

Supplemental information for :
"Spontaneous localized fluid release on swelling fibers"

Pierre Van de Velde, Julien Dervaux, Camille Duprat, Suzie Protière

May 21, 2022

1 Correlation between drop height and fiber tension

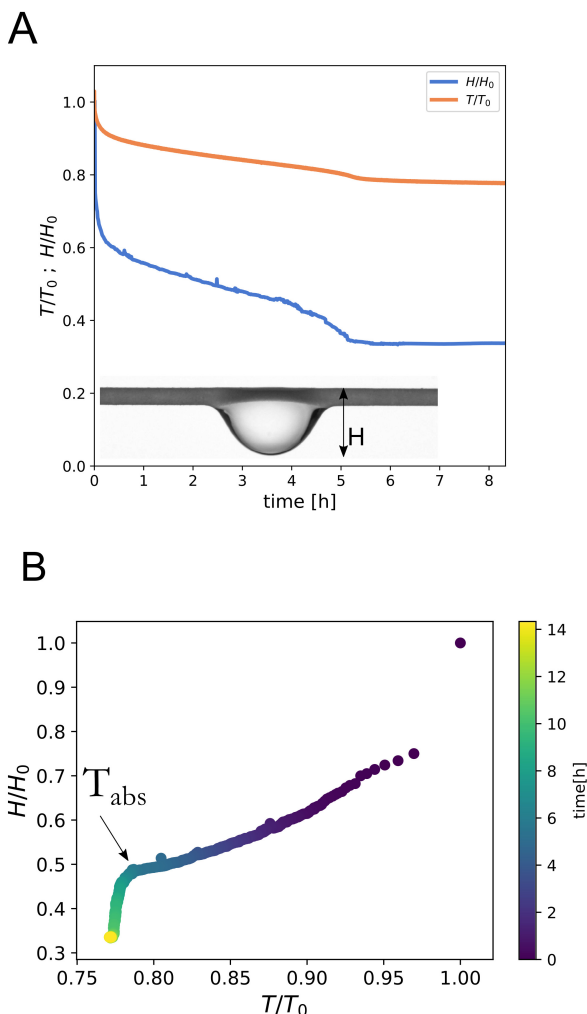


Figure 1 Correlation between drop height and fiber tension **A** Normalized fiber tension and drop height during the absorption of a single drop of $2 \mu\text{L}$ on a fiber of initial radius $R_0 = 250 \mu\text{m}$. Both the tension and the drop height (measured from apex to apex, including the fiber radius) decrease following very similar dynamics. **B** Direct correlation of the two values. Once the drop is fully absorbed (at $t = T_{abs}$), the fiber slowly relaxes, which decreases H , while the tension remains almost constant.

2 Model development

We model our experiment in the framework of linear poroelasticity. We consider a poroelastic gel-like material which is not subjected to any mechanical load in its reference state. The initial concen-

tration of solvent molecules in the poroelastic material is homogeneous and given by c_0 while the chemical potential is μ_0 . In the deformed state, the system is described by the solvent concentration c , chemical potential μ and displacement field \vec{u} . In response to the application of an external force, or when the poroelastic material is brought into contact with a reservoir of solvent molecules at a chemical potential different from μ_0 , the solvent is not in diffusive equilibrium and will penetrate the material causing it to swell. The conservation of the number of solvent molecules writes:

$$\frac{\partial c}{\partial t} + \vec{\nabla} \cdot \vec{J} = 0 \quad (1)$$

where \vec{J} is the flux of the solvent in the gel and is driven by spatial differences of the chemical potential. For simplicity, we will assume that the flux \vec{J} of small molecules is given by Darcy's law:

$$\vec{J} = - \left(\frac{k}{\eta \Omega^2} \right) \vec{\nabla} \mu \quad (2)$$

where k is the permeability, η is the viscosity of the solvent and Ω is the molar volume of the solvent.

