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Gel mechanics: A comparison of the theories of Biot and Tanaka, Hocker, and Benedek

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This article compares the Biot [J. Appl. Phys. **12**, 155 (1941)] and Tanaka, Hocker, and Benedek (THB) [J. Chem. Phys. **59**, 5151 (1973)] theories of gel deformation. Biot's theory treats the gel as a continuum with the pore pressure as a state variable whereas the THB theory treats the gel as a mixture of solid and liquid phases. We revisit the problem of gel deswelling and use this example to show that there can be substantial differences between the two theories. The THB theory is not a complete mixture theory since the displacements of the liquid are assumed to be negligibly small in comparison with the displacements of the network. We propose a simple extension of the THB model, which takes into account the momentum transfer of the liquid phase. We show that with this simple addition and some very reasonable assumptions, the extended THB theory is identical to the Biot theory. © 2005 American Institute of Physics.
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I. INTRODUCTION

There are essentially two approaches to study the three-dimensional deformation of elastic gels. The first is due to Scherer¹ who treated the gel as a continuum (as opposed to a mixture of solid and liquid phases) with the pore pressure as a state variable. The model proposed by Scherer is actually based on a theory proposed by Biot² to study soil consolidation. Using this model, Scherer has provided insights on a wide range of phenomenon related to the deformation of gels, including drying, swelling/deswelling, and cracking.^{3,4} The second approach is due to Tanaka, Hocker, and Benedek,⁵ henceforth abbreviated as THB. In this theory, the gel is treated as a continuum mixture of a solid and liquid. In reality, the THB theory is not a complete mixture theory since the displacements of the fluid are assumed to be negligibly small in comparison with the displacements of the network. The use of mixture theory is not new, for example, Truesdell and Toupin⁶ had developed a general theory for the mechanics of a mixture of several constituents but THB seemed to be unaware of this development and Biot's earlier work.

Although many investigators have used the Biot and THB theories to study gel mechanics for a long time,^{7–10} no attempt has been made to contrast/unify these two approaches with the exception of Johnson.¹¹ Johnson¹¹ analyzed wave propagation in an infinite elastic gel in detail using Biot's model^{12,13} and showed that THB's displacement fields are in agreement with his analysis in the limit of low polymer density. His comparison was focused on different modes of wave propagation and displacement fields in infinite domains. In many gel problems, the deformations are quasistatic and since the specimens are finite, boundary conditions play a significant role in determining the displace-

ments and the stresses. The aim of this work is to compare and contrast the Biot and THB theories. We show that there can be considerable differences between the two theories, even in very simple boundary-value problems. We propose a simple extension of the THB model, which takes into account the momentum transfer of the liquid phase. We show that with this simple addition and some very reasonable assumptions, the extended THB theory is identical to the Biot theory.

The outline of this paper is as follows. We first summarize the Biot approach in Sec. II. The THB theory is summarized in Sec. III. Section IV contrasts these two approaches by comparing the exact solutions of two gel deswelling experiments. We discuss the extensions to the THB approach in Sec. V and show that it becomes identical to the Biot approach by supplementing it with the momentum balance for the gel liquid. Section VI summarizes our results.

II. BIOT APPROACH

We briefly summarize the Biot formulation.^{2,14,15} In his original work, the gel was treated as a porous continuum with the pore pressure as a state variable.² Subsequently, Biot adopted a mixture theory approach, where the solid network and the gel liquid are treated separately.¹⁴ He then showed that these two theories are entirely equivalent. We mention the mixture theory of Biot since later we will extend the THB theory to the Biot theory, and it is easier to establish the equivalence between the extended THB theory and Biot's mixture theory.¹⁴ Table I summarizes these two different versions of the Biot theory, along with a list of the variables involved and their physical interpretation. In both the THB and Biot theories, because of the much higher compressibility of the network, the solid and liquid phases are considered incompressible. This assumption will be assumed throughout this work.

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TABLE I. Difference between Biot's original formulation (Ref. 2) and mixture theory (Ref. 14).

