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Strains and Stresses Induced by Water Transport in Hygroscopic Elastic Media

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Abstract. We propose to analyse the phenomenological equation of water transport in a biconstituent medium with stresses. The work is limited to the case of elastic, isotropic behaviour and an isothermal transfer process.

1 Introduction

Water transport in certain heterogeneous media such as gels and biopolymers may be accompanied by considerable deformation causing internal mechanical stresses which interact with water transport mechanisms. In previous work (Auria R and Bénét JC, 1990) on transfer of water in natural rubber, the pattern of variation of the transport coefficient with the water content remained unexplained; the hypothesis of interaction between transfer and deformation was mentioned. It has been shown that internal strain occurring during transfer affect the expression of local diffusion flux (Larché FC and Cahn JW, 1982). Following these publications, we propose to analyse the phenomenological equation of water transport in a biconstituent medium with stresses. The work is limited to the case of elastic, isotropic behaviour and an isothermal transfer process.

2 Phenomenological transport equation

2.1 General equation, elastic case

The thermodynamics of irreversible linear processes gives the following transport equation in a biconstituent medium during an isothermal process in which evolution is assumed to be quasi-static (Mrani I, 1993) :

$$\mathbf{J} = \rho_e(\mathbf{u} - \boldsymbol{\vartheta}) = -\frac{L}{T} \mathbf{grad} \mu_e \quad (1)$$

Gravity and accelerations have been ignored in this equation. \mathbf{J} is the transport flux defined from velocities of water \mathbf{u} and the solid component $\boldsymbol{\vartheta}$, ρ_e is the apparent bulk mass density of water, L is the phenomenological

transport coefficient, T is the temperature and μ_e is the chemical mass potential of the liquid.

The state variables used to describe the local state of the system are water content (w) and the strain tensor (ε_{ij}). In the neighbourhood of a reference state ($w^\circ, \varepsilon_{ij}$), the increment of the chemical potential of water μ_e is related to the infinitesimal increments in state variables (Coussy O, 1993) by the equation:

$$\rho_s \Delta \mu_e = d_w \Delta w - 3K \beta \Delta \text{tr} \varepsilon \quad (2)$$

where ρ_s is the apparent bulk mass density of the solid phase. d_w is defined by $d_w = \rho_s (\partial \mu_e / \partial w)$ with constant deformation. K is the coefficient of compressibility: $3K = E / (1 - 2\nu)$, where E is Young's modulus and ν is Poisson's ratio. β is the hygrometric expansion coefficient; $\text{tr} \varepsilon$ is the trace of the strain tensor.

Within the framework of infinitesimal changes around the reference state, the elastic mechanical behaviour of the medium accounts for the incremental variation in stress tensor σ_{ij} according to increments in state variables (Coussy O, 1991) :

$$\Delta \sigma_{ij} = \lambda \Delta \text{tr} \varepsilon \delta_{ij} + 2\mu \Delta \varepsilon_{ij} - 3K \beta \Delta w \delta_{ij} \quad (3)$$

where δ_{ij} is the unit tensor and λ and μ are Lamé's coefficient. According to (3), coefficient β is defined by: $3K \beta = -(\partial \sigma_{ij} / \partial w) \delta_{ij}$, with constant deformation.

Using (2), transport equation (1) becomes:

$$J = -D_w \mathbf{grad} w + D_\varepsilon \mathbf{grad} \text{tr} \varepsilon \quad (4)$$

Transport coefficients D_w and D_ε are related to the phenomenological coefficient L by the following equations:

$$D_w = L \frac{dw}{T \rho_s}; \quad D_\varepsilon = L \frac{3K \beta}{T \rho_s} \quad (5)$$

2.2 Transport equation for a plate

In general, equation (4) is difficult to analyse experimentally. In the case of a plate, the increment of the trace of the stress tensor depends solely on the water content increment (Mrani I. 1993):

$$\Delta \mu_e = \frac{1}{\rho_s} [d_w^* + \frac{2E\beta^2}{1-\nu}] \Delta w \quad (6)$$

