Deformation of Fluid and Solid Interfaces in Viscous Flow

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Abstract

This dissertation covers a variety of problems in viscous fluid mechanics, with a uniting theme of deformable interfaces. The work is divided into three separate sections exploring three distinct geometries: a liquid film on a rough surface, a liquid bridge between two flexible solids, and a flexible fiber in a confined viscous flow.

In the first section, we explore the dynamic of a liquid film on a rough or patterned surface. A liquid-infused surface like this displays many of the same useful properties as conventional gas-cushioned superhydrophobic surfaces. However, liquid-infused surfaces may drain due to an external shear flow, causing the surface to lose its advantageous properties. Using experiments and analytical theory we examine shear-driven drainage of these surfaces, with the goal of understanding and thereby mitigating this failure mode. On patterned surfaces exposed to a known shear stress, we find that a finite length of the surface remains wetted indefinitely, despite the fact that no physical barriers prevent drainage. We then consider the effects of physical barriers, and explore how they modify the drainage of the film. We conclude this section with a study of how drainage can be prevented through the use of chemical patterning, and a brief extension where we propose a new technique for measuring the velocity profile of a thin film flow.

In the next section, we consider how a wetting droplet trapped in the thin gap between two elastic bodies will deflect the bodies towards one another. The deformation increases the total capillary adhesion force between the bodies by increasing the contact radius and narrowing the gap height. We present experiments, scalings, and closed-form solutions that describe the deformation.

In the final section, we present a mathematical model and corresponding series of microfluidic experiments examining the flow of a viscous fluid past an elastic fibre in a three-dimensional channel. Experiments show that there is a linear relationship between deflection and flow rate for highly confined fibers at low flow rates, which
inspires an asymptotic treatment of the problem in this regime. The analysis yields insight into the competing effects of flexion and leakage.
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This dissertation carries T-3302 in the records of the Department of Mechanical and Aerospace Engineering.
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Chapter 1

Shear-driven failure of liquid-infused surfaces

The majority of this chapter was recently published as a journal article with the same title 1. The research was performed in collaboration with Ian Jacobi and Howard Stone; they are co-authors of the article.

1.1 Introduction

Liquid-infused surfaces demonstrate a remarkable array of useful properties, from omniphobicity 2, 3, 4 and bio-fouling resistance 5, to enhanced heat transfer 6, 7 and drag reduction 8, 9, 10. Unlike traditional superhydrophobic materials, liquid-infused surfaces are robust against pressure-induced failure, making them particularly attractive for submerged applications 11, 12, 13, 14, 15, 16, 17. However, when these surfaces are immersed in dynamic fluid environments, external flow can shear away the infused liquid layer that is responsible for their unique properties.

Robust implementation of liquid-infused surfaces thus requires a thorough understanding of the dynamics of a liquid lubricant trapped within a patterned substrate that is exposed to shear. This fundamental shear-driven drainage problem also ap-
plies to a wide variety of structurally similar situations, including cleaning oily surface contaminants from textiles [18], extraction of residual oil from permeable rocks [19], liquid-vapor interactions in micro-patterned heat pipes [20, 21], and the shear flow over hydrophobic mucus trapped on rough biological tissue [22, 23]. Existing theories that govern liquid films trapped in rough or patterned surfaces are able to describe the process of imbibition [24, 25, 26, 27], the flow of a superficial fluid film above the height of the underlying surface pattern [28, 29], and the static configurations of wetting drops [30]. In addition, the steady-state shape of a shear- or gravity-driven film that coats individual surface features has been explored [31, 32, 33, 34]. Despite these advances, current theories are unable to predict whether a patterned surface will retain an infused liquid when subject to an external flow.

1.2 Experiments

We report a series of experiments to study the behavior of liquid-infused patterned surfaces exposed to the flow of an immiscible liquid. A microfluidic flow cell was constructed from transparent epoxy [35] with a patterned surface imprinted on a section of its floor (Fig. 1.1(a)-(c)). The surface pattern in this experiment consists of 50 streamwise grooves with width \( w = 8.8 - 9.2 \, \mu m \) and height \( h = 10.0 \, \mu m \) (Fig. 1.1(d)) that end upstream in a 1 mm by 1 mm well of equal depth to create the open end shown schematically in Fig. 1.2(c). The pattern is initially filled with silicone oil mixed with fluorescent dye, and connects to a downstream reservoir of oil at the terminus of the flow cell. The external aqueous fluid enters the upstream inlet of the device, and exits through a slot-shaped outlet that is upstream of the terminus. This configuration ensures that the draining oil does not block the flow of the external phase, and that the external flow is not constricted as it exits the device.
Figure 1.1: (a) Cross-section of the microfluidic flow cell, showing the configuration at the beginning of an experiment. Distances in the $y$-direction are exaggerated. The aqueous solution (blue) flows in the left inlet and out the first (slot-shaped) outlet. The grooves are filled with oil (green), and connect to the reservoir of oil at the flow cell terminus. (b) This planform view shows the entire device before drainage commences. Low viscosity oil fills the 50 longitudinal grooves at the center of the device and fluoresces green. (c) Snapshots of a sample shear-driven drainage experiment subject to an aqueous flow of $Q = 2$ mL/min ($\tau_{yx} = 5.2$ Pa). (d) Micrograph of the silicon wafer micro-pattern that is used to mold the grooves, including the surface profile (purple). Grooves appear dark gray and walls appear light gray.

The external aqueous phase consists of a 1:1 weight mixture of glycerol and water, with a viscosity of $\mu_{aq} = 5.4$ mPa·s and density of $\rho_{aq} = 1.125$ g/mL. Two different silicone oils with high index of refraction are used for the infused phase: 1) 1,1,5,5-Tetraphenyl-1,3,3,5-tetramethyltrisiloxane (Gelest PDM-7040), with viscosity $\mu_o = 42.7$ mPa·s, density $\rho_o = 1.061$ g/mL, and interfacial tension with the aqueous solution $\gamma = 29.0$ mN/m; and 2) 1,1,3,5,5-Pentaphenyl-1,3,5-trimethyltrisiloxane (Gelest PDM-7050) with $\mu_o = 201$ mPa·s, $\rho_o = 1.092$ g/mL, and $\gamma = 28.2$ mN/m.
For both oils, the receding contact angle $\theta = 56 \pm 4^\circ$. Note that in both cases the trapped oil is more viscous than the external aqueous solution, by a factor of either 5.3 or 37. These oils were chosen because their index of refraction is close to that of the epoxy that is used to construct the microfluidic device, and because silicone oils are less susceptible to contamination from surface-active components than hydrocarbon oils. To aid in visualization for the macro- and micro-scale experiments, the oils are mixed with Tracer Products TP-4300 UV Fluorescent Dye in a volume ratio of 1000:2. We assume that the aqueous solution in the flow cell has a planar Poiseuille profile and hence that the wall shear stress imposed on the textured substrate is given by $\tau_{yx} = 6\mu_{aq} Q/WH^2$, where the cell has width $W$ and height $H$, in the same cardinal directions as $w$ and $h$ in the groove. The maximum Reynolds number in the aqueous phase, defined as $\rho_{aq} Q/\mu_{aq} W$, is $\mathcal{O}(1)$, whereas the maximum Reynolds number of the oil in the grooves, defined as $\rho_o \tau_{yx} h^2/\mu_o^2$, is $\mathcal{O}(10^{-2})$. The maximum Bond number of the oil in the grooves, defined as $w^2 g (\rho_{aq} - \rho_o)/\gamma$, is $\mathcal{O}(10^{-4})$. Thus, surface tension and viscous forces dominate flow within the grooves.

Experiments were performed to determine whether the fluorescent dye may act as a surfactant and thereby influence the drainage behavior. Using the confocal microscope setup that is described later, experiments were performed with a variety of dye concentrations, ranging from 0 : 1000 to 10 : 1000 by volume (with the confocal setup, it is possible to measure the interface location without any dye, by looking at reflection). The drainage behavior in all cases was identical within the resolution of our measurements, indicating that the dye had negligible effect on flow. However, one drawback of the dye was that the brightness of the dye decreased over time; over the course of a two-hour experiment, the fluorescence intensity decreased by roughly a factor of two. This occurred regardless of whether the experiment was illuminated throughout that time interval, leading us to conclude that it was not a photobleaching effect, and that the dye may slowly partition into the external aqueous
Figure 1.2: (a) Representative groove cross-sections from the steady-state configuration of an experiment conducted at $Q = 2$ mL/min with low viscosity oil, taken at the outlet slot ($x = 0$ mm) and the far upstream end of the wetted groove ($x = -5.5$ mm). (b) The steady-state deflection at the center of the groove, $\delta(x)$, with theoretical predictions from Eq. (1.15) plotted as dashed lines. Gray is low viscosity oil at $Q = 2$ mL/min, red is high viscosity oil at the same flow rate, and blue is low viscosity oil at $Q = 1$ mL/min. (c) Schematic of one groove, showing geometric parameters and the shape of the interface deduced from (b).

phase. However, the dye was never observed to absorb into the solid microstructure, in contrast to other dyes that were used such as Nile Red and DiI.

The entire microfluidic device, including the roughness microstructure, is molded from Norland Optical Adhesive (NOA 81) using the ‘sticker’ technique[35]. The flow cells are 7 mm wide by 180 $\mu$m deep, and 45 mm long, with the roughness pattern positioned near the spanwise center of the device. There is an inlet port at the upstream end of the device, and two ports at the downstream end: one filling port far downstream at the terminus, and a second slot-shaped port that serves as the outlet and is 10 mm upstream of the terminus. The micro-pattern is 36 mm long, and is located such that the downstream ends of the grooves lie at the downstream terminus of the flow cell. The pattern is entirely filled with oil at the beginning of a drainage
experiment, and connects to a downstream reservoir of oil, while the remainder of the device is filled with the aqueous solution, whose flow will generate the external shear stress.

The flow cell geometry is molded out of the epoxy onto a piece of glass with dimensions $25 \times 75 \times 2$ mm (black glass is used to block background fluorescence in the experiments). The roughness pattern is molded out of epoxy on a clear glass No. 1 coverslip of dimensions $24 \times 60$ mm, before the flow cell and pattern are stuck together. To create the pattern used for the flow cell, a negative of the geometry is cut from a sheet of adhesive Kapton film (thickness 180 µm) using a laser cutter. The adhesive pattern is then stuck onto a 4” diameter silicon wafer. Photolithography and deep reactive ion etching (Bosch process, 46 cycles) are used to etch the roughness pattern into a separate silicon wafer. An inverse mold of both patterns is created using PDMS (Sylgard 184), and then the epoxy is molded from the PDMS. To ensure high fidelity, the PDMS molds that are used for the micropattern are degassed under vacuum overnight before each use and cleaned with a 1 M solution of NaOH after each use.

To create the connection ports, holes and slots are drilled into the black glass using a diamond-coated bit prior to molding. During the molding process, the holes are partially blocked with HDPE tubing, and the slot is filled with a strip of HDPE (254 µm thick, 9 mm wide). The HDPE does not stick to the epoxy and is removed after curing, to create the ports. The circular ports are trimmed with a countersink drill bit, and the slot-shape port is manually trimmed with a razor blade. Then, the channel side is plasma treated for five minutes to make it hydrophilic and prevent oil drops from adhering to that side.

The cure time of the patterned slide is set so that the epoxy remains slightly tacky. Then, the two epoxy-glass laminates are pressed together to create the microfluidic device. Inlet and outlet tubing are inserted into PDMS blocks that are hole-punched
and then bonded to the black glass above the pre-formed holes in the epoxy. Devices made of Norland epoxy are much stiffer than those made of PDMS and thus do not deform appreciably at the high flow rates that occur in our experiments. Furthermore, the oils used in the experiments would swell PDMS, but they do not swell the Norland epoxy.

A unique setup allows us to image the oil in the grooves with a resolution as small as the width of one groove (10 µm), but with a field of view as large as the whole device (45 mm). The microfluidic device is placed upside down and illuminated with an array of LEDs with peak wavelength 395 nm. The device is imaged from above with a vertically-mounted Nikon D90 DSLR camera outfitted with a Nikon AF Micro-Nikkor 200 mm lens and a Tiffen Yellow 8 filter (Wratten 8 transmission spectrum) that blocks the excitation light but transmits the fluorescence from the UV dye. We shroud the entire setup in black fabric to block external light. The aqueous solution is pumped into the device using one or two 140 mL syringes mounted on a syringe pump (Harvard Apparatus, PHD-2000).

Using micro-fabricated grooves to study lubricant drainage ensures a controlled and reproducible surface topography that is invariant in the streamwise direction, thereby providing a system amenable to a fluid dynamical description. The lessons learned from studying this geometry can be applied to predicting the drainage behavior of surfaces with more complicated topographies, as we demonstrate below. Indeed, there is a strong precedent in the study of capillary flows for such a ‘reduced-order’ approach to treating complicated geometries [24, 29]. The particular geometry of streamwise grooves also represents what appears to be the ‘worst-case’ surface configuration for oil retention. This worst-case scenario is instructive for evaluating many real-world liquid-infused surfaces, since rough surfaces inevitably have some degree of streamwise connectivity – either by accident or by design – that allows fluid in upstream portions of the pattern to drain downstream.
At the beginning of an experiment, the entire device is first filled with oil through the filling port at the flow cell terminus; this port is then clamped shut with locking forceps and the aqueous solution is pumped into the inlet at 2.5 µL/min, slowly clearing the bulk oil from the portion of the device between the inlet and the outlet slot, but leaving the oil infused in the pattern. Next, the clamp is switched from the filling port to the outlet slot, so that the interface between the oil and aqueous solution retreats towards the flow cell terminus. When the interface reaches a location midway between the outlet slot and the terminus, the clamp is switched back to the filling port, so that the interface halts, a reservoir of oil is established at the terminus, and the flow of the aqueous solution is redirected back to the outlet slot.

A time series of photographs from a typical experiment is shown in Fig. 1.1(c), demonstrating the characteristic drainage behavior (see Movie 1): under the influence of shear from the aqueous phase, the oil in the upstream portion of the pattern dewets first, with a dewetting front that propagates downstream. The front initially propagates rapidly, before slowing and eventually stopping at a steady-state streamwise position; the length of fluid retained in the pattern between this final front location and the slot-shaped outlet is defined as the steady-state length, $L_\infty$ (Fig. 1.1(c)).

Since the streamwise grooves terminate in a fluid reservoir, there is no physical barrier to drainage of the oil, and thus the existence of steady-state oil retention may seem non-intuitive. To clarify the mechanism that leads to oil retention, we perform identical experiments using a confocal microscope, and observe the steady-state configuration of the oil at the scale of the pattern itself. Cross-sectional ($yz$-plane) images of the steady-state oil distribution are taken at regular intervals in the streamwise ($x$) direction along the length of the filled portion of the groove. Two representative images are shown in Fig. 1.2(a). The fluorescent oil (represented as red) is index-matched with the solid so that the interface between the oil/solid and aqueous phase is visible in reflection (represented as green).
A Leica TCS-SP5 confocal microscope is used to capture the images of the groove cross-sections (63× oil objective, 514 nm laser). The same dye and concentration as above is used for these experiments, though the fluorescence is used only to locate the grooves. The y-location of the interface is determined entirely from the peak in reflection intensity so as to eliminate any ambiguity in data processing. The index of refraction of the oils is closely matched to the index of refraction of the solid, which in turn is closely matched to the index of refraction of the immersion oil of the microscope.

1.3 Discussion and Theory

The oil-aqueous interface is deflected inward towards the substrate and appears to have a constant curvature, $\kappa$, in the cross-sectional ($yz$) plane. Because the length of the filled portion of the groove is much longer than the width or height of the groove, this cross-sectional interfacial curvature dominates over curvature in the streamwise/wall-normal ($xy$) plane. The pressure drop across a curved liquid-liquid interface is equal to $\kappa$ multiplied by the surface tension of the interface, $\gamma$. Since the interface is deflected inwards, the pressure is lower in the oil than in the aqueous phase. Thus, the pressure within the oil decreases in the direction opposite the flow of the external phase. This adverse pressure gradient drives recirculation of the oil trapped in the groove, countering the external shear stress, and provides the physical mechanism for a steady-state wetting configuration under shear.

We note that this explanation of the oil retention mechanism rests on a number of assumptions about the system: the Reynolds number in the oil $\rho_o \tau_{yx} h^2/\mu_o^2 \ll 1$, indicating negligible inertial effects, and the Bond number $w^2 g (\rho_{aq} - \rho_o)/\gamma \ll 1$, indicating that gravity is negligible. These assumptions apply to most applications of liquid-infused surfaces. Furthermore, we ignore long-range forces (such as van der
Waals); though this assumption is valid for the micro-scale patterns of the current experiment, long-range forces may be relevant for certain chemistries on surfaces with nano-scale geometries. Finally, we assume that $\mu_o \gg \mu_{aq}$, so that the shear stress imposed by the aqueous flow is effectively unchanged by the oil.

The adverse pressure gradient driving oil in the upstream direction depends on the gradient in curvature of the interface over the length of the groove. At the downstream end, where the aqueous fluid exits the flow cell, the interface is flat, indicating zero pressure drop across the interface. At the upstream end, the minimum radius of curvature, $r_{\text{min}}$, is determined by the groove width, $w$, and the receding contact angle, $\theta$, or, for wider grooves, the aspect ratio of the groove, $w/h$. The interfacial deflection at the groove center, $\delta$, varies as $\delta \sim x$ between these two limits, as shown in Fig. [1.2](b). Since $\delta \sim \kappa$ for small deflections, $d\kappa/dx$ is approximately constant, indicating that the pressure gradient within the oil is constant.

We now construct a quantitative model to predict the dynamics of drainage from the grooved pattern based on the flow reversal mechanism we inferred from interfacial measurements. Our goal is to predict how the wetted length of the groove, $L(t)$, evolves under the action of an applied shear stress, $\tau_{yx}$. The drainage dynamics are modeled as a function of flow and geometrical properties in order to determine the rate at which the fluid drains from the grooves, and the shape of the fluid-fluid interface once a steady state is established. Each groove has width $w$, height $h$, and initial filled length $L_0$. The instantaneous filled length of the groove, $L(t)$, is defined as the distance between the outlet slot of the flow cell and the location where the fluid-fluid interface first touches the bottom of the groove (see Figure 1 in main text).

The shear stress from the flow of the external aqueous solution drags the oil downstream. At the fluid-fluid interface, the velocities and shear stresses in the two phases are equal. However, since the depth of the flow cell is much greater than the depth of the groove, and the trapped oil is more viscous than the aqueous solution,
the velocity of the former is assumed negligible compared to typical velocities in the latter. The flow of the aqueous solution can therefore be considered unperturbed by the presence of a non-solid boundary; it passively imposes a shear stress, \( \tau_{yx} \), on the trapped oil. It follows that the velocity-matching condition can be neglected, since the aqueous solution will accommodate whatever (much lower) velocity occurs at the fluid-fluid interface.

If oil is to be retained under the action of shear at steady state, there must be recirculation in the streamwise direction, driven by an adverse pressure gradient within the groove, \( dp/dx \). We assume low-Reynolds-number unidirectional flow within the groove, so that the streamwise velocity in the groove \( u(y,z) \) satisfies

\[
\mu_o \nabla^2 u = \frac{dp}{dx}. \tag{1.1}
\]

To make the problem analytically tractable, we assume that the groove is filled completely in the \( yz \)-plane, and thus that the fluid-fluid interface is flat with the top of the groove. The no-slip condition applies at the side and bottom walls of the groove, and the external shear is specified at the fluid-fluid interface, where \( \mu_o \partial u/\partial y = \tau_{yx} \) and \( \tau_{yx} \) is determined from the Poiseuille flow of the aqueous liquid.