	Biot ^a	Biot ^b , mixture theory
Network or gel displacement	u_i	u_i
Fluid displacement	Not applicable	v_i
Strain-displacement relationship	$\varepsilon_{ij} = \frac{1}{2}(u_{i,j} + u_{j,i})$	$\varepsilon_{ij} = \frac{1}{2}(u_{i,j} + u_{j,i})$
Stresses	σ_{ij} (Force per unit gel area)	$\sigma_{ij} = \sigma_{ij}^s$ (Network stress) + $p\delta_{ij}$ (pore pressure)
Constitutive law	$\sigma_{ij} = 2G\varepsilon_{ij} + \frac{2G\nu\varepsilon}{1-2\nu}\delta_{ij} + p\delta_{ij}$	$\sigma_{ij}^s = 2G\varepsilon_{ij} + \frac{2G\nu\varepsilon}{1-2\nu}\delta_{ij}$
Equilibrium equations	$\sigma_{ij,j} = 0$	$\sigma_{ij,j} = 0$
Darcy's law	$J_i = D_p p_{,i} / \eta$	$u_{i,t} - v_{i,t} = D_p p_{,i} / \eta$

^aReference 2.^bReference 14.

In the absence of body forces and ignoring inertia, the stresses σ_{ij} in the gel satisfy the equilibrium equation,

$$\sigma_{ij,j} = 0. \quad (1)$$

Throughout this work, we use summation convention where repeated indices are summed and a comma denotes partial differentiation. In the original theory of Biot, the stresses are the forces acting on a unit area of gel. In the mixture theory, the stress is decomposed into two parts: one caused by the hydrostatic pressure of water in pores and the other caused by the average stress of the solid network. In Biot's theory, the shear stresses in the liquid are assumed to be zero. For a saturated isotropic elastic gel, the strains ε_{ij} and the stresses are related by

$$\sigma_{ij} = 2G\varepsilon_{ij} + \frac{2G\nu\varepsilon}{1-2\nu}\delta_{ij} + p\delta_{ij}, \quad (2)$$

where p is the pore pressure, G and ν denote the shear modulus and Poisson's ratio of the drained network, and $\varepsilon = \varepsilon_{ii}$ is the volumetric strain. In Eq. (2), we have used the convention where a positive pore pressure corresponds to hydrostatic tension.¹⁶ Substituting the constitutive law (2) and the usual small-strain strain-displacement relationship, $\varepsilon_{ij} = \frac{1}{2}(u_{i,j} + u_{j,i})$ in the equilibrium equation (1) results in

$$G\nabla^2 u_i + \frac{G}{1-2\nu}\varepsilon_{,i} + p_{,i} = 0, \quad (3)$$

where u_i are the gel displacements. The pore pressure gradient is related to the flux of the gel liquid J_i (volume of gel liquid flowing per second per unit area), by Darcy's law,

$$J_i = D_p p_{,i} / \eta, \quad (4)$$

where D_p is the permeability of the network¹⁷ and η is the viscosity of the gel liquid. Using Darcy's law and mass balance for the pore liquid, Biot² showed

$$D_c \nabla^2 \varepsilon = \frac{\partial \varepsilon}{\partial t}, \quad (5)$$

where

$$D_c = E_L D_p / \eta, \quad E_L = \frac{2G(1-\nu)}{(1-2\nu)}.$$

Here E_L is the longitudinal elastic modulus, obtained, for example, from an experiment where the gel is compressed uniaxially while maintaining a constant cross-sectional area. Note that for a homogeneous gel, D_c is the cooperative diffusion coefficient that is measured in a light-scattering experiment. Equations (3) and (5), together with the appropriate initial and boundary conditions, completely determine the quasistatic deformation of an elastic gel.

III. THB APPROACH

In THB theory, the displacement of the solvent is assumed to be zero, so that the equilibrium equation for a gel with no body force and negligible inertia is

$$\sigma_{ij,j} - f u_{i,t} = 0, \quad (6)$$

where t denotes time, u_i are the displacements of the network (also that of the solid gel), f is the friction constant, and σ_{ij} are the network stresses. The second term in (6) is supposed to model the drag force caused by the motion of the network through the pore liquid. The stress σ_{ij} is related to the network strains via (2) with $p=0$. Combining the constitutive law (2) with $p=0$, the strain-displacement relationship and the equilibrium equation (6) result in

$$G\nabla^2 u_i + \frac{G}{1-2\nu}\varepsilon_{,i} - f u_{i,t} = 0. \quad (7)$$

The differences in the two approaches are quite obvious. The THB theory does not consider fluid flow explicitly and hence cannot enforce boundary conditions involving flow. It is to be noted here that THB do explicitly state that they are concerned only with the structural fluctuations of the solid network rather than the gel liquid. The question is how strongly fluid transport in gels affects network deformation. Geissler and Hecht,⁷ henceforth abbreviated as GH, attempted to extend the THB theory to model their gel deswelling experiments. In their approach, an additional term was added to take solvent displacement into account. However, their formulation has many deficiencies and a detailed critic of their approach is given in Appendix A.