According to (6), equation (1) becomes:

$$J = -D^* \frac{\partial w}{\partial x} \quad (7)$$

This equation is similar to Fick's law. Apparent transport coefficient D^* depends on the hygroscopic and mechanical parameters of the material:

$$D^* = \frac{L}{T\rho_s} \left[d_w^* + \frac{2E\beta^2}{1-\nu} \right] \quad (8)$$

With :

$$d_w^* = d_w - 9K\beta^2 \quad (9)$$

3 Experimental study of the phenomenological equation

3.1 Experimental verification

Validation of transport law (7) and deduction of transport coefficient values D^* are proposed. The experimental study concerned water transport in agar gel plates initially 15 mm thick and with a uniform water content of 10. The samples were placed in a polyethylene glycol (PEG) solution with controlled temperature (30°C). The difference in concentration at the interface between the sample and the solution caused movement of water from the material to the solution, accompanied by shrinkage of the material. Transfer was one-directional along axis ox perpendicular to the facets of the agar gel plate. Experimental conditions are those of the conditions of application of equation (7).

Samples were taken at intervals of time and cut into fine slices 0.6 mm thick at right angles to the transfer direction. Water contents were measured in the slices. This makes it possible to draw water content profiles against time and to deduce the flux J and the water content gradient.

Figure 1 shows the variation in transport flux according to water content gradient for different values of w . This figure shows that the transport flux can be considered to be proportional to the water content gradient. This validates equation (7) in the water content range studied.

3.2 Apparent transport coefficient D^*

Apparent transport coefficients were calculated using the slopes of the lines in Fig 1. Figure 2 shows the variation in coefficient D^* according to the water

content for an interval $1.5 < w < 8$. It can be seen that the coefficient depends strongly on water content. At $w > 6$, the transport coefficient fell rapidly to a minimum of about $w = 6$. Similar movements of coefficient D^* to those in Fig 2 were observed in the case of biphasic media (Auria R, 1988, Do Amaral S.P.J, 1992, Gehrmann D and Kast W, 1978) and triphasic media (Constock GL, 1963, Crausse P et al., 1984, Ketelaars AAJ, 1992)

In triphasic media, the rise in transport coefficient can be attributed to the preponderance of transfers in the gas phase with low water contents. This explanation cannot be used here because of the biphasic nature of agar gel. Interpretation based on the equation for coefficient D^* (8) is proposed here. This requires analysis of the term related to contraction of the medium: $2E\beta^2/(1 - \nu)$.

The Young's modulus E of agar gel was determined by one-directional compression tests (Mrani I, 1993). Its variation according to water content reveals considerable rigidification of the material at below $w=400\%$. Ultrasonic measurements of Poisson's ratio ν (Mrani I, 1993) show that this coefficient is practically constant at 0.5 in the water content range investigated. In the hypothesis of a biphasic medium, coefficient β can be expressed as follows (Mrani I, 1993): $\beta = 1/3(\alpha + w)$; the coefficient increases when w decreases. In the case of agar gel, these variations cause very strong increase in the term $2E\beta^2/(1 - \nu)$ at low water contents, which may account for the increase in coefficient D^* in Figure2.

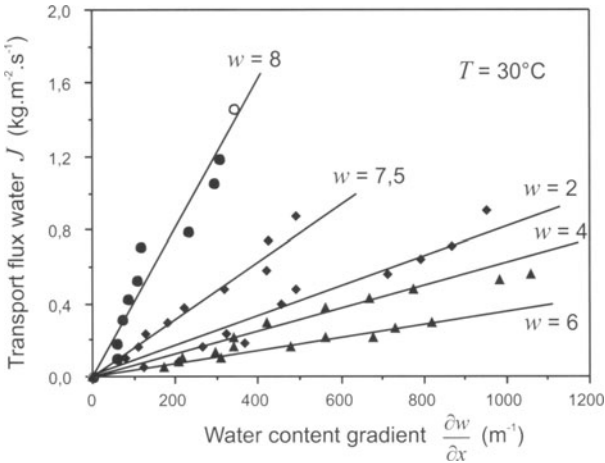


Fig.1. Variation of water transport flux coefficient according to water content gradient for different values of w

