

# Gel Debonding from a Rigid Substrate

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July 10, 2019

## Abstract

We consider the problem of debonding of a thin gel domain from a rigid substrate. Starting with a variational approach involving the total energy of a gel, we formulate the boundary value problem of the governing equations in two-space dimensions. We consider the case that the aspect ratio,  $\eta$ , the quotient of the thickness of the membrane with respect to its length, is very small. We assume that the gel is partially debonded at the dimensionless horizontal location denoted by  $0 < \delta < 1$ . The appropriate limiting problem with respect to  $\eta$ , with fixed  $\delta$ , yields an approximate solution corresponding to a deformation that is homogeneous both on the bonded part and on the debonded part of the gel, but whose gradient and vertical component are discontinuous across the interface  $x = \delta$ . This approximate solution determines, up to first order, the energy release rate on  $\delta$ , giving the critical value for the gel thickness at which it becomes unstable against debonding.

## A Introduction

A variational model is presented for the debonding of a polymer gel from a rigid substrate. Our motivation comes from the design of the synthetic polymers which protect various implants in the medical industry (such as pacemakers, bone implants or intervertebral disks). Due to the high moisture environment of the body, the materials forming the device become gel, and swell at often very different ratios. As a result, high stress builds at interfaces between different materials that may result in debonding.

We consider a gel as an interactive mixture of polymer and fluid, although most materials also exhibit gel behavior. A signature mechanical response of gels when no boundary constraints are imposed is the free swelling in the presence of solvent: it absorbs the liquid and swells until it reaches (or tends to reach) a certain equilibrium in which each subpart of the gel increases its volume by an optimal factor (as explained in the celebrated Flory-Huggins theory[6] [1] [10] [14] [3] [9] [13]). Dynamical models of gel swelling have been studied by Doi and collaborators [4] and are extensively discussed in [5]. Models that follow the point of view of mixture theory have been explored in [3]. The formation of wrinkles in swollen gels is studied in [12].

In contrast with free swelling, when the polymer is attached to a rigid substrate, it is allowed to grow only perpendicularly to the substrate instead of uniformly in every direction. The swelling ratio depends on temperature and, on ionic content in the solvent and electric charge in the polymer, in the case of polyelectrolyte gels. This confinement and its consequences are more significant when the polymer is only a thin film, leading to stresses at the interface having the potential to trigger the debonding.

Based on the free-discontinuity approach to fracture in elasticity [8] [7] [2], we study the debonding of the gel as the competition between a mechanical energy (comprising both the elastic and the mixing terms) and a fracture energy (required for the gel to continue to detach from the substrate). As a result, the debonding will initiate, and/or progress, only if the mechanical energy released by this increase of free surface (which reduces the confinement and allows the gel to expand isotropically, balancing the osmotic pressure in an optimal manner) is sufficient to overcome the fracture energetic barrier. A simple 2-dimensional situation is considered in which the gel/substrate interface is planar and the film is allowed to debond only from its ends. For every  $0 < \delta < 1$ , minimizers of the mechanical energy are sought among all deformations of the gel film that remain attached along a fraction  $\delta$  of the original interface. For small aspect ratio,  $\eta > 0$ , we propose a formal asymptotic expansion of the solution as the sum of an outer contribution, valid away from the transition region  $x = \delta$ , plus an interior one that regularizes the former.

The relevant information on the deformation map and the energy released by delamination is contained in the leading term of the outer expansion. Indeed, approximating the governing equations for small  $\eta$  away from the interface, yields an approximate solution  $(u_0, v_0)$  whose vertical component has a jump discontinuity at the interface. The components  $(u_{0_x}, v_{0_y})$  of the gradient of the approximated deformation map are constant on each side of the interface  $x = \delta$ , which is consistent with the gel experiencing a volume phase transition. The dependence of the total energy on  $\delta$  is studied, giving the maximum gel thickness allowed for the system to be predicted to be stable against debonding.

In section 2, we present the model to be analyzed and derive the expressions of the Piola-Kirchhoff stress tensors. In section 3, we formulate the equilibrium boundary values problem corresponding to a plane gel that has detached from the substrate along the portion of the boundary  $\delta < x < 1$ . We formally compute the solution of the dimensionless form of the governing equations in the limit as the aspect ratio  $\eta$  of the gel vanishes. The energy of the system as a function of the debonded length  $(1 - \delta)$  is studied in section 4. The case of a clamped boundary condition is also presented. Numerical simulations are presented in section 5, together with data consistent with medical device applications. The conclusions are presented in section 6.

## B Model

We assume that a gel is a saturated, incompressible and immiscible mixture of elastic solid and fluid. In the reference configuration, the polymer occupies a domain  $\Omega_B \subset \mathbf{R}^3$ . Deformation of the polymer may occur by external forces and by the gain or loss of fluid. The solid undergoes a static deformation according to the one-to-one, differentiable map

$$\mathbf{y} = \varphi_B(\mathbf{x}), \quad \text{such that } \det(\nabla \varphi_B) > 0, \quad \mathbf{x} \in \Omega_B. \quad (1)$$

The domain occupied by the gel at time  $t \geq 0$  is  $\varphi_B(\Omega_B)$ . We label the polymer and fluid components with indices 1 and 2, respectively. According to the theory of mixtures, a point  $\mathbf{y} \in \varphi_B(\Omega_B)$  is occupied by, both, solid and fluid at volume fractions  $\phi_1 = \phi_1(\mathbf{y})$  and  $\phi_2 = \phi_2(\mathbf{y})$ , respectively. An immiscible mixture is such that the constitutive equations depend explicitly on the volume fractions  $\phi_i, i = 1, 2$ . We let  $\rho_i$  denote the mass density of the  $i$ th component (per unit volume of gel). It is related to the intrinsic density,  $\gamma_i$ , by the equation  $\rho_i = \gamma_i \phi_i, i = 1, 2$ . Moreover  $\gamma_i = \text{constant}, i = 1, 2$  define an incompressible mixture. The assumption of saturation of the mixture expressing that no species other than polymer and fluid are present, is expressed by the constraint

$$\phi_1 + \phi_2 = 1. \quad (2)$$

As a consequence, it suffices to know the volume fraction of polymer, which henceforth will be denoted simply by  $\phi$ . We assume that the reference configuration of the gel corresponds to the domain  $\Omega_B$  occupied by the dry polymer. The equation of balance of mass of the polymer states that, for every subset  $\tilde{\omega} \subset \Omega_B$ ,

$$\int_{\varphi_B(\tilde{\omega})} \phi(\mathbf{y}) d\mathbf{y} = \int_{\tilde{\omega}} d\mathbf{x}. \quad (3)$$

Formally, it reduces to a pointwise relation

$$\phi(\varphi_B(\mathbf{x}))J(\mathbf{x}) = 1, \quad \text{for all } \mathbf{x} \in \Omega_B, \quad (4)$$

where we have set

$$J(\mathbf{x}) = \det F(\mathbf{x}), \quad \text{with} \quad F(\mathbf{x}) = \nabla \varphi_{\mathcal{B}}(\mathbf{x}). \quad (5)$$

We point out that the volume fractions  $\phi_1$  and  $\phi_2$  are defined in the deformed configuration. This is also the case to the Flory-Huggins energy. Therefore, we must formulate this balance law and Eulerian energies of the problem assuming, for the time being, the deformation map does not embody formation of cavities.

