INTERACTION BETWEEN AQUEOUS SOLUTION TRANSPORT AND STRESS/STRAIN IN A DEFORMABLE POROUS MEDIUM

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Abstract   Liquid phase transport in heterogeneous media such as gels and biopolymers may induce very large strains. These produces internal mechanical stresses that interact with water transport mechanisms. We analyze transfers in porous media saturated with an ionic solution using the linear thermodynamics of irreversible processes. The interaction between mass transfer and stress/strain is analyzed using free energy. A large number of coefficients appears and we proposed theoretical and experimental method for their determination. A validation of the model is given in the simplified case of osmotic dehydration of Agar gel.

Keywords: aqueous solution, ions, coupling, Darcy coefficient, free energy, stress, strain, dehydration, Agar gel.

Introduction

When a sphere of Agar gel is placed in an aqueous solution of polyethylene glycol (PEG), a variation of its diameter can be observed during time, which reveals mass exchanges between the sphere and the solution [8]. This phenomenon depends on the PEG concentration and on the molecule size. It is possible to add to this transport mechanism, related to the presence of concentration gradients, the effect of an electric field [3]. These exchanges come along with stresses and strains that can induce cracks in the gel (fig. 1 and 2). We propose to establish a model coupling the transfer of an ionic solution and the stress/strain in an elastic, isotropic, and two-phase (solid-liquid) porous medium under isothermal conditions. Experiments will validate the model in the case of Agar gel saturated by pure water.
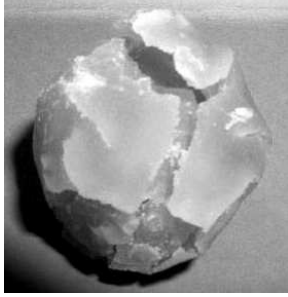
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Figure 1. View of a sphere of Agar gel after fast dehydration.

Figure 2. Appearance of a gel crack during electro-osmosis tests.

1. Electro-osmotic mass fluxes

We consider a system made of a solid phase (denoted by s) containing a liquid phase (denoted by L). The latter is composed by water (denoted by e) and by two kinds of ions (denoted by $+$ and $-$). An electric field is applied. The methods of the linear thermodynamics of irreversible processes permits the description of transport phenomena by linear relations. For the liquid phase [9] (in this paper, the indices or exponents k and m refer to cartesian coordinates):

$$v_L^k - v_s^k = -\frac{L}{T}\left(\rho_e\mu_{e,k} + \rho_+\mu_{+,k} + \rho_-\mu_{-,k} + \rho_L g z_{,k} + Z_L\phi_{,k}\right) \qquad (1)$$

where v_L^k and v_s^k are the velocities of the liquid and solid phases, respectively, L is a phenomenological coefficient, T is the temperature, ρ_i are the apparent mass densities ($i = e, +, -, L$), μ_e, μ_+ and μ_- are the chemical potentials of water, cation and anion, g is the gravity acceleration, z is the height, Z_L is the electric charge of the liquid phase per unit mass, and ϕ is the electric potential.

With the expression of the chemical potentials, equation (1) gives the Darcy's law [9]:

$$v_L^k - v_s^k = -K_w\left(\frac{P_{L,k}^*}{\rho_e^* g} + z_{,k} + \frac{Z_L\phi_{,k}}{g}\right) \qquad (2)$$

where K_w is the Darcy coefficient defined by: $K_w = Lg/\rho_L T$, P_L^* is the pressure of the liquid phase at the pores scale and ρ_e^* is the real mass density of water.

2.   Coupling with the solid phase mechanics

The method used here [2, 10, 4, 6] consists of assuming the existence of a free energy function Φ that depends on state variables of the medium. With the above assumptions, the chosen state variables are w, the water content of the medium, ε^{km}, the strain of the skeleton, n_+ and n_-, the numbers of cations and anions per unit mass of the solid:

$$\Phi = \Phi\left(w, \varepsilon^{km}, n_+, n_-\right) \tag{3}$$

The second order development near a reference state (referred to by exponent 0) leads to the following state relations:

$$\mu_e = \mu_e^0 + \frac{1}{\rho_s}\left(d_e\Delta w - 3K\beta\delta^{km}\Delta\varepsilon^{km} - d_+^e\Delta n_+ - d_-^e\Delta n_-\right) \tag{4}$$

