Linear stability analysis for the formation of wrinkles on confined swelling hydrogels

Interpore 2023, Edinburgh

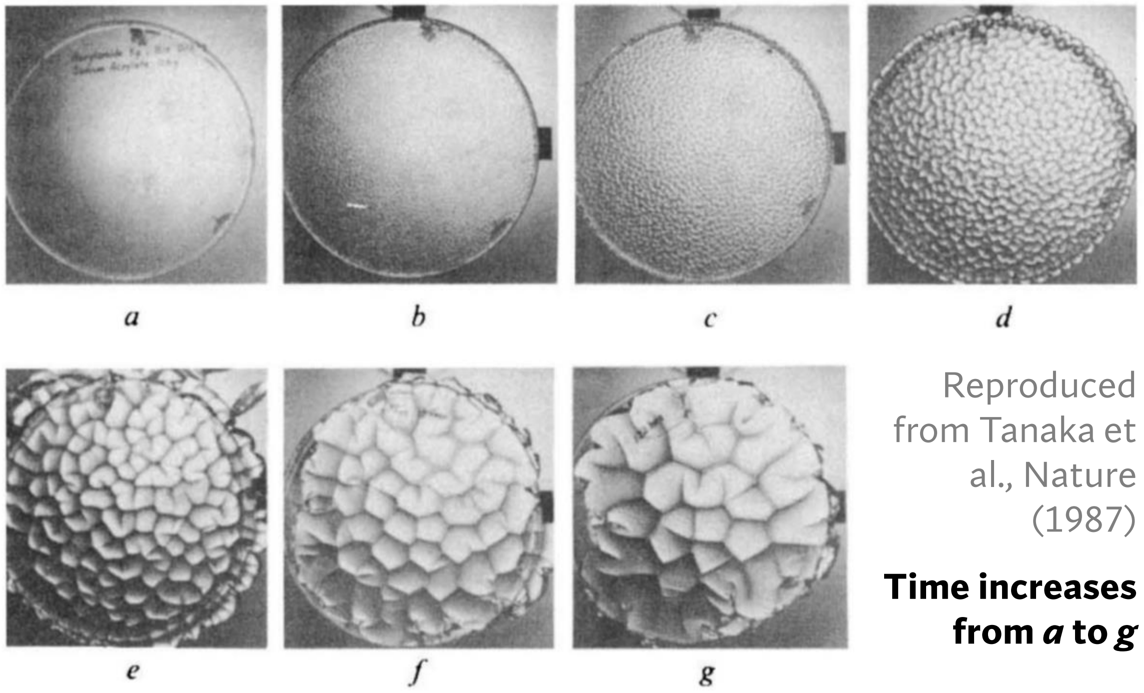
UNIVERSITY OF CAMBRIDGE

UKRI Natural Environment Research Council

Joseph Webber *and* Grae Worster

When do gels wrinkle?

- Wrinkling and buckling is observed in a number of different contexts, and not just in soft gels.
- Horizontal compressive stresses are relieved by the formation of surface buckles and wrinkles



Reproduced from Tanaka et al., Nature (1987)

Time increases from *a* to *g*

- In general:
  - Patterns smooth out in time ($\lambda \sim t^{1/2}$, Tanaka *et al.* (1992))
  - Start as wrinkles and then crease or fold.
- Conditions for the onset of instability are well-studied

LENS theory for hydrogels

Webber & Worster (2023)
A linear-elastic-nonlinear-swelling theory for hydrogels. Part 1. Modelling of super-absorbent gels *(J Fluid Mech)*

Webber, Etzold & Worster (2023)
A linear-elastic-nonlinear-swelling theory for hydrogels. Part 2. Displacement formulation *(J Fluid Mech)*

- Existing investigations use fully-nonlinear energy minimisation approaches but we use a linear-elastic-nonlinear-swelling approach.

