

Asymptotic description of confined hydrogel swelling

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Hydrogels are soft, poroelastic solids, comprising hydrophilic polymer scaffold surrounded by adsorbed water that is free to move through the pore spaces. Hydrogels are typically biocompatible, and are therefore used in a wide variety of medical contexts. For instance, they are used to make soft contact lenses, and have potential as advanced wound dressings, targeted drug delivery devices, and deployable intestinal stents. In all these applications, the hydrogel tends to be confined by or adhered to its surroundings, and while hydrogel dynamics has received increasing mathematical attention recently, most studies have focused on free swelling. In this paper, we mathematically analyze the swelling and deswelling behavior of a hydrogel confined between two plates, in both the case where the hydrogel can slip parallel to the wall and the case where it cannot. In the slip case, we numerically solve for a full nonlinear solution based on uniaxial deformation. In the adhered case, focusing on small deformation, we find that the gel may swell either completely or incompletely (i.e., the water only penetrates partially through the gel). We give a leading-order closed-form solution for the evolution of the porosity, and thus the force on the two plates, in both cases. The partial swelling may greatly impact the efficacy of hydrogels as actuators for deployable medical devices, so may need to be a consideration in design.

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I. INTRODUCTION

Hydrogels comprise a hydrophilic polymer scaffold, surrounded by adsorbed water [1]. Remarkably, the water content of hydrogels can be up to 99% [2], and yet they may still be treated as a (soft) deformable solid. Because of their ability to take on a large volume of water, as hydrogels swell their volume may increase several-fold between the initial and fully swollen states [3]. Everyday examples of hydrogels include the children's sensory toy Orbeez (or water beads) [4], and the inner lining of disposable diapers [5].

Because they are soft, often biocompatible, and can be loaded with solute, hydrogels are useful for a wide range of medical applications [6]. For instance, soft contact lenses are made from hydrogels [7], hydrogel-based wound dressings promote healing [8], and there is considerable research interest in hydrogels as scaffolds in tissue engineering [9]. Hydrogels can also be designed to swell in response to changes in their environment such as temperature and pH [10], and as a result they are effective actuators; for example, they can be used to design a delivery mechanism for targeted drug therapies [11], a trigger mechanism for deployable medical devices such as

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gastrointestinal stents [12,13], or a gripping mechanisms in soft robotics [14]. In the vast majority of applications, the hydrogel does not swell in free space, but is rather adhered to or otherwise confined by some external surface or tissue, which significantly affects the swelling dynamics. The aim of this paper is therefore to investigate how these constraints influence swelling theoretically.

Hydrogels are typically modelled as a porous material since water is able to flow through the polymer scaffold [15]. The dynamics of hydrogel swelling is described by a competition between the elastic and chemical parts of their free energy [16,17]; from the chemical point of view, it is favourable for the gel to absorb water, but this requires work to be done to move the water and results in an elastic penalty since it stretches the gel's polymer chains. It eventually reaches equilibrium when the elastic and chemical forces are in balance. This energetic formulation coupled with poromechanical theory (i.e., Darcy's law) produces a framework of the same form as classical poroelasticity, wherein the total stress can be split into an effective stress [18] (which captures the stress in the elastic matrix separately from the fluid) and a fluid pressure, which in hydrogels has an additional osmotic component due to the gel hydrophilicity. Using a linear elastic constitutive law for the effective stress in the style of Ref. [19] then produces the linear poroelastic gel model, which has been used to derive analytic and semi-analytic solutions for gel swelling in the limit of small deformation [15]. However, hydrogels often undergo large deformation, and the linear regime may not adequately approximate swelling in this case. This motivates the nonlinear constitutive laws taken by, for example, Refs. [2,20–22]. These models have the advantage of allowing large deformations but this comes at the expense of greater analytical complexity. A third approach is taken by Webber and Worster [23], which allows for nonlinear isotropic swelling while assuming linear deviatoric strain. This is an appropriate and useful assumption in the case of free swelling, but becomes invalid in certain practical cases, including (most relevantly to the present work) in cases of adhesion and confinement where the deviatoric strain may perhaps even be the dominant contribution.

Swelling under adhesion and/or confinement has received significantly less attention than free swelling, despite its direct relevancy to numerous practical applications. Experiments have been carried out by, for example, Refs. [24–26], and some authors have used finite element methods to numerically compute fully nonlinear solutions [27,28]. From an analytical point of view, there is a large body of research on surface instabilities in infinite gel layers adhered along one surface [29,30], which typically arise beyond a critical swelling ratio of around 1.5 to 3 depending on gel properties, but these typically focus on this single canonical example. Doi [15] uses a linear poroelastic formulation to investigate swelling of gel layers in several cases, including under confinement by rigid plates and under mechanical load, but analytical investigations of this kind are relatively rare.

Our aim is therefore to develop the theory of gel swelling under confinement from a mathematical viewpoint, by focusing on gel layers confined laterally by rigid plates. In terms of practical applications, this setup could represent a subcutaneous drug delivery or wound healing device, or an actuation mechanism. We take a particular interest in the force the gel can exert on the plates with the actuation application in mind. We investigate two scenarios: (i) confinement with slip, i.e., the gel may have a displacement parallel to the plates at the gel-plate boundary, and (ii) gels which are fully adhered to the plates, i.e., no displacement is permitted at the boundary. Both of these scenarios are shown schematically in Fig. 1. The latter case constrains swelling more severely than the former. A related problem was considered analytically by Yamaue and Doi [31] using linear poroelasticity to explain experimental results by Suzuki and Hara [24], who considered a fully adhered thin gel layer between two rigid plates but treated the plates as free to move. A parabolic gel shape profile is observed and reproduced by analysis. However, as noted by the authors, the approach neglects the fast-time process of the parabola formation, instead focusing on the slower process of the vertical motion of the plates; it is this faster process which is of interest here. In both problems, we derive our analysis from the fully nonlinear model of Refs. [2,32]. In case (i) this allows us to capture large deformations, and in case (ii), although we assume small deformation, we will see that the potential for large displacement gradients mean it is important to treat nonlinear terms with care.

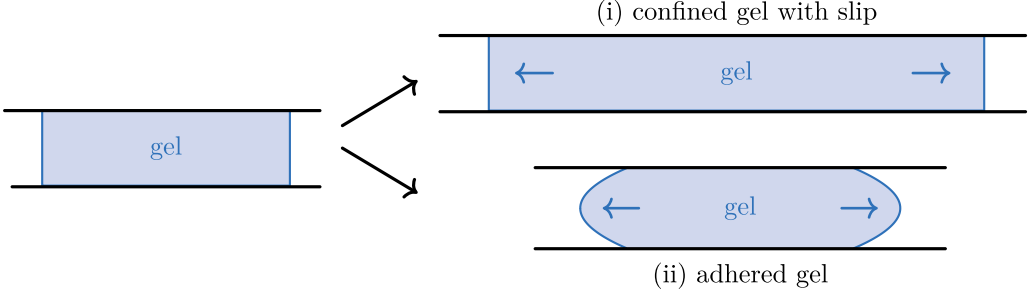


FIG. 1. Schematic of a hydrogel swelling in confinement. We consider two different cases for the wall boundary condition. In (i), we permit slip at the wall. In (ii), we treat the hydrogel as fully adhered to the wall.

The paper is set out as follows. In Sec. II, we describe the large deformation poroelastic gel framework. In Sec. III, we use the framework to solve the problem of confined gels with slip at the wall, both numerically for large deformations and analytically for small deformations. In Sec. IV, we consider the case of fully adhered gels. Finally, we discuss our results and provide closing remarks in Sec. V.

II. LARGE DEFORMATION POROELASTIC GEL MODEL

We use the framework of large deformation poroelasticity to model the hydrogel, as laid out by Bertrand *et al.* [2] and MacMinn *et al.* [32]. We first set out the equations in their full dimensional form before proceeding to nondimensionalize in Sec. II A.

Here and throughout the paper, tildes ($\tilde{\cdot}$) indicate a dimensional quantity and the absence thereof indicates a nondimensional quantity. We use an Eulerian formulation, with displacement vector given by

$$\tilde{\xi}(\tilde{\mathbf{x}}, \tilde{t}) = \tilde{\mathbf{x}} - \tilde{\mathbf{x}}_0(\tilde{\mathbf{x}}, \tilde{t}), \quad (1)$$

where $\tilde{\mathbf{x}}_0$ is the reference position of the material point which is at position $\tilde{\mathbf{x}}$ at time $\tilde{t}$. This then allows us to define the inverse deformation gradient tensor,

$$\mathbf{F}^{-1} = \tilde{\nabla} \tilde{\mathbf{x}}_0 = \mathbf{I} - \tilde{\nabla} \tilde{\xi}, \quad (2)$$

and the Jacobian $J = \det(\mathbf{F})$, which by mass conservation is related to the porosity ϕ by

$$J = \frac{1 - \phi_d}{1 - \phi}. \quad (3)$$

Here ϕ_d is the constant porosity of the reference state. We take the reference state to be the initial state of the gel rather than the fully dry state, so $\phi_d > 0$. This is because choosing a reference state in a confined geometry imposes restrictions on the possible states of hydrogel (i.e., taking the reference state to be fully dry in a confined space presumes that the fully dry hydrogel fits in exactly said space).

