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Analysis of Dilatation in a Multilayered Spherical Hydrogel

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Abstract

The stresses and displacements in a dual-layer spherical hydrogel subjected to a constant dilatation in the outer layer are derived analytically. Both the inner core and the outer layer are described by a second-order nonlinear elasticity model. The results show the importance of nonlinearity, and indicate that the surface displacements or swelling and the interfacial radial or tangential stresses can be significantly moderated by a judicious choice of the elastic constants of the two layers.

Keywords: hydrogel composite; nonlinear elasticity; dilatation; swelling; interfacial stresses

1. Introduction

The properties of hydrogels have received considerable attention due to their diverse applications in medicine and engineering. For instance, hydrogels have been used as drug-delivery vehicles, scaffolds for skin and vascular tissues, sensors and actuators, and bio-mimicking materials [1,2]. Their properties, such as hydrophilicity and hydrophobicity (which determine the drug release profile), bio-degradability, mechanical strength/integrity, and electrical conductivity, can be controlled and manipulated. Generally speaking, they are super-absorbent, non-cytotoxic, and are responsive to moisture, temperature, pH, electric potential, and ion concentration. There are also attempts to impart several distinct properties, e.g., high mechanical strength and strong hydrophilicity, to hydrogels through the use of multi-layered composites [3,4]. Hydrogels with a single composition may not possess such desirable combination of properties.

This paper presents an analytical model for a dual-layered spherical hydrogel composite, and solves for the displacement and stress fields within it. It employs a second-order non-linear elasticity model for the

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material layers undergoing large deformation. As a first study, a constant dilatation resulting from some stimulus such as temperature or solvent concentration is applied to the outer layer only. The formulation and the solutions are presented in Section 2, selected results are explained in Section 3, and a set of conclusions are given in Section 4.

2. Second-Order Nonlinear Elasticity Model

Figure 1 shows a spherical hydrogel composite consisting of an inner layer or core (denoted by 1) of radius r_1 and an outer layer or shell (denoted by 2) of radius r_2. The composite may be of a hybrid nature, e.g., the core is metallic and the shell is hydrogel. The outer surface of the composite is traction-free and the bonding between the core and shell is assumed to be perfect at the interface. A dilatation of arbitrary profile $\vartheta(r)$, where r is the radial coordinate, may be applied to the composite. In the current study, a constant dilatation of magnitude $\vartheta = \vartheta_0$ is applied to the shell. The geometry and loading constitute a spherically symmetric system where only the dependence of the physical variables on r is of interest. The objective is to calculate the displacement field and the stress field using the second-order elasticity theory of Murnaghan [5]. In the following, an outline of the formulation and the solutions is presented as the mathematical details are rather cumbersome.

In [5], the energy density W of a nonlinear material is written in terms of the first, second and third invariants J_1, J_2 and J_3 of the Lagrangian strain tensor **E**:

$$W = \frac{\lambda + 2\mu}{2} J_1^2 - 2\mu J_2 + \frac{l + 2m}{3} J_1^3 - 2m J_1 J_2 + n J_3 , \tag{1}$$

where λ, μ are the second-order and l, m, n the third-order elastic constants, respectively. For the dual-layered composite, a set of ten elastic constants describe the material properties: λ_1, μ_1, l_1, m_1, n_1 and λ_2, μ_2, l_2, m_2, n_2. Using a perturbation procedure, the radial displacement u_r is written as the sum of the first- and second-order displacements u and w:

$$u_r = u + kw , \tag{2}$$

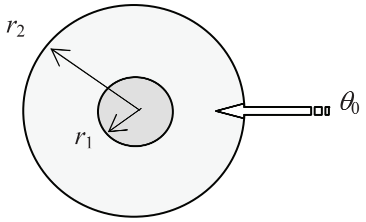

Fig. 1. A spherical hydrogel composite consisting of a core of radius r_1 and a shell of outer radius r_2. A constant dilatation θ_0 is applied to the shell.