Since (1.1) is linear, we consider \( u(y,z) \) as a superposition of two components: a shear-driven contribution \( u_s(y,z) \) and a pressure-driven contribution \( u_p(y,z) \). The flow fields for both components are determined using separation of variables and an eigenfunction expansion over the rectangular domain \( h \times w \), with appropriate boundary conditions applied for calculating each component of \( u(y,z) \) \[36\]. To calculate the shear-driven flow field, the pressure gradient is set to zero (\( dp/dx = 0 \)) and the boundary-value problem is solved with the prescribed shear-stress at the top interface and no-slip along the walls and floor of the groove. Then, integrating \( u_s(y,z) \) over
the cross-section, an expression for the shear-driven fluid flux, \( q_s \), is found:

\[
q_s = c_s \frac{\tau_{yz} \lambda h^2}{\mu_o} \text{ with } c_s = \frac{1}{2} - \frac{4h}{w} \sum_{n=0}^{\infty} \frac{(-1)^n}{\lambda_n^4} \tanh\left(\frac{\lambda_n w}{2h}\right),
\]

(1.2)

where \( \lambda_n = (n + \frac{1}{2}) \pi \). To determine the pressure-driven flow field, the boundary condition at the top interface is set to be shear-free (\( \mu_o \partial u / \partial y = 0 \)), no-slip is again specified along the walls and floor of the groove, and the same procedure is employed with non-zero \( dp/dx \) to solve for \( u_p(y, z) \). The pressure-driven flux, \( q_p \), is then

\[
q_p = -c_p \frac{wh^3 dp}{\mu_o dx} \text{ with } c_p = \frac{1}{3} - \frac{4h}{w} \sum_{n=0}^{\infty} \frac{1}{\lambda_n^5} \tanh\left(\frac{\lambda_n w}{2h}\right),
\]

(1.3)

with the same eigenvalues \( \lambda_n \) as before.

At the length-scale of the current experiment, long-range forces (such as van der Waals) are negligible, so that surface tension forces alone induce the pressure gradient through the capillary pressure. As mentioned in the main text, experiments performed using the confocal microscope indicate that at steady-state the interface shape varies smoothly along the length of the groove. Towards the downstream end of the groove, the interface is effectively flat since it must match the interfacial curvature of the reservoir, which is essentially zero. At the upstream end the interface bends inward, producing a negative capillary pressure within the groove. The length of the filled portion of the groove is much longer than the width or height of the groove, so interfacial curvature in the \( yz \)-plane dominates over curvature in the \( xy \)-plane. We assume that interfacial curvature in the \( yz \)-plane is constant at a given \( x \)-location since flow in the groove is unidirectional and the Bond number based on the groove width is small, indicating that gravity has a negligible effect (see Methods in main text).

The pressure change corresponding to the change in interfacial curvature between upstream and downstream ends of the filled portion of the groove is

\[
\Delta p \approx \frac{\gamma}{r_{\text{min}}},
\]
where \( r_{\text{min}} \) is the minimum radius of curvature in the \( yz \)-plane at the upstream end. The pressure drop is approximated as being constant along the filled length of the groove, so that \( dp/dx \approx \Delta p/L \). For grooves with an aspect ratio smaller than \( w/h = 2(\sec \theta + \tan \theta) \), \( r_{\text{min}} \) is determined by the receding contact angle, \( \theta \). If the groove has a larger aspect ratio, however, the curved interface touches the bottom of the groove before the receding contact angle is reached. Solving for \( r_{\text{min}} \), we substitute into equation (1.3) to find

\[
q_p = -c_p \frac{wh^3 \gamma}{\mu_o r_{\text{min}} L},
\]

(1.4)

with

\[
r_{\text{min}} = \begin{cases} 
\frac{w}{2 \cos \theta} & \text{for } w/h \leq 2(\sec \theta + \tan \theta) \\
\frac{h \left(1 + \left(\frac{w}{2h}\right)^2\right)/2}{w} & \text{for } w/h > 2(\sec \theta + \tan \theta) 
\end{cases}
\]

(1.5)

Note, however, that if the dewetting happens very rapidly, with a Capillary number that is \( \mathcal{O}(1) \), the contact line will not retreat with the equilibrium receding contact angle, \( \theta \), and will instead retreat with a dynamic contact angle that is lower than the equilibrium value. This could result in the oil draining slower than it would otherwise, due to the greater Laplace pressure. This dynamic contact angle should not effect the steady-state length, though, since the contact line must slow down as the oil in the groove approaches an equilibrium wetting state.

Now, with expressions for both the shear-driven fluid flux, \( q_s \), and the pressure-driven flux, \( q_p \), conservation of volume requires that the sum of these fluxes equals the flux of fluid into or out of the system, which is denoted \( q_d \), the drainage flux, so that

\[
q_s + q_p = q_d.
\]

(1.6)
If the groove has reached its steady-state length, \( q_d = 0 \). Substituting this condition into (1.6), we find for the steady-state length,

\[
L_\infty = \frac{c_p h}{c_s r_{\min} \tau_{yx}} \gamma,
\]

where the pre-factor contains all effects of the groove geometry.

The groove drains when \( q_s \) and \( q_p \) are not balanced, resulting in a change in volume of the oil trapped in the groove. Thus, \( q_d = -dV/dt \), where \( V(t) \) is the instantaneous volume of oil in the groove. To calculate \( V(t) \) in a groove of filled length \( L(t) \), we assume that the curvature of the interface varies linearly within the filled portion of the groove (see Figure 2 in main text), corresponding to a constant pressure gradient. We integrate the cross-sectional area from \( x = -L(t) \) to \( x = 0 \) to calculate \( V(t) \), and take \( dV/dt \), to arrive at

\[
q_d = -c_d w h \frac{dL}{dt}.
\]

(1.8)

The constant \( c_d \) is another geometric prefactor representing the fraction of the groove cross-sectional area \( w h \) that is filled by the oil, averaged over the length \( L \), and is given by

\[
c_d = 1 - \frac{r_{\min}}{h} \left( 1 - \sqrt{\frac{1}{4} - \frac{w^2}{16r_{\min}^2}} \right) + \frac{r_{\min}^2}{wh} \csc^{-1} \left( \frac{2r_{\min}}{w} \right).
\]

(1.9)

Substituting the relationships for \( q_s \), \( q_p \), and \( q_d \) into equation (1.6) results in a Lucas-Washburn-type equation for the wetted length of the groove [37, 38]. We non-dimensionalize length as \( \bar{L} = L/L_\infty \) and time as \( \bar{t} = t/t_c \), with

\[
t_c = \frac{C_d C_p H_o \gamma}{C_s^2 r_{\min} \tau_{yx}^2},
\]

(1.10)

so that the non-dimensional balance equation (1.6) becomes

\[
1 - \frac{1}{\bar{L}} = -\frac{d\bar{L}}{d\bar{t}}.
\]

(1.11)
Equation (1.11) with initial condition $\tilde{L}(\tilde{t} = 0) = \tilde{L}_0$ can be integrated trivially but the solution remains an implicit function of $\tilde{L}$ and $\tilde{t}$,

$$\tilde{L} = \tilde{L}_0 - \tilde{t} + \log \left( \frac{\tilde{L}_0 - 1}{\tilde{L} - 1} \right). \quad (1.12)$$

For $\tilde{L}_0 \gg 1$, (1.12) asymptotes to $\tilde{L} = \tilde{L}_0 - \tilde{t}$ as $\tilde{t} \to 0$, meaning that the length decreases linearly at early times for long grooves. The rate of decrease, which is unity in dimensionless terms, is $c_s \tau_{yz} h / (\mu_0 c_d)$ dimensionally. This value is the average fluid velocity from (1.2) for shear-driven flow in the groove, divided by $c_d$, to compensate for the fact that the groove is not filled uniformly to the top. When the length of the fluid plug is very large the pressure gradient due to the capillary pressure is negligible, so the groove freely drains under the influence of shear only. As $\tilde{t} \to \infty$ on the other hand, (1.12) approaches $\tilde{L} = 1$ exponentially, with a time constant of unity.

The fact that the system displays Lucas-Washburn dynamics is not a coincidence; the Lucas-Washburn equation is typically associated with imbibition in a porous media or rough surface against the influence of gravity (so-called capillary rise), and indeed a direct analogy can be made between shear- and gravity-driven drainage from a rough surface. Carrying through the analysis, it becomes clear that the equilibrium length in our experiment is a shear-driven equivalent to the classical capillary rise height.

To calculate the expected shape of the steady-state interface, we rewrite the pressure-driven flux in terms of a local pressure gradient rather than an average pressure gradient. With $p(x) = \gamma / r(x)$, where $r(x)$ is the local radius of curvature in the $yz$-plane (again assuming that curvature in the $xy$-plane is negligible), we then relate the local curvature $r(x)$ to the local deflection of the center of the groove, $\delta(x)$, through

$$r(x) = \frac{\delta(x)}{2} \left( 1 + \left( \frac{w}{2\delta(x)} \right)^2 \right). \quad (1.13)$$
Substituting \( p(x) = \gamma/r(x) \) into (1.3), and equating with (1.2) for the steady-state balance as before, we arrive at

\[
\frac{c_s \tau_{yx}}{c_p \gamma h} = \frac{d}{dx} \left( \frac{2}{\delta} \left[ 1 + \left( \frac{w}{2\delta} \right)^2 \right]^{-1} \right). \tag{1.14}
\]

We non-dimensionalize with \([\delta] = w/2\) and \([x] = 4c_p h \gamma/c_s w \tau_{yx}\) so that \(\tilde{\delta} = \delta/|\delta|\) and \(\tilde{x} = x/|x|\). Integrating in \(\tilde{x}\), we set the boundary condition that \(\tilde{\delta}(0) = 0\) to account for the flat interface at the downstream end of the groove. Thus, the interface shape is given by

\[
\tilde{\delta} = \frac{-1 + \sqrt{1 - 4\tilde{x}^2}}{2\tilde{x}} \tag{1.15}
\]

between \(\tilde{x} = 0\) and the upstream end of the wetted portion of the groove, at \(x = -L_\infty\), or \(\tilde{x} = -L_\infty c_s w \tau_{yx}/4c_p h \gamma = -w/4r_{\text{min}}\).

The proposed model for groove drainage was validated against macroscale measurements of how the wetted length of the grooves changes as a function of time. Fig. 1.3(a)-(b) show the measured drainage behavior at two different shear rates, and how the non-dimensional scales collapse both drainage trends towards the universal theoretical prediction. An important consequence of the analysis is that the steady-state length, \(L_\infty\), does not depend on the viscosity of the fluid in the groove, \(\mu_o\) (see Eq. (1.7)). Thus, \(\mu_o\) can be used as a design parameter in the construction of liquid-infused materials without influencing the oil retention properties. The viscosity independence was validated by repeating the above experiments with two different oils whose viscosities differ by an order of magnitude. Despite the different drainage rates between the two oils, the steady-state wetted lengths were roughly the same, as shown in Fig. 1.3(c)-(d).

Another significant design consequence is that the steady-state length is independent of the groove size. Though the groove aspect ratio enters the formula for \(L_\infty\), through \(c_p, c_s,\) and \(h/r_{\text{min}}\), the magnitude of the groove size is not important. Grooves
with the same aspect ratio should have identical steady-state lengths, regardless of whether the depth is 1 µm or 1 mm. However, when $w$ or $h$ falls into the nano-scale range, long-range forces may modify the retention behavior; conversely, if these dimensions or $\tau_{yx}$ become too large, the Reynolds number or Bond number may no longer be low enough for our analysis to remain valid.

Within the parameter range of our analysis, $L_\infty$ depends on only the surface tension, contact angle (through $r_{\text{min}}$), and groove aspect ratio, so that options for designing a surface to retain lubricant are limited. In most cases, the surface tension and the contact angle cannot be considered adjustable parameters because of the need to prevent the oil from “cloaking” sessile drops [3, 4, 39, 40]. Thus, the aspect ratio of
Figure 1.4: (a) Steady-state length, $L_\infty$, normalized by $\gamma/\tau_{yx}$, for varying groove aspect ratio, with low viscosity oil; $Q = 2$ mL/min (red), $Q = 1$ mL/min (blue), and $Q = 0.5$ mL/min (orange). Squares are for grooves with $h \approx 10\ \mu$m, and circles are for grooves with $h \approx 20\ \mu$m. The theoretical prediction from Eq. (1.7), $L_\infty\tau_{yx}/\gamma = 2c_p h/c_s\tau_{\text{min}}$ is given by the dashed line. (b) Groove resistance coefficients $c_p$ (green), $c_s$ (purple), and the ratio $c_p/c_s$ (black) as a function of aspect ratio $w/h$. Each curve asymptotes to the dashed line of the same color.

Figure 1.5: (a) Snapshots of a shear-driven drainage experiment on a substrate consisting of randomly placed posts. A script was written in MATLAB to define the uniformly random locations of the cubes on a 1-µm grid subject to two conditions: 1) that the area density of the posts is 25%, and 2) that the minimum space between posts is 3 µm (to aid with photolithography). The pattern is initially filled completely with low viscosity oil (green). The oil drains due to an external aqueous flow of $Q = 0.5$ mL/min. (b) Micrograph of the silicon wafer micro-pattern that is used to mold the posts (light gray), including a surface profile (purple).

the grooves, $w/h$, is the primary means of tuning oil retention, and the steady-state length in Eq. (1.7) depends strongly on this parameter.

To explore the dependence of drainage behavior on aspect ratio, surfaces with grooves of different width and different depth were fabricated and tested at three flow rates (see Table 1.1 for groove dimensions). The steady-state length, $L_\infty$, is plotted in Fig. 1.4(a) along with the theoretical prediction, where $L_\infty$ has been normalized
by $\gamma/\tau_{yx}$ in order to isolate effects of the aspect ratio. These results demonstrate that narrower and deeper grooves result in longer $L_{\infty}$. Fig. 1.4(b) shows how $c_p$ and $c_s$ vary as a function of $w/h$. Note that $2/3 < c_p/c_s < 1$, so that groove geometry affects $L_{\infty}$ in Fig. 1.4(a) primarily through $h/r_{\text{min}}$.

<table>
<thead>
<tr>
<th>$w$ (µm)</th>
<th>$h$ (µm)</th>
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</thead>
<tbody>
<tr>
<td>4.7 – 4.9</td>
<td>8.8</td>
</tr>
<tr>
<td>9.9 – 10.1</td>
<td>9.3</td>
</tr>
<tr>
<td>19.7 – 20.0</td>
<td>9.7</td>
</tr>
<tr>
<td>39.4 – 39.6</td>
<td>10.0</td>
</tr>
<tr>
<td>11.1 – 11.7</td>
<td>18.9</td>
</tr>
<tr>
<td>20.7 – 20.9</td>
<td>20.4</td>
</tr>
<tr>
<td>40.7 – 51.1</td>
<td>21.4</td>
</tr>
</tbody>
</table>

Table 1.1: Dimensions of grooves of different width and different depth. Grooves with similar depth were fabricated and tested in the same pattern.

1.4 Extensions and Conclusions

We noted earlier that longitudinal grooves provide a ‘reduced-order’ perspective on a variety of more complicated patterned surfaces, and thus we expect surfaces with a random pattern to follow a similar drainage behavior. To demonstrate the general nature of our findings, we repeated the drainage experiments on a surface with a micro-fabricated geometry consisting of randomly placed 10-µm cubes. The film drains from the random posts following a similar behavior: as with the grooves, a finite length of the pattern remains fully wetted (see Fig. 1.5 and Movie 2). The retention of fluid under shear is therefore not unique to well-controlled surface geometries, and may be expected on surfaces with industrially fabricated patterns that are inherently more random.

Our findings suggest a methodology for designing liquid-infused materials capable of retaining their lubricant up to a design-limited shear stress. We have shown that the value of this limiting shear can be tuned by manipulating the aspect ratio of the
surface pattern. Extrapolating further, the theory suggests a method by which any existing patterned surface may be made resistant to shear-driven drainage: we predict that oil can be retained indefinitely if surface features are interrupted by periodic barriers with a period less than or equal to $L_\infty$. For instance, if a grid of barriers with period $L_\infty$ is overlaid on a random rough pattern, the surface will be expected to retain its lubricant up to the shear value used to determine $L_\infty$. Such minimally structured geometries, designed according to the insights from this study, will allow for greater adoption of liquid-infused surfaces by enabling their use in applications where they would otherwise fail.
Chapter 2

Overflow cascades in liquid-infused substrates

The majority of this chapter has been submitted as a journal article of the same title \cite{41}. The research was performed in collaboration with Ian Jacobi and Howard Stone; Ian Jacobi led the majority of the research and wrote the majority of the paper, and is thus the first author of the article. I contributed the section on surfactant effects, as well as some experimental results.

2.1 Background

Liquid imbibition, spreading, and drainage from rough or patterned surfaces are important processes that appear in a variety of engineered and natural systems, including a new generation of liquid-infused omniphobic coatings \cite{42, 2, 3, 4}. The dynamics \cite{25, 29, 43} and stability \cite{32, 44, 45} of flows within patterned substrates have been studied extensively when the fluid flow in the complex substrate is subject to gravitational forcing. However, in many practical scenarios, such as using liquid-infused coatings for drag reduction \cite{9, 10}, the liquid imbibed within the patterned or rough...
surface is subject to an external flow of immiscible fluid, which exerts a shear stress on the fluid interface and can result in loss of the infused liquid.

In Chapter [1] we explored the mechanism of shear-driven loss of fluid and proposed substrate design criteria that can mitigate this loss. Because this study is important to the findings presented here, we provide a short summary of the main ideas. In the earlier experiments, a single longitudinal groove was filled with oil as a simple model for non-uniform but longitudinally-oriented, geometric features of patterned surfaces. The filled groove was subject to an external shear flow of an immiscible liquid and the infused liquid was allowed to drain freely into a reservoir of excess lubricant. For a fixed shear stress, $\tau$, interfacial tension $\gamma$, and groove cross-sectional shape, defined by width, $w$, and height, $h$, we observed that a finite length of lubricant, $L_\infty$, was retained in the groove at steady state, despite the external flow. The lubricant retention was caused by an adverse capillary pressure gradient in the lubricant phase, driven by interfacial deformation: as the groove began to drain due to the external shear stress, the interfacial radius of curvature at the upstream end of the groove, $r_{\text{min}}^u$, decreased, creating a pressure gradient, $\Delta p = \gamma/r_{\text{min}}^u$, between the upstream end and the downstream reservoir which suppressed further drainage.

By cross-sectionally averaging the flow field within the groove, it was shown that the net flux, $q_p$, of lubricant with viscosity $\mu$ in the upstream direction generated by this pressure gradient is given by

$$q_p = -c_p \frac{w h^3 \Delta p}{\mu L_\infty}, \quad (2.1)$$

where $c_p$ is an $O(1)$ parameter that represents the appropriate boundary conditions of the series solution of the governing Poisson equation within the groove. The calculation assumes that the steady interface shape is only a small perturbation of the original flat interface. This same approach was used to calculate the downstream flux of lubricant due to the action of the external shear flow, $q_s$, given by
where $c_s$ is another $O(1)$ geometric parameter depending only on the shape of the groove. The coefficients $c_p$ and $c_s$ are derived in Chapter 1 and for all streamwise groove geometries $2/3 \leq c_p/c_s \leq 1$. Balancing these two fluid fluxes, $q_s$ and $q_p$, at steady state yields the predicted length of retained fluid,

$$L_\infty = \left( \frac{c_ph}{c_s u_{\text{min}}} \right) \frac{\gamma}{\tau},$$

which was verified by experiment. Therefore, as long as the length, $L$, of the groove containing the oil is less than $L_\infty$, the oil within the groove will be retained indefinitely. The question raised by this analysis is what happens when the groove is longer than the maximum retention length, $L_\infty$?

In this study, we explore through experiments and modeling the overflow behavior of infused liquids when this retention criterion is violated. We consider a longitudinal groove filled with lubricant that terminates abruptly, without any connection to a downstream lubricant reservoir. When such a substrate is immersed in an immiscible shear flow, the basic fluid geometry of interest is a finite liquid rivulet (or ridge) with its base contained within the longitudinal groove and its interface exposed to the external flow. The central question above can then be expressed in terms of the finite fluid rivulet: what is the mechanism by which the finite, constrained rivulet overflows the ‘dead-end’ terminus of the groove due to the external shear flow?