Displacement-strain relations

Deviatoric strain ($\boldsymbol{\epsilon}$)

$$\boldsymbol{\epsilon} = \frac{1}{2}[\boldsymbol{\nabla}\boldsymbol{\xi} + \boldsymbol{\nabla}\boldsymbol{\xi}^{\mathrm{T}}] - \left[1 - \left(\frac{\phi}{\phi_0}\right)^{\frac{1}{n}}\right]\boldsymbol{I}\,; \qquad \boldsymbol{\nabla}\cdot\boldsymbol{\xi} = n\left[1 - \left(\frac{\phi}{\phi_0}\right)^{1/n}\right]$$

Number of dimensions (n)

Constitutive relation

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

with $\mu_s(\phi) = \mu_s$ and $\Pi(\phi) = K(\phi - \phi_0)/\phi_0$

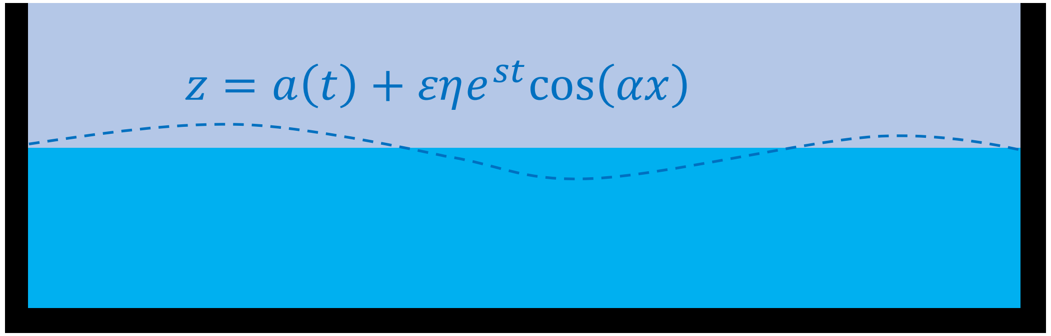
Polymer fraction evolution equation

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

with $k(\phi) = k$, $n = 2$, $\frac{\phi}{\phi_0} = \Phi$

Setup for linear stability analysis

- Gel of uniform polymer fraction Φ^*, depth a^* held between horizontal confines and brought into contact with water.
- Slowly swells to uniform polymer fraction Φ_1 and thickness a_1, starting at the interface and propagating through.
- Perturb the interface with a sinusoidal displacement and seek the growth rate s as a function of the wavenumber α.



$$\begin{pmatrix} \xi \\ \eta \\ p \\ \xi' \\ \eta' \\ p' \end{pmatrix}' = \underline{\underline{\boldsymbol{M}}}(z) \begin{pmatrix} \xi \\ \eta \\ p \\ \xi' \\ \eta' \\ p' \end{pmatrix}$$

No solutions where $\boldsymbol{\nabla} \cdot \boldsymbol{\xi} = 0$ permitted – all involve swelling or drying.

Boundary conditions at the interface

- On the lower surface:
  - No interstitial flow ($p' = 0$)
  - No normal displacement ($\eta = 0$)
  - No tangential stress ($\xi' = 0$)
- On the upper surface:
  - **Continuity of normal stress**

$$\sigma_{zz} = 0 \Rightarrow \left(\Phi^{1/2} - \frac{\mu_s}{K}\right)\alpha\xi + \left(\Phi^{1/2} + \frac{\mu_s}{K}\right)\eta' = 0$$

  - **Continuity of tangential stress**

$$\sigma_{xz} = 0 \Rightarrow \frac{\mu_s}{K}\xi' - \left[\frac{\mu_s}{K} + 2(\Phi - 1)\right]\alpha\eta = 0$$

  - **Continuity of pervadic pressure**

    Assume that $p = 0$ in the water

$$p + \eta\frac{\partial p_{base}}{\partial z} = 0 \Rightarrow p = \eta\left(1 + \frac{\mu_s}{K}\Phi^{-1/2}\right)\Phi'$$