Forthcoming calculations involve expressing vectors and tensors in components. For this, we let  $\{\mathbf{e}_1, \mathbf{e}_2, \mathbf{e}_3\}$  denote the canonical basis of the Euclidean space  $\mathbb{R}^3$  and  $\{x_1, x_2, x_3\}$  the associated coordinate system.

## B.1 Energy of the Gel

Three different contributions to the total mechanical energy of a gel will be considered. First, there is an internal elastic energy  $\int_{\Omega_{\mathcal{B}}} W(F(\mathbf{x})) d\mathbf{x}$  associated to the deformation of the polymer.

For simplicity, we assume that

$$W(F) := \frac{\mu}{2} |F|^2, \quad \mu > 0, \quad (6)$$

with an elasticity modulus  $\mu$  that has dimensions of energy density. However, part of the analysis could still be carried out for any  $W$  of Hadamard form:

$$W(F) = \mu \frac{|F|^p}{p|I|^{p-2}} + G(\det F), \quad \det F \geq \phi_0, \quad (7)$$

$p \geq 2$ , and  $G(J)$  is convex and such that

$$\lim_{J \rightarrow \infty} G(J) = +\infty. \quad (8)$$

Second, we consider the Flory-Huggins energy of mixing ([5], page 18; [11], page 143),  $\int_{\varphi_{\mathcal{B}}(\Omega_{\mathcal{B}})} \mathcal{W}_{\text{FH}}(\phi(\mathbf{y}), 1 - \phi(\mathbf{y})) d\mathbf{y}$ , where

$$\mathcal{W}_{\text{FH}}(\phi_1, \phi_2) := \frac{K_B \theta}{V_m N_1} \phi_1 \ln \phi_1 + \frac{K_B \theta}{V_m N_2} \phi_2 \ln \phi_2 + \frac{\chi K_B \theta}{2V_m} \phi_1 \phi_2. \quad (9)$$

Here  $V_m$  represents the volume occupied by one monomer,  $K_B$  the Boltzmann constant,  $N_1, N_2$  the number of lattice sites occupied by the polymer and the solvent, respectively, and  $\frac{\chi}{2}$  is the Flory parameter. The first and second terms in (9) correspond to the entropies of the polymer and fluid, respectively, and the third term represents repulsive forces between the two components.

Note that the mixing energy density must be integrated on  $\varphi_{\mathcal{B}}(\Omega_{\mathcal{B}})$  since it is in the current (swollen) configuration where the interaction between the two species takes place. So as to be able to compare it against the elastic energy, we map it back to the original (Lagrangian) variables, obtaining

$$\int_{\Omega_{\mathcal{B}}} \mathcal{W}_{\text{FH}}(\phi \circ \varphi_{\mathcal{B}}, 1 - \phi \circ \varphi_{\mathcal{B}}) \cdot \det F(\mathbf{x}) d\mathbf{x}.$$

Finally, we assume that the gel is subject to a prescribed constant external pressure  $P_0 > 0$ . The total energy of the system associated to a deformation  $\varphi_{\mathcal{B}}$  needs to take into account the mechanical work done against this external pressure. Since we assume the pressure to be constant, the total work is given by  $P_0 \cdot \text{volume}(\varphi_{\mathcal{B}}(\Omega_{\mathcal{B}}))$  regardless of the speed or intermediate states in the evolution of the gel from the initial state to the final state given by  $\varphi_{\mathcal{B}}$ . All in all, the energy of the gel is

$$E = \int_{\Omega_{\mathcal{B}}} \left( W(F(\mathbf{x})) + \left( \mathcal{W}_{\text{FH}}(\phi \circ \varphi_{\mathcal{B}}, 1 - \phi \circ \varphi_{\mathcal{B}}) + P_0 \right) \cdot \det F(\mathbf{x}) \right) d\mathbf{x}. \quad (10)$$

By introducing the notation

$$\nu := \frac{K_B \theta}{V_m N_2}, \quad J := \det F,$$

and the dimensionless expression

$$\begin{aligned} H(J) &:= \frac{1}{\nu} J \mathcal{W}_{\text{FH}}\left(\frac{1}{J}, 1 - \frac{1}{J}\right) \\ &= \frac{N_2}{N_1} \ln \frac{1}{J} + (J - 1) \ln\left(1 - \frac{1}{J}\right) + \frac{\chi N_2}{2} \left(1 - \frac{1}{J}\right), \quad J > 1 \end{aligned} \quad (11)$$

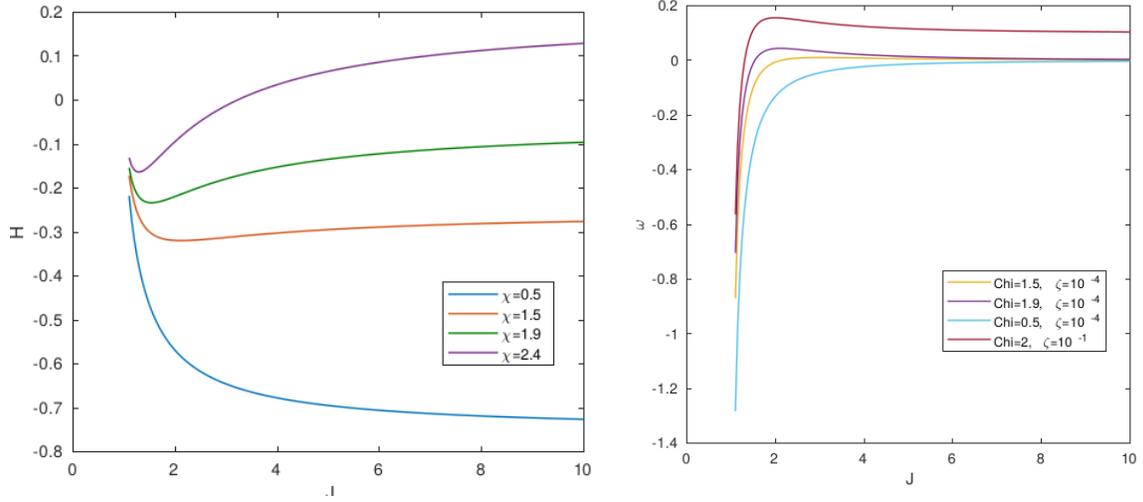


Figure 1: Plots of  $H(\phi)$  (left) and  $\omega(\phi)$  (right) as functions of the Jacobian determinant  $J$ . Note the loss of convexity of  $H$  as  $\chi$  increases. Likewise, the profiles of  $\omega$  become monotonic for  $\chi$  large.

the total energy can be rewritten as

$$E = \int_{\Omega_B} \left( \frac{\mu}{2} |F|^2 + \nu H(J) + P_0 J \right) dx. \quad (12)$$

Hereforth, we take values

$$N_1 = 1000, \quad N_2 = 1, \quad 0 < \chi < 1.0326, \quad (13)$$

as is customary in applications of interest. In particular, the choice of  $N_2$  is standard when the second component of the mixture is a solvent. The last inequality corresponds to mixing regimes [11] where  $H$  is convex and minimizers of (12) are expected to exist under appropriate boundary conditions. However, note that convexity of  $H$  may be lost in physical situations where phase separation occurs. Determining the values of  $\chi$  such that  $H(J)$  is monotonic and finding the critical,  $\chi_c$ , for which  $H(J)$  loses monotonicity is very relevant to our problem ([5], chapter 3). In the later case, the gel separates into the polymer and the fluid phases, which is well outside the regime relevant to the applications that we consider.