The gel dynamics are governed by the interaction of elastic and chemical forces. The chemical energy gain from absorbing water is countered by the elastic energy penalty from stretching polymer chains. The free energy density of the hydrogel per unit reference volume is then given by the sum of its elastic component and the energy due to chemical mixing [17],

$$\tilde{W} = \tilde{W}_{\text{elast}} + \tilde{W}_{\text{mix}}. \quad (4)$$

We assume a neo-Hookean constitutive law for the elastic free energy,

$$\tilde{W}_{\text{elast}} = \frac{\tilde{\mu}_s}{2} [\text{tr}(\mathbf{F}\mathbf{F}^T) - 3 - 2 \log J], \quad (5)$$

where $\tilde{\mu}_s$ is the shear modulus of the gel. The mixing contribution to free energy is derived from Flory-Huggins polymer theory (see Doi [33]),

$$\tilde{W}_{\text{mix}} = \tilde{K}J[\phi \log \phi + \chi \phi(1 - \phi)], \quad (6)$$

where the constant $\tilde{K} = \tilde{k}_B \tilde{T} / \tilde{\Omega}_f$ is the characteristic osmotic energy of the gel (where $\tilde{k}_B$ is the Boltzmann constant, $\tilde{T}$ is the temperature and $\tilde{\Omega}_f$ is the volume of fluid per fluid molecule), and χ is the Flory mixing parameter, which quantifies the interaction energy between polymer and solvent. We take the mixing parameter to be a constant with the value 0.4 [2]. The stress tensor can then be computed as

$$\tilde{\sigma} = J^{-1} \mathbf{F} \frac{\partial \tilde{W}}{\partial \mathbf{F}} - \frac{\tilde{\mu}}{\tilde{\Omega}_f} \mathbf{I}, \quad (7)$$

where $\tilde{\mu}$ is the chemical potential of the gel, which measures the energy required to move a molecule of fluid from the environment to the mixture. The quantity $\tilde{\mu} / \tilde{\Omega}_f$ can be thought of as equivalent to the fluid pore pressure in poromechanics [34]. Computing this then yields the stress tensor

$$\tilde{\sigma} = \tilde{\sigma}' - [\tilde{\Pi}(\phi) + \tilde{\mu}] \mathbf{I}, \quad (8)$$

where

$$\tilde{\sigma}' = J^{-1} \mathbf{F} \frac{\partial \tilde{W}_{\text{elast}}}{\partial \mathbf{F}} = \tilde{\mu}_s J^{-1} (\mathbf{F} \mathbf{F}^T - \mathbf{I}) \quad (9)$$

is the Terzaghi effective stress [18], and

$$\tilde{\Pi}(\phi) = - \frac{\partial \tilde{W}_{\text{mix}}}{\partial J} = -\tilde{K}[\log \phi + \chi(1 - \phi)^2 + (1 - \phi)] \quad (10)$$

is the osmotic pressure, which can be thought of as the additional pressure arising from the hydrophilic properties of the gel. Labelling the total pressure (osmotic pressure and chemical potential/pore pressure) as

$$\tilde{p} = \tilde{\Pi} + \frac{\tilde{\mu}}{\tilde{\Omega}_f}, \quad (11)$$

then recasts the model in the form of Biot poroelasticity [19],

$$\tilde{\sigma} = \tilde{\sigma}' - \tilde{p} \mathbf{I}. \quad (12)$$

Conservation of momentum in the system is governed by Cauchy's momentum equation, which reads

$$\tilde{\nabla} \cdot \tilde{\sigma} = 0 \Rightarrow \tilde{\nabla} \tilde{p} = \tilde{\nabla} \cdot \tilde{\sigma}'. \quad (13)$$

These are coupled to the governing equations of the poroelastic flow through the gel. We treat the gel as consisting of two phases, fluid and solid, and assume each phase to be individually incompressible (though the gel overall is compressible as it may absorb or expel fluid). Mass conservation of the fluid and solid phases, respectively, necessitate the balances

$$\frac{\partial \phi}{\partial \tilde{t}} + \tilde{\nabla} \cdot (\phi \tilde{\mathbf{v}}_f) = 0, \quad (14)$$

and

$$\frac{\partial}{\partial \tilde{t}}(1 - \phi) + \tilde{\nabla} \cdot [(1 - \phi) \tilde{\mathbf{v}}_s] = 0, \quad (15)$$

where $\tilde{\mathbf{v}}_{f,s}$ are the fluid and solid velocities, respectively, and

$$\tilde{\mathbf{v}}_s = \frac{\partial \tilde{\boldsymbol{\xi}}}{\partial \tilde{t}} \cdot \mathbf{F}. \quad (16)$$

Together, Eqs. (14) and (15) imply

$$\tilde{\nabla} \cdot \tilde{\mathbf{q}} = 0, \quad (17)$$

where $\tilde{\mathbf{q}} = \phi \tilde{\mathbf{v}}_f + (1 - \phi) \tilde{\mathbf{v}}_s$. The flow is driven by the chemical potential gradient in the hydrogel, as governed by Darcy's law,

$$\phi(\tilde{\mathbf{v}}_f - \tilde{\mathbf{v}}_s) = -\frac{\tilde{k}(\phi)}{\tilde{\eta}_f} \tilde{\nabla} \left(\frac{\tilde{\mu}}{\tilde{\Omega}_f} \right). \quad (18)$$

Here $\tilde{k}(\phi)$ is the permeability of the hydrogel, which depends on porosity, and $\tilde{\eta}_f$ is the dynamic viscosity of the fluid. Throughout the paper, we adopt $\tilde{k}(\phi) = \tilde{k}_0 \phi / (1 - \phi)^{3/2}$, following Ref. [2]. Combining Eqs. (14), (15), (17), and (18), we arrive at a nonlinear advection-diffusion equation for the porosity of the gel,

$$\frac{\partial \phi}{\partial \tilde{t}} + \tilde{\mathbf{q}} \cdot \tilde{\nabla} \phi = \tilde{\nabla} \cdot \left[\frac{(1 - \phi) \tilde{k}(\phi)}{\tilde{\eta}_f} \tilde{\nabla} \left(\frac{\tilde{\mu}}{\tilde{\Omega}_f} \right) \right]. \quad (19)$$

The gel dynamics are then largely governed by the coupling of Eqs. (13) and (19).

A. Nondimensionalization

We nondimensionalize our equations according to

$$\begin{aligned} (\tilde{\mathbf{x}}, \tilde{\boldsymbol{\xi}}, \tilde{\mathbf{x}}_0) &= \tilde{L}(\mathbf{x}, \boldsymbol{\xi}, \mathbf{x}_0), \quad \left(\tilde{\sigma}, \tilde{p}, \frac{\tilde{\mu}}{\tilde{\Omega}_f}, \tilde{\Pi} \right) = \tilde{K}(\sigma, p, \mu, \Pi), \\ \tilde{k} &= \tilde{k}_0 k, \quad \tilde{t} = \frac{\tilde{L}^2 \tilde{\eta}_f}{\tilde{k}_0 \tilde{K}} t, \quad (\tilde{\mathbf{v}}_{f,s}, \tilde{\mathbf{q}}) = \frac{\tilde{k}_0 \tilde{K}}{\tilde{\eta}_f \tilde{L}} (\mathbf{v}_{f,s}, \mathbf{q}), \end{aligned} \quad (20)$$

where $\tilde{L}$ is some measure of the size of the hydrogel, and $\tilde{k}_0$ is the permeability scale. To give some indication of typical values for the dimensional parameters used in our nondimensionalization, Bertrand *et al.* [2] estimated $\tilde{\Omega}_f \approx 3 \times 10^{-29} \text{ m}^3$ (which yields $\tilde{K} = 1.36 \times 10^8 \text{ Pa}$ at $\tilde{T} = 295 \text{ K}$) and $\tilde{k}_0 = 8 \times 10^{-20} \text{ m}^2$. Given the viscosity of water is $\tilde{\eta}_f = 10^{-3} \text{ Pa s}$, the result is a characteristic timescale for the gel swelling of between 10 s and 1000 s for a gel with size in the millimeter to centimeter range.

The momentum equation then becomes

$$\nabla p = \nabla \cdot \boldsymbol{\sigma}', \quad (21)$$

where $p = \mu + \Pi$ and $\Pi(\phi) = -[\log \phi + \chi(1 - \phi)^2 + (1 - \phi)]$. This is coupled to the neo-Hookean constitutive law,

$$\boldsymbol{\sigma}' = GJ^{-1}(\mathbf{F}\mathbf{F}^T - \mathbf{I}), \quad (22)$$

where $G = \tilde{\mu}_s / \tilde{K} = \tilde{\Omega}_p / \tilde{\Omega}_f$ is the ratio of shear modulus to osmotic modulus, a dimensionless number which captures the relative importance of elastic to chemical forces. Since hydrogels are soft and highly absorbent materials, G is typically a small parameter, estimated as around 10^{-4} for some gels [35]. The Jacobian and deformation gradient are related to displacement and porosity via

$$\mathbf{F}^{-1} = \mathbf{I} - \nabla \boldsymbol{\xi}, \quad J = \det \mathbf{F} = \frac{1 - \phi_d}{1 - \phi}. \quad (23)$$

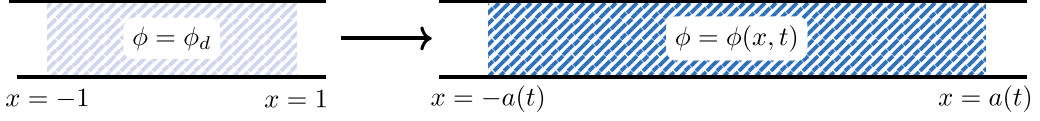


FIG. 2. Schematic of a hydrogel swelling in confinement when it can slip along side walls. The hydrogel will swell symmetrically about its midpoint.

This is coupled to the advection-diffusion equation for porosity,

$$\frac{\partial \phi}{\partial t} + \mathbf{q} \cdot \nabla \phi = \nabla \cdot [(1 - \phi)k(\phi)\nabla \mu], \quad (24)$$

where

$$\mathbf{q} = -k(\phi)\nabla \mu + \mathbf{v}_s, \quad \mathbf{v}_s = \frac{\partial \boldsymbol{\xi}}{\partial t} \cdot \mathbf{F}, \quad \nabla \cdot \mathbf{q} = 0. \quad (25)$$

B. Initial and boundary conditions

In this paper, we consider the swelling of an initially rectangular hydrogel confined between two walls; see Fig. 7. The hydrogel is initially at uniform porosity $\phi = \phi_d$, where $0 < \phi_d < 1$, and is submerged in water at $t = 0$ and allowed to swell. The initial conditions are therefore

$$\phi(\mathbf{x}, 0) = \phi_d, \quad \boldsymbol{\xi}(\mathbf{x}, 0) = \mathbf{0}. \quad (26)$$

The boundary conditions at the free surface are [2,15]

$$\mu = \mu_*, \quad \boldsymbol{\sigma} \cdot \mathbf{n} = \mathbf{0}, \quad (27)$$

where μ_* is the ambient chemical potential in the surrounding water and $\mathbf{n}$ is the normal vector to the free surface. In both of the problems we consider in this paper, we will find that these two conditions combine to enforce an effective condition on the porosity at the free surface, i.e., $\phi = \phi_w$ there.