The overflow behavior of a liquid rivulet in a dead-end geometry depends both on the chemistry and the geometry of the fluid-fluid interface. The geometrical problem of a liquid rivulet on a substrate has been studied extensively due to its ubiquity – the rivulets on car windshields during a rainstorm are a classic example of this fluid geometry. A static liquid ridge or rivulet of initially uniform cross-section on
a substrate exhibits an instability in which bulges form along the length of the ridge, known as a ‘pearling instability’. This instability is a variation of the classical Rayleigh-Plateau instability that includes the influence of the solid substrate. The stability of these rivulets has been analyzed by classical energetic and hydrodynamic stability methods [46], which established that for pinned contact lines, the rivulet is linearly stable to all perturbations if the contact angle of the rivulet fluid with the surface, $\theta < \pi/2$. The pearling instability has been observed when the contact lines of the rivulets were pinned by chemical discontinuities in the surface, e.g. hydrophilic stripes [47, 48, 49] and by topographic patterning of the substrate [30]. The pearling instability criterion was also extended to the scenario of an external flow parallel to the liquid ridge [50, 51]. Qualitative observations of deformations in rivulet shape for cases of pressure-driven flow within the rivulet itself have also been reported [52], and related instabilities have been used to generate microscale droplets from the rivulet terminus [53].

While the stability of liquid rivulets has been studied extensively from a geometrical perspective, the chemistry of the rivulet fluid interface (i.e. surfactants) has not received extensive treatment, except in a case without any external flow that involved the spreading of surfactants [54, 55]. However, the importance of surface chemistry in a multiphase system with flow has been explored previously in studies of bubble and droplet motion in quiescent fluids [56] and under vertical temperature gradients [57]. When a bubble or drop is moving in an immiscible fluid, shear from the external fluid advects any surfactant molecules present on the interface to the downstream end of the bubble or drop. The surface tension then becomes lower at the downstream end, generating a Marangoni stress opposing the direction of advection. In the simplest cases, the effect of the surface tension gradient on the finite surface of the droplet is manifested by an immobilized interface over the downstream end of the droplet and a clean, mobile interface on the upstream end [56]. Because most real systems
have some degree of impurities capable of modifying the interfacial tension\(^5^8\), we can expect a similar surface tension gradient to develop on the interface of the rivulet in most practical environments.

In this chapter, we explore how the geometric pearling instability and the chemical Marangoni stresses both contribute to the shear-driven fluid overflow predicted for dead-end, liquid-infused substrates. Also, we examine the subsequent behavior of those substrates after the fluid overflow generates droplets on the substrate surface, and we consider the conditions under which the droplets depin and slide downstream.

## 2.2 Overflow

### 2.2.1 Experiments

The worst case scenario for the retention of infused liquid on a substrate is when the substrate pattern is aligned with the external flow as longitudinal grooves, as was explored in Chapter 1; this configuration also happens to be the most productive orientation for purposes of drag reduction\(^5^9\). Following the approach outlined in Chapter 1 microfluidic flow cells 7 mm wide by 180 µm deep, and 45 mm long were assembled from Norland Optial Adhesive (NOA 81) using the ‘sticker’ technique\(^3^5\). The floor of the flow cells were patterned with a set of 50 parallel longitudinal grooves with width \(w = 9.0 \, \mu m\), depth \(d = 9.8 \, \mu m\) and length \(L = 36 \, mm\).

A schematic of the flow cell is included in figure 2.1. The outer fluid flowing through the flow cell was a 1:1 wt. mixture of glycerol and water, with viscosity \(\mu_w = 5.4 \, mPa \cdot s\), and was controlled by a syringe pump (Harvard PHD2000). The grooves themselves were pre-filled with an immiscible silicone oil (Gelest PDM 7040) with viscosity \(\mu_o = 42.7 \, mPa \cdot s\) that was dyed with a small amount of fluorescent dye in order to image the oil dynamics on fluorescent light and confocal microscopes. The interfacial tension was measured by the pendant drop method to be \(\gamma = 28.7 \, mN/m\).
Figure 2.1: (a) Cross-sectional schematic of the microfluidic flow cell used to expose the lubricant-infused grooves to a controlled shear stress. Aqueous solution is represented as blue and the infused lubricant/oil is represented as green. (b) Schematic of the downstream, ‘dead-end’ substrate microstructure of two neighboring grooves, with a flat interface in the absence of flow; the flow direction is noted for orientation.

A top view of some of the initially filled grooves is shown in figure 2.2(a). As the flow rate, \( Q \), of the external aqueous solution is increased, the outer flow exerts a shear stress, \( \tau \), on the interface of infused oil, which can be estimated assuming Poiseuille flow in the channel, where \( \tau = \frac{6 \mu w Q}{WH^2} \).

At sufficiently high shear stress, the oil within the grooves (downstream of the inlet) begins to bulge slightly, out of the uniform rivulet geometry, in a way reminiscent of the pearling instability, figure 2.2(b). The bulge then overflows the groove, figure 2.2(c), and eventually spreads to neighboring grooves, figure 2.2(d). The bulge and overflow nucleate just upstream of the dead-end terminus of the groove.

The overflow process can be understood mechanistically by viewing it in cross-section, at a streamwise position approximately 50 \( \mu \text{m} \) upstream of the grooves’ termini. Using a confocal microscope, cross-sectional images encompassing a number of
Figure 2.2: The sequence of images preceding an overflow event of a single longitudinal groove. Shown are streamwise/spanwise ($x$-$z$ plane, top-down) views of the microfluidic flow cell, captured by the confocal microscope and thresholded to produce two-color images where green represents the lubricating oil and black represents the substrate. External flow is from top to bottom; the grooves (9 $\mu$m wide) terminate near the bottom of the image frame. Note how the rivulet begins to bulge near its downstream end in (b); the contact line is depinned and the overflow begins to occur in (c); and the oil spreads to neighboring grooves in (d). The steady-state flow produces a shear stress of $\tau = 2.58$ Pa although these images were recorded during the pump transient and thus the instantaneous shear stress is less.

grooves and a fraction of the overall flow cell height were recorded at 1 s time intervals under an applied shear stress of $\tau = 2.58$ Pa. Initially, before the outer flow is turned on, the grooves are completely filled with oil and the oil/water interface appears flat, as shown in figure 2.3(a). As the outer flow rate increases, the oil/water interface begins to swell until, at some point, the longitudinal contact lines become unstable,
figure 2.3(b), and the oil overflows the groove and joins oil in neighboring grooves, figure 2.3(c). As time progresses, the droplets of oil coalesce on the substrate to form larger droplets until a significant fraction of the flow cell height and width is blocked by the growing droplets, figure 2.3(d).

2.2.2 Geometrical Overflow Criterion

The destabilization and overflow of the rivulets are qualitatively similar to the pearling instability that fluid rivulets are generally susceptible to on flat substrates when the instantaneous contact angle of the rivulet, $\theta$, exceeds $\pi/2$. In order to test the hypothesis that the pearling instability is the mechanism responsible for overflow of these liquid-infused surfaces, the stability criterion for the pearling instability can be used to predict the shear stress at which overflow is expected to occur, which can be compared with measurements.

In the case of liquid-infused grooves subject to an external shear flow, the relevant contact angle for the rivulet is measured from the inside, vertical wall of the groove. Therefore, the pearling instability in the groove should destabilize the contact lines on either side of the rivulet when $\theta = \pi$ at the downstream end of the rivulet, corresponding to a configuration of a rivulet on a flat substrate with a contact angle relative to the substrate of $\pi/2$. This instantaneous contact angle of the rivulet at the downstream end of the flow cell determines the Laplace pressure gradient within the rivulet, due to the gradient in the oil-water interfacial curvature between the bulging downstream end of the groove and the draining upstream end. Therefore, unlike the analysis in the reservoir-draining grooves discussed above, two different radii of curvature are relevant to the finite rivulet: the upstream radius where the drainage begins, $r_{\text{min}}^{u}$, and the downstream radius where the bulging occurs, $r_{\text{min}}^{d}$. The total pressure difference in the fluid rivulet is then given by
Figure 2.3: An experimental overflow event in cross-section. Shown are spanwise/wall-normal ($y$-$z$ plane) cross-sections of the microfluidic flow cell, transecting the initially filled longitudinal grooves, captured by the confocal microscope and thresholded where green represents lubricating oil, blue the aqueous external flow, and black the substrate. Flow is into the page and the grooves are 9 $\mu$m wide. At $t = 0$ s, the pump is started and the flow begins to accelerate, producing a shear stress at the water-oil interface at the surface of the grooves. The steady-state flow produces a shear stress of $\tau = 2.58$ Pa. Note how quickly the overflow transition occurs between (b) and (c).

$$\Delta p = \gamma (1/r_{\text{min}}^u - 1/r_{\text{min}}^d).$$  \hspace{1cm} (2.4)
The radii of curvature are related geometrically to the size of the groove and the effective contact angle of the interface, $\theta$, at each streamwise location, according to

$$r_{\text{min}} = \frac{w}{2\cos\theta},$$

(2.5)

where the upstream radius of curvature, $r_{\text{min}}^u$ is associated with the receding contact angle $\theta_{\text{rec}}$ and the downstream radius of curvature $r_{\text{min}}^d$ is associated with the instantaneous contact angle of the bulging rivulet at the downstream end of the groove. For the upstream radius of curvature, we assume that $w/h \leq 2(\sec\theta_{\text{rec}} + \tan\theta_{\text{rec}})$, which ensures that the interface does not contact the bottom of the groove.

Using this pressure difference (equations (2.4) and (2.5)), we can rewrite the flux balance between the flow in the groove driven downstream by the applied shear flow (equation (2.2)) and that driven upstream by the Laplace pressure gradient (equation (2.1)), in order to relate the pearling instability criterion to the applied shear over the liquid-infused surface. For the dead-end groove, this balance yields

$$\tau = \frac{2c_p h \gamma}{c_s L w} \left( \cos\theta_{\text{rec}} - \frac{w}{2r_{\text{min}}^d} \right).$$

(2.6)

At the onset of instability, the instantaneous contact angle at the downstream end is $\theta = \pi$ and using equation (2.5), we can then write the critical shear stress for the onset of the instability directly in terms of the receding upstream contact angle as

$$\tau_{\text{crit}} = \frac{2c_p h \gamma}{c_s L w} (\cos\theta_{\text{rec}} + 1).$$

(2.7)

Equation (2.7) provides an experimental means of verifying the hypothesis that the pearling instability is the mechanism of overflow by comparing the observed shear stress for overflow with the prediction. Using the experimental parameters listed above and the receding contact angle, measured in-situ, listed in the first row of table 2.1, yields $\tau_{\text{crit}} = 2.35 \pm 0.05$ Pa. By adjusting the flowrate in the syringe pump,
different shear stresses can be applied experimentally in order to identify the critical shear stress at which the overflow actually occurs. The overflow can be observed through microscopic observations, as reported above, or by means of a macroscopic lens that allows simultaneous observation of the entire length of the groove, as shown in figure 2.4. Over repeated experiments, the observed overflow appears to occur between $\tau = 1.5 - 1.9$ Pa, which is the same order of magnitude as the geometrical prediction, but slightly smaller; a possible cause of this discrepancy is discussed in section 2.2.3.

However, we must consider another possible mechanism for the overflow. Instead of the pearling instability, the overflow may occur simply due to exceeding the maximum contact angle sustainable by the sharp corner, which is the sum of the advancing contact angle and the corner angle, $\theta_{\text{adv}} + \pi/2$, following Gibbs’s formulation [60, 61]. Because this maximum contact angle of $158 \pm 1^\circ$ is close to the value of $180^\circ$ for the pearling instability, it is difficult to conclude based on the shear measurements alone that the pearling instability is the cause. However, the observation that the overflow first exhibits a localized, transverse bulging, as shown in figure 2.2, makes the pearling instability appear to be the most likely explanation.

After overflow occurs, droplets form at the downstream of each individual groove and then these droplets coalesce with the overflowed oil from neighboring grooves to form even larger droplets, as shown in figure 2.3(c). These droplets are at least an order of magnitude larger than the individual grooves (see details in section 2.3) and thus are expected to produce a nearly zero surface curvature at the downstream end, just like the case of drainage into a reservoir. This curvature condition should, in principle, produce the same capillary pressure gradient to retain fluid in both the post-overflow, dead-end and the reservoir-end groove scenarios. However, the oil retention length, $L_\infty$, observed in dead-end experiments was much larger than that predicted by the previously derived theory that was based on the reservoir-end case. For example,
Figure 2.4: Overflow behavior observed using a macroscopic lens over the entire groove length, with external flow from left to right. The view is again top down (x-z plane). Oil fluoresces green, but appears white when the groove overflows into a droplet due to the higher intensity of the emitted light (image saturation). Note that (a) exhibits some drainage at the upstream end of the grooves, but no droplet generation at the downstream end, whereas (b) shows extensive droplet formation due to overflow. The predicted critical shear stress $\tau_{\text{crit}} = 2.35 \pm 0.05$ Pa and the discrepancy is discussed in section 2.2.3. Below each top-down image is a cross-sectional cartoon (x-y plane) illustrating the groove geometry and droplet location.

Table 2.1: Contact angle measurements

<table>
<thead>
<tr>
<th>Measurement type</th>
<th>$\theta_{\text{adv}}$</th>
<th>$\theta_{\text{rec}}$</th>
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<tbody>
<tr>
<td>Microscale (within the grooves under flow)</td>
<td>$52 \pm 2^\circ$</td>
<td>$68 \pm 1^\circ$</td>
</tr>
<tr>
<td>Macroscale (on the substrate without flow)</td>
<td>$21 \pm 2^\circ$</td>
<td>$21 \pm 2^\circ$</td>
</tr>
</tbody>
</table>

the experiment shown in figure 2.4(b) displayed $L_\infty \approx 22$ mm; the theory, on the other hand predicts $L_\infty \approx 14$ mm. In order to explain these discrepancies, it is necessary to take into account the effect of surface-active contaminants at the water-oil interface within the flow cell, in addition to purely geometric considerations.

2.2.3 Surfactant Effects in Dead-End Grooves

In the experiments of Chapter with oil-filled grooves draining into a static reservoir of oil, the oil retention was produced by an adverse capillary pressure gradient in the oil phase, which was able to balance the shear applied by the external aqueous phase. The pressure gradient was measured on a confocal microscope indirectly, by means of the deflection, $\delta$, of the fluid-fluid interface at the center of the grooves.
The downstream static reservoir enforced an effective zero-curvature interface at the downstream end of the streamwise grooves, and thus the interfacial deflection was close to zero. Moving upstream of the outlet, the deflection, $\delta$, was observed to increase linearly with distance, indicating a constant adverse pressure gradient within the oil phase due to increasing interfacial curvature.

In order to understand why the oil retention predictions for the dead-end grooves differ from the theory developed for the static reservoir end condition, we performed microscopic measurements of the interfacial deflection for the case of dead-end grooves, illustrated in figure 2.5. Figure 2.6 shows $\delta$ as a function of the streamwise coordinate, $x$, where it is clear that the variation of $\delta$ in the dead-end groove is no longer entirely linear. A linear region is offset from the end of the grooves by a stretch of fluid with an undeformed interface (for this experiment, from $x = 0$ mm to $x = -3$ mm). We will refer to the length of the undeformed region as $L_u^\infty$, and the length of the deformed region as $L_d^\infty$, with the total steady-state length $L_\infty = L_u^\infty + L_d^\infty$, as shown in figure 2.5.

The existence of an undeformed region of interface in the dead-end grooves corresponds to a lack of adverse capillary pressure gradient in the oil phase within that
Figure 2.6: Measured deflection at the center of the interface for a dead-end groove with applied external shear $\tau = 5.2$ Pa after overflow of the grooves has produced droplets at the terminus. The black line represents the theoretical prediction for a groove ending in a stagnant reservoir, based on the assumption of a clean interface as in Chapter 1. Note that the length of fluid retention in the dead-end groove case is significantly longer, and that the deflection profile is no longer linear.

region. But without an adverse pressure gradient driven by interfacial deformation, what explains the oil retention – and indeed even enhanced oil retention – within the dead-end groove? There must be another physical mechanism countering the external shear applied to the interface. We propose that the natural presence of surface-active molecules within the fluidic system produces a Marangoni stress that is capable of retaining oil even in the absence of interfacial deformation.

Real-world systems contain a variety of impurities, including amphiphilic molecules that adsorb to fluid-fluid interfaces and act to decrease the interfacial tension. We refer to these molecules as surfactants, for simplicity. These surfactant molecules can be rearranged, by self-induced or external flows, into a configuration where their concentration varies along the interface, thereby generating Marangoni stresses (figure 2.5). In a fluid-filled groove, if the surfactants are arranged such that there is a constant gradient in surface tension along the rivulet interface, the stress generated by this gradient can exactly cancel the stress imposed by an external flow, leading to zero motion at the interface. Given the materials used in our experiments, we hypothesize that surfactant contaminants leach from the epoxy that
is used to construct the microfluidic flow cell, and that these contaminants produce a Marangoni stress in the undeformed, downstream region.

Based on the hypothesis that naturally occurring surfactants are responsible for the undeformed region of the rivulet, we attempted to eliminate the surfactants from the experimental system. The flow cells were washed thoroughly using a variety of solvents, and substitute materials were also employed in the fabrication process, but in all cases a finite length of the rivulet remained undeformed. Indeed, there is strong evidence in the literature supporting the fact that removing trace surfactants from a fluid system can be exceedingly difficult[62].

However, if contaminating surfactants are unavoidable in our experiments, why is the undeformed region observed only in dead-end grooves and not in experiments where the grooves emptied into a reservoir? We reasoned that the finite dimension of the groove must be responsible for the accumulation of surfactant molecules at the interface of the rivulet; when a stagnant reservoir is available, the surfactants are able to collect in the reservoir without generating a significant gradient over the rivulet interface.

In order to test this interdependence between the dead-end geometry and the interfacial chemistry, we performed another set of experiments in the flow cell with a reservoir as in Chapter 1, but with additional, ‘non-native’ surfactants intentionally added to the system, in an attempt to saturate the stagnant reservoir and thereby generate a surfactant gradient on the rivulet interface upstream of the reservoir. Experiments in the grooves with a static reservoir were conducted at identical flow rates, in one case with no added surfactant and another with 0.1 wt.% of Span-80 dissolved into the oil phase. The interfacial deflection, shown in Figure 2.7, indicates that the additional surfactant was capable of producing an undeformed region in the rivulet upstream of the reservoir, confirming that surfactants are capable of generating
oil retention in both reservoir-end and dead-end geometries even in the absence of capillary pressure gradients.

However, these effects are dependent on both the concentration and type of surfactant. Span-80 at a concentration of 0.01 wt.% showed negligible difference to the experiments that were performed with no additional surfactants, and 1 wt.% produced irregular dewetting behavior indicative perhaps of some interfacial instabilities. Experiments performed with the surfactants Triton X-100 and sodium dodecyl sulfate dissolved in the water phase produced interfacial instabilities as well and this remains a topic of ongoing work.

It is difficult to estimate the length of the undeformed region, $L_u^\infty$, (see figure 2.5) since this length depends on an unknown concentration of an unknown surfactant, both of which certainly depend on the particular operating environment of the liquid-infused surface. However we can provide an upper bound on the length of this region. We assume that at the upstream end of the wetted groove, where the interface is deflected, the surface tension is equal to that of the clean fluid-fluid interface, $\gamma$. At the downstream end of the groove, at $x = 0$, the surface tension must be greater than
zero. Therefore, over the entire undeformed length of the rivulet $L_u^\infty$, the surface tension varies by at most $\gamma$. The gradient of interfacial tension then is no greater than $\gamma/L_u^\infty$, and this gradient must exactly cancel the applied shear stress, $\tau$. With this physical picture, we have the inequality

$$L_u^\infty < \gamma/\tau. \quad (2.8)$$

We showed in Chapter 1 that $L_d^\infty$ is an $O(1)$ multiple of $\gamma/\tau$. For the experiments reported in figures 2.6 and 2.7, $\gamma/\tau = 5.6$ mm, which is greater than the measured $L_u^\infty$ for both cases. Given a more accurate estimate of the change in $\gamma$ over the length of the rivulet from specific knowledge of the operating environment, a more precise estimate of the Marangoni enhancement of oil retention can be made, resulting in a more accurate prediction of $L_\infty$.

In addition to explaining the enhanced oil retention in dead-end grooves, the surfactant effect can also explain the observation above that oil overflows from dead-end grooves at lower shear stresses than predicted by the purely geometric theory. The critical shear stress for overflow scales linearly with the interfacial tension, $\gamma$, so that as surfactant accumulation at the downstream end of the rivulet suppresses $\gamma$, the critical shear stress for overflow decreases.

