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INTRA-EXTRAFIBRILLAR MIXTURE FORMULATION OF SOFT CHARGED HYDRATED TISSUES

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The volume of the intrafibrillar water space – i.e. the water contained inside the collagen fibers – is a key parameter that is relevant to concepts of connective tissue structure and function. Because existing theories of finite deformation of cartilaginous tissues do not distinguish between intra- and extrafibrillar water, we derive a chemo-electro-mechanical formulation of quasi-static finite deformation including such distinctions. The model features a porous solid saturated with two fluid compartments, in which an arbitrary number of solutes are dissolved. Each fluid compartment has its own fixed charge density. Incompressible deformation is assumed. Each fluid compartment is assumed to be locally electroneutral. Balance laws are derived for each constituent and for the mixture as a whole. A Lagrangian form of the second law of thermodynamics for incompressible porous media is used to derive the constitutive restrictions of the medium. The material properties are shown to be contained in one strain energy function and a matrix of friction tensors. The formulation is consistent with the experimental finding (Maroudas et al., 1991) that the intrafibrillar water content is regulated by the osmotic pressure gradient between the extra- and intrafibrillar compartments.

Key words: mixture theory, porous media, cartilage, intervertebral disc

Notation

$\boldsymbol{b}_i^\alpha$ – body force of constituent α in phase i per constituent unit volume
$\boldsymbol{b}$ – body force of the mixture per mixture unit volume
$\mathbf{B}^{\beta\gamma}$ – frictional tensor between the constituents β and γ

C_i^α – concentration of constituent α in phase i per current mixture unit volume
$\mathbf{E}$ – Green strain tensor of the solid
F – Faraday constant
$\mathbf{F}$ – deformation gradient tensor of the solid
J – ratio of current mixture volume to initial mixture volume
n_i^α – ratio of current volume of constituent α in phase i to current mixture volume
N_i^α – ratio of current volume of constituent α in phase i to initial mixture volume
p – pressure
$\boldsymbol{q}_i^\alpha$ – heat flux of constituent α in phase i
$\boldsymbol{q}$ – an energy flux of the mixture
r_i^α – heat supply of constituent α in phase i per constituent unit volume
r – heat supply of the mixture per mixture unit volume
$\boldsymbol{v}_i^\alpha$ – velocity of constituent α in phase i
$\boldsymbol{v}^{\beta s}$ – Lagrangian velocity of constituent β relative to the solid
$\boldsymbol{v}^{qs}$ – Lagrangian form of energy flux $\boldsymbol{q}$
$\overline{V}^\alpha$ – molar volume of constituent α
W – Helmholtz free energy per initial mixture unit volume
z^α – valence of constituent α
ϵ_i^α – internal energy of constituent α in phase i per constituent unit volume
ϵ – internal energy of the mixture per unit volume
η_i^α – entropy of constituent α in phase i per constituent unit volume
η – entropy of the mixture per unit volume
η_0 – entropy of the mixture per initial mixture unit volume
μ_i^β – electrochemical potential of constituent β in phase i per constituent unit volume
π_i^α – momentum interaction of constituent α in phase i
Ψ_i^α – Helmholtz free energy of constituent α in phase i per constituent unit volume
ψ_i^α – Helmholtz free energy of constituent α in phase i per mixture unit volume
σ_i^α – partial Cauchy stress of constituent α in phase i
θ_i^α – temperature of constituent α in phase i
θ – temperature of the mixture
∇_0 – gradient tensor in initial configuration

1. Introduction

Chondrocytes occupy only a small fraction of the total volume of cartilaginous tissues. Therefore, the extracellular matrix determines the instantaneous electro-chemomechanical properties of the tissue. Although the tissue has a solid appearance, the extracellular matrix consists mainly of water (60 to 85% wet weight). The non-aquous components of the extracellular matrix are: collagen fiber network (30-60% dry weight), molecular network of proteoglycans (10-60% dry weight), small ions (3% dry weight) and some larger solutes. The entanglement of the collagen and proteoglycan network force them to behave as a single porous solid within which water and ions are free to move (Maroudas and Schneiderman, 1987). Experiments on solute transport have shown that larger solutes including the proteoglycan macromolecules cannot penetrate into the water present within the collagen fibrils (Wells, 1973). The strong water binding capacity of the tissue is caused by ionisation of the proteoglycans (Urban et al., 1979). The ensuing negative charge fixed to the solid is counteracted by the corresponding small positive counterions in the fluid. These charges – mainly Na^+ – do not bind to the proteoglycans and result in a variety of features including swelling, electro-osmosis, streaming potentials and streaming currents. Conversely, the osmotic activity inside the fibrillar collagen must be very low. The collagen molecules have many ionisable groups, but most of them are self compensated via electrostatic linkages (Li and Katz, 1976). It is, therefore, obvious that the fluid volume contained in the extracellular matrix is composed of two functionally distinct compartments, the intrafibrillar water inside the collagen fibrils and the extrafibrillar water outside the collagen network. Maroudas et al. (1991) showed by means of X-ray diffraction measurements that the intrafibrillar water content is a function of the osmotic pressure difference between the extrafibrillar and intrafibrillar spaces and ranges between 0.8 and 1.3 g/g dry collagen. Since the proteoglycans are restricted to the extrafibrillar compartment, it is not their overall concentration in the matrix which is relevant, but their actual concentrations in the extrafibrillar space. This effective concentration depends on the proportions of water in the extra- and intrafibrillar space compartments. In particular for compressed cartilage – a condition particularly relevant for its mechanical integrity – the calculations based on total water content can suffer from a seriously error (Maroudas et al., 1991).