IV. GEL DESWELLING EXPERIMENTS: COMPARISON OF THE BIOT AND THB THEORIES

To contrast these two approaches, we analyze these two models in the context of a gel deswelling experiment carried out by GH. Figure 1 shows a schematic of the setup used by GH. The gel is enclosed in a rigid cylinder with frictionless walls. The length of the cylinder is much larger than its radius. The perforated piston (which allows liquid flow) is in contact with the gel at time $t=0$ and a known compressive load P is applied. Since the cylinder is rigid, the only non-vanishing displacement is in the Y direction. We first solve this problem using the Biot formulation, followed by the THB approach.

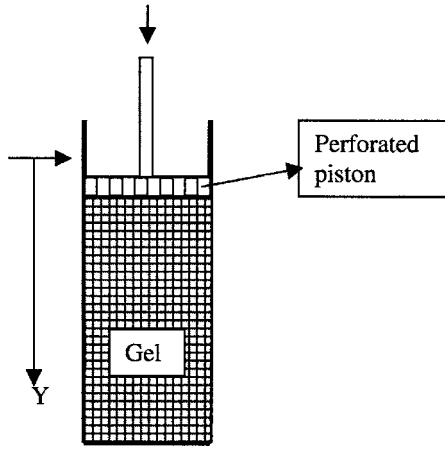


FIG. 1. Schematic of the experimental setup used to measure gel permeability. A compressive load P is applied through the perforated piston on top of the gel at $t > 0$.

A. Case 1: Biot approach

Since the piston is permeable, solvent can flow so that the pore pressure p is zero at $y=0$ (for $t > 0$). On the other hand, at infinity, the pore pressure p and the network stress are the same and equal $-P/A$, where A is the cross-sectional area of the cylinder. Following Scherer,¹ the governing equation for the pore pressure is

$$\frac{\partial p}{\partial t} = D_c \frac{\partial^2 p}{\partial y^2}. \quad (8)$$

The initial and boundary conditions are

$$p(y > 0, t = 0^+) = -P/A, \quad (9a)$$

$$p(y = 0, t > 0) = 0, \quad (9b)$$

$$p(y \rightarrow \infty, t > 0) = -P/A. \quad (9c)$$

Equation (8) with the conditions (9) has an exact closed-form solution. Once we obtain the pore pressure, it is straightforward to find the displacement and stresses in the gel. The solution of (8) subjected to (9) can be obtained using Laplace transforms and is

$$p = -\frac{P}{A} \operatorname{erfc}\left(\frac{y}{2\sqrt{D_c t}}\right). \quad (10)$$

The strain is given by

$$\varepsilon = \varepsilon_{22} = \left(-\frac{P}{A} - p\right)/E_L = -\frac{P}{AE_L} \operatorname{erfc}\left(\frac{y}{2\sqrt{D_c t}}\right), \quad (11)$$

B. Case 2: THB approach

Using the THB approach, the governing equation is

$$\frac{\partial u_2}{\partial t} = D_c \frac{\partial^2 u_2}{\partial y^2}, \quad (12)$$

with $D_c = E_L/f$. The boundary conditions are

$$u_2(y = \infty, t) = 0, \quad (13a)$$

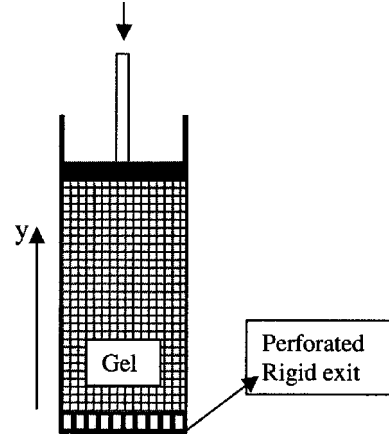


FIG. 2. Gel in a rigid cylinder pushed by a piston at one end and with a permeable outlet at the other end.

