APS DFD 2022
Sunday 20th November, Session G11

A linear-elastic-nonlinear-swelling theory for hydrogels: displacements and differential swelling

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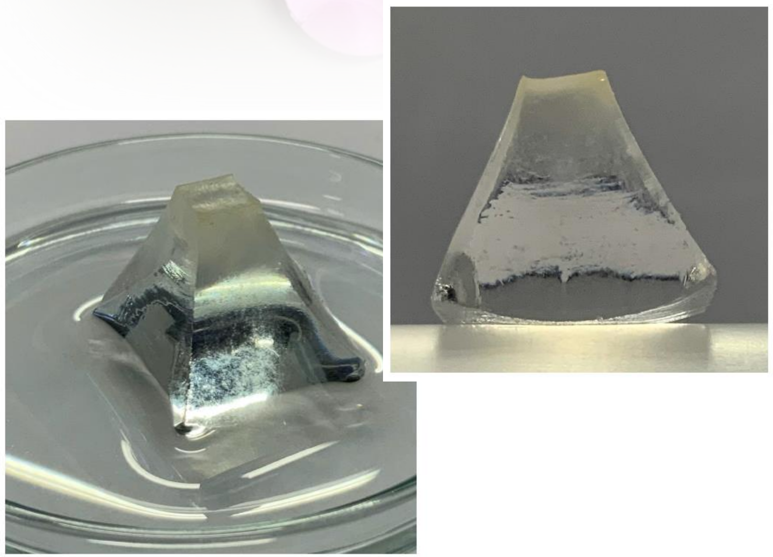

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LENS model for hydrogels

$$\frac{\partial \phi}{\partial t} + \boldsymbol{q} \cdot \boldsymbol{\nabla} \phi = \boldsymbol{\nabla} \cdot \left[\frac{k(\phi)}{\mu_l} \left\{ K(\phi) + \frac{4\mu_s(\phi)}{3} \left(\frac{\phi}{\phi_0} \right)^{1/3} \right\} \boldsymbol{\nabla} \phi \right]$$

Permeability

Fluid viscosity

Shear modulus

$$\underline{\underline{\boldsymbol{\sigma}}} = -[p + \Pi(\phi)]\underline{\underline{\boldsymbol{I}}} + 2\mu_s(\phi)\underline{\underline{\boldsymbol{\epsilon}}}$$

$$\underline{\underline{\boldsymbol{\epsilon}}} = \frac{1}{2}\left[\boldsymbol{\nabla}\boldsymbol{\xi} + (\boldsymbol{\nabla}\boldsymbol{\xi})^{\mathrm{T}}\right] - \left[1 - (\phi/\phi_0)^{1/3}\right]\underline{\underline{\boldsymbol{I}}}$$

Total (material) flux

$$\boldsymbol{q} = \left(\frac{\phi}{\phi_0} \right)^{1/3} \frac{\partial \boldsymbol{\xi}}{\partial t} - \frac{k(\phi)}{\mu_l} \boldsymbol{\nabla} p$$

POLYMER VELOCITY — FLUID VELOCITY

- Require boundary conditions on displacements, stresses and interstitial quantities

- But the first two require knowledge of the displacement field; we don't have this in our framework

Displacement formulation

- In incompressible linear elasticity, $\boldsymbol{\xi}$ satisfies the biharmonic equation

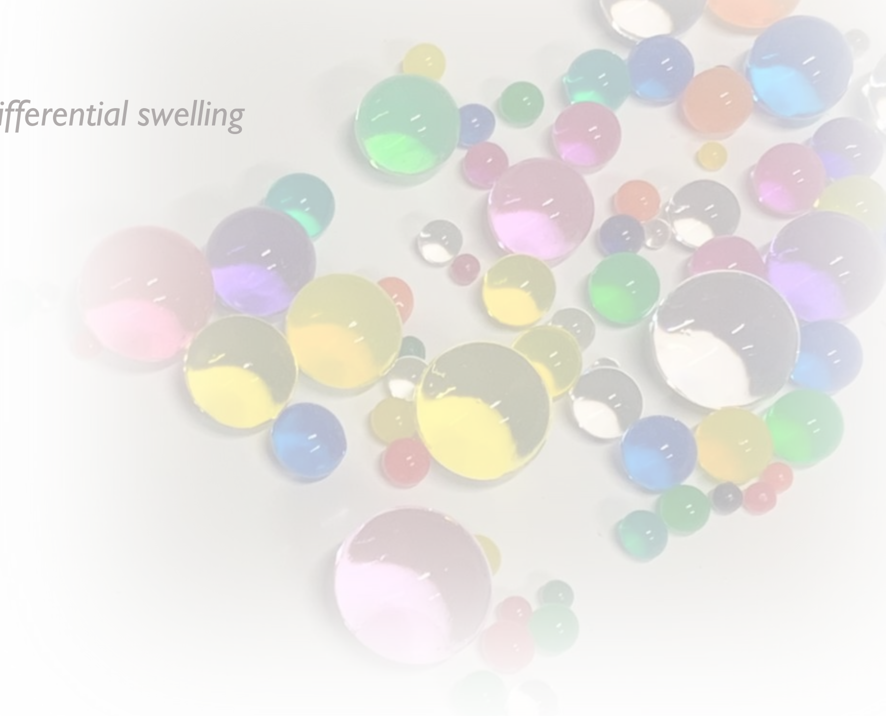
- Volumetric change → polymer fraction change and so