To the best of our knowledge, all models dealing with cartilage electrochemomechanics (Lai et al., 1991; Simon et al., 1996; Huyghe and Janssen, 1997) base their calculations on the total water content. For this reason the present study aims at developing a blueprint of a mixture model including a distinction between the two fluid compartments. Earlier work in the field of geomechanics has been dealing with fluid-solid mixtures in which the fluid was subdivided into two or more compartments. Aifantis (1977) introduced the concept of multiporosity for deforming media that are characterised by several distinct families of diffusion or flow paths. A special case of this concept, in which only two degrees of diffusivity were included, was applied to fissured rock formations, in which most of the fluid volume is located in low permeability pores of the rock and most of the permeability is associated with the fissures (Wilson and Aifantis, 1982). Huyghe and van Campen (1995a) and later Vankan et al. (1996) derived a multiporosity model for blood perfused soft tissue. This model features a spectrum of porosities intercommunicating with one another through anisotropic interfaces and has been applied successfully to several biological situations (Vankan et al., 1997, 1998). Murad and Cushman (1997) developed a dual porosity model for clays in which the vicinal water between clay platelets is treated separately from the bulk water. They include swelling through adsorption forces between solid and fluid. They do not include electrostatic forces associated with the ionisation of the clay platelets, although they acknowledge the need for it. The present paper develops a dual porosity model of a saturated deforming porous medium including ionisation of the solid in both porosities. Although the primary field of application that we have in mind in this paper is the separation of the extracellular space of cartilaginous tissues in a intrafibrillar compartment with low ionisation and an extrafibrillar space with high ionisation, the theory may also find application in other fields. These include modelling of clays and shales or the modelling of biological tissues including a separation between the intracellular space and the extracellular space.

2. Theory

We consider a porous solid (s) saturated by two fluid compartments, one intrafibrillar (1) the other extrafibrillar (2). We assume that the solid and all constituents α constituting the fluid compartments are intrinsically incompressible, i.e., if one adds a mass m of constituent α to the mixture, the added volume of the mixture is always proportional to the mass m, irrespective of its value m, of the initial composition of the mixture or of the state of deformation. The proportionality constants are the intrinsic densities

ρ^{si} and $\rho_j^{\alpha i}$

$$\rho^{si} = \frac{\rho^s}{n^s}\rho_j^{\alpha i} = \frac{\rho_j^\alpha}{n_j^\alpha} \qquad j = 1,2 \tag{2.1}$$

In Eq (2.1) the superscript s means solid, the subscript j specifies the compartment $j = 1$ intrafibrillar and $j = 2$ extrafibrillar, while the superscript α specifies the constituent. ρ_j^α represents the apparent density of the αth constituent in the jth phase. n_j^α is the volume fraction of the αth constituent in the jth phase. We use the word incompressible after Bowen (1980); i.e., neither deformation of the constituents nor changes in mixing ratios result in changes of intrinsic densities. Although it is more common to express solute quantities in molar or mass values than in volume values, we will use volume fractions throughout this paper, to reduce the number of symbols to introduce. Due to incompressibility, the ratio between mass and volume is constant, and the two quantities are interchangable in the equations. Excluding chemical reactions between constituents, but including mass transfer between the same constituents in different phases, the mass balance of each phase is then written as

$$\frac{\partial n_i^\alpha}{\partial t} + \nabla \cdot (n_i^\alpha \boldsymbol{v}_i^\alpha) = c_i^\alpha \qquad i = 1,2 \tag{2.2}$$

in which $\boldsymbol{v}_i^\alpha$ is the velocity of constituent α of phase i, c_i^α the volume of constituent α added into phase i from the other phase per unit time and per unit current mixture volume. Conservation of mass of each constituent requires

$$\sum_{i=1,2} c_i^\alpha = 0 \quad \text{for all constituents } \alpha \tag{2.3}$$

while for the solid we find

$$c^s = 0 \tag{2.4}$$

As we assume saturation, we find

$$n^s + \sum_{i=1,2}\sum_{\alpha} n_i^\alpha = 1 \tag{2.5}$$

The summation over α is taken over all constituents of the ith phase. Differentiation of Eq (2.5) with respect to t and substitution of the mass balance equations (2.2) yields the mass balance of the mixture

$$\nabla \cdot \boldsymbol{v}^s + \sum_{i=1,2}\sum_{\beta} \nabla \cdot \left[n_i^\beta(\boldsymbol{v}_i^\beta - \boldsymbol{v}^s)\right] = 0 \tag{2.6}$$

We refer current descriptors of the mixture to an initial state of the porous solid. The deformation gradient tensor $\mathbf{F}$ transforms an infinitesimal material line segment in the initial state of the solid into the corresponding line segment in the current state. The relative volume change from the initial state to the current one is the determinant of the deformation gradient tensor $J = \det \mathsf{F}$. If we introduce the volume fractions

$$N_i^\alpha = J n_i^\alpha \tag{2.7}$$

per initial unit volume, we can rewrite the mass balance equation (2.2) as follows

$$\frac{D^s N_i^\alpha}{Dt} + J \nabla \cdot [n_i^\alpha (\boldsymbol{v}_i^\alpha - \boldsymbol{v}^s)] = J c_i^\alpha \tag{2.8}$$

using the identity

$$\frac{D^s J}{Dt} = J \nabla \cdot \boldsymbol{v}^s \tag{2.9}$$

In Eqs (2.8) and (2.9), $\frac{D^s}{Dt} = \frac{\partial}{\partial t} + \boldsymbol{v}^s \cdot \nabla$ is the time derivative for an observer moving with the solid. The electrostatic interactions between ions and the charged solid are accounted for by means of two electroneutrality conditions, one for the intrafibrillar space and one for the extrafibrillar one. The electroneutrality conditions require that

$$C_i^{fc} + \sum_\alpha z^\alpha C_i^\alpha = 0 \qquad i = 1,2 \tag{2.10}$$

in which z^α are the valences, C_i^{fc} the fixed charge density per initial mixture unit volume and C_i^α the current molar concentration of mobile ions in the ith phase per initial mixture unit volume

$$C_i^\alpha = \frac{N_i^\alpha}{\overline{V}^\alpha} \tag{2.11}$$

$\overline{V}^\alpha$ are the partial molar volumes of the solvents and solutes. Eq (2.10) is rewritten in a more convenient differentiated form, using the fixity of the fixed charges $(D^s C_i^{fc}/Dt = 0,\ i = 1,2)$ and Eq (2.11) (Huyghe and Janssen, 1997)

$$\sum_\alpha \frac{z^\alpha}{\overline{V}^\alpha} \left\{ \nabla \cdot [n_i^\alpha (\boldsymbol{v}_i^\alpha - \boldsymbol{v}^s)] - c_i^\alpha \right\} = 0 \tag{2.12}$$