$$\sigma_{22}(y = 0, t) = E_L \frac{\partial u_2}{\partial y} = PH(t)/A, \quad (13b)$$

where $H(t)$ is the Heaviside function. With the initial condition assumed to be

$$u_2(y, t = 0) = 0, \quad (13c)$$

Eq. (12) with the conditions (13) can be solved in a closed form. Using the Laplace transforms, it can be shown that the gel displacement is given by

$$u_2 = -\frac{P}{E_L A} \left[2\sqrt{D_c t / \pi} e^{-y^2/4D_c t} - y \operatorname{erfc}\left(\frac{y}{2\sqrt{D_c t}}\right) \right]. \quad (14)$$

The dilation strain ε which is the same as ε_{22} , is

$$\varepsilon = u_{2,y} = -\frac{P}{AE_L} \operatorname{erfc}\left(\frac{y}{2\sqrt{D_c t}}\right). \quad (15)$$

It is interesting to compare the predictions of Biot and THB for this simple one-dimensional problem [see (11) and (15)]. The strains are identical but the stresses in the gel are different. In Biot's theory, the stress σ_{22} is uniform and equals the applied load divided by the area; whereas in THB, the stress is a function of position $\sigma_{22} = E_L \varepsilon = -(P/A) \operatorname{erfc}(y/2\sqrt{D_c t})$. Thus, the stress tensor in the THB model must not be interpreted as the stress in the gel, but as the stress acting on the network. In this simple problem, the stress σ_{22} acting on the gel is known and equals $-P/A$, so the network stress according to the Biot theory should be $-(P/A) - p$, which is indeed the network stress predicted by the THB theory.

This example shows that there are situations where the predictions of both theories are identical, provided that the THB stress tensor is interpreted as the network stress. In problems involving complex geometries, the THB formulation cannot determine the actual stress in the gel, since the pore pressure cannot be determined.

In the above example, the displacement and strain were the same in both theories. This is not always true, as illustrated by the following example. To illustrate this, consider a slightly different experimental setup (similar to the setup used by Scherer¹). A gel layer of thickness h is placed inside a rigid container with frictionless walls, as shown in Fig. 2.

One end of the container has an outlet that is permeable. At the other end, the gel is pushed downward with a frictionless rigid piston with velocity V . The initial and boundary conditions for the two formulations are

$$\left. \begin{aligned} u_2(y=h, t>0) &= -Vt \\ u_2(y=0, t>0) &= 0 \\ u_2(y, t=0) &= 0 \end{aligned} \right\} \quad (\text{THB}), \quad (16)$$

$$\left. \begin{aligned} u_2(y=h, t>0) &= -Vt \\ p(y=0, t>0) &= 0 \\ p_y(y=h, t>0) &= 0 \\ p(y, t=0) &= 0 \\ u_2(y=0, t>0) &= 0 \\ u_2(y, t=0) &= 0 \end{aligned} \right\} \quad (\text{Biot}). \quad (17)$$

It can be shown that the gel displacement for the THB and Biot formulations are given by (see Appendix B)

$$\begin{aligned} u_{2\text{THB}}(y, t) = & -V \left[\frac{y}{h} \left(t + \frac{y^2 - h^2}{6D_c} \right) \right. \\ & + 2 \frac{h^2}{D_c} \sum_{n=1}^{\infty} (-1)^{n+1} \frac{e^{-(n^2 \pi^2)/(h^2/D_c)t}}{n^3 \pi^3} \\ & \left. \times \sin\left(\frac{n \pi y}{h}\right) \right], \end{aligned} \quad (18a)$$

$$u_{2\text{BS}}(y, t) = -Vt - u_{2\text{THB}}(h - y, t) \quad (18b)$$

so that the displacements are not the same. Also, the strains are different and are related by

$$\varepsilon_{\text{BS}}(y, t) = \varepsilon_{\text{THB}}(h - y, t). \quad (19)$$

