant voltage.

2.2 Visco-hyperelastic model of dielectric elastomer

Some experiments show that the response of a dielectric elastomer is time-dependent and dissipative when subjected to force or voltage (Wissler and Mazza, 2005; 2007; Plante and Dubowsky, 2006; Qu *et al.*, 2012; Tagarielli *et al.*, 2012; Li *et al.*, 2013; Liu L. *et al.*, 2014). This behavior results from the slippage between the long polymers and the rotation of joints between monomers (Suo, 2010).

Guided by non-equilibrium thermodynamics, the following inequality can be obtained (Suo, 2010; Zhao *et al.*, 2011):

$$\boldsymbol{s}\delta\boldsymbol{F} + \tilde{\boldsymbol{E}}\delta\tilde{\boldsymbol{D}} \le \delta W(\boldsymbol{F}, \tilde{\boldsymbol{D}}, \xi_1, \xi_2, \ldots), \tag{5}$$

where ξ_1, ξ_2, … are known as internal variables. If we assume the system is in mechanical and electrostatic equilibrium, then Eq. (5) can be rewritten as

$$\boldsymbol{s} = \frac{\partial W(\boldsymbol{F}, \tilde{\boldsymbol{D}})}{\partial \boldsymbol{F}}, \quad \tilde{\boldsymbol{E}} = \frac{\partial W(\boldsymbol{F}, \tilde{\boldsymbol{D}})}{\partial \tilde{\boldsymbol{D}}}, \quad \sum_i \frac{\partial W}{\partial \xi_i}\delta\xi_i \le 0. \tag{6}$$

The inequality in Eq. (6) may be satisfied by adopting a suitable kinetic mode of the type:

$$\frac{\mathrm{d}\xi_i}{\mathrm{d}t} = -\sum_j M_{ij}\frac{\partial W(\boldsymbol{F}, \tilde{\boldsymbol{D}}, \xi_1, \xi_2, \ldots)}{\partial \xi_j}, \tag{7}$$

where $\boldsymbol{M}$ is a positive-definite matrix.

The total free energy function can be treated as (Hong, 2011; Park and Nguyen, 2013)

$$W(\boldsymbol{F}, \tilde{\boldsymbol{D}}, \xi_1, \xi_2, \ldots) = W_{\mathrm{EQ}}(\boldsymbol{F}, \tilde{\boldsymbol{D}}) + W_{\mathrm{NEQ}}(\boldsymbol{F}, \xi_1, \xi_2, \ldots), \tag{8}$$

where $W_{\mathrm{EQ}}(\boldsymbol{F}, \tilde{\boldsymbol{D}})$ is named as the equilibrium Helmholtz free-energy in the local equilibrium state and takes the form $W_s(\boldsymbol{F}, \tilde{\boldsymbol{D}})$ for hyperelastic dielectric elastomers. $W_{\mathrm{NEQ}}(\boldsymbol{F}, \xi_1, \xi_2, \ldots)$ is the non-equilibrium Helmholtz free energy and is used to describe the dissipative process. Some specific models have been presented in the open literature (Foo *et al.*, 2012a; 2012b; Li T.F. *et al.*, 2012; Wang *et al.*, 2013; Bai *et al.*, 2014). For an incompressible dielectric elastomer thin membrane subjected to a voltage between the two compliant electrodes, Foo *et al.* (2012a) constructed a specific viscoelastic model by using two parallel units. One unit consists of a spring α and the other unit consists of a spring β and a dashpot. Let λ_1 and λ_2 be the stretches of the elastomer of spring α and ξ_1 and ξ_2 be stretches in the dashpot. By employing the Gent model (Gent, 1996), they obtained:

$$\begin{cases} \sigma_1 = -\varepsilon E^2 + \dfrac{\mu_\alpha(\lambda_1^2 - \lambda_1^{-2}\lambda_2^{-2})}{1 - (\lambda_1^2 + \lambda_2^2 + \lambda_1^{-2}\lambda_2^{-2} - 3)/J_\alpha^{\lim}} \\ \qquad + \dfrac{\mu_\beta(\lambda_1^2\xi_1^{-2} - \xi_1^2\xi_2^2\lambda_1^{-2}\lambda_2^{-2})}{1 - (\lambda_1^2\xi_1^{-2} + \lambda_2^2\xi_2^{-2} + \xi_1^2\xi_2^2\lambda_1^{-2}\lambda_2^{-2} - 3)/J_\beta^{\lim}}, \\ \sigma_2 = -\varepsilon E^2 + \dfrac{\mu_\alpha(\lambda_2^2 - \lambda_1^{-2}\lambda_2^{-2})}{1 - (\lambda_1^2 + \lambda_2^2 + \lambda_1^{-2}\lambda_2^{-2} - 3)/J_\alpha^{\lim}} \\ \qquad + \dfrac{\mu_\beta(\lambda_2^2\xi_2^{-2} - \xi_1^2\xi_2^2\lambda_1^{-2}\lambda_2^{-2})}{1 - (\lambda_1^2\xi_1^{-2} + \lambda_2^2\xi_2^{-2} + \xi_1^2\xi_2^2\lambda_1^{-2}\lambda_2^{-2} - 3)/J_\beta^{\lim}}, \end{cases} \tag{9}$$

where μ_α and μ_β are the shear moduli of the two springs, and $J_\alpha^{\lim}$ and $J_\beta^{\lim}$ are the constants related to the limiting stretches of the two springs. σ_1 and σ_2 are the true stresses and E is the true electric field. They further modeled the dashpot as a Newtonian fluid and the corresponding evolution equations were obtained:

$$\begin{cases}\dfrac{\mathrm{d}\xi_1}{\mathrm{d}t}=\dfrac{\xi_1}{3\eta}\times\left[\dfrac{\mu_\beta(\lambda_1^2\xi_1^{-2}-\xi_1^2\xi_2^2\lambda_1^{-2}\lambda_2^{-2})}{1-(\lambda_1^2\xi_1^{-2}+\lambda_2^2\xi_2^{-2}+\xi_1^2\xi_2^2\lambda_1^{-2}\lambda_2^{-2}-3)/J_\beta^{\lim}}\right. \\ \qquad\left.-\dfrac{\mu_\beta(\lambda_2^2\xi_2^{-2}-\xi_1^2\xi_2^2\lambda_1^{-2}\lambda_2^{-2})/2}{1-(\lambda_1^2\xi_1^{-2}+\lambda_2^2\xi_2^{-2}+\xi_1^2\xi_2^2\lambda_1^{-2}\lambda_2^{-2}-3)/J_\beta^{\lim}}\right], \\ \dfrac{\mathrm{d}\xi_2}{\mathrm{d}t}=\dfrac{\xi_2}{3\eta}\times\left[\dfrac{\mu_\beta(\lambda_2^2\xi_2^{-2}-\xi_1^2\xi_2^2\lambda_1^{-2}\lambda_2^{-2})}{1-(\lambda_1^2\xi_1^{-2}+\lambda_2^2\xi_2^{-2}+\xi_1^2\xi_2^2\lambda_1^{-2}\lambda_2^{-2}-3)/J_\beta^{\lim}}\right. \\ \qquad\left.-\dfrac{\mu_\beta(\lambda_1^2\xi_1^{-2}-\xi_1^2\xi_2^2\lambda_1^{-2}\lambda_2^{-2})/2}{1-(\lambda_1^2\xi_1^{-2}+\lambda_2^2\xi_2^{-2}+\xi_1^2\xi_2^2\lambda_1^{-2}\lambda_2^{-2}-3)/J_\beta^{\lim}}\right],\end{cases} \tag{10}$$

where η is the viscosity of the dashpot.

Foo *et al.* (2012a) also presented a model of leakage current. In that model, the dielectric elastomer was considered as a capacitor with a resistor in parallel. Based on the Kelvin-Voigt model, a visco-hyperelastic model was proposed by Lochmatter *et al.* (2007) by fitting the uniaxial tensile-creep-relaxation test data of acrylic VHB 4910 film.