An important design consequence of surfactant gradients in grooves is that surfactants can actually be used to enhance the oil retention in practical liquid-infused systems. The presence of naturally occurring surfactants of known concentration could be accounted for in the design of liquid-infused substrates, with the result of easing the geometric constraints on the design of the substrate assuming a surfactant-free system. These geometric design constraints could potentially be further eased by selecting substrate materials that naturally leach surfactant molecules over time,
thereby supplementing the natively occurring surfactants in the fluids. The precise behavior of surfactant emitting surfaces is a topic of current investigation.

2.3 Droplet Detachment

2.3.1 Qualitative Observations

Once the threshold shear stress is reached and the grooves overflow, forming droplets at the downstream end of the grooves (e.g. figure 2.4), a significant fraction of the oil initially in the grooves collects within the droplets. But in the absence of external flow, that oil could potentially re-infuse into the pattern, similar to imbibition processes on rough substrates [63, 64], and thus the oil is not necessarily ‘lost’ from the grooves when it is aggregated into the droplets. However, for high enough sustained external shear stress, the droplets of oil are depinned from the downstream end of the pattern and swept away by the external flow, after which the oil is permanently lost.

The shear stress at which the depinning occurs in the flow cell was determined by repeated experiments in which the external flow rate was increased via the syringe pump and the droplet behavior was again observed by means of a macroscopic lens. Figure 2.8 illustrates two different shear stresses that produce, for the lower stress, a droplet on the verge of depinning, but still connected to the grooves, and for the high stress, a train of depinned and sliding droplets.

Estimating the shear stress at the point of depinning is difficult due to size limitations of the flow cell itself. As the droplets grow at the downstream end of the grooves, their height can exceed half the flow cell height, and thus their projected area is a significant fraction of the overall external flow field. Therefore, we expect that the effective shear stress at the droplet interface is actually significantly larger than the value that can be calculated assuming the external flow is a standard, undeformed
Figure 2.8: Droplet depinning behavior observed over the entire groove length using a macroscopic lens, with external flow from left to right. The view is again top down (x-z plane). Green represents fluorescent dyed oil; white is the overflowed oil droplets (which saturate the image). Note that (a) shows a very large droplet (the white blob) pinned to the downstream end of the grooves, containing a significant fraction of the total oil initially in the grooves, whereas (b) exhibits two droplets that have depinned and disconnected from the grooves. Below each top-down image is a cross-sectional cartoon (x-y plane) illustrating the groove geometry and droplet location.

Poiseuille flow, as in the overflow analysis above. Nevertheless, taking the naive Poiseuille assumption yields a depinning shear stress in the range of $\tau = 6 - 7 \text{ Pa}$.

After the droplet depins and slides downstream along the substrate, the oil remaining in the grooves can overflow again, resulting in the formation of additional droplets. This cascade process can be seen in figure 2.8(b) where new droplets are visible at the downstream end of the grooves from which previous droplets have already depinned. On the microscopic scale, it is possible to note the stretching of each droplet as it depins and slides downstream, shown in figure 2.9, and the resulting cascade of oil loss from the grooves, as overflow and depinning events repeat. However, the critical shear stress required for overflow $\tau_{\text{crit}} \sim L^{-1}$ (equation (2.7)), so as the groove drains, the shear necessary for additional overflow events increases, until eventually a steady amount of fluid is left within the grooves for a fixed external shear stress.
Figure 2.9: A new overflow event after droplet depinning. Shown are streamwise/spanwise ($x$-$z$ plane, top-down) views of the microfluidic flow cell, captured by the confocal microscope and thresholded to produce two-color images where green represents the lubricating oil and black the substrate. Flow is from top to bottom; the grooves terminate near the middle of the image frame. Each individual groove is 9 µm wide.

### 2.3.2 Quantitative Modeling

Besides the practical difficulty in quantifying the depinning transition in a microfluidic system with macro-scale droplets, there are fundamental difficulties in estimating the shear stress necessary for depinning droplets. The traditional problem of dislodging a droplet from a no-slip substrate by the application of an external shear flow has been studied for three-dimensional droplets in certain asymptotic flow regimes \[65\] and for a general two-dimensional droplet \[66\]. Assuming the contact angles of the droplet on a surface are small, $\theta_{\text{rec}}, \theta_{\text{adv}} \ll 1$, the lubrication approximation can be applied to construct a force balance for the yield stress of a droplet. For the two-dimensional analysis, the yield stress for depinning is \[66\]
\[ \tau_{\text{depin}}^{2D} = \frac{\gamma}{R} \left( \frac{2}{27\pi} \right)^{1/2} \theta_{\text{adv}}^{3/2}(\theta_{\text{adv}} - \theta_{\text{rec}}), \]  

(2.9)

where \( R \) is a typical length scale associated with the droplet (here, the radius of the spherical equivalent volume of the droplet). For three-dimensional droplet displacement, the critical shear stress is given by [65]

\[ \tau_{\text{depin}}^{3D} = \frac{\gamma}{R} (0.28) \theta_{\text{adv}}^{4/3}(\theta_{\text{adv}} - \theta_{\text{rec}}). \]  

(2.10)

These depinning criteria involve two key assumptions: that the droplet is sitting on a solid, no-slip surface unlike the actual grooved, liquid-filled surface of direct interest here; and that the interface between the droplet and external flow is uncontaminated by surfactants, again unlike the system of interest, which is quite sensitive to the presence of surfactant molecules, as shown in the earlier experiments on overflow.

The grooved substrate geometry can be incorporated into the force balance by accounting for the increased surface area of substrate in contact with the oil, which, in the case of the evenly spaced longitudinal grooves, means a doubling in total wall shear stress per spanwise unit width of the flow cell. However, the surfactant effect is subtler as it affects the stress distribution and contact angle hysteresis over the length of the droplet.

In the discussion of the overflow criterion, the receding contact angle was measured \textit{in-situ} within the grooves themselves, using the confocal microscope, as reported in the first row of table [2.1]. That measurement was performed at the upstream end of the grooves, where surfactant concentrations are expected to be negligible. To get some sense of the effect of surfactants on the contact angles, the advancing and receding contact angles were measured macroscopically, on a sample of the substrate itself, as reported in the second row of table [2.1], where the presence of surfactant is not negligible, since there is no flow to wash the surfactant away from the measurement.
location, as in the microscopic measurement. In this macroscopic measurement, the receding contact angle was significantly smaller than the microscopic case. In the flow cell itself, we expect the receding contact angle of the droplet to be similarly affected by surfactants, since the length-scale of the droplet prior to depinning is similar to the length-scale over which the surfactant gradient was observed in the undeformed rivulet interface. Therefore, we expect there to be a significant gradient in surfactant concentration over the length of the droplet itself. Not only should the receding contact angle be affected by surfactants, but further downstream, the advancing contact angle should be even more strongly diminished due to the high surfactant concentration there. Such a gradient could also induce a Maragoni flow in the droplet itself.

The macroscopic measurements of the receding contact angle provide a sense of the uncertainty involved in estimating the contact angle hysteresis in this droplet system or similar experimental configurations. If we take the advancing contact angle as the value measured at the macroscale, in the presence of surfactants, \( \theta_{\text{adv}} = 68 \pm 1^\circ \), we can estimate that the receding contact angle should be in the range of the two receding measurements, one without surfactant effects measured \textit{in-situ} and the other including those effects, \( \theta_r = 21 - 52^\circ \). This produces a range of hysteresis values \( \Delta \theta = 16 - 47^\circ \). For a droplet half the width of the set of grooves, \( L_d = 0.50 \) mm, then the radius \( R \approx 0.10 - 0.16 \) mm. Using equations (2.9) and (2.10), the range for the critical shear stress for droplet dislocation is then between \( \tau_{\text{d depin}}^{2D} = 10 - 43 \) Pa and \( \tau_{\text{d depin}}^{3D} = 18 - 77 \) Pa, which spans nearly an order of magnitude in shear stress. Of course, given more detailed information about the precise surfactant concentration, it would be possible to provide more accurate values of the contact angle hysteresis, in addition to revising the actual force balance to take account of the Marangoni stress on the surface of the droplet. Despite the wide range of critical depinning stresses, all
of the values remain consistent with the lower bound estimate from our experiments of $\tau = 6 - 7$ Pa.

### 2.4 Conclusions

When liquid-infused substrates and open-microfluidic devices are subjected to an external shear stress, the fluid infused within the system can be lost through a cascade mechanism. By considering the optimal drag-reducing configuration of liquid-infused longitudinal grooves, two distinct stages of fluid loss were observed: an instability-driven overflow resulting in droplet formation, followed by droplet depinning and liquid loss. For a given substrate geometry, the critical shear stress at which overflow occurs was predicted (equation (2.7)) and verified by experiment. Similarly, the range for the critical shear stress at which the subsequent drop depins was also predicted (equations (2.9) and (2.10)) and tested experimentally, although the verification was more challenging. Both the overflow and depinning mechanisms were found to depend strongly on the presence of naturally occurring surfactant molecules in the flow, which tend to encourage groove overflow at lower shear stress, but also tend to enhance fluid retention after the overflow occurs, and affect the shear stress at which droplets depin and are lost from the substrate. It was shown that the cascade of liquid loss is itself limited by the shear stress such that each ‘round’ of the cascade results in increased resistance to further overflow. Understanding these critical levels of shear stress and the nature of this cascade will enable more robust design of liquid-infused and open-microfluidic devices.
Chapter 3

Robust liquid-infused surfaces through patterned wettability

The majority of this chapter has been submitted as a journal article of the same title [67]. The research was performed in collaboration with Abigail Grosskopf, Melissa Chow, Yuyang Fan, Ian Jacobi, and Howard Stone; they are all coauthors of the article.

3.1 Introduction

Liquid-infused surfaces have received much attention in recent years due to their protective [2, 3, 5, 4] and drag-reducing properties [8, 9, 10]. A major benefit of these surfaces is that they resist the pressure-induced failure mode associated with conventional gas-cushioned superhydrophobic surfaces, since the oil that is infused into the substrate is essentially incompressible and immiscible with the external phase. However, if a liquid-infused surface is oriented vertically, liquid that is initially located above the capillary rise height, $L_\infty$, will drain due to gravity [24, 27]. Moreover, even in the absence of gravity, liquid-infused surfaces are subject to failure; in Chapters 1 and 2 we show that shear stress from an external flow can drain an infused lubricating
liquid from a patterned or rough surface. As with drainage due to gravity, a finite length of the surface, $L_{s\infty}$, remains wetted when exposed to a given shear stress. This steady-state length is analogous to the capillary rise height, $L_{g\infty}$. However, just as with a vertical surface, if the infused region of a surface is longer than the retainable length, $L_{s\infty}$, the liquid that is upstream of the stable region will drain.

In both cases, the infused liquid is retained within the stable region by a balance between the force that causes drainage, be it gravity or shear stress, and a capillary pressure gradient within the infused liquid that develops due to deformation of the fluid-fluid interface. Since the pressure gradient is inversely proportional to the wetted length, surfaces that are too long will have a pressure gradient that is too weak to counterbalance the draining force, and will drain until the wetted length reaches $L_{s\infty}$ or $L_{g\infty}$, and the capillary-induced pressure gradient becomes high enough to resist further drainage. To create stable surfaces that are longer than $L_{s\infty}$ or $L_{g\infty}$, the texture must be interrupted by physical barriers with a period less than $L_{s\infty}$ or $L_{g\infty}$. The barriers create patches of disconnected texture that, independently, are able to resist the draining force. For applications with precisely fabricated textures, this design criterion represents a promising method for increasing the robustness of liquid-infused surfaces. However, when producing surfaces at an industrial scale, it may be prohibitively expensive to fabricate the structures needed to prevent the infused liquid from draining. Indeed, with conventional superhydrophobic surfaces, the need to produce precise micro- or nanoscale surface features that raise the threshold for pressure-driven failure has in the past been a major impediment towards widespread implementation of these surfaces.

We propose that the infused liquid can be retained, with considerably less expense, by instead patterning regions with differing wettability on a textured surface. Surfaces with patterned wettability have been shown to be a powerful tool for manipulating droplets, rivulets, and multiphase flows and thus there is reason to expect that
patterned wettability on textured surfaces can be used to influence the behavior of an infused film. In an early example of patterning wettability on flat surfaces, a chemical gradient of wettability was used to move droplets against the direction of gravity [69]. Since then, advances in lithographic techniques have made it possible to create sharply defined regions of differing wettability. These sharp regions constrain static rivulets to shapes that would otherwise be unstable [47, 70]; hence, stripes of preferential wettability have been used to guide rivulet flows in open microfluidic applications. For these chemically-patterned devices the flow is either imposed by a pump [71, 52] or, more frequently, driven by an external force such as a surface tension gradient [72, 73, 74], an applied shear stress [51], or gravity [50, 53].

In a more limited set of cases, wettability patterning has been combined with structural patterning for added functionality. For example, superhydrophobic surfaces demonstrate improved condensation performance if the top surface of the texture elements is made hydrophilic [75]. Similar patterning allows for precise control of ice nucleation [76], and in recent experiments, wettability patterning was used to control fluid invasion in liquid-infused textured surfaces [77]. In all of these cases, however, the wettability patterning has been used to add additional functionality to textured surfaces, but the idea of using such patterning to enhance the fundamental robustness of the surfaces has not been explored.

We propose that chemical patterning can be used to prevent drainage of liquid films that are infused in textured surfaces. Rather than relying on physical barriers to prevent liquid from draining under the influence of shear stress or gravity, we pattern thin stripes of a differing chemistry on the sample, separated by a distance less than the stable capillary length, $L^\infty_\infty$ or $L^g_\infty$. The chemistry of the stripes is such that they are preferentially wetted by the external phase, be it air, water, or another fluid that is immiscible with the lubricating liquid. This chemical patterning induces the infused liquid to dewet from select regions, thereby limiting the length of
continuous wetted regions to be less than the characteristic length-scale for drainage. By inducing sacrificial regions of the surface to dewet intentionally, the remainder of the surface may be made more robust. This design concept is analogous to that of a firebreak, whereby thin strips of a forest are burned intentionally in order to prevent the remainder of a forest from burning.

Patterning wettability can be accomplished through a variety of low-cost scalable techniques that are adaptable to numerous immiscible fluid systems. In this chapter, we demonstrate that substrates designed to be infused with either an oily liquid or an aqueous liquid can be made resistant to either shear- or gravity-driven drainage through patterned wettability. To retain infused oil against shear, we expose select regions of a hydrophobic epoxy surface to deep-UV light through a photomask, rendering these regions hydrophilic \cite{78}. To retain an infused aqueous solution against gravitational drainage, we spray-coat select regions of a hydrophilic acrylic surface through a stencil to achieve a desired hydrophobic pattern. The treated and untreated contact angles for both surfaces are shown in Table 3.1, indicating that strong contrast in wettability was achieved with these techniques.

### 3.2 Protecting against shear-driven drainage

We start with a set of experiments to explore how patterned wettability can be used to prevent drainage of a liquid-infused substrate due to shear. A microfluidic flow

<table>
<thead>
<tr>
<th>substrate</th>
<th>infused</th>
<th>external</th>
<th>$\theta_{\text{adv}}$</th>
<th>$\theta_{\text{rec}}$</th>
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</thead>
<tbody>
<tr>
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<tr>
<td>acrylic</td>
<td>aqueous</td>
<td>air</td>
<td>$114 \pm 2^\circ$</td>
<td>$72 \pm 2^\circ$</td>
</tr>
</tbody>
</table>

Table 3.1: Measured wetting properties between the substrate and the infusing liquid for shear- and gravity-driven drainage experiments. Shaded rows are for the substrates that were treated to be non-wetting by the infused liquid and unshaded rows are for untreated substrates.
Figure 3.1: (a) Schematic showing the microfluidic flow cell used to test shear-driven drainage of a liquid-infused substrate. (b) Closeup of the surface texture showing a sacrificial region. Oil is colored green and the aqueous solution is colored blue. Hydrophilic regions are denoted with orange stripes.

The flow cell is constructed with a texture imprinted on its floor, as in Fig. 3.1. This flow cell geometry is similar to that used for the shear-driven drainage experiments discussed in Chapters 1 and 2. A surface texture consisting of grooves running in the streamwise direction was chosen for the experiments, because this reduced-order geometry is conducive to theoretical understanding but still captures the characteristic behavior of more complicated geometries.

The flow cell is constructed from Norland Optical Adhesive, a UV-cured epoxy, following the microfluidic sticker technique [35], with a depth \( H = 180 \, \mu m \), a width \( W = 7 \, \text{mm} \), and a length of 45 mm. The grooves have a width \( w = 8.8 - 9.2 \, \mu m \), a depth \( h = 10.0 \, \mu m \), and are 35 mm long (Fig. 3.1). There are fifty grooves in
Figure 3.2: Planform view of shear-driven drainage experiments conducted on surfaces with fifty streamwise grooves. Oil fluoresces green, and the drained portions of the texture reflect the blue light used for excitation. White blobs indicate the presence of overflowing oil droplets. Each image set in (a)-(d) shows the initial state (top) and the steady-state (bottom) that develops due to the designated shear stress. (a)-(b) Experiments on treated samples; hydrophilic regions are denoted in the initial state images with orange stripes. (c)-(d) Experiments on untreated samples. The scale bar in (d) applies to all images.

the texture, each separated by walls of width 11.8 – 12.2 \( \mu \)m, for a total width of 1 mm. The pattern is positioned near the spanwise and streamwise center of the flow cell. Since the flow cell is very wide, with an aspect ratio of 40:1, and is much deeper than the pattern, the flow profile is approximately uniform through the width of the flow cell and parabolic through its depth. The infused liquid is silicone oil (Gelest PDM-7040, viscosity \( \mu_o = 43 \text{ mPa}\cdot\text{s} \)) mixed with 0.2 vol.% fluorescent dye (Tracer Products, TP-3400), and the outer aqueous liquid is a 1:1 weight mixture of glycerol and water (\( \mu_{aq} = 5.2 \text{ mPa}\cdot\text{s} \)). The surface tension between the two phases is \( \gamma = 29 \text{ mN/m} \). The outer fluid, pumped with a syringe pump at a flow rate \( Q \), imposes a shear stress of approximately \( \tau = 6\mu_{aq}Q/WH^2 \) on the texture.

Norland Optical Adhesive is naturally slightly hydrophobic, so the surface must be modified in select regions to create the desired sacrificial regions of hydrophilicity.
We use a method\cite{78} that relies on deep-UV exposure to modify the surface chemistry and make the epoxy hydrophilic. The hydrophilic regions can be defined precisely through the use of a photomask. To create a photomask with micron-scale pattern geometry, a 100 nm layer of chromium is sputtered onto a bare quartz wafer; quartz is transparent in the deep-UV range. A 500 nm layer of photoresist is then spin-coated onto the quartz, before being selectively exposed and developed. After, the wafer is etched with a chromium etchant, so that unprotected regions of the chromium are dissolved. Then, the hardened photoresist is removed, leaving only the chromium photomask bonded to the quartz wafer.

The microfluidic flow cells are molded in two separate halves – an upper half with the flow cell geometry and a lower half with the grooves – before being bonded together to create a closed flow cell. Before the two sides are sealed, but after the epoxy is cured, the grooved side of the flow cell is exposed for 30 minutes under the photomask in a deep-UV lamp (Jelight bulb, intensity $\approx 30$ mW/cm$^2$ at 253.7 nm). For the experiments presented here, the mask has transparent stripes that are 3 mm long and 250 $\mu$m wide, running across the texture with a streamwise period of 8 mm. Thus the hydrophobic untreated regions have an approximate length of $L = 7.75$ mm. The mask is elevated $100 - 200$ $\mu$m above the surface of the epoxy during the patterning step, because the mask was found to damage the surface texture if it contacted the epoxy. This offset resulted in hydrophilic regions with diffuse boundaries, as well as minor variations in the geometry of the hydrophilic regions due to a non-uniform gap height. After the hydrophilic treatment the two sides of the flow cell are bonded together and the flow cells are left in an oven at 70$^\circ$C overnight prior to being used.