Neglecting inertia, the momentum balance takes the form

$$\nabla \cdot (\boldsymbol{\sigma}^s)^C + \boldsymbol{\pi}^s = n^s \boldsymbol{b}^s \qquad\qquad \nabla \cdot (\boldsymbol{\sigma}_i^\alpha)^C + \boldsymbol{\pi}_i^\alpha = n_i^\alpha \boldsymbol{b}_i^\alpha \tag{2.13}$$

which after summation over all constituents, yields

$$\nabla \cdot \boldsymbol{\sigma}^C - \sum_{i=1,2} \sum_\alpha \rho_i^{\alpha i} c_i^\alpha \boldsymbol{v}_i^\alpha = \nabla \cdot (\boldsymbol{\sigma}^s)^C + \sum_{i=1,2} \sum_\alpha \left[\nabla \cdot (\boldsymbol{\sigma}_i^\alpha)^C - \rho_i^{\alpha i} c_i^\alpha \boldsymbol{v}_i^\alpha \right] = \boldsymbol{b} \tag{2.14}$$

if use is made of the balance condition

$$\boldsymbol{\pi}^s + \sum_{i=1,2} \sum_{\alpha} (\boldsymbol{\pi}_i^\alpha + \rho_i^{\alpha i} c_i^\alpha \boldsymbol{v}_i^\alpha) = \boldsymbol{0} \tag{2.15}$$

where $\boldsymbol{\sigma}^s$ is the partial Cauchy stress of the solid, $\boldsymbol{\sigma}_i^\alpha$ is the partial Cauchy stress tensor of constituent α of the ith phase, $\boldsymbol{\pi}^s$ and $\boldsymbol{\pi}_i^\alpha$ represent the momentum interaction with other constituents, $\boldsymbol{b}_i^\alpha$ the body force of constituent α of the ith phase per unit volume and $\boldsymbol{b} = \boldsymbol{b}^s + \sum_{i=1,2} \sum_\alpha n_i^\alpha \boldsymbol{b}_i^\alpha$ the body force of the mixture per unit volume. The term appearing in Eqs (2.14) and (2.15) involving the volume interaction c_i^α is non-zero in both equations, but negligibly small compared to other terms in the equations in the case of a permeation and diffusion as slow as in cartilaginous tissues. Therefore, we replace Eq (2.14) by

$$\nabla \cdot \boldsymbol{\sigma}^C = \nabla \cdot (\boldsymbol{\sigma}^s)^C + \sum_{i=1,2} \sum_{\alpha} \nabla \cdot (\boldsymbol{\sigma}_i^\alpha)^C = \boldsymbol{b} \tag{2.16}$$

and Eq (2.15) by

$$\boldsymbol{\pi}^s + \sum_{i=1,2} \sum_{\alpha} \boldsymbol{\pi}_i^\alpha = \boldsymbol{0} \tag{2.17}$$

Balance of moment of momentum requires that the stress tensor $\boldsymbol{\sigma}$ be symmetric. If no moment of momentum interaction between components occurs, the partial stresses $\boldsymbol{\sigma}_i^\alpha$ also are symmetric. In this paper we assume all partial stresses to be symmetric. The balance of energy for the αth constituent of the ith phase reads

$$n_i^\alpha \frac{D_i^\alpha \epsilon_i^\alpha}{Dt} = \boldsymbol{\sigma}_i^\alpha : \nabla \boldsymbol{v}_i^\alpha - \nabla \cdot \boldsymbol{q}_i^\alpha + n_i^\alpha r_i^\alpha + \widehat{\epsilon}_i^\alpha \tag{2.18}$$

in which ϵ_i^α is the partial internal energy of the αth constituent of the ith phase per unit volume, $\boldsymbol{q}_i^\alpha$ the heat flux of constituent α of the ith phase, r_i^α the partial heat supply per constituent unit volume and $\widehat{\epsilon}_i^\alpha$ the energy interaction with other constituents per unit volume. D_i^α/Dt is the time derivative for an observer fixed to the αth constituent of the ith phase. A similar energy balance holds for the solid

$$n^s \frac{D^s \epsilon^s}{Dt} = \boldsymbol{\sigma}^s : \nabla \boldsymbol{v}^s - \nabla \cdot \boldsymbol{q}^s + n^s r^s + \widehat{\epsilon}^s \tag{2.19}$$

Total energy balance requires

$$\widehat{\epsilon}^s + \boldsymbol{v}^s \cdot \boldsymbol{\pi}^s + \sum_{i=1,2} \sum_{\alpha} (\widehat{\epsilon}_i^\alpha + \boldsymbol{v}_i^\alpha \cdot \boldsymbol{\pi}_i^\alpha + c_i^\alpha \epsilon_i^\alpha) = 0 \tag{2.20}$$

