A Systematic Study of Swelling-Induced Instabilities of Hydrogels

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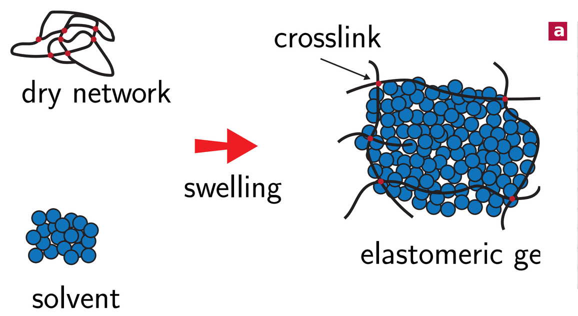
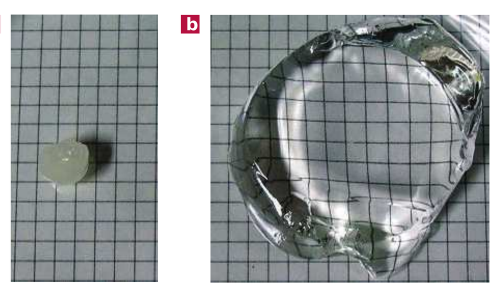
Introduction

Hydrogels and their intrinsic instabilities have a wide variety of applications due to their high levels of hydrophilicity, biocompatibility, and stimulus-responsive behaviours against light, temperature, chemical, and electric fields, including:

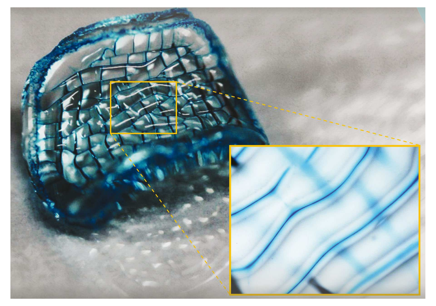
- Actuators
- Stretchable electronics
- Tissue engineering
- Biosensing
- Drug delivery
- Tunable wettability or adhesiveness ...



Hydrogel = polymer + water.



Large swelling of hydrogels[1].



Global and local instability modes[2].

The objective of this work includes three novel studies:

- develop a mixed & stable computational tool based on isogeometric analysis,
- provide an efficient framework for stability analysis of hydrogels,
- present numerical examples to show the robustness of the formulation.

Hydrogel Formulation

Coupled solid-deformation & fluid-diffusion

The governing equations are derived through the variational principle. Through the incompressibility constraint $\det(\boldsymbol{F}) = 1 + \nu C$ and $p := \mu/\nu$, we consider the mixed incremental potential;

$$\Pi(\boldsymbol{\varphi}, p) = \int_{\mathcal{B}} \Big[\psi - p(J - J_n) - \Delta t \ \phi^{\text{dissipation}}(-\nabla p; \boldsymbol{F}_n) \Big] \, dV - \Pi^{\text{EXT}}(\bar{\boldsymbol{\varphi}}, \bar{p})$$

with the coupled deformation $\boldsymbol{\varphi}$ ($\boldsymbol{F} = \nabla \boldsymbol{\varphi}$) and fluid pressure p. C: concentration of solvent molecules, ν: volume of a solvent molecule.
The stationarity of the potential implies

$$D\Pi[\delta \boldsymbol{\varphi}] = \int_{\mathcal{B}} \big[(\partial_{\boldsymbol{F}} \psi - p \ J \boldsymbol{F}^{-T}) : \delta \boldsymbol{F} \big] dV - D\Pi^{\text{EXT}}[\boldsymbol{\varphi}] = 0,$$

$$D\Pi[\delta p] = \int_{\mathcal{B}} \big[-(J - J_n)\delta p - \Delta t \partial_{\nabla p} \phi^{\text{dissipation}} \cdot \nabla \delta p \big] dV - D\Pi^{\text{EXT}}[\delta p] = 0 \ .$$

Material model & solvent diffusion:

We adopt the free energy in [3]:

$$\psi(\boldsymbol{F}) = \underbrace{\frac{1}{2} N k_B T (\boldsymbol{F} : \boldsymbol{F} - 3 - 2\ln(J))}_{\text{stretching part}} \underbrace{- \frac{k_B T}{\nu} \Big[(J-1)\ln(\frac{J}{J-1}) + \frac{\chi}{J} \Big]}_{\text{mixing part}}$$