$$\mu_+ = \mu_+^0 + \frac{1}{\rho_s M_+}\left(-d_+^e\Delta w + d_+^\varepsilon\delta^{km}\Delta\varepsilon^{km} + d_+\Delta n_+ - d_-^+\Delta n_-\right) \tag{5}$$

$$\mu_- = \mu_-^0 + \frac{1}{\rho_s M_-}\left(-d_-^e\Delta w + d_-^\varepsilon\delta^{km}\Delta\varepsilon^{km} - d_-^+\Delta n_+ + d_-\Delta n_-\right) \tag{6}$$

$$\sigma^{km} = \sigma^{km0} - 3K\beta\Delta w\delta^{km} + \lambda\Delta\varepsilon^{jj}\delta^{km} + 2\mu\Delta\varepsilon^{km} + d_+^\varepsilon\Delta n_+\delta^{km}$$
$$+ d_-^\varepsilon\Delta n_-\delta^{km} \tag{7}$$

in which appear the molar mass of the cation M_+ and anion M_-, the unit tensor δ^{km}, the coefficient of compressibility K, the Lamé coefficients λ and μ, and nine partial derivatives of free energy (d_e, β, d_+^e, d_-^e, d_+, d_-, d_+^ε, d_-^ε and d_-^+). Introducing (4), (5) and (6) in (1) gives an new expression of the liquid phase transport:

$$v_L^k - v_s^k = -D_w w_{,k} + D_\varepsilon\left(tr\varepsilon\right)_{,k} - D_+ n_{+,k} - D_- n_{-,k} - \frac{L\rho_L g}{T}z_{,k} - \frac{L\rho_L Z_L}{T}\phi_{,k} \tag{8}$$

where D_w, D_ε, D_+ and D_- can be expressed using K_w and coefficients appearing in (4), (5), (6) and (7). This equation provides the coupling between the transport phenomena and the mechanics of the medium.

Although the equation (7) restricts the consideration to small strains, it is possible to consider large strains by moving the state reference (w^0, ε^{km0}, n_+^0, n_-^0) and by updating the coefficients, which depend all on state variables.

In the general case, the model depends on two mechanical coefficients (the Young's modulus E and the Poisson's ratio ν), one transport coefficient (L or K_w) and the nine coefficients defined in the equations (4) to (7). Coefficients d_+, d_-, d_+^e, d_-^e, d_+^ε, and d_-^ε were determined in the case of an ideal solution [8].

3. Experimental determination of some coefficients - Case of pure water in Agar gel

In order to analyze coefficients D_w, D_ε, d_e, and β, the liquid phase is assumed to be pure water. In this case, the chemical potentiel of water is given by: $\mu_e = \mu_e^0 + P_L^*/\rho_e^*$. The relation (4) takes the shape proposed by Biot [1]:

$$\Delta P_L^* = \frac{\rho_e^*}{\rho_s} \left(d_e \Delta w - 3K\beta\delta^{km}\Delta\varepsilon^{km} \right) \qquad (9)$$

The two coefficients d_e and β, which can be related to the two physical coefficients introduced by Biot in the theory of consolidation [1], are defined by:

$$d_e = \rho_s \left(\frac{\partial\mu_e}{\partial w} \right) \qquad ; \qquad 3K\beta = -\frac{\partial\left(tr\sigma\right)}{\partial w} \qquad (10)$$

For Agar gel, experimental study of these coefficients were performed [7]. The coefficient E was determined by compressive tests. Ultrasonic measurements of the Poisson's ratio ν showed that it is almost equal to 0.5. Compressibility K (fig. 3) is deduced from $K = E/(3(1 - 2\nu))$.

Agar gel is a hygroscopic medium for $w < 60\%$ [7]. In this range, the equation (10) allows the determination of d_e from the chemical potential of water given by the desorption isotherm. Assuming that the two-phase structure of the medium is conserved, the coefficient β can be expressed by: $\beta = 1/3\left(\alpha + w\right)$, where α is the ratio between the specific mass of the water and the specific mass of the solid. This expression is confirmed by experimental tests (fig. 4). The coefficient K_w (fig. 5) was measured for a plate of gel placed in a PEG solution [6]. D_w and D_ε (fig. 6) are deduced from:

$$D_w = \frac{K_w d_e w}{g} \qquad ; \qquad D_\varepsilon = \frac{K_w 3K\beta w}{g} \qquad (11)$$