Substitution of Eq (2.18) into Eq (2.20) yields an equivalent form of the total energy balance

$$\begin{aligned} \frac{D^s \epsilon}{Dt} &= \boldsymbol{\sigma}^s : \nabla \boldsymbol{v}^s - \nabla \cdot \boldsymbol{q}^s - \boldsymbol{v}^s \cdot \boldsymbol{\pi}^s - \epsilon \nabla \cdot \boldsymbol{v}^s + r + \\ &+ \sum_{i=1,2} \sum_{\alpha} \Big\{ \boldsymbol{\sigma}_i^\alpha : \nabla \boldsymbol{v}_i^\alpha - \nabla \cdot [\boldsymbol{q}_i^\alpha + n_i^\alpha \epsilon_i^\alpha (\boldsymbol{v}_i^\alpha - \boldsymbol{v}^s)] - \boldsymbol{v}_i^\alpha \cdot \boldsymbol{\pi}_i^\alpha - c_i^\alpha \epsilon_i^\alpha \Big\} \end{aligned} \tag{2.21}$$

in which

$$\epsilon = \epsilon^s + \sum_{i=1,2} \sum_{\alpha} n_i^\alpha \epsilon_i^\alpha \tag{2.22}$$

is the internal energy of the mixture per unit volume and

$$r = r^s + \sum_{i=1,2} \sum_{\alpha} n_i^\alpha r_i^\alpha \tag{2.23}$$

is the total heat supply of the mixture per unit volume. Under incompressible conditions, the entropy inequality for a unit volume of mixture reads (Bowen, 1980)

$$\frac{D^s \eta}{Dt} - \eta \nabla \cdot \boldsymbol{v}^s + \sum_{i=1,2} \sum_{\alpha} \Big\{ \nabla \cdot \Big[\frac{\boldsymbol{q}_i^\alpha}{\theta_i^\alpha} + n_i^\alpha \eta_i^\alpha (\boldsymbol{v}_i^\alpha - \boldsymbol{v}^s) \Big] - \frac{n_i^\alpha r_i^\alpha}{\theta_i^\alpha} \Big\} \geqslant 0 \tag{2.24}$$

in which η_i^α is the entropy of constituent α per constituent unit volume, $\eta = \sum_{i=1,2} \sum_\alpha n_i^\alpha \eta_i^\alpha$ is the entropy of the mixture per mixture unit volume and θ_i^α the temperature of the constituent α in the ith phase. If the temperature field is assumed the same for all phases, we find

$$\frac{D^s \eta}{Dt} - \eta \nabla \cdot \boldsymbol{v}^s + \nabla \cdot \Big\{ \frac{1}{\theta} \sum_{i=1,2} \sum_{\alpha} [\boldsymbol{q}_i^\alpha + n_i^\alpha \eta_i^\alpha \theta (\boldsymbol{v}_i^\alpha - \boldsymbol{v}^s)] \Big\} - \frac{r}{\theta} \geqslant 0 \tag{2.25}$$

in which θ is the temperature of the mixture. Substitution of the energy balance (2.21) into the entropy inequality eliminates the heat supply r from the inequality

$$-\eta \frac{D^s \theta}{Dt} - \frac{1}{\theta} \boldsymbol{q} \cdot \nabla \theta + \sum_{i=1,2} \sum_{\alpha} \Big(-n_i^\alpha \frac{D_i^\alpha \Psi_i^\alpha}{Dt} + \boldsymbol{\sigma}_i^\alpha : \nabla \boldsymbol{v}_i^\alpha - \boldsymbol{\pi}_i^\alpha \cdot \boldsymbol{v}_i^\alpha \Big) \geqslant 0 \tag{2.26}$$

in which

$$\Psi_i^\alpha = \epsilon_i^\alpha - \theta \eta_i^\alpha \tag{2.27}$$

is the Helmholtz free energy of constituent α per its unit volume and

$$\boldsymbol{q} = \sum_{i=1,2} \sum_{\alpha} [\boldsymbol{q}_i^\alpha + n_i^\alpha \eta_i^\alpha \theta (\boldsymbol{v}_i^\alpha - \boldsymbol{v}^s)] \tag{2.28}$$

is an energy flux vector of the mixture. We introduce the strain energy function

$$W = J \sum_{i=1,2} \sum_{\alpha} n_i^\alpha \Psi_i^\alpha = J \sum_{i=1,2} \sum_{\alpha} \psi_i^\alpha \tag{2.29}$$

as the Helmholtz free energy of a mixture volume which in the *initial* state of the solid equals unity. ψ_i^α is the Helmholz free energy of constituent α in phase i per mixture unit volume. Rewriting Eq (2.26) for the entropy production per initial mixture volume – i.e. we multiply inequality (2.26) by the relative volume change J – we find

$$\begin{gathered}
-J\eta \frac{D^s\theta}{Dt} - \frac{J}{\theta}\boldsymbol{q} \cdot \nabla\theta - \frac{D^sW}{Dt} + J\boldsymbol{\sigma} : \nabla \boldsymbol{v}^s - J\nabla \cdot \sum_{i=1,2} \sum_{\beta} (\boldsymbol{v}_i^\beta - \boldsymbol{v}^s)\psi_i^\beta + \\
+J \sum_{i=1,2} \sum_{\beta} [\nabla(\boldsymbol{v}_i^\beta - \boldsymbol{v}^s) : \boldsymbol{\sigma}_i^\beta - (\boldsymbol{v}_i^\beta - \boldsymbol{v}^s) \cdot \boldsymbol{\pi}_i^\beta] \geqslant 0
\end{gathered} \tag{2.30}$$

The entropy inequality should hold for an arbitrary state of the mixture, complying with the balance laws, incompressibility, saturation and electroneutrality. The balance laws and incompressibility conditions (2.1) are accounted for by means of substitution. The differentiated forms of the saturation condition (2.6) and of the electroneutrality conditions (2.12) are accounted for by means of Lagrange multipliers. The inequality (2.30) shows that the apparent densities ρ_i^α, the body forces $\boldsymbol{b}_i^\alpha$ and the heat supplies r_i^α are already eliminated from the inequality. Therefore the conditions of incompressibility (2.1) are fulfilled by choosing $\rho^s = \rho^{si} n^s$ and $\rho_i^\alpha = \rho_i^{\alpha i} n_i^\alpha$, the momentum balances (2.13) are fulfilled by choosing $\boldsymbol{b}^s$ and $\boldsymbol{b}_i^\alpha$ accordingly and the energy balance is fulfilled by choosing the heat supply r accordingly. Therefore, restrictions still to be fulfilled are the mass balances, saturation and the electroneutralities. The differentiated form of the saturation condition (2.6) is substituted in (2.30) by means of the Lagrange multiplier p