On normalizing the displacements, we obtain

$$\begin{aligned} u_{2\text{THB}}^*(\xi, t^*) = & - \left[\xi \left(t^* + \frac{\xi^2 - 1}{6} \right) \right. \\ & + 2 \sum_{n=1}^{\infty} (-1)^{n+1} \frac{e^{-n^2 \pi^2 t^*}}{n^2 \pi^3} \sin(n \pi \xi) \left. \right], \end{aligned} \quad (20a)$$

$$\begin{aligned} u_{2\text{BS}}^*(\xi, t^*) = & -t^* + \left[(1 - \xi) \left(t^* + \frac{\xi^2 - 2\xi}{6} \right) \right. \\ & + 2 \sum_{n=1}^{\infty} (-1)^{n+1} \frac{e^{-n^2 \pi^2 t^*}}{n^3 \pi^3} \sin(n \pi (1 - \xi)) \left. \right], \end{aligned} \quad (20b)$$

where

$$u_2^* = \frac{u_2}{Vh^2/D_c}, \quad t^* = \frac{t}{h^2/D_c}, \quad \xi = \frac{y}{h} \quad (20c)$$

Figure 3 plots the normalized displacements as a function of normalized position for the Biot and THB approaches. Clearly there is a difference between the two approaches and this simple example shows the need for taking into account

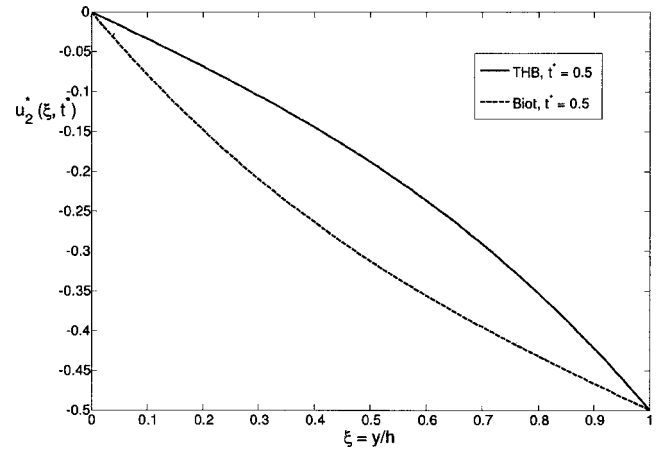


FIG. 3. Plot of normalized displacement of network vs normalized gel position for the Biot and THB theories.

the coupling between the solid network deformation and fluid transport.

V. EXTENDED THB THEORY

As pointed out previously,^{10,18} in certain situations, Darcy's law is a special case of linear momentum balance. We borrow their insight to show that the THB theory can be extended to the Biot theory. Since (6) is the equation of momentum balance for the solid network, there must be a similar equation for the pore liquid; this equation is, neglecting body forces and inertia,

$$\sigma_{ij,j}^L - m_i^L = 0, \quad (21a)$$

where σ_{ij}^L is the partial stress of the liquid phase and m_i^L is the momentum vector due to the interaction between the solid network and the fluid. In the THB formulation, the interaction between the network and pore fluid is viscous drag, so

$$m_i^L = -f(u_{i,t} - v_{i,t}), \quad (21b)$$

where $u_{i,t}$ and $v_{i,t}$ are the velocities of the network and the pore liquid, respectively. For a more detailed discussion of the various interaction effects during the diffusion of one constituent through other, the reader is referred to Rajagopal and Tao.¹⁸ Following Biot,² we assume that the stress in the liquid is purely hydrostatic, i.e., $\sigma_{ij}^L = p \delta_{ij}$, then (21a) becomes

$$-f(u_{i,t} - v_{i,t}) = p_{,i}. \quad (22)$$

Equation (22) is Darcy's law in three dimensions for an isotropic material (Biot,¹⁴ see Table I) and is a special case of the momentum balance for the fluid. The corresponding momentum balance for the solid is given by

$$\sigma_{ij,j}^s - m_i^s = 0, \quad (23)$$

where σ_{ij} in Eq. (6) must be interpreted as the stress of the solid network, denoted by σ_{ij}^s , and $m_i^s = -m_i^L = f(u_{i,t} - v_{i,t})$ by Newton's third law. Note that in the THB formulation, $m_i^s = fu_{i,t}$, since the pore liquid is stationary. Adding (21a) and (23), we obtain the momentum balance equation for the gel as a whole

$$\sigma_{ij,j} = 0, \quad (24)$$

where $\sigma_{ij} = \sigma_{ij}^s + \sigma_{ij}^L$ is the total stress on the gel. Equation (24) is identical to (1) in the Biot theory. Since, (21a) and (23) are actually equivalent to (22) and (24), the THB formulation has been extended to the Biot formulation. Of course, the stress σ_{ij} in (6) must be interpreted as the network stress σ_{ij}^s , so Eq. (6), in the extended version, is

$$\sigma_{ij}^s = \sigma_{ij} - p\delta_{ij} = 2G\varepsilon_{ij} + \frac{2G\nu\varepsilon}{1-2\nu}\delta_{ij}, \quad (25)$$

which is identical to (2).