Let us, then, investigate changes of sign of  $H'(\phi)$  by finding the zeros of  $H''(J)$ , for  $J > 1$ . A simple calculation gives

$$H''(J) = -\left(1 - \frac{N_2}{N_1}\right)J^{-2} + \frac{1}{J(J-1)} - \chi N_2 J^{-3}.$$

Note that  $H''(J) = 0$ , for  $J > 1$ , if and only if

$$\left(1 - \frac{N_2}{N_1}\right)J^2 + \left(1 - \frac{N_2}{N_1} - \chi N_2\right)J + \chi N_2 = 0.$$

This quadratic equation has a solution  $J_0$  with  $J_0 > 1$  provided  $\chi \geq \frac{1}{N_2} + \frac{1}{N_1} + \frac{1}{\sqrt{N_1 N_2}}$ . Consequently, the monotonicity regime of  $H(J)$  corresponds to

$$\chi < \frac{1}{N_2} + \frac{1}{N_1} + \frac{1}{\sqrt{N_1 N_2}}. \quad (14)$$

From now on, we will assume that (14) holds.

## B.2 Cauchy and Piola-Kirchhoff stress tensors

We now introduce the Piola-Kirchhoff stress tensor, which can be obtained by differentiating the energy density in (10) with respect to the deformation gradient. Denoting the cofactor matrix of  $F$  by  $F^c$  we obtain

$$P = \mu F + \nu \omega(J) F^c, \quad (15)$$

where we have set

$$\omega(J) := H'(J) + \frac{P_0}{\nu} = \left(1 - \frac{N_2}{N_1}\right) J^{-1} + \ln(1 - J^{-1}) + \frac{\chi}{2} J^{-2} + \zeta, \quad \zeta := \frac{P_0}{\nu}. \quad (16)$$

Note that  $\nu H'(J)$  corresponds to the osmotic pressure and that  $\omega(J) = H'(J) + \frac{P_0}{\nu}$  takes also into account the external pressure.

The traction boundary conditions due to the external pressure correspond to the balance of internal and external forces at the boundary, namely, the relation  $P \mathbf{n}_0 = 0$ , where  $\mathbf{n}_0$  denotes the unit normal to  $\partial\Omega$ . Using (15), the condition can be formulated as

$$\omega(J) F^c \mathbf{n}_0 = -\gamma F \mathbf{n}_0, \quad (17)$$

where

$$\gamma := \frac{\mu}{\nu}. \quad (18)$$

## B.3 Minimum energy solutions

As a consequence of the isotropy assumption on the gel, the pressurized states for it (those in which the internal stresses balance out the external pressure  $P_0$  but otherwise swell freely) are uniform expansions, that is, homogeneous solutions with a constant deformation gradient of the form

$$F = \lambda_{\text{free}} R, \quad R \in SO(3) \quad (19)$$

such that  $P = 0$  (see [5], chapter 3;[3]). From (15) we obtain

$$\omega(J) F^c = -\gamma F. \quad (20)$$

Taking  $F$  as in (19) gives the equation

$$\lambda_{\text{free}} \omega\left((\lambda_{\text{free}})^3\right) = -\gamma, \quad (21)$$

or

$$\phi^{*-1/3} \omega(\phi^{*-1}) = -\gamma, \quad \text{with } \phi^* := (\lambda_{\text{free}})^{-3}. \quad (22)$$

The equilibrium swelling rate,  $\phi^*$ , depends on the Flory parameter and has a prominent role in the study of gels ([5], chapter 3).

In this paper we shall consider thin gels with a cross-section in the  $x_1$  and  $x_2$  coordinates that is allowed to swell, but that at the same time are confined between two parallel plates, of equation  $x_3 = \text{constant}$ , with respect to which the gel is allowed to slide freely. (The gels are thin in the  $x_2$  direction, which we will refer to as the vertical direction.) It can be seen that in the absence of a bonding rigid substrate the global minimizer is a homogeneous deformation with

$$F = \text{diag}(\lambda^*, \lambda^*, 1), \quad (23)$$

the optimal stretch factor  $\lambda^*$  being determined, by (17) for  $\mathbf{n}_0 = \mathbf{e}_1$  and  $\mathbf{n}_0 = \mathbf{e}_2$ , as the unique solution to

$$\omega\left((\lambda^*)^2\right) = -\gamma. \quad (24)$$

Another type of uniform solutions that will play a role are those with a uniaxial extension along the free direction

$$F = \text{diag}(1, \lambda_*, 1), \quad (25)$$

with  $\lambda_*$  given by the pressure boundary condition (17) along the free direction  $\mathbf{n}_0 = \mathbf{e}_2$ :

$$\omega(\lambda_*) = -\gamma \lambda_*. \quad (26)$$

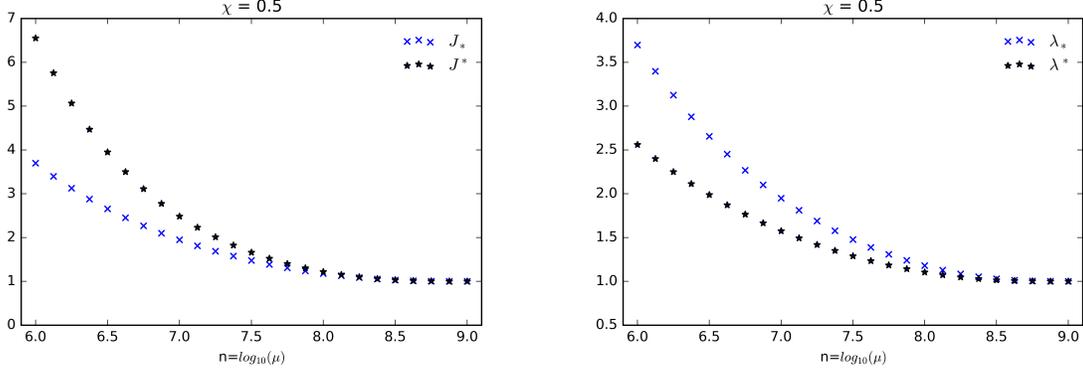


Figure 2: The left and right plots, respectively, show that  $J_* < J^*$  and  $\lambda_* > \lambda^*$  in the range  $10^6 < \mu < 10^9$ .

#### B.4 Properties of the uniform uniaxial extension and isotropic expansion ratios

We now study the properties of the quantities  $\lambda_*$  and  $\lambda^*$ .