$$\begin{aligned}
&-\eta_0 \frac{D^s\theta}{Dt} - \frac{J}{\theta}\boldsymbol{v}^{qs} \cdot \nabla_0\theta - \frac{D^sW}{Dt} + J\boldsymbol{\sigma}^{eff} : \nabla \boldsymbol{v}^s + \\
&+J \sum_{i=1,2} \sum_{\beta} [\boldsymbol{\sigma}_i^\beta + (pn_i^\beta - \psi_i^\beta)\mathsf{I}] : \nabla(\boldsymbol{v}_i^\beta - \boldsymbol{v}^s) + \\
&+J \sum_{i=1,2} \sum_{\beta} (\boldsymbol{v}_i^\beta - \boldsymbol{v}^s) \cdot (-\nabla\psi_i^\beta + p\nabla n_i^\beta - \boldsymbol{\pi}_i^\beta) \geqslant 0
\end{aligned} \tag{2.31}$$

in which the effective stress $\boldsymbol{\sigma}^{eff}$ is defined as

$$\boldsymbol{\sigma}^{eff} = \boldsymbol{\sigma} + p\mathsf{I} \tag{2.32}$$

the heat flux $\boldsymbol{v}^{qs}$ through an area equal to unity in the initial state of the solid as

$$\boldsymbol{v}^{qs} = \mathsf{F}^{-1} \cdot \boldsymbol{q} \tag{2.33}$$

and the entropy per initial unit volume as

$$\eta_0 = J\eta \tag{2.34}$$

Introducing the restrictions (2.12) into inequality (2.31) by means of Lagrange multipliers λ_i, $i = 1, 2$ and eliminating c_2^β by means of Eq (2.3), yields

$$\begin{aligned}
&-\eta_0 \frac{D^s\theta}{Dt} - \frac{J}{\theta}\boldsymbol{v}^{qs} \cdot \nabla_0\theta - \frac{D^sW}{Dt} + J\boldsymbol{\sigma}^{eff} : \nabla\boldsymbol{v}^s - J\sum_\beta \frac{z^\beta c_1^\beta}{\overline{V}^\beta}(\lambda_1 - \lambda_2) + \\
&+J\sum_{i=1,2}\sum_\beta \left\{\boldsymbol{\sigma}_i^\beta + \left[\left(p + \frac{z^\beta\lambda_i}{\overline{V}^\beta}\right)n_i^\beta - \psi_i^\beta\right]\mathsf{I}\right\} : \nabla(\boldsymbol{v}_i^\beta - \boldsymbol{v}^s) + \\
&+J\sum_{i=1,2}\sum_\beta (\boldsymbol{v}_i^\beta - \boldsymbol{v}^s) \cdot \left[-\nabla\psi_i^\beta + \left(p + \frac{z^\beta\lambda_i}{\overline{V}^\beta}\right)\nabla n_i^\beta - \boldsymbol{\pi}_i^\beta\right] \geqslant 0
\end{aligned} \tag{2.35}$$

in which $\nabla_0 = \mathsf{F}^C \cdot \nabla$ is the gradient operator with respect to the initial configuration. We choose as independent variables the Green strain E, the temperature θ, the Lagrangian form of the volume fractions of the fluid and the ions N_i^β, of the volume interaction terms Jc_1^β and of the relative velocities $\boldsymbol{v}_i^{\beta s} = \mathsf{F}^{-1} \cdot (\boldsymbol{v}_i^\beta - \boldsymbol{v}^s)$ and $\boldsymbol{v}^{qs}$. The dependent variables are the Helmholtz free energy per initial mixture unit volume W, the entropy per initial mixture unit volume η_0, the effective stress $\boldsymbol{\sigma}^{eff}$ of the mixture, the Helmholtz free energy ψ_j^γ of constituent γ, the effective partial stresses $\boldsymbol{\sigma}_j^\gamma + (p + z^\gamma\lambda_j/\overline{V}^\gamma)n_j^\gamma\mathsf{I}$ the effective momentum interactions $\boldsymbol{\pi}_j^\gamma - (p + z^\gamma\lambda_j/\overline{V}^\gamma)\nabla n^\gamma$ and the temperature gradient $\nabla_0\theta$. We apply the principle of equipresence, i.e. all dependent variables depend on all independent variables, unless the entropy inequality requires otherwise

$$\begin{aligned}
&W = W(\mathsf{E}, \theta, N_i^\beta, \boldsymbol{v}_i^{\beta s}, c_1^\beta, \boldsymbol{v}^{qs}) && \eta_0 = \eta_0(\mathsf{E}, \theta, N_i^\beta, \boldsymbol{v}_i^{\beta s}, c_1^\beta, \boldsymbol{v}^{qs}) \\
&\psi_j^\gamma = \psi_j^\gamma(\mathsf{E}, \theta, N_i^\beta, \boldsymbol{v}_i^{\beta s}, c_1^\beta, \boldsymbol{v}^{qs}) && \nabla_0\theta = \boldsymbol{g}_0(\mathsf{E}, \theta, N_i^\beta, \boldsymbol{v}_i^{\beta s}, c_1^\beta, \boldsymbol{v}^{qs}) \\
&\boldsymbol{\sigma}^{eff} = \mathsf{F} \cdot \mathsf{S}^{eff}(\mathsf{E}, \theta, N_i^\beta, \boldsymbol{v}_i^{\beta s}, c_1^\beta, \boldsymbol{v}^{qs}) \cdot \mathsf{F}^C &&
\end{aligned} \tag{2.36}$$

$$
\begin{aligned}
&\boldsymbol{\sigma}_j^\gamma + \Big(p + \frac{z^\gamma \lambda_j}{\overline{V}^\gamma}\Big) n_j^\gamma \mathsf{I} = \mathsf{F} \cdot \mathsf{S}_j^\gamma(\mathsf{E}, \theta, N_i^\beta, \boldsymbol{v}_i^{\beta s}, c_1^\beta, \boldsymbol{v}^{qs}) \cdot \mathsf{F}^C \\
&\boldsymbol{\pi}_j^\gamma - \Big(p + \frac{z^\gamma \lambda_j}{\overline{V}^\gamma}\Big) \nabla n^\gamma = \mathsf{F} \cdot \boldsymbol{p}_j^\gamma(\mathsf{E}, \theta, N_i^\beta, \boldsymbol{v}_i^{\beta s}, c_1^\beta, \boldsymbol{v}^{qs})
\end{aligned}
$$