We consider two different cases for the wall boundary conditions:

(1) In the first case, treated in Sec. III, we allow the gel to slip at the wall, and therefore the deformation reduces to uniaxial at leading order.

(2) In the second case, we consider gels which are fully adhered to both walls, and thus we impose $\boldsymbol{\xi} = \mathbf{0}$ there. This boundary condition severely restricts the swelling of the gel since it cannot expand vertically, and horizontal expansion results in large displacement gradients (and hence stresses). We treat this problem linearly in Sec. IV, and use the solution we obtain to derive insights into the nonlinear swelling case.

III. CONFINED GELS WITH SLIP

In this section, we consider the scenario in which the hydrogel can slip against the confining walls. We show this schematically in Fig. 2. In this case, the hydrogel undergoes uniaxial deformation [15,36], without loss of generality in the x direction, and then the deformation matrix reduces to $\mathbf{F} = \text{diag}(J(x, t), 1, 1)$. The Jacobian then satisfies

$$J^{-1} = \frac{1 - \phi}{1 - \phi_d} = 1 - \frac{\partial \xi}{\partial x}. \quad (28)$$

This can be integrated once to obtain the displacement in terms of the porosity,

$$\xi(x, t) = \frac{\phi_d}{1 - \phi_d}x + \int_0^x \frac{\phi(s, t)}{1 - \phi_d} ds. \quad (29)$$

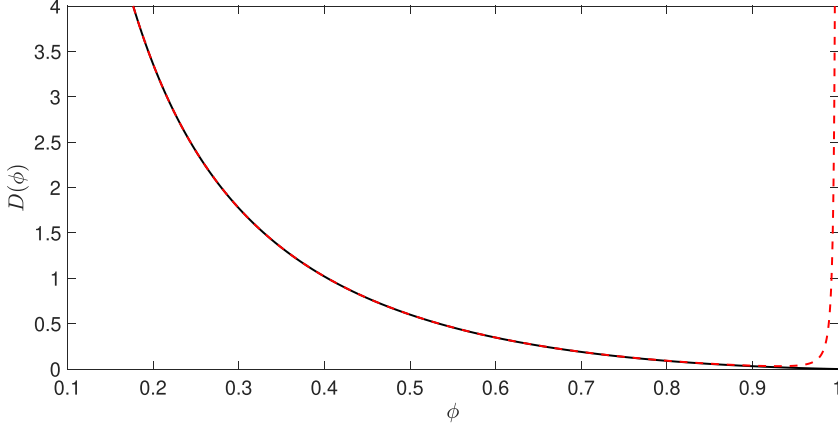


FIG. 3. The nonlinear diffusion coefficient $D(\phi)$, for $G = 0$ (black), and $G = 10^{-4}$ (red).

Under uniaxial swelling, the effective elastic stress (22) becomes

$$\sigma'_{xx} = G(J - J^{-1}) = G\left(\frac{1 - \phi_d}{1 - \phi} - \frac{1 - \phi}{1 - \phi_d}\right), \quad (30)$$

which is a function of only the porosity, ϕ . This allows us to write the momentum equation (75) in the form

$$\frac{\partial p}{\partial x} = \frac{\partial \sigma'_{xx}}{\partial x} \Rightarrow \frac{\partial \mu}{\partial x} = \left(-\frac{d\Pi}{d\phi} + \frac{d\sigma'_{xx}}{d\phi}\right) \frac{\partial \phi}{\partial x}. \quad (31)$$

We can then substitute (31) into the advection-diffusion equation (19) to obtain an equation solely for the porosity. In uniaxial swelling (17) implies $\mathbf{q} = q(t)\hat{\mathbf{x}}$ (where $\hat{\mathbf{x}}$ is the unit vector in the x direction), and since $q = 0$ at $x = 0$ by symmetry, we have $q \equiv 0$ throughout the dynamics. This results in the nonlinear diffusion equation for the porosity,

$$\frac{\partial \phi}{\partial t} = \frac{\partial}{\partial x} \left(D(\phi) \frac{\partial \phi}{\partial x} \right), \quad (32)$$

where

$$\begin{aligned} D(\phi) &= (1 - \phi)k(\phi) \left(-\frac{d\Pi}{d\phi} + \frac{d\sigma'_{xx}}{d\phi} \right), \\ &= (1 - \phi)k(\phi) \left[\left(\frac{1}{\phi} - 1 - 2\chi(1 - \phi) \right) + G \left(\frac{1}{1 - \phi_d} + \frac{1 - \phi_d}{(1 - \phi)^2} \right) \right], \end{aligned} \quad (33)$$

is the nonlinear diffusion coefficient. We plot the diffusion coefficient, $D(\phi)$, in Fig. 3, for $G = 0$ and $G = 10^{-4}$. We observe that the leading order diffusion coefficient ($G = 0$) provides a good approximation for $0 < \phi < 0.9$, however in a small region near $\phi = 1$, the contribution due to the elastic stress dominates the behavior. Since hydrogels can reach large values of porosity around $\phi \approx 0.99$ [2], the contribution from the elastic stress is potentially significant even for small values of G .

We close the porosity equation (32) by supplying the boundary conditions described in Sec. II B. Since in our uniaxial case we can write the elastic stress σ_{xx} as a function of ϕ [see Eq. (30)], the stress-free condition on the outer edge of the hydrogel (27) is effectively a nonlinear equation for the value of ϕ at $x = a(t)$. We denote the solution to this equation by ϕ_w ; more concretely, ϕ_w is the solution of $\mu_* = -\Pi(\phi_w) + \sigma'_{xx}(\phi_w)$. Thus, we supply the following boundary

conditions:

$$\phi = \phi_w \quad \text{on} \quad x = a(t), \quad (34)$$

$$\frac{\partial \phi}{\partial x} = 0 \quad \text{on} \quad x = 0. \quad (35)$$

The first represents the stress balance condition, and the second condition ensures symmetry. Due to the free end of the hydrogel, we require another boundary condition to determine its position. We achieve this by setting the solid velocity to equal the velocity of the free end, i.e.,

$$\frac{da}{dt} = v_s \Rightarrow \frac{da}{dt} = \frac{D(\phi)}{(1-\phi)} \frac{\partial \phi}{\partial x} \quad \text{at} \quad x = a(t), \quad (36)$$

where we have used Eqs. (18) and (31) to write v_s as a function of porosity, ϕ , and its derivatives.

Finally, we supply the initial condition

$$\phi(x, 0) = \phi_d. \quad (37)$$

A. Numerical solution

In this section, we provide an overview of our numerical scheme to solve (32). To account for the moving boundary we rescale our spatial coordinate, as $y = x/a(t)$, which maps $x \in [0, a(t)]$ onto $y \in [0, 1]$. With this scaling the system (32)–(37) becomes

$$\frac{\partial \phi}{\partial t} + \frac{\dot{a}y}{a} \frac{\partial \phi}{\partial y} = \frac{1}{a^2} \frac{\partial}{\partial y} \left(D(\phi) \frac{\partial \phi}{\partial y} \right), \quad (38)$$

where $\dot{a} = da/dt$. The boundary and initial conditions then become

$$\phi = \phi_w \quad \text{on} \quad y = 1, \quad (39)$$

$$\frac{da}{dt} = \frac{D(\phi)}{a(1-\phi)} \frac{\partial \phi}{\partial y} \quad \text{on} \quad y = 1, \quad (40)$$

$$\frac{\partial \phi}{\partial y} = 0 \quad \text{on} \quad y = 0, \quad (41)$$

$$\phi(y, 0) = \phi_d. \quad (42)$$

The system Eqs. (38)–(42) is then solved numerically, using central differences in space, and a forward Euler scheme in time. We use $N = 100$ discretization points in space and use a timestep of 10^{-5} . The convergence of the numerical scheme has been compared to both greater spatial and time resolutions.

B. Equilibrium state

At late times, Eq. (32) admits a spatially uniform solution in which the hydrogel has fully swollen and thus $\phi \equiv \phi_w$. Then by conservation of polymer we can deduce that

$$a \rightarrow \frac{1 - \phi_d}{1 - \phi_w} \quad \text{as} \quad t \rightarrow \infty. \quad (43)$$

C. Force exerted on the walls

Once we have solved for the porosity using Eq. (38), we can use Eq. (31) and stress balance at the free end of the hydrogel to find that the pressure in the hydrogel is given by $p = \sigma'_{xx}$. Then the force exerted on the confining walls is given by the integral of the pressure inside the hydrogel along

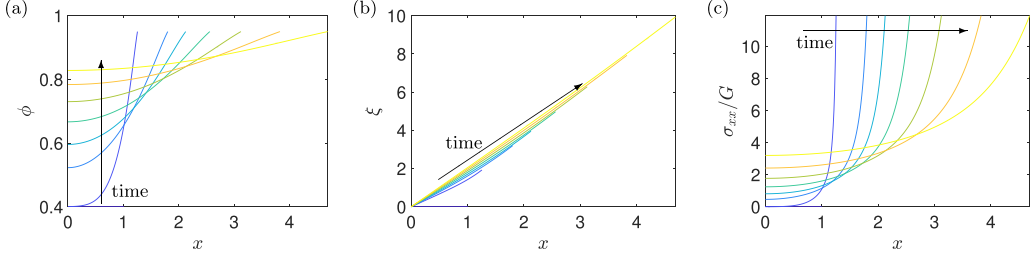


FIG. 4. Spatial distribution of (a) porosity, ϕ , (b) the displacement, ξ , (c) the scaled effective stress, σ'_{xx}/G , taken at the time points $t = 0, 0.1, 1, 2.4, 8, 16, 32$, with an initial uniform porosity of $\phi_d = 0.4$, equilibrium porosity of $\phi_w = 0.95$, and dimensionless ratio of shear modulus to osmotic modulus $G = 10^{-4}$.

the upper confining wall:

$$\mathbf{F}(t) = G\hat{\mathbf{z}} \int_0^{a(t)} \left(\frac{1 - \phi_d}{1 - \phi} - \frac{1 - \phi}{1 - \phi_d} \right) dx. \quad (44)$$

In the limit as $t \rightarrow \infty$, we find there is a constant residual force on the upper wall, due to the build up of elastic stresses as the hydrogel swells, given by

$$\mathbf{F}_\infty = G \left[\left(\frac{1 - \phi_d}{1 - \phi_w} \right)^2 - 1 \right] \hat{\mathbf{z}}. \quad (45)$$

Intuitively, this can be thought of the force required to make the gel swell uniaxially rather than isotropically.

