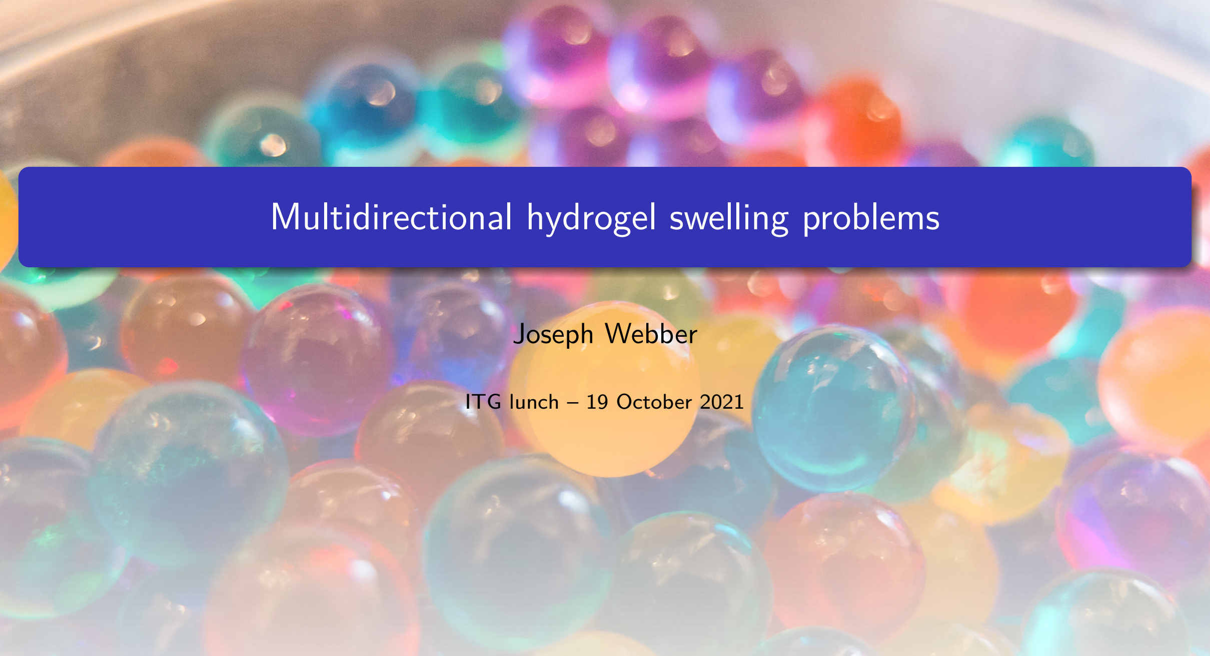

Multidirectional hydrogel swelling problems

Joseph Webber

ITG lunch – 19 October 2021

Outline

- Recap of existing models for hydrogel swelling and drying, and an introduction to the linear-elastic-nonlinear-swelling approach.

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- Displacement formulation – derivation and comparison with linear elasticity.
- Drying of a cylinder of gel:
  - Drying from the top, leveraging small deviatoric strains
  - Comparison with the classical theory of thin elastic plates
  - Full multidirectional problem with evaporation from the sides

The state of play – models for hydrogel swelling

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Fully-nonlinear models

Derive an energy-density function $\mathcal{W}$ with contributions from intermolecular interactions and stretching of polymer chains (Bertrand *et al.* 2016) or use Biot poroelasticity but with a fully-nonlinear constitutive relation for the stress (MacMinn *et al.* 2016)

Linear-elastic-nonlinear-swelling model

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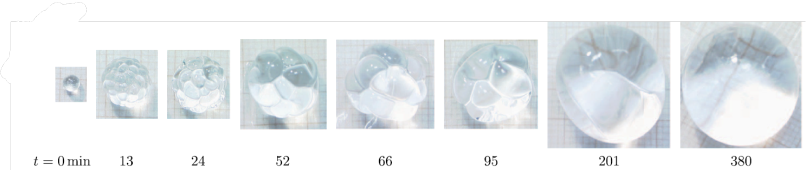

