Continuum Mechanics group seminar
University of Strathclyde, Wednesday 1st October 2025, 13:00

A tractable framework for modelling hydrophilic large-swelling gels

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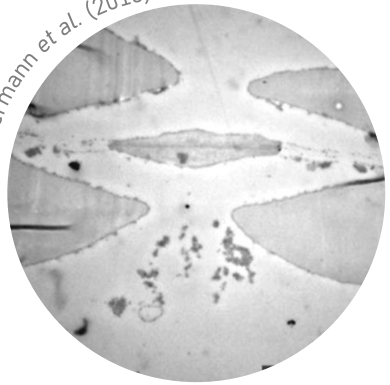
Hydrogels

Formed of a hydrophilic polymer scaffold surrounded by adsorbed water molecules

- can comprise >99% water by volume but remain solid
- behave elastically with low shear modulus
- swell or dry to extreme degrees when water is added or removed

Royal Horticultural Society



Pittermann *et al.* (2010)

Hydrogels

Fully-nonlinear models

$$W = W_{\text{mix}} + W_{\text{elastic}}$$

- Energy density function with contributions from mixing (entropy, electrostatic interactions, temperature-dependence, …) and elasticity (of individual polymer chains).
- Accurate, models large strains
- Not analytically tractable, parameters hard to determine

Flory & Rehner (1943a,b), Cai & Suo (2012), Bertrand et al. (2016), Butler & Montenegro-Johnson (2022)

Fully-linear models

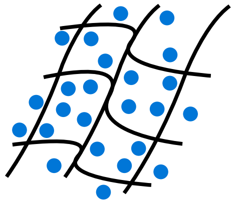
$$\frac{\partial \phi}{\partial t} = D \frac{\partial^2 \phi}{\partial x^2} \quad \left(D = K + \frac{4}{3}\mu \right)$$

- Based on linear poroelasticity, interstitial flow via Darcy's law. Treats gel as a linear-elastic material.
- Analytically tractable, clear physics, 'macroscopic' parameters
- Can't deal with large swelling strain

Biot (1941), Tanaka & Fillmore (1979), Doi (2009)

A detour into chemical physics

Usual approach: an energy density function with contributions from everything that could affect behaviour.



def. gradient from dry state

polymer volume fraction

$$\mathcal{W} = \underbrace{\frac{k_B T}{2\Omega_p}\left[\mathrm{tr}\left(\mathbf{F_d}\,\mathbf{F_d^T}\right) - 3 + 2\log\phi\right]}_{\text{Gaussian-chain elasticity}} + \underbrace{\frac{k_B T}{\Omega_f}\left[\frac{1-\phi}{\phi}\log(1-\phi) + \chi(\phi,\,T)(1-\phi)\right]}_{\text{Mixing of polymer and water}}$$

At equilibrium, elasticity balances mixing ($\partial W/\partial\lambda_i = 0$)

$$\phi_0 \approx \left[\frac{\Omega_p}{\Omega_f}\left(\frac{1}{2} - \chi(T)\right)\right]^{-3/5}$$

Often, chi parameter is very close to ½, but polymer molecules are much bigger than water

Away from equilibrium, things are nastier!

$$\sigma_{\mathrm{ij}}^{\mathrm{eff}} = \phi\frac{\partial\mathcal{W}}{\partial\mathsf{F}_{\mathrm{ik}}}\mathsf{F}_{\mathrm{jk}}$$

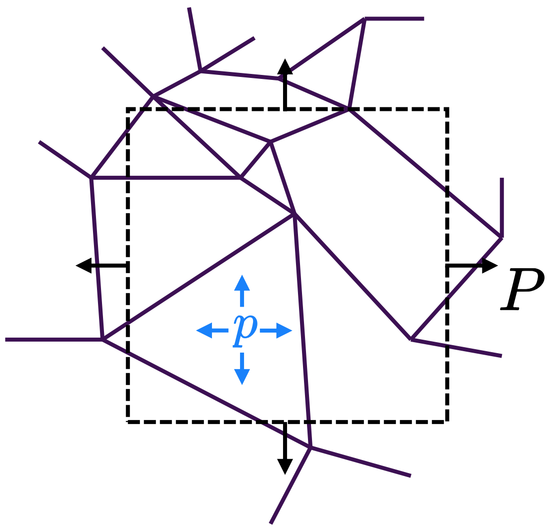
Flory & Rehner (1943) *J. Chem. Phys.* ***11***:*512-520*; Flory & Rehner (1943) *J. Chem. Phys* ***11***:*521-526*; Cai & Suo (2011) *Europhys. Lett.* ***97***:*34009*

Poromechanics

A geophysicist's approach: separate contributions from stress into a 'pore pressure' and an 'effective stress'

$$\boldsymbol{\sigma} = -p\mathbf{I} + \boldsymbol{\sigma}_{\text{eff}}$$

Bulk pressure (or thermodynamic pressure) the isotropic stress exerted by a sample of gel; our familiar concept of pressure



Pervadic pressure (or Darcy pressure, "pore" pressure) is the pressure as would be measured by a transducer separated by a partially-permeable membrane from the gel.

In soil science: p is the pore pressure, P is the overburden pressure
In colloids: p is [related to] the chemical potential, Π is the osmotic pressure

$$P = p + \Pi$$

osmotic effects?
isotropic elasticity?
generalised osmotic pressure

p drives flows by Darcy's law:

permeability

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

relative fluid flux
$\boldsymbol{u} = (1-\phi)(\boldsymbol{u_w} - \boldsymbol{u_p})$

(dynamic) viscosity of fluid

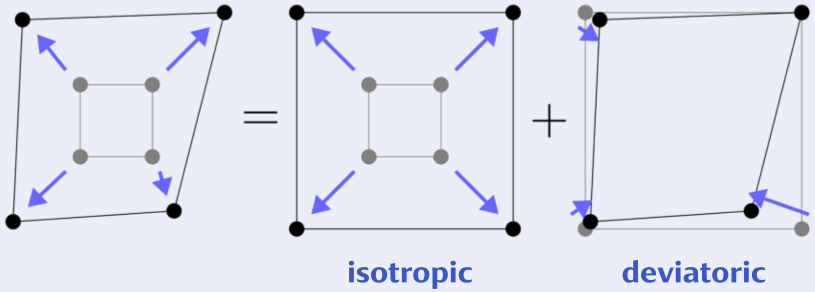
Biot (1941) *J. Appl. Phys.* ***12****:155–164*; Peppin *et al.* (2004) *Phys. Fluids* ***17****:053301*

Poromechanics

Linear (Biot) poroelasticity specifies a linear-elastic constitutive relation linking strains to effective stresses. Hydrogels swell a lot, with potentially large strains: linear is no good!

One way around this: use **finite strain (nonlinear) elastic models** for effective stress.

e.g. Hencky model $$\boldsymbol{\sigma}_{\text{eff}} = \frac{\Lambda\phi}{2}\,\mathrm{tr}\big(\ln(\mathbf{F}\mathbf{F}^{\mathrm{T}})\big)\mathbf{I} + \frac{M-\Lambda}{2}\ln(\mathbf{F}\mathbf{F}^{\mathrm{T}})$$



assume linearity only in the **deviatoric strain** from some **fully-swollen reference state**

"linear-elastic materials with properties dependent on swelling state"

Biot (1941) *J. Appl. Phys.* ***12**:155-164*; Tanaka *et al.* (1973) *J. Chem Phys.* ***59**:5151-5159*; MacMinn *et al.* (2016) *Phys. Rev. Appl.* ***5**:044020*

The linear-elastic-nonlinear-swelling model

Therefore, the deviatoric part of the effective stress tensor must depend linearly on the deviatoric part of the Cauchy strain (the isotropic part could be huge)

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

deviatoric strain
assumed small

isotropic strain
depends only on degree to which gel is swollen

This allows us to construct a stress tensor like usual

shear modulus depends on swelling!
dry gels will probably be stiffer

$$\boldsymbol{\sigma}_{\mathrm{eff}} = -\Pi(\phi)\mathbf{I} + 2\mu_s(\phi)\boldsymbol{\epsilon}$$

isotropic part must be (-) osmotic pressure
since the isotropic part of the total stress tensor is (-) the bulk pressure

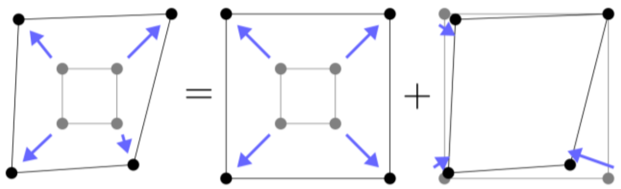
depends on swelling alone
isotropic strains lead to isotropic stresses – see the isotropic part of strain tensor

physically intuitive result

JJW & Worster (2023) *J. Fluid Mech.* ***960:****A37*

The linear-elastic-nonlinear-swelling model

$$\boldsymbol{\sigma} = -p\mathbf{I} + \boldsymbol{\sigma}_{\text{eff}}$$
$$P = p + \Pi$$



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

$$\boldsymbol{\sigma}_{\text{eff}} = -\Pi(\phi)\mathbf{I} + 2\mu_s(\phi)\boldsymbol{\epsilon}$$
$$\boldsymbol{u} = (1-\phi)(\boldsymbol{u_w} - \boldsymbol{u_p})$$
$$\boldsymbol{q} = (1-\phi)\boldsymbol{u_w} + \phi\boldsymbol{u_p}$$