$$\boldsymbol{\nabla} \cdot \boldsymbol{\xi} = 3\left[1 - \left(\frac{\phi}{\phi_0}\right)^{1/3}\right]$$

- Combine this with Cauchy's momentum equation to find that $\mu_s \boldsymbol{\nabla} \cdot \boldsymbol{\epsilon}$ is given by $\boldsymbol{\nabla} P$ and therefore, taking curls,

$$\nabla^4 \boldsymbol{\xi} = -3 \boldsymbol{\nabla} \nabla^2 \left(\frac{\phi}{\phi_0}\right)^{1/3}$$

- Reduces to linear elasticity when polymer fraction is uniform
- Can be interpreted like classical plate theory – deviatoric deformation is forced due to gradients in curvature of surfaces of constant polymer fraction



Displacement formulation

$$\frac{\partial \phi}{\partial t} + \boldsymbol{q} \cdot \boldsymbol{\nabla} \phi = \boldsymbol{\nabla} \cdot \left[\frac{k(\phi)}{\mu_l} \left\{ K(\phi) + \frac{4\mu_s(\phi)}{3} \left(\frac{\phi}{\phi_0} \right)^{1/3} \right\} \boldsymbol{\nabla} \phi \right]$$

$$\boldsymbol{q} = \left(\frac{\phi}{\phi_0} \right)^{1/3} \frac{\partial \boldsymbol{\xi}}{\partial t} - \frac{k(\phi)}{\mu_l} \boldsymbol{\nabla} p$$

$$\boldsymbol{\nabla} \cdot \boldsymbol{\xi} = 3 \left[1 - \left(\frac{\phi}{\phi_0} \right)^{1/3} \right]$$

$$\nabla^4 \boldsymbol{\xi} = -3 \boldsymbol{\nabla} \nabla^2 \left(\frac{\phi}{\phi_0} \right)^{1/3}$$

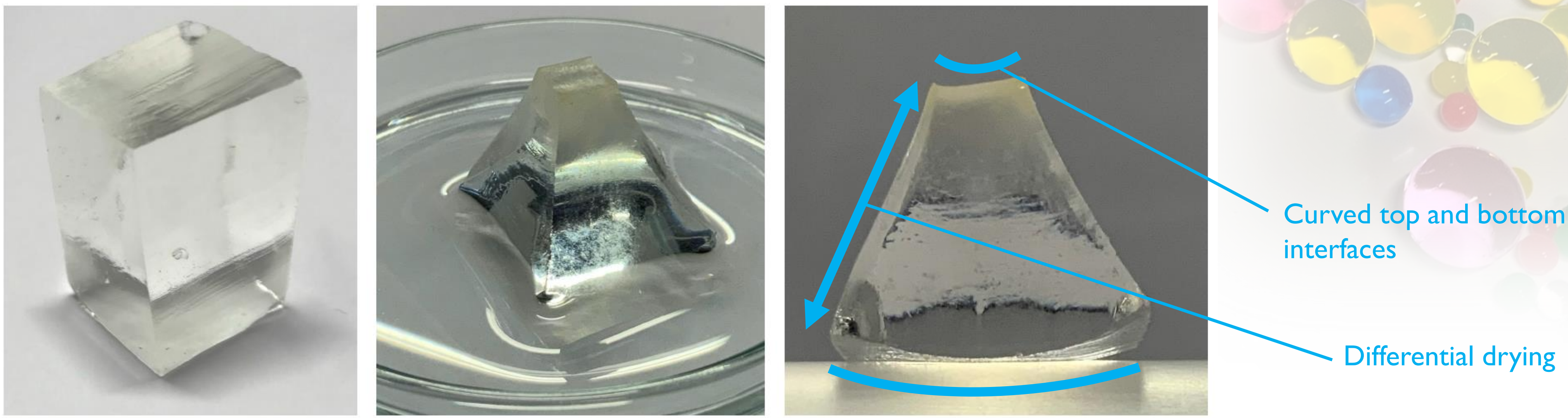
- Mechanical boundary conditions on stress and/or displacement
- Interstitial boundary conditions on pervadic pressure

