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Supplementary information

Bubble casting soft robotics

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Supporting Information:
Bubble Casting Soft Robotics

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In this supplementary document we provide the derivation of the flow equations predicting (S1) the fluid deposition on the wall following bubble injection and (S2) the film thickness from gravitational drainage.

S1 Bretherton's Problem

We leverage the large literature on the displacement of bubbles in channels [1, 2, 3, 4, 5] to rationalize the thickness h_i of polymer melt left on the tubular mold wall after the bubble injection. Below we present two ways of obtaining Eq. 1 of the main text using either scaling arguments [4] or lubrication theory [5].

S1.1 Scaling

As the bubble progresses with a velocity U, the melt viscous drag near the wall results in the deposition of a film of constant thickness h_i. This deposition deforms the liquid-air interface such that capillary forces resist the motion. A transition region of length λ appears between the thin film and the bubble front whose curvature is $2/R$ with R the tube radius. Balancing viscous and capillary forces in the transition region yields: $\mu U/h_i^2 \sim \gamma/R\lambda$, with μ the viscosity of the liquid film. The transition length λ is found by matching the curvature of the bubble front with the one in the transition region $2/R \sim 1/R + h_i/\lambda^2$. Replacing λ and rearranging provides the classic Bretherton's Law:

$$\frac{h_i}{R} \sim \mathrm{Ca}^{2/3}. \tag{S1}$$

At high capillary number $\mathrm{Ca} = \mu U/\gamma$, the film becomes thick and the assumption that the bubble front curvature is $2/R$ breaks down. Using the better approximation $2/(R - h_i)$ for the curvature changes the force balance to $\mu U/h_i^2 \sim \gamma/(R - h_i)\lambda$ and the transition length to $\lambda \sim \sqrt{(R - h_i)h_i}$. Rearranging gives the functional form:

$$\frac{h_i}{R} \sim \frac{\mathrm{Ca}^{2/3}}{1 + \mathrm{Ca}^{2/3}}. \tag{S2}$$

Using Eq. S2 Aussillous and Quéré justified the empirical fit for Bretherton's Problem at moderate capillary number [4]:

$$\frac{h_i}{R} = \frac{1.34\mathrm{Ca}^{2/3}}{1 + 1.34 \cdot 2.5\mathrm{Ca}^{2/3}}. \tag{S3}$$

S1.2 Theory

An extended Bretherton's model by Klaseboer et al. [5] provides a rigorous derivation of Eq. S3. Because the film thickness $h(z)$ is small compared to the tube radius R, we can use the lubrication approximation which simplify the Navier-Stokes Equations to $\frac{\mathrm{d}p}{\mathrm{d}z} = \mu\frac{\mathrm{d}^2 u}{\mathrm{d}r^2}$. Here, $u(r)$ is the velocity profile in the liquid film, in the frame of the moving bubble and $p(z)$ is the pressure. Using the no slip (wall moving at $u(0) = -U$) and shear-free ($\mathrm{d}u/\mathrm{d}r|_{r=h} = 0$) boundary conditions allow us to recover the velocity:

$$u(r) = \frac{1}{\mu}\frac{\mathrm{d}p}{\mathrm{d}z}\left(\frac{r^2}{2} - rh\right) - U. \tag{S4}$$

Mass conservation imposes that the flux in any section along the film must be constant and equal to $Q = -Uh_i$ such that:

$$\int_0^{h(z)} u(r)dr = -\frac{h(z)^3}{3\mu}\frac{\mathrm{d}p}{\mathrm{d}z} - Uh(z) = -Uh_i. \tag{S5}$$