- Have an expression for stress in the gel, so conservation of momentum links pressure gradients to deviatoric strains,

$$\boldsymbol{\nabla}\cdot\boldsymbol{\sigma} = 0 \quad \text{so} \quad \underbrace{\boldsymbol{\nabla} p = -\boldsymbol{\nabla}\Pi(\phi)}_{\text{pervadic pressure gradients oppose osmotic ones}} + 2\boldsymbol{\nabla}\cdot[\mu_s(\phi)\boldsymbol{\epsilon}]$$

- Since gradients in pervadic pressure drive flows, this allows us to describe gel reconfiguration (when coupled with conservation of polymer and water)

depends on swelling

$$\frac{\partial\phi}{\partial t} + \boldsymbol{q}\cdot\boldsymbol{\nabla}\phi = \boldsymbol{\nabla}\cdot(\phi\boldsymbol{u}) \quad \text{alongside} \quad \boldsymbol{u} = -\frac{k(\phi)}{\mu_l}\boldsymbol{\nabla} p$$

phase-averaged (gel and water) flux

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

JJW & Worster (2023) *J. Fluid Mech.* ***960****:A37*; JJW et al. (2023) *J. Fluid Mech.* ***960****:A38*

Characterising a gel

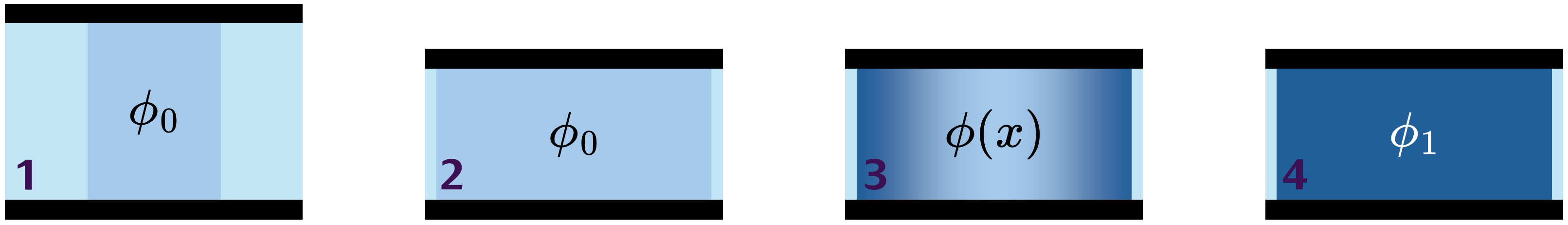
$$\boxed{\frac{\partial\phi}{\partial t} + \boldsymbol{q}\cdot\boldsymbol{\nabla}\phi = \boldsymbol{\nabla}\cdot\left\{\frac{k(\phi)}{\mu_l}\left[\phi\frac{\partial\Pi}{\partial\phi} + \frac{4\mu_s(\phi)}{3}\left(\frac{\phi}{\phi_0}\right)^{1/3}\right]\boldsymbol{\nabla}\phi\right\}}$$

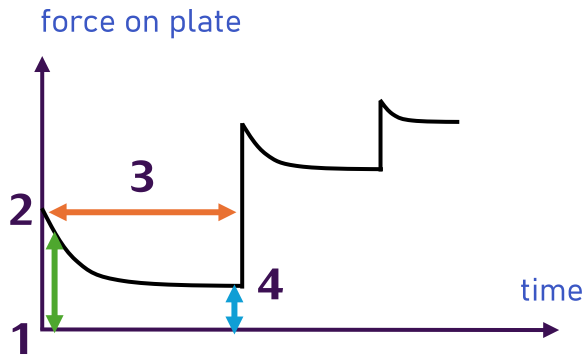
$$\boldsymbol{\sigma} = -[p + \Pi(\phi)]\mathbf{I} + 2\mu_s(\phi)\boldsymbol{\epsilon} \qquad \boldsymbol{u} = \frac{k(\phi)}{\mu_l}\left[\frac{\partial\Pi}{\partial\phi} + \frac{4\mu_s(\phi)}{3\phi}\left(\frac{\phi}{\phi_0}\right)^{1/3}\right]\boldsymbol{\nabla}\phi$$

Shear modulus characterises the stiffness of a hydrogel and describes the initial elastic response before water diffuses through the structure

Osmotic pressure characterises the affinity for water (‘desire’ to swell or deswell)

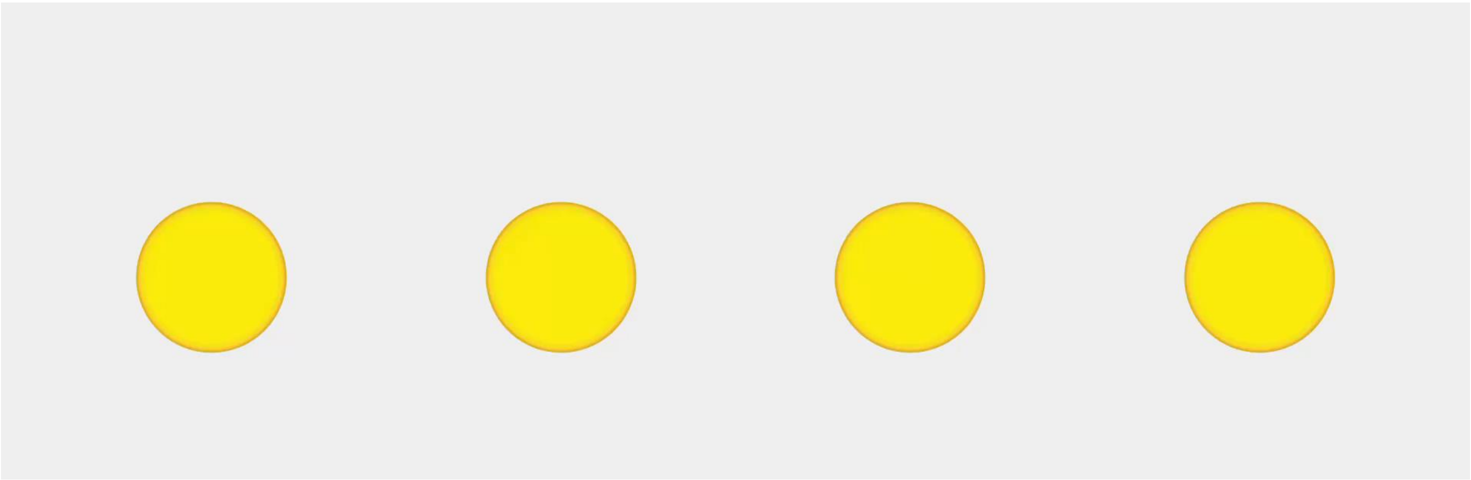
Permeability describes the resistance to viscous flow through the pore scaffold





JJW & Worster (2023) *J. Fluid Mech.* ***960**:A37*; JJW et al. (2023) *J. Fluid Mech.* ***960**:A38*; JJW (2024) *PhD thesis, U. Cambridge*

Insights from LENS modelling



Uniaxial problems are easy: shape set by polymer conservation

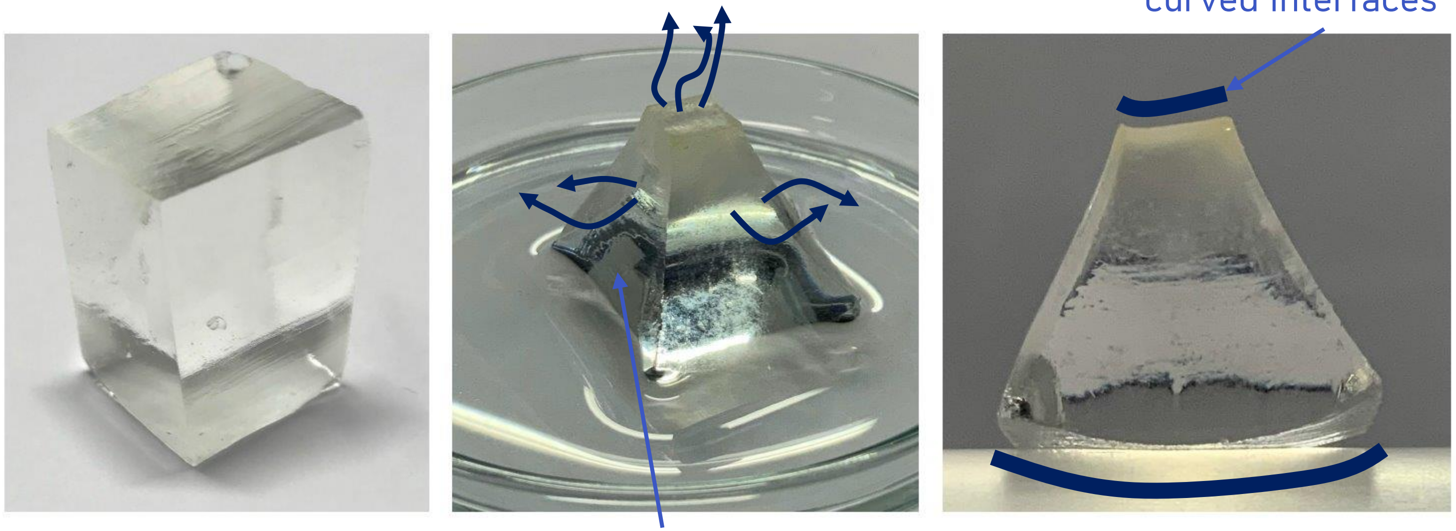
1+1 dimensional advection-diffusion equation

BC at gel-water interface set by stress and pressure balance

More complex geometries = hard!