The strain tension ε is defined as:

$$\varepsilon = \frac{1}{2} \left(\vec{\nabla} \vec{u} + (\vec{\nabla} \vec{u})^t \right) \quad (3)$$

In the framework of linear poroelasticity, the stress tensor σ is given by:

$$\sigma = 2G \left(\varepsilon + \frac{\nu}{1-2\nu} \text{Tr}(\varepsilon) \mathbf{I} \right) - \frac{\mu - \mu_0}{\Omega} \mathbf{I} \quad (4)$$

where G is the shear modulus, ν is the poroelastic Poisson ratio that characterizes the ability of a gel to absorb its solvent and \mathbf{I} is the identity tensor. We assume that solvent and polymer molecules are incompressible and consequently the local volume variation is given by the local variation of the solvent concentration. This molecular incompressibility condition reads:

$$\text{Tr}(\varepsilon) = (c - c_0) \Omega \quad (5)$$

The mechanical equilibrium in the bulk of the poroelastic material is described by the Navier equations:

$$\vec{\nabla} \cdot \sigma = \vec{0} \quad (6)$$

Combining the equations above we get:

$$\frac{\partial c}{\partial t} = D^* \Delta c \quad (7)$$

where

$$D^* = \frac{2(1-\nu)Gk}{(1-2\nu)\eta} \quad (8)$$

is an effective diffusion coefficient (also called the cooperative diffusion coefficient) and Δ is the Laplace operator. Note that the material parameters G , k and thus D^* are effective parameters that depend on the initial state of the gel. In a nonlinear theory, they

are also functions on the local deformation of the gel. Within the framework of linear poroelasticity however, we will assume that the deformed state is close enough to the initial state such that G , k and thus D^* can be treated as constant material parameters. Finally, combining (3, 4, 5) and (6) and under the assumption that c_0 is homogeneous, we get:

$$G\Omega \left(\Delta \bar{u} + \frac{\Omega}{1-2\nu} \bar{c} \right) = \vec{\nabla} \mu \quad (9)$$

We opt for a traditional dimensionality reduction and we will limit ourselves to very thin fibers and focus on timescales larger than R^2/D^* . The goal of this approximation is to focus on a regime where the concentration, and hence also the chemical potential, are approximately constant throughout the radius of the fiber such that the problem will become effectively 2D (z and time t). Hence we hypothesize that:

$$c(r, z, t) = \bar{c}(z, t) \quad (10)$$

$$\mu(r, z, t) = \mu(\bar{c}, t) \quad (11)$$

For consistency with our approximation, we assume that physical quantities vary over a typical lengthscale L in the axial direction z while they vary over a typical lengthscale R in the radial direction r with of course $L \gg R$; at leading order in R/L , the r -component of the mechanical equilibrium equations (9) thus implies:

$$\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial u_r}{\partial r} - \frac{u_r}{r^2} \right) = 0 \quad (12)$$

The only solution of (12) that vanishes at $r = 0$ is of the form:

$$u_r(r, z, t) = r \bar{u}_r(z, t) \quad (13)$$

The molecular incompressibility (5) further implies that the axial displacement is independent of r at leading order:

$$u_z(r, z, t) = \bar{u}_z(z, t) \quad (14)$$

From the results above, we deduce that the radial stress σ_{rr} and shear stress σ_{rz} are solely functions of z and t , at leading order in $R=L$. Since both of these quantities vanish at the free surface, by virtue of the stress free boundary condition at the surface ($\sigma \cdot \vec{n} = \vec{0}$ at $r = R$), we have, again at leading order in $R=L$:

$$\sigma_{rr} = \sigma_{rz} = 0 \quad (15)$$

We can thus write:

$$u_r(r, z, t) = r \frac{2(\bar{c} - c_0)G\Omega - \sigma_{zz}(t)}{6G} \quad (16)$$

$$\frac{\partial \bar{u}_z}{\partial z} = \frac{(\bar{c} - c_0)G\Omega - \sigma_{zz}(t)}{6G} \quad (17)$$

$$\bar{\mu}(z, t) = \mu_0 + \frac{2G\Omega^2(1+\nu)}{3(1-2\nu)}(\bar{c} - c_0) - \frac{\Omega\sigma_{zz}(t)}{3} \quad (18)$$

