Freezing soft porous gels

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Yang et al., arXiv 2401.12871 (2024)







Why model the freezing of gels?

- Damage to tissues in freezing causes problems in transplantation, food science and plant physiology.
- Formation of ice crystals damages the structure of tissues whether in the form
 of small dispersed crystals, large chunks of pure ice, or mushy tissue-ice
 mixtures.
- Hydrogels provide a good analogue for soft tissue, with tunable properties and
 - High (>50%) water content
 - Porous structure
 - Elastic response
 - Can be manufactured to be clear

Mechanisms of damage



...but water only expands by around 10% in volume when freezing. Hence, strains are of the order of 2%.

 ${{\rm fracture\ strength}\over{
m shear\ modulus}}\lesssim 0.02$

From the literature, both of these values are of the order of 50 MPa, so tissue is **too strong** to be damaged this way.

Ice-entry temperature

• In micro- or nano-porous media, the freezing temperature is depressed owing to surface tension in the small pores



- Hence ice forms **outside** of the pores and it grows by drawing water from the porous medium.
- At the phase boundary, ice growth rates are set by heat fluxes.

$$ho \mathcal{L} V_n = - [\mathcal{K} \left({f n} \cdot
abla T
ight)]_{
m ice}^{
m gel}$$

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Cryosuction



There is then suction of water from the gel to grow ice more rapidly than pure thermal expansion alone.

This allows for the formation of large strains in the gel that can form cracks. Water freezing inside permeable hydrogel



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- Cooled lower boundary with a linear temperature profile from $T_c < T_m$
- Expect the formation of an ice lens, thicker in cooler regions

ENS theory for gels
$$\frac{D_{q}\phi}{Dt} = \boldsymbol{\nabla} \cdot \left\{ \frac{k(\phi)}{\mu_{l}} \times \left[\frac{K\phi}{\phi_{0}} + \mu_{s}(\phi) \left(\frac{\phi}{\phi_{0}} \right)^{1/2} \right] \boldsymbol{\nabla}\phi \right\}$$

 $\mathbf{u} \propto \nabla \phi$

Swelling and drying are driven by interstitial flows from swollen to dry regions, with effects from shear.

Webber & Worster and Webber, Etzold & Worster JFM, 2023



• Can solve the equations in the LENS formulation to get the ice thickness and polymer fraction field as the gel dries





Mode-II crack opening



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• Then deduce the displacement field of the hydrogel that is left behind, with two different boundary conditions





Mode-II crack opening



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• This shows that the crack opening depends heavily on boundary conditions





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• This shows that the crack opening depends heavily on boundary conditions





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- Soft hydrogels can be tuned to have similar properties to biological tissues, so are a good model for freezing damage
- Water does not freeze in place in the pores, so freezing damage is not caused by thermal expansion, but instead by cryosuction
- LENS modelling allows us to find that freezing damage is caused by mode-I crack opening when the tissue is free to slip (but confined) and mode-II when pinned

Webber & Worster (in prep) Cryosuction and freezing hydrogels *Proc. Roy. Soc. A*



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