A new formulation and computation of the triphasic model for mechano-electrochemical mixtures

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Abstract  From the generalized first law of thermodynamics for an irreversible thermodynamical system, a new set of governing equations for the mixture theory is derived based on the triphasic model for mechano-electrochemical mixtures. It is shown that, in the case of electroneutral solution, a new biphasic mixture theory including the electrochemical effects can be derived from the new governing equations. The chemical-expansion stress representing both the influences of deformation on the fixed charge density and the electric potential of fixed charge field is given. For comparison and verification purposes, the numerical solution for a confined compression problem of a charged hydrated soft tissue is computed using the multiquadric method.

Introduction  Biological tissues are multiphasic materials consisting of various amounts of living cells, extracellular matrix, and interstitial fluid. To a large extent, the biorheological properties of connective tissue are dependent upon the compositional and ultrastructural properties of the extracellular matrix. For soft connective tissues, the extracellular matrix is composed of a collagenous fibrous network which is swollen by a highly hydrophilic glycosaminoglycan gel. All connective tissues exhibit some viscoelastic behaviors to various degrees. Some of these apparent viscoelasticity is ascribable to the intrinsic viscoelasticity of the matrix macromolecules, namely, collagen and glycosaminoglycan.

Biological tissues consist of a large proportion of water. Much of this water is movable and could move as a result of deformation of the tissue matrix. The frictional drag as the interstitial fluid moves relates to the tissue matrix has been regarded as one of the most important mechanisms of energy dissipation during tissue deformation. The physics of interstitial flows is important not only in tissue rheology, but also in other electro-mechanical phenomena as the electrolytic interstitial fluid carrying mobile ions/charges moves relative to the tissue matrix with fixed charges, as well as in other chemomechanical phenomena during the concomitant movement of the interstitial water as small mobile ions move down a concentration gradient. This understanding is important to our appreciation of the transport mechanisms in biological tissues as well as the transduction mechanisms of physical stimuli in the microenvironment within the tissue.

In this paper, a new set of governing equations for the triphasic mixture theory based on the triphasic model proposed by Lai et al. [1] is derived from the generalized first law of thermodynamics for an irreversible thermodynamical system. In the newly derived model, the ionic concentration is expressed by the chemical energy and the chemical expansion stress is represented by the electrostatic energy. The effect of the diffusive resistance is then included in the energy dissipation. This greatly simplifies the governing equations for the triphasic model given by Lai et al. [1]. For an electroneutral solution, i.e. in absence of electric current, the new governing equations can be simplified to a new biphasic mixture model which includes the electrochemical effects. For comparison and verification purposes, the numerical solution for a one-dimensional confined compression problem of a charged hydrated soft tissue is computed using the multiquadric (MQ) method which was developed by Hon et al. [12] for solving the biphasic mixture model.

Preliminary assumptions and notations in triphasic model  Hydrated biological soft tissues such as articular cartilage have been idealized by a triphasic model [1, 2], which consists of three phases: (1) a solid phase including the collagen fibres, proteoglycan aggregates (PGA) with its fixed negative charges, superscribed by s, (2) an interstitial water phase, superscribed by w, and (3) an ion phase of dissolved salt consisting of cations (Na$^+$) and anions (Cl$^-$), superscribed by i. Each phase is assumed to be intrinsically incompressible. The solution, which consists of
solvent water and solute ions, is called fluid phase, superscribed by f (f = w ∪ i). It is assumed that the solution is electroneutral and no electric current occurs. The electrostatic effect of fixed charge groups along the glycosaminoglycan (GAG) chains is considered in this paper.

(A) Density and concentration

The volume fraction of each phase \( x \) (\( x = s, w, i \)) is given by

\[
\phi^x = \frac{V^x}{V} \quad (x = s, w, i)
\]

in which \( V^x \) is the true volume of phase \( x \), \( V \) the mixture volume, which is equal to the apparent volume of each phase \( x \), \( \phi^x(x = s, w, i) \) satisfy the continuity condition of mixture:

\[
\sum_{x=s,w,i} \phi^x = 1 .
\]

The apparent mass density of phase \( x \) (\( x = s, w, i \)) is

\[
\rho^x = \phi^s \rho^s + \phi^w \rho^w \quad (x = s, w, i),
\]

where \( \rho^s \) is the true mass density of phase \( s \). The total density of the mixture, i.e., the tissue, is

\[
\rho = \sum_{x=s,w,i} \rho^x = \sum_{x=s,w,i} \phi^x \rho^x .
\]

In the study of the chemical effects, the molar concentration per unit solution (fluid phase) volume,

\[
c^x = \frac{\rho^x}{\phi^x M^x} \quad (x = s, w, i),
\]

is used. Here, \( M^x \) is the molecular weight of species \( x \) and \( \phi^x \) is assumed to be comparatively very small so that \( \phi^x \approx \phi^s \).