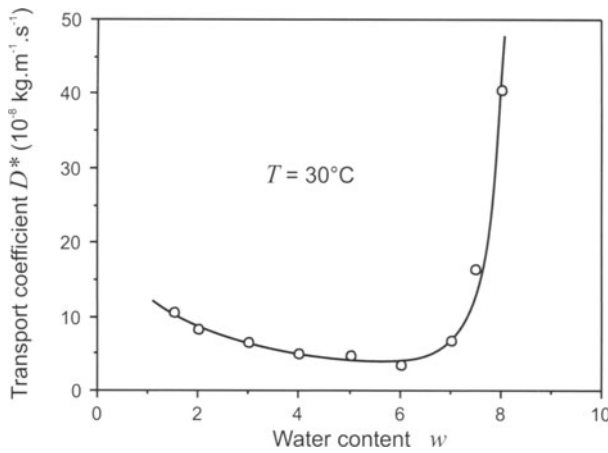


Fig. 2. Variation of transport coefficient of water in agar gel

3.3 Identification of intrinsic transport coefficients

Unlike the transport coefficient D^* defined in the case of a plate, coefficients D_w and D_ε are intrinsic to the material. It is thus necessary to determine these coefficients to describe transfers for any geometrical case.

Using expressions (5) and (8), D_w and D_ε are given by:

$$D_w = \frac{3K\beta}{d_w^* + \frac{2E\beta^2}{1-\nu}} D^* \quad ; \quad D_\varepsilon = \frac{d_w^* + 9K\beta^2}{d_w^* + \frac{2E\beta^2}{1-\nu}} D^* \quad (10)$$

Coefficient d_w^* is determined using the desorption isotherm (Mrani I, 1993) in the case of agar gel. Variation of D_w and D_ε according to water content are shown in Figure 3. This figure shows that unlike the transport coefficient D^* , coefficients D_w and D_ε display monotonic decreasing on the water content and approach zero when the material is dry. Rise in coefficient D^* (Fig 2) with $w < 6$ did not affect the rate of variation of coefficients D_w et D_ε , given the intrinsic nature of the two coefficients.

4 Numerical simulation of the dehydration of a sphere and a cylinder of agar gel

A sphere and a cylinder of gel are placed in a solution of polyethylen glycol (PEG). In these conditions, the water is eliminated of the gel by effet of concentration difference between the sample and the solution. The flux of water imposed as condition to the limit has been calculated from the dehydration kinetic. A numerical model gives water content and stresses fields and deformation of the sphere and the cylinder.

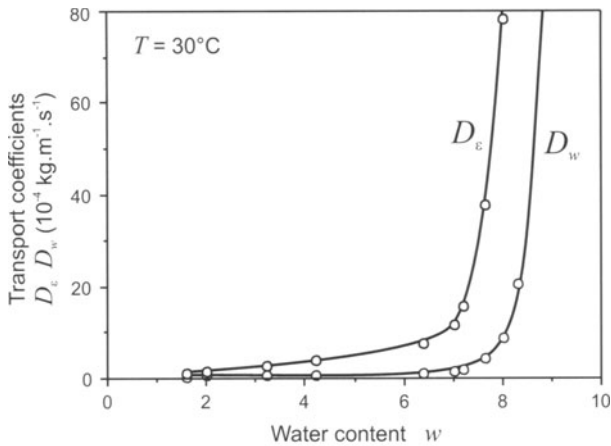


Fig. 3. Variation of transport coefficients D_w and D_ϵ in agar gel according to water content