Boundary conditions at the interface

- On the lower surface:
  - No interstitial flow ($p' = 0$)
  - N
  - N
- On th
  - **C**
  - **C**

$$(\mathcal{M}+\Phi^{1/2})\left[\frac{\mathcal{M}s}{2}\sinh(2\alpha)+\alpha(\mathcal{M}+\Phi_1-1)\left(s+\mathcal{M}\Phi_1^{1/2}\alpha\sinh(2\alpha)\right)\right]=$$

$$2\mathcal{M}\Phi_1^{1/4}\alpha(\mathcal{M}+\Phi_1-1)\cosh^2(\alpha)\tanh\left(\sqrt{\alpha^2+\frac{s\Phi_1^{-1/2}}{\mathcal{M}+\Phi_1^{1/2}}}\right)\times$$

$$\sqrt{\left(\mathcal{M}+\Phi_1^{1/2}\right)\left(s+\Phi_1^{1/2}\left(\mathcal{M}+\Phi_1^{1/2}\right)\alpha^2\right)}$$

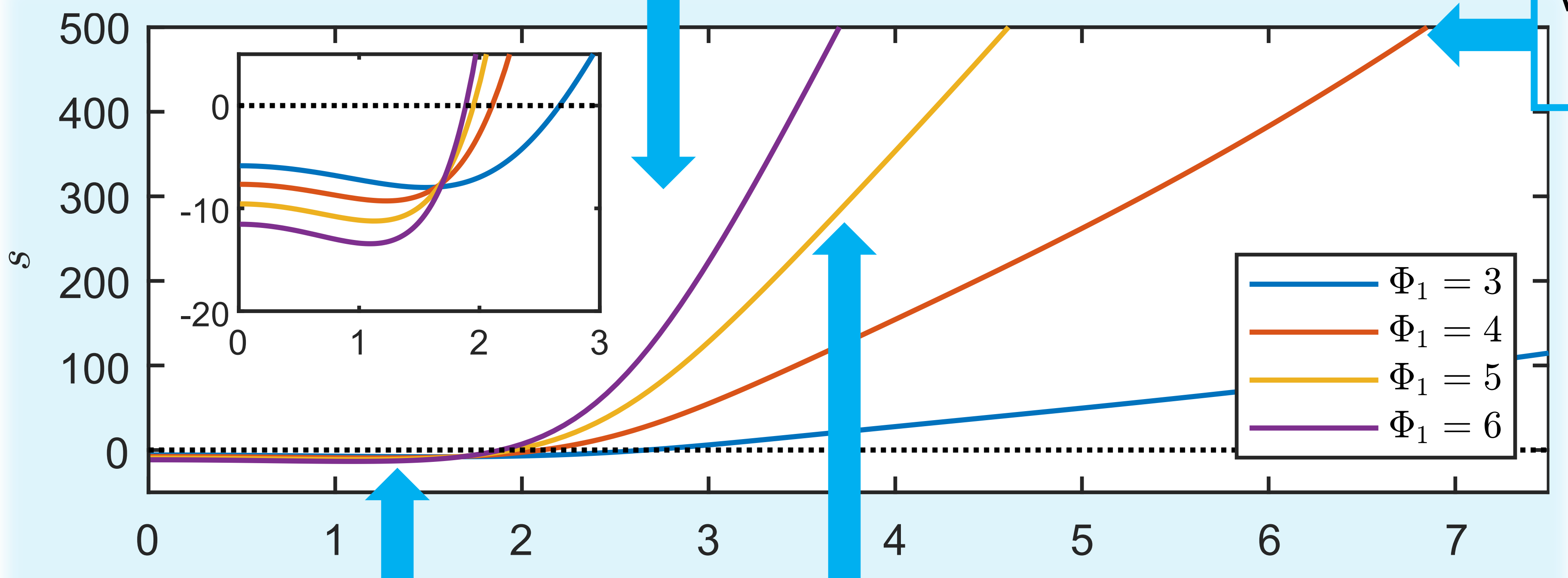
  - **Continuity of pervadic pressure**
    Assume that $p = 0$ in the water

$$p+\eta\frac{\partial p_{base}}{\partial z}=0 \Rightarrow p=\eta\left(1+\frac{\mu_s}{K}\Phi^{-1/2}\right)\Phi'$$