Eq. S5 relate the pressure gradient in the film to its thickness: $\frac{\mathrm{d}p}{\mathrm{d}z} = -3\mu U\frac{h(z)-h_i}{h(z)^3}$. The pressure in the thin film is given by Laplace's law: $p(z) = -\gamma\frac{\mathrm{d}^2h}{\mathrm{d}z^2} - \frac{\gamma}{R-h_i} + P_{\text{air}}$. Differentiating the pressure and replacing it in the equation for the thickness yields:

$$\gamma\frac{\mathrm{d}^3h}{\mathrm{d}z^3} = 3\mu U\frac{h-h_i}{h^3}, \tag{S6}$$

which is the Landau-Levich Equation $\frac{\mathrm{d}^3\bar{h}}{\mathrm{d}\bar{z}^3} = \frac{\bar{h}-1}{\bar{h}^3}$ after rescaling the height $h = h_i\bar{h}$ and axial length $z = h_i(3\mathrm{Ca})^{-1/3}\bar{z}$. The Landau-Levich Equation has no known analytic solutions but posses two asymptotic solutions that match the geometry of the problem. First, as $z \to -\infty$ the thickness approaches the final thickness such that $\bar{h} = 1$ ($h = h_i$). Second, as $z \to \infty$ then $h \gg h_i$ such that Eq. S6 simplifies to $\mathrm{d}^3h/\mathrm{d}z^3 = 0$. Integrating twice gives:

$$h(z) = \frac{1}{2h_i}(3\mathrm{Ca})^{2/3}C_1(z-z_0)^2 + h_iC_2. \tag{S7}$$

Equation S7 is a parabolic approximation of a sphere with curvature $C_1(3\mathrm{Ca})^{2/3}/h_i$ whose apex is at z_0. With a bubble front curvature $2/R_b$ one could match the curvatures and rearrange to arrive at:

$$\frac{h_i}{R_b} = C_1(3\mathrm{Ca})^{2/3}. \tag{S8}$$

Note that Eq. S8 has the same form as Eq. S1. The prefactors were calculated by Bretherton [3] and found to be $C_1 = 0.643$ and $C_2 = 2.79$. The difference with Eq. S1 lies in the bubble front curvature R_b which is found by matching the transition region to the bubble nose [5] at the matching point z_0: $h(z_0) = h_iC_2 = R - R_b$. Combining this with Eq. S8 and rearranging recovers Eq. 1:

$$\frac{h_i}{R} = \frac{1.34\mathrm{Ca}^{2/3}}{1 + 1.34C_2\mathrm{Ca}^{2/3}}. \tag{S9}$$

The small discrepancy between the calculated and fitted value of C_2 is discussed in [5] and was attributed to the matching leading to Eq. S7 being done at $\bar{h} \to \infty$ rather than the large but finite experimental value.

S2 Film drainage of Newtonian fluids

Here, we describe how the drainage of the annulus of thickness h_i left by the bubble forms the upper membrane of our actuators.

We introduce the cylindrical coordinates $\{r, \psi, z\}$, assume that the Reynold's number is low (since we deal with viscous fluids), and that the film is thin $\delta = h_i/R \ll 1$ which allows us to use the lubrication approximation. Assuming no flow in the z direction, the Navier-Stokes equations simplifies to:

$$0 = -\frac{\partial p}{\partial r} - \rho g \cos\psi, \tag{S10a}$$

$$0 = -\frac{1}{r}\frac{\partial p}{\partial \psi} + \rho g \sin\psi + \mu\left(\frac{\partial}{\partial r}\left[\frac{1}{r}\frac{\partial (ru)}{\partial r}\right] + \frac{1}{r^2}\frac{\partial u^2}{\partial \psi^2}\right), \tag{S10b}$$

with $u(r, \psi)$ the fluid velocity and $p(r, \psi)$ the pressure.

The pressure at the liquid-air interface is the Laplace pressure: $p(r = R - h, \psi) = P_{\text{air}} - \gamma\kappa$ where γ is the surface tension, and κ is the interface curvature. Integrating Eq. S10a gives the pressure in the fluid: $p(r, \psi) = P_{\text{air}} - \gamma\kappa + \rho g(R - r - h)\cos\psi$. Under the assumption $\delta \ll 1$, introducing the change of coordinates $\bar{r} = R - r$ and keeping first-order terms greatly simplifies Eq. S10b to [6]

$$\frac{\partial^2 u}{\partial \bar{r}^2} = -\frac{\gamma}{\mu R}\frac{\partial \kappa}{\partial \psi} + \frac{\rho g}{\mu R}\frac{\partial h}{\partial \psi}\cos\psi - \frac{\rho g}{\mu}\sin\psi. \tag{S11}$$