where k tracks the different orders of physical quantities in the mathematical expressions. The stresses can be determined from the derivative of the W with respect to $\mathbf{E}$, and written in terms of the displacements. The relevant governing equation is the stress equilibrium equation along the radial direction. Separating this equilibrium equation into its first- and second-order parts (k and k^2 terms) yields:

$$(\lambda+2\mu)(u''+2\frac{u'}{r}-2\frac{u}{r^2})=(3\lambda+2\mu)\frac{d\vartheta}{dr}, \tag{3}$$

and

$$\begin{aligned}&(\lambda+2\mu)(w''+2\frac{w'}{r}-2\frac{w}{r^2})\\ &=-2(\lambda+3\mu+2m)[(u''+2\frac{u'}{r}-2\frac{u}{r^2})u'-\frac{1}{r}(u'-\frac{u}{r})^2]\\ &-(\lambda+2l)(u''+2\frac{u'}{r}-2\frac{u}{r^2})(u'+2\frac{u}{r})-(\lambda+\frac{2}{3}\mu-2l-\frac{2n}{9})9\vartheta\frac{d\vartheta}{dr}.\end{aligned} \tag{4}$$

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The dilatation enters the equilibrium equation as a body force, and the associated terms are placed on the right-hand side of Eqs. (3) and (4). General solutions for the linear displacement u can be solved from Eq. (3) for an arbitrary $\vartheta(r)$, and the nonlinear part w can next be solved from Eq. (4). These two equations are to be solved in both the core and the shell. Appropriate boundary/interface conditions are used, i.e.: (i) the outer surface of the composite is traction-free, (ii) the linear and nonlinear radial displacements and radial stresses must be continuous across the interface at $r=r_1$.

The analytical, closed-form solutions determined include (subscripts 1 and 2 denote the core and the shell respectively): (i) the linear displacements u_1 and u_2, (ii) the nonlinear displacements w_1 and w_2, (iii) the linear radial stresses T_{1r}^{L} and T_{2r}^{L}, the nonlinear radial stresses T_{1r}^{NL} and T_{2r}^{NL}. For brevity, only the expressions for the linear and nonlinear displacements in the shell are given below:

$$u_2=C_1r+C_{-2}\frac{r_1^3}{r^2}, \tag{5}$$

$$w_2=D_1r+\frac{D_{-2}}{r^2}+\frac{D_{-5}}{r^5}, \tag{6}$$

where the constants C_1, C_{-2}, D_1, D_{-2} and D_{-5} are dependent on the ten elastic constants, the radii r_1 and r_2, and the dilatation ϑ_0. These expressions show that the dependence of the linear and nonlinear displacements on r in the outer layer may be dominated by an inverse power law at small r (for a small core) and by a linear law at the surface. The importance of nonlinearity can be judged through the comparison of Eqs. (5) and (6) for specific elastic parameters.

3. Numerical Results

The elastic constants can be taken from theoretical estimates [6]. For the current work, the following reference set of elastic constants is adopted: $\lambda_1=\lambda_2=3570NkT$, $\mu_1=\mu_2=1034NkT$, $l_1=l_2=-3560NkT$, $m_1=m_2=-2427NkT$ and $n_1=n_2=-2355NkT$. The grouped parameter NkT is in the range of 10^{-5} to 10^{-2}

GPa; for biogels it can be smaller. Also, *N*, *k* and *T* are respectively the number of polymer chains in the gel divided by the volume of the dry polymer, the Boltzmann's constant and the absolute temperature. A composite is formed when the two layers have differences in one or more of the five elastic constants. The dilatation is taken to be $\vartheta_0 = 0.35$ for all numerical results below. The displacement results are normalized against the radius r_1, with $r_2 = 2r_1$, while the stress results are normalized with respect to *NkT*.

Figure 2 plots u_r (radial displacement), T_r (radial stress) and T_θ (meridional stress, which equals the azimuthal stress for spherical loading and geometry) as well as their linear and nonlinear parts against r/r_1 for the homogeneous case where the inner and outer layers have the same elastic constants. All plotted quantities are normalized as stated previously. The linear and nonlinear parts add up to the total or actual displacements and stresses. The nonlinear contribution is very significant, as can be expected for a gel. Note that u_r and T_r are continuous but exhibit a change in gradient, while T_θ is discontinuous at r/r_1, the spherical boundary beyond which a constant dilatation is applied. It is also interesting to observe that T_θ is tensile for $r/r_1 < 1$ (core) and compressive for $1 < r/r_1 \leq 2$ (shell). The compressive stress at $r = r_1$, of the order of -1500 *NkT*, is large compared to the elastic constants, and suggests the possibility of instability or buckling there. Such buckling phenomenon may model the instability of tumor growth.