Uniformly-swollen gels

Higher polymer fractions go unstable at smaller wavenumbers

For short wavelengths, $s \sim \alpha^2$



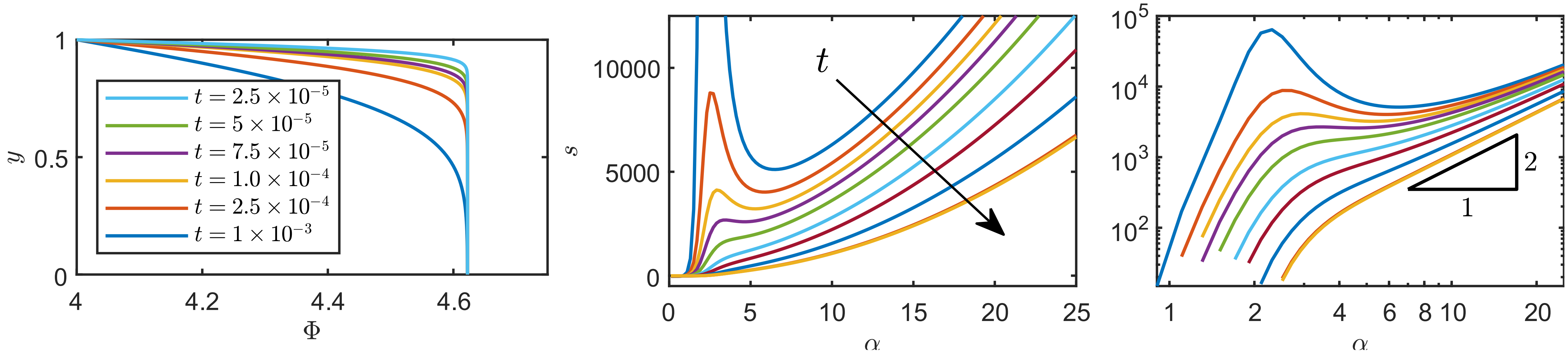
Stabilisation at low wavenumbers due to anchoring from the base of the gel

Growth rate increases with Φ

Stable **at all wavenumbers** if $\Phi_1 < \Phi_{crit} = (3+\sqrt{5})/2.$

Effect of the transient state

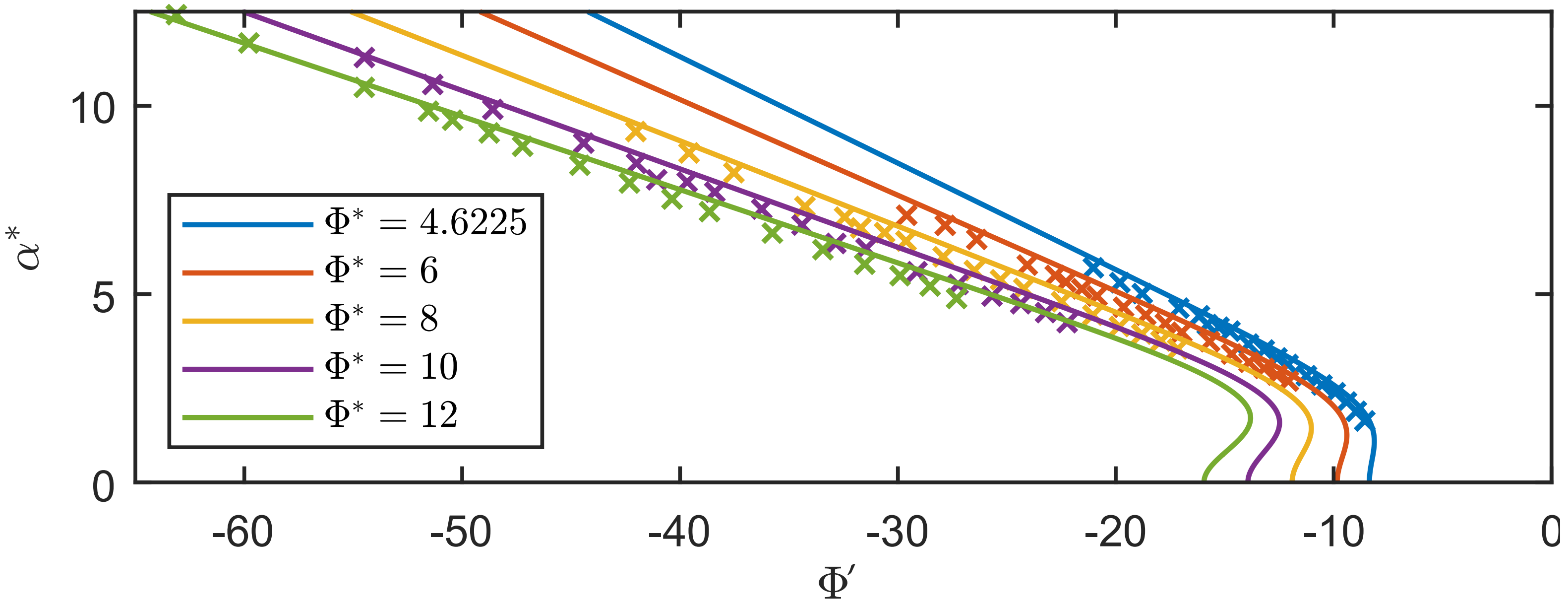
- Growth of wrinkles is driven by swelling: scaling vertical displacements with $\bar{\eta}$, the vertical interstitial velocity $\bar{u} \sim \bar{\eta}s$.
- Darcy's law gives $\bar{u} \sim \partial p/\partial z$ thus $\bar{\eta}s \sim \bar{p}/L$, where L is the vertical lengthscale, scaling like $1/\alpha$.
- Interfacial boundary condition on pervadic pressure: $\bar{p}/L$ increases in magnitude as $\Phi' \to -\infty$