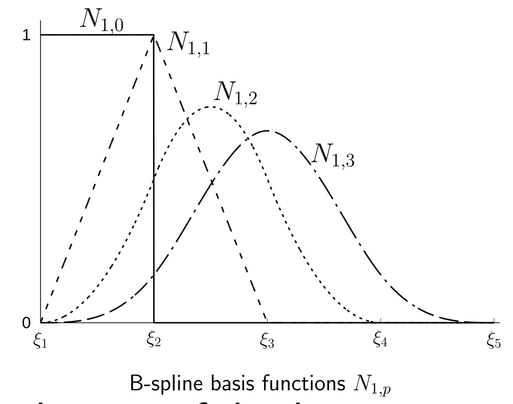
with N polymeric chains per volume, k_B Boltzmann factor, T temperature, χ polymer-solvent interaction parameter, D coefficient of diffusion.
The Cauchy stress & the fluid flux:

$$\boldsymbol{\sigma} = \frac{1}{J} \mathbf{P} \boldsymbol{F}^T \quad \text{and} \quad \mathbf{q} = \partial_{\nabla p} \phi^{\text{dissipation}} = -\frac{J_n - 1}{J_n} \frac{D\nu}{k_B T} \nabla p.$$

Mixed Isogeometric Analysis

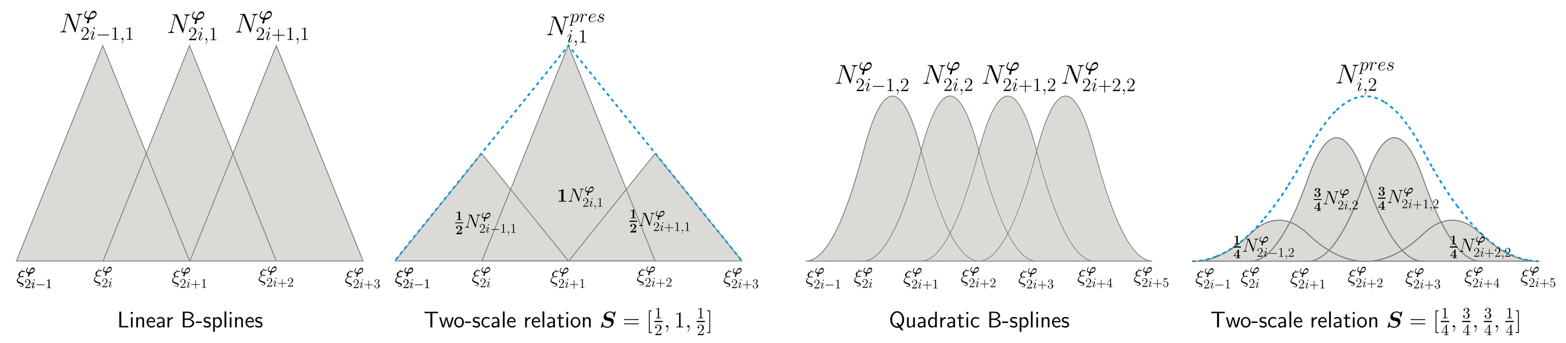
The displacements of the solid phase and the fluid pressure are interpolated independently using higher order NURBS.

$$\boldsymbol{\varphi}^h = \sum_{I=1}^{n_{en}^{\varphi}} N_I^{\varphi} \mathbf{u}_I \quad \text{and} \quad p^h = \sum_{I=1}^{n_{en}^{p}} N_I^{pres} p_I$$



Subdivision stabilization: Stable & efficient interpolations of displacement and pressure are achieved; the global definition of B-splines is conserved.
The two-scale relation is used to form the coarse basis (pressure interpolation)

$$N_{i,p}^{pres}(\xi) = \frac{1}{2^p} \sum_{k=0}^{p+1} \binom{p+1}{k} N_{2i+k-1,p}^{\varphi}(\xi) \quad \rightarrow \quad \boldsymbol{N}^{pres}(\boldsymbol{\xi}) = \boldsymbol{S} \ \boldsymbol{N}^{\varphi}(\boldsymbol{\xi})$$



Stability Analysis for Saddle-Point Formulation

We take the second derivative of the potential $\Pi(\boldsymbol{\varphi}, p)$ for stability assessment. Linearization, discretization, and assembly of finite elements in a discrete system:

$$\underbrace{\begin{bmatrix} \boldsymbol{K}_{\varphi\varphi} & \boldsymbol{K}_{\varphi p} \\ \boldsymbol{K}_{p\varphi} & \boldsymbol{K}_{pp} \end{bmatrix}}_{\boldsymbol{K}} \begin{bmatrix} \Delta \boldsymbol{u} \\ \Delta p \end{bmatrix} = \begin{bmatrix} \boldsymbol{R}_{\varphi} \\ \boldsymbol{R}_p \end{bmatrix}$$

