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Surface instability of a gel disc in swelling

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Abstract. The swelling of a soft disc made of polymeric gel and attached to a fixed substrate is modeled using a variational method in nonlinear elasticity. A linear stability analysis is performed to detect the onset of a surface instability. An exact solution of the perturbed disc is found, and both the threshold values of the growth rates and the surface morphology are derived analytically.

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1 Introduction

Since the pioneering work of Tanaka et al. [1], the interest of soft matter physicists on the mechanical instabilities of polymeric gels has flourished. As underlined in the extensive review of Dervaux and Ben Amar [2], an incredible amount of articles has been published in the last decades on this subject, lying at the crossroad between several scientific communities. In fact, polymeric gels have been taken as reliable system models not only for studying the mechanisms of pattern formation in living tissues [3], but also for guiding the fabrication of micro-patterned surfaces [4, 5]. Novel experimental techniques have allowed a better comprehension of the dynamics of pattern formation in gels, attracting over the last years considerable attention from a theoretical viewpoint, especially concerning the buckling transition towards surface folds or creases [6].

2 The model

The aim of this work is to investigate the instability properties of a polymeric gel using the same experimental conditions originally studied by Tanaka et al. [1]. For this purpose, a disc made of a soft polymeric gel is initially considered having thickness H, being enclosed in a Petri dish of radius R_0. Choosing a polar coordinate system (R, Θ, Z) in the reference configuration C^r, the disc is attached at $Z = 0$. The gel undergoes a generic homogeneous swelling process, which is modeled using a virtual grown configuration C^g such that, in absence of geometrical constraint, its reference position $\mathbf{X}$ at time t is given by $R(t) = g_r(t)R(0)$ and $Z(t) = g_z(t)Z(0)$, where g_r and g_z are the growth rates in the radial and axial directions, respectively.
Considering that the soft gel is attached to the bottom substrate and radially confined at the disc side, the swollen position $\mathbf{x}$ in the spatial configuration C^s is given by $r(t) = R(0)$ and $z(t) = g_r^2 g_z(t)Z(0)$, so that the volume of the disc increase of a factor $J = g_r^2 g_z$. Assuming a neo-Hookean elastic behavior for the gel, the total elastic energy E_v reads:

$$E_v = 2\pi J \int\int \Big[\frac{\mu}{2}(\lambda_r^2 + \lambda_\theta^2 + \lambda_z^2 - 3) \\ -p(\lambda_r\lambda_\theta\lambda_z - 1)\Big]\, RdRdZ \qquad (1)$$

where λ_i $(i = r, \theta, z)$ are the elastic strains, μ is the shear modulus and p is the Lagrange multiplier arising from the incompressibility constraint, which acts like an hydrostatic pressure. The principal components of the Cauchy stress tensor inside the disc read $\sigma_{ij} = (\mu\lambda_i^2 - p)\delta_{ij}$, with δ_{ij} being the Kronecker delta, and the equilibrium equation reads:

$$r\sigma_{rr,r} + \sigma_{rr} - \sigma_{\theta\theta} = 0 \qquad (2)$$

where comma denotes differentiation. A basic swollen solution of Eq.(2) is given by $\lambda_r = \lambda_\theta = 1/g_r$, $\lambda_z = g_r^2$, representing an homogeneous deformation of the elastic disc. From the stress-free boundary condition $\sigma_{zz}(H) = 0$ one gets $p = g_r^4$, so that the swollen gels is subjected to a equi-biaxial stress state with $\sigma_{rr} = \sigma_{\theta\theta} = (g_r^{-2} - g_r^4)$, which is compressive in the case of swelling (i.e. for $g_r > 1$).
Having the aim to study the pattern formation at the free surface of the disc, it is now useful to perform a linear stability analysis of this basic swollen configuration. Instead of using the classical method of incremental elastic deformations, a variational formulation is used for solving exactly the incompressibility constraint. Assuming that the generic perturbation is plane and axial-symmetric, it is possible to define a canonical transformation using a virtual configuration C_v, made of mixed coordinates (r, Z, θ) [7]. In fact, an incompressible mapping can be defined using a non-linear stream function $\Psi(r, Z)$, as follows:

$$\mathrm{R}^2 = 2\ \Psi_{,\mathrm{Z}}; \qquad \mathrm{z} = \frac{\mathrm{J}\Psi_{,\mathrm{r}}}{\mathrm{r}}; \qquad \Theta = \theta \qquad (3)$$