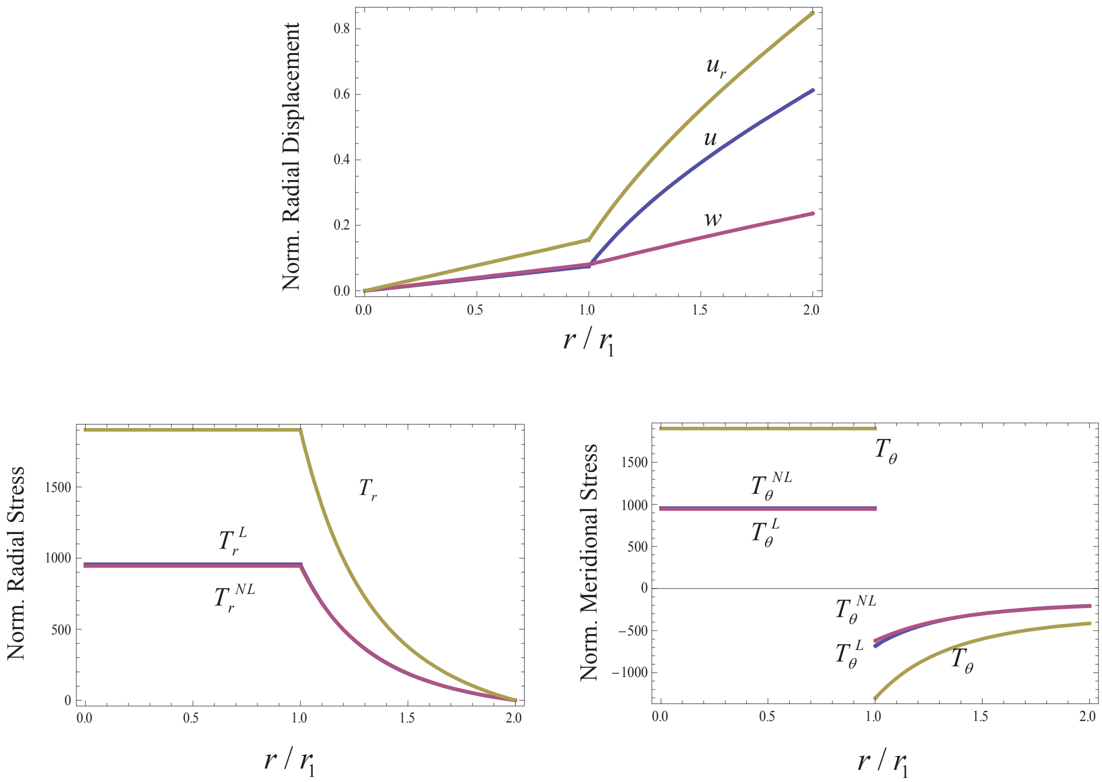

Fig. 2. Variation of the radial displacement, radial stress and meridional stress (all normalized) with the radial coordinate r/r_1 in a homogeneous hydrogel. The linear and nonlinear stresses are comparable in magnitude.

Figure 3 shows how the dependence of T_θ on r/r_1 can be substantially modified by changing just one elastic constant μ_2 from 1034 *NkT* to 103.4 *NkT*. All other parameters are kept at the reference values. The one-order-of-magnitude decrease in μ_2 not only reduces T_θ significantly but also flips the sign of T_θ on the two sides of $r = r_1$. The core is now under compression while the shell is in tension. Finally, Fig. 4 plots the dependence of u_r at the surface on r/r_1. It can be seen that increasing the third-order elastic constant l_2 from -3560 *NkT* to -35600 *NkT*, while keeping all other elastic parameters fixed, substantially increases the swelling of the composite.

These results suggest that the mechanics of hydrogel composites can be manipulated for specific applications. An example is designing patterned hydrogel substrates with spatially varying moduli for stem cells, whose proliferation, morphology and differentiation are strongly sensitive to the substrate moduli [7]. It was demonstrated in [7] that a two order-of-magnitude change of the hydrogel modulus from ~ 3 to 100 kPa resulted in an increased spreading and proliferation of human mesenchymal stem cells. The current results show that multiple desirable properties can likely be obtained from a careful design of the elasticity (especially the nonlinearity) and the composite constituents.