4. Experimental validation

Associated with the mass balance of water and the mechanical equilibrium equation, relations (7) and (8) give a model of solution transport in a deformable porous medium. The result of an incremental numerical method was compared to experiments accomplished for the case of a sphere with an initial radius of 2 cm placed in a PEG solution. Figure 7 gives an example of the comparison between experimental and numerical water content profiles. The model permits the analysis of the evolution of stress profiles. Figure 8 shows the evolution of the circumferential
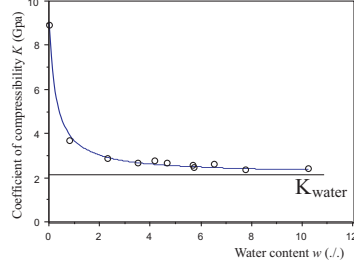
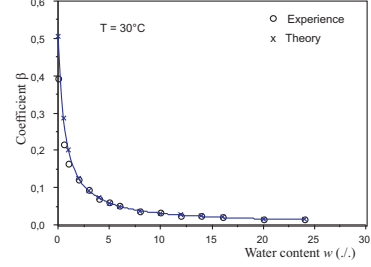*Figure 3.* Variation of the coefficient K with water content.

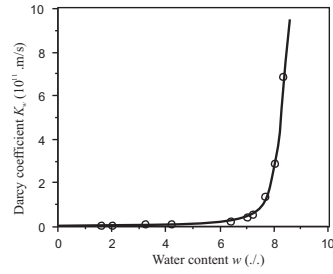*Figure 4.* Variation of the coefficient β with water content.

Figure 5. Variation of Darcy coefficient K_w with water content.
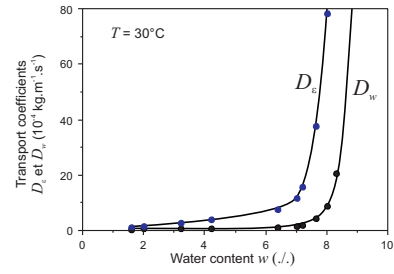

Figure 6. Variation of transport coefficients D_w and D_ε with water content.

stress at different times in the case of the sphere. Figure 9 represents the final experimental and theoretical deformations of a cylinder. There is a good agreement between experience and theory.
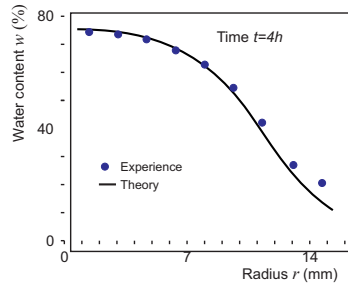

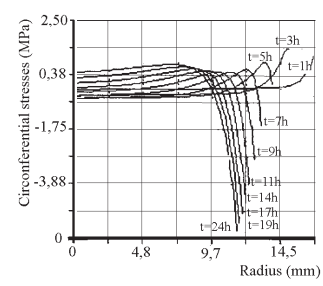*Figure 7.* Experimental an theoretical water content profiles in the sphere.

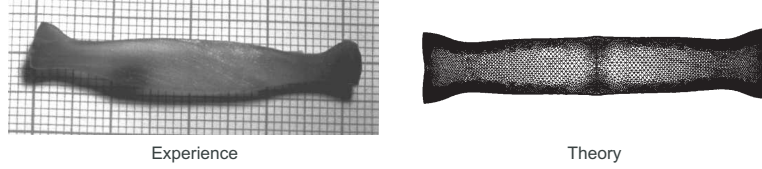*Figure 8.* Circumferential stresses in the sphere at different times.

Figure 9. Experimental and theoretical deformations of a cylinder after dehydration.

Conclusion

In this paper, we give a model coupling mass transfer and stress/strain in an elastic porous medium whose pores are filled by an ionic solution. In this model, the chemical potential and mechanical behaviour laws are developed from a free energy potential. A large number of coefficients appears and we proposed theoretical and experimental method for their determination. In the simple case of pure water, we have shown that it is possible to analyze the coefficients of the model in Agar gel. A good agreement between the experiments and the numerical model was found in the case of dehydration of a sphere. This suggests to extend the analysis on the effect of ionic species on the coefficients of the model and the simulation of electro-osmosis in deformable media.

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