We find the functions c and $\sigma_{zz}(t)$ from the boundary conditions and transport equation which reads in cylindrical coordinates:

$$\frac{\partial \bar{c}}{\partial t} + \frac{1}{r} \frac{\partial(rJ_r)}{\partial r} + \frac{\partial J_z}{\partial z} = 0 \quad (19)$$

where J_z is the flux in the lengthwise direction. Integrating (19) over the section of the fiber we find:

$$\frac{\partial \bar{c}}{\partial t} + \frac{2J_r^s}{R} + \frac{\partial J_z}{\partial z} = 0 \quad (20)$$

where J_r^s is the surface flux from the drop to the fiber. Beneath the drops, it is simply proportional to the difference between the

chemical potential in the drop μ_d and the chemical potential in the fiber μ_0 . Outside the drops, we assume that the fiber is impermeable and this flux is zero. Here we express the flux in term of the concentration field \bar{c} :

$$\begin{aligned} J_r^s &= -\frac{k1_d(z, t)}{\eta\Omega^2 h} (\mu_b - \bar{\mu}) \\ &= -\frac{k1_d(z, t)}{\eta\Omega^2 h} \left(\mu_b - \mu_0 + \frac{\Omega\sigma_{zz}(t)}{3} - \frac{2G\Omega^2(1+\nu)}{3(1-2\nu)}(\bar{c} - c_0) \right) \end{aligned} \quad (21)$$

where h is a length (presumably microscopic) that characterizes the interface such that the quantity $k/2h$ is a surface permeability. The function $1_d(z, t)$ is the indicative of the drops: it is a function which value is 1 if there is a drop on the fiber at position z and time t and which value is zero otherwise. We finally obtain the following equation which is essentially a diffusion equation with a source term at the position of the drops:

$$\frac{\partial \bar{c}}{\partial t} = D \frac{\partial^2 \bar{c}}{\partial z^2} + \frac{2D}{Rh} (c_{max}(t) - \bar{c}) 1_d(z, t) \quad (22)$$

where the time dependent equilibrium concentration $c_{max}(t)$ is:

$$c_{max}(t) = c_0 + \frac{3(1-2\nu)}{2G\Omega^2(1+\nu)} \left(\mu_b - \mu_0 + \frac{\Omega\sigma_{zz}(t)}{3} \right) \quad (23)$$

The effective diffusion coefficient D appearing in (22) is $D = \frac{2kG(1+\nu)}{3\eta(1-2\nu)}$. Finally we need an equation for the tension in the fiber. This equation is provided by the boundary condition at the end of the fiber. Here we focus on a fiber that is initially stretched and whose ends are subsequently held fixed. Since the displacements $u_z(-L, t)$ and $u_z(L, t)$ at the ends of the fiber are fixed, we can set $u_z(0, t) = 0$ without loss of generality and write $-u_z(-L, t) = u_z(L, t) = \varepsilon L$. Integrating (16) with respect to z in the interval $[-L, L]$ we obtain:

$$\sigma_{zz}(t) = 3G\varepsilon - \frac{G\Omega}{2L} \int_{-L}^L (\bar{c} - c_0) dz \quad (24)$$

3 Supplementary Movies

S1: Spontaneous localized fluid release: Two drops of $V = 2\mu l$ are placed on a PVS Fiber of initial radius $R = 250\mu m$. After fully swelling the region below and between the two drops, we see a new drop forming in the center as fluid is released from the fiber (localized deswelling). The two initial drop continue to be absorbed and/or interact with the center drop. The time is indicated in hh:mm:ss

S2: Swelling induced coalescence: Two drops of $V = 1.5\mu l$ are placed on a PVS Fiber of initial radius $R = 250\mu m$. After fully swelling the region below and between the two drops, one of the drops becomes very mobile. Eventually, the two drops coalesce completely forming a single drop that will slowly be absorbed. The time is indicated in hh:mm:ss

S3: Fluid release when reducing the tension in a fiber: A fiber of unstretched radius $R = 250\mu m$ is fully swollen under very high tension (initial stretch $\varepsilon_0 = 1$). It is then dried on the surface. At $t = 0$ (start of the video) the length of the fiber is reduced within a few seconds so that the tension drops almost instantly. Within minutes we observe drops appearing on the fiber as fluid is released. The time is indicated in hh:mm:ss

S4: Provoked coalescence: Two drops of $V = 2\mu\text{ l}$ are placed on a PVS Fiber of initial radius $R = 250\mu\text{m}$. The time is indicated in hh:mm:ss. The movie starts as the center at $z = 0$ is already saturated with fluid. As drops are added far from the center at $t = 40$ min they are quickly absorbed by swelling the fiber radially. The tension in the fiber is thus dropping very quickly. This decrease in tension provokes the coalescence of the two initial drops right after this tension drop at $t = 45$ min.