3 Constitutive models of responsive gel

A cross-linked polymer can take up the solvent molecules from the surrounding environment and form a gel. Fig. 3 illustrates the schematics and photos of a responsive gel in dry and swollen states.

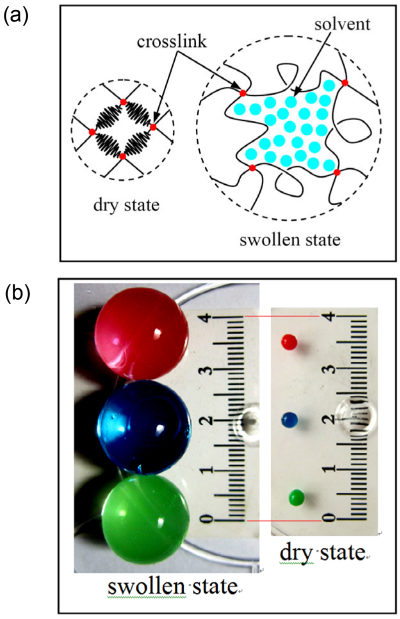

Fig. 3 Schematics (a) and photos (b) of a responsive gel

The stimuli of the responsive gels can be temperature, pH, light, electric field humidity, stress, etc. (Gerlach *et al.*, 2005; Yamaue and Doi, 2005; Lawrence *et al.*, 2007; Rotzetter *et al.*, 2012; Zarzar *et al.*, 2012; Chu *et al.*, 2013; Wang *et al.*, 2013; Lim *et al.*, 2014). Hong *et al.* (2008) presented a framework of coupled diffusion and large deformation in polymeric gels. Based on non-equilibrium thermodynamic theory, one can obtain (Hong *et al.*, 2008; Toh *et al.*, 2013):

$$\boldsymbol{s}=\frac{\partial W(\boldsymbol{F},C)}{\partial \boldsymbol{F}},\quad \mu=\frac{\partial W(\boldsymbol{F},C)}{\partial C}. \tag{11}$$

It should be specially mentioned that in this section, μ denotes the chemical potential and C is the concentration of the solvent. $W(\boldsymbol{F},C)$ is the free energy of the polymeric gel. Following the treatment introduced by Flory and Rehner (1943), $W(\boldsymbol{F},C)$ takes the form:

$$W(\boldsymbol{F},C)=W_s(\boldsymbol{F})+W_m(C), \tag{12}$$

where $W_s(\boldsymbol{F})$ and $W_m(C)$ are the free energy functions of stretching and mixing, respectively, and can be written as (Hong *et al.*, 2008; 2009)

$$\begin{aligned}W_s(\boldsymbol{F})&=0.5NkT\left[\lambda_1^2+\lambda_2^2+\lambda_3^2-3-2\log(\lambda_1\lambda_2\lambda_3)\right],\\ W_m(C)&=-\frac{kT}{\Omega}\left[\Omega C\log\left(1+\frac{1}{\Omega C}\right)+\frac{\chi}{1+\Omega C}\right],\end{aligned} \tag{13}$$

where χ is a dimensionless measure of mixing enthalpy. The product kT is the temperature in the unit of energy. N is the number of the polymeric chain divided by the volume of the dry network and Ω is the volume of a solvent molecule. The condition of molecular incompressibility demands:

$$1+\Omega C=\lambda_1\lambda_2\lambda_3. \tag{14}$$

That is, the free energy function (12) should be enforced by the constraint (14). By using the Legendre transformation:

$$\tilde{W}(\boldsymbol{F},\mu)=W(\boldsymbol{F},C)-\mu C, \tag{15}$$

we have (Hong *et al.*, 2009; Toh *et al.*, 2013):

$$\boldsymbol{s}=\frac{\partial\tilde{W}(\boldsymbol{F},\mu)}{\partial\boldsymbol{F}},\quad C=-\frac{\partial\tilde{W}(\boldsymbol{F},\mu)}{\partial\mu}.\tag{16}$$