To image the drainage dynamics over the length of the entire texture, the flow cells are mounted upside-down under a Nikon D90 camera outfitted with a 200 mm f/4 macro lens. The flow cells are illuminated with 470 nm blue LEDs that cause the
oil to fluoresce. Since the fluorescence from the oil is rather weak, a number of steps are taken to enhance the image quality: a photographic filter is mounted on the lens to block the excitation light (Wratten 12 transmission spectrum), the flow cells are molded with black glass on the back side to block background light, and black fabric is wrapped around the whole setup to block light from the room.

The threshold for initial drainage can be estimated from measured properties of the substrate-fluid system though the theory derived in Chapter 2 that balances the external shear stress, \( \tau \), against capillary stresses (indicated by the surface tension, \( \gamma \)). We express the stability criterion as an inequality involving geometric and fluid properties of the substrate,

\[
L < \frac{2c_p h \gamma}{c_s w \tau} \left( 1 + \frac{w}{2r_{\text{min}}} \right),
\]

where \( L \) is the length of continuous wetted regions. The parameters \( c_p \) and \( c_s \) are dimensionless functions of the groove aspect ratio, \( w/h \), and \( r_{\text{min}}^{u} \) is a length-scale that is a function of \( w, h, \) and the receding contact angle, \( \theta_{\text{rec}} \), as in Chapters 1 and 2. If inequality (3.1) is satisfied, the infused liquid should be stable; otherwise, it should drain.

Based on \( L = 7.75 \text{ mm} \) in our experiments, and the other measured parameters of the system, we calculate that at a shear stress of \( \tau = 12.2 \text{ Pa} \) the grooves should start to drain. However, as explained in Chapter 2 there is some amount of variability in the measured contact angles for this experimental setup, and the transitional value could be significantly lower.

At the beginning of an experiment, the entire flow cell is first filled with silicone oil. The silicone oil invades the texture, including regions that are treated to be hydrophilic, because these regions are also lipophilic in the presence of air. Then, the external aqueous solution is slowly pumped into the device at 5 \( \mu \text{L/min} \), corre-
sponding to a calculated shear stress of $\tau = 1.2 \times 10^{-3}$ Pa. The aqueous solution displaces the oil from the main portion of the channel, leaving the oil trapped in the unexposed hydrophobic regions of the texture. As the aqueous-oil interface reaches an exposed hydrophilic region, however, the aqueous solution displaces the oil from the hydrophilic region and preferentially wets this region. Thus, regions of the texture that are located between hydrophilic regions are disconnected from one another. This wetting state is shown in the top images of Fig. 3.2a-b, including orange stripes that indicate the exposed hydrophilic regions.

After the main portion of the flow cell has been cleared of its initial oil, the flow rate of the aqueous solution is increased rapidly to a much higher level. Experiments were performed at shear stresses of $\tau = 4.8$ and $19.0$ Pa, which are below and above the estimated critical value of $12.2$ Pa. Steady-state images from these two experiments are shown in Fig. 3.2a-b. On a surface with patterned wettability, the oil resists drainage entirely when exposed to the lower shear stress. At the higher shear stress, roughly half of each hydrophobic region is drained of oil. Control cases are shown in Fig. 3.2c-d, where untreated textures are exposed to the same shear stress values. In both cases, a significant portion of the texture is drained entirely of oil, indicating that our proposed method successfully prevents shear driven-drainage below a design-limited shear stress threshold, eqn (3.1).

A surprising feature is that the resulting wetted length in Fig. 3.2d is less than the individual wetted lengths in Fig. 3.2b. If the chemical barriers functioned exactly the same as physical barriers, we would expect these lengths to be equal. Examining the interface shape on a confocal microscope reveals that the advancing and receding contact angles in the hydrophobic regions of the treated texture are modified from those of the untreated texture, and exactly compensate for the increased wetted lengths, if these measured contact angle values are used with the theories presented in Chapters 1 and 2. It is unclear why the contact angle in the unexposed region
would be modified; the deep-UV treatment converts oxygen in the atmosphere to ozone, so we speculate that the ozone generated in the exposed regions diffuses into the unexposed regions and interacts with the substrate there.

Various other deep-UV treatment schemes were also tested. Experiments were performed on textures that were exposed for 15 minutes and 60 minutes. The 60 minute exposure produced similar results to those shown in Fig. 3.2a-b, but the 15 minute exposure resulted in only partial oil retention. The widths of the transparent stripes on the mask were also varied. When the widths were increased to 1 mm, the method worked as before. For widths of 50 μm, however, the treatment resulted in only partial oil retention, even for a 60 minute exposure time. We hypothesize that the reason for this failure is that the stripe width was significantly less than the gap between the mask and the sample, so that the UV light that reached the sample was too diffuse. This issue could potentially be resolved if a columnated light source were used instead. Alternatively, use of a photomask could be avoided entirely if a laser with a narrow beam was scanned across the sample to produce the surface treatment.

3.3 Protecting against gravity-driven drainage

To demonstrate the general nature of patterned wettability as a technique to retain fluids on textured surfaces, we present a second set of experiments to test gravity-driven rather than shear-driven drainage, see Fig. 3.3. As a further test of the retainment mechanism, we choose to design the substrate to retain an aqueous solution in the presence of air, rather than an oil in the presence of an aqueous solution. Also, an entirely different technique is utilized to achieve the contrasting chemistry: hydrophilic acrylic plastic is spray-coated through a stencil with a hydrophobic spray, resulting in regions with differing wettability. Though the drainage mechanisms, fluid systems, and surface treatments are different between these two sets of experiments,
Figure 3.3: (a) Schematic showing the vertical texture used to test gravity-driven drainage of a liquid-infused substrate. (b) Closeup of the surface texture showing a sacrificial hydrophobic region. Imbibed aqueous solution is colored green, and hydrophobic regions are denoted with orange stripes.

The underlying physics is analogous, and thus we expect the two sets of experiments to behave similarly.

Grooves were again used as the test geometry for the reasons stated previously, but their size was scaled up by more than an order of magnitude to demonstrate the range of length-scales at which this technique can be employed. The grooves were milled to have width $w = 0.51$ mm and depth $h = 0.51$ mm, running along the entire length of a 100 mm ×150 mm piece of black acrylic. The infused liquid is a 5:1 weight mixture of glycerol and water, mixed with 0.1 wt.% fluorescein sodium salt ($\rho_{aq} = 1.125$ g/mL, $\mu_{aq} = 52$ mPa·s, $\gamma = 66$ mN/m).

Carrying through the analogy with shear-driven drainage, we estimate the threshold for initial gravity-driven drainage of a liquid in a groove. Again, the stability criteria is expressed as an inequality,

$$L < \frac{2\gamma}{\rho_{aq}gw} \left(1 + \frac{w}{2r_{\text{min}}} \right),$$

(3.2)
where $L$ is again the length of continuous wetting regions. The parameter $r_{\text{min}}^u$ is a length-scale that is a function on $w$, $h$, and $\theta_{\text{rec}}$, as defined in Chapters 1 and 2. If inequality (3.2) is satisfied the liquid in the groove should be stable against gravitational drainage; otherwise, it should drain.

The stability criterion in Section 3.2 was tested by varying the magnitude of the forcing, $\tau$, such that eqn (3.1) was either satisfied or not. Rather than varying the magnitude of $g$ (which could effectively be accomplished by adjusting the tilt-angle of the substrate), we test two different values of $L$ that are above and below the critical value given by eqn (3.2). Substituting the measured parameters into eqn (3.2), we arrive at a threshold of $L = 46$ mm, so we test hydrophilic lengths of $L = 25$ mm and $L = 50$ mm.

To create the desired pattern of hydrophilicity and hydrophobicity, the machined acrylic was first thoroughly cleaned using isopropyl alcohol, followed by rinsing with water. The texture was then treated in an oxygen plasma chamber for 10 minutes to make it hydrophilic. To create the hydrophobic design on the texture, 1-mm thick
glass microscope slides were placed over the acrylic to act as a stencil, and the glass-acrylic sandwich was spray-coated with Penguin Water & Stain Repellent. The glass microscope slides were placed such that they covered either 25-mm or 50-mm long sections of the groove, with a 2-mm exposed gap between the sections – the acrylic beneath the gap would then become hydrophobic. In order to ensure that the sprayed regions are coated uniformly, and to prevent leakage of the hydrophobic spray under the glass, the acrylic was lightly sprayed repeatedly (approximately 15 times) with 10 second intervals between each spray, which allowed for the spray to evaporate between repetitions and prevented pooling. The glass slides were then removed and the treated acrylic was allowed to dry in an oven at 70°C for 30 minutes.

The acrylic substrate was mounted on a stand that allowed it to be quickly rotated between a horizontal and a vertical position. While the acrylic was in the horizontal position, 1 mL of aqueous glycerol solution was distributed across one end of the texture using a syringe. The solution was then pushed along the grooves using a squeegee at approximately 20 mm/s to allow the grooves to fill with liquid. The solution only filled the hydrophilic sections of the grooves and was repelled by the hydrophobic sections. Occasionally the solution did not initially enter the hydrophilic sections, and the process was repeated until all hydrophilic sections of the grooves were filled. Drainage was initiated by quickly flipping the acrylic to the vertical position. The setup was imaged using the same macroscale fluorescent imaging as before.

Results from gravity-driven drainage experiments are shown in Fig. 3.4. The top images in Fig. 3.4a-b show the configuration for the treated substrates immediately after the acrylic is tilted vertically, with the hydrophobic regions designated using orange stripes. The hydrophilic regions in Fig. 3.4a have a length $L = 25$ mm, which is less than the critical value, and the hydrophilic regions in Fig. 3.4b have an unstable length $L = 50$ mm. Note that the groove in Fig. 3.4b has already started draining by
the time the first image is recorded. The bottom images of 3.4a-b show the wetting configuration after the aqueous solution has reached a steady-state configuration. An untreated control case is included in Fig. 3.4c.

It is apparent that the hydrophobic stripes succeeded in preventing the infused aqueous solution from draining from the texture. The final state in Fig. 3.4c shows that most of the liquid has drained from the untreated groove, whereas the final state in Fig. 3.4a shows negligible drainage from the properly spaced regions. In Fig. 3.4b, which was treated to have an unstable length, \( L \), the infused liquid has partially drained. This wetting state is very similar to the partially drained state observed in Fig. 3.2b, further confirming the analogy between the two sets of experiments.

### 3.4 Conclusions

Drainage from textured surfaces can be prevented by creating sacrificial regions with differing chemistry. By exploring two different drainage mechanism (shear and gravity), with two different fluid chemistries (oil/aqueous and aqueous/air), at two different length scales (10 \( \mu \)m and 500 \( \mu \)m), we have demonstrated how the sacrificial patterning technique can be broadly applied to retain liquid in a variety of liquid-infused surfaces. Both techniques for applying the surface treatment were facile and easily scalable, showing the advantage of using chemical patterning for liquid retention. In addition, a wide variety of other chemical treatment techniques potentially could be used, including conventional photolithography [77], UV-based monolayer modification [79], and laser-based processing [80]. By using this strategy of chemical patterning at length-scales comparable to the length-scale suggested by the shear-and gravity-driven drainage criteria, we have demonstrated that these two formerly limiting failure mechanisms of liquid-infused surfaces can be eliminated through min-
imal surface processing. Thus liquid-infused surfaces may be used in a variety of applications that were previously inaccessible, at only minimal additional expense.
Chapter 4

Thin film velocimetry using FRAP

This chapter has not been submitted for publication as a journal article. The research was performed in collaboration with Ian Jacobi and Howard Stone, and was initially inspired by the research that is presented in the previous chapters.

4.1 Introduction

The lubrication equation, which governs the flow in a thin film, admits only linear and parabolic velocity profiles through its depth. Shear stress on the top surface of the film, $\tau_{yx}$, can cause the linear component, and a pressure gradient within the film, $\frac{dp}{dx}$, can cause the parabolic component. Provided that the lower surface of the film satisfies the no-slip condition, the flow in a two-dimensional thin film can thus be uniquely characterized by three independent local parameters,

1. Film thickness, $h(x)$,
2. Applied shear stress at the surface, $\tau_{yx}(x)$,
3. Pressure gradient in the film, $\frac{dp}{dx}(x)$,

in addition to the viscosity of the film, $\mu$. 
Therefore, when studying the flow in a thin film it may not be necessary to perform standard velocimetry techniques such as particle image velocimetry or laser Doppler velocimetry, since the velocity profile is so simple. We propose that it may be possible to fully characterize a thin film flow through the three quantities listed above, by monitoring the diffusion of a spot of solute in the flowing film.

To create a precisely defined spot, we propose using Fluorescence Recovery After Photobleaching (FRAP), a technique that was originally developed for microbiology. To perform FRAP in a thin film, a high-intensity laser is used to bleach a spot in a dyed film. By monitoring the advection of the center of the spot and the diffusive recovery of the dye to the spot, one can determine the shear stress applied to the film, and the pressure gradient within the film. In the analysis below, we first predict how a line of solute will behave in a thin film, and then generalize this behavior to a spot before discussing the details of experimental implementation.

### 4.2 Thin Film Flow

We consider a thin film of liquid with viscosity \( \mu \) flowing on a solid surface, as shown in Fig. 4.1. Restricting our analysis to a region of the film with uniform depth, \( h \),
and a unidirectional velocity field that varies only through the depth of the film, we choose a local coordinate axis with $x$ in the streamwise direction, $y$ in the positive normal direction from the solid surface, and $z$ in the direction transverse to flow, so that the flow profile within the film is $u = u(y)$. A shear stress $\tau_{yx}$ is applied to the top surface of the liquid, and the fluid particles adjacent to the solid flow satisfy the zero-slip condition. Thus, for boundary conditions we have

$$u = 0 \quad \text{at } y = 0 \quad (4.1)$$
$$\frac{\partial u}{\partial y} = \frac{\tau_{yx}}{\mu} \quad \text{at } y = h. \quad (4.2)$$

We consider a thin film flow with characteristic velocity, $U$, and characteristic length-scale in the flow direction, $\ell$. Provided that the effective Reynolds number within the film is small, $\rho U h^2 / \mu \ell \ll 1$, and the flow aspect ratio is small, $h/\ell \ll 1$, flow within the thin film satisfies the lubrication equation, $0 = -\frac{dp}{dx} + \mu \frac{\partial^2 u}{\partial y^2}$. Integrating twice in $y$ yields a generic equation for the velocity profile satisfying the boundary conditions in (4.1),

$$u(y) = \frac{1}{2\mu} \frac{dp}{dx} y(y - 2h) + \frac{\tau_{yx}}{\mu} y.$$

From this expression we also compute the depth-averaged velocity, which we express in terms of the depth-averaged pressure-driven velocity, $u_p$, and the depth-averaged shear-driven velocity, $u_s$,

$$\langle u \rangle = -\frac{h^2}{3\mu} \frac{dp}{dx} + \frac{\tau_{yx} h}{2\mu} = u_p + u_s, \quad (4.3)$$

We will also need an expression for the deviation from the depth-averaged velocity, $u'(y) = u(y) - \langle u \rangle$,

$$u'(y) = -\frac{3u_p}{2} \left( \left( \frac{y}{h} \right)^2 - 2 \left( \frac{y}{h} \right) + \frac{2}{3} \right) + 2u_s \left( \frac{y}{h} - \frac{1}{2} \right).$$
4.3 A Diffusing Line

We start by analyzing the case where an infinite line of solute appears in the film at some time \( t = 0 \). The line runs in the spanwise \( z \)-direction and is initially distributed uniformly through the depth of the film. The solute diffuses with diffusively, \( D \), according to the two-dimensional advection-diffusion equation,

\[
\frac{\partial C}{\partial t} + u(y) \frac{\partial C}{\partial x} = D \left( \frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial y^2} \right).
\]

We transform to a coordinate system, \( \tilde{x} = x - \langle u \rangle t \) that advects with the average speed of the thin film. In this frame of reference, the advection-diffusion equation becomes

\[
\frac{DC}{Dt} + (u(y) - \langle u \rangle) \frac{\partial C}{\partial \tilde{x}} = D \left( \frac{\partial^2 C}{\partial \tilde{x}^2} + \frac{\partial^2 C}{\partial y^2} \right).
\]

Now we decompose the concentration, \( C \), into a depth-averaged term and a deviation, \( C(y) = \langle C \rangle + C'(y) \). At large Peclet number, \( Pe = \langle u \rangle h / D \), it can be shown that the advected average concentration satisfies the relationship \( \frac{\partial\langle C \rangle}{\partial t} = -\langle u \rangle \frac{\partial C'}{\partial x} \). Performing the usual manipulations, we arrive at

\[
\frac{D\langle C \rangle}{Dt} = D \left( 1 + \left( \frac{14u_s^2 + 21u_s u_p + 8u_p^2}{420} \right) \frac{h^2}{D^2} \right) \frac{\partial^2 \langle C \rangle}{\partial \tilde{x}^2}.
\]

This equation satisfies the two limits for pure shear flow and pure pressure-driven thin film flow [81]. Written in a stationary reference frame, the equation reads

\[
\frac{d\langle C \rangle}{dt} + \langle u \rangle \frac{d\langle C \rangle}{dx} = D_{\text{eff}} \frac{\partial^2 \langle C \rangle}{\partial x^2}, \quad (4.4)
\]

where the effective diffusion coefficient in the streamwise direction is

\[
D_{\text{eff}} = D \left( 1 + \left( \frac{14u_s^2 + 21u_s u_p + 8u_p^2}{420} \right) \frac{h^2}{D^2} \right). \quad (4.5)
\]
Alternatively, the effective diffusivity could have been computed directly using a formula that is valid for any rectilinear flow,

\[ D_{\text{eff}} = D + \frac{1}{D} \int_0^h \left[ \int_0^y (u(y') - \langle u \rangle) \, dy' \right]^2 \, dy. \]

4.4 A Diffusing Spot

If, instead of creating a line of solute perpendicular to the flow at time \( t = 0 \), we create a discreet spot in the film, the recovery process will proceed similarly in the \( x \)-direction, but less rapidly in the \( z \)-direction, since the molecular diffusivity alone will induce recovery in that direction. Thus, a spot that is initially circular will evolve to be elliptical, with the major axis of the ellipse aligned with the flow in the \( x \)-direction (Fig. 4.1). The \( y \)-averaged diffusion-advection equation in the \( xz \)-plane is thus

\[ \frac{\partial C}{\partial t} + \langle u \rangle \frac{\partial C}{\partial x} = D_{\text{eff}} \frac{\partial^2 C}{\partial x^2} + D \frac{\partial^2 C}{\partial z^2}. \]

Transforming again to a coordinate \( \tilde{x} \) that advects with the average film speed \( \langle u \rangle \), this equation becomes

\[ \frac{\partial C}{\partial t} = D_{\text{eff}} \frac{\partial^2 C}{\partial \tilde{x}^2} + D \frac{\partial^2 C}{\partial z^2}. \] (4.6)

We assume that the spot initially has a a Gaussian intensity profile in the \( \tilde{x}z \)-plane, with width, \( w \). The concentration is normalized so that \( C(\tilde{x} \to \infty, z \to \infty, t) = 1 \), and \( C(\tilde{x} = 0, z = 0, t = 0) = 0 \). The width is defined as the location at which the solute concentration drops to \( 1/e \). Thus, the initial solute concentration is

\[ C(\tilde{x}, z, t = 0) = 1 - \exp \left[ -\frac{\tilde{x}^2 + z^2}{w^2} \right]. \] (4.7)
Figure 4.2: The change in the aspect ratio of an ellipse with respect to time, \( r(\bar{t}) \), for \( D_{\text{eff}}/D = 2 \). The curve approaches the dashed line, \( r(\bar{t}) = \sqrt{D_{\text{eff}}/D} = \sqrt{2} \) as \( \bar{t} \to \infty \). The dotted line shows the initial behavior, \( r(\bar{t}) = 1 + \frac{D_{\text{eff}} - D}{2D \bar{t}} \).