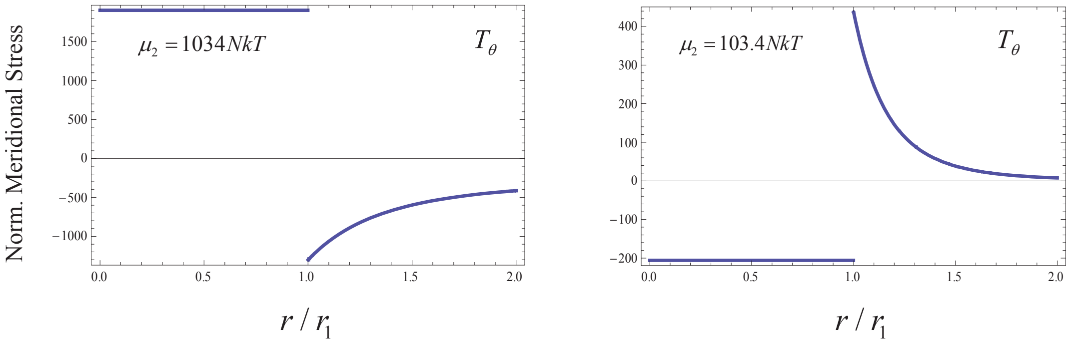

Fig. 3. Variation of the normalized meridional stress with r/r_1 in a hydrogel composite: reducing the shear modulus μ_2 of the shell by an order of magnitude reduces the meridional stress and reverses the signs of the stresses on either side of the interface at $r/r_1 = 1$.

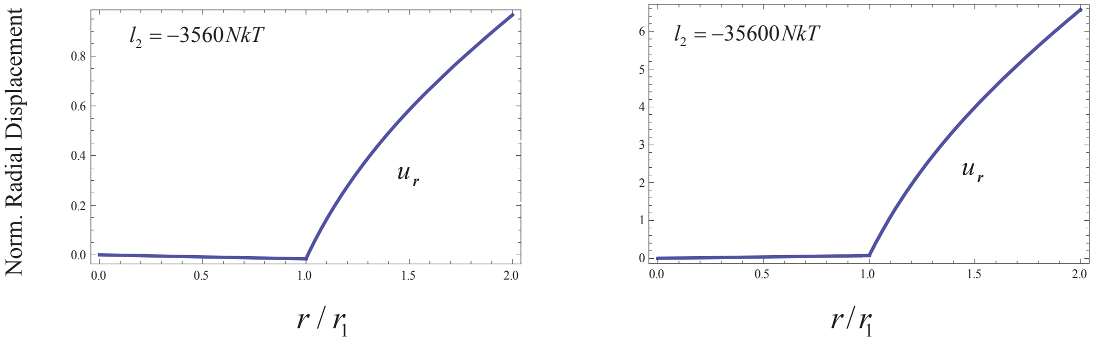

Fig. 4. Variation of the normalized surface radial displacement with r/r_1 in a hydrogel composite: increasing the third-order elastic constant l_2 of the shell by an order of magnitude substantially increases the displacement.

4. Conclusions

Analytical closed-form solutions based on second-order nonlinear elasticity theory have been obtained for a dual-layered hydrogel composite. The results show the essential role of nonlinearity and that a wide range of mechanical behaviors, in terms of stresses and displacements, can be obtained through various combinations of the second- and third-order elastic constants of the composite constituents. From the perspective of designing hydrogel composites for tailored properties and behaviors, these analytical solutions serve as a valuable predictive tool. For instance, it is known that biological growth, morphology and cell differentiation are all significantly influenced by the mechanics of the environment, and hence the use of hydrogels as biological substitutes, substrates and the like requires an in-depth investigation into the mechanical properties of hydrogels.

Investigations are under way to obtain a detailed understanding of the differences in the dependence of hydrogel mechanics on the second- and third-order elastic constants. The coupling between material nonlinearity and composite heterogeneity is unclear and will be of practical interest. A complex dilatation profile mimicking the effect of the stimulus detected by the hydrogel should also be considered. Further research is also needed to explore the specific material combinations that will yield multiple desired properties such as low interfacial stress and high degree of swelling.

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