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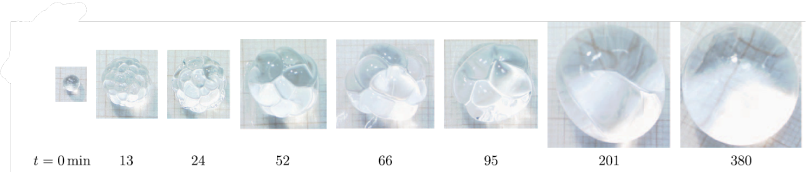

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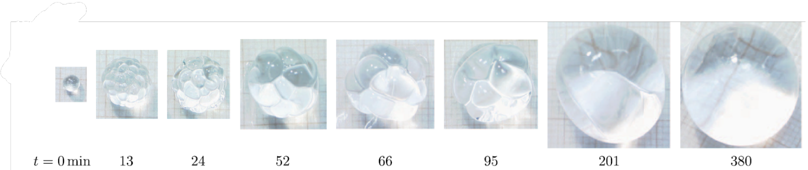
$$\boldsymbol{e} = \frac{1}{2}\left[\nabla\boldsymbol{\xi} + (\nabla\boldsymbol{\xi})^{\mathsf{T}}\right] = \left[1 - \left(\frac{\phi}{\phi_0}\right)^{1/n}\right]\boldsymbol{I} + \boldsymbol{\epsilon}.$$



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Linear-elastic-nonlinear-swelling model

These assumptions allow us to derive a constitutive relation

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

Here, p is pervadic pressure (Peppin *et al.* 2005), Π is osmotic pressure, $\mu_s(\phi)$ is shear modulus. Cauchy's momentum equation $\boldsymbol{\nabla} \cdot \boldsymbol{\sigma} = \mathbf{0}$ relates pressure gradients to matrix deformation, and then

$$\boldsymbol{u} = -\frac{k(\phi)}{\mu_l}\boldsymbol{\nabla} p$$

gives the interstitial flux. This is sufficient to derive an equation governing the time-evolution of polymer fraction in any given gel which satisfies our starting assumptions.

Using Cauchy,

$$\boldsymbol{\nabla} p = -\boldsymbol{\nabla} \Pi(\phi) + 2\boldsymbol{\nabla} \cdot \left[\mu_s(\phi)\boldsymbol{\epsilon}\right].$$

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$$\frac{\partial \phi}{\partial t} + \boldsymbol{q} \cdot \nabla \phi = \nabla \cdot (\phi \boldsymbol{u}) \, ,$$

Polymer transport equation

$$\frac{D_{\boldsymbol{q}}\phi}{Dt} = \frac{\partial \phi}{\partial t} + \boldsymbol{q} \cdot \nabla \phi = \nabla \cdot \left[\frac{\phi k(\phi)}{\mu_l} \left\{ \nabla \Pi(\phi) - 2\nabla \cdot [\mu_s(\phi)\boldsymbol{\epsilon}] \right\} \right]$$

Aside: gradients in polymer fraction

- Isotropic swelling corresponds to a uniform polymer fraction, since $\boldsymbol{e} = \left[1 - (\phi/\phi_0)^{1/n}\right] \boldsymbol{I}$.

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- Amongst other things, this allows us to rewrite $\boldsymbol{\nabla} \cdot [f(\phi)\boldsymbol{\epsilon}] = f(\phi)\boldsymbol{\nabla} \cdot \boldsymbol{\epsilon}$ at leading order, and therefore rephrase our conservation equation in terms of ϕ alone.

$$\boldsymbol{\nabla} \cdot \boldsymbol{\epsilon} = \frac{1}{2}\nabla^2\boldsymbol{\xi} + \left(1 - \frac{n}{2}\right)\boldsymbol{\nabla}\left(\frac{\phi}{\phi_0}\right)^{1/n} = \frac{1}{2}\nabla^2\boldsymbol{\xi}_{\text{dev}} + (1-n)\,\boldsymbol{\nabla}\left(\frac{\phi}{\phi_0}\right)^{1/n}.$$