Basically there are two mechanisms for tissue swelling effect resulted from the ionic phase. The first is the well known Donnan osmotic pressure caused by the concentration difference between the interstitial solution and the external bathing solution. The other is the so-called chemical-expansion stress effect by the electrostatic repulsive forces of the fixed charges. It is assumed that the number of fixed charges per unit mass of solid phase is kept constant at any time [1].

The molar concentration of the negative fixed charges based on the apparent volume \( V \) of the solid phase is then defined by:

\[
c^f = \frac{\mathrm{d}n^f}{\mathrm{d}V} = c_0^f J ,
\]

where \( n^f \) is the total molar number of fixed charges, \( c_0^f = \frac{\mathrm{d}n^f}{\mathrm{d}V_0} \) is the value of \( c^f \) at the reference configuration. The volume ratio of apparent solid phase,

\[
J = \frac{\mathrm{d}V_0}{\mathrm{d}V},
\]

is related to the Green strain tensor \( E \) of the apparent solid phase as follows:

\[
\frac{1}{J} = 1 + 2 \mathcal{F}_1(E) + 4 \mathcal{F}_2(E) + 8 \mathcal{F}_3(E) \approx 1 + \mathcal{F}_1(E) ,
\]

where \( \mathcal{F}_1(E) = \mathrm{tr}(E) \), \( \mathcal{F}_2(E) \), and \( \mathcal{F}_3(E) \) are the first, second, and third invariant of Green strain tensor \( E \). We note here that the element of the triphasic mixture is referred to the porous solid matrix, i.e., the element of the apparent solid phase, so that \( J = J, E = E \). For infinitesimal deformation, we obtain from (7) that

\[
J = 1 - \mathrm{tr}(E) .
\]

Substituting Eq. (8) into (6) we have

\[
c^f = c_0^f (1 - \mathrm{tr}(E)) ,
\]

The fixed negative charges on the GAG attract mobile cations \( Na^+ \) to maintain the macroscopic electroneutral state. By assuming that each negative fixed charge attracts a cation \( Na^+ \), the molar concentration of the attracted cations \( Na^+ \) per unit fluid volume can be defined as

\[
c^f = \frac{\mathrm{d}n^f}{\mathrm{d}V} = c_0^f \left( 1 + \frac{\phi^s}{\phi^f \mathrm{tr}(E)} \right) ,
\]

and for infinitesimal deformation,

\[
\phi^f = 1 - \phi^s J = \phi^f \left( 1 + \frac{\phi^s}{\phi^f \mathrm{tr}(E)} \right) .
\]

(B) Electroneutrality

Denote the concentration of free cations \( (Na^+)_f \) and anions \( Cl^- \) in a solution of electroneutral salt per unit fluid volume by \( c^+_f = c^- = c^i \). Due to the presence of additional attracted cations, the concentration of \( Na^+ \) is

\[
c^+ = c^+_f + c^a = c^i + c^a .
\]

The electric current density, denoted by \( I \), is calculated by

\[
I = F_C \phi^w \left[ c^+_f (v^+ - v^s) + c^a (v^a - v^s) - c^- (v^- - v^s) \right] ,
\]

where \( F_C = 96.485 \) C/mol is the Faraday constant and \( v^+, v^a, v^- , v^s \) are the velocities of the free cation, attracted cation, anion, and solid matrix respectively. Due to the absence of electric current and the fact that the velocity of attracted cations is macroscopically equal to \( v^s \), Eq. (13) becomes

\[
0 = F_C \phi^w \left[ c^i (v^+ - v^s) + c^a (v^a - v^s) - c^- (v^- - v^s) \right] .
\]

Hence,

\[
v^+ = v^- \equiv v^i .
\]

In other words, if the electric current density in an electroneutral solution is zero, the ion phase can be treated as an uniform phase for the consideration of mechanical effects such as movement and deformation.

(C) Continuity of mixture consisting of incompressible components

The velocities \( v^s, v^w \) and \( v^i \) of phases \( s, w, i \) respectively must satisfy the continuity condition. The continuity equation for each phase \( x \) is

\[
\frac{\partial \rho^x}{\partial t} + \nabla \cdot (\rho^x v^x) = 0 \quad (x = s, w, i) ,
\]

i.e.

\[
\frac{\partial \rho^x}{\partial t} + \nabla \cdot (\rho^x v^x) = 0 \quad (x = s, w, i) .
\]