Need to relate swelling state to displacement to find how the shape evolves

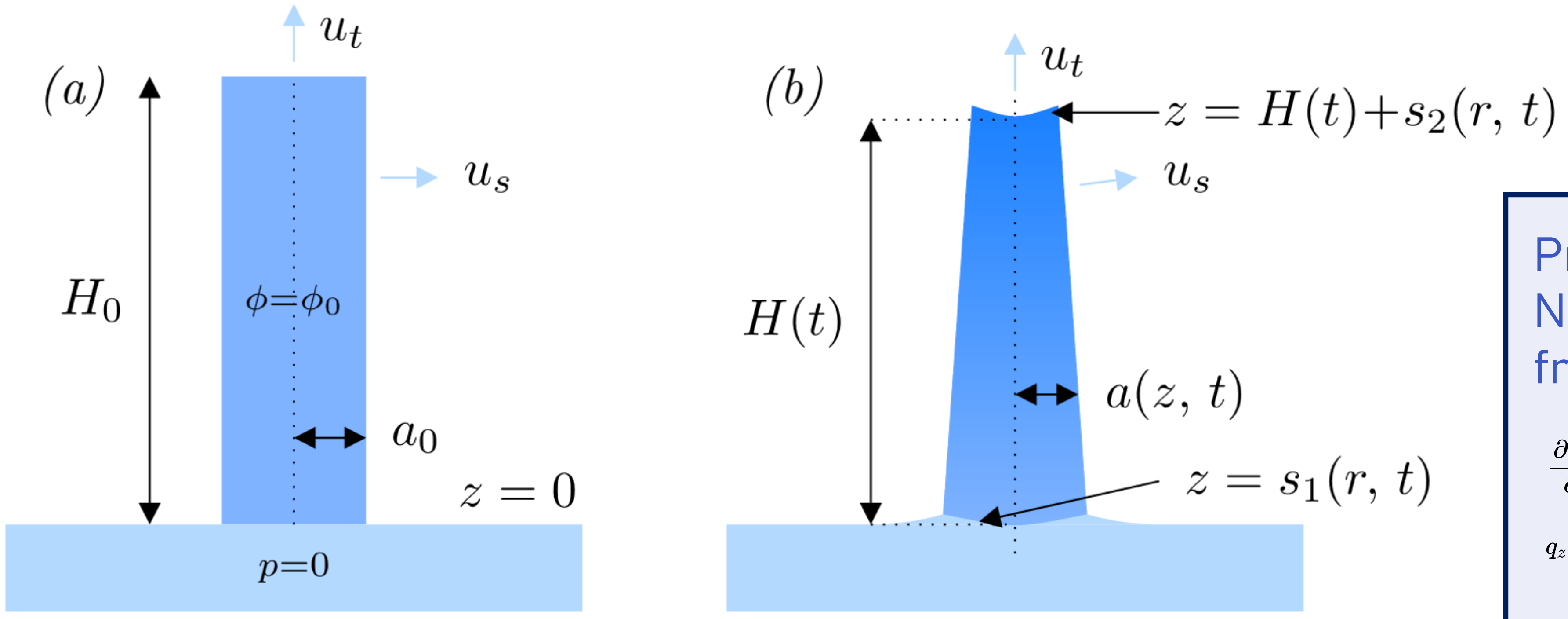
$$\nabla^4 \boldsymbol{\xi} = -3\boldsymbol{\nabla}\nabla^2(\phi/\phi_0)^{1/3}$$



JJW & Worster (2023) *J. Fluid Mech.* ***960**:A37*; JJW *et al.* (2023) *J. Fluid Mech.* ***960**:A38*

Drying of slender cylinders

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



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

Prescribed flux gives a Neumann BC on polymer fraction field

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

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

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

+ use polymer conservation to set the geometry of the cylinder as it dries

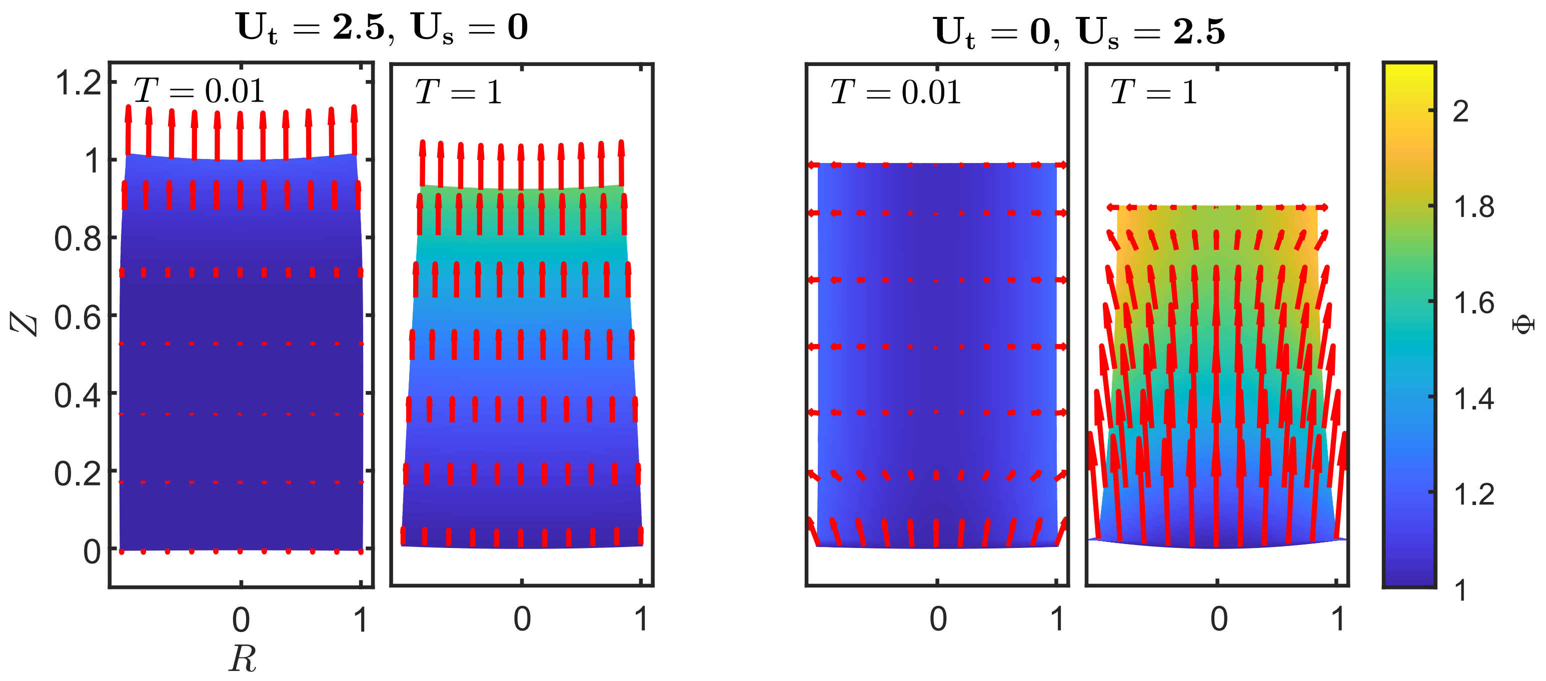
$$\phi(r,\, z,\, t) = \phi_C(z,\, t) + \varepsilon^2\phi_1(r,\, z,\, t)$$

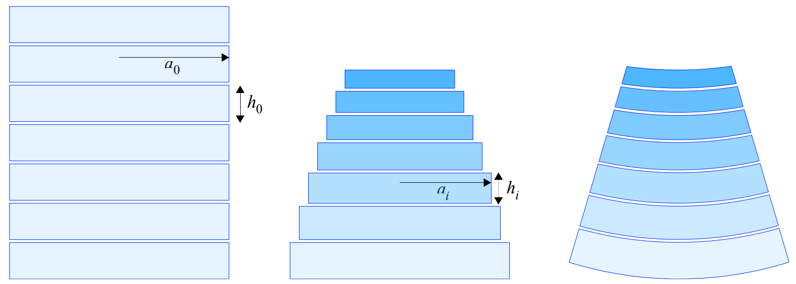
separation of variables implies that

- $\varepsilon = a_0/H_0$ so only square of the aspect ratio should be small
- $\phi_1 \propto r^2$ parabolic polymer fraction profiles

JJW et al. (2023) *J. Fluid Mech.* ***960***:A38

Drying of slender cylinders





Expression for radius suggests isotropic contraction at a fixed vertical position

Height follows from polymer conservation

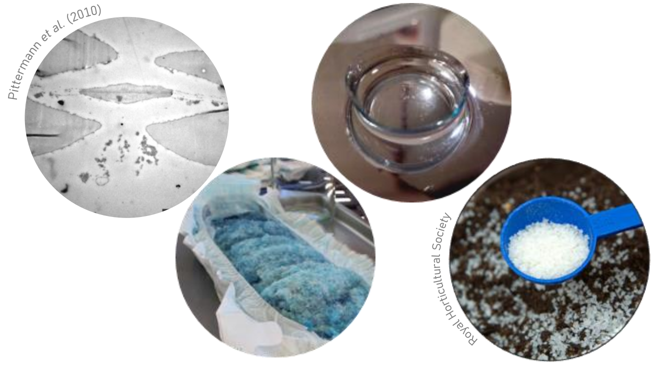
Differential drying creates the curved shapes at the top and bottom

$$a(z,\, t) = (\phi_C/\phi_0)^{-1/3} a_0 \qquad h_0 = \int_0^{H(t)} \left[1 - (\phi_C/\phi_0)^{1/3}\right] \mathrm{d}z'$$

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

JJW *et al.* (2023) *J. Fluid Mech.* ***960:****A38*

Where can we go from here?