We apply the chain rule to time differentiation of W, which implies

$$
\begin{aligned}
&-\Big(\eta_0 + \frac{\partial W}{\partial \theta}\Big)\frac{D^s\theta}{Dt} - \frac{1}{\theta}\boldsymbol{v}^{qs} \cdot \nabla_0 \theta + \Big(J\boldsymbol{\sigma}^{eff} - \mathsf{F} \cdot \frac{\partial W}{\partial \mathsf{E}} \cdot \mathsf{F}^C\Big) : \nabla \boldsymbol{v}^s + \\
&-\frac{\partial W}{\partial \boldsymbol{v}^{qs}} \cdot \frac{D^s \boldsymbol{v}^{qs}}{Dt} - \sum_\beta \Big\{ \Big[\frac{\partial W}{\partial \boldsymbol{v}^{\beta s}} \cdot \frac{D^s \boldsymbol{v}^{\beta s}}{Dt} + \frac{\partial W}{\partial c_1^\beta} \frac{D^s c_1^\beta}{Dt} + J c_1^\beta (\mu_1^\beta - \mu_2^\beta)\Big] + \\
&+J \sum_{i=1,2} \sum_\beta [\boldsymbol{\sigma}_i^\beta + (\mu_i^\beta n_i^\beta - \psi_i^\beta)\mathsf{I}] : \nabla(\boldsymbol{v}_i^\beta - \boldsymbol{v}^s) + \\
&+J \sum_{i=1,2} \sum_\beta (\boldsymbol{v}_i^\beta - \boldsymbol{v}^s) \cdot (-\nabla \psi_i^\beta + \mu_i^\beta \nabla n_i^\beta - \boldsymbol{\pi}_i^\beta) \Big\} \geqslant 0
\end{aligned}
\tag{2.37}
$$

in which μ_i^β are the electrochemical potentials of constituents other than the porous solid

$$
\mu_i^\beta = \frac{\partial W}{\partial N_i^\beta} + p + \frac{z^\beta \lambda_i}{\overline{V}^\beta}
\tag{2.38}
$$

Comparison of the above equation to the classical equations of electrochemistry indicates that the Lagrange multiplier p can be interpreted as a pressure, λ_1 as the intrafibrillar electrical potential multiplied by the Faraday constant and λ_2 as the extrafibrillar electrical potential multiplied by the Faraday constant. In Eq (2.37) should be true for any value of the state variables. In view of the set of independent variables, the first term of (2.37) is linear in the time derivative of the temperature $D^s\theta/Dt$, the third term is linear in the solid velocity gradient $\nabla \boldsymbol{v}^s$, the fourth term linear in $D^s\boldsymbol{v}^{qs}/Dt$, the fifth term linear in $D^s\boldsymbol{v}^{\beta s}/Dt$, the sixth term linear in $\frac{D^s}{Dt}c_1^\beta$ and the eighth term linear in the relative velocity gradients $\nabla(\boldsymbol{v}_i^\beta - \boldsymbol{v}^s)$. Therefore, by a standard argument (Coleman and Noll, 1963), we find

$$
\begin{aligned}
&\eta_0 = -\frac{\partial W}{\partial \theta} && \boldsymbol{\sigma}^{eff} = \frac{1}{J}\mathsf{F} \cdot \frac{\partial W}{\partial \mathsf{E}} \cdot \mathsf{F}^C \\
&\frac{\partial W}{\partial \boldsymbol{v}^{qs}} = \mathbf{0} && \frac{\partial W}{\partial \boldsymbol{v}^{\beta s}} = \mathbf{0} \\
&\frac{\partial W}{\partial c_1^\beta} = 0 && \boldsymbol{\sigma}_i^\beta = (\psi_i^\beta - \mu_i^\beta n_i^\beta)\mathsf{I}
\end{aligned}
\tag{2.39}
$$

leaving as inequality

$$-\frac{1}{\theta}J\boldsymbol{v}^{qs}\cdot\nabla_0\theta+J\sum_{\beta}\Big[c_1^{\beta}(\mu_2-\mu_1)+\sum_{i=1,2}(\boldsymbol{v}_i^{\beta}-\boldsymbol{v}^{s})\cdot(-\nabla\psi_i^{\beta}+\mu_i^{\beta}\nabla n_i^{\beta}-\boldsymbol{\pi}_i^{\beta})\Big]\geqslant 0 \tag{2.40}$$

Eq $(2.39)_2$ indicates that the effective stress of the mixture can be derived from a strain energy function W which represents the free energy of the mixture. Eqs $(2.39)_{3,4}$ and $(2.39)_5$ show that the strain energy function cannot depend on the heat flux, the relative velocities or the mass interactions between phases. Thus, the effective stress of a charged porous medium can be derived from a regular strain energy function, which physically has the same meaning as in single phase or biphasic media, but which can depend on both strain and solute concentrations in the phases of the medium. According to Eq $(2.39)_6$ the partial stresses of the fluid and the solutes are scalars. Transforming the relative velocities to their Lagrangian equivalents, we can rewrite (2.40)

$$-\frac{1}{\theta}\boldsymbol{v}^{qs}\cdot\nabla_0\theta+\sum_{\beta}\Big[c_1^{\beta}(\mu_2^{\beta}-\mu_1^{\beta})+\sum_{i=1,2}\boldsymbol{v}^{\beta s}\cdot(-\nabla_0\psi_i^{\beta}+\mu_i^{\beta}\nabla_0 n_i^{\beta}-\mathbf{F}^{C}\cdot\boldsymbol{\pi}_i^{\beta})\Big]\geqslant 0 \tag{2.41}$$