Utilizing Eqs. (12)–(14), Eq. (15) can be specified as

$$\begin{aligned}\tilde{W}(\boldsymbol{F},\mu)=&\frac{1}{2}NkT\left[\lambda_1^2+\lambda_2^2+\lambda_3^2-3-2\log\left(\lambda_1\lambda_2\lambda_3\right)\right]\\&-\frac{kT}{\Omega}\left[\left(\lambda_1\lambda_2\lambda_3-1\right)\log\left(\frac{\lambda_1\lambda_2\lambda_3}{\lambda_1\lambda_2\lambda_3-1}\right)+\frac{\chi}{\lambda_1\lambda_2\lambda_3}\right]\\&-\frac{\mu}{\Omega}\left(\lambda_1\lambda_2\lambda_3-1\right).\end{aligned}\tag{17}$$

Based on the framework presented by Hong *et al.* (2008), some models for special cases have been constructed. The inhomogeneous and anisotropic equilibrium state of a swollen hydrogel containing a hard core has been solved by Zhao *et al.* (2008). Hong *et al.* (2009) also presented the model for inhomogeneous swelling of a gel in equilibrium with a solvent and mechanical load. Wu and Zhong (2013) simulated the inhomogeneous equilibrium swelling of core-shell-coating gels. Dai and Song (2011) obtained some analytical solutions of the equilibrium states of a swollen hydrogel shell under spherical deformation by the perturbation method. Chen and Dai (2013) obtained the asymptotic solutions of cylinder and core-shell polymer gels for the equilibrium states. Cai *et al.* (2010) investigated the kinetic swelling process of a polymeric tube. Cai and Suo (2012) presented the equations of state for ideal elastomeric gels. Cai and Suo (2011) dealt with the mechanics and chemical thermodynamics of phase transition in temperature-sensitive hydrogels. In their model, $W_s(\boldsymbol{F})$ also takes the form as in Eq. (13), while $W_m(C)$ is modified as (Cai and Suo, 2011)

$$W_m(C)=\frac{kT}{\Omega}\left[\Omega C\log\left(\frac{\Omega C}{1+\Omega C}\right)+\frac{\chi\Omega C}{1+\Omega C}\right],\tag{18}$$

with

$$\begin{aligned}&\chi(T,\phi)=\chi_0+\chi_1\phi,\\&\chi_0=A_0+B_0T,\quad \chi_1=A_1+B_1T,\\&\phi=(1+\Omega C)^{-1}.\end{aligned}\tag{19}$$

For PNIPAM–water solutions, Afroze *et al.* (2000) fitted their experimental data by using the following parameters:

$$\begin{aligned}&A_0=-12.97,\quad B_0=0.04496K^{-1},\\&A_1=-17.92,\quad B_1=-0.0569K^{-1}.\end{aligned}\tag{20}$$

Hong *et al.* (2010) developed a model for large deformation and electrochemistry of polyelectrolyte gels when immersed in ionic solution. In this model, the free energy is treated as

$$W=W_{\text{net}}+W_{\text{sol}}+W_{\text{ion}}+W_{\text{pol}},\tag{21}$$

where W_{net}, W_{sol}, W_{ion}, and W_{pol} are the free energy functions due to stretching the network, mixing the solvent with the polymers, mixing the ions with the solvent, and polarizing the gel. The specific form of each term has been presented in Hong *et al.* (2010). Then the constitutive model for polyelectrolyte gels can be obtained:

$$\begin{aligned}&\boldsymbol{s}=\frac{\partial W(\boldsymbol{F},\tilde{\boldsymbol{D}},C^1,C^2,\ldots)}{\partial\boldsymbol{F}},\quad \tilde{\boldsymbol{E}}=\frac{\partial W(\boldsymbol{F},\tilde{\boldsymbol{D}},C^1,C^2,\ldots)}{\partial\tilde{\boldsymbol{D}}},\\&\mu^\alpha=ez^\alpha\Phi+\frac{\partial W(\boldsymbol{F},\tilde{\boldsymbol{D}},C^1,C^2,\ldots)}{\partial C^\alpha}\quad(\alpha=1,2,\ldots),\end{aligned}\tag{22}$$

where e is the elementary charge, z^α is the valence of species α, and Φ is the electric potential.