$$\underline{\underline{\boldsymbol{\sigma}}} = -[p + \Pi(\phi)] \underline{\underline{\boldsymbol{I}}} + 2\mu_s(\phi) \underline{\underline{\boldsymbol{\epsilon}}}$$

$$\underline{\underline{\boldsymbol{\epsilon}}} = \frac{1}{2} \left[\boldsymbol{\nabla} \boldsymbol{\xi} + (\boldsymbol{\nabla} \boldsymbol{\xi})^{\mathrm{T}} \right] - \left[1 - (\phi/\phi_0)^{1/3} \right] \underline{\underline{\boldsymbol{I}}}$$

This provides a complete set of Galilean-invariant equations and boundary conditions to solve for the evolution of a gel in any geometry

Drying of slender cylinders



- Model as a cylinder of initial radius a_0 and height h_0 with its base immersed in water and sides open to the air to dry

- Describe drying through the use of a **fixed** evaporative flux from top and sides (e.g. in an environment of fixed humidity)

Boundary conditions

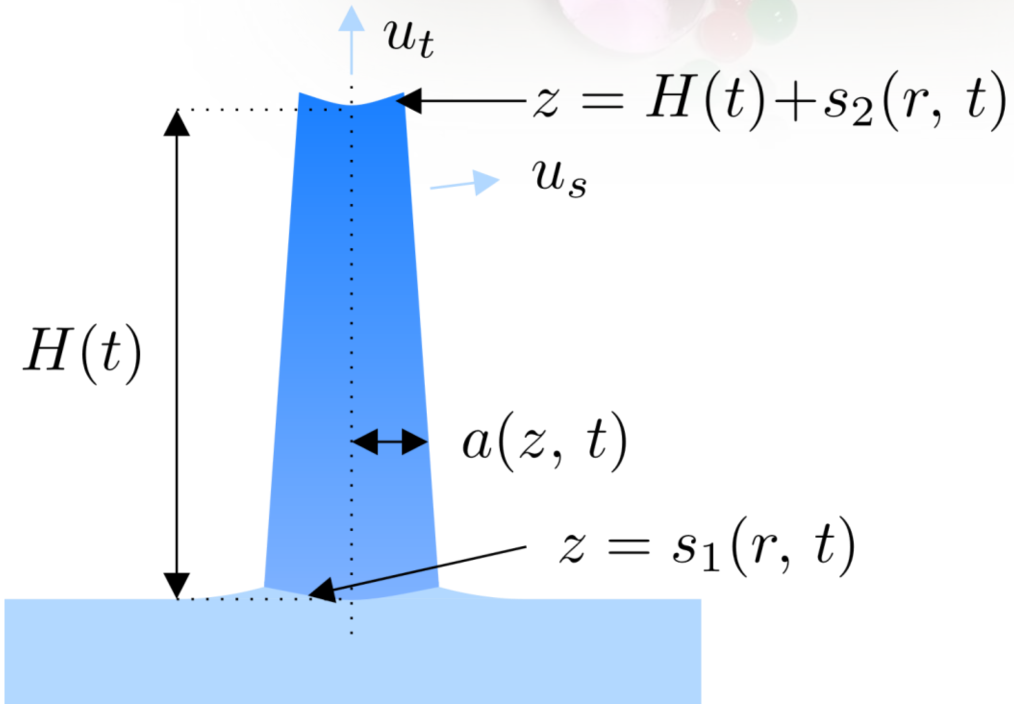
- On the base, no normal stress and continuity of pervadic pressure combine to imply that $\phi = \phi_0$ - the gel is fully-swollen.