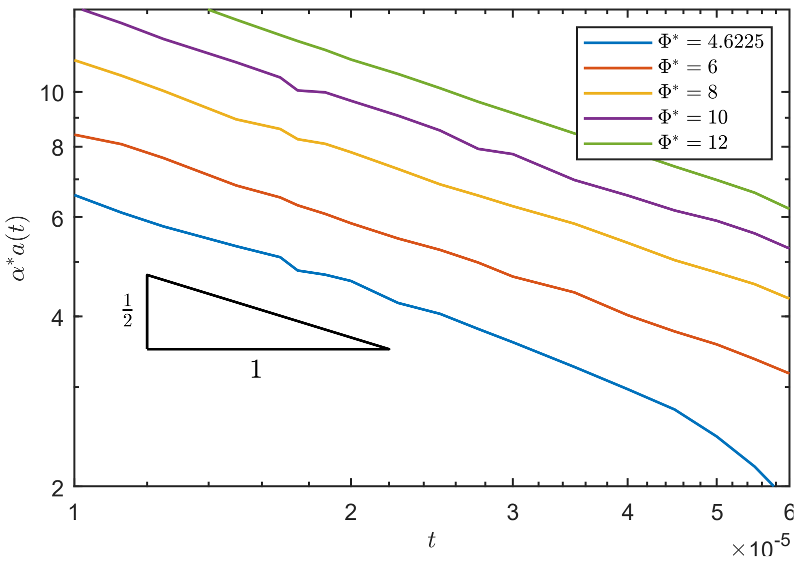


Rapid wrinkling at early times

- At early times, there is a most rapid growth at a finite wavenumber.

- Can we match up with the behaviour seen in experiments? Solve the uniformly-swollen problem with the pervadic pressure boundary condition and get a good approximation to the full problem.