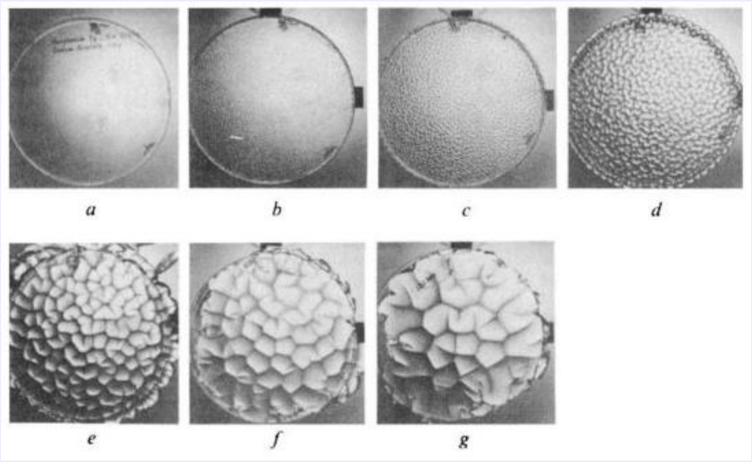


Wrinkling instabilities

JJW & Worster *Phys. Rev. E* **109**:044602 (2024)

Wrinkles form and coarsen as gels swell, owing to anchoring from a partially-dried core.

Tanaka *et al.*, Nature **325:**796-798, 1987



The LENS model

Responsive hydrogels

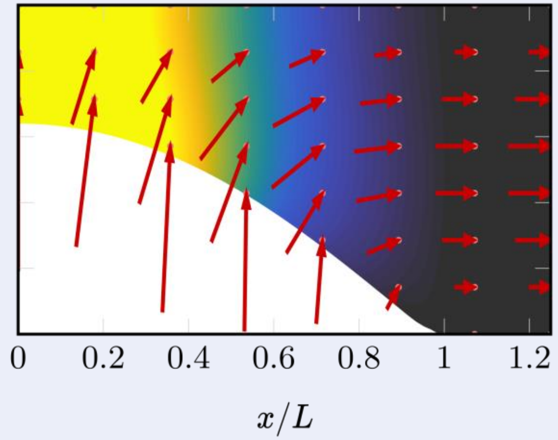
JJW & Montenegro-Johnson *J. Fluid Mech.* **1009:**A38 (2025)

Gels that swell or deswell in response to external stimuli

Freezing soft gels

JJW & Worster *Proc. Roy. Soc. A* **481:**20240721 (2025)

Ice can't form inside hydrogel pores owing to capillarity, so it forms as segregated 'chunks' that grow through a process known as cryosuction.



Self-oscillating gels

Chemically-responsive gels deswell once the concentration of a certain species (usually an ion of some kind) crosses some critical threshold.

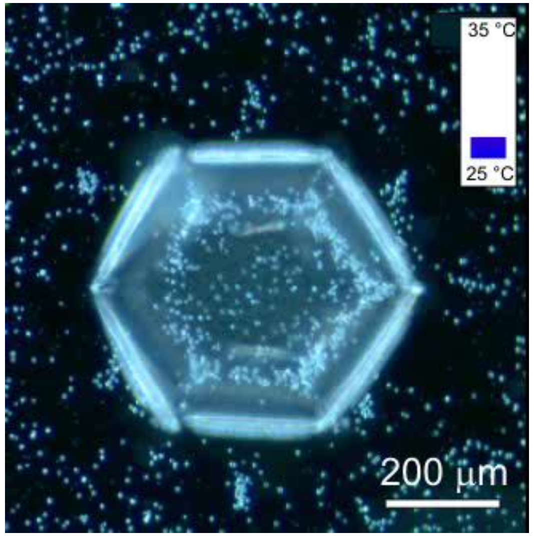
Mechanism: ions are attracted to charged parts of the polymer chains, which they stick to and the chains 'refold', driving out water.

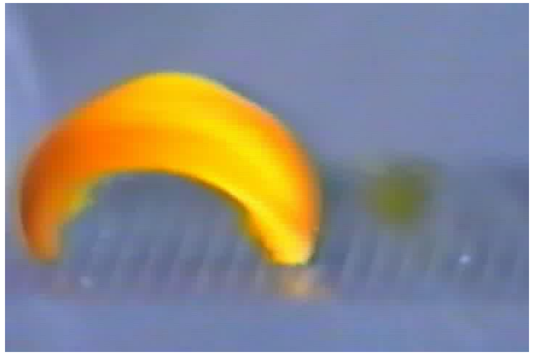
This process is reversible, and so chemical signals can be converted to mechanical changes.

$$\mathcal{W} = \frac{k_B T}{2\Omega_p}\left[\mathrm{tr}\left(\mathsf{F_d}\,\mathsf{F_d^T}\right) - 3 + 2\log\phi\right] + \frac{k_B T}{\Omega_f}\left[\frac{1-\phi}{\phi}\log(1-\phi) + \boxed{\chi(\phi, T)}(1-\phi)\right]$$

Recall that $\sigma^{\mathrm{eff}}_{\mathrm{ij}} = \phi \dfrac{\partial \mathcal{W}}{\partial \mathsf{F}_{\mathrm{ik}}}\mathsf{F}_{\mathrm{jk}}$ and thus the influence of the chi parameter (after much algebra) is found to be only in the isotropic part

Chemical response just affects the osmotic pressure



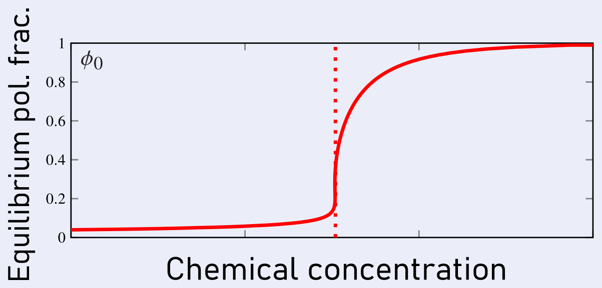


1. **Stoychev *et al.*** Soft Matter **7** (2011)
2. **Maeda *et al.*** Advanced Materials **19** (2007)