- Stress boundary conditions on the sides give

$$\sigma_{rr} = \sigma_{rz} = 0 \qquad (r = a(z,\, t))$$

- Evaporative flux boundary conditions require, at leading order,

$$\hat{\boldsymbol{n}} \cdot \boldsymbol{\nabla} p = -\mu_l u_t / k(\phi) \qquad (z = H(t))$$
$$\hat{\boldsymbol{n}} \cdot \boldsymbol{\nabla} p = -\mu_l u_s / k(\phi) \qquad (r = a(z,\, t))$$



Slenderness and polymer fraction

- Requiring small deviatoric strain enforces small gradients in polymer fraction, and therefore, with

  $$\partial\phi/\partial z \sim \Delta\phi/h_0 \ll \Delta\phi/a_0$$

  so order-unity differences in polymer fraction can be supported along the axis, but not radially. This motivates taking

  $$\phi(r,\, z,\, t) = \phi_C(z,\, t) + \phi_1(r;\, z,\, t)$$

- Here, $\phi_1 \sim (a_0/h_0)\,\phi_C$ and separation of variables in the polymer-fraction evolution equation gives

  $$\phi_1(r;\, z,\, t) = f(z,\, t)r^2$$

Slenderness and polymer fraction

- Imposing the evaporative flux boundary condition and assuming constant material parameters K, μ_s and k implies that

$$\phi_1 = \frac{\phi_C}{2a}\left\{\frac{\mu_l \phi_C u_s}{k}\left[\frac{K\phi_C}{\phi_0} + \frac{4\mu_s}{3}\left(\frac{\phi_C}{\phi_0}\right)^{1/3}\right]^{-1} + \frac{1}{\phi_C}\frac{\partial \phi_C}{\partial z}\frac{\partial a}{\partial z}\right\} r^2$$

- Then,

$$\frac{\partial \phi_C}{\partial t} + q_z \frac{\partial \phi_C}{\partial z} = \frac{1}{a^2}\frac{\partial}{\partial z}\left[a^2 D(\phi_C)\frac{\partial \phi_C}{\partial z}\right] + \frac{2\phi_C u_s}{a}$$

$$q_z = \frac{D(\phi_C)}{\phi_C}\frac{\partial \phi_C}{\partial z} - \left(\frac{\phi_C}{\phi_0}\right)^{1/3}\int_0^z \frac{\partial}{\partial t}\left(\frac{\phi_C}{\phi_0}\right)^{1/3} \mathrm{d}z'$$

$$D(\phi_C) = \frac{k}{\mu_l}\left[\frac{K\phi_C}{\phi_0} + \frac{4\mu_s}{3}\left(\frac{\phi_C}{\phi_0}\right)^{1/3}\right]$$

$$\phi_C = 0 \qquad (z = 0)$$

$$\partial \phi_C/\partial z = \phi_C u_t / D(\phi_C) \qquad (z = H(t))$$

… but also need the shape of the gel at any given time

Displacements and cylinder shape

- Using the equation derived earlier for the displacements, the vertical and horizontal displacement fields can be deduced, giving the shape of the gel

$$a(z,\,t) = (\phi_C/\phi_0)^{-1/3} a_0$$

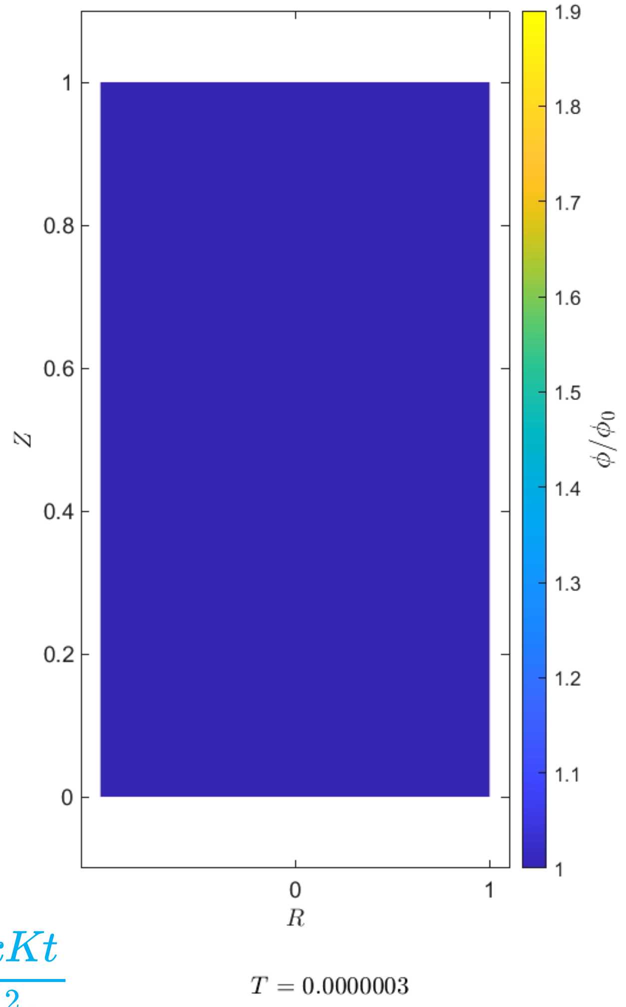
$$h_0 = \int_0^{H(t)} 1 - (\phi_C/\phi_0)^{1/3}\,\mathrm{d}z'$$