According the volume-preserving mapping in Eq.(3), the elastic deformation gradient $\mathbf{F}_e = \partial \mathbf{x}/\partial \mathbf{X} : C^g \rightarrow C^s$ can be written as a function of the stream function, being:

$$\mathbf{F}_e = \begin{bmatrix} \frac{\sqrt{2\Psi_{,Z}}}{g_r \Psi_{Zr}} & -\frac{\Psi_{,ZZ}}{g_z \Psi_{Zr}} & 0 \\ \frac{J\sqrt{2\Psi_{,Z}}}{g_r \Psi_{Zr}}\left(\frac{\Psi_{,r}}{r}\right)_{,r} & \frac{g_r^2 \Psi_{,Zr}}{r} - \frac{g_r^2 \Psi_{,ZZ}}{\Psi_{,Zr}}\left(\frac{\Psi_{,r}}{r}\right)_{,r} & 0 \\ 0 & 0 & \frac{r}{g_r \sqrt{2\Psi_{,Z}}} \end{bmatrix} \quad (4)$$

Using a canonical transformation there is no need to introduce a Lagrange multiplier for imposing the incompressibility constraint, and the elastic energy in C^v takes the following simplified form:

$$E_v = 2\pi J \int_0^{R_0} \int_0^H \frac{\mu}{2}(F^e_{\alpha\beta} F^e_{\beta\alpha} - 3)\Psi_{,Zr} \; drdZ \quad (5)$$

where Einstein's summation on the repeated indices $\beta, \alpha = (r, Z, \theta)$ is assumed. Furthermore, a surface energy E_s exists at the free surface of the gel, which reads:

$$E_s = 2\pi\gamma \int_0^{R_0} r \sqrt{1 + \left(\frac{\Psi_{,r}}{J\ r}\right)^2_{,r}} \, dr \quad (6)$$

where γ is the surface tension at the free interface of the polymeric gel, so that we can define a characteristic capillary length of the disc as $L_{cap} = \gamma/\mu$. In general, such a surface energy also accounts for the soft skin created by oxygen inhibition of gelification in hydrogels.
Using a variational approach, the boundary value problem in nonlinear elasticity can be transformed into the minimization of the total potential energy of the swelling gel, such that $(\delta E_v + \delta E_s) = 0$. Considering that the energy terms are function of the stream function and its partial derivatives, the elastic equilibrium is given by the following volumetric Euler-Lagrange equation:

$$\left(\frac{\partial E_v}{\partial \Psi_{,lm}}\right)_{,lm} - \left(\frac{\partial E_v}{\partial \Psi_{,k}}\right)_{,k} = 0 \quad (7)$$

where the indices mean either r or Z. Two surface integrals represent the Euler-Lagrange conditions at the free surface $Z = H$ for arbitrary variations on Ψ and on $\Psi_{,Z}$, and read:

$$\frac{\partial E_v}{\partial \Psi_{,Z}} - \left(\frac{\partial E_v}{\partial \Psi_{,ZZ}}\right)_{,Z} - \left(\frac{\partial E_v}{\partial \Psi_{,rZ}} + \frac{\partial E_s}{\partial \Psi_{,r}}\right)_{,r} + \left(\frac{\partial E_s}{\partial \Psi_{,rr}}\right)_{,rr} = 0 \quad (8)$$

$$\frac{\partial E_v}{\partial \Psi_{,ZZ}} = 0 \quad (9)$$

3 Linear stability analysis

In order to perform a linear stability analysis of the swelling disc, let me consider a perturbation of the basic axial-symmetric solution by imposing:

$$\Psi(r, Z) = \frac{r^2 Z}{2} + \epsilon \cdot \Phi(r, Z) \quad (10)$$

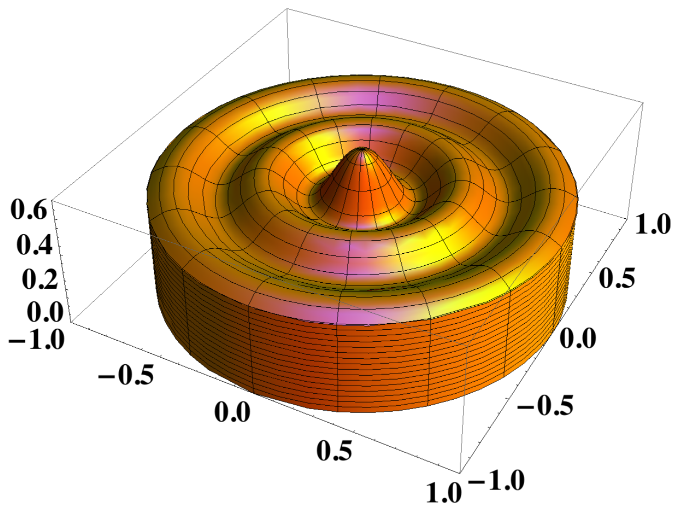

Fig. 1. Perturbed shape of the disc from Eq.(13). Parameters are set at ϵ=0.02, $JH = 0.5$, $R_0 = 1$, $u(H) = 3/2$, $u_{,Z}(H) = 1$, and $k_r = 16.4706$.

where $|\epsilon| \ll 1$ is the small amplitude of the perturbation defined by the infinitesimal stream function $\Phi(r, Z)$. Therefore, the bulk equilibrium equation for the perturbed state is given by substituting Eq.(10) in Eq.(7), and reads:

$$3J^2 g_z^2(-\Phi_{,r} + r\Phi_{,rr}) + r^2\left(J^2 g_z^2(-2\Phi_{,rrr} + r\Phi_{,rrrr}) - (1 + g_r^6)g_z^2(\Phi_{,ZZr} - r\Phi_{,ZZrr}) + g_r^2 r\Phi_{,ZZZZ}\right) = 0 \quad (11)$$

A solution of Eq.(11) can be found by searching for a separate variable form of the stream function. Recalling that:

$$r(rI_{n,r})_{,r} + (r^2 - n^2)I_n = 0 \quad (12)$$

where $I_n = I_n(r)$ indicates the Bessel function of the first kind of order n, it is possible to simplify Eq.(11) assuming that $\Phi(r, Z) = rI_1(k_r r) \cdot u(Z)$. In this case, using Eq.(3) the imposed perturbation at first order in ϵ reads:

$$r = R - \epsilon I_1(k_r r)u_{,Z}(Z); \; z = J(Z + \epsilon k_r I_0(k_r r)u(Z)) \quad (13)$$

Recalling the boundary condition at the disc side, being $r(t) = R_0$, from Eq.(13) k_r must belong to the discrete set of positive reals such that $I_1(k_r R_0)$=0. The shape of such a perturbed configuration of the gel disc is depicted in Fig. 1. Using Eq.(13), we can write the equilibrium condition in Eq.(11) as an ordinary differential equation on $u(Z)$, which reads:

$$g_r^2 u_{,ZZZZ} - (1 + g_r^6)g_z^2 k_r^2 u_{,ZZ} + J^2 g_z^2 k_r^4 u = 0 \quad (14)$$

and whose solution is given by:

$$u(Z) = a_1 e^{\lambda_1 Z} + a_2 e^{-\lambda_1 Z} + a_3 e^{\lambda_2 Z} + a_4 e^{-\lambda_2 Z} \quad (15)$$

where λ_1=$k_r g_z/g_r$, λ_2=Jk_r, and $a_1, .., a_4$ are constant parameters that must be determined through the four boundary conditions. Two of them are imposed by Eq.(13) considering the presence of the fixed bottom substrate, and read:

$$u(Z) = 0; \quad u_{,Z}(Z) = 0 \quad \text{at Z} = 0 \quad (16)$$

The two other boundary conditions are given by the Euler-Lagrange surface terms in Eqs.(8, 9), which can be simplified as follows:

$$J g_r^4 u_{,ZZZ} - J^2 g_z (1 + 2g_r^6) k_r^2 u_{,Z} - L_{cap} k_r^4 = 0 \qquad (17)$$

$$u_{,ZZ} + J^2 k_r^2 u = 0 \qquad (18)$$

which apply for $Z = H$, and correspond to vanishing incremental stresses at the free surface. Using the solution given by Eq.(15) with the four boundary conditions in Eqs.(16-18), we get the following expression for the incremental displacement:

$$u(Z) = \sinh(\lambda_1 Z) - \frac{\sinh(\lambda_2 Z)}{g_r^3} - (\cosh(\lambda_1 Z) \\ -\cosh(\lambda_2 Z)) \frac{(1+g_r^6)\sinh(\lambda_1 Z) + 2g_r^3 \sinh(\lambda_2 Z)}{(1+g_r^6)\cosh(\lambda_1 Z) - 2g_r^6 \cosh(\lambda_2 Z)} \qquad (19)$$

whilst the dispersion relation for the disc instability is given by:

$$g_r^3 \sinh(\lambda_1 H) \left[(g_r^6 - 1) k_r L_{cap} \cosh(\lambda_2 H) \\ - g_r^6 g_z^4 A_1 \sinh(\lambda_2 H) \right] - 4 g_r^4 J^4 (1 + g_r^6) + \cosh(\lambda_1 H) \cdot \\ \left[g_r^6 g_z^4 A_2 \cosh(\lambda_2 H) - (g_r^6 - 1) k_r L_{cap} \sinh(\lambda_2 H) \right] = 0 \qquad (20)$$

with $A_1 = (1 + 6g_r^6 + g_r^{12})$ and $A_2 = (1 + 2g_r^6 + 5g_r^{12})$.

4 Results and discussion

Neglecting the presence of a surface energy and setting $L_{cap} = 0$ in Eq.(20), one gets the classical surface instability described by Biot [8]. In fact, a radial growth threshold g_r^* exists when $k_r \to \infty$, given by the condition $(A_2 - g_r^3 A_1) = 0$, and reads:

$$g_r^* = \sqrt[3]{1 + \frac{(54 - 6\sqrt{33})^{\frac{1}{3}}}{3} + \frac{(18 + 2\sqrt{33})^{\frac{1}{3}}}{3^{\frac{2}{3}}}} \sim 1.5011 \qquad (21)$$

which is the real root of $(-1 - g_r^3 - 3g_r^6 + g_r^9) = 0$.
Interestingly, this surface instability mechanism only depends on the radial growth of the disc, and the critical value g_r^* is the same found after sinusoidal perturbation of a square gel layer under biaxial compression [9]. The problem of having a vanishing wavelength for the Biot surface instability is overcome when taking into account the presence of a surface tension. As shown in Figure 2 in the case of isotropic growth ($g_Z = g_r$), setting a non-zero L_{cap} in Eq.(20) one gets not only an increased growth threshold, if compared to Eq.(21), but also a finite critical wavenumber k_r^* which determines the morphology of the perturbed disc. Solving numerically Eq. (20), the threshold value for the growth rate and the wavelength $\Lambda = 2\pi/k_r^*$ of the surface instability are depicted in Figure 3 as a function of the capillary ratio L_{cap}/H. Although such results are shown as continuous curves for the sake of simplicity, it is important to recall that an instability can only occur for the discrete values of k_r^* such that $I_1(k_r^* R_0)$=0, in order to respect the boundary condition at the disc side.
In particular, we find that increasing the surface tension one always obtains a higher instability threshold, whose value is dependent on the growth anisotropy ratio g_z/g_r. Moreover, taking the limit $L_{cap}/H \ll 1$, one finds logarithmic corrections to the Biot formula of the growth

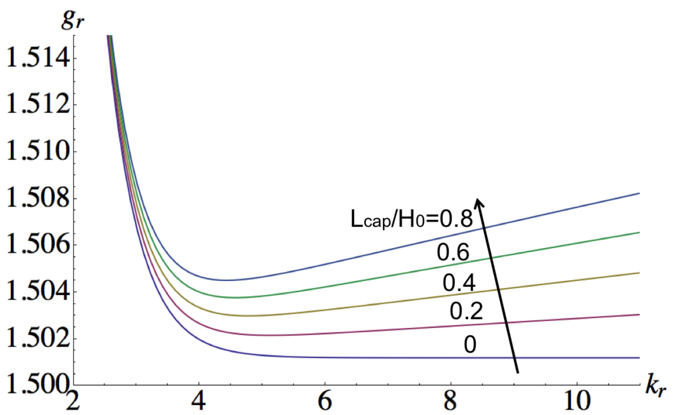

Fig. 2. Solution of the dispersion relation in Eq.(20) given as the radial growth rate g_r versus the wavenumber k_r. The results are obtained setting $g_z = g_r$ and $H = 1$, and depicted for the labeled values of L_{cap}.