1. Results

We plot the spatial distributions of the porosity ϕ , displacement ξ , and the scaled stress σ'_{xx}/G in Fig. 4 at different time points, for a model hydrogel that has an initial uniform porosity of $\phi_d = 0.4$, equilibrium porosity, $\phi_w = 0.95$, and dimensionless ratio of shear modulus to osmotic modulus $G = 10^{-4}$. Initially, we observe large gradients in the porosity near the free end of the hydrogel where the water is imbibed. The hydrogel then expands as it equilibrates to account for the larger volume of water. We show the time evolution of the free end of the hydrogel $x = a(t)$ in Fig. 5(a).

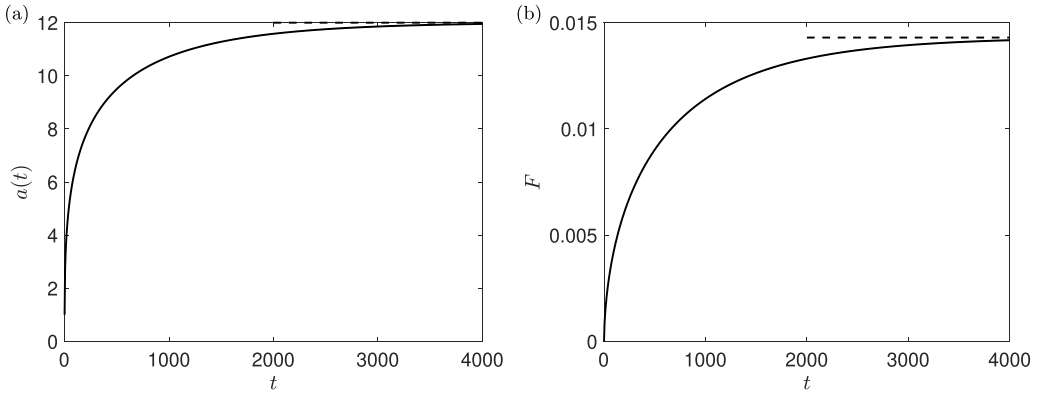


FIG. 5. (a) Position of the edge of the hydrogel $x = a(t)$ and (b) the force on the upper plate exerted by the hydrogel, $\mathbf{F}(t) = F\hat{\mathbf{z}}$, for a hydrogel with $\phi_d = 0.4$, $\phi_w = 0.95$, and $G = 10^{-4}$. The dashed lines indicate the limiting behavior as $t \rightarrow \infty$.

There is an initial fast expansion of the hydrogel and a slow relaxation to the new fully swollen equilibrium state. This expansion leads to a buildup of elastic stresses within the hydrogel [see Fig. 4(c)]. This stress is directly proportional to the pressure inside the hydrogel and thus exerts a force, $F(t)$, on the confining walls, which we plot in Fig. 5(b). We find that once the hydrogel is fully swollen it exerts a residual force on the walls given by Eq. (45).

2. Linearized solution

If the difference between the initial porosity of the hydrogel and the equilibrium porosity is small, i.e., $|\phi_w - \phi_d| \ll 1$, then we can linearize Eq. (32) by expanding $\phi = \phi_d + (\phi_w - \phi_d)\Phi$, where $\Phi = O(1)$. Also if the change in porosity is small then we expect the change in a to also be small, so we further expand $a(t) = 1 + (\phi_w - \phi_d)a_1(t)$. These expansions then lead to the linearized system

$$\frac{\partial \Phi}{\partial t} = D \frac{\partial^2 \Phi}{\partial x^2}, \quad (46)$$

with boundary conditions

$$\Phi(1, t) = 1, \quad (47)$$

$$\frac{\partial \Phi}{\partial x} = 0, \quad (48)$$

$$\Phi(x, 0) = 0, \quad (49)$$

$$\left. \frac{da_1}{dt} = \frac{D}{1 - \phi_d} \frac{\partial \Phi}{\partial x} \right|_{x=1}, \quad (50)$$

where $D = D(\phi_d)$. We observe that in the linear regime, the equation for the position of the free end of the hydrogel decouples, allowing us to solve for the porosity on a fixed domain. The porosity can be found using separation of variables and expressed as a Fourier series, resulting in

$$\Phi = 1 - \sum_{n=0}^{\infty} \frac{4(-1)^n}{(2n+1)\pi} \cos\left(\frac{(2n+1)\pi x}{2}\right) \exp\left(-\frac{D(2n+1)^2\pi^2 t}{4}\right). \quad (51)$$

We plot an example solution of the linearized porosity, Φ , in Fig. 6. We observe qualitatively similar dynamics to solutions of the full system shown in Fig. 4(a), with initially large porosity gradients near the edge of the hydrogel and a slow relaxation to the equilibrium porosity. The correction to the position of the free end of the hydrogel, a_1 , can then be calculated using

$$\frac{da_1}{dt} = \frac{D}{1 - \phi_d} \sum_{n=0}^{\infty} 2 \exp\left(-\frac{D(2n+1)^2\pi^2 t}{4}\right), \quad (52)$$

which can be integrated analytically to obtain

$$a_1(t) = \frac{1}{1 - \phi_d} \sum_{n=0}^{\infty} \frac{8}{(2n+1)^2\pi^2} \left[1 - \exp\left(-\frac{D(2n+1)^2\pi^2 t}{4}\right) \right]. \quad (53)$$

In the linear regime we find that, in the limit $t \rightarrow \infty$,

$$a \rightarrow 1 + \frac{\phi_w - \phi_d}{1 - \phi_d}, \quad (54)$$

which reproduces the first two terms in the asymptotic expansion of Eq. (43) for small $\phi_w - \phi_d$, as expected.

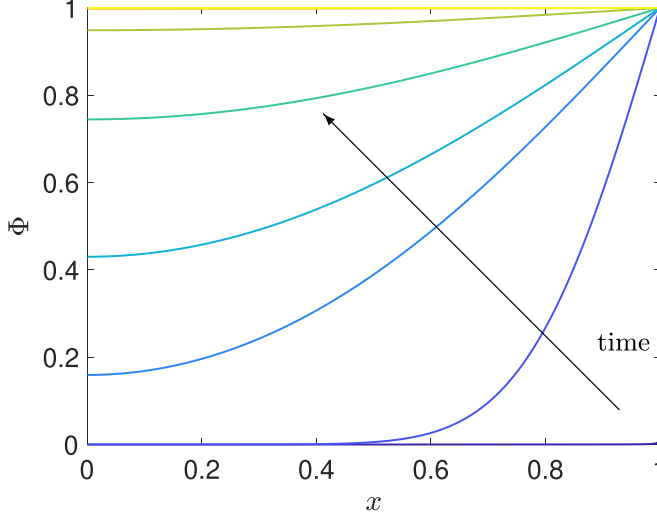


FIG. 6. Linearized solution for the porosity, Φ , given by Eq. (51) at different times $t = 0, 0.1, 1, 24, 8, 16, 32$, with $\phi_d = 0.8$, and $G = 10^{-4}$.

IV. ADHERED GELS

In this section, we turn our attention to gels which are fully adhered at both walls, and focus on the case of a layer of gel with aspect ratio $\epsilon \ll 1$. See Fig. 7. The adhesion boundary condition $\xi = 0$ means that swelling is no longer uniaxial and so we model the gel as two-dimensional in the (x, z) plane (i.e., assuming symmetry in the y direction) with length $2\tilde{L}$ and height $2\epsilon\tilde{L}$. As before, the gel is initially rectangular and in a relatively dry state, with constant porosity $\phi = \phi_d$, and is submerged in water at $t = 0$ and allowed to swell. The inverse deformation gradient tensor is given by

$$\mathbf{F}^{-1} = \begin{pmatrix} 1 - \frac{\partial \xi}{\partial x} & -\frac{\partial \eta}{\partial x} \\ -\frac{\partial \xi}{\partial z} & 1 - \frac{\partial \eta}{\partial z} \end{pmatrix}, \quad (55)$$

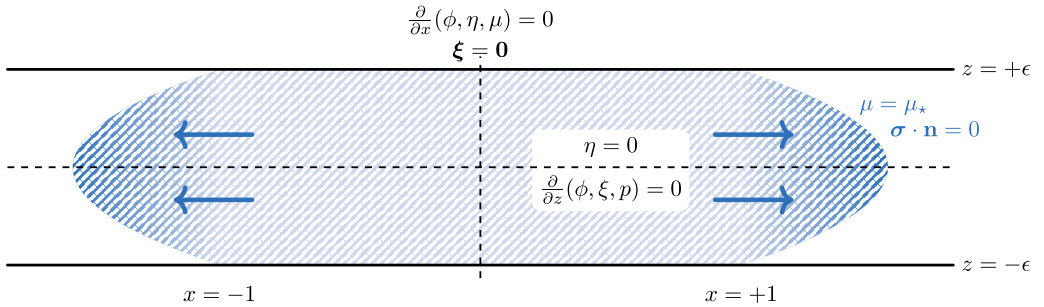


FIG. 7. Schematic of a 2D hydrogel with aspect ratio $\epsilon \ll 1$ swelling in water while confined between two walls. The problem has two lines of symmetry at $x = 0$ and $z = 0$, which dictates that displacement and gradients in the horizontal and vertical directions respectively are zero there. At the interface between the gel and the water, the chemical potential μ and stress $\sigma \cdot \hat{\mathbf{n}}$ must be zero [15]. The gel is fully adhered to the walls so $\xi = 0$ there.