A theory of constrained swelling of a pH-sensitive hydrogel was developed by Marcombe *et al.* (2010). The free energy function accounting for the stretching of the network W_{net}, mixing of the network with the solution W_{sol}, mixing the ions with the solvent W_{ion}, and dissociating the acidic groups W_{dis} is shown below as (Marcombe *et al.*, 2010; Toh *et al.*, 2014)

$$W=W_{\text{net}}+W_{\text{sol}}+W_{\text{ion}}+W_{\text{dis}}.\tag{23}$$

The constitutive model was also specified by Marcombe *et al.* (2010).

Yamaue and Doi (2004) proposed a linearized stress-diffusion coupling model for the 1D swelling dynamics of polymer gels. The constitutive equation involves the bulk modulus and the shear modulus of gels. Based on the stress-diffusion model, Lucantonio and Nardinocchi (2012) presented a reduced model of swelling-induced bending of gel bars under the assumption of plane stress. In response to the stimuli, the elastomeric gels show both viscoelasticity and poroelasticity. Hu and Suo (2012) developed a theory of concurrent viscoelasticity and poroelasticity. The kinetics of viscoelasticity are described by using the homogeneous states of a material element, while the kinetics of poroelasticity are described by using the inhomogeneous states in a body.

Reactive gels have also been investigated. Boissonade (2003; 2009) presented the chemical reaction-diffusion models to simulate the self-oscillation of gels. Due to the swelling-deswelling process, the gel can switch between "reacted state" and "unreacted state" which leads to temporal shape changes. Yashin and Balazs (2006) and Kuksenok *et al.* (2008) developed models for the polymer gels involved in the Belousov-Zhabotinsky (BZ) reaction. Yashin *et al.* (2012) reviewed the coupling models between non-linear chemical dynamics and mechanics for reactive gels.

4 Conclusions

The constitutive models of artificial muscles have been developed and widely used in analyzing the electromechanical or chemomechanical behaviors of dielectric elastomer and gel based actuators, sensors, energy harvesters, soft robots, and soft tissues, etc. The constitutive models are an important foundation for predicting and improving the performance of artificial muscles.

The constitutive models of artificial muscles are complicated due to the large deformation and the multi-field coupling effect. Based on equilibrium and non-equilibrium thermodynamics, the constitutive models of dielectric elastomers and responsive gels can be effectively constructed by using free energy functions. In the models presented, the material parameters can be determined by fitting to the experimental data.

There are a number of constitutive models for rubber-like materials. It is important to select a suitable model for theoretical analysis of soft materials.

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中文概要

题　目：人工肌肉本构模型的综述

摘　要：人工肌肉是指具有类似肌肉特性的材料，这些材料在外界激励下，可以实现大变形，且响应速度快。本文总结两类人工肌肉本构模型的研究成果：一类是介电高弹体，另一类是响应性凝胶。本文中提到的本构模型仅限于用自由能函数导出的情形。对于介电高弹体材料，分别综述超弹性模型和粘性超弹性模型。在超弹性模型中，列出目前研究中使用较多的一些本构模型的自由能函数具体表达式；比较 neo-Hookean、Gent、Arruda-Boyce 和 Ogden 四种模型在单轴拉伸和等双轴拉伸两种情形下的名义应力-伸长曲线；给出了考虑一些重要因素的研究模型，这些因素包括材料可压缩性、取向极化、变介电常数、热耦合、受纤维约束、流体耦合以及空气耦合等。对于响应性凝胶，分别综述水凝胶、pH 敏感性凝胶、温度敏感性凝胶、聚电解质凝胶以及反应性凝胶等的本构模型。这些精确、可靠和有效的本构模型，将有助于开展人工肌肉系统的性能分析和预测，甚至揭示其内在特性和本质规律。

关键词：本构模型；人工肌肉；介电高弹体；响应性凝胶；自由能函数