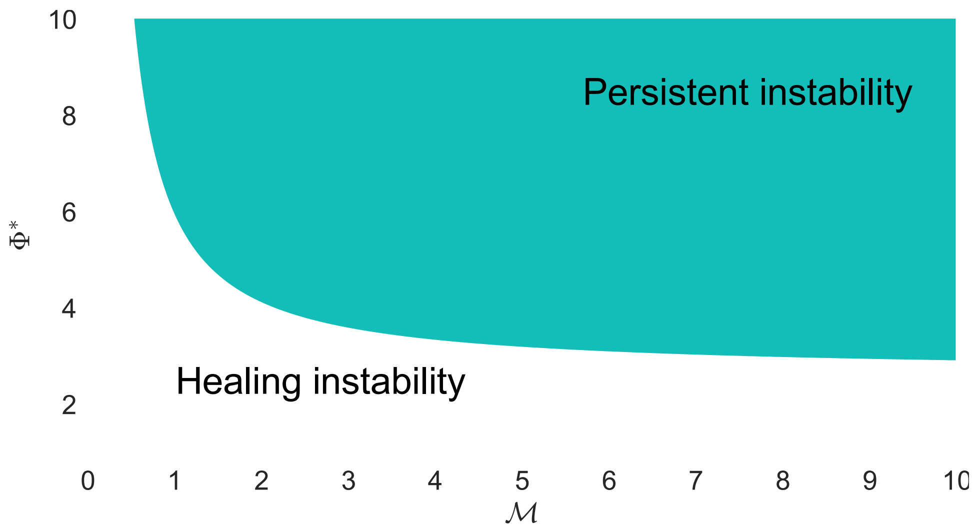


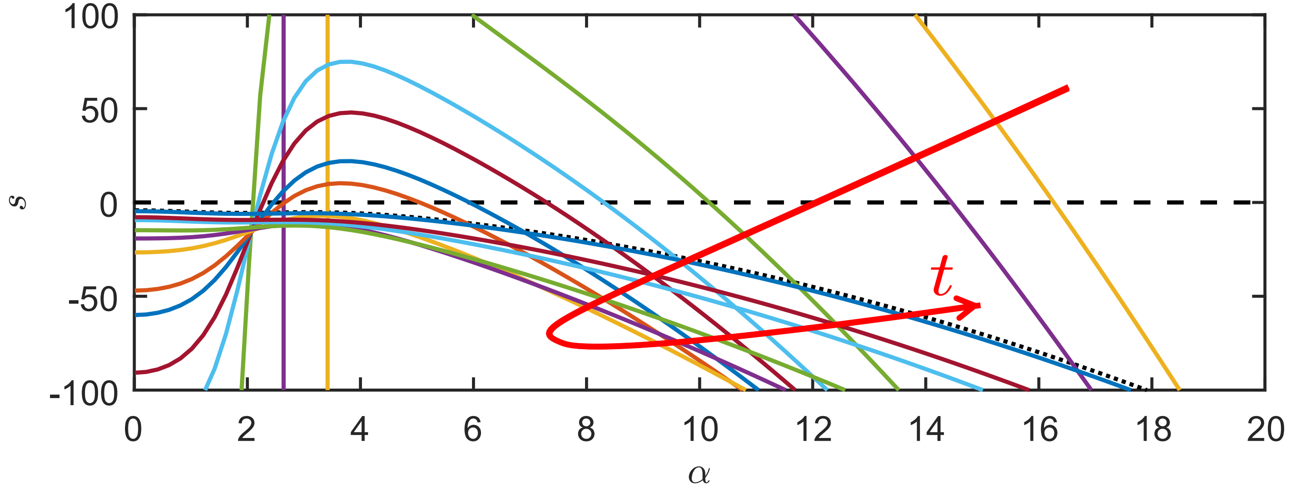


Healing of wrinkles

- Some experiments show a complete healing of wrinkles; from an initially unstable configuration, a stable steady state results.

- This can be explained using our model for wrinkle formation; the transient state destabilises the gel and its effects ease off as time progresses. Start with a $\Phi^* > \Phi_{\text{crit}} = \left(3+\sqrt{5}\right)/2$ and pick parameters such that $\Phi_1 < \Phi_{\text{crit}}$.





Conclusions

- Taking a model that linearises around small deviatoric strains but allows for arbitrarily large isotropic (swelling) strains allows us to model the wrinkling instability in compressed gels.
- It becomes clear that this instability is swelling-driven and we can derive growth rates as a function of material properties and wavenumber.
- The transient swelling state destabilises wrinkle formation, an effect seen most clearly at early times when a finite most unstable mode appears. Our model predicts that the wavenumber of this mode decreases like $t^{-1/2}$, as seen in experiments.
- The model also explains the healing behaviour seen in existing studies, where wrinkles form, smooth (by the behaviour described above) and then disappear.

UNIVERSITY OF CAMBRIDGE

UKRI Natural Environment Research Council