$$s_1(r,\,t) = \frac{r^2}{2}\frac{\partial}{\partial z}\left(\frac{\phi_C}{\phi_0}\right)^{1/3}\Bigg|_{z=0}$$

$$s_2(r,\,t) = \frac{r^2}{2}\frac{\partial}{\partial z}\left(\frac{\phi_C}{\phi_0}\right)^{1/3}\Bigg|_{z=H}$$

- The expression for the radius suggests isotropic contraction at fixed vertical position
- The height then follows from polymer conservation
- Differential drying creates the curved top and bottom interfaces

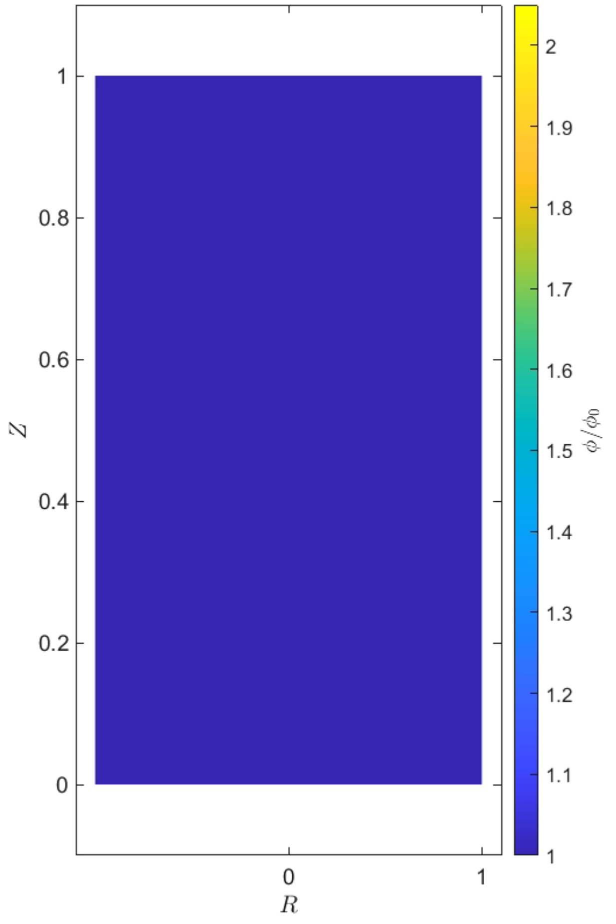
Drying from the top



$$T = \frac{kKt}{h_0^2 \mu_l}$$

- Curved top and bottom surfaces form due to differential drying
- Notice the small radial gradients $\partial\phi/\partial r < 0$ that arise from the need to impose no evaporation from the sides
- A steady state is reached where uptake of water from the base matches evaporative flux from the top

Drying from the sides



$T = 0.0000003$

- In this case, radial shrinkage dominates axial shrinkage, and at early times, the shrinkage is axially-uniform
- Here, $\partial\phi/\partial r > 0$ to drive flow radially owing to the imposed evaporation flux
- A different steady state is reached in this case; there are no vertical gradients in polymer fraction on the top surface so it remains flat

Conclusions

- Can deduce an expression for the displacement field, describing the shape of a hydrogel as it swells or dries
- We can use this result to find stresses throughout the gel, and the interstitial flows throughout
- Differential drying leads to small deviatoric strains which result in curved surfaces of constant polymer fraction

Webber, J. J., Etzold, M. A. and Worster, M. G. A linear-elastic-nonlinear-swelling theory for hydrogels. Part 2. Displacement formulation *(J. Fluid Mech., submitted)*

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NE/S007164/1 reference 2436164

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