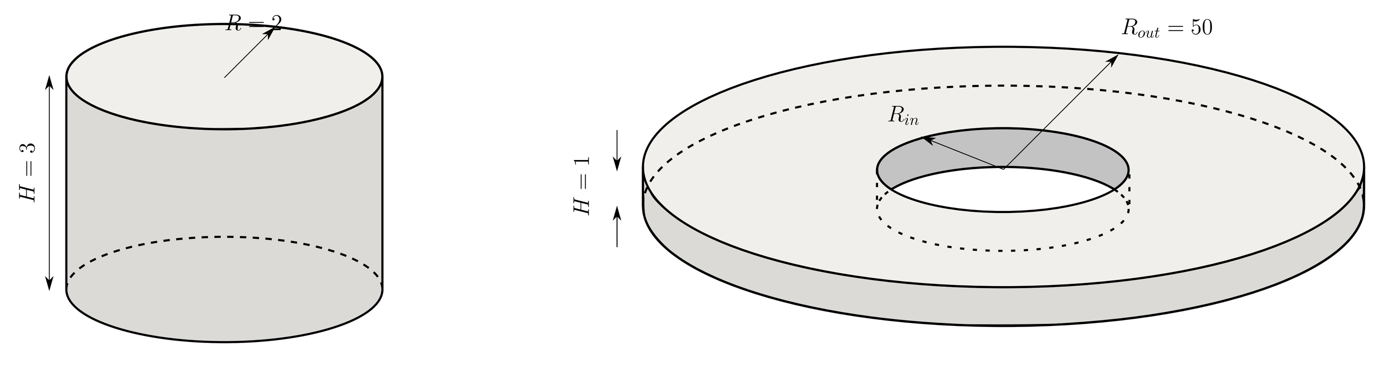
The resulting system $\boldsymbol{K}$ is ❶ symmetric ❷ indefinite (due to saddle point nature).
Eigenvalue analysis: Eigenvalues of saddle-point problems are not suitable for stability analysis. Use the Schur compliments of $\boldsymbol{K}$ for the eigenvalue analysis

$$\boldsymbol{K}^* = \boldsymbol{K}_{\varphi\varphi} - \boldsymbol{K}_{\varphi p} \boldsymbol{K}_{pp}^{-1} \boldsymbol{K}_{p\varphi} \quad \text{and} \quad \boldsymbol{K}^{**} = -\boldsymbol{K}_{pp} \ \text{(both symm. and PSD)}$$

Numerical Examples

Example I shows the rubostness of the current formulation in a compressed hydrogel disc and the stability is assessed via *numerical inf-sup test*.

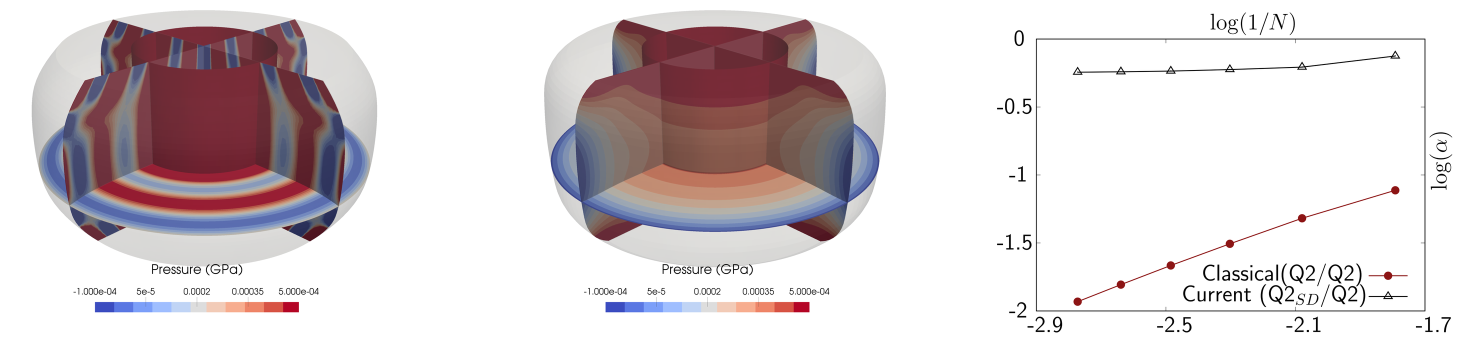
Example II emphasizes the proper structural stability criteria and captures the instabilities due to the free-swelling of a hydrogel corona.



Part I: Fully swollen hydrogel disc ($\lambda_0 = 3.3$) Part II: Thin hydrogel ring immersed in water ($\lambda_0 = 1.1$)

Ex I: Compression of a Fully Swollen Hydrogel

- Compressed by 25%.
- No-flow boundary conditions on the boundaries.