VI. DISCUSSION AND CONCLUSION

This article compares the Biot and THB theories of gel deformation. We revisit the problem of gel deswelling and use this example to show that there can be substantial differences between the two theories. By taking into account of momentum transfer of the pore liquid, we show that the THB theory can be extended to the Biot theory. As a result, the THB theory is incomplete.

The usual application of the Biot theory assumes that the cooperative diffusion coefficient D_c is independent of the polymer concentration, while the experiments have demonstrated that D_c depends strongly on it.^{7,19,20} For example, it has been shown that D_c obeys a power-law relationship with the polymer weight fraction.^{7,19} However, this dependence can be easily included in Biot's formulation. Indeed, if we assume that the polymer mass per unit gel volume ρ_s , which increases if the gel fluid escapes, and hence is a monotonic function of the volumetric strain ε [see (A5) and (A7)], then Biot's formulation [i.e., Eqs. (3) and (5)] is still applicable provided that Eq. (5) is modified to

$$\nabla \cdot (D_c(\varepsilon) \nabla \varepsilon) = \frac{\partial \varepsilon}{\partial t}. \quad (26)$$

The only set back is that (26) is nonlinear [since D_c is a function of polymer weight fraction c , which is related to ε by $c \approx \rho_s^0(1-\varepsilon)/\rho_L$, where ρ_s^0 , ρ_L are the initial polymer mass and gel fluid mass per unit gel volume, respectively] and therefore in general it is difficult to obtain closed-form analytic solutions. However, numerical techniques such as finite element method can be used to obtain the solution of these coupled equations. Since the strains are assumed to be small in the Biot and THB theories, the volumetric strain cannot be too large in order for the theory to be self-consistent. The question is how large can the deformation be before the small-strain theory breaks down. Our experience is that the answer to this question is problem dependent, but one would expect that for most problems, a 10% strain could be tolerated. For example, GH have shown that $D_c = D_0(c/c_0)^{0.5}$. A simple calculation shows that D_c changes by 10% for a 10% change in volumetric strain. Unfortunately, there is very little or no literature on the effect of large deformation of gels on its mechanical properties. We hope to address this question in a future work.

The assumption of a constant D_c , though restrictive, has proven to be very useful in explaining the mechanical behav-

ior of elastic gels. For example, Scherer has successfully used Biot's theory to model experiments on silica gels to measure its permeability and elastic modulus.^{3,21} Using Biot's theory to model his beam bending experiment, Scherer²¹ has found for a silica gel containing ethanol, $\nu \approx 0.22$, $G \approx 1.23$ MPa, and $D_c \approx 2.2 \times 10^{-11}$ m²/s. He has also used Biot's theory to explain a wide range of phenomenon such as drying, deswelling, and cracking.^{3,4}

The conclusion of this work can be easily extended to include linear viscoelastic gels (e.g., using the correspondence principle in viscoelasticity theory) as well as problems involving inertia. Body forces can also be included in the formulation as long as they are conservative, that is, they are the gradient of a potential.

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APPENDIX A: GH APPROACH

First, to clarify GH's equations, we introduce the Eulerian coordinate $y = Y + u_2(Y, t)$ in (12) to conform to standard notation in mechanics, where Y denotes the Lagrangian coordinate of a material point. It should be noted that in the THB and Biot formulations, small displacements and strains are assumed so that either y or Y can be used in (12). In GH, the solvent velocity $v_{,t}$ is added to (12) resulting in

$$u_{2,t} - v_{,t} = Du_{2,yy}. \quad (A1)$$

To determine v , GH introduced the following equations [Eqs. (4a) and (4b) in GH]. These equations are supposed to express the mass conservation in the convected or Eulerian coordinates; they are

$$\frac{\partial c}{\partial t} + \frac{\partial(cu_{2,t})}{\partial y} = 0, \quad (A2a)$$

$$\frac{\partial a}{\partial t} + \frac{\partial(av_{,t})}{\partial y} = 0, \quad (A2b)$$