Figure 4 gives an exemple of comparison between experimental and numerical water content profiles in the case of a sphere presenting initially a radius of 2 cm. The model make it possible to analyse the evolution of stresses profiles; figure 5 shows the evolution of the circunferential stress at different times in the case of the sphere. Two phases can be seen is the figure. At the begenning of the dehydration ($t < 5\text{h}$) the circunferential stress was positive at the surface of the sphere (traction) and negative in the material (compression). If the dehydration is to fast, this may cause cracks at the surface. In a second phase ($t > 7\text{h}$) the circunferential stress become mainly in compression at the surface and traction in the centre. Figure 6 represents the final deformation of the cylinder. There is a good agreement between experience and theory

5 Conclusion

The study shows the influence of mechanical actions on diffusion fluxes. Transfer is not governed solely by water content gradient but also by the deformation gradient. It has been shown that in an infinite plate with elastic, isotropic behaviour, transport is governed by an equation similar to Fick's law: water flux is proportional to the water content gradient. In this case, the transport coefficient expression contains a part related to the hygroscopic feature of the medium and a part related to its rheology.

Experimental study of water content profiles in an agar gel plate justified the use of a transport law of the Fick type for the water content range investigated. The apparent transport coefficient decreases with the water content and reaches a minimum at $w = 6$ and then increases with low water contents. The theoretical expression proposed makes it possible to attribute the

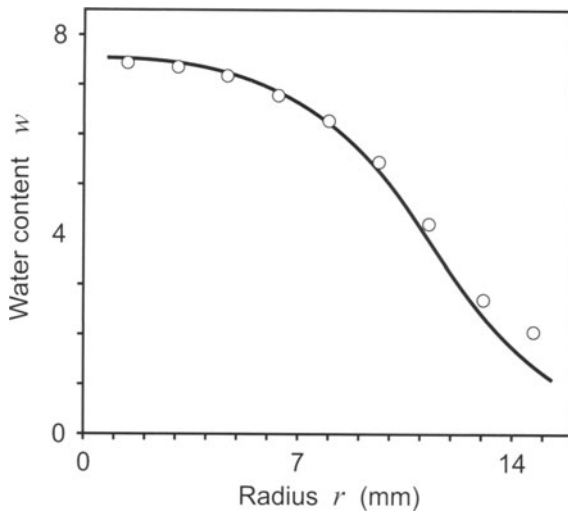


Fig. 4. Theoretical and experimental water content profiles in the sphere at $t = 4h$

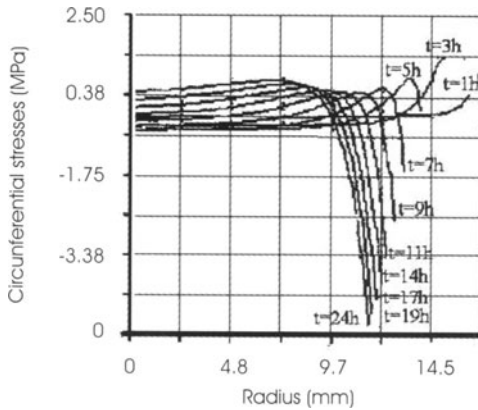
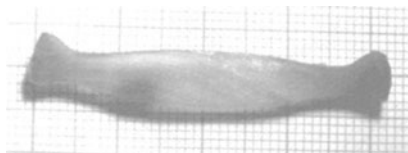


Fig. 5. Circumferential stresses in the sphere at different times

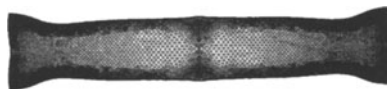
increase in apparent transport coefficient to variations in elastic and hygro-metric coefficients.

Use of the variation of the apparent transport coefficient enabled determination of the two coefficients related to the water content and deformation gradients. These coefficients are intrinsic to the material and make it possible to describe water transport in agar gel for any geometrical layout.

The numerical results show good agreement between experience and theory in the case of dehydration of a sphere and a cylinder concerning water content profiles, stresses and deformation.



Experience



Theory

Fig. 6. Comparison of experimental and theoretical deformation of a cylinder at the end of dehydration

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