Assuming that the system is not too far from equilibrium, we express the dissipation (2.41) associated with relative flow of fluid, solutes and heat as a quadratic function of the relative velocities and volume interactions

$$\begin{aligned}
&-\nabla_0\psi_i^{\beta}+\mu_i^{\beta}\nabla_0 n_i^{\beta}-\mathbf{F}^{C}\cdot\boldsymbol{\pi}_i^{\beta}=\sum_{\gamma}\Big(\sum_{j=1,2}\mathbf{B}_{ij}^{\beta\gamma}\cdot\boldsymbol{v}_j^{\gamma s}+\boldsymbol{b}_i^{\beta\gamma}c_1^{\gamma}\Big)+\mathbf{B}_i^{\beta q}\cdot\boldsymbol{v}^{qs}\\
&\mu_2^{\beta}-\mu_1^{\beta}=\sum_{\gamma}\Big(\sum_{j=1,2}\boldsymbol{b}_j^{\beta\gamma}\cdot\boldsymbol{v}_j^{\gamma s}+b^{\beta\gamma}c_1^{\gamma}\Big)+\boldsymbol{b}^{\beta q}\cdot\boldsymbol{v}^{qs}\\
&-\frac{1}{\theta}\nabla_0\theta=\sum_{i=1,2}\sum_{\gamma}\Big(\sum_{j=1,2}\mathbf{B}_j^{\gamma q}\cdot\boldsymbol{v}^{\gamma s}+\boldsymbol{b}^{\gamma q}c_1^{\gamma}\Big)+\mathbf{B}^{qq}\cdot\boldsymbol{v}^{qs}
\end{aligned} \tag{2.42}$$

in which

$$\begin{bmatrix}\mathbf{B}_{ij}^{\beta\gamma} & \boldsymbol{b}_i^{\beta\gamma} & \mathbf{B}_i^{\beta q}\\ \boldsymbol{b}_j^{\beta\gamma} & b^{\beta\gamma} & \boldsymbol{b}^{\beta q}\\ \mathbf{B}_j^{q\gamma} & \boldsymbol{b}^{\gamma q} & \mathbf{B}^{qq}\end{bmatrix} \tag{2.43}$$

is a positive definite frictional matrix. Substituting Eqs (2.13) and $(2.39)_6$ into Eq $(2.42)_1$ yields

$$
\begin{aligned}
&-n_i^\beta \nabla_0 \mu_i^\beta - \mathsf{F}^C \cdot n_i^\beta \boldsymbol{b}_i^\beta = \sum_\gamma \Big(\sum_{j=1,2} \mathsf{B}_{ij}^{\beta\gamma} \cdot \boldsymbol{v}_j^{\gamma s} + \boldsymbol{b}_i^{\beta\gamma} c_1^\gamma \Big) + \mathsf{B}_i^{\beta q} \cdot \boldsymbol{v}^{qs} \\
&\mu_2^\beta - \mu_1^\beta = \sum_\gamma \Big(\sum_{j=1,2} \boldsymbol{b}_j^{\beta\gamma} \cdot \boldsymbol{v}_j^{\gamma s} + b^{\beta\gamma} c_1^\gamma \Big) + \boldsymbol{b}^{\beta q} \cdot \boldsymbol{v}^{qs} \qquad (2.44) \\
&-\frac{\nabla_0 \theta}{\theta} = \sum_\gamma \Big(\sum_{j=1,2} \mathsf{B}_j^{\gamma q} \cdot \boldsymbol{v}^{\gamma s} + \boldsymbol{b}^{\gamma q} c_1^\gamma \Big) + \mathsf{B}^{qq} \cdot \boldsymbol{v}^{qs}
\end{aligned}
$$

The dynamic boundary conditions are given by a no-jump condition of the temperature, the intra- and extrafibrillar electrochemical potential of the ions and the fluid across the boundary and the momentum balance of the boundary. The corresonding kinematic boundary conditions are the heat balance of the boundary, the intra- and extrafibrillar ion and fluid flux balance across the boudary and the no-jump condition for the solid displacement.

3. Discussion

The present derivation demonstrates that the constitutive properties of a dual porosity charged porous medium subject to large deformations is entirely contained in the strain energy function W and the frictional matrix (2.43). The definition of the function W as the Helmholtz free energy of a mixture volume equal to unity in the initial state of the solid is consistent with the classical definition of a strain energy function of a single phase material or of a saturated porous medium by Biot (1972). The swelling properties of the porous medium originate from the follows three distinct micromechanical phenomena:

1. Adsorption of fluid along the fluid-solid interface
2. Adsorption of fluid by solutes or osmosis
3. Direct solute-solute ineraction.

The first phenomenon has been formulated for saturated porous media by Biot (1972), later derived from mixture theory by Bowen (1980) and introduced into a dual porosity model by Murad and Cushman (1997). It is believed to play a primary role in low porosity media such as shales subject to high overburden pressures (Heidug and Wong, 1996) and human skin subject to high evaporation forces (van Kemenade, 1998). Adsorption is incorporated into porous media theories through the inclusion of a matric potential into the formula for the fluid chemical potential. Typically, the potential used in Darcy's equation is the sum of the pressure p and the matric potential $\partial W/\partial N_i^f$ which depends upon the fluid volume content. The relationship between the fluid content N^f and the matric potential is experimentally quantified by means of sorption experiments, involving the measurement of fluid content of samples equilibrated with a gaseous environment with known relative humidity. In terms of the strain energy function W the adsorption forces contribute as non-linear terms to the water content N^f.

The second phenomenon is significant for charged porous media, as the electrical charge of the solid attracts counterions in the fluid which in turn attracts water through osmosis. This swelling phenomenon, known as Donnan osmosis, plays an important role in the extrafibrillar compartment of cartilaginous tissues (Maroudas and Bannon, 1981) and explains almost all swelling in acrylic acid-acrylamid copolymer hydrogels (Oomens, 1994). In terms of the strain energy function W, the osmosis forces appear as mixed terms of the water content N^f and the solute content N^β, $\beta \neq f$. In the case of Donnan osmosis fixed charge density is a key quantity, which is often quantified by means of radiotracer techniques.