JJW & Montenegro-Johnson (2025) *J. Fluid Mech.* ***1009**:A38*

Self-oscillating gels

Sharp transition

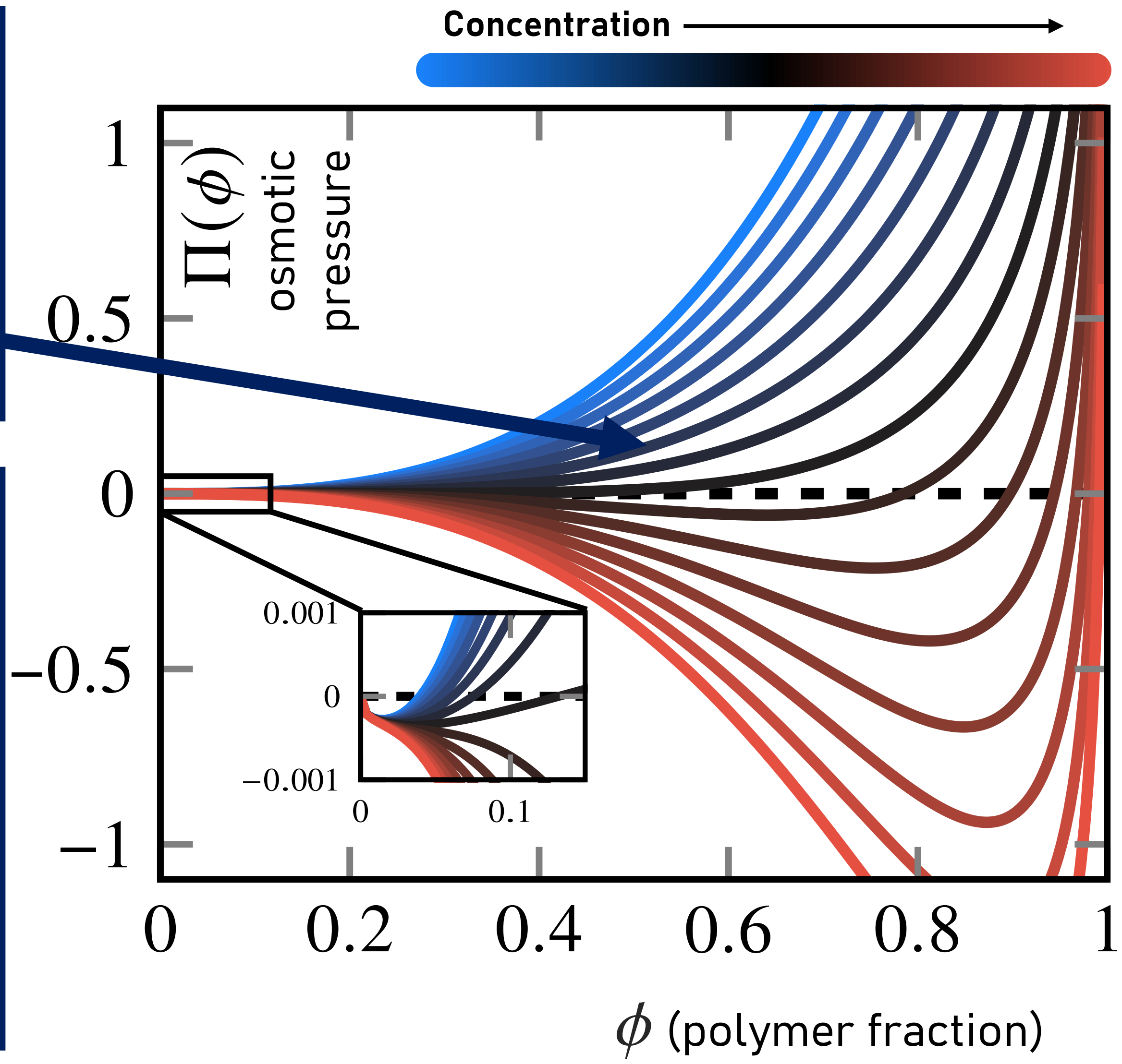


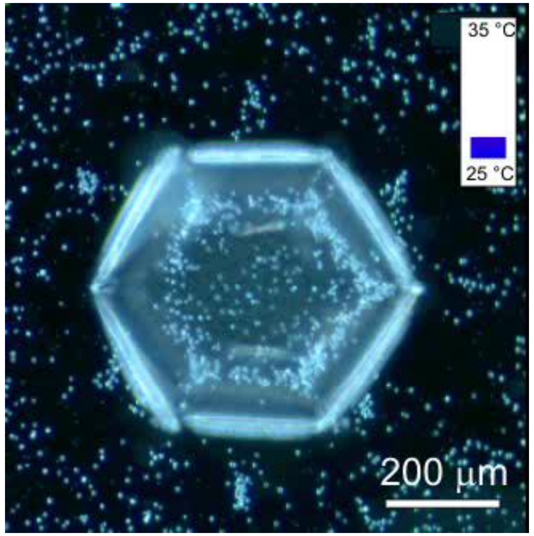
Phenomenology informs the model

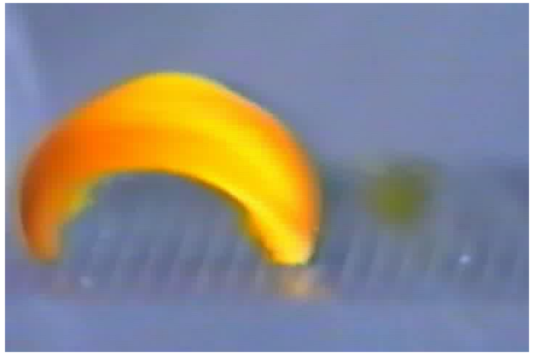
$$\phi_0(Y) = \begin{cases} \phi_{00} & Y \leq Y_C \\ \phi_{0\infty} & Y > Y_C \end{cases}$$

With a linear osmotic pressure for simplicity

$$\Pi(\phi) = \Pi_0 \frac{\phi - \phi_0(Y)}{\phi_0(Y)}$$







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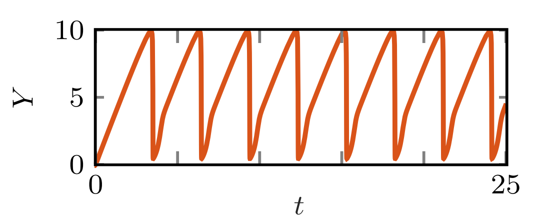
Self-oscillating gels

Brusselator model for reaction

$$A \to X$$
$$2X + Y \to 3X$$
$$B + X \to Y + D$$
$$X \to E$$



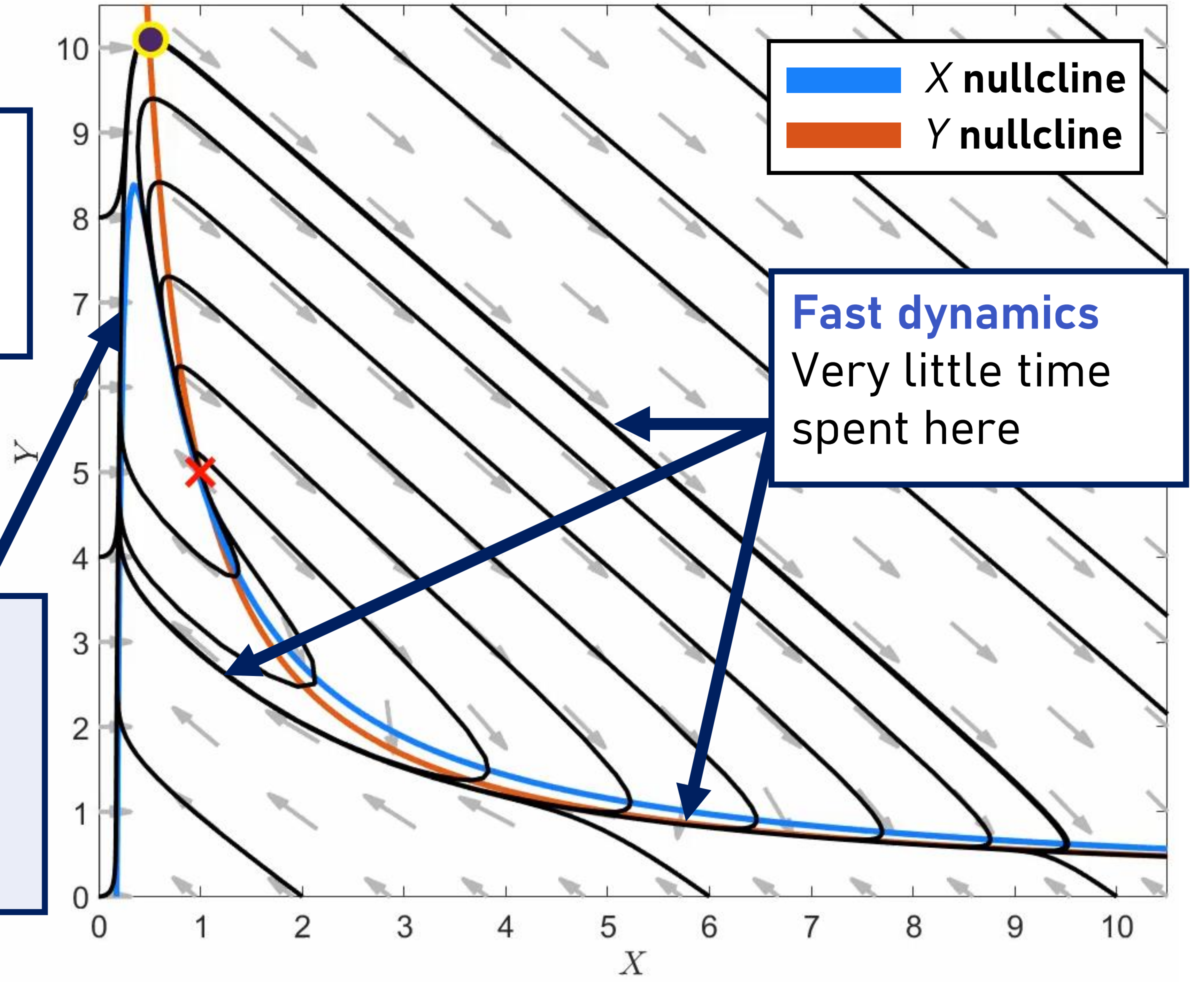
assume A and B are in excess

$$\frac{\mathrm{d}X}{\mathrm{d}t} = k\left[A + X^2Y - (1+B)X\right]$$
$$\frac{\mathrm{d}Y}{\mathrm{d}t} = k\left[BX - X^2Y\right]$$

One fixed point at $(X, Y) = (A, B/A)$**,**
unstable provided $B > 1 + A^2$

Slow dynamics
On X nullcline

$$T \approx \frac{(B+1)^2}{4kA^2}$$



Fast dynamics
Very little time spent here

JJW & Montenegro-Johnson (2025) *Phys. Rev. Res.* ***1009***:*A38*

Self-oscillating gels

Gel dynamics
Decoupled, but drives interstitial flows!

Reaction (only in gel)

$$k\left[A + X^2Y - (1+B)X\right] \quad (c{=}X)$$

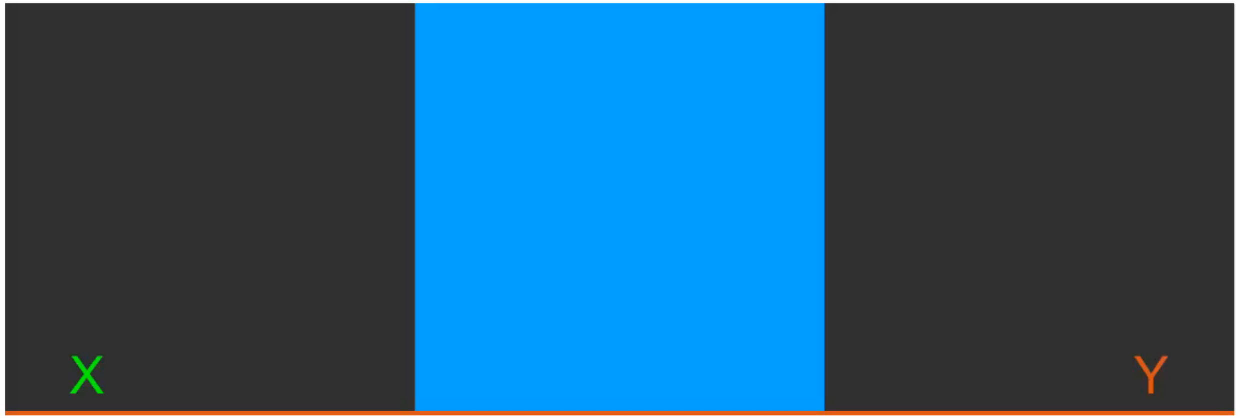
$$k\left[BX - X^2Y\right] \quad (c{=}Y)$$