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$$\frac{D_{\boldsymbol{q}}\phi}{Dt} = \frac{\partial \phi}{\partial t} + \boldsymbol{q} \cdot \nabla \phi = \boldsymbol{\nabla} \cdot \left[\frac{\phi k(\phi)}{\mu_l} \left\{ \boldsymbol{\nabla} \Pi(\phi) + 2\,(n-1)\,\mu_s(\phi) \boldsymbol{\nabla} \left(\frac{\phi}{\phi_0} \right)^{1/n} \right\} \right]$$

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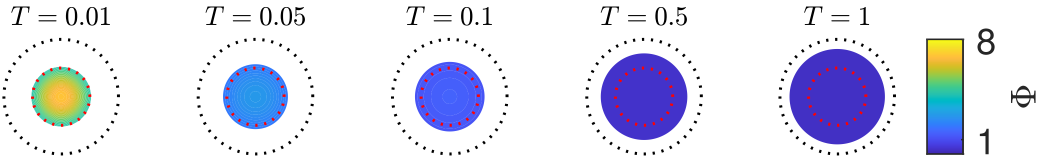
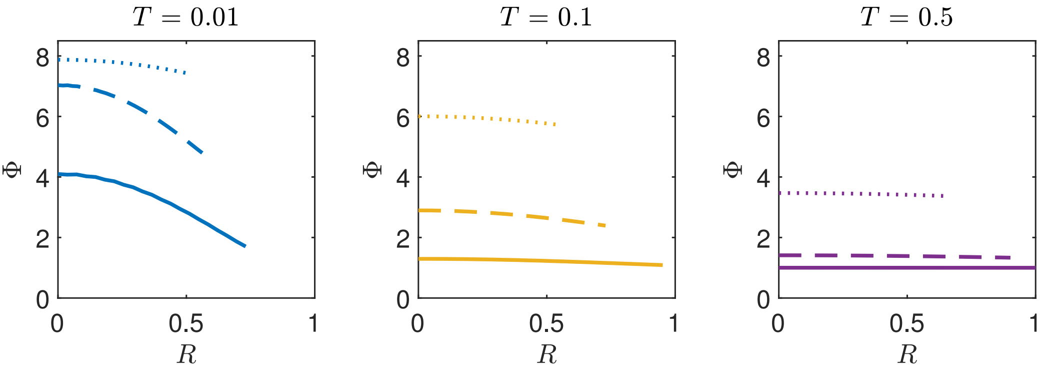
N.B. $\boldsymbol{q} \cdot \boldsymbol{\nabla}\phi$ can cause issues too! We have an expression for $\boldsymbol{u}$ but not $\boldsymbol{u_p}$ – scaling arguments clear this up.





Solid: $\mathcal{M} = 1$, dashed: $\mathcal{M} = 10$, dotted: $\mathcal{M} = 100$.

Determining the displacement field

- In a one-dimensional problem, can get $\boldsymbol{\xi}$ from ϕ – take the case of a sphere as an example, where $\boldsymbol{\xi} = \xi \hat{\boldsymbol{r}}$.

$$\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 \xi\right) = \left[1 - \left(\frac{\phi}{\phi_0}\right)^{1/3}\right] \quad \text{with} \quad \xi\big|_{r=0} = 0.$$

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- This allows us to fully-determine the shape of the gel as it swells, since its radius is given implicitly by $a(t) - a_0 = \xi\big|_{r=a(t)}$.
- In general, of course, we can't repeat this process for any three-dimensional swelling problem, so we need to exploit the specific geometry of a problem, alongside our assumption of small deviatoric strains, to determine this in more generality.

Learning from linear elastostatics

The displacement field in linear elastostatic problems can be seen to satisfy the biharmonic equation $\nabla^4 \boldsymbol{\xi} = \mathbf{0}$. This arises from Cauchy's momentum equation $\boldsymbol{\nabla} \cdot \boldsymbol{\sigma} = \mathbf{0}$, and taking curls.