As a consequence of Taylor's theorem, for all  $J > 1$

$$\begin{aligned}
 -\omega(J) + \zeta &= -\left(1 - \frac{N_2}{N_1}\right)J^{-1} - \ln(1 - J^{-1}) + \frac{\chi}{2}J^{-2} \\
 &> -\left(1 - \frac{N_2}{N_1}\right)J^{-1} - J^{-1} - \frac{1}{2}J^{-2} + \frac{\chi}{2}J^{-2} \\
 &= \frac{N_2}{N_1}J^{-1} + \frac{1 - \chi}{2}J^{-2} > \frac{1 - \chi}{2}J^{-2}.
 \end{aligned}$$

Substituting  $(\lambda^*)^2$  and  $\lambda_*$  for  $J$  it follows that

$$\frac{1 - \chi}{2}(\lambda^*)^{-4} < \gamma + \zeta \quad \text{and} \quad \frac{1 - \chi}{2}(\lambda_*)^{-2} < \gamma\lambda_* + \zeta < (\gamma + \zeta)\lambda_*,$$

that is,

$$\lambda^* > \left(\frac{1 - \chi}{2(\gamma + \zeta)}\right)^{-1/4} \quad \text{and} \quad \lambda_* > \left(\frac{1 - \chi}{2(\gamma + \zeta)}\right)^{-1/3}.$$

This shows that  $\lambda^*$  grows like  $\gamma^{-1/4}$  as  $\gamma$  grows smaller, at least under the assumption  $\zeta \leq \gamma$ . That is to be expected since  $\gamma$  is small for very soft materials, which are able to swell notoriously more than the more rigid ones. As for  $\lambda_*$ , the growth is as  $\gamma^{-1/3}$  instead. These calculations suggest that

$$J_* := \lambda_* < (\lambda^*)^2 =: J^* \quad \text{whereas} \quad \lambda_* > \lambda^*$$

(the gel absorbs more fluid when detached, yet in the bonded region it stretches vertically more than in the detached part). Those relations are verified for a large range of parameters, as shown in Figure 2 and in Table 1.

When  $\gamma$  grows large the gel grows stiffer, preventing it to swell significantly. This is confirmed by the observation that for all  $J > 1$

$$\omega(J) - \zeta > \ln(1 - J^{-1})$$

and that  $\omega(\lambda_*) = -\gamma\lambda_* < -\gamma$ , so that both

$$J_* - 1 < \frac{e^{-(\gamma + \zeta)}}{1 - e^{-(\gamma + \zeta)}} \quad \text{and} \quad J^* - 1 < \frac{e^{-(\gamma + \zeta)}}{1 - e^{-(\gamma + \zeta)}}.$$

Hence, the increase in volume due to fluid absorption decays exponentially as  $\gamma$  increases (see Figure 3).

Table 1: Optimal stretches and Jacobians – up to an error of  $10^{-5}$

		$\mu = 10^4$	$\mu = 10^5$	$\mu = 10^6$	$\mu = 10^7$	$\mu = 10^8$	$\mu = 10^9$
$\chi = 0.3$	$\lambda^*$	7.29088	4.69619	2.73109	1.6409	1.11611	1.00011
	$\lambda_* = J_*$	17.15694	8.19052	3.97878	2.04031	1.19642	1.00023
	$J^* = (\lambda^*)^2$	53.15704	22.05423	7.45888	2.69258	1.24571	1.00023
$\chi = 0.5$	$\lambda^*$	6.76277	4.35705	2.55945	1.57566	1.10477	1.00009
	$\lambda_* = J_*$	15.51538	7.47787	3.69736	1.9489	1.17976	1.00020
	$J^* = (\lambda^*)^2$	45.73512	18.98392	6.55082	2.4827	1.22053	1.00020
$\chi = 0.9$	$\lambda^*$	4.97289	3.29415	2.10909	1.43793	1.0844	1.00008
	$\lambda_* = J_*$	10.60023	5.5376	3.02236	1.75776	1.14893	1.00017
	$J^* = (\lambda^*)^2$	24.72963	10.85146	4.44827	2.06765	1.17594	1.00017

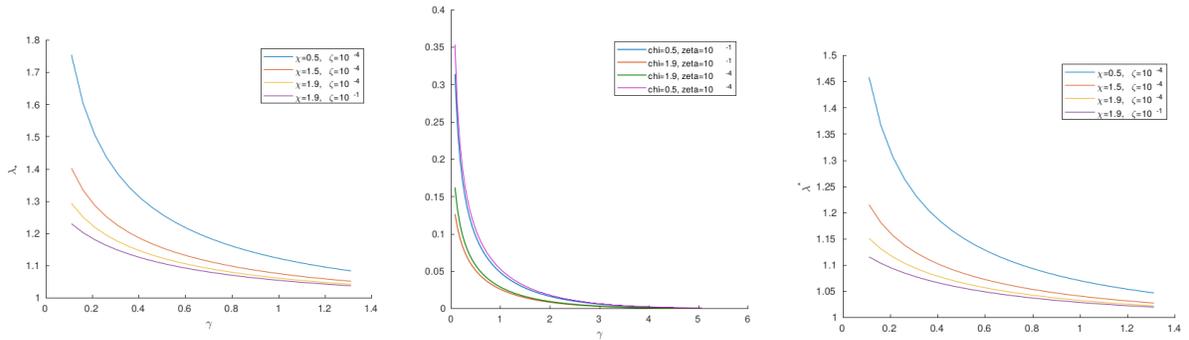


Figure 3: The left and the right plots represent  $\lambda_*$  and  $\lambda^*$ , respectively, depicting their monotonicity and exponential decay to 1. The middle plot represents their difference, which approaches 0 as  $\gamma$  grows.

## C Boundary value problem

Given positive numbers  $d, L, L_z$ , we consider the domain

$$\begin{aligned}\Omega_{\mathcal{B}} &= \Omega \times (-L_z, L_z), \text{ with} \\ \Omega &= \{(x_1, x_2) : -L < x_1 < L, \ 0 < x_2 < d\},\end{aligned}\tag{27}$$

and the map  $\varphi_{\mathcal{B}}(\mathbf{x}) = \varphi(x_1, x_2) + x_3 \mathbf{e}_3$ , where

$$\varphi = \sum_{i=1}^2 \varphi_i(x_1, x_2) \mathbf{e}_i,$$

represents a two-dimensional deformation map. In this geometry, the total energy becomes

$$\hat{E} = \int_{\Omega} \left\{ \frac{\mu}{2} \sum_{i,j=1}^2 \varphi_{ix_j}^2 + \nu H(J) + P_0 J \right\} d\mathbf{x} + \frac{\mu}{2}.\tag{28}$$

Omitting the last constant term in (28), the problem becomes that of a two dimensional deformation of a plane domain. The Euler-Lagrange equations of the system are

$$\frac{\partial}{\partial x_1} \left( \mu \varphi_{1x_1} + \nu \omega(J) \varphi_{2x_2} \right) + \frac{\partial}{\partial x_2} \left( \mu \varphi_{1x_2} - \nu \omega(J) \varphi_{2x_1} \right) = 0\tag{29}$$

$$\frac{\partial}{\partial x_1} \left( \mu \varphi_{2x_1} - \nu \omega(J) \varphi_{1x_2} \right) + \frac{\partial}{\partial x_2} \left( \mu \varphi_{2x_2} + \nu \omega(J) \varphi_{1x_1} \right) = 0.\tag{30}$$

Our goal is to analyze the equations (29)-(30) for the domain (27) in the case when  $0 < d \ll L$ , i.e., when the aspect ratio  $\eta := \frac{d}{L}$  is very small. We focus our attention to the case when the domain is partially delaminated from the substrate. More precisely, we shall assume that for some fixed  $0 < \delta < 1$ , the gel is attached to the substrate on the part of the boundary

$$-\delta L < x_1 < \delta L, \quad x_2 = 0, \quad -L_z < x_3 < L_z.$$

Moreover, we seek solutions that are symmetric with respect to the  $X_2$ - $X_3$  plane, so in the sequel the variational problem will only be studied in the two-dimensional domain  $[0, L] \times [0, d]$ .

