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A nonlinear acoustomechanical field theory of polymeric gels

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ABSTRACT

A nonlinear acoustomechanical field theory is presented for polymeric gels undergoing large deformation coupled with diffusion mass transport of solvent molecules in and out of the gel. The theory is developed by combining the acoustic radiation stress theory with the nonlinear elasticity theory of polymeric gels. Explicit velocity and acoustic fields are determined by solving the elastodynamical equations of wave propagation in Eulerian coordinates, which are then employed to determine the distribution of acoustic radiation stresses inside the gel. The nonlinear elasticity of gels is modeled by adopting the Flory–Rehner free energy functions for network stretching and molecules-polymer mixing. For illustration, the developed theory is applied to a layer of polymeric gel immersed in external solvent subjected to two counterpropagating acoustic waves. The acoustically actuated large deformation of the gel is analyzed under three different constraint conditions. Unique acoustomechanical behaviors of polymeric gels are revealed, such as periodical response and nonlinear chaos. This work is expected to enable novel design of ultrasound-triggered sensors and actuators made of polymeric gels, and can also enlighten the application of ultrasonic waves in biomedical engineering.

1. Introduction

Polymeric gel is often immersed in a liquid environment and can be regarded as an assembly of three-dimensional cross-linked networks and small molecule solvent. With the migration of small molecules in and out of the gel when subjected to external stimuli, it is capable of undergoing large and recoverable elastic deformation. This functionality of polymeric gel enables applications such as carriers for drug delivery (Duncan, 2003; Fischel-Ghodsian et al., 1988; Jeong et al., 1997), actuators and sensors in microfluidic devices (Beebe et al., 2000; Calvert, 2009), and tissue engineering matrices (Lee and Mooney, 2001; Luo and Shoichet, 2004). Recently, there is considerable interest in harnessing acoustic waves to actuate large and nonlinear deformation in polymeric gels, with promising applications in medical devices, microfluidic manipulation devices, adaptive robots, etc. (Huebsch et al., 2014; Xin and Lu, 2016b). With regard to large deformation of gels coupled with diffusion mass transport under either mechanical or electric loading, there already exist a great deal of theoretical works (Biot, 1941; Chester and Anand, 2010; Hong et al., 2008; Tanaka and Fillmore, 1979). However, there is yet a comprehensive acoustomechanical field theory of acoustic-triggered deformation and mass transport of polymeric gels. Such a theory is formulated in the present study. This work should be of paramount importance for gain a fundamental insight into a series of ultrasound techniques and thus can enlighten the widespread application of ultrasound technique in biomedical engineering (De Cock et al., 2016; Zhang et al., 2016).

The Poynting theory says that an acoustic wave propagating in a medium also carries momentum flux in the direction of wave passing on, which gives rise to a back acoustic radiation pressure on the source (Post, 1953, 1960). Generally, the radiation stress is a field concept since it is induced by acoustic momentum transfer between adjacent medium particles and directly deduced from the nonlinearity of acoustic field. Lord Rayleigh firstly gave the expression of acoustic radiation stress in compressional fluids (Rayleigh, 1905), while Brillouin firstly pointed out the second-rank tensor nature of acoustic radiation stress (Beyer, 1978). They all concluded that the radiation stress is proportional to the mean mechanical energy density $\langle E \rangle$ of medium particle motion. Subsequently, enormous efforts are devoted to investigating radiation forces acting on rigid or compressible spheres (Doinikov, 1994; Hasegawa and Yosioka, 1969), acoustical trapping and tweezers (Caleap and Drinkwater, 2014; Shi et al., 2009), acoustic levitation and contactless handling of matter (Brandt, 2001; Foresti and Poulikakos, 2014), deforming fluid interface and biological tissue (Issenmann et al., 2008; Mishra et al., 2014; Walker, 1999), etc. All these researches demonstrate the fact that acoustic radiation stress generated by focused acoustic input is sufficiently large to levitate a metallic sphere and induce material deformation.