which yields the Jacobian

$$J^{-1} = \frac{1 - \phi}{1 - \phi_d} = \left(1 - \frac{\partial \xi}{\partial x}\right) \left(1 - \frac{\partial \eta}{\partial z}\right) - \frac{\partial \xi}{\partial z} \frac{\partial \eta}{\partial x}, \quad (56)$$

and hence computing the effective stress tensor using Eq. (22), we have

$$\boldsymbol{\sigma}' = G \begin{pmatrix} J \left[\left(1 - \frac{\partial \eta}{\partial z}\right)^2 + \left(\frac{\partial \eta}{\partial x}\right)^2 \right] - J^{-1} & J \left[\frac{\partial \xi}{\partial z} \left(1 - \frac{\partial \eta}{\partial z}\right) + \frac{\partial \eta}{\partial x} \left(1 - \frac{\partial \xi}{\partial x}\right) \right] \\ J \left[\frac{\partial \xi}{\partial z} \left(1 - \frac{\partial \eta}{\partial z}\right) + \frac{\partial \eta}{\partial x} \left(1 - \frac{\partial \xi}{\partial x}\right) \right] & J \left[\left(1 - \frac{\partial \xi}{\partial x}\right)^2 + \left(\frac{\partial \xi}{\partial z}\right)^2 \right] - J^{-1} \end{pmatrix}. \quad (57)$$

The momentum equation (13) then yields the balances

$$\begin{aligned} \frac{\partial p}{\partial x} = G & \left[\frac{\partial}{\partial x} \left(J \left[\left(1 - \frac{\partial \eta}{\partial z}\right)^2 + \left(\frac{\partial \eta}{\partial x}\right)^2 \right] - J^{-1} \right) \right. \\ & \left. + \frac{\partial}{\partial z} \left(J \left[\frac{\partial \xi}{\partial z} \left(1 - \frac{\partial \eta}{\partial z}\right) + \frac{\partial \eta}{\partial x} \left(1 - \frac{\partial \xi}{\partial x}\right) \right] \right) \right], \end{aligned} \quad (58)$$

and

$$\begin{aligned} \frac{\partial p}{\partial z} = G & \left[\frac{\partial}{\partial x} \left(J \left[\frac{\partial \xi}{\partial z} \left(1 - \frac{\partial \eta}{\partial z}\right) + \frac{\partial \eta}{\partial x} \left(1 - \frac{\partial \xi}{\partial x}\right) \right] \right) \right. \\ & \left. + \frac{\partial}{\partial z} \left(J \left[\left(1 - \frac{\partial \xi}{\partial x}\right)^2 + \left(\frac{\partial \xi}{\partial z}\right)^2 \right] - J^{-1} \right) \right]. \end{aligned} \quad (59)$$

The momentum equations (75) and (76) are coupled to the advection-diffusion equation (19), the form of which is unchanged.

To close the problem, we require boundary conditions. We formulate the problem within the bottom right quadrant in Fig. 7, with the knowledge that the solutions in the three remaining quadrants can be deduced by symmetry. The boundary conditions are thus

$$\xi = 0, \quad \frac{\partial}{\partial x}(\phi, \eta, \mu) = 0 \quad \text{at } x = 0, \quad (60)$$

$$\eta = 0, \quad \frac{\partial}{\partial z}(\phi, \xi, \mu) = 0 \quad \text{at } z = 0, \quad (61)$$

$$\mu = \mu_\star, \quad \boldsymbol{\sigma} \cdot \hat{\mathbf{n}} = 0 \quad \text{on } S, \quad (62)$$

where $\mu_\star$ is the ambient chemical potential in the surrounding fluid, and S defines the free surface between the gel and the water.

As mentioned previously, we assume the gel is initially rectangular and in a relaxed state, so the initial conditions are

$$\phi(\mathbf{x}, 0) = \phi_d, \quad \text{and} \quad (63)$$

$$\boldsymbol{\xi}(\mathbf{x}, 0) = \mathbf{0}. \quad (64)$$

In this section, we consider the case of fully adhered hydrogels, and hence apply the wall boundary condition

$$\boldsymbol{\xi} = \mathbf{0} \quad \text{at } z = -\epsilon. \quad (65)$$

The system given by Eqs. (19), (56), and (60)–(65), (75), and (76) can then be simplified as follows.

We work in scaled coordinates $(x, z) = (x, \epsilon Z)$. By geometry, we also have $(\xi, \eta) = (\xi, \epsilon \eta_1)$. The Jacobian is then given by

$$J^{-1} = \frac{1 - \phi}{1 - \phi_d} = \left(1 - \frac{\partial \xi}{\partial x}\right) \left(1 - \frac{\partial \eta_1}{\partial Z}\right) - \frac{\partial \xi}{\partial Z} \frac{\partial \eta_1}{\partial x}, \quad (66)$$

while the momentum equations reduce to

$$\frac{\partial p}{\partial x} = \frac{G}{\epsilon^2} \frac{\partial}{\partial Z} \left[J \frac{\partial \xi}{\partial Z} \left(1 - \frac{\partial \eta_1}{\partial Z}\right) \right] + O(G), \quad (67)$$

$$\frac{\partial p}{\partial Z} = \frac{G}{\epsilon^2} \frac{\partial}{\partial Z} \left[J \left(\frac{\partial \xi}{\partial Z} \right)^2 \right] + O(G). \quad (68)$$

We see then that we expect pressure gradients to be of order G/ϵ^2 . We note that at equilibrium we must have $\mu = \mu_\star$ everywhere, since chemical potential gradients always induce a flow, and hence

$$\nabla p = \Pi'(\phi) \nabla \phi = O(G/\epsilon^2) \quad (\text{at equilibrium}). \quad (69)$$

Two distinct regimes of thin confined gel swelling thus arise:

(1) If $G/\epsilon^2 \gg 1$, then the balance (69) necessitates large ϕ gradients. Since $\phi_d < \phi < 1$ everywhere, this is only possible by introducing a small length scale into the problem; this regime corresponds to “incomplete swelling,” where the porosity variation is confined to a small region near the edge.

(2) Otherwise, if $G/\epsilon^2 \lesssim 1$, then we have $\nabla \phi$ being of order unity or smaller at equilibrium, corresponding to “complete swelling.”

We will investigate the gel dynamics in each of these limits in turn in Secs. IV A and IV B.

Because the fully adhered boundary condition severely constrains the swelling, we will focus on the solutions in each regime in the case of a small volumetric swelling ratio (though we comment briefly on the nonlinear case in Sec. IV C). We thus expand

$$J = 1 + \delta J_1 + \dots, \quad (70)$$

where $\delta \ll 1$, which we will see enables the problem to be solved analytically. The scalings differ according to the size of G/ϵ^2 , so we treat each case separately below. Since J and ϕ are related by Eq. (56), this correspondingly means there will only be $O(\delta)$ variations in porosity.

A. Incomplete swelling, $G/\epsilon^2 \gg 1$

In the case of incomplete swelling, to balance Eq. (69), it must be the case that the water only permeates a short distance $L = \epsilon/\sqrt{G}$. Hence, we work in rescaled coordinates $(X, Z) = ((x - 1)/L, z/\epsilon)$. Note that since we assume $G \ll 1$ that $\epsilon \ll L$. Let $\xi = \delta L \hat{\xi}$ and $\eta = \delta \epsilon \hat{\eta}$. Then by Eq. (56),

$$J^{-1} = \frac{1 - \phi}{1 - \phi_d} = 1 - \delta \left(\frac{\partial \hat{\xi}}{\partial X} + \frac{\partial \hat{\eta}}{\partial Z} \right) + O(\delta^2). \quad (71)$$

As a result, we scale

$$\phi = \phi_d + \delta(1 - \phi_d)\Phi, \quad \text{where} \quad \Phi = \frac{\partial \hat{\xi}}{\partial X} + \frac{\partial \hat{\eta}}{\partial Z} + O(\delta). \quad (72)$$

Since the osmotic pressure $\Pi(\phi)$ is a function of only ϕ , we expect pressure gradients to scale with ϕ gradients. Hence, we scale $p = \delta P$ and $\mu = \mu_\star + \delta \hat{\mu}$. The diffusion Eq. (19) then demonstrates at leading order that $\hat{\mu}$ is independent of Z and thus we expand

$$\hat{\mu} = \hat{\mu}_0(X) + G \hat{\mu}_1(X, Z) + \dots, \quad (73)$$

to obtain the leading order balance

$$\frac{\partial \Phi}{\partial T} = k_d \left(\frac{\partial^2 \hat{\mu}_0}{\partial X^2} + \frac{\partial^2 \hat{\mu}_1}{\partial Z^2} \right), \quad (74)$$

where we have defined the scaled time coordinate $T = t/L^2$ and $k_d = k(\phi_d)$ is the constant permeability.

From Eqs. (67) and (68) we obtain the momentum balance

$$\frac{\partial P}{\partial X} = \frac{\partial^2 \hat{\xi}}{\partial Z^2} + O(G), \quad (75)$$

$$\frac{\partial P}{\partial Z} = O(\delta + G), \quad (76)$$

where we have made no assumption on the relative sizes of δ and G . Thus P is independent of Z at leading order, and expanding $P = P_0(X) + O(\delta + G)$ we obtain

$$\hat{\xi} = \frac{\partial P_0}{\partial X} (Z^2 - 1), \quad (77)$$

where we have applied the boundary conditions $\hat{\xi} = 0$ at $Z = -1$ and $\partial \hat{\xi} / \partial Z = 0$ at $Z = 0$.