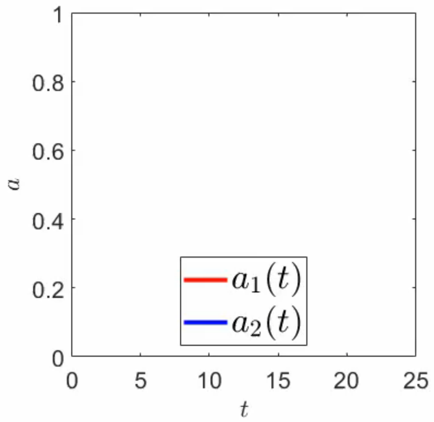
Advection with flow
Assume no flow in water, Darcy flow in gel

$$u = -\frac{k(\phi)}{\mu_l}\frac{\partial p}{\partial x} = -\frac{D(\phi, Y)}{\phi}\frac{\partial \phi}{\partial x}$$

$$\frac{\partial c}{\partial t} + u\frac{\partial c}{\partial x} = \mathcal{R} + D_c\frac{\partial^2 c}{\partial x^2}$$

Diffusion
Different coefficient in water and gel

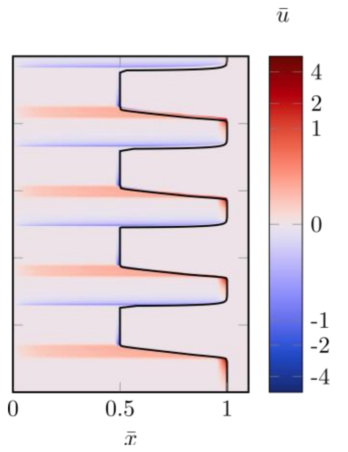




Reaction rate constant 5 times higher than in left-hand gel

JJW & Montenegro-Johnson (2025) *Phys. Rev. Res.* ***1009****:A38*

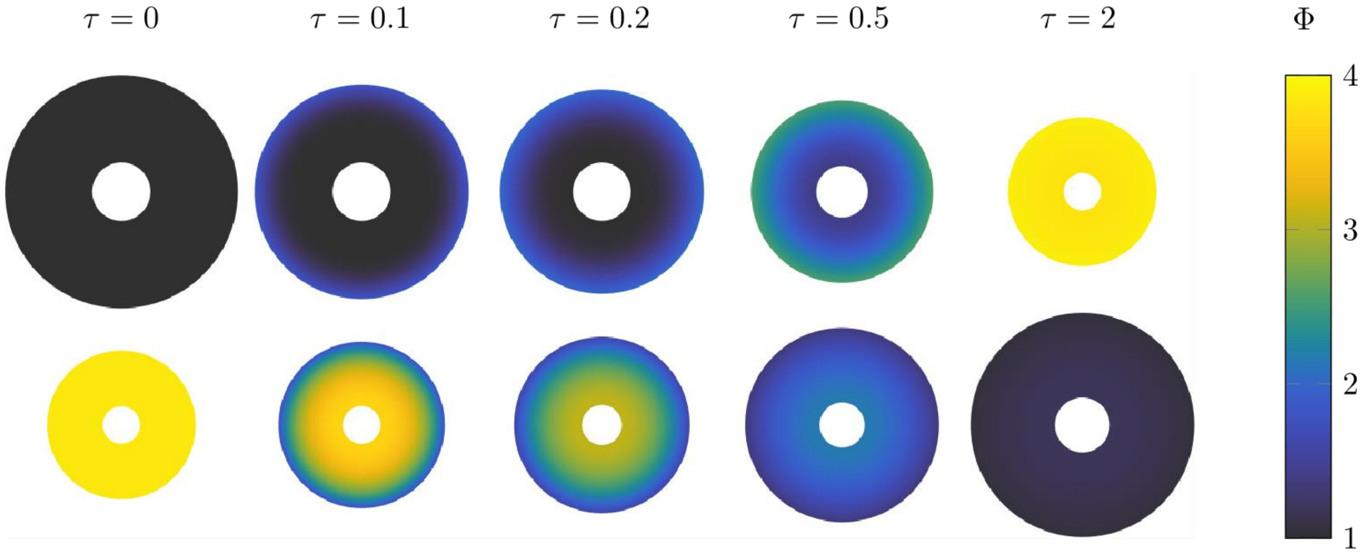
Self-oscillating pumps?

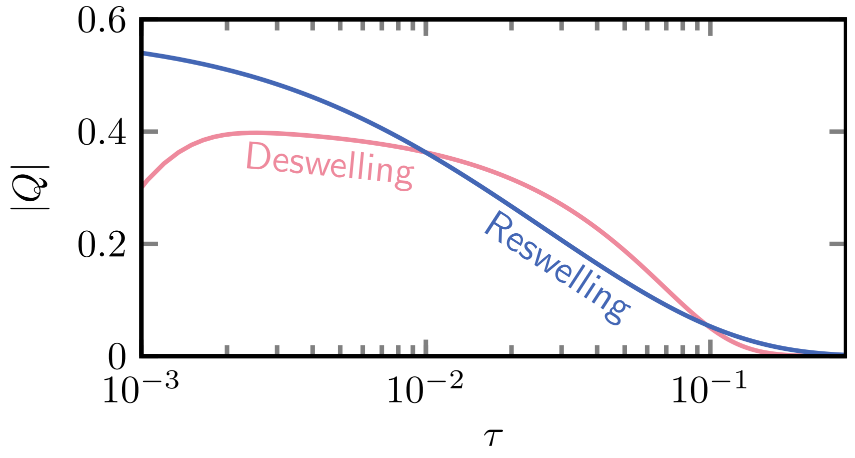


Where does the water go when a gel deswells?

- Pumped out of the matrix into the bath (flux $(1-\phi)\boldsymbol{u_w}$)
- Occupies the space where the gel once was – interface moves at $\boldsymbol{u_p}$

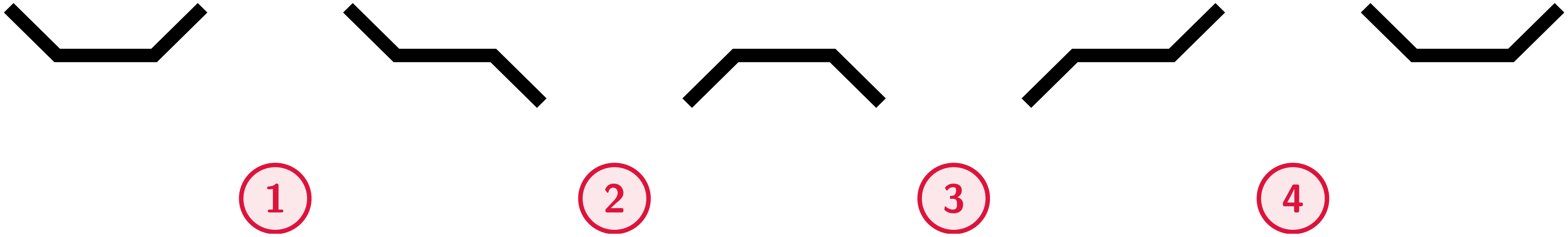
Net flux is thus $(1-\phi)\boldsymbol{u_w} - \phi\boldsymbol{u_p} = (1-\phi)\left(\boldsymbol{u_w} - \boldsymbol{u_p}\right) + \boldsymbol{u_p} = \boldsymbol{u} + \boldsymbol{u_p} = \boldsymbol{q}$ which is a solenoidal quantity. In most uniaxial geometries, this has to be zero everywhere if it's zero somewhere **so these gels can't pump!**



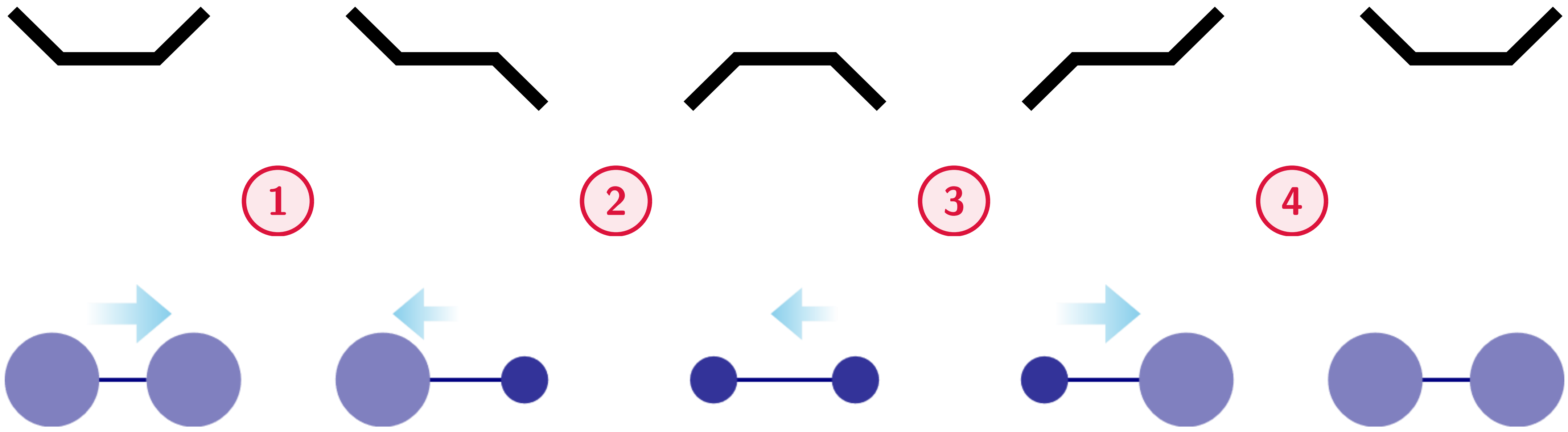


$$\boldsymbol{q} = \frac{A}{r^2}\hat{\boldsymbol{r}} \qquad \boldsymbol{q} = \left(\frac{\phi_{\text{in}}}{\phi_0}\right)^{-1/3} \frac{r_{\text{in}}^2}{r^2} \frac{\mathrm{d}r_{\text{in}}}{\mathrm{d}t}\hat{\boldsymbol{r}}$$