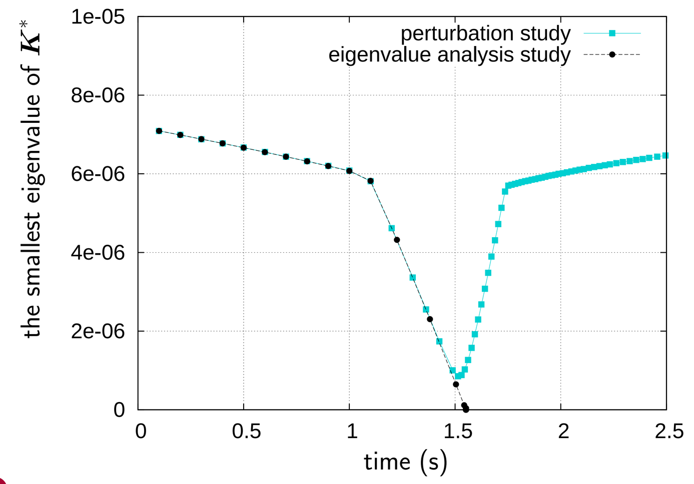


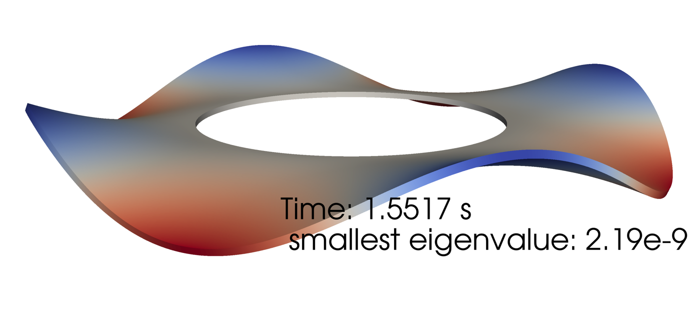
$Q2/Q2$ (Classical formulation) $Q2_{SD}/Q2$ (Current formulation) The inf-sup parameter α.

Observations: ❶ Classical ($Q2/Q2$) formulation suffers from pressure oscillations; failed the inf-sup test. ❷ Current ($Q2_{SD}/Q2$) formulation shows oscillation-free results; satisfies the inf-sup condition. ❸ Current stable $Q2_{SD}/Q2$ benefits from equal order interpolation & higher order regularity.

Ex II: Diffusion-Induced Buckling of a Hydrogel

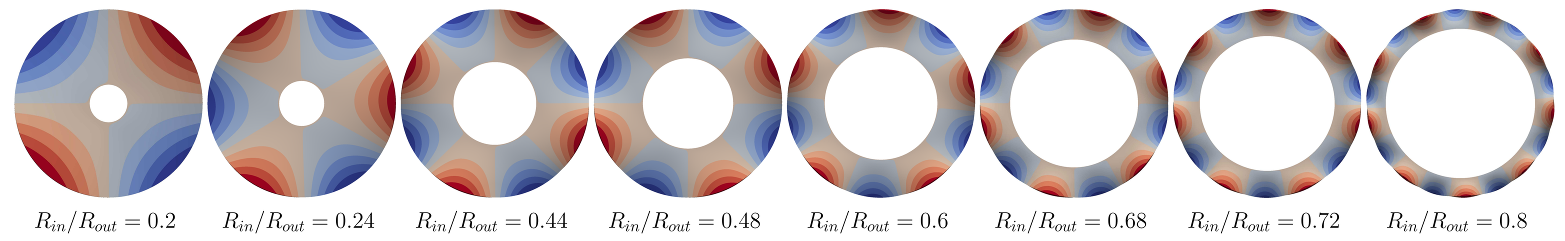
- **Study 1:** Eigenvalue analysis on $\boldsymbol{K}^*$. **Aim:** capture the onset & the shape.
- **Study 2:** Prescribing perturbations. **Aim:** capture the post-buckling curve.





First eigenmode at the onset of instability

- **Study 3:** Systematic investigation. **Aim:** determine the critical conditions.



Observations: ❶ The critical conditions are accurately captured for R_{in}/R_{out}. ❷ Concurrent eigenvalue analysis provides insights for prescribing perturbations. ❸ Eigenvalue analysis on $\boldsymbol{K}^*$ proves to be efficient for systematic studies.

Future Goals

Adopting the mixed and stable isogeometric analysis and the suitable stability citeria, we aim to investigate snap–through and snap–back instabilities recently observed in hydrogel applications, e.g. fringe, fingering, cavitation, and crease.

References

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