We first scale the variables and fields of the problem according to the dimensions of the domain:

$$x = \frac{x_1}{L}, \quad y = \frac{x_2}{d}, \quad u = \frac{\varphi_1}{L}, \quad v = \frac{\varphi_2}{d}.\tag{31}$$

The previous expressions represent a geometric scaling of the domain and render the deformation fields dimensionless. We will find that they yield an approximation ansatz of the deformation map linear in the *thin direction*  $y$ -variable, as commonly assumed in thin film approaches. (The above is a special choice among exponents  $\alpha, \beta$  such that  $u = \eta^\alpha \frac{\varphi_1}{L}$ ,  $v = \eta^\beta \frac{\varphi_2}{L}$  with  $\alpha + \beta = -1$ , as required so that  $\frac{\partial(u,v)}{\partial(x,y)}$  coincides with  $J = \det \nabla \varphi$  and remains of order 1 in the limit as  $\eta \rightarrow 0$ ; see, e.g., Equation (36) below.)

The total energy written in terms of the dimensionless variables is

$$\frac{\hat{E}/|\Omega_{\mathcal{B}}|}{\nu} = \int_0^1 \int_0^1 \left( \frac{\gamma}{2} (1 + u_x^2 + \eta^2 v_x^2 + \eta^{-2} u_y^2 + v_y^2) + H(J) + \zeta J \right) dx dy.\tag{32}$$

The dimensionless form of the Euler-Lagrange equations of  $\mathcal{E}$  is

$$\frac{\partial}{\partial x} (\gamma u_x + \omega(J) v_y) + \frac{\partial}{\partial y} \left( \frac{\gamma}{\eta^2} u_y - \omega(J) v_x \right) = 0,\tag{33}$$

$$\frac{\partial}{\partial x} (\eta^2 \gamma v_x - \omega(J) u_y) + \frac{\partial}{\partial y} (\gamma v_y + \omega(J) u_x) = 0.\tag{34}$$

Let us denote

$$F = \begin{bmatrix} u_x & \frac{1}{\eta}u_y & 0 \\ \eta v_x & v_y & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad F^c = \begin{bmatrix} v_y & -\eta v_x & 0 \\ -\frac{1}{\eta}u_y & u_x & 0 \\ 0 & 0 & u_x v_y - u_y v_x \end{bmatrix}. \quad (35)$$

Note that

$$J = u_x v_y - u_y v_x \quad (36)$$

denotes the Jacobian determinant (5) with respect to the scaled variables (31). Hence,

$$P = \mu \begin{bmatrix} u_x & \frac{1}{\eta}u_y & 0 \\ \eta v_x & v_y & 0 \\ 0 & 0 & 1 \end{bmatrix} + \nu \omega(J) \begin{bmatrix} v_y & -\eta v_x & 0 \\ -\frac{1}{\eta}u_y & u_x & 0 \\ 0 & 0 & J \end{bmatrix}. \quad (37)$$

For a given  $0 < \delta < 1$ , we look for solutions satisfying mixed displacement and traction boundary conditions. We consider the case when apart from being perfectly bonded to a rigid substrate on the left part of its bottom surface:

$$u(x, 0) = x, \quad v(x, 0) = 0, \quad 0 \leq x \leq \delta, \quad (38)$$

and being subject to the external pressure described by Equation (17):

$$\omega(J^-)(F^-)^c \mathbf{n}_0 = -\gamma(F^-) \mathbf{n}_0, \quad \mathbf{n}_0 = \mathbf{e}_2, \quad 0 < x < \delta, \quad y = 1, \quad (39)$$

$$\omega(J^+)(F^+)^c \mathbf{n}_0 = -\gamma(F^+) \mathbf{n}_0, \quad \mathbf{n}_0 = \mathbf{e}_2, \quad \delta < x < 1, \quad y = 1, \quad (40)$$

$$\omega(J^+)(F^+)^c \mathbf{n}_0 = -\gamma(F^+) \mathbf{n}_0, \quad \mathbf{n}_0 = -\mathbf{e}_2, \quad \delta < x < 1, \quad y = 0, \quad (41)$$

$$\omega(J^+)(F^+)^c \mathbf{n}_0 = -\gamma(F^+) \mathbf{n}_0, \quad \mathbf{n}_0 = \mathbf{e}_1, \quad x = 1, \quad 0 < y < 1, \quad (42)$$

the gel is also free to slide along the side  $x = 0$ :

$$u(0, y) = 0, \quad y \in [0, 1], \quad (43)$$

$$P^- \mathbf{n}_0 \cdot \mathbf{n}_0^\perp = 0, \quad \mathbf{n}_0 = -\mathbf{e}_1, \quad \mathbf{n}_0^\perp = \mathbf{e}_2, \quad x = 0, \quad 0 < y < 1. \quad (44)$$

(We employ superindex notation  $-$  and  $+$  to represent  $u$ ,  $v$ ,  $F$ ,  $J$ , and  $P$  for  $x < \delta$  and  $x > \delta$ , respectively.) Note that equations (39) and (40) become

$$\eta^2 \omega(J) v_x(x, 1) = \gamma u_y(x, 1), \quad (45)$$

$$\omega(J) u_x(x, 1) = -\gamma v_y(x, 1), \quad x \in (0, 1). \quad (46)$$

Equation (41) yields also the previous equations but for  $x \in (\delta, 1), y = 0$ . Likewise, for (42) we have,

$$\omega(J) v_y(1, y) = -\gamma u_x(1, y), \quad (47)$$

$$\omega(J) u_y(1, y) = \gamma \eta^2 v_x(1, y), \quad y \in (0, 1). \quad (48)$$

Equation (44) becomes

$$\gamma \eta v_x(0, y) - \omega(J) \eta^{-1} u_y(0, y) = 0, \quad y \in (0, 1). \quad (49)$$

Taking now the limit as  $\eta \rightarrow 0$  on each of them, we get

$$u_y(x, 1) = 0, \quad (50)$$

$$\omega(J) u_x(x, 1) = -\gamma v_y(x, 1), \quad x \in (0, 1). \quad (51)$$

Next, we write the components of equation (41) giving also

$$u_y(x, 0) = 0, \quad (52)$$

$$\omega(J)u_x(x, 0) = -\gamma v_y(x, 0), \quad x \in (\delta, 1). \quad (53)$$

As for (47)-(48), we find that

$$\omega(J)v_y(1, y) = -\gamma u_x(1, y), \quad (54)$$

$$\omega(J)u_y(1, y) = 0, \quad y \in (0, 1). \quad (55)$$

The pressure boundary conditions are summarized in equations (50)-(55). Taking limit as  $\eta \rightarrow 0$  as well in equation (49), we get

$$\omega(J)u_y(0, y) = 0, \quad y \in (0, 1). \quad (56)$$

The governing equations (33)-(34) for  $(u, v)$ , in the  $\eta \rightarrow 0$  limit, become

$$u_{yy} = 0, \quad (57)$$

$$\gamma v_{yy} + H''(J)(J_y u_x - J_x u_y) = 0. \quad (58)$$

Integrating the first of the previous equations gives  $u_y(x, y) = u_y(x, 1)$ , and taking (50) into account, yields