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$$\boldsymbol{\nabla} \times \boldsymbol{\nabla} \cdot [\mu_s(\phi)\boldsymbol{\epsilon}] \approx \boldsymbol{\nabla} \times [\mu_s(\phi)\boldsymbol{\nabla} \cdot \boldsymbol{\epsilon}] \approx \mu_s(\phi)\boldsymbol{\nabla} \times \boldsymbol{\nabla} \cdot \boldsymbol{\epsilon} = \mathbf{0}.$$

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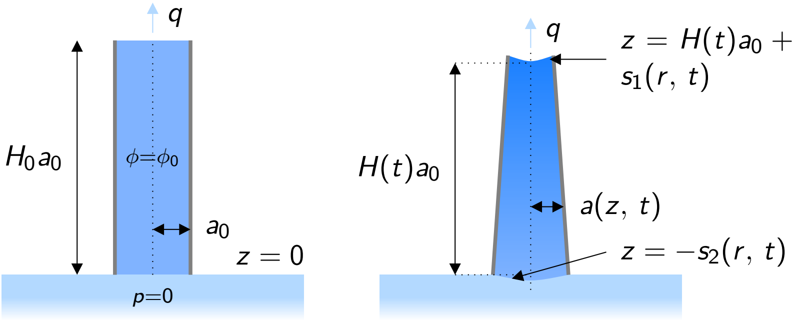
Take curls again and use $\boldsymbol{\nabla} \times \boldsymbol{\nabla} \times \boldsymbol{f} = \boldsymbol{\nabla} (\boldsymbol{\nabla} \cdot \boldsymbol{f}) - \nabla^2 \boldsymbol{f}$,

$$\nabla^2 \left[\nabla^2 \boldsymbol{\xi} + n \boldsymbol{\nabla} \left(\frac{\phi}{\phi_0} \right)^{1/n} \right] = \mathbf{0},$$

using the definition of $\boldsymbol{\epsilon}$ in terms of $\boldsymbol{\xi}$ and ϕ. This reduces to elastostatics when $\boldsymbol{\nabla}\phi = \mathbf{0}$.

Drying of a slender cylinder

Start by considering a slender cylinder which is allowed to dry only from the top, with its base immersed in water.



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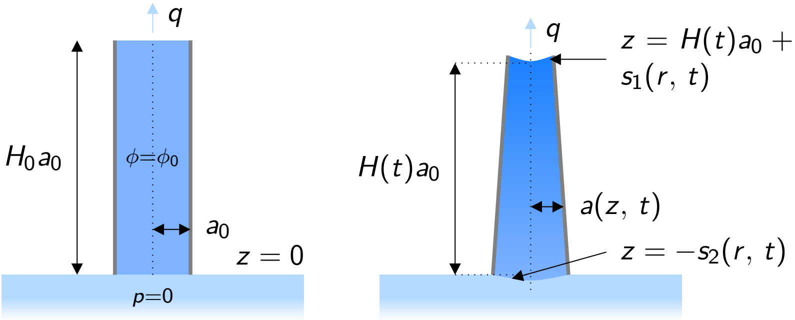

- Constant evaporation flux q
- Slenderness implies we can assume $\phi(r,\ z,\ t) \approx \phi(z,\ t)$ here.
- Introduce $\varepsilon = 1/H_0 \ll 1$.

Drying of a slender cylinder

Take a series expansion of the displacement field and the polymer fraction,

$$\phi(r,\, z,\, t) = \phi^{(0)}(z,\, t) + \varepsilon\phi^{(1)}(r,\, z,\, t) + \ldots$$
$$\xi_r(r,\, z,\, t) = \xi_r^{(0)}(r,\, z,\, t) + \varepsilon\xi_r^{(1)}(r,\, z,\, t) + \ldots$$
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Furthermore, assume the elements of $\boldsymbol{\epsilon}$ scale like ε and therefore

$$\frac{\partial \xi_r^{(0)}}{\partial r} = \frac{\xi_r^{(0)}}{r} = \frac{\partial \xi_z^{(0)}}{\partial z} = 1 - \left(\frac{\phi^{(0)}}{\phi_0}\right)^{1/3} \quad \text{and} \quad \frac{\partial \xi_z^{(0)}}{\partial r} + \frac{\partial \xi_r^{(0)}}{\partial z} = 0$$

at leading order. This corresponds to swelling which is isotropic at each z, as we would expect.