Since by Eqs. (73) and (76), we have that both $\hat{\mu}$ and P are independent of Z at leading order, we must have that Π and hence Φ are also independent of Z . Hence, we may expand $\Phi = \Phi_0(X) + O(\delta + G)$. As a result, integrating Eq. (71) yields for the vertical displacement

$$\eta = \Phi_0 Z - \frac{1}{2} \frac{\partial^2 P_0}{\partial X^2} \left(\frac{1}{3} Z^3 - Z \right). \quad (78)$$

We then require $\eta = 0$ at $Z = \pm 1$, which yields

$$0 = \Phi_0 + \frac{1}{3} \Pi'(\phi_d)(1 - \phi_d) \frac{\partial^2 \Phi_0}{\partial X^2} + \frac{1}{3} \frac{\partial^2 \hat{\mu}_0}{\partial X^2}. \quad (79)$$

Returning to the diffusion equation (74), we see that since $\hat{\mu}_0$ and Φ_0 are independent of Z at leading order, by averaging the equation in Z we obtain a one-dimensional equivalent,

$$\frac{\partial \Phi_0}{\partial T} = k_d \frac{\partial^2 \hat{\mu}_0}{\partial X^2}, \quad (80)$$

and thus, combining this with Eq. (79), we have that Φ_0 satisfies the reaction diffusion equation

$$\frac{\partial \Phi_0}{\partial T} = D \frac{\partial^2 \Phi_0}{\partial X^2} - 3k_d \Phi_0, \quad (81)$$

where $D = -k_d \Pi'(\phi_d)(1 - \phi_d)$.

We now require appropriate boundary conditions on Φ_0 . The center of the gel remains fully dry, and so we have

$$\Phi_0 \rightarrow 0 \quad \text{as} \quad X \rightarrow -\infty. \quad (82)$$

We denote the free surface S by $x = 1 + \delta La(Z)$ where $\hat{\xi}(1 + \delta a) = a$. Taylor expanding, the stress boundary condition (62) states that, at leading order,

$$P = 0 \quad \text{at} \quad X = 0. \quad (83)$$

Since we also have fixed chemical potential at the boundary, we must have

$$\Phi_0 = \Phi_w \quad \text{at} \quad X = 0, \quad (84)$$

where $\Pi(\Phi_w) = -\mu_\star$. We also have the initial condition,

$$\Phi_0 = 0 \quad \text{at} \quad T = 0. \quad (85)$$

Equations (81), (82), (84), and (85) define a linear boundary value problem that can be solved analytically by separation of variables and the method of images (substituting $\Phi = e^{-3kT} \Psi$ reduces it to the heat equation). The solution is

$$\Phi_0 = \Phi_w e^{X\sqrt{3k_d/D}} + \frac{\Phi_w}{2} \left[e^{-X\sqrt{3k_d/D}} \operatorname{erfc}\left(\frac{-X + 2\sqrt{3k_d DT}}{2\sqrt{DT}}\right) - e^{X\sqrt{3k_d/D}} \operatorname{erfc}\left(\frac{X + 2\sqrt{3k_d DT}}{2\sqrt{DT}}\right) \right], \quad (86)$$

where the first term gives the steady-state solution, $\Phi_0 = \Phi_w e^{X\sqrt{3k_d/D}}$. This is a full analytic solution which can be used to obtain the leading order displacement and stress fields exactly. The results are shown in Fig. 8.

1. Force exerted on the walls

The force exerted on the walls is given by

$$\mathbf{F}(t) = \delta \int_{-1}^1 P_0(x, t) \hat{\mathbf{z}} dx + O(\delta^2 + \delta\sqrt{G}). \quad (87)$$

Since the pressure integral is dominated by the constant contribution from the bulk of the gel, at equilibrium,

$$p = \mu_\star + \Pi(\phi_d) = -\Pi(\phi_w) + \Pi(\phi_d) = -\delta(1 - \phi_d)\Pi'(\phi_d)\Phi_w + \dots \quad (88)$$

in the bulk, and hence

$$\mathbf{F}_\infty = -2\delta(1 - \phi_d)\Pi'(\phi_d)\Phi_w \hat{\mathbf{z}}. \quad (89)$$

Note that since this is $O(\delta)$, the swelling ratio as opposed to $O(G)$ as found in Sec. III C for nonadhered gel swelling, we expect the force in this case to be significantly larger provided $\delta \gg G$, even though this solution assumes small displacement. The time evolution of the force is displayed in Fig. 9.

B. Complete swelling, $G/\epsilon^2 \lesssim 1$

We now consider the case $G/\epsilon^2 \lesssim 1$; we will begin by treating G/ϵ^2 as an order-unity parameter. We thus set $L = 1$ in the above derivation, and scale $p = G\delta/\epsilon^2 P$ to obtain the the same momentum balance as in the previous case, Eqs. (75) and (76). The vertical derivatives are still the dominant terms in the diffusion equation, and so we expand

$$\hat{\mu} = \hat{\mu}_0(x) + \epsilon^2 \hat{\mu}_1(x, Z) + \dots, \quad (90)$$

as before to obtain the diffusion equation (74) which we again may average in Z to obtain the one-dimensional form (80). The analysis then follows as before, where integrating Eq. (71) and applying the boundary conditions on η now yields

$$\Phi_0 + \frac{1}{3} \frac{\partial^2 P_0}{\partial x^2} = 0. \quad (91)$$

Since $\mu = -\Pi + (G\delta/\epsilon^2)P$, Eq. (80) becomes

$$\frac{\partial \Phi_0}{\partial t} = D \frac{\partial^2 \Phi_0}{\partial x^2} - \frac{3k_d G}{\epsilon^2} \Phi_0, \quad (92)$$

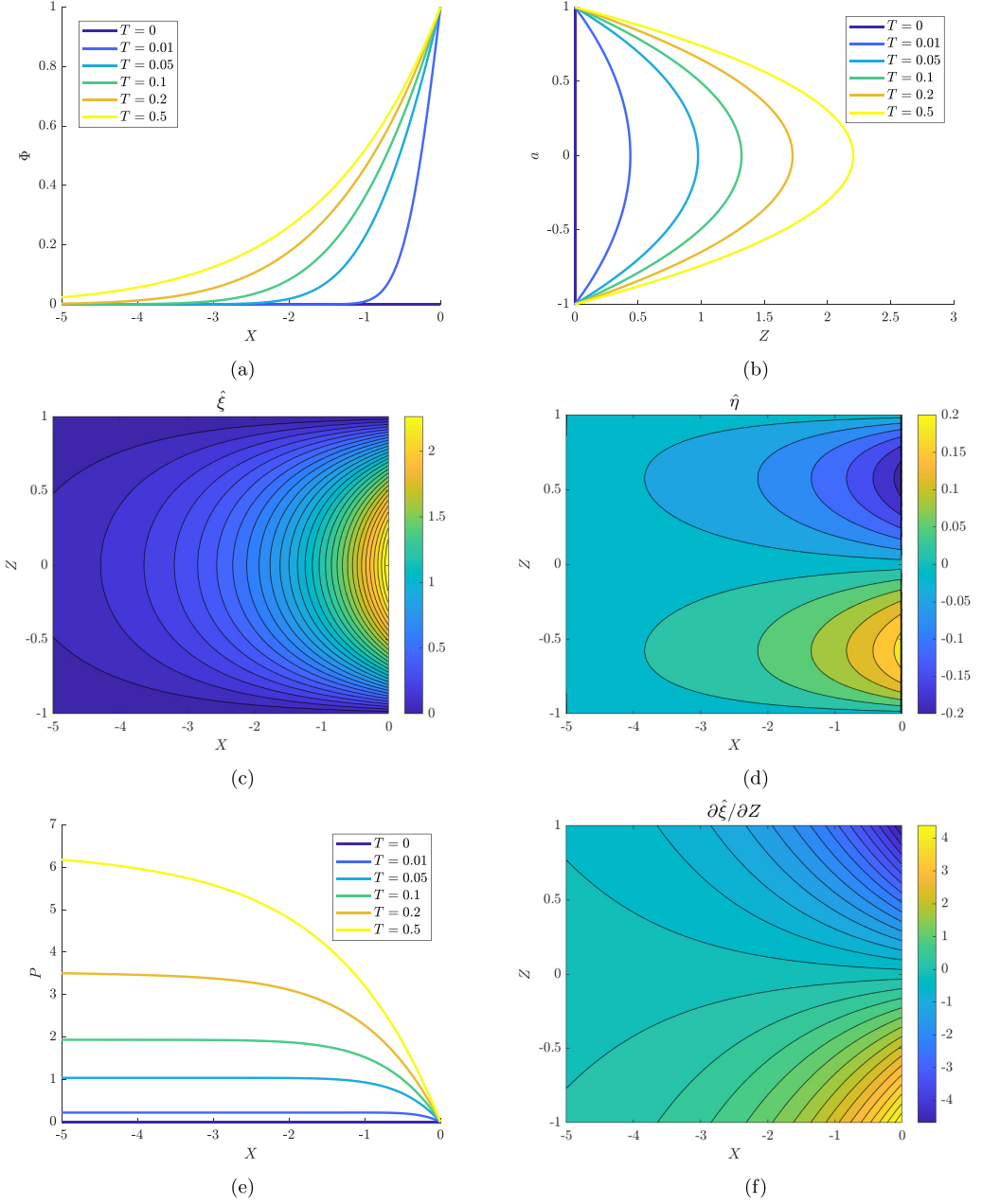


FIG. 8. Top: The time evolution of scaled porosity Φ_0 and shape a . Middle: Contour plots of (c) $\hat{\xi}$ and (d) $\hat{\eta}$. Bottom: (e) time evolution of pressure p , and (f) contour plot of $\partial \hat{\xi} / \partial Z$ at equilibrium (related to tangential stress by scale factor $\sqrt{G\delta}$). Parameter values: $\phi_d = 0.1$, $k_d = 1$, $\Phi_w = 1$, $\chi = 0.4$.