JJW & Montenegro-Johnson (2025) *Phys. Rev. Fluids (accepted)*



JJW & Montenegro-Johnson (2025) *Phys. Rev. Fluids (accepted)*



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Flow field around the two-sphere swimmer

Each sphere acts like a source or sink of strength q – if there were just one sphere with velocity $\boldsymbol{U_1}$, the surrounding flow field would be

$$\boldsymbol{u} = \frac{3}{4}\boldsymbol{U_1}\left[\frac{a_1}{r} + \frac{a_1^3}{3r^3}\right] + \frac{3}{4r^2}(\boldsymbol{U_1}\cdot\boldsymbol{x})\boldsymbol{x}\left[\frac{a_1}{r} - \frac{a_1^3}{r^3}\right] + \frac{q_1\boldsymbol{x}}{4\pi r^3}$$

We can then use the Faxén relations for a sphere to find the motion of the other sphere, calculating the "first reflection"

$$\boldsymbol{U} = \frac{\boldsymbol{F}}{6\pi\mu_l a} + \boldsymbol{u_\infty}(\boldsymbol{0}) + \frac{a^2}{6}\nabla^2\boldsymbol{u_\infty}(\boldsymbol{0})$$

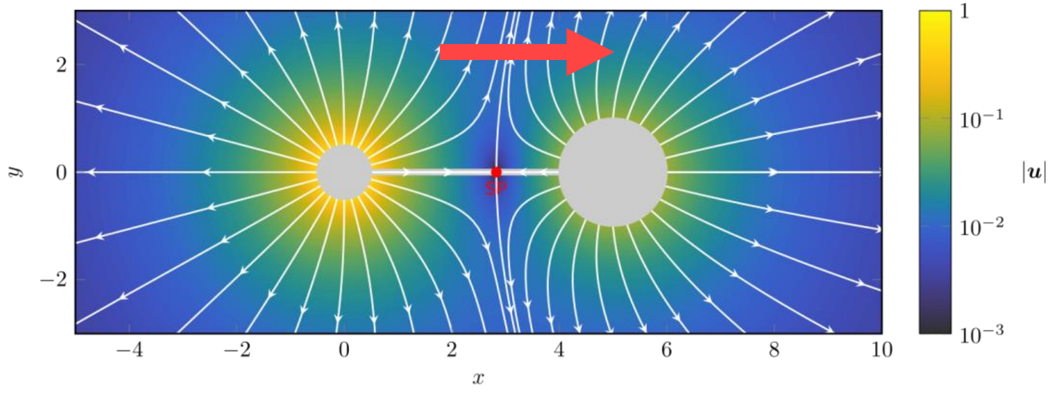
measured in coordinates centred on sphere 2

instantaneous swim speed

$$U = \frac{1}{4\pi\ell^2}\frac{a_2q_1 - a_1q_2}{a_1+a_2}\left[1 - \frac{1}{\ell}\frac{a_1a_2}{a_1+a_2}\left(\frac{2}{3} - \frac{a_1^2+a_2^2}{\ell^2}\right)\right]^{-1}$$

separation distance

Then, do the same the other way around and equate velocities…



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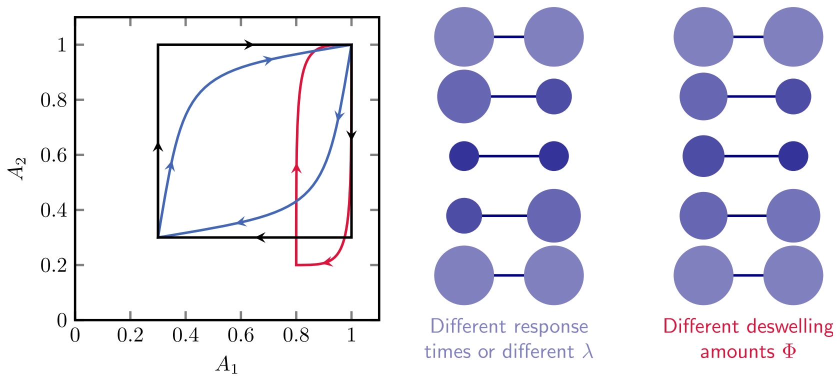
Drifting

The strength of the point source can be related to the radii of the two spheres through the parameter λ (the ratio of inner to outer radius of gel in the sphere) and the initial outer radius ρ_{20}

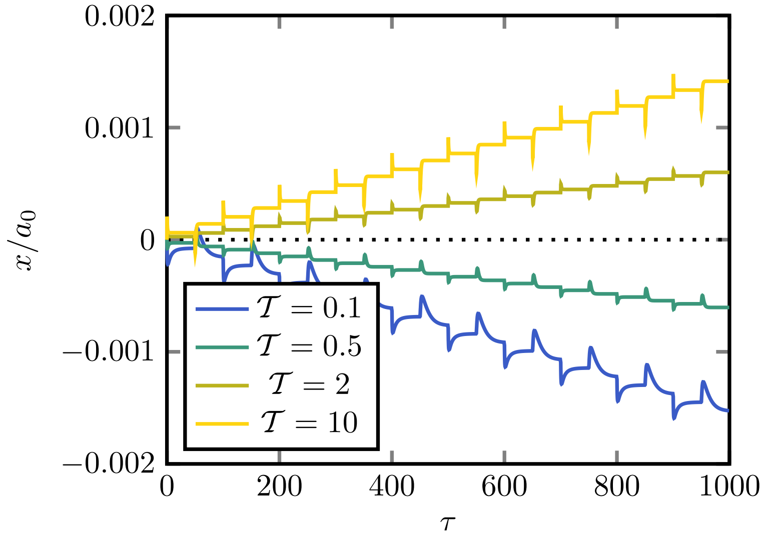
$$U = \frac{1}{3\ell^2}\frac{a_1 a_2}{a_1 + a_2}\left[1 - \frac{1}{\ell}\frac{a_1 a_2}{a_1 + a_2}\left(\frac{2}{3} - \frac{a_1^2 + a_2^2}{\ell^2}\right)\right]^{-1}\frac{\mathrm{d}}{\mathrm{d}t}\left(\frac{\lambda_1^3 a_1^3}{(\rho_{20})_1} - \frac{\lambda_2^3 a_2^3}{(\rho_{20})_2}\right)$$

Integrate over one stroke cycle to find a net drift, and can investigate how this occurs in more physically realisable sources of asymmetry (e.g. different extents of drying, different rates of drying, different thicknesses of gel.



In this case, the response times for each sphere are set differently, but they are otherwise identical

$\mathcal{T} = t_1/t_2$



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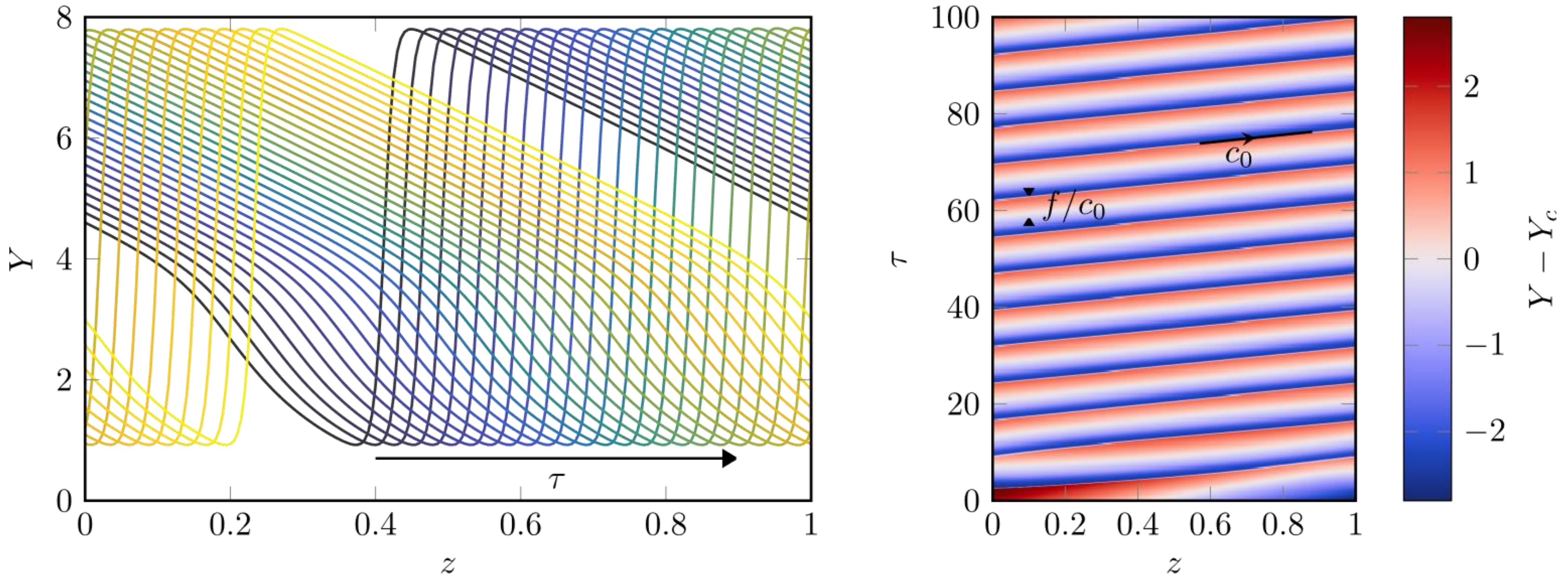
Surfing

With spatial variation the BZ reaction can have travelling wave-like solutions with a duration f/c_0 and speed c_0



What happens if the asymmetry or rate of deswelling is sufficiently high that the instantaneous speed outpaces the chemical wave?