The third phenomenon has been hypothesised as a mayor component of swelling in cartilaginous tissues by Lai et al. (1991) who coined the term chemical expansion stress for it and attribute the effect to electrostatic interactions between the charges present within the tissue. In terms of the strain energy function W, the chemical expansion forces appear as mixed terms of strain E and charged solute content N^β and yield according to Eq $(2.39)_2$ an effective stress dependent upon the local solute concentration. The corresponding strain dependence of the electrochemical potential μ^β results from Eq (2.38) and is negligible according to Lai et al. (1991).

Frijns et al. (1997) simulated one-dimensional transient swelling and compression curves of canine annulus fibrosus samples by means of a finite element model including only the Donnan osmosis as a swelling mechanism and found no evidence that a chemical expansion stress was needed to fit the experimental data. Maroudas and Bannon (1981) found that 85% of the swelling pressure was explained by the Donnan osmosis while the remainder 15% was attributable to an entropic effect poorly dependent upon salt concentration and therefore belonging to the first category outlined above. It is beyond the scope of this paper to derive an expression for the strain energy function W that is suitable for a cartilaginous tissue. However, to give some insight into the suitability of the present approach in interpreting experimental data on intra- extrafibrillar water transport, we shall evaluate its potential to reproduce at least quantitatively the experimental facts of this transport. Estimates of the intrafibrillar water content of different connective tissues published in the literature range between 0.7 and 1.33 g water per g collagen at neutral pH and physiological saline (Katz and Li, 1972; Maroudas and Bannon, 1981; Urban and McMullin, 1985; Maroudas et al., 1991). All these authors deal with a piece of tissue in equilibrium with an external solution and without significant body forces $\boldsymbol{b}_i^\beta$ acting on them. As equilibrium is reached, all relative velocities and mass exchange terms in Eq $(2.44)_3$ vanish

$$-n_i^\beta \nabla_0 \mu_i^\beta = \mathbf{0} \qquad \mu_2^\beta - \mu_1^\beta = 0 \qquad -\frac{\nabla_0 \theta}{\theta} = \mathbf{0} \tag{3.1}$$

yielding a tissue sample where temperature and electrochemical potentials of all constituents are the same everywhere. Putting $\beta = f$ in Eq $(3.1)_2$, yields the balance of forces between on the one hand the proteoglycan network in the extrafibrillar space drawing the water towards the extrafibrillar compartment – mainly through the Donnan osmosis – and on the other hand the collagen exerting some opposite force on the water. Because there is evidence that the collagen fibrils are electroneutral (Li and Katz, 1976), this force cannot be a Donnan osmotic force or a chemical expansion stress. Therefore we infer that it should be primarily an adsorption force. This force should be sufficiently large to counteract the Donnan osmosis of the dense proteoglycan network and ensure that almost one third of the water content of the tissue remains intrafibrillar as found experimentally (Maroudas et al., 1991). As such force is generally only poorly dependent upon solute concentration, it is essentially a function of intrafibrillar water content N_1^f and we name it $f(N_1^f)$. Therefore the chemical potential of the intrafibrillar water is

$$\mu_1^f = p + \frac{\partial W}{\partial N_1^f} = p - f(N_1^f) - \pi_1 \tag{3.2}$$

in which π_1 is the osmotic pressure of the intrafibrillar solutes. Minus signs appear in front of the adsorption and osmotic terms in Eq (3.2) because they are both suction forces. The chemical potential of the extrafibrillar compartment consists of the pressure p, and an osmotic potential $-\pi_2$ containing the Donnan osmotic and the entropic contribution

$$\mu_2^f = p - \pi_2 \tag{3.3}$$

Combining Eqs (3.2) and (3.3), yields

$$f(N_1^f) = \pi_2 - \pi_1 \tag{3.4}$$

showing that intrafibrillar water content N_1^f is modulated by the difference in osmotic pressure between the extrafibrillar and intrafibrillar compartments, which is consistent with the experiment (Maroudas et al., 1991).

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Opis naładowanych i uwodnionych tkanek miękkich w postaci mieszaniny uwzględniającej wodę zawartą wewnątrz włókien kolagenu i poza nimi

Streszczenie

Objętość wody zawartej wewnątrz włókien kolagenu jest kluczowym parametrem związanym z pojęciami struktury i funkcji tkanki łącznej. Ponieważ w istniejących teoriach odkształceń skończonych tkanek chrząstkowych nie przeprowadza się rozróżnienia pomiędzy wodą zawartą wewnątrz włókien kolagenu i poza nimi, przeto w niniejszej pracy podajemy chemo-elektro-mechaniczne sformułowanie quasi-statycznych odkształceń skończonych uwzględniające takie rozróżnienia. Opracowany model opisuje ośrodek porowaty z dwoma przedziałami cieczy, w których rozpuszczona jest dowolna liczba substancji. Każdy przedział cieczy posiada własną gęstość ładunków ustalonych. Przyjęto, że odkształcenia są nieściśliwe. Założono, że każdy przedział cieczy jest lokalnie elektrycznie obojętny. Wyprowadzono równania bilansu dla każdego składnika i dla mieszaniny jako całości. Wykorzystano postać lagranżowską drugiego prawa termodynamiki dla nieściśliwych ośrodków porowatych w celu wyprowadzenia ograniczeń konstytutywnych. Wykazano, że własności materiałowe są zawarte w jednej funkcji energii odkształcenia i macierzy tensorów tarcia. Przedstawione sformułowanie jest zgodne z danymi doświadczalnymi (Maroudas i inni, 1991), w których stwierdzono, że zawartość wody wewnątrz włókien kolagenu jest regulowana przez gradient ciśnienia osmotycznego pomiędzy przedziałami wewnątrz tych włókien i poza nimi.

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