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A nonlinear, transient finite element method for coupled solvent diffusion and large deformation of hydrogels

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ABSTRACT

Hydrogels are capable of coupled mass transport and large deformation in response to external stimuli. In this paper, a nonlinear, transient finite element formulation is presented for initial boundary value problems associated with swelling and deformation of hydrogels, based on a nonlinear continuum theory that is consistent with classical theory of linear poroelasticity. A mixed finite element method is implemented with implicit time integration. The incompressible or nearly incompressible behavior at the initial stage imposes a constraint to the finite element discretization in order to satisfy the Ladyzhenskaya–Babuska–Brezzi (LBB) condition for stability of the mixed method, similar to linear poroelasticity as well as incompressible elasticity and Stokes flow; failure to choose an appropriate discretization would result in locking and numerical oscillations in transient analysis. To demonstrate the numerical method, two problems of practical interests are considered: constrained swelling and flat-punch indentation of hydrogel layers. Constrained swelling may lead to instantaneous surface instability for a soft hydrogel in a good solvent, which can be regulated by assuming a stiff surface layer. Indentation relaxation of hydrogels is simulated beyond the linear regime under plane strain conditions, in comparison with two elastic limits for the instantaneous and equilibrium states. The effects of Poisson's ratio and loading rate are discussed. It is concluded that the present finite element method is robust and can be extended to study other transient phenomena in hydrogels.

1. Introduction

Hydrogels consist of crosslinked polymer chains and solvent molecules (e.g., water). The crosslinked polymer chains form a three-dimensional network structure through which the smaller solvent molecules can migrate. The response of hydrogels to external forces or chemical stimuli is generally a transient process involving solvent diffusion and deformation coupled via the chemo-mechanical interactions between the polymer network and the solvent. Both linear and nonlinear theories have been proposed to model the transient responses of hydrogels subject to various mechanical and chemical conditions. Tanaka et al. (1973) derived a linear diffusion equation by treating the gel as a mixture of solid and liquid with a coefficient of friction for the interaction. Another linear approach was proposed by Scherer (1989), who extended the linear poroelasticity theory to model the gel as a continuum phase with solvent concentration and pore pressure. Recently, the theory of linear poroelasticity has been used extensively in combination with experimental measurements for characterizing the

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mechanical and transport properties of polymer gels (Hui et al., 2006; Galli and Oyen 2009; Hu et al., 2010; Yoon et al., 2010; Chan et al., 2012; Kalcioglu et al., 2012). In spite of remarkable success, it is well known that the linear theory is limited to relatively small deformation, while large deformation is common for hydrogels. On the other hand, a variety of nonlinear approaches have been proposed for coupling large deformation and transport processes in gels (Dolbow et al., 2004; Hong et al., 2008; Birgersson et al., 2008; Doi, 2009; Chester and Anand, 2010; Duda et al., 2010; Wang and Hong, 2012). In particular, the nonlinear continuum theory proposed by Hong et al. (2008) was found to be consistent with Biot's linear poroelasticity theory (Biot, 1941) for small deformation of an isotropically swollen gel (Bouklas and Huang, 2012).

The present study aims to develop a transient finite element method for large deformation of hydrogels based on a nonlinear continuum theory. Previously, finite element methods for equilibrium analyses of hydrogels have been developed (Hong et al., 2009a; Kang and Huang, 2010a), without considering the diffusion kinetics. More recently, several implementations of transient finite element methods for hydrogels have been reported (Zhang et al., 2009; Wang and Hong, 2012; Lucantonio et al., 2013; Toh et al., 2013). These implementations have taken advantage of commercial software packages (ABAQUS or COMSOL), which are however less flexible for addressing specific numerical issues. As noted by Wang and Hong (2012), when implementing the finite element method, a mixed formulation must be used with different shape functions for discretization of the displacement and chemical potential in a hydrogel. In fact, such a requirement has been known for finite element methods in linear poroelasticity, which have been studied primarily in the context of geomechanics. The most common method used in the studies of linear poroelasticity is the mixed continuous Galerkin formulation for displacement and pore pressure (Borja, 1986; Wan, 2002; White and Borja, 2008). For either poroelasticity or hydrogels, the transient response can often be decomposed into three stages (Rice and Cleary, 1976; Yoon et al., 2010; Bouklas and Huang 2012): the instantaneous response at $t \to 0$, the transient evolution, and the equilibrium or steady state as $t \to \infty$. At the instantaneous limit, the poroelastic response is similar to the linear elastic response of an incompressible or nearly incompressible solid, which requires the mixed finite element method to satisfy the Ladyzhenskaya–Babuska–Brezzi (LBB) condition (Murad and Loula, 1994; Wan, 2002). First noted by Babuška (1971) and Brezzi (1974), the LBB condition requires that the finite element discretization for incompressible linear elasticity and Stokes flow satisfy the incompressibility constraints in order to produce stable results. An example is provided by the elements in the Taylor–Hood family (Taylor and Hood, 1973) where the spatial discretization for pressure is one order lower than for displacement. If the LBB condition is not satisfied, numerical oscillations in the form of spurious pressure modes would be obtained for the instantaneous response of a nearly incompressible poroelastic medium. As discussed in Murad and Loula (1992), Wan (2002), and Phillips (2005), the numerical oscillations are prevalent in the early stage of transient responses, which would decay in time and eventually converge towards the equilibrium or steady state. In the present study, we show that the implementation of a transient finite element method for hydrogels with coupled diffusion and large deformation should also satisfy the LBB condition to avoid numerical oscillations.

Most of the previous works on transient finite element methods for hydrogels have assumed incompressibility for both the polymer network and the solvent, which imposes a volume constraint on the hydrogel with a relationship between solvent concentration and deformation. In the present study, we relax the volume constraint by introducing a bulk modulus as an additional material property in the nonlinear continuum theory proposed by Hong et al. (2008), which is briefly summarized in Section 2. Section 3 presents the finite element formulation and implementation of a mixed finite element method using the backward Euler scheme for time integration and the Newton–Raphson method for solving the nonlinear problem iteratively. Both equal-order elements and Taylor–Hood elements are implemented for comparison. Numerical results are presented for constrained swelling and flat-punch indentation of hydrogel layers (Sections 4 and 5). The numerical examples are chosen to demonstrate the capability of the finite element method for simulating the transient behavior of hydrogels under various initial/boundary conditions. Section 6 summarizes the findings in terms of both the numerical method and the transient responses of hydrogels.

2. A nonlinear theory

Following Hong et al. (2008), the constitutive behavior of a polymer gel is described through a free energy density function based on the Flory–Rehner theory, which takes the form

$$U(\mathbf{F}, C) = U_e(\mathbf{F}) + U_m(C) \tag{2.1}$$

with

$$U_e(\mathbf{F}) = \frac{1}{2} N k_B T [F_{iK} F_{iK} - 3 - 2 \ln(\det(\mathbf{F}))] \tag{2.2}$$

$$U_m(C) = \frac{k_B T}{\Omega} \left(\Omega C \ln \frac{\Omega C}{1 + \Omega C} + \frac{\chi \Omega C}{1 + \Omega C} \right) \tag{2.3}$$

where $\mathbf{F}$ is the deformation gradient with Cartesian components $F_{ij} = \partial x_i / \partial X_J$, describing the deformation kinematics of the polymer network from a reference frame X_J (i.e., the dry state) to the current frame x_i, and C is the nominal concentration of