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Since the elastic moduli of polymeric gels typically range from dozens of times Pa to several times kPa, acoustic radiation stress with magnitude of mega pascal can induce large deformation in the gel (Mishra et al., 2014; Xin and Lu, 2016b). As previously mentioned, acoustically triggered gel deformation is attractive for a variety of applications. Therefore, it is necessary to formulate a nonlinear acoustomechanical theory for polymeric gels. In the present study, a nonlinear acoustomechanical field theory is developed in Eulerian coordinates for polymeric gels coupled with diffusion mass transport, by combining the nonlinear elasticity theory of gels and the acoustic radiation stress theory. For illustration, the large deformation and mass transport behavior of gels is separately analyzed under biaxial constraint, uniaxial constraint and freestanding conditions. For convenience, acoustic radiation stress/force will be called as acoustic stress/force. This work provides a theoretical guideline for designing novel acoustically triggered sensors and actuators made of polymeric gels.

2. Inhomogeneous field theory of gels

An inhomogeneous field theory of polymeric gels is formulated below in three-dimensional space. The Eulerian coordinates are adopted, since it is more intuitive and convenient for theoretical analyses when all the field quantities are expressed in true values. The polymeric gel is actually an assembly of a three-dimensional cross-linked network of long polymers and one type of small molecules, which is formed when dry networks immerse in and imbibe external solvent. The external solvent is in self-equilibrium and can be regarded as a virtual polymeric gel with networks of vanishing elastic moduli, so that the whole system can be modeled using a uniform field theory. In the field theory, the volume integrals cover both the gel and the solvent, and the surface integrals cover all the interfaces between the gel and the solvent. Let the dry network at undeformed state be the reference configuration. Let each material particle be labeled using its coordinate $\mathbf{X}$ in the reference configuration, and let each spatial point be labeled using its coordinate $\boldsymbol{x}$ in the current configuration. The deformation gradient $\mathbf{F}=\partial\boldsymbol{x}/\partial\mathbf{X}$ maps the reference configuration $\mathbf{X}$ to the current configuration $\boldsymbol{x}$. The Cauchy stress is related to the first Piola–Kirchhoff stress as $\sigma=\mathbf{s}\cdot\mathbf{F}^{\mathbf{T}}/\det(\mathbf{F})$. Let $dV(\boldsymbol{x})$ be a volume element with mass density $\rho(\boldsymbol{x})$ and body force $\mathbf{f}^b(\boldsymbol{x},t)$. Let $\mathbf{n}(\boldsymbol{x})dA(\boldsymbol{x})$ be a surface element with surface force $\mathbf{f}^s(\boldsymbol{x},t)$, where $dA(\boldsymbol{x})$ is the area of the element and $\mathbf{n}(\boldsymbol{x})$ is the unit vector normal to the interface between two materials, labeled as − for pointing toward outside and + for pointing toward inside. Force balance of the volume element is represented by $\partial\sigma/\partial\mathbf{x}+\mathbf{f}^b=\rho\partial^2\mathbf{u}/\partial t^2$, with force boundary condition $\boldsymbol{\sigma}\cdot\mathbf{n}=\mathbf{f}^s$ and displacement field $\mathbf{u}(\boldsymbol{x},t)$.

Now consider an acoustomechanical problem coupled with diffusion mass transport. Essentially, the acoustomechanical problem is a static field problem on the basis of a dynamic field problem. Because the acoustic (radiation) stress is intrinsically a time-averaged stress over one oscillation cycle of an acoustic wave, it is a static stress regardless of time. This static acoustic stress eventually causes static material deformation. Consequently, the acoustomechanical problem is a dynamic field problem for wave propagation but a static field problem for large material deformation. In the present article, all the terms associated with large material deformation are pure static terms without including any inertial terms for wave propagation. In the current configuration, the volume force on an element is $f_i^b dV$ and the surface force on an element is $f_i^s dA$. Force balance dictates that the combination of Cauchy stress and acoustic stress should satisfy:

$$\int\left(\sigma_{ij}-\tau_{ij}\right)\frac{\partial\xi_i}{\partial x_j}dV=\int f_i^b\xi_i dV+\int f_i^s\xi_i dA \tag{1}$$

for arbitrary test function $\xi_i(\mathbf{x})$ where τ_{ij} is the acoustic stress (pressure), representing compression when it is positive and tension when it is negative. Here the acoustic (radiation) stress is generated due to the momentum transfer between adjacent particles in the process of ultrasonic wave propagation, which can be calculated from the acoustic fields inside and outside of the material. While the Cauchy stress is generally equal to outside mechanical stress. Following the Gauss divergence theorem, one has:

$$\int\left(\sigma_{ij}-\tau_{ij}\right)\frac{\partial\xi_i}{\partial x_j}dV=\int\left(\sigma_{ij}^{-}-\sigma_{ij}^{+}\right)n_i\xi_i dA-\int\left(\tau_{ij}^{-}-\tau_{ij}^{+}\right)n_i\xi_i dA-\int\frac{\partial\left(\sigma_{ij}-\tau_{ij}\right)}{\partial x_j}\xi_i dV \tag{2}$$

The test function $\xi_i(\mathbf{x})$ is considered to be continuous across the material interface, while the stress can be discontinuous at the interface. The weak form of the force balance condition in Eq. (1) can be rewritten as:

$$\frac{\partial\left(\sigma_{ij}(\boldsymbol{x},t)-\tau_{ij}(\boldsymbol{x},t)\right)}{\partial x_j}+f_i^b(\boldsymbol{x},t)=0 \tag{3}$$

in the volume, and

$$\left[\left(\sigma_{ij}^{-}(\boldsymbol{x},t)-\sigma_{ij}^{+}(\boldsymbol{x},t)\right)-\left(\tau_{ij}^{-}(\boldsymbol{x},t)-\tau_{ij}^{+}(\boldsymbol{x},t)\right)\right]n_i(\boldsymbol{x},t)=f_i^s(\boldsymbol{x},t) \tag{4}$$

at the interface. These equations describe the general force balance condition of the acoustomechanical system.

Next consider the diffusion phenomenon of mass transport between polymeric gel and external solvent. The small molecules penetrating in and out of the gel with chemical potential $\mu(\boldsymbol{x},t)$ give rise to the change of the chemical potential and the swelling stress. In the current configuration, let $r(\boldsymbol{x},t)$ be the number of small molecules generated by sources (e.g., chemical reaction) inside the gel per unit time and volume, and let $i(\boldsymbol{x},t)$ be the number of small molecules generated by sources in the interface element per unit time and area. Mass conservation of the small molecules requires:

$$\frac{\partial c(\boldsymbol{x},t)}{\partial t}+\frac{\partial j_i(\boldsymbol{x},t)}{\partial x_i}=r(\boldsymbol{x},t) \tag{5}$$

in the volume, where $c(\boldsymbol{x},t)$ is the concentration of small molecules in a volume element, $j_i(\boldsymbol{x},t)$ is the number flux of small molecules that migrate through unit area of the interface element. Also, at the interface:

$$\left(j_i^{+}(\boldsymbol{x},t)-j_i^{-}(\boldsymbol{x},t)\right)n_i(\boldsymbol{x},t)=i(\boldsymbol{x},t) \tag{6}$$

These diffusion balance conditions can be rewritten in a weak form by applying an arbitrary test function $\zeta(\boldsymbol{x})$. Specifically, multiplying this test function on Eq. (5), integrating over the gel volume and adopting Eq. (6), one obtains:

$$\int\frac{\partial c}{\partial t}\zeta dV=\int j_i\frac{\partial\zeta}{\partial x_i}dV+\int r\zeta dV+\int i\zeta dA \tag{7}$$

This weak form of mass conservation is completely equivalent to Eqs. (5) and (6).

To complement the above field equations expressed in the form of force balance law and mass conversation law, the material law is proposed below by applying the principle of minimization potential energy. Generally, the constitutive relation of hyperelastic matters can be expressed using the Helmholtz free energy function, which is dependent upon the strain or concentration of molecules. With an arbitrary small virtual perturbation, the virtual changes of the free energy can be described as:

$$\delta W_e=\frac{\partial W_e(\boldsymbol{\varepsilon})}{\partial\varepsilon_{ij}}\delta\varepsilon_{ij},\ \delta W_a=\frac{\partial W_a(\boldsymbol{\varepsilon})}{\partial\varepsilon_{ij}}\delta\varepsilon_{ij},\ \delta W_m=\frac{\partial W_m(c)}{\partial c}\delta c \tag{8}$$

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