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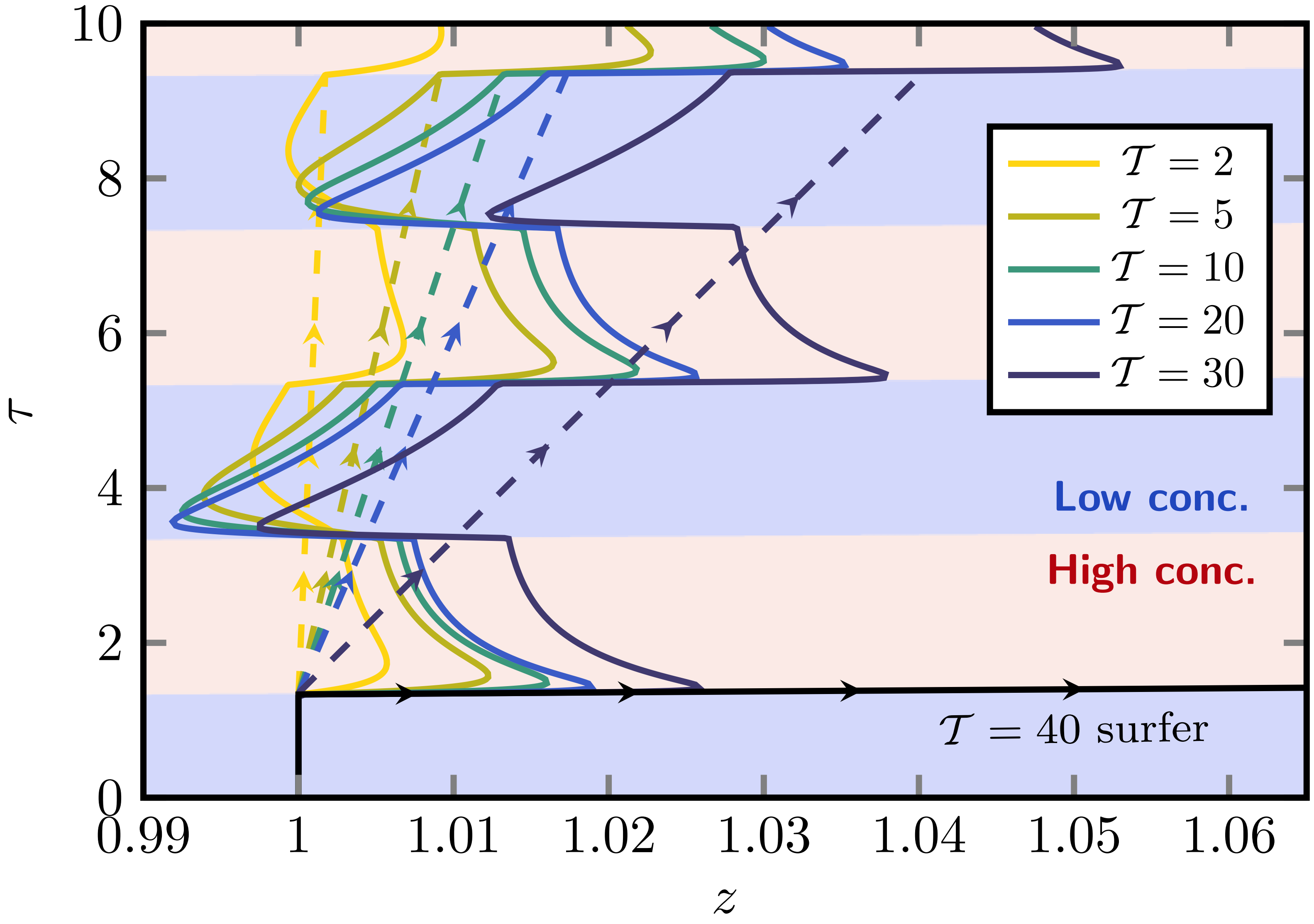
Surfing



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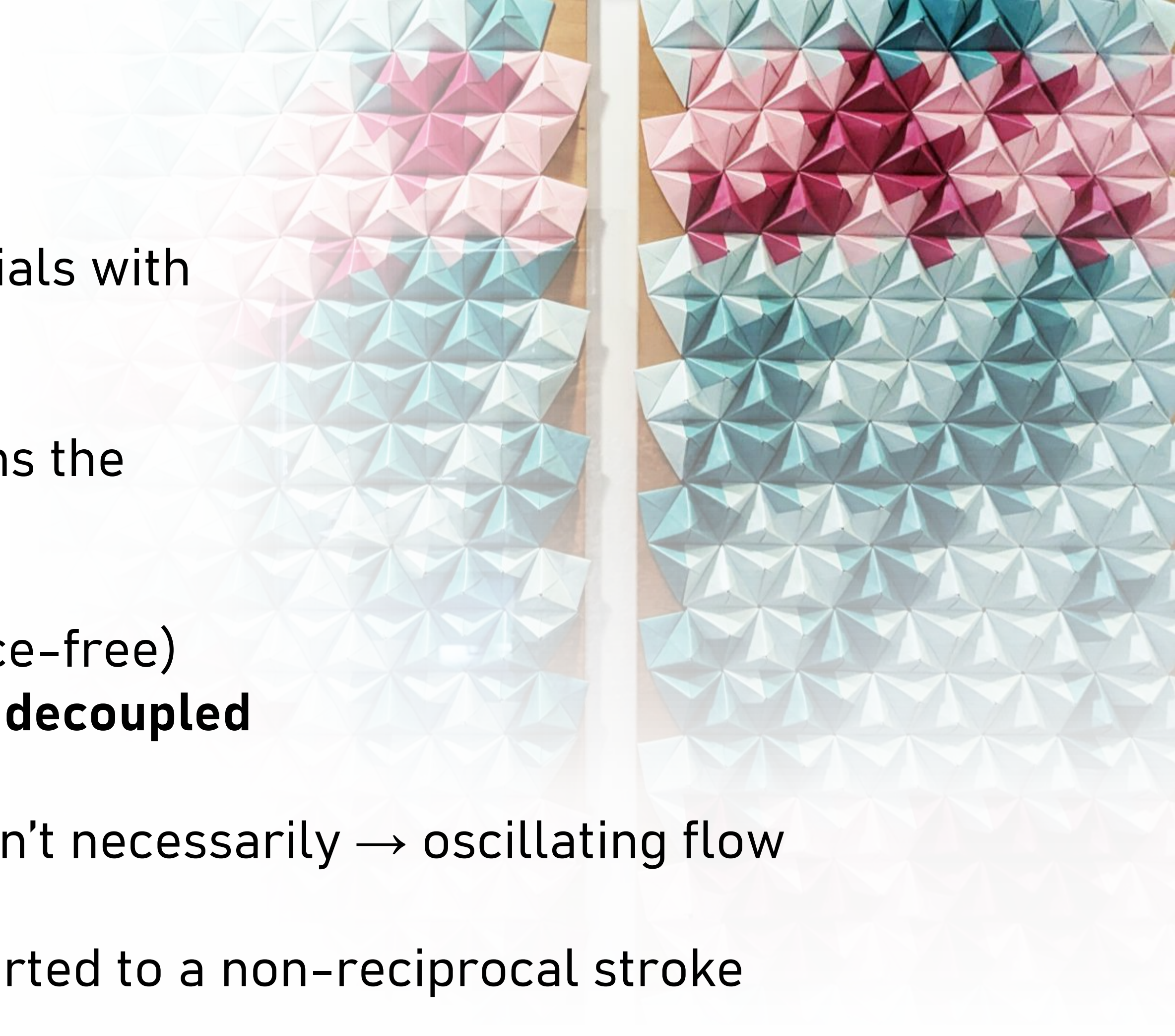
Conclusions and applications



- Hydrogels can be modelled as linear-elastic materials with properties that depend on swelling state

- This captures the nonlinearity of swelling but retains the analytic tractability of linear poroelastic models

- Easy to extend to responsive gels: equilibrium (force-free) state just varies with stimulus **so the problems are decoupled**

- Oscillating stimulus → oscillating gel, but this doesn't necessarily → oscillating flow

- Adding anisotropy lets a reciprocal signal be converted to a non-reciprocal stroke

- Jellies can surf

Continuum Mechanics group seminar
University of Strathclyde, Wednesday 1st October 2025, 13:00

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Warwick

Grae Worster
Cambridge

more details can be found in

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