The PDE (4.6), with initial condition (4.7) and boundary condition \( C(\bar{x} \to \pm \infty, z \to \pm \infty, t) = 1 \), admits the solution

\[
C(\bar{x}, z, t) = 1 - \frac{w^2}{[(4D_{\text{eff}} t + w^2)(4D t + w^2)]^{1/2}} \cdot \exp \left[ -\frac{\bar{x}^2}{4D_{\text{eff}} t + w^2} - \frac{\bar{y}^2}{4D t + w^2} \right],
\]

which is a modified version of the solution for a diffusing point-source. We can non-dimensionalize (4.8) with \( \bar{x} = \bar{x}/w \), \( \bar{z} = z/w \), and \( \bar{t} = t/(w^2/4D) \). Thus, (4.8) becomes

\[
C(\bar{x}, \bar{z}, \bar{t}) = 1 - \frac{1}{[(\bar{t} + 1)(\frac{D_{\text{eff}} \bar{t}}{D} + 1)]^{1/2}} \cdot \exp \left[ -\frac{\bar{x}^2}{\frac{D_{\text{eff}} \bar{t}}{D} + 1} - \frac{\bar{z}^2}{\bar{t} + 1} \right].
\]

(4.9)

Examining the expression in the exponent reveals that the shape of the spot evolves from a circular gaussian profile to an elliptical gaussian profile that is stretched in the \( x \)-direction. The ratio of the major to the minor axis is

\[
r(\bar{t}) = \sqrt{\frac{D_{\text{eff}} \bar{t}}{D} + 1},
\]

(4.10)
Figure 4.3: Fluorescence recovery at the advected center of the spot with respect to time, for $D_{\text{eff}}/D = 2$.

showing that the ellipticity becomes more apparent as time progresses. The ratio $r$ will eventually reach a steady-state value of precisely $\sqrt{D_{\text{eff}}/D}$, but by that point the diffused spot will have disappeared. The intensity at the center of the spot recovers according to

$$C(\bar{x} = 0, \bar{z} = 0, t) = 1 - \frac{1}{\left[(\bar{t} + 1)(\frac{D_{\text{eff}}}{D} \bar{t} + 1)\right]^{1/2}}. \quad (4.11)$$

These two curves are plotted in Figure 4.3, for $D_{\text{eff}}/D = 2$. Note that the rate of recovery is similar to the rate that the ellipticity grows. This presents a potential challenge with the experiment, since the spot will become harder to see as it becomes more elliptical, as is apparent in Figure 4.1. Perhaps it would be sufficient to measure the initial change towards ellipticity, rather than the steady-state value. The dimensionless slope as $t \to 0$ is given by $\frac{dr}{dt} = \frac{D_{\text{eff}}-D}{2D}$.

### 4.5 Connection to Experiments

The line or spot of solute could be generated by injecting a diffusible substance into the thin film at a specific time. However, it would be difficult to accomplish this without disrupting the flow of the film.
Figure 4.4: Normalized value of $u_p$, plotted against $D_{\text{eff}}/D$, for $\text{Pe} = \langle u \rangle h/D = \{0, 5, 10, 15, 25\}$. Increasing values of $\text{Pe}$ move further away from the $x$-axis. Eq. (4.12) is plotted as a solid line, and eq. (4.14) is plotted as a dashed line.

Figure 4.5: Normalized value of $u_s$, plotted against $D_{\text{eff}}/D$, for $\text{Pe} = \langle u \rangle h/D = \{0, 5, 10, 15, 25\}$. Increasing values of $\text{Pe}$ move further away from the $x$-axis. Eq. (4.13) is plotted as a solid line, and eq. (4.15) is plotted as a dashed line.
We propose instead that Fluorescence Recovery After Photobleaching (FRAP) could be used to generate the line or spot of solute. FRAP is a process that was originally developed to measure the diffusive properties of biological systems. To perform a FRAP experiment, fluorescent molecules are included in a system of interest. The molecules fluoresce under low intensity illumination, allowing their presence to be readily measured, but are bleached under high intensity illumination. In a FRAP experiment, a small region of a sample is illuminated with high intensity light, which bleaches the molecules in that region. Then, when the entire sample is illuminated with low intensity light, the bleached region does not fluoresce. By observing the recovery of fluorescence to the bleached region, the diffusivity of the fluorescent molecules can be determined.

In our proposed FRAP experiments, a laser is used to bleach a spot in the thin film, and the film is observed as the spot simultaneously diffuses and advects downstream. In such an experiment, an observer can easily determine three macroscopic parameters governing the behavior of the spot: the average velocity of the spot, $\langle u \rangle = u_s + u_p$, the effective diffusivity in the streamwise direction, $D_{\text{eff}}$, and the molecular diffusivity in the spanwise direction, $D$. These are the only parameters in (4.6), the depth-averaged diffusion-advection equation for a spot. The film thickness, $h$, could also be deduced, either by correlating to the total fluorescence intensity of the unbleached thin film, or by measuring with other methods (such as interferometry).

From these macroscopic parameters we aim to calculate values for $u_s$ and $u_p$, which will uniquely describe the flow within the film. We simultaneously solve (4.3) and (4.5) for $u_s$ and $u_p$, and express the solution in terms of $\text{Pe} = \langle u \rangle h/D$ and $D_{\text{eff}}/D$. Unfortunately, there are two branches to the solution for all combinations of
Pe = ⟨u⟩h/D and \( D_{\text{eff}}/D \),

\[
    u_p = \frac{D}{h} \left( \frac{7}{2} \text{Pe} + \sqrt{420} \left( \frac{D_{\text{eff}}}{D} - 1 \right) - \frac{7}{4} \text{Pe}^2 \right),
\]

(4.12)

\[
    u_s = \frac{D}{h} \left( -\frac{5}{2} \text{Pe} - \sqrt{420} \left( \frac{D_{\text{eff}}}{D} - 1 \right) - \frac{7}{4} \text{Pe}^2 \right)
\]

(4.13)

and

\[
    u_p = \frac{D}{h} \left( \frac{7}{2} \text{Pe} - \sqrt{420} \left( \frac{D_{\text{eff}}}{D} - 1 \right) - \frac{7}{4} \text{Pe}^2 \right),
\]

(4.14)

\[
    u_s = \frac{D}{h} \left( -\frac{5}{2} \text{Pe} + \sqrt{420} \left( \frac{D_{\text{eff}}}{D} - 1 \right) - \frac{7}{4} \text{Pe}^2 \right)
\]

(4.15)

For some applications of this technique it may be possible to differentiate between the branches if, for example, the approximate value of \( u_s \) or \( u_p \) is known. However, the ambiguity may limit the scenarios in which this technique yields valuable information.

### 4.6 Conclusions

Preliminary experiments were performed on a thin film of oil, driven by shear among an array of posts, as shown in the schematic in Figure 4.6 and described in earlier chapters of this thesis. For these experiments, a confocal microscope (Leica TCS SP5) was used to bleach a spot in the thin oil film and observe the fluorescence recovery. In these experiments, the film of oil was approximately 2 \( \mu \)m deep; conventional velocimetry techniques will not provide useful results for flows of this length-scale. The experiments displayed non-isotropic diffusion as predicted here, but correlating the enhanced diffusivity to flow properties is challenging for a number of reasons, including: 1) the possible ambiguity discussed above, and 2) the fact that the presence of the posts will modify the expected Taylor dispersion from its ideal case. Thus, these experimental results are omitted here.
Figure 4.6: Schematic of the experimental setup that was used to test the thin-film velocimetry technique.

We await a compelling application for this velocimetry technique before moving forward. The challenges associated with this technique could likely be overcome for a given application, but the approach will probably depend on the specific goals of the application. One additional application of this experimental technique that could prove promising would be the measurement of slip in a thin film flow. If either the shear- or pressure-driven velocity component were zero, FRAP velocimetry could be used to measure the slip velocity at the wall.
Chapter 5

Capillary bridges between soft substrates

The majority of this chapter has been published as a journal article of the same title [82]. The research was performed in collaboration with Tiara Heard and Howard Stone; they are coauthors of the article.

5.1 Introduction

Capillary adhesion is a classic example of the strength of surface tension forces; when a drop of wetting liquid bridges a gap separating two nearby surfaces, large normal forces can develop if the gap is very thin [83]. These forces may dominate a wide variety of physical systems, ranging from the stability of granular structures [84] and the adhesion of insects [85] to a glass of cold water stuck to a table. If the adhering objects are soft, the trapped droplet can pull the two surfaces of the gap together as shown in Fig. 5.1, which increases the adhesion force by magnifying the Laplace pressure and enlarging the contact radius. Previous efforts to understand capillary bridges between elastic materials have focused on interactions between spheres, extending the Hertz theory of contact [86] to include effects of a capillary meniscus.
We focus on the case of a droplet trapped in a thin gap, and report experiments, scalings, and closed-form solutions for the deformation.

A sessile droplet resting unconstrained on an elastic substrate deforms the solid underneath it by an amount $O(\gamma/E)$ which is termed the elasto-capillary length [90], where $\gamma$ is the surface tension of the fluid-fluid interface and $E$ is the Young’s modulus of the substrate [91, 92, 93, 94]. Most everyday liquids have a surface tension on the order of 10–100 mN/m. Therefore, in order for a sessile droplet to deform a solid substrate by even a few microns the material must be very soft, with a Young’s modulus in the range of 10–100 kPa [95, 96, 97].

We consider a drop trapped between two elastic surfaces (Fig. 5.1). A scaling analysis demonstrates that the vertical deformations, $u_z$, are orders of magnitude larger for this configuration than for a similarly sized sessile drop. For the case of a sessile drop the Laplace pressure scales as $\gamma/a$, where $a$ is the droplet radius. The pressure is balanced by elastic stresses within the solid $O(Eu_z/a)$ leading to deformations $O(\gamma/E)$ as indicated above. However, for a droplet between two surfaces separated by a distance $h$, as in Fig. 5.1, the Laplace pressure scales instead as $\gamma/h$, while the elastic stress scales as before. Balancing pressure and surface stress leads to deformations $O(\gamma a/Eh)$, which can be substantial if the droplet is flat ($h \ll a$).

Certain organisms with flexible appendages rely on moisture to form an adhesive capillary bridge to an underlying substrate [98]. The system we are studying is a simplified version of this case, and could bring an understanding to the different forces at play in these complex systems. In addition, our results extend the elasto-capillary instability of thin MEMS structures [99] to include high aspect-ratio microfluidic or other devices fabricated from flexible (but non-thin) polymeric materials. Microfluidic devices are known to deform at high flow rates [100], but may also be susceptible to deformation in a static configuration due to capillary forces alone. Our study also provides insight into crack propagation in the presence of moisture. Moisture at the
tip of a crack can arrest crack growth [101, 102], but the effect of moisture at some intermediary location is unknown.

5.2 Theory

We begin by describing in detail the geometry of our study, as shown in Fig. 5.1. We specify a cylindrical coordinate system whose origin lies at the center of a circular droplet. The width of the gap approaches $h$ as $r \to \infty$. The liquid-air interface has a surface tension $\gamma$ and is assumed to wet the solid with a contact angle $\theta_c \approx 0$. We limit our study to small Bond numbers, where gravitational effects are negligible compared to capillary forces. The two solids are identical, with Young’s modulus $E$ and Poisson’s ratio $\nu$. The vertical deflection of the surface of each substrate in the outward-normal direction is denoted $u_z$, and for an axisymmetric geometry we have $u_z = u_z(r)$. The radius of the droplet contact line, after the substrates have deformed and the droplet radius has equilibrated, is $a$. We start with the case of an infinitely thick substrate, and follow with an analysis of substrates with finite thickness, $d$.

Inside the contact radius, the droplet exerts a constant Laplace pressure, $\Delta p$, on the substrate. At the contact line a force/length equal to $\gamma$ pulls on the substrate.

![Figure 5.1: Cross-section of the axisymmetric geometry, where a droplet bridges two elastic substrates.](image)
In the limit of a very flat droplet, however, the deformation generated by the contact line force is negligible compared to the deformation induced by the Laplace pressure. Thus, the stress boundary condition at the surface of the elastic substrate is

$$\sigma_{zz} = \begin{cases} 
\Delta p & \text{for } r < a \\
0 & \text{for } r > a,
\end{cases} \quad (5.1)$$

where the stress is a function of the local strains by the standard linear constitutive relations \[103\].

In the limit of $d/a \to \infty$, the Navier equations with boundary conditions \[5.1\] have a well-known analytical solution \[91\]. The vertical deformation at the surface is

$$u_z(r) = -\frac{2(1-\nu^2)a \Delta p}{E I(r/a, d/a \to \infty)}, \quad (5.2)$$

where

$$I(r/a, d/a \to \infty) = \begin{cases} 
\frac{2}{\pi} E(r/a) & \text{for } r < a \\
\frac{2r}{\pi a} (E(a/r) - (1 - (a/r)^2)K(a/r)) & \text{for } r > a,
\end{cases} \quad (5.3)$$

and $K(\cdot)$ and $E(\cdot)$ are respectively, the complete elliptic integrals of the first and second kind. The function $I(r/a, d/a)$ is dimensionless, and has been introduced to simplify the presentation.

For a substrate with finite thickness, we follow a previously introduced method \[94\]. Briefly, the stress and displacement fields are converted to Fourier space using Hankel transform methods. In Fourier space, the deformation is linearly related to the stress by a second-order matrix. We specify the surface stresses given in Eq. (5.1), and perform the necessary manipulations to obtain a displacement in the form of
Eq. (5.2), but with
\[
I(\tau/a, d/a) = \int_0^{\infty} \frac{1}{s} \left[ (3 - 4\nu) \sinh(2sd) - 2sd \right] J_1(sa)J_0(sr) ds, \tag{5.4}
\]
where \( J_k(\cdot) \) is the \( k \)th order Bessel function of the first kind. It can be shown that Eq. (5.4) approaches Eq. (5.3) as \( d/a \to \infty \).

The next task is to self-consistently determine the Laplace pressure, \( \Delta p \), inside the droplet. We have already specified that the droplet perimeter is circular; this is the equilibrium shape for an ideal system devoid of surface imperfections, gap irregularities, etc. As shown in Fig. 5.1, the liquid-air interface has two principle radii of curvature, \( \rho \) and \( a_1 \). Thus, the Laplace pressure is \( \Delta p = \gamma (a^{-1} - \rho^{-1}) \) where
\[
\rho = (\frac{h^2}{2} - u_z(a)) \sqrt{1 + \left( \frac{du_z}{dr} \bigg|_{r=a} \right)^2}.
\]

We substitute this expression into Eq. (5.2) to determine the deformation for a given droplet size. The equations reveal natural non-dimensionalizations for the radial coordinate and deformation: \( [r] = a \) and \( [u_z] = \frac{4\pi(1-\nu^2)a}{Eh} \), which agree with the scaling argument given earlier. Thus, the non-dimensional deformation (denoted \( \tilde{\cdot} \)) is
\[
\tilde{u}_z(\tilde{r}) = \left( -\frac{h}{2a} + \frac{1}{\beta \left[ 1 - \frac{8\ell_{EC}a}{h^2} \tilde{u}_z(1) \right]} \right) \tilde{I}(\tilde{r}, d/a), \tag{5.5}
\]
where \( \beta = \sqrt{1 + \left( \frac{4\ell_{EC}}{h} \frac{du_z}{dr} \bigg|_{r=1} \right)^2} \), and \( \ell_{EC} = (1 - \nu^2)\gamma/E \) is a modified version of the elasto-capillary length introduced earlier.

We have already assumed that \( h \ll a \), since this limit produces the most dramatic deformations. Furthermore, we assume that \( \ell_{EC} \ll h \), because otherwise the gap would close with only the slightest amount of moisture. In these limits, Eq. (5.5)
simplifies considerably, and becomes

\[ \tilde{u}_z(\tilde{r}) = \frac{1}{1 - \Lambda \tilde{u}_z(1)} \cdot I(\tilde{r}, d/a), \quad (5.6) \]

with \( \Lambda = 8 \ell_{EC} a / h^2 \) as the ratio between the expected deformation and the half-height of the gap.\(^2\)

We now solve for the deformation at the contact line, \( \tilde{u}_z(1) \), by evaluating Eq. (5.6) at \( \tilde{r} = 1 \). This step produces a quadratic equation for \( \tilde{u}_z(1) \), with the two solutions \( \tilde{u}_z(1) = \left( 1 \pm \sqrt{1 - 4 \Lambda I(1, d/a)} \right) / 2 \Lambda \). The upper solution is in fact unstable, as we show next, but first we substitute the expression for the lower solution of \( \tilde{u}_z(1) \) back into Eq. (5.6) to arrive at

\[ \tilde{u}_z(\tilde{r}) = \frac{2}{1 + \sqrt{1 - 4 \Lambda I(1, d/a)}} \cdot I(\tilde{r}, d/a), \quad (5.7) \]

which gives the surface deformation in terms of only \( \Lambda \) and \( d/a \).

To examine the stability of the deformed system, we re-express the problem in terms of a minimization of energies rather than a balance of forces. The dominant contributions to the total energy of the system are 1) the elastic energy of the deformed substrate and 2) the reduction in energy that results from the droplet wetting the two solids. We neglect the energy of the liquid-gas interface since \( h \ll a \), which is equivalent to neglecting the secondary curvature \( a^{-1} \) in the prior calculations.

For a given droplet volume, \( V \), we calculate the total energy, \( U \), in terms of the deformed droplet radius, \( a \), as

\[ U = \frac{\pi Eah^2}{16(1 - \nu^2)V(d/a)} \cdot \left( 1 - \left( \frac{a_s}{a} \right)^2 \right) - 2 \pi \gamma a^2. \quad (5.8) \]

\(^2\)Examination of Fig. 5.4 reveals that \( \frac{du}{d\tilde{r}} \), and hence \( \frac{d\tilde{u}_z}{d\tilde{r}} \), is steepest at the contact line, \( \tilde{r} = 1 \). In fact, for a substrate with infinite thickness \( a_s \) is logarithmically singular at this point. The weak nature of this singularity, however, means that even large values of \( d/a \) produce \( \frac{d\tilde{u}_z}{d\tilde{r}} \) that is order one. Furthermore, it is likely that local effects due to the contact line force and the surface tension of the solid will dampen the singularity [94].
with the first term resulting from elastic contributions and the second resulting from wetting. Here, \( a_s = \sqrt{V/\pi h} \), is the radius of a droplet of volume \( V \) in an undeformed geometry, and \( V(d/a) = \int_0^1 I(\tilde{r}, d/a) \tilde{r} d\tilde{r} \).

The non-dimensional energy \( \tilde{U} = U/Eh^4 \pi^{128} \gamma (1 - \nu^2)^2 \) is plotted in Fig. 5.2 against the drop radius \( \Lambda \) for various non-dimensional drop volumes, \( \Lambda_s = \frac{8\ell_{EC} h}{\pi h} \sqrt{V/\pi h} \). Note that Fig. 5.2 and the following analysis are for a substrate of infinite thickness, i.e. \( d/a \to \infty \), but similar behavior occurs when the substrate is finite. For the smallest drop volume plotted, \( \Lambda_s = 0.05 \), we see that only one equilibrium state exists, corresponding to an energy minimum near the undeformed state of \( \Lambda = \Lambda_s \). Then, as the drop volume increases, i.e. \( \Lambda \) increases, a second unstable equilibrium appears (see the curve for \( \Lambda_s = 0.2 \)). This unstable equilibrium corresponds to the neglected negative root in Eq. (5.7). Finally, when \( \Lambda_s = \frac{\pi - 2}{2\pi} \sqrt{\frac{8 - \pi}{3\pi}} \approx 0.261 \), the minimum becomes a saddle point, and the gap collapses into a lower energy state corresponding to solid-solid contact.

Surprisingly, the maximum deformation at the critical value of \( \Lambda_s \) is only \( u_z(0)/h = \frac{\pi - 2}{4} \approx 0.285 \), meaning that collapse is triggered when the gap is barely half-way closed. This abrupt collapse is not reversible; indeed, the problem exhibits significant hysteresis that depends on the interfacial energy for solid-solid contact.

For the experimental conditions described below, we have observed that solid-solid contact, once initiated, is maintained after the drop of liquid evaporates completely. A similar bifurcation and pattern of hysteresis occurs in the elasto-capillary adhesion of thin structures \([99]\), and has been suggested for spheres \([88]\).

5.3 Experiments

We now present a series of experiments to test the predicted deformations. Our model experiment mimics the geometry of Fig. 5.1 with polydimethylsiloxane (PDMS) as
the elastic material, and a droplet of deionized water in the gap. The droplet is allowed to evaporate slowly so that a variety of different radii may be tested in identical experimental conditions. By varying the drop radius we change both non-dimensional parameters, $\Lambda$ and $d/a$.