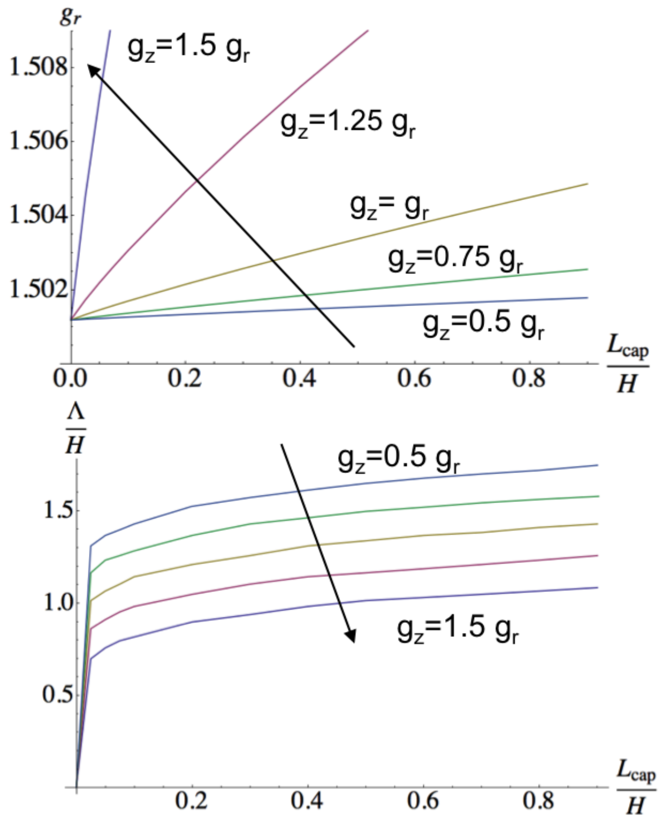

Fig. 3. (Top) Threshold value of radial growth rate g_r versus the capillary ratio L_{cap}/H. (Bottom) Wavelength $\Lambda = 2\pi/k_r$ of the surface instability as a function of L_{cap}/H. The curves are depicted for the labeled values of anisotropic growth (g_z/g_r= 0.5, 0.75, 1, 1.25, 1.5)

threshold and critical wavenumber, as reported in previous theoretical and experimental studies [9, 10].
For higher ratios L_{cap}/H, the wavelength Λ of the surface pattern is finally about the order of the thickness of the gel disc, in accordance with the original observations of Tanaka et al. [1], as well as with more recent experiments [11].
Although the analysis of the nonlinear regime of the disc instability if out of the scopes of this study, it is useful to add a final consideration about the nature of the bifurcation of the growing disc. In particular, from Eqs.(5,6) the series expansion of the total potential energy E_p at the leading orders in ϵ reads:

$$E_p = E_v + E_s = \left(\frac{\mu}{2}(2 - 3g_r^2 + g_r^6)g_z H + \gamma\right) R_0^2/2 \\ +\epsilon^2/2 \cdot E_2 + \epsilon^3/3 \cdot E_3 + \epsilon^4/4 \cdot E_4 \qquad (22)$$

where E_2, E_3, E_4 are integration terms [12]. At a weakly non-linear regime the amplitude ϵ of the perturbation can be fixed by minimizing the potential energy of the disc, so that $\delta E_p/\delta\epsilon = 0$. Therefore, the presence of a non-zero term E_3 in Eq.(22) indicates that the bifurcation of the elastic stability is more likely subcritical, and the amplitude will be fixed by $E_2 + E_3\epsilon + E_4\epsilon^2 = 0$. The subcriticality of the bifurcation would explain the hysteresis in the crease formation observed experimentally. In fact, the onset, the morphology and the cycle-to-cycle memory of surface folds in compressed hydrogels seem to be dominated by the distribution of heterogeneous defects [13].

5 Conclusion

In summary, an analytical solution of the linear stability analysis for the constrained swelling of a gel disc has been derived. Performing an axial-symmetric perturbation, both the total volume change and the growth anisotropy are found to control the onset of the instability. The ratio between the capillary length L_{cap} and the thickness H of the disc is found to determine the wavelength of the surface pattern. Further nonlinear treatments are needed to explain the observed transition from simple surface undulations to both tree-like and honeycomb structures [14]. A deeper understanding of the mechanisms regulating pattern formation in swelling polymeric materials is of utmost importance for designing smart materials with tunable surface properties.

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