which can be solved analytically by the same methods as before. The boundary condition at the free surface (84) is unchanged, while along the center line of the gel we have

$$\frac{\partial \Phi_0}{\partial x} = 0 \quad \text{at } x = 0 \quad (93)$$

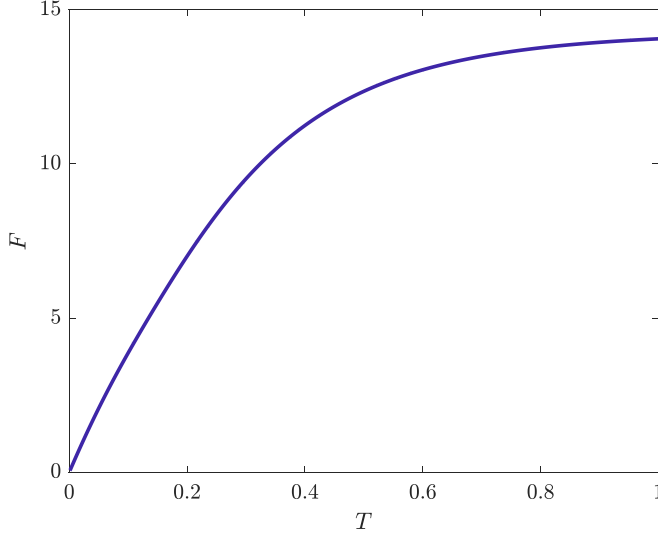


FIG. 9. The time evolution of the scaled force F exerted on the upper wall by the gel, where $\mathbf{F} = \delta F \hat{\mathbf{z}}$. Parameter values: $\phi_d = 0.1$, $k_d = 1$, $\Phi_w = 1$, $\chi = 0.4$.

by symmetry. Since this is of the same form as Eq. (86), this can be solved by separation of variables to obtain

$$\Phi_0 = \frac{\Phi_w \cosh\left(\frac{x\sqrt{3k_d G}}{\epsilon\sqrt{D}}\right)}{\cosh\left(\frac{\sqrt{3Gk_d}}{\epsilon\sqrt{D}}\right)} + \Phi_w e^{-3k_d Gt/\epsilon^2} \sum_{n=0}^{\infty} \frac{(-1)^{n+1} (2n+1)\pi}{\frac{(2n+1)^2 \pi^2}{4} + \frac{3Gk_d}{\epsilon^2 D}} e^{-(2n+1)^2 \pi^2 D t/4} \cos\left[\frac{(2n+1)\pi x}{2}\right]. \quad (94)$$

The above solution is valid provided $G/\epsilon^2 \gg \delta$. In the case $G/\epsilon^2 \ll \delta$, the G/ϵ^2 term will only enter the dynamics at late times once $\Phi = \Phi_w + O(G/\epsilon^2)$, and until this point the leading order solution simply obeys the heat equation with diffusion coefficient D , which can be obtained by setting $G/\epsilon^2 = 0$ in Eq. (94).

We display the results for the complete swelling case with $G/\epsilon^2 = 1$ and the same parameters as in the incomplete results in Figs. 11 and 10. Note that the scalings are different across the two cases, so although it appears that there is smaller displacement in Fig. 10 than Fig. 8, the true displacement in the latter case is smaller by a factor of $L \ll 1$. The porosity and (since we have chosen $G/\epsilon^2 = 1$) the pressures and forces are however directly comparable across the two cases.

1. Force exerted on the walls

The force exerted on the wall in this case is given by

$$\mathbf{F}(t) = \frac{G\delta}{\epsilon^2} \int_{-1}^1 P_0(x, t) \hat{\mathbf{z}} dx = \frac{6\delta G}{\epsilon^2} \int_0^1 \int_{x'}^1 \int_0^{x''} \Phi_0 \hat{\mathbf{z}} dx dx' dx''. \quad (95)$$

At equilibrium, this can be evaluated as

$$\mathbf{F}_{\infty} = -2\delta \Pi'(\phi_d)(1 - \phi_d)\Phi_w \left(1 - \frac{\epsilon\sqrt{D}}{\sqrt{3k_d G}} \tanh\left(\frac{\sqrt{3k_d G}}{\epsilon D}\right)\right) \hat{\mathbf{z}}. \quad (96)$$

Note that in the limit of $G/\epsilon^2 \gg 1$ we obtain the incomplete swelling result, Eq. (89) (since $\tanh(x)/x \rightarrow 0$ as $x \rightarrow \infty$). In the limit $G/\epsilon^2 \ll 1$, we can Taylor expand to

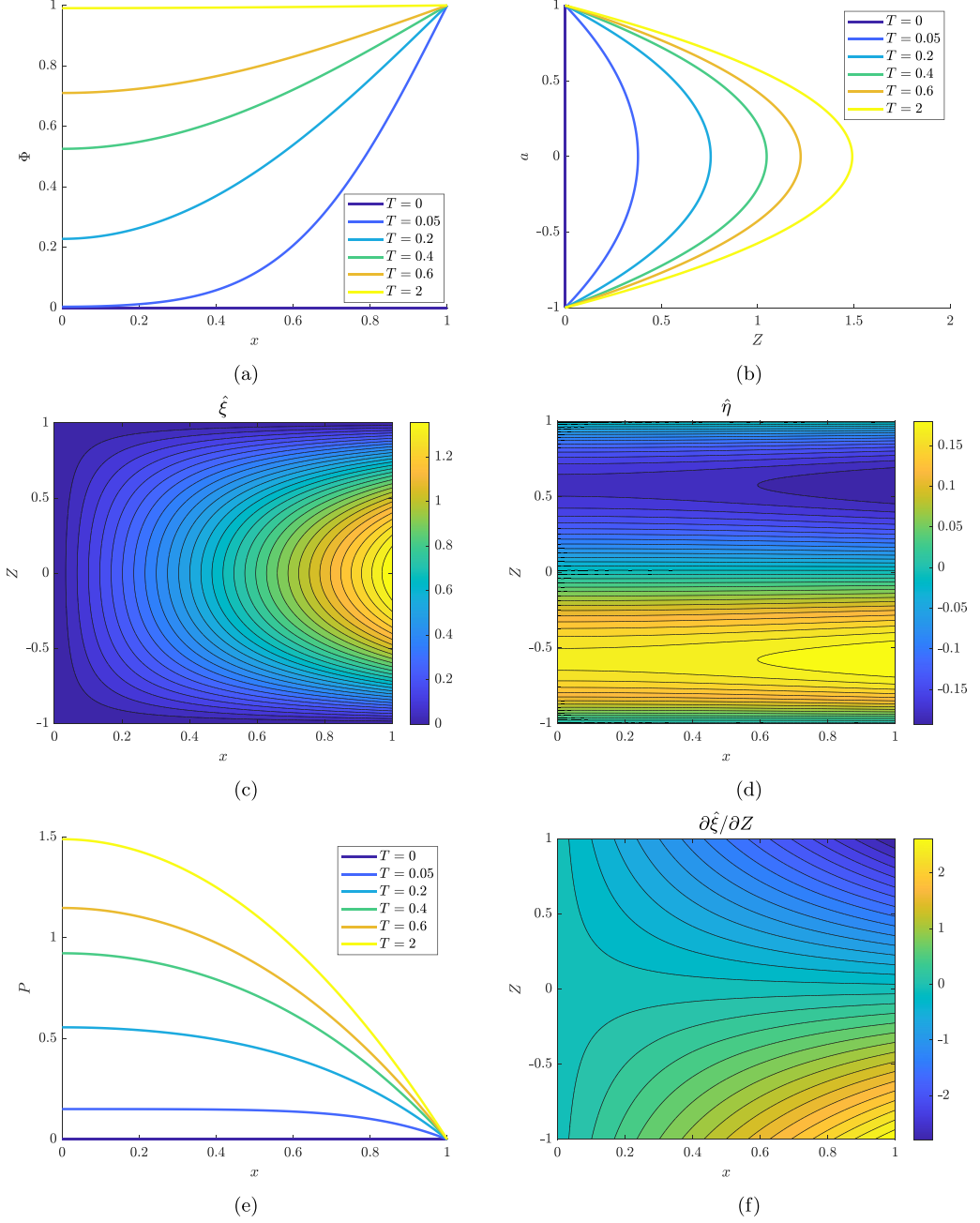


FIG. 10. Top: The time evolution of scaled porosity Φ_0 and shape a . Middle: Contour plots of (c) $\hat{\xi}$ and (d) $\hat{\eta}$. Bottom: (e) time evolution of pressure p , and (f) contour plot of $\partial \hat{\xi} / \partial Z$ at equilibrium (related to tangential stress by scale factor $\sqrt{G\delta}$). Parameter values: $\phi_d = 0.1$, $k_d = 1$, $\Phi_w = 1$, $\chi = 0.4$, $G/\epsilon^2 = 1$.

obtain

$$\mathbf{F}_\infty = \left(-\frac{2\delta\Phi_w G}{\epsilon^2} + O(G^2/\epsilon^4) \right) \hat{\mathbf{z}}, \quad (97)$$

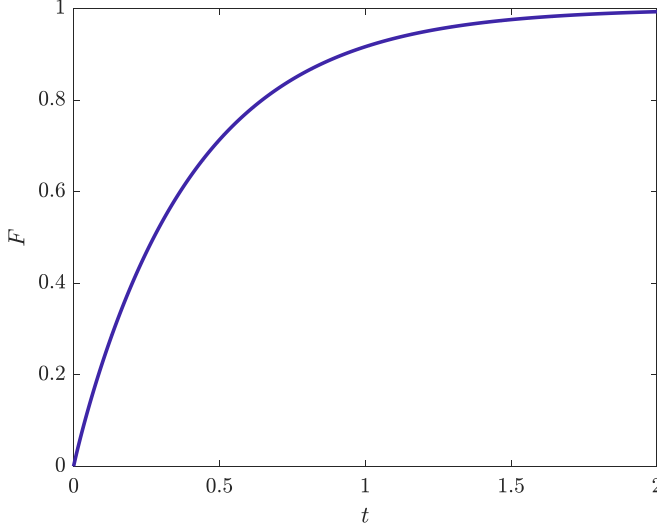


FIG. 11. The time evolution of the scaled force F exerted on the upper wall by the gel, where $\mathbf{F} = G\delta/\epsilon^2 F \hat{\mathbf{z}}$. Parameter values: $\phi_d = 0.1$, $k_d = 1$, $\Phi_w = 1$, $\chi = 0.4$, $G/\epsilon^2 = 1$.

since $D/k_d = \Pi'(\phi_d)(1 - \phi_d)$. We see in Fig. 11 that the force exerted in our complete case is around 10 times smaller than that exerted in the incomplete case, even though we selected $G/\epsilon^2 = 1$.