The PDMS (Sylgard 184) is mixed in a base-to-curing-agent ratio of 10:1, 20:1, or 40:1, and baked at 70°C until it is fully cured (time varies depending on ratio). These recipes result in measured Young’s moduli of $E = 1250 \pm 50, 620 \pm 30, 87 \pm 4$ kPa, respectively. The highest value is two orders of magnitude larger than that used to observe elastocapillary deformations in previous studies of sessile droplets [95, 96, 97]. The back side of the PDMS is plasma-bonded to a rigid glass slide (1 mm thick) to achieve a zero-displacement boundary condition.

We use fluorescent beads (1.01 µm diameter, Bangs Laboratories) adsorbed to each PDMS surface to indicate the precise $z$-location of the surfaces. To attach the beads, the substrate is first treated with oxygen plasma for 20 s, and then 1 mL of a 250:1 dilution of the stock bead solution is deposited before being aspirated off. PDMS is by default slightly hydrophobic, so the surface must be modified to alter its
wetting characteristics. We follow the method of [104] to graft polyvinylpyrrolidone (PVP) polymer brushes to the surface of the PDMS, which results in a contact angle of $\theta_c \approx 4^\circ$. A ring-shaped spacer (ID = 2 mm, Kapton film) is placed between the two pieces of PDMS to set a constant gap height. We use different thicknesses of Kapton film to adjust the gap height.

A 25–250 nL droplet of deionized water is placed in the gap between the two surfaces, and the assembly is viewed from below with an inverted confocal microscope (Leica TCS SP5). We measure the deformation as the droplet slowly evaporates. The vacated gap is then used as the reference state for the undeformed geometry. Using interferometry we obtain precise measurements of differences in gap height over large $r$-distances, which are combined with a high-magnification confocal $z$-stack to give us the absolute gap height at a known reference point.

We report interferometry fringe patterns (2.5×, NA 0.25) in Fig. 5.3 which result from differences in gap height that cause either constructive or destructive interference [105]. Adjacent light bands (or dark bands) correspond to a difference in gap height of $\Delta h = \lambda/(2n)$, where $\lambda = 488$ nm is the wavelength of the light used, and $n$ is the refractive index of the material in the gap; $n = 1.00$ for air and $n = 1.33$ for water at our experimental conditions.

A 40× objective (NA 0.6) is used for the confocal $z$-stack, with a stack taken before and after each interferometric image (see Figs. 5.3A and 5.3B for an indication of the location and relative size of this query region). The fluorescent beads provide a precise indication of the absolute location of the top and bottom surfaces of the gap. The high magnification objective allows for a narrow depth of field, and hence precise $z$-measurements.

We analyze the gap profile with a droplet present (Figs. 5.3A,C) and after the droplet has evaporated (Fig. 5.3B). Notice that the height of the vacant undeformed gap in Fig. 5.3B is not constant; if it were constant we would not see the alternating
fringes. We determine the gap profile along a line that intersects the center of the droplet (Fig. 5.3A), and along an identical line in the undeformed image (Fig. 5.3B). We then subtract the profile of the deformed case from the undeformed case to obtain $2u_z(r)$.

The results provided in Fig. 5.4 show characteristic deformation profiles, $u_z(r)$, for different droplet sizes in a given gap geometry ($h = 58.3 \pm 0.4 \text{ µm}$, and $d = 3.2 \pm 0.1 \text{ mm}$). Given $d$ and $a$, we have a prediction from Eq. (5.4) for the shape of the deformation, $I(r/a, d/a)$. The shape can be expressed as

$$u_z(r/a) = u_z^m \cdot I(r/a, d/a) + u_z^o$$

(5.9)

with a magnitude $u_z^m$, and an offset $u_z^o$. We fit Eq. (5.9) to intersect the two data points outside of the droplet that are nearest and furthest from the contact line. As evident in Fig. 5.4, the intermediate points fit the theoretical shape for a range of $a$ (and hence $d/a$).
Figure 5.4: Deformation for \( a = 249,391,598 \) \( \mu m \). Experimental measurements are shown with symbols. The gap in the data corresponds to the unresolvable region beneath the meniscus. The solid lines denote the fit to Eq. (5.9) as described in the text. Inset: Deformation magnitude, \( u_z^m \), plotted against droplet size, \( a \). 40:1 PDMS is used for the pink points, 20:1 for the blue and gray, and 10:1 for the orange. From top to bottom, \( h = 66.4, 43.8, 58.3, 89.5 \pm 0.4 \) \( \mu m \), and \( d = 3.1, 3.8, 3.2, 3.0 \pm 0.1 \) mm. The colored lines denote the theoretical prediction, with their spread denoting uncertainty in geometric and material parameters. The non-circular gray points correspond to their counterparts in the main plot.

The data inside of the droplet requires special treatment. There is an unknown jump in the deformation between the inside and the outside of the droplet, since the fringes beneath the circular meniscus are obscured by refraction. To estimate this jump, we specify that the deformation at the inner fringe closest to the meniscus is equal to the prediction from the fit to Eq. (5.9). The remaining inner points are offset by the same amount, and follow the theory as can be seen in Fig. 5.4.

Experimental \( u_z^m \), determined from fitting to Eq. (5.9), are plotted against droplet radius, \( a \), in the inset of Fig. 5.4. Included with this data are theoretical predictions from Eq. (5.7), \( u_z^m = \frac{8\ell ECa}{h} \left( 1 + \sqrt{1 - \frac{32\ell ECa}{h^2} I(1, d/a)} \right)^{-1} \), which are calculated from measured geometric and material parameters \( (a, h, d, E, \nu, \gamma) \). The theoretical pre-
dictions for $u^m_2$ match the measurements quite well for a wide range of experimental conditions, described in the caption of Fig. 5.4. The deviations from theory that do occur are likely due to variations in $h$ or $\theta_c$, difficulties in measuring $E$, or secondary curvature effects as $a \rightarrow h/2$. The offset, $u^0_2$, in Eq. (5.9) is due to the limited $z$-stack resolution involved in determining the absolute deflection, and in all cases $|u^0_2| < 0.4 \, \mu m$.

5.4 Conclusions

As predicted by theory and proven with experiments, the geometry of a capillary bridge acts to magnify the deformation caused by capillary forces. This deformation, in turn, is expected to enhance the adhesion force through the mechanisms mentioned earlier: an increased drop radius and a decreased gap height. By applying these ideas to a variety of physical systems ranging from soft robotics [106] to the construction of granular structures [107], substrate softness may be used as a convenient tool to amplify the force of capillary adhesion.
Chapter 6

Bending of elastic fibres in viscous flows: the influence of confinement

The majority of this chapter has been published as a journal article of the same title [108]. The research was performed in collaboration with Phil Trinh, Helene Berthet, Nawal Quennouz, Olivia du Roure, Herbert Huppert, Anke Lindner, and Howard Stone; they are all coauthors of the article. The asymptotic analysis presented in Sections 6.4 and 6.9 is almost entirely the work of Phil Trinh.

6.1 Introduction

An anchored elastic fibre will bend when held perpendicular to the flow of a viscous fluid. The degree of bending varies with material properties of the fibre and the fluid, the flow rate, and the surrounding geometry. In particular, we consider a fibre that is anchored in a rectangular channel, with the walls of the channel positioned near the fibre and thereby confining the flow. We present experiments for this system and demonstrate how the three-dimensional geometry can be reduced to a two-dimensional model, and how quantities such as fibre deflection and velocity fields can be derived with the use of asymptotic and numerical methods.
The study of fluid-structure interactions has a rich history, though the majority of research has focused on flows at high Reynolds numbers [109]. In a recent example giving insight into such problems, it was shown how the drag induced by an anchored deformable fibre differs from the traditional drag induced by rigid objects [110]. In contrast, a number of papers study the dynamics of an elastic fibre submerged in a flow at low Reynolds number (Stokes flow). The fibre can be, for example, freely flowing [111, 112, 113], experiencing a body force [114], forced with a prescribed end motion [115, 116, 117], or held with one end anchored [118, 119, 120, 121].

A complication not emphasised in the above low-Reynolds-number studies is the effect of a confining geometry on the resultant flow fields, and the corresponding influence on fibre deformation [122]. In examples ranging from bacteria motion [123] to micro-pumps [124], confinement has been shown to have a significant effect. Indeed, it was recently shown that the drag on a fixed rigid cylinder is a strong function of the degree of geometrical confinement [125]. Our goal then is to study the influence of confinement on the dynamics of an elastic fibre in low-Reynolds-number flow.

Our study has practical implications, with many possible applications in the field of microfluidics [126, 127]. For example, thin deformable structures with a spring-like geometry, anchored in the center of a microfluidic channel, can be used as sensors to measure flow rate [128]. In this study the deformation of the sensor is calibrated experimentally against known flow rates, to obtain a relationship between deformation and flow rate. Thin flexible structures are also prevalent in biology, with examples including cilia and flagella, which are primary appendages for feeding and propulsion for many types of cells [129]. Our analysis could bring insight to the dynamics of cell motility and feeding in confined flow. As an additional example, it was recently reported that flexible fibre-like biofilm structures originate from wall-anchored biofilms located near the corners of micro-channels [130].
We begin this chapter by describing, in section 6.2, our microfluidic experiments that motivate the study. The geometry, shown schematically in figure 6.1, is chosen to highlight the effects of confinement on fibre bending, as well as potentially to offer a method of flow measurement [128]. In section 6.3, we propose a mathematical model for the corresponding low-Reynolds-number flow, which focusses on the particular case of weakly deflected fibres, when only a small amount of fluid passes between the fibre and the laterally confining channel walls. This step allows a reduction of the three-dimensional geometry to a two-dimensional model, which is further studied using asymptotic analysis in section 6.4, as well as numerical methods in section 6.5. We offer a comparison to experimental results in section 6.6 and conclude in section 6.7 with a summary of the implications of our analysis.

6.2 Experiment

The experimental geometry consists of a long rectangular channel of high aspect ratio, with a rectangular fibre extending from one of the sidewalls as shown schematically in figure 6.1. The fluid flows in the $x$-direction, the fibre extends in the $y$-direction,
and the channel is thin in the $z$-direction. The fibre is flexible, both because of its slenderness and because it is made of a soft material. We pump a Newtonian fluid through the channel at known flow rates, and record the shape that the fibre takes as in the inset of figure 6.2. Experiments are performed over a broad range of flow rates, but in this work we focus on small deflections of the fibre.

### 6.2.1 Experimental Methods

The rectangular microfluidic channel is made of polydimethylsiloxane (PDMS, General Electric) and has a depth $D = 66 \, \mu m$ and a height $H = 400 \, \mu m$ (see figure 6.1). The channels are approximately 3 cm long and are moulded on a silicone wafer using standard soft-lithography techniques. The fabrication allows for features such as walls and pillars that span the entire depth of the channel, but features such as the fibre, which only partially block the depth, present a difficulty and necessitate a different fabrication method.

We implement the technique of “stop-flow lithography” [131] and succeed in making highly confined fibres that are anchored in the channel. Following previously introduced polymerisation methods [122], the channel is first filled with a photo-curable solution of 90 wt% Polyethylene glycol diacrilate, with average molecular weight 575 (PEGDA-575, Aldrich Chemistry), and 10 wt% 2-hydroxy-2-methylpropiophenone photo-initiator (Aldrich Chemistry). Great care is taken in preparing the solution so as to eliminate irreproducibilities in the polymerisation process. A fresh solution is mixed before each experiment, nitrogen is run through the solution for 30 minutes to purge dissolved oxygen, and then the solution is degassed for 30 minutes to remove nitrogen bubbles. A photomask with the desired fibre geometry is placed in the light path of the fluorescence lamp (X-Cite) on a microscope (Zeiss) at $10 \times$ magnification, and the shutter is opened for 225 ms. This procedure cures the unmasked portion
of the solution in the channel. The exposure time was determined to give the most accurate reproduction of the desired fibre geometry with our specific setup.

When the shutter is open, the entire depth of the channel not blocked by the mask is exposed to ultraviolet light. The polymerisation reaction can only occur away from PDMS surfaces however, since the reaction is inhibited by dissolved oxygen and PDMS is permeable to oxygen. Therefore, next to all surfaces there exists a thin “inhibition layer” that cannot be polymerised [132]. The existence of the inhibition layer allows us to create a fibre that partially blocks the channel depth, but it also presents a difficulty in mounting the fibre because there is a similar inhibition layer next to lateral walls. To overcome this issue, we polymerise the fibre so that it attaches to a wall that is also polymerised from PEGDA. The wall, in turn, is anchored with multiple PDMS posts, and gives the fibre a clamped boundary condition at its base (see [128] for a similar example using one post).

We observe the inhibition layer to be approximately 5–6 µm, based on observations that \( D - d \approx 10–12 \mu m \) (accounting for an inhibition layer on both the top and bottom surfaces), where \( D \) is the depth of the channel, and \( d \) is the depth of the fibre (see figure 6.1). The channel depth is measured on the silicon mould using a mechanical profilometer (Dektak). The fibre depth is measured optically, by polymerising un-anchored fibres in the channel, and applying flow so that they flip to their side. The cross-section of the fibre is observed to be rectangular, but the very tip is slightly rounded in the xy-plane as shown in the microscope images of figure 6.2. In our experiments, the fibre width, \( w \), varies from 22 to 34 µm, and the fibre height, \( h \), varies from 144 to 293 µm. A value for the Young’s modulus of a material polymerised under these specific cross-linking conditions could not be found in the literature (measurements under different cross-linking conditions exist [122]). Thus, we use our model to extract the Young’s modulus as discussed in section 6.6.
Figure 6.2: Particle paths from a channel with fibre height $h = 226 \, \mu m$, fibre width $w = 31 \, \mu m$, and fibre depth $d = 34 \, \mu m$. The channel has height $H = 400 \, \mu m$, depth $D = 45 \, \mu m$, and flow rate $Q = 3 \, \mu L/min$. The inset displays phase contrast images of the same fibre without tracer particles, for flow rates of $Q = 0, 3, 8, 15,$ and $30 \, \mu L/min$ (left to right). The higher flow rates are included here for illustrative purposes, and are not considered to be in the linear regime of our model. Scale bars are 100 \, \mu m.

In our experiments, we pump a solution of 100 wt% PEGDA-575 through the channels, and measure the deflection of the tip of the fibre at varying flow rates. We use this solution to guarantee that the polymerised fibre, which is a gel, does not swell, and to avoid accidental polymerisation that might occur if the photo-initiator were included in the solution. The density of the solution is reported by the manufacturer to be $\rho = 1.12 \, \text{g/mL}$. The kinematic viscosity of the solution was measured with a capillary viscometer (Schott) to be $\mu/\rho = 50 \, \text{mm}^2/\text{s}$, which agrees well with the value provided by the manufacturer. The fluid is contained in a glass syringe (500 \, \muL, SGE Analytical Science), and injected with a precision syringe pump (Cetoni) into the microfluidic device. The fibres are imaged at a magnification of $20 \times$ (see inset of figure 6.2) with the same microscope used for polymerisation, and a digital camera (PixeLINK) connected to a computer. The fibre deflects to a steady shape, $u(y)$, and we measure with ImageJ (NIH) the deflection at the tip, $u(h_y)$, where $h_y$ is the $y$-coordinate of the center of the fibre tip as in figure 6.1.
6.2.2 Observations

To gain qualitative insight into the velocity field in the channel, we seed the fluid with 2 µm polystyrene particles (Duke Scientific) and observe the particle traces. The suspension is pumped through the device using the same setup as for the deflection experiments, and the particles are imaged through the microscope with high-speed videography (3000 images/sec, Photron Fastcam). The resulting images are superimposed to obtain the pathlines shown in figure 6.2. Note that the particles are visible through the entire depth of the channel, so the image that we see is a projection of the pathlines in the $z$-direction. Some of the pathlines appear to pass through the fibre, while others are bent around it. The particles that appear to pass through the fibre actually flow in the thin gap above and below it, and their traces are faintly visible as the white lines on the fibre perpendicular to its axis. The balance between flow through the thin gaps above and below the fibre, and flow deflected around the fibre, is a defining feature of the problem. The competing confinement, and hence resistance, of flow in both directions determines this balance, and influences the degree of bending of the fibre.

The focus of this chapter is predicting the fibre deflection, and figure 6.3 shows the observed deflection versus imposed flow rate for a representative data set. A log-log plot of the data reveals that at low flow rates the deflection of the fibre tip, $u(h_y)$, is linearly proportional to the flow rate, $Q$. Though the current work focuses on this deflection regime, experiments were done over a wide range of flow rates extending far beyond the initial linear regime, and the data is included here both for completeness and to allow us to assess how far our linearized model is valid. At the flow rates deemed to be in the linear regime (up to $Q = 1.2$ µL/min), no deflection of the channel was observed; indeed, a theoretical argument balancing pressure forces against channel elasticity predicts that the channel deformation in this flow regime
Figure 6.3: (a) Fibre tip deflection, $u(h_y)$, versus flow rate, $Q$, for $d = 56 \mu m$, $D = 66 \mu m$, $H = 400 \mu m$, and varying values of $w$ and $h$. (b) The same experiments with a greater range of data plotted on logarithmic axes, which emphasise the linear trend at low flow rates.

is less than 1% \cite{100}. The average Reynolds number, $Re_D$, calculated from the flow rate and channel depth, does not exceed $10^{-3}$ for flow rates in this regime.

### 6.3 A two-dimensional mathematical model

Before beginning we note that the full problem presented in figure 6.1 could be solved for individual cases using a finite-element package or a boundary integral approach. These solution methods could, in principle, capture high degrees of fibre deflection and the corresponding three-dimensional flow fields. However, we choose to use mainly analytical methods. Our solution technique fully encompasses the regime of our experiments, and allows general insight into related problems. We are able to extract scalings, and to obtain individual solutions through analytical and simple numerical calculations.

#### 6.3.1 Dimensional Problem

In this section we reduce the three-dimensional geometry of figure 6.1 to a two-dimensional flow approximation. To start with, we define the parameters of the
problem using figure 6.1 as a guide. The dimensions of the channel are given in upper case, with height $H$ and depth $D$. The volumetric flow rate is $Q$. For the fibre, the dimensions are given in lower case, with undeflected height $h$, depth $d$, and width $w$. Note that $h$ also corresponds to the total arc-length of the deflected fibre, which is assumed to be inextensible. The height of the deflected fibre, projected on the $y$-axis, is given by $h_y$.

We use an Euler-Bernoulli model of beam deflection in which the fibre is approximated as being infinitely thin, with its deflection in the $x$-direction relative to $x = 0$ given by $u(y)$, where $y$ ranges from 0 to $h_y$. If the fibre is not doubled-over, the coordinate $y$ uniquely defines a point along the fibre path. We also define a local coordinate system along the fibre path, shown in the inset of figure 6.1, with tangent $s$ and normal $n$. For an undeflected fibre, the positive $n$-direction corresponds to positive $x$, and the positive $s$-direction corresponds to positive $y$. Note that the curvilinear coordinate system is only used for derivatives; the absolute location of the fibre is described in cartesian coordinates in order to relate it to the pressure, $p(x, y)$. The fibre curvature, $\kappa(y)$, is defined as positive in the positive $n$-direction.

In our analysis, the flow in the $xy$-plane is assumed to correspond to Hele-Shaw flow, which requires that the channel is thin and that the reduced Reynolds number is small,

\begin{align}
D &\ll H, \\
(D/H)^2 Re_D &= (D/H)^2 (\rho D v/\mu) \ll 1.
\end{align}

From the Hele-Shaw approximation, the out-of-plane component of the velocity $v_z$ is set to zero, while the depth-averaged velocities, $\bar{v}_x$ and $\bar{v}_y$, satisfy Darcy’s law with $p$ as the pressure,
\[ \mathbf{v} = (v_x, v_y) = -\frac{D^2}{12\mu} \nabla p. \] (6.2)

Since the flow is incompressible, \( \nabla \cdot \mathbf{v} = 0 \), and we obtain the Laplace equation for the pressure in the \( xy \)-plane

\[ \nabla^2 p = 0, \] (6.3)

with \( \nabla^2 = \partial_{xx} + \partial_{yy} \). For the remainder of the chapter, we shall always refer to depth-averaged velocities and write \( \mathbf{v} = \mathbf{v} \).