where $u_{2,t}$ is the velocity of the network, $v_{,t}$ is the velocity of the solvent, and a and c are the weight fraction of polymer and solvent, respectively. These two equations are incorrect. Equations (A2a) and (A2b) would have been correct if a and c are replaced by the mass density of polymer (ρ_s) and solvent in the gel (ρ_L), respectively. The correct mass balance equations in Eulerian coordinates are

$$\frac{\partial \rho_s}{\partial t} + \frac{\partial(\rho_s u_{2,t})}{\partial y} = 0, \quad (A3a)$$

$$\frac{\partial \rho_L}{\partial t} + \frac{\partial(\rho_L v_{,t})}{\partial y} = 0. \quad (A3b)$$

It is important to note that ρ_s is not the mass density of the pure polymer but is the mass of the polymer network per unit gel volume. It should also be noted that Eqs. (A3a) and

(A3b) are unaffected by replacing the densities ρ_s and ρ_L by their respective volume fractions. The polymer and solvent weight fractions are related to the densities by

$$c = \frac{\rho_s}{\rho_s + \rho_L}, \quad a = \frac{\rho_L}{\rho_s + \rho_L}, \quad (\text{A4a})$$

$$a + c = 1. \quad (\text{A4b})$$

Note $\rho_s + \rho_L = \rho$, where ρ is the mass density of the gel. During deformation, both ρ_s and ρ_L are unknowns, whereas the weight fractions are related by (A4b).

First, we note that in the formulation of GH, there are four unknowns a , c , u_2 , and ν and there are enough equations relating these unknowns, namely, (A1), (A2a), (A2b), and (A4b) or [to be correct, (A1), (A3a), (A3b), and (A4b)]. Note that Eq. (7) in GH is not an independent equation, since it is simply mass conservation of the network solid expressed in Lagrangian coordinates. Indeed, by considering the motion of a slab of gel material between Y and $Y+dY$, it can be easily shown that

$$\rho_s^0 = \rho_s \left[1 + \frac{\partial u_2}{\partial Y} \right], \quad (\text{A5})$$

where ρ_s^0 and ρ_s are the average density of the polymer before and after deformation. Since (A5) is nothing but the conservation of mass, it can be shown to be equivalent to (A3a) provided we take into account the difference between the Eulerian and Lagrangian coordinates. If one assumes $\partial u_2 / \partial Y \ll 1$, then (A5) implies that

$$\frac{\partial \rho_s}{\partial Y} + \rho_s \frac{\partial^2 u_2}{\partial Y^2} = 0, \quad (\text{A6})$$

where is Eq. (7) in GH with ρ_s in (A6) replaced by c . This replacement relies on the following assumptions: (1) the gel liquid is water; (2) $c \ll a$; and (3) ρ_L is exactly 1 g/cm^3 . To quote GH: “For the small value of c encountered in this experiment, the polymer weight fraction and the polymer density in g cm^{-3} are nearly equal.” As a result of this assumption, the validity of their Eq. (8) depends on units. In addition, ρ_L is not really the density of the solvent (it is the mass of solvent per unit gel volume) and hence is not exactly 1 g/cm^3 . Indeed, if one assumes the average density of the solvent to be exactly constant, then the velocity of the solvent must be identically zero and nothing is gained.

It seems to us that the fluctuation in network density in their experiment is simply $\rho_s \approx \rho_s^0 [1 - \varepsilon]$, where $\varepsilon = u_{2,Y} \approx u_{2,y}$ [see (A5)]. Since the network strain can be obtained using the THB formulation, (15), the density fluctuations $\Delta \rho = \rho_s - \rho_s^0 \approx -\varepsilon \rho_s^0$ are simply

$$\Delta \rho = \frac{P \rho_s^0}{A E_L} \operatorname{erfc} \left(\frac{y}{2 \sqrt{D_c t}} \right). \quad (\text{A7})$$

Indeed, one cannot expect to do any better, since the THB formulation is based on small-strain theory.