C. Toward nonlinear swelling

It is interesting to consider what happens to the incomplete and complete solutions as we increase δ , to provide information about the nonlinear problem. Most of our variables are not affected by the value of δ in their scaled form, with the exception of the reference position, which is $X_0 = X - \delta \hat{\xi}$ in the incomplete case and $x_0 = x - \delta \hat{\xi}$ in the complete case. We plot both in Fig. 12 for two values of δ . Contours of x_0 or X_0 physically represent the deformed position of initially vertical lines. As seen in Fig. 12, there is a bifurcation after which a saddle point appears. We can calculate this onset of this saddle point as occurring at $\delta = 2/(3\Phi_w)$ in both cases by equating the horizontal derivative of $\hat{\xi}$ to $1/\delta$. In the incomplete case, the saddle point forms at $X = 0$ (i.e., at the edge of the domain) and moves inward as δ increases, whereas in the complete case it may only form at the gel midpoint. While we expect that nonlinear terms would likely have become significant at smaller values of δ and the saddle point solutions are certainly invalid since the map from $\mathbf{x}_0$ to $\mathbf{x}$ is no longer bijective, it seems to suggest that as the gel swells sufficiently far from the walls it may ‘bulge’ rather than retain its parabolic shape (though this would depend on whether or not the walls continue beyond $-1 < x < 1$). If this happened, it would no longer be possible to express the free surface in the form $X = \delta a(Z)$, as this would not be a single-valued function of Z . Elastic bulging problems have received limited attention, but the phenomenon is known to induce large stresses in the bulge region and thus potentially cause damage to surroundings [37]. Since hydrogels are relatively soft materials however, they may be more likely to simply fracture; this has been observed experimentally by Plummer *et al.* [26].

V. DISCUSSION AND CONCLUSION

In this paper, we have developed and analysed models for the swelling of a hydrogel confined between two rigid plates. We considered two scenarios: (i) the hydrogel is allowed to slip against the confining walls, (ii) the gel is adhered to the walls. We used the large deformation poroelasticity framework of Ref. [32], but introduced the assumption that G (the ratio of shear modulus to osmotic

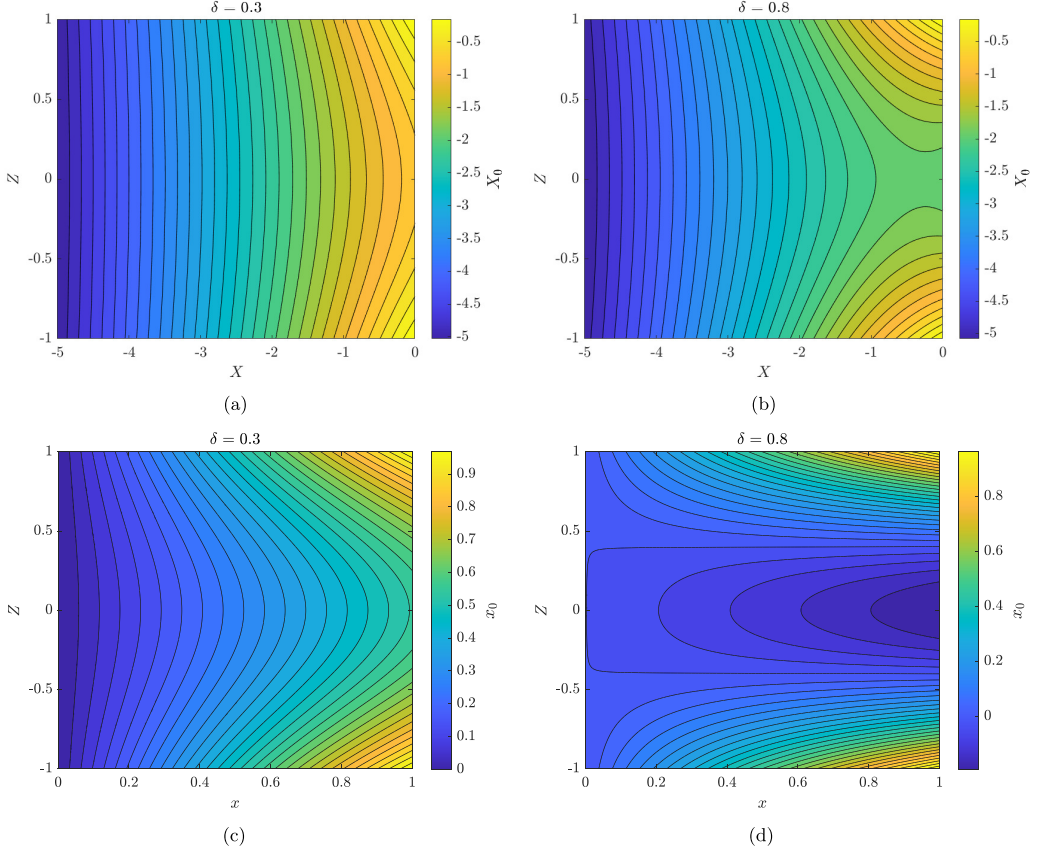


FIG. 12. Equilibrium contour plots of reference position $X_0 = X - \delta \hat{\xi}$ in the incomplete case (top) and $x_0 = x - \delta \hat{\xi}$ in the complete case (bottom) for (a), (c) $\delta = 0.3$ and (b), (d) $\delta = 0.8$. These represent the deformed position of initially vertical material lines. As δ increases, there is a bifurcation at $\delta = 2/3$ after which a saddle point appears, as seen in panels (b), (d). Parameter values: $\phi_d = 0.1$, $k_d = 1$, $\Phi_w = 1$, $\chi = 0.4$, and, in the complete case, $G/\epsilon^2 = 1$.

modulus) is a small parameter, based on experimental observations of G being 10^{-4} [2], which aided analytical progress.

In case (i), the swelling dynamics reduces to uniaxial deformation. This allows us to write the displacement, and elastic stresses as functions of the porosity which is governed by a single partial differential equation. We find that as the hydrogel swells, the internal pressure supplies a force on the confining walls. However, the resultant force is proportional to the size of the ratio between the shear modulus and the osmotic modulus G , which has been measured for typical hydrogels as being very small, approximately 10^{-4} . In case (ii), assuming small aspect ratio ϵ and small deformation, the gel swelling can be split into two regimes according to the size of G/ϵ^2 , which we term “incomplete” and “complete” swelling, both of which a solveable analytically. In incomplete swelling, the water only permeates a distance $L = \epsilon/\sqrt{G} \ll 1$. The force exerted on the walls in this case scales with δ , the perturbation of the volumetric swelling ratio from 1. In complete swelling, where $G/\epsilon^2 \ll 1$, the water permeates fully through the gel. The force in this case scales with $\delta G/\epsilon^2$, so smaller than the incomplete case, but providing $\delta/\epsilon^2 \geq 1$ it remains significantly larger than the slip case. While this paper focused on two-dimensional (2D) geometry, we anticipate the core insights would be generalizable to suitable three-dimensional geometries. The axisymmetric case is an obvious

example, which we expect to be qualitatively similar in the confined case and effectively identical in the adhered case (given the linearized analysis). It would also be expected to apply for cuboidal gels sufficiently far from the corners.

The findings have potentially significant implications for the design of hydrogel-based actuation devices. Adhesion increased the force exerted by the gel on its surroundings (potentially by orders of magnitude) but also may limit swelling. It is also worth noting that in the case of adhered gels, the gel exerts a significant tangential force on the wall as well as perpendicular. The magnitude of the increase in force provided by adhesion is perhaps surprising, especially given in the adhered case we assumed small deformation whereas in the slip case we did not. As discussed in Sec. IV C, while the equivalent nonlinear problem for adhered gel swelling is significantly more complicated, the linear results in the limit $\delta \rightarrow 1$ and the results of experiments [26] indicate that (assuming the walls do not continue beyond the length of the hydrogel) the hydrogel may form a “bubble” just outside the confined region and fracture at relatively modest swelling ratios. In practice, we might imagine that more realistic boundary conditions for confined and adhered gels involve a compromise between the two scenarios considered here, for example a finite adhesion energy or a slip condition with friction. It would be interesting to investigate the effect of these on the applied forces in future work; in the former case, we can envisage that the results of the adhered case might enable a ‘delamination condition’ to be derived, while for a partial slip condition a hybrid of the two solutions from this paper may apply.

Investigating what happens in the nonlinear limit of the adhered case is a natural further question arising from this work, which may be best achieved by finite element simulations. Furthermore, one might expect the eventual onset of wrinkling or buckling instabilities. While these are in principle permitted in our 2D adhered gel analysis, the focus on small displacement means they have not arisen, and the possibility may have been excluded from the confined-with-slip case by our focus on uniaxial swelling. The conditions of onset of such instabilities would be another interesting topic for further work. Other potential extensions to the work include considering other aspect ratios in the adhered case, and in both cases permitting the walls to move in response to the gel swelling. This could also be developed into detailed models of realistic hydrogel actuators by adding a model for the gel responsivity (as in Refs. [20,38]) which could then be paired with relevant environmental data. It may also be of interest, with for example subcutaneous wound-healing/drug delivery devices in mind, to couple the gel swelling to a surrounding soft material.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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