The velocity tends to a uniform value \( v_x = Q/DH \) as \( |x| \to \infty \), far away from the fibre. We integrate the momentum equation (6.2) to obtain a far-field boundary condition on pressure,

\[ p \to \left( -\frac{12\mu v_x}{D^2} \right) x = \left( -\frac{12\mu Q}{D^3 H} \right) x \] (6.4)
as \( |x| \to \infty \). The boundaries at the base of the fibre and the opposite wall are impermeable. Thus, to satisfy no flux at both surfaces we specify that

\[ \frac{\partial p}{\partial y} = 0 \quad \text{on } y = 0, H. \] (6.5)

The pressure field is defined within an arbitrary constant, so without loss of generality we set the pressure at the tip of the fibre, \( y = h_y \), to be zero,

\[ p = 0 \quad \text{on } x = u(h_y) \text{ and } y = h_y, \] (6.6)

where the deflected fibre is given by \( x = u(y) \).

Since the fibre only partially blocks the channel depth, some fluid can flow through the narrow gap above and below the fibre (see cross-section in the inset of figure 6.1 and the flow visualisation shown in figure 6.2). Noting that the depth of the gap is
\( \frac{1}{2}(D - d) \), and that the width of the gap is the width of the fibre, \( w \), we observe for the parameters in the experiments that

\[ \frac{1}{2}(D - d) \ll w, \]  

(6.7)

allowing us to use the lubrication approximation to determine the flow profile within the gap. In addition, the depth of the channel, \( D \), and the depth of gap, \( \frac{1}{2}(D - d) \), satisfy the condition

\[ \frac{1}{2}(D - d) \ll D, \]  

(6.8)

implying that the majority of the pressure drop occurs within the gap, since the resistance to flow within the gap is much higher than the resistance to flow in the main channel. This observation allows us to take the pressure on either side of the gap to be approximately constant in \( z \) but varying along the fibre length, denoted \( p_{\pm}(y) \), where the subscript refers to either the right or left side of the fibre, respectively.

Furthermore we require that the width of the fibre is much less than its length, or

\[ w \ll h, \]  

(6.9)

because we approximate the fibre as an infinitely thin barrier with a certain permeability when we solve for the flow in the \( xy \)-plane. Under this approximation, the entering flux at one point on the fibre equals the exiting flux directly opposite it. Leakage dynamics that are more complex cannot be captured by a permeable barrier approximation. If condition (6.9) is met, then in the gap the resistance to flow perpendicular to the fibre is much less than the resistance parallel to the fibre, and the predominant flux through the fibre is in the perpendicular direction. Thus, the width of the fibre may be shrunk to zero while maintaining accurate leakage from the perspective of the outside flow.
The lubrication approximation is applied within the gap to relate the pressure drop across the fibre to the flow through the gap. We define the local two-dimensional flow rate perpendicular to the fibre and summed over both gaps as \( q(y) \). With the height of each gap as \( \frac{1}{2}(D - d) \), the length of each gap as \( w \), and the flow rate through one gap as \( \frac{1}{2}q(y) \), the lubrication equation within one gap (either above or below the fibre) integrates to

\[
\frac{p_+(y) - p_-(y)}{w} = -6\mu q(y) \left( \frac{2}{D - d} \right)^3. \tag{6.10}
\]

By continuity, the local flow rate above and below the fibre must equal the flow rate perpendicular to the fibre (in the \( n \)-direction) immediately adjacent to it. We take the inner product of (6.2) with the vector in the \( n \)-direction to calculate the average velocity normal to the fibre, and multiply by the channel depth, \( D \), to obtain

\[
q(y) = -\frac{D^3}{12\mu} \frac{\partial p(y)}{\partial n}. \tag{6.11}
\]

Thus, combining equations (6.10) and (6.11) yields a mixed-type (Robin) boundary condition on the pressure to be satisfied along the length of the fibre in the \( xy \)-plane,

\[
p_+(y) - p_-(y) = \left( \frac{4wD^3}{(D - d)^3} \right) \frac{\partial p(y)}{\partial n} \quad \text{for} \quad 0 < y < h_y. \tag{6.12}
\]

Note that the derivative of the pressure is continuous, so it is not necessary to specify on which side of the fibre it is calculated. With boundary condition (6.12) we are no longer concerned with the specifics of the flow in the gap. Effectively, the fibre is modeled as being infinitely thin with a certain permeability governed by the coefficient linking \([p_+(y) - p_-(y)]\) to \( \partial p(y)/\partial n \) in the boundary condition. In physical terms, (3.12) flattens the leakage dynamics into the \( xy \)-plane, by equating the flux entering and exiting the fibre gap to the flux through the fibre gap itself.
In deriving (6.12) we have approximated the flow as transitioning directly from Hele-Shaw flow away from the fibre to lubrication flow through the fibre itself. However, a transition region with a more complex three-dimensional velocity field must exist. For simplicity we require that the flow in this region is governed by Stokes’ equations, necessitating that the Reynolds number here is small (not just the reduced Reynolds number as in (6.1b)). It is sufficient to specify this condition on the average Reynolds number calculated from the bulk flow rate and channel depth,

\[ R_e D = \frac{\rho D v}{\mu} = \frac{\rho Q}{\mu H} \ll 1. \]  

(6.13)

If (6.13) is satisfied then we estimate that the size of the transition region scales with the fibre depth, \( d \) (a similar idea appears in [133]). Thus, in the transition region, a pressure drop of order \( \mu v / d \) occurs and is not accounted for in (6.12). In order for our approximation to be valid we require that this additional pressure drop is small compared to both the pressure drop in the Hele-Shaw region, and the pressure drop through the gap itself. A scaling argument shows that this condition may be reduced to the requirement that the depth of the channel is much less than the height of the fibre,

\[ D \ll h, \]  

(6.14)

along with inequalities (6.7) and (6.8).

To calculate the degree of bending of the fibre, we compute the force per unit length acting normal to the fibre, \( f_n(y) \), by combining the pressure forces on the left and right faces, \( f_p(y) \) with the viscous forces on the top and bottom faces, \( f_v(y) \). The pressure is already specified, and the viscous force may be found by calculating the
shear stress within the gap, $\tau_{nz}$, from the lubrication equation (6.10). Consequently,

$$
fn(y) = f_v(y) + f_p(y) = \left( 2\tau_{nz}w \right) - \left( \left[ p_+(y) - p_-(y) \right] d \right)
= \left( -\frac{p_+(y) - p_-(y)}{w} \right) \cdot \left( \frac{D - d}{2} w \right) - \left( \left[ p_+(y) - p_-(y) \right] d \right)
= -\frac{1}{2} (D + d) \left[ p_+(y) - p_-(y) \right].
$$

(6.15)

The fibre is narrower than it is deep, with $w < d$, so it only deflects in the $xy$-plane, with no torsion. We treat the displacement of the fibre using a simplified version of the beam-bending equation that is derived in the first Appendix, 6.8.

$$
\frac{d^2\kappa}{ds^2} + \frac{1}{2} \kappa^3 = -\frac{(D + d)}{2EI} \left[ p_+(y) - p_-(y) \right].
$$

(6.16)

where $s$ is the arc-length and $\kappa(y)$ is the local curvature of the fibre. The shape of the deflected fibre determines $s(y)$. The Young’s modulus of the fibre is $E$, and the moment of inertia is

$$
I = \frac{dw^3}{12}.
$$

(6.17)

Finally, to complete the problem statement four boundary conditions are required on the fibre; the end of the fibre clamped to the channel wall requires $u = \frac{du}{ds} = 0$ at $y = 0$, and since the fibre is slender the other end can be assumed to be moment- and shear-free, with $\frac{d^2u}{ds^2} = \frac{d^3u}{ds^3} = 0$ at $y = h_y$.

### 6.3.2 Nondimensional equations

We nondimensionalise the spatial coordinates and deflection by the channel height, $H$, and we scale the pressure so as to normalise the far-field condition. The scale of
each variable, given in square brackets, is

\[ x = y = s = n = \kappa^{-1} = H \quad \text{and} \quad p = \frac{12\mu Q}{D^3}, \tag{6.18} \]

and we maintain the same notation for dimensionless quantities. The nondimensionalised \( y \)-coordinate of the tip of the deflected fibre is \( c_y = h_y/H \). After algebraic steps the nondimensional equations become

\[
\begin{align*}
\nabla^2 p &= 0 \quad \text{within the cell} \tag{6.19a} \\
\frac{\partial p}{\partial y} &= 0 \quad y = 0, 1 \tag{6.19b} \\
p &\to -x \quad x \to \pm \infty \tag{6.19c} \\
p &= 0 \quad x = u(c_y) \text{ and } y = c_y \tag{6.19d} \\
\frac{\partial p}{\partial n} &= \beta (p_+ - p_-) \quad x = u(y) \text{ and } y \in (0, c_y) \tag{6.19e} \\
\frac{d^2 \kappa}{ds^2} + \frac{1}{2} \kappa^3 &= -\epsilon (p_+ - p_-) \quad x = u(y) \text{ and } y \in (0, c_y) \tag{6.19f} \\
u &= \frac{du}{ds} = 0 \quad x = 0 \text{ and } y = 0 \tag{6.19g} \\
\frac{d^2 u}{ds^2} &= \frac{d^3 u}{ds^3} = 0 \quad x = u(c_y) \text{ and } y = c_y, \tag{6.19h}
\end{align*}
\]

with three dimensionless parameters defining the problem,

\[
\begin{align*}
\text{fibre height} & \quad c = \frac{h}{H} \tag{6.20a} \\
\text{permeability parameter} & \quad \beta = \left( \frac{(D - d)^3}{4wD^3} \right) H \tag{6.20b} \\
\text{bending parameter} & \quad \epsilon = \left( \frac{D + d}{2EI} \right) \left( \frac{12\mu Q}{D^3} \right) H^3. \tag{6.20c}
\end{align*}
\]

Not surprisingly, there are multiple ways to nondimensionalise the distances \( x, y, s, n, u, \) and \( \kappa^{-1} \) in (6.18): for example, one can choose the length scale so the channel height \( (H) \) is fixed, or choose the length scale so the fibre height \( (h) \) is fixed.
Since the focus of this chapter is confinement, we choose to nondimensionalise by $H$ (fixing the non-dimensional channel height) and so $H$ appears in all three of the dimensionless parameters, $c$, $\beta$, and $\epsilon$. This also isolates all effects of the fibre height to one variable, $c$.

For some readers, however, it might be more natural for $h$ to appear in the bending parameter, $\epsilon$. Had we chosen the length scale to be $h$ (fixing the non-dimensional fibre height) the pre-factor on the right-hand side of (6.19f) would contain an $h^4$ factor due to the second derivative of the curvature. In section 6.4.3, we find the proper scaling of beam deflection (nondimensionalised by $H$) for small fibres, $c \rightarrow 0$, to be $\epsilon c^5$. In dimensional terms, this deflection is exactly equivalent to the $h^4$ scaling that would have appeared had the lengths been nondimensionalised by $h$.

### 6.4 Asymptotic analysis

We focus on our experiments that are in the regime of small permeability and small deflection, so we provide an asymptotic analysis of the limit where $\epsilon \ll 1$ and $\beta \ll 1$. This analysis offers an analytical solution for the leading-order pressure field and fibre deflection, and insight into the scalings of the problem. We shall see that there are two distinguished limits depending on whether $1 \gg \beta \gg \epsilon$, referring to permeability dominated flows, or $1 \gg \epsilon \gg \beta$, referring to flexion dominated flows.

Before continuing we note that for small deflections, $\epsilon \ll 1$, the leading-order problem can also be solved for arbitrary $\beta$, albeit numerically. For this technique, an asymptotic expansion is performed using the variable $\epsilon$, with $\beta$ left as a parameter in the problem, including at leading order. The numerical solution technique is described in section 6.5.2.
In this section however, we consider an expansion of the pressure, $p(x, y)$, and deflection, $u(y)$, according to

$$p = \sum_{m=0}^{\infty} \delta^m p_m \quad \text{and} \quad u = \sum_{m=1}^{\infty} \epsilon^m u_m,$$  

(6.21a, b)

where $\delta (\ll 1)$ is to be determined. At each order, $p_m$ satisfies the Laplace equation, with impermeable walls at $y = 0$ and 1, and the reference pressure being set to zero at the fibre tip, as in (6.19a)–(6.19d). Because of inextensibility, the total length of the fibre, $c = h/H$, can be related to the projected height of the fibre, $c_y$, through

$$c = \int_0^{c_y} \sqrt{1 + \left(\frac{du}{dy}\right)^2} \ dy = c_y + \mathcal{O}(\epsilon^2),$$  

(6.22)

so all instances of $c_y$ in (6.19) can be replaced with $c$ to $\mathcal{O}(\epsilon^2)$. 

Figure 6.4: (a) Constant pressure curves (gray), and streamlines (black), for flow past an undeflected, impermeable fibre of length $c = 0.5$. This is the leading-order solution, $p_0$ and $\psi_0$, for flow past a lightly flexed and weakly permeable fibre. (b) Leading-order pressure versus $x$ at various $y$-heights in the channel.
At zeroth order the bending parameter $\epsilon = 0$, so the fibre is undeflected and vertical. Therefore, the leading-order geometry is symmetric about $x = 0$. We expect that the streamlines of the leading-order flow, corresponding to $p_0$, are also symmetric about $x = 0$, because the Laplace equation is linear in $x$. For reference, we show in figure [6.4] the leading-order streamlines and curves of constant pressure calculated using the methods of this and the following sections.

Due to this symmetry there is no vertical component to the zeroth-order velocity in the region above the fibre, so $\partial p_0 / \partial y = 0$ at $x = 0$. We integrate in $y$, applying the boundary condition that the pressure at the tip of the fibre is set to zero by (6.19d), to obtain

$$p_0 = 0 \quad x = 0 \text{ and } y \in (c, 1).$$

(6.23)

This condition, combined with the streamlines being symmetric about $x = 0$, requires that $p_0$ must be antisymmetric about $x = 0$.

Now we examine the bending equation (6.19f) using the expansion of (6.21). Applying the standard definition of curvature in cartesian coordinates, and assuming asymmetry in $p_0$ about $x = 0$, we see that the bending equation reduces to the usual small-deflection form,

$$\epsilon \frac{d^4 u_1}{dy^4} + \mathcal{O}(\epsilon^2) = -\epsilon \left( p_0 \bigg|_{x=0+} - p_0 \bigg|_{x=0-} \right) + \mathcal{O}(\epsilon \delta, \epsilon^2) \sim \mp 2\epsilon p_0 \bigg|_{x=0\pm}. \quad (6.24)$$

This shows that, as expected on physical grounds, the leading- and first-order deflection is determined entirely by the zeroth-order pressure field, $p_0$.

Next we write the normal derivatives in the fibre permeability condition (6.19e) as

$$\left. \frac{\partial p}{\partial n} \right|_{x=u\pm} = \left[ 1 + \left( \frac{du}{dy} \right)^2 \right]^{-1} \left( \frac{\partial p}{\partial x} - \frac{du}{dy} \frac{\partial p}{\partial y} \right)_{x=u\pm}. \quad (6.25)$$
Using (6.25) in the fibre condition (6.19e), and substituting the asymptotic expansions for $u$ and $p$ (6.21), we obtain

$$
\left(1 - O(\epsilon^2)\right) \left[ \left( \frac{\partial p_0}{\partial x} + \delta \frac{\partial p_1}{\partial x} + O(\delta^2) \right)_{x=0^\pm} + \left( \epsilon u_1 + O(\epsilon^2) \right) \left( \frac{\partial^2 p_0}{\partial x^2} + O(\delta) \right)_{x=0^\pm} \\
- \left( \epsilon \frac{du_1}{dy} + O(\epsilon^2) \right) \left( \frac{\partial p_0}{\partial y} + O(\delta) + O(\epsilon) \right)_{x=0^\pm} \right] = \beta \left( p_0 \big|_{x=0^+} - p_0 \big|_{x=0^-} \right) + O(\beta \delta) + O(\beta \epsilon). 
$$

(6.26)

Replacing the permeability condition on $x = u(y)$ by the expansion (6.26) about $x = 0$ is formally correct, as long as the linear beam theory of (6.24) holds, and in particular the expansion (6.21) remains well-ordered. However, beam deflections that are too large may cause the series to converge slowly.

From (6.26), and with $\epsilon \ll 1$, $\beta \ll 1$, the leading-order boundary condition requires that at the fibre

$$
\frac{\partial p_0}{\partial x} \bigg|_{x=0^\pm} = 0 \quad \text{at } x = 0 \text{ and } y \in (0, c),
$$

(6.27)

which is the standard no-flux constraint. Combining the boundary conditions (6.23) and (6.27) at $x = 0$ with the boundary conditions (6.19b) at $y = 0, 1$, and (6.19c) at $x \to \pm\infty$, the $p_0$ problem is fully defined.

Obtaining appropriate boundary conditions for the first-order correction $p_1$ requires us to specify the relative magnitudes of the permeability, $\beta$, and flexion, $\epsilon$, and this leads to three distinguished limits of (6.26).

The first limit is marked by permeability dominated effects, with $1 \gg \beta \gg \epsilon$. In this case, we choose $\delta = \beta$ in (6.26), and recalling the aforementioned symmetry of
$p_0$, the first-order boundary condition requires that

$$\frac{\partial p_1}{\partial x} \bigg|_{x=0}\at x = 0 \text{ and } y \in (0, c).$$ (6.28)

The result is a dominant balance between the horizontal gradient of $p_1$ and the permeability. We see that the first-order correction to the pressure field is determined by the leakage through the fibre, and is in fact independent of the deflection. Therefore, we may assume that above the straight fibre the corrected streamlines remain horizontal, so

$$p_1 = 0 \quad \text{ at } x = 0 \text{ and } y \in (c, 1),$$ (6.29)

similar to the leading-order pressure field $p_0$.

The second distinguished limit is marked by flexion dominated effects, with $1 \gg \epsilon \gg \beta$. Thus, we choose $\delta = \epsilon$, and the first-order boundary condition requires at $O(\epsilon)$ that

$$\frac{\partial p_1}{\partial x} \bigg|_{x=0}\at x = 0 \text{ and } y \in (0, c).$$ (6.30)

We see here that an advective flux around the deflected fibre leads to the gradient in $p_1$. In order to apply this boundary condition, the deflection, $u_1(y)$, must first be computed using (6.24).

The third distinguished limit is marked by equal magnitudes of permeability and flexion, with $\delta = \epsilon = O(\beta)$, and the boundary condition on $\frac{\partial p_1}{\partial x}$ is the sum of (6.29) and (6.30). The linearity of the asymptotic approximation then implies that the solutions of the permeability and flexion dominated limits can be independently superimposed to form a solution of the third type.
It is worthwhile to note that for a given channel and fibre geometry, all three distinguished limits may be reached by varying the flow rate, $Q$. Since $\epsilon$ is the only term that contains $Q$, and $\epsilon$ is proportional to $Q$, it should be possible, starting with low $Q$ and increasing, to satisfy in a sequential manner the conditions that $\epsilon \ll \beta$, $\epsilon = \mathcal{O}(\beta)$, and $\epsilon \gg \beta$. In other words, if the flow rate in an experiment is gradually increased from a very low value, the first-order pressure correction will transition from being permeability-dominated to flexion-dominated.

### 6.4.1 The leading-order pressure, $p_0$

The leading-order pressure, $p_0$, consists of potential flow through a finite channel past a infinitely thin, impermeable fibre. In solving for the potential flow contained within an impermeable boundary, complex variable methods known as *conformal mapping* can be used to determine analytic solutions in certain cases. These methods rely upon mapping the physical geometry to an alternate and simpler geometry (such as a disc or upper half-plane) where the problem is hopefully tractable. Once this new problem is solved, inverting the map produces the solution in the original geometry.

The case of flow past a thin, vertical barrier in an unconfined, upper half-plane is well known as a standard exercise in conformal mapping (p. 355 in [35]). We have found that an explicit formula can be written for the confined geometry as well. The method begins by noting that since $p_0$ satisfies the Laplace equation, we can first define the complex potential

$$ w_0(z) \equiv p_0(x, y) + i\psi_0(x, y), \quad (6.31) $$

where $\psi_0$ is the stream function and harmonic conjugate of $p_0