APPENDIX B: DERIVATION OF (18)

In this problem, the shear strains are identically zero and $\varepsilon_{11} = \varepsilon_{33} = 0$. This implies that the shear stresses are identi-

cally zero and the gel is subjected to in-plane biaxial compression, i.e., $\sigma_{11} = \sigma_{33} = \sigma$. It can be shown that the strain is related to the pore pressure and stress by $\varepsilon_{22} = (\sigma_{22} - p)/E_L$ for this simple problem. Since the shear stresses are identically zero, equilibrium in the y direction dictates that σ_{22} is a function of time only. With Biot's approach, the volumetric strain satisfies (5). For our special case, $\varepsilon = \varepsilon_{22}$, so using the relation between ε_{22} and pore pressure, we have

$$\frac{\partial p}{\partial t} - D_c \frac{\partial^2 p}{\partial y^2} = \frac{\partial \sigma_{22}}{\partial t}. \quad (\text{B1a})$$

Hence, even in this simple case, the pressure field is coupled to the gel deformation. We need to solve (B1a) subjected to the conditions (17) for pore pressure. Let $p = p_1 + p_c$, where $p_c = \sigma_{22}(t)$ is the particular solution. Then, the governing equation (B1a) and pore pressure boundary conditions in (17) become

$$\frac{\partial p_1}{\partial t} - D_c \frac{\partial^2 p_1}{\partial y^2} = 0, \quad (\text{B1b})$$

$$p_1(y=0, t > 0) = -p_c(t),$$

$$p_{1,y}(y=h, t > 0) = 0,$$

$$p_1(y, t=0) = 0. \quad (\text{B1c})$$

Taking the Laplace transform of (B1b) and applying the conditions (B1c) result in

$$\tilde{p}_1 = -\frac{\tilde{\sigma}_{22}(s)}{\cosh \beta h} \cosh \beta(y-h), \quad (\text{B2})$$

where $\beta = \sqrt{s/D_c}$. Now, using the relationship between ε_{22} and pore pressure, we have

$$\frac{\partial \tilde{u}_2}{\partial y} = \frac{\tilde{\sigma}_{22} - \tilde{p}}{E_L} = -\frac{\tilde{p}_1}{E_L} \Rightarrow \tilde{u}_2 = -\frac{1}{E_L} \int_0^y \tilde{p}_1(y', s) dy' + c(s). \quad (\text{B3})$$

Applying the displacement boundary conditions in (17), we obtain $c(s) = 0$ and

$$\tilde{\sigma}_{22}(s) = -\frac{E_L V}{\sqrt{D_c s^{3/2}}} \coth \beta h. \quad (\text{B4})$$

Using (B4) in (B2) and substituting the resulting expression in (B3), we obtain

$$\tilde{u}_2(\text{BS}) = -\frac{V}{s^2} \left[1 - \frac{\sinh \beta(h-y)}{\sinh \beta h} \right]. \quad (\text{B5})$$

Let us now consider the THB formulation, which is much easier to solve. We basically need to solve (8) subjected to (16). Using the Laplace transforms it can be shown that

$$\widetilde{u}_2(\text{THB}) = -\frac{V \sinh \beta y}{s^2 \sinh \beta h}. \quad (\text{B6})$$

APPENDIX C: INVERSION OF (B6)

Let $a=y^2/D_c$ and $b=h^2/D_c$. Using the inversion integral and residue theorem,

$$\ell^{-1} \left[\Lambda(s) = \frac{1}{s^2} \frac{\sinh \sqrt{as}}{\sinh \sqrt{bs}} \right] = \sum_n \text{Res} \frac{e^{st}}{s^2} \frac{\sinh \sqrt{as}}{\sinh \sqrt{bs}} \Big|_{s_n}, \quad (\text{C1})$$

where ℓ^{-1} denotes the inverse Laplace transform and s_n are the singular points of $\Lambda(s)$. From the expansion of $\Lambda(s)$, it can be seen that the singularities are located at $s=0$ (double pole) and zeroes of $\sinh \sqrt{bs}$, which are given by $s_n = -n^2 \pi^2 / b$. On evaluating the residues and substituting in (B6), we obtain

$$u_{2\text{THB}}(y,t) = -V \left[\frac{y}{h} \left(t + \frac{y^2 - h^2}{6D_c} \right) + 2 \frac{h^2}{D_c} \times \sum_{n=1}^{\infty} (-1)^{n+1} \frac{e^{-(n^2 \pi^2 / (h^2/D))t}}{n^3 \pi^3} \sin \left(\frac{n \pi y}{h} \right) \right]. \quad (\text{C2})$$

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