Integrating Eq. S11 twice with no-slip boundary condition $u(\bar{r} = 0) = 0$ at the solid-liquid interface and zero-shear stress $\partial u/\partial \bar{r}|_{\bar{r}=h} = 0$ at the air-liquid interface yields the tangential velocity:

$$u(\bar{r}, \psi) = \left(\frac{\gamma}{\mu R}\frac{\partial \kappa}{\partial \psi} - \frac{\rho g}{\mu R}\frac{\partial h}{\partial \psi}\cos\psi + \frac{\rho g}{\mu}\sin\psi\right)\left(h\bar{r} - \frac{\bar{r}^2}{2}\right). \tag{S12}$$

From the velocity, we compute the volumetric flow rate $Q = \int_0^h u(\bar{r}, \psi) d\bar{r}$. Using mass conservation in cylindrical coordinates $\partial h/\partial t + R^{-1}\partial Q/\partial \psi = 0$, we eventually obtain the lubrication equation:

$$\frac{\partial h}{\partial t} + \frac{1}{3\mu R}\frac{\partial}{\partial \psi}\left[h^3\left(\underbrace{\frac{\gamma}{R}\frac{\partial \kappa}{\partial \psi}}_{\textbf{I}} - \underbrace{\frac{\rho g}{R}\frac{\partial h}{\partial \psi}\cos\psi}_{\textbf{II}} + \underbrace{\rho g \sin\psi}_{\textbf{III}}\right)\right] = 0. \tag{S13}$$

Where the leading order curvature derivative term is $\frac{\partial \kappa}{\partial \psi} = (\frac{\partial^3 h}{\partial \psi^3} + \frac{\partial h}{\partial \psi})/R^2$. In the spatial variation of the flux, term **I** corresponds to the surface tension effects, term **II** corresponds to the hydrostatic pressure distribution, and term **III** corresponds to the gravitational drainage.

The lubrication equation can be nondimensionalized by rescaling the height by the initial film thickness $h = h_i\hat{h}$ and the time by the initial drainage time $t = \tau_d^*\hat{t}$ (see Eq. 8 in Methods section 'Timescale separation'). The new equation reads:

$$\frac{\partial \hat{h}}{\partial \hat{t}} + \frac{1}{3}\frac{\partial}{\partial \psi}\left[\hat{h}^3\left(\frac{\delta^2}{\text{Bo}}\left(\frac{\partial^3 h}{\partial \psi^3} + \frac{\partial h}{\partial \psi}\right) - \delta\frac{\partial h}{\partial \psi}\cos\psi + \sin\psi\right)\right] = 0, \tag{S14}$$

where $\text{Bo} = \rho g h_i R/\gamma$ is a modified Bond number.

Keeping first order terms in Eq. S14, we obtain the zeroth order solution (which is the exact solution at the apex) $\hat{h}(\hat{t}) = (1 + 2\hat{t}/3)^{-1/2}$ which can be simplified to $\hat{h}(\hat{t}) = \sqrt{3/(2\hat{t})}$ at late time $t \gg \tau_d^*$ [6] or in dimensional form:

$$h(t) = \sqrt{\frac{3\mu R}{2\rho g t}}. \tag{S15}$$

References

[1] Fred Fairbrother and Alfred E Stubbs. 119. studies in electro-endosmosis. part vi. the “bubble-tube” method of measurement. *Journal of the Chemical Society (Resumed)*, pages 527–529, 1935.

[2] GI Taylor. Deposition of a viscous fluid on the wall of a tube. *Journal of Fluid Mechanics*, 10(2):161–165, 1961.

[3] Francis Patton Bretherton. The motion of long bubbles in tubes. *Journal of Fluid Mechanics*, 10(2):166–188, 1961.

[4] Pascale Aussillous and David Quéré. Quick deposition of a fluid on the wall of a tube. *Physics of fluids*, 12(10):2367–2371, 2000.

[5] Evert Klaseboer, Raghvendra Gupta, and Rogerio Manica. An extended bretherton model for long taylor bubbles at moderate capillary numbers. *Physics of Fluids*, 26(3):032107, 2014.

[6] Daisuke Takagi and Herbert E Huppert. Flow and instability of thin films on a cylinder and sphere. *Journal of fluid mechanics*, 647:221, 2010.