$$u(x, y) = \alpha(x), \quad x \in (0, 1), y \in (0, 1), \quad (59)$$

with  $\alpha(x)$  to be determined (and possibly having different expressions for  $x < \delta$  and  $x > \delta$ ). Consequently, the Jacobian determinant is given by  $J = \alpha'(x)v_y(x, y)$  and (58) becomes

$$v_{yy}(\gamma + H''(J)(\alpha'(x))^2) = 0. \quad (60)$$

The positivity of  $H''$  that follows from (14) yields

$$v_{yy} = 0. \quad (61)$$

Integration of the previous equation gives

$$v(x, y) = \beta(x)y + \beta_0(x), \quad x, y \in (0, 1). \quad (62)$$

As a consequence of the first boundary condition in (38),

$$u(x, y) = x, \quad x \in (0, \delta), y \in (0, 1). \quad (63)$$

Note that it trivially satisfies boundary conditions (43) and (56). We require  $v$  to satisfy the second boundary condition in (38) giving

$$\beta_0^-(x) = 0, \quad 0 < x < \delta. \quad (64)$$

We now write  $\beta_0^+(x) = \beta_0(x)$ .

Summarizing, the components of the deformation map are

$$u(x, y) = \alpha(x), \quad v(x, y) = \beta(x)y + \beta_0(x), \quad x \in (0, 1), y \in (0, 1), \quad (65)$$

with

$$\alpha(x) = x, \quad \beta_0(x) = 0, \quad 0 < x < \delta. \quad (66)$$

The deformation gradients (35) and their cofactors are then given by

$$F^- = \begin{bmatrix} 1 & 0 & 0 \\ \eta\beta'_-(x)y & \beta_-(x) & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad (67)$$

$$F^+ = \begin{bmatrix} \alpha'_+(x) & 0 & 0 \\ \eta(\beta'_+(x)y + \beta'_0(x)) & \beta_+(x) & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad (68)$$

$$(F^-)^c = \begin{bmatrix} \beta_-(x) & -\eta\beta'_-(x)y & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \beta_-(x) \end{bmatrix}, \quad (69)$$

$$(F^+)^c = \begin{bmatrix} \beta_+(x) & -\eta(\beta'_+(x)y + \beta'_0(x)) & 0 \\ 0 & \alpha'_+(x) & 0 \\ 0 & 0 & \alpha'_+(x)\beta_+(x) \end{bmatrix}. \quad (70)$$

The Jacobian determinant has the form

$$J^-(x, y) = \beta_-(x), \quad 0 < x < \delta, \quad (71)$$

$$J^+(x, y) = \alpha'(x)\beta_+(x), \quad \delta < x < 1. \quad (72)$$

Equations (50)-(55) for the piecewise continuous deformation gradient become

$$\omega(\beta_-(x)) = -\gamma\beta_-(x), \quad x \in (0, \delta), \quad (73)$$

$$\omega(\alpha'(x)\beta_+(x))\alpha'(x) = -\gamma\beta_+(x), \quad x \in (\delta, 1), \quad (74)$$

$$\omega(\alpha'(1)\beta_+(1))\beta_+(1) = -\gamma\alpha'(1). \quad (75)$$

From (73) we deduce that  $\beta_-(x)$  is constant and equal to the value  $\lambda_*$  defined in (26). Concerning the debonded part  $x > \delta$ , observe that when  $\eta \rightarrow 0$  the energy functional (32) becomes

$$\begin{aligned} \frac{\hat{E}/|\Omega_B|}{\nu} &= \delta \cdot \left( \frac{\gamma}{2}(2 + \lambda_*^2) + H(\lambda_*) + \zeta\lambda_* \right) \\ &\quad + \int_{\delta}^1 \left( \frac{\gamma}{2}(\alpha'(x)^2 + \beta_+(x)^2) + H(J^+(x)) + \zeta J^+(x) \right) dx. \end{aligned} \quad (76)$$

It is easy to see that the integrand  $(s, t) \mapsto \frac{\mu}{2}(s^2 + t^2) + \nu H(st) + P_0 st$  is minimized when both  $s$  and  $t$  coincide with the value  $\lambda^*$  defined in (24). Hence, the minimum total energy is attained when both  $\alpha'(x)$  and  $\beta_+(x)$  are constant and equal to  $\lambda^*$ . Note that, when this is the case, the traction boundary conditions (74) and (75) are automatically satisfied.

In conclusion, the solution is of the form

$$\begin{aligned} 0 < x < \delta : \\ u &= x, \quad v = \lambda_* y, \end{aligned} \quad (77)$$

$$\begin{aligned} \delta < x < 1 : \\ u &= \lambda^* x + \delta(1 - \lambda^*), \\ v &= \lambda^* y + \beta(\delta^+). \end{aligned} \quad (78)$$

The constants in (78) are chosen so as to guarantee the continuity of the horizontal component  $u$  of the displacement at  $x = \delta$ . The vertical displacement  $v$ , in contrast, is discontinuous at that interface; this may indicate that the gel is undergoing a volume phase transition. The corresponding deformation gradient matrices are

$$F^- = \begin{bmatrix} 1 & 0 & 0 \\ 0 & \lambda_* & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad F^+ = \begin{bmatrix} \lambda^* & 0 & 0 \\ 0 & \lambda^* & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad (79)$$

for  $0 < x < \delta$  and  $\delta < x < 1$ , respectively. The associated Jacobian determinants have values

$$J^- = \lambda_*, \quad J^+ = \lambda^{*2}. \quad (80)$$

We now calculate the components of the stress evaluated at the uniform uniaxial extension (77) and at the uniform expansion (78) states:

$$\begin{aligned} P_{11}^- &= \mu + \nu\omega(\lambda_*)\lambda_* = \mu(1 - \lambda_*^2), \\ P_{22}^- &= \mu\lambda_* + \nu\omega(\lambda_*) = 0, \\ P_{21}^- &= 0, \quad P_{12}^- = 0, \\ P^+ &= 0. \end{aligned} \quad (81)$$

Note that  $P^- \mathbf{e}_1$  does not vanish since the bonding to the substrate prevents that part of the gel from stretching in the  $x_1$  direction, as it would prefer. In particular, the horizontal component of the force is discontinuous across  $x = \delta$ .

Let us calculate the components of the force needed to find equilibrium conditions at the interface:

$$\mathbf{e}_1 \cdot P \mathbf{e}_1 = \mu u_x + \nu\omega(J)v_y, \quad \mathbf{e}_2 \cdot P \mathbf{e}_1 = \mu\eta v_x - \frac{\nu}{\eta}\omega(J)u_y. \quad (82)$$

We conclude this section by replacing the expressions (78) in the formula (76) for the dimensionless energy:

$$\begin{aligned} \tilde{\mathcal{E}}(\delta) := \frac{\hat{E}}{4LL_z d\nu} &= \delta \cdot \left( \frac{\gamma}{2}(2 + \lambda_*^2) + H(\lambda_*) + \zeta\lambda_* \right) \\ &+ \left( \gamma\lambda^{*2} + H(\lambda^{*2}) + \zeta\lambda^{*2} \right)(1 - \delta). \end{aligned} \quad (83)$$

## D A debonding criterion

We have obtained the energy of a partially debonded gel, confined between two parallel walls but free to slide along them, in the zero thickness limit,  $\eta \rightarrow 0$ . Since the total energy of the gel  $\hat{E}$  is related to the dimensionless energy  $\tilde{\mathcal{E}}(\delta)$  according to

$$\hat{E} = 4LL_z d\nu \tilde{\mathcal{E}}(\delta),$$

and since the debonded region has an area of  $(2L_z)(2L - 2\delta L)$ , in this model the energy release rate (energy released per unit increase in debonded area) is given by

$$\left| \frac{-\frac{d}{d\delta}(4LL_z d\nu \tilde{\mathcal{E}}(\delta))}{-\frac{d}{d\delta}(4LL_z - 4\delta LL_z)} \right| = d\nu R, \quad \text{with} \quad R := \frac{d}{d\delta} \tilde{\mathcal{E}}(\delta)$$

(there are minus signs on both the numerator and the denominator because in order for the debonding to progress the parameter  $\delta$  must decrease). In particular, the energy release rate is proportional to the thickness of the gel.

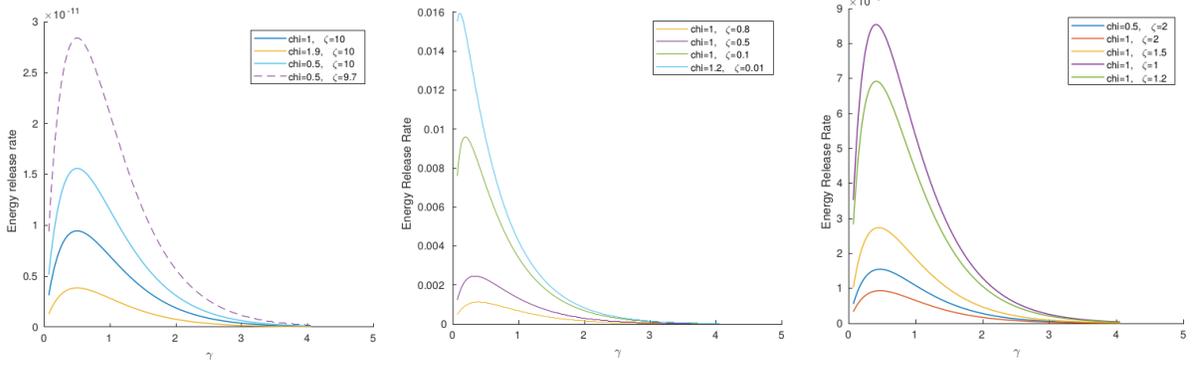


Figure 4: The above plots show the energy release rate  $R$  as function of  $\gamma$ . It is found to increase as the external pressure,  $\zeta$ , decreases. The graphs also show that  $R$  decreases with increasing  $\chi$ . They also establish that  $R \rightarrow 0$  as  $\gamma$  grows large.

The expression (83) that we obtained is linear with respect to  $\delta$ . Consequently, the quantity  $R$  above, which corresponds to the energy release rate per unit thickness, measured using  $\nu$  as the energy unit, is independent of the debonded length:

$$R := \left( \frac{\gamma}{2}(1 + \lambda_*^2) + H(\lambda_*) + \zeta\lambda_* \right) - (\gamma\lambda^{*2} + H(\lambda^{*2}) + \zeta\lambda^{*2}). \quad (84)$$

According to Griffith's theory of crack propagation [8][7][2], the debonding criterion is obtained by comparing the energy released rate  $d\nu R$  against the adhesive toughness  $\sigma$  (the energy per unit area that the adhesive applies in order to keep the membrane bonded). That implicitly defines, in terms of the parameters  $\chi, \gamma, \zeta$  and  $\sigma$  of the problem, the threshold thickness  $d_{\max} > 0$  for which the membrane remains bonded:

$$d_{\max} := \frac{\sigma}{\nu R(\chi, \gamma, \zeta)}. \quad (85)$$

## E Parameter values and numerical simulations

Recall that the basic parameter groups of the model are

- $K_B = 1.3806 * 10^{-23} \frac{\text{m}^2 K_g}{\text{s}^2 K}$ ,  $V_m = 3 * 10^{-29} \text{m}^3$ ,  $T = 300K$ ,
- $\nu := \frac{K_B T}{V_m N_2} = 1.38 * 10^8$ , ( $N_2 = 1$ ), with dimensions of energy per unit volume,
- $P_0 = 10^4$  Pa (arterial pressure),  $\zeta = \frac{P_0}{\nu} = 0.72 * 10^{-4}$ ,
- $\mu_E = 10^4$  to  $10^9$  Pa,
- $\gamma := \frac{\mu_E}{\nu}$ ; it takes values in the range  $10^{-4}$  to 10,
- $\sigma = 1 * 10^3$  to  $4 * 10^3$ ,  $\frac{\text{joules}}{\text{m}^2}$ , so  $\frac{\sigma}{\nu} = 0.72 * 10^{-5}$  to  $4 * 10^{-5} m$ .

Figures 4–6 illustrate the behaviour of the energy release rate (per unit thickness)  $R$  and of the threshold thickness  $d_{\max}$  with respect to changes in the parameters  $\gamma$  and  $\chi$ . Figures 7–8 shows how the liberty to stretch horizontally by the optimal factor  $\lambda^*$  is taken advantage of by the debonded part of the gel. Note that the vertical stretch  $\lambda_*$  in the bonded region (where the horizontal distances are kept fixed) is larger than the stretch  $\lambda^*$  in the free region. The width of the transition layer is of the same order as the gel thickness.

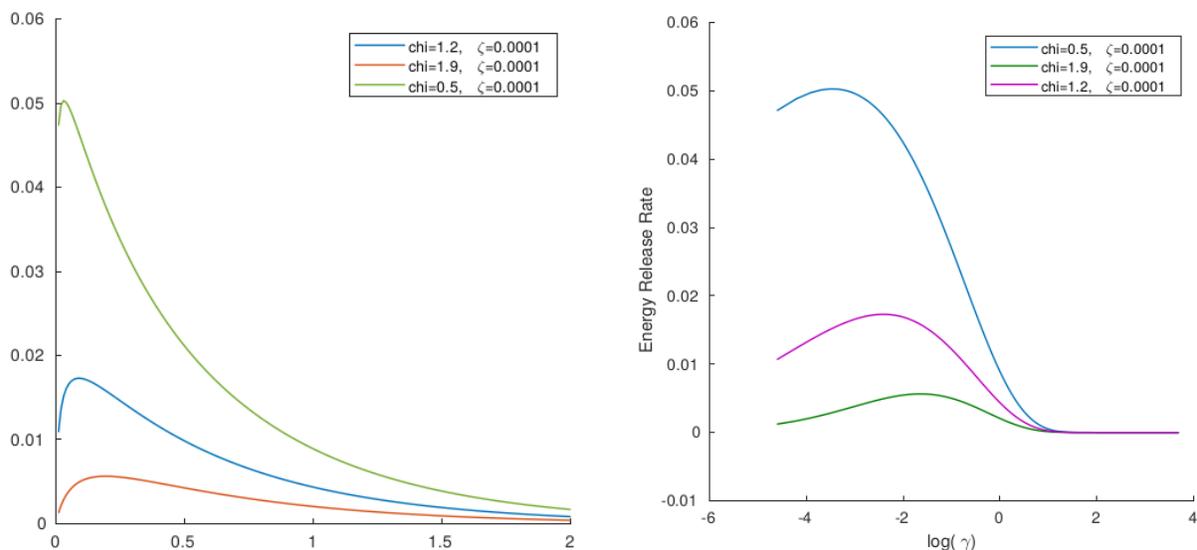


Figure 5: The above plots show the energy release rate  $R$  as function of  $\gamma$ . The left plot corresponds to pressure values found in device applications. The right graph shows the energy release rate as a function of the logarithm of  $\gamma$  depicting its approach to 0 as  $\gamma$  grows large.

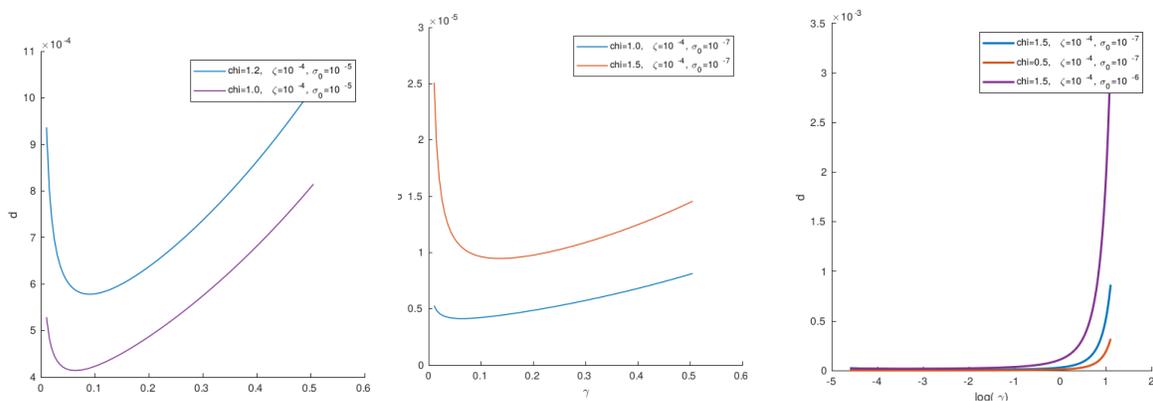


Figure 6: These graphs represent the threshold thickness  $d$  of the sample as a function of  $\gamma$ . The greater adhesive toughness allows for larger membrane thickness. It is also observed that  $d$  is first decreasing and then increasing with  $\gamma$ . The logarithmic plot on the right brings out a special behavior of  $d$  with respect to  $\gamma$  for large values of  $\gamma$ . It is also observed that larger values of  $\chi$ , still within the mixing regime, allow for greater thickness.

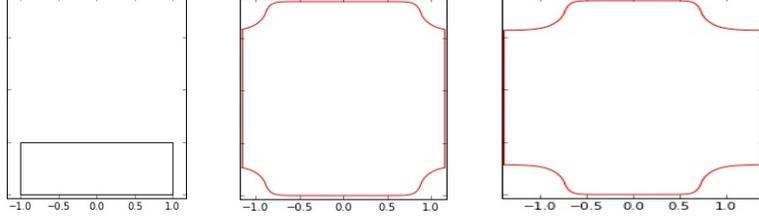


Figure 7: Change in the final shape of the gel as the debonding progresses ( $L = 1$ ,  $d = 0.05$ ,  $\mu = 10^6$ ,  $\chi = 0.5$ ). On the left, the cross-section in the  $X_1$ - $X_2$  plane of the dry polymer (the reference configuration  $\Omega$ ). In the middle and on the right, cross-section of the deformed configuration for  $\delta = 0.9$  and  $\delta = 0.75$ , respectively. The axes have the same scaling in the three plots, for ease of comparison.

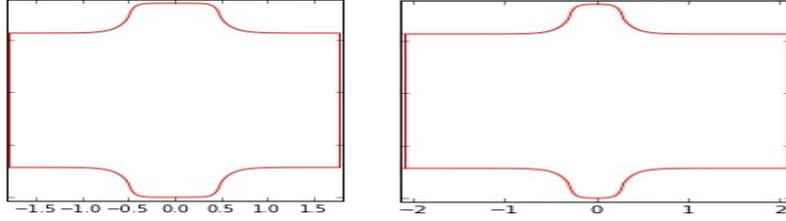


Figure 8: Change in the final shape of the gel as the debonding progresses ( $L = 1$ ,  $d = 0.05$ ,  $\mu = 10^6$ ,  $\chi = 0.5$ ). On the left, the cross-section the cross-section in the  $X_1$ - $X_2$  of the deformed configuration for  $\delta = 0.5$ . On the right, cross-section of the deformed configuration for  $\delta = 0.3$ . The axes have the same scaling in the three plots, for ease of comparison.

## F Conclusions

This article analyzes debonding of a thin, two dimensional gel layer from a rigid substrate, by calculating the energy released by the system upon debonding. One main aspect of the analysis is the construction of an approximate solution, valid at the limit of small aspect ratio. We have found that the vertical component of the approximating deformation map is discontinuous at the interface between the bound and the debonded regions. In future work, we will address the construction of the internal layer corrections that guarantee the continuity of the deformation map at the interface. For this, we will consider the formal expansion of the solution as

$$u(x, y, \eta) = \hat{u}(x, y, \eta) + \eta U(\tilde{x}, y, \eta), \quad (86)$$

$$v(x, y, \eta) = \hat{v}(x, y, \eta) + V(\tilde{x}, y, \eta), \quad (87)$$

$$\tilde{x} = \frac{x - \delta}{\eta}. \quad (88)$$

The terms  $(\hat{u}, \hat{v})$  correspond to the approximation of the solution away from the transition layer. They admit a formal asymptotic expansion in terms of the small parameter  $\eta$ , whose leading terms have been calculated in the previous sections. The scaling factor  $\eta$  multiplying  $U$  is consistent with the continuity of the horizontal component of the deformation map of the previously constructed approximating solution. Our preliminary study suggests existence of solutions of the form

$$U(\tilde{x}, \cdot) = O(e^{-\lambda|\tilde{x}|}), \quad V(\tilde{x}, \cdot) = O(e^{-\lambda|\tilde{x}|}), \quad \lambda > 0. \quad (89)$$

The latter suggest that the contribution of the internal layer terms  $(U, V)$  to the energy is of higher order in  $\eta$ , and it does not alter the debonding analysis. The study of the convergence of the asymptotic expansions (86, 87) is the subject of a forthcoming article.

## Acknowledgements

We are grateful to M. Barchiesi, M. Doi, D. Golovaty, O. Lavrentovich, R. Siegel, S. Walker, N. Walkington & W. Zhang for their insight and valuable comments. M. C. Calderer acknowledges the support of the National Science Foundation, grant N° DMS-1616866. D. Henao is indebted to the Institute for Mathematics and its Applications of the University of Minnesota and its program ‘Multiscale Mathematics and Computing in Science and Engineering’, where he was received as a long-term visitor, for greatly facilitating this research. He also acknowledges the School of Mathematics of the University of Minnesota and the City Library of San Fernando, VI Región, Chile for their warm hospitality, as well as the Chilean Ministries of Economy and of Education for partially funding his research through Millennium Nucleus NC130017 CAPDE and FONDECYT projects 1150038 and 1190018.

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