Drying of a slender cylinder

Imposing $\xi_r = 0$ at $r = 0$, and fixing $\xi_z = 0$ at $z = 0$, $r = a_0$, these equations have solutions

$$\xi_r = \left[1 - \left(\frac{\phi}{\phi_0}\right)^{1/3}\right] r \quad \text{and}$$

$$\xi_z = \int_0^z \left[1 - \left(\frac{\phi}{\phi_0}\right)^{1/3}\right] \mathrm{d}z + \frac{r^2}{2}\frac{\partial}{\partial z}\left(\frac{\phi}{\phi_0}\right)^{1/3} - \frac{a_0^2}{2}\left.\frac{\partial}{\partial z}\left(\frac{\phi}{\phi_0}\right)^{1/3}\right|_{z=0}.$$

We can thus describe the aspect ratio at time t as well, recalling that $\partial/\partial z \sim 1/H_0 a_0 \sim \varepsilon/a_0$,

$$H_0 a_0 = \int_0^{H(t)a_0} \left(\frac{\phi}{\phi_0}\right)^{1/3} \mathrm{d}z.$$

Drying of a slender cylinder

For simplicity's sake, take k, μ_s to be constants, and let $\mathit{\Pi} = K(\phi - \phi_0)/\phi_0$. Then, the governing equation for polymer fraction is

$$\frac{\partial \phi}{\partial t} = \frac{kK}{\mu_l} \frac{\partial}{\partial z} \left\{ \phi \frac{\partial}{\partial z} \left[\frac{\phi}{\phi_0} + \frac{4\mu_s}{K} \left(\frac{\phi}{\phi_0} \right)^{1/3} \right] \right\},$$

with $\sigma_{zz} = 0$ on the base and $\partial p / \partial z = -\mu_l q / k$ on the top setting boundary conditions. These become, at leading order,

$$\phi\big|_{z=0} = \phi_0 \quad \text{and} \quad \left.\frac{\partial \phi}{\partial z}\right|_{z=H(t)a_0} = \frac{\phi_0 \mu_l q}{kK} \left[1 + \frac{4\mu_s}{3K} \left(\frac{\phi}{\phi_0} \right)^{-2/3} \right]^{-1}.$$

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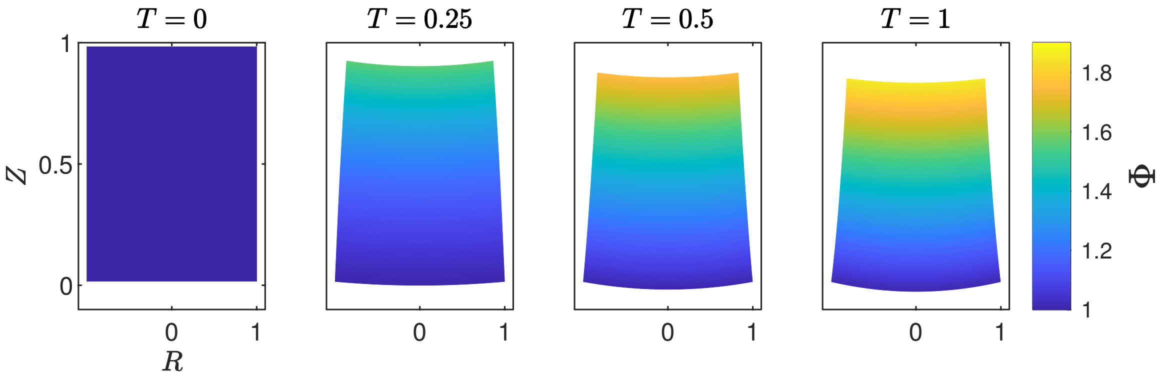
Polymer fraction ✓; shape of gel ✓(via ξ_r and ξ_z)



Notice, especially, the shape of the top surface:

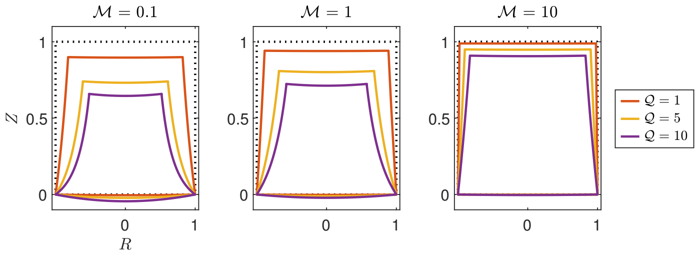
$$z - H(t)a_0 = \frac{r^2}{2} \frac{\partial}{\partial z} \left(\frac{\phi}{\phi_0} \right)^{1/3} \Bigg|_{z=H(t)a_0}$$

Steady state

The gel does not continue drying forever – eventually a steady state is reached where the evaporation flux from the top surface matches the rate at which water is drawn up from the base. In this state,

$$\frac{\phi - \phi_0}{\phi_0} + \frac{4\mu_s}{K}\left[\left(\frac{\phi}{\phi_0}\right)^{1/3} - 1\right] = \frac{\mu_l q}{kK}\phi_{\text{top}} z$$

with H_∞ and ϕ_{top} set by considering polymer conservation.



Origin of curvature

Without appealing to our theory for hydrogel swelling, can we explain why there is a curvature induced in the cylinder as it dries?

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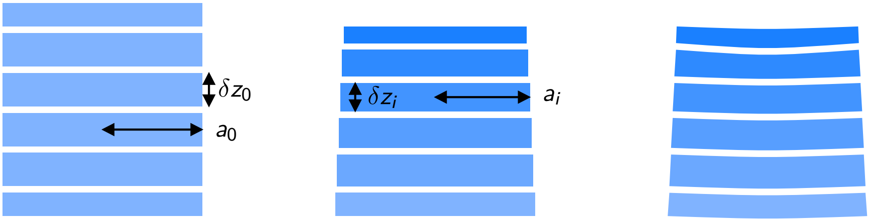
- Under the assumption of isotropy, $\delta z_i = (\phi_i/\phi_0)^{-1/3}\,\delta z_0$ and $a_i = (\phi_i/\phi_0)^{-1/3}\,a_0$.
- In order to match radial strains from the differential drying, we must introduce a curvature of each slice:



Origin of curvature

Appeal to classical plate theory to describe this curvature. This is valid since the gel is instantaneously incompressible and linear-elastic. Let the deflection displacements be $u\hat{\boldsymbol{r}} + w\hat{\boldsymbol{z}}$.

$$\nabla^4 w = 0 \quad \text{with } w = w' = 0 \text{ at } r = 0.$$

This comes from the Föppl-von Kármán equation in the absence of any loading, since $\partial\sigma_{zz}/\partial z = 0$. Then $w = \alpha r^2$ for some α.

Classical plate theory requires $\partial w/\partial r + \partial u/\partial z = 0$, and use this to derive a form for u. Then match radial strains across adjacent slices in the limit $\delta z_0 \to 0$ and find

$$w = \frac{r^2}{2}\frac{\partial}{\partial z}\left(\frac{\phi}{\phi_0}\right)^{1/3}.$$

Drying from the sides – a true multidirectional problem

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- Instead allow drying from the sides of the cylinder, which necessarily requires $\partial\phi/\partial r \neq 0$.

Non-dimensionalising $\Phi = \phi/\phi_0$, $R = r/a_0$, $Z = z/H_0 a_0$, $\mathcal{M} = \mu_s/K$ and $T = kKt/a_0^2 H_0^2 \mu_l$ as before,

Transport equation

$$\frac{\partial \Phi}{\partial T} = \frac{\partial}{\partial Z}\left[\Phi \frac{\partial}{\partial Z}\left(\Phi + 4\mathcal{M}\Phi^{1/3}\right)\right] + \frac{1}{R}\frac{\partial}{\partial R}\left[\Phi R \frac{\partial}{\partial R}\left(\Phi + 4\mathcal{M}\Phi^{1/3}\right)\right].$$

Drying from the sides – a true multidirectional problem

- The slenderness assumption made above essentially reduces the drying problem down to a unidirectional one – all flow of water is vertical, and radial contraction is purely isotropic.
- Instead allow drying from the sides of the cylinder, which necessarily requires $\partial\phi/\partial r \neq 0$.

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Boundary conditions

$$\Phi\big|_{Z=0} = 1; \quad \left.\frac{\partial \Phi}{\partial Z}\right|_{Z=\mathcal{H}(T)} = 0 \quad \text{and} \quad \left.\frac{\partial \Phi}{\partial R}\right|_{R=0} = 0; \quad \left.\frac{\partial \Phi}{\partial R}\right|_{R=A(Z,T)} = \frac{\mathcal{Q} - \frac{3\mathcal{M}}{A(Z,T)}\left(1 - \Phi^{1/3}\right)}{1 + (4\mathcal{M}/3)\Phi^{-2/3}}.$$

Leading-order displacement field

Can no longer make the assumption of 'isotropic' radial swelling, but still have small deviatoric strains. So, working at leading order,

$$\frac{1}{r}\frac{\partial}{\partial r}(r\xi_r) + \frac{\partial \xi_z}{\partial z} = 3\left[1 - \left(\frac{\phi}{\phi_0}\right)^{1/3}\right] \quad \text{and} \quad \frac{\partial \xi_r}{\partial z} + \frac{\partial \xi_z}{\partial r} = 0.$$

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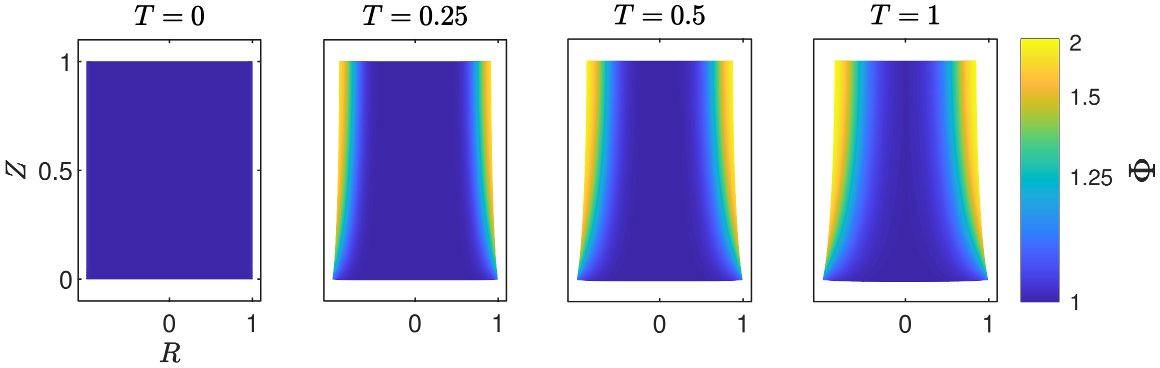
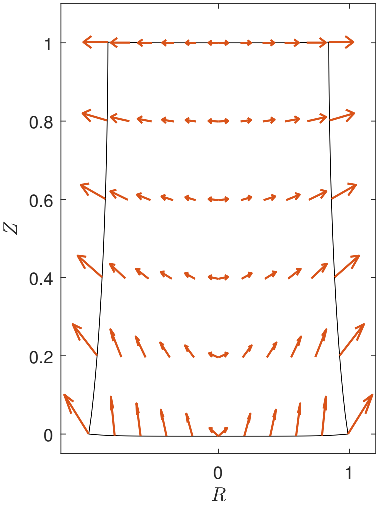
Polymer fraction ✓; shape of gel ✓



Curved surfaces present again but negligible contraction in the height of the gel since the bulk of the shrinkage is radial. Here, top surface is

$$Z - \mathcal{H}(T) = \frac{1}{H_0^2} \int_0^R \frac{3}{u} \int_0^u s \left. \frac{\partial \Phi^{1/3}}{\partial Z} \right|_{Z=\mathcal{H}(T)} \mathrm{d}s \, \mathrm{d}u \approx \frac{3R^2}{4H_0^2} \left. \frac{\partial \Phi^{1/3}}{\partial Z} \right|_{Z=\mathcal{H}(T)}$$



$\mathcal{M} = 0.1,\ \mathcal{Q} = 7.5,\ H_0 = 10$ at $T = 0.5.$

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- Such an approach works – it describes the behaviour seen in experiments and we are able to solve simple three-dimensional problems analytically.
- Current work: Rayleigh-Plateau-like instability of drying cylindrical gels, as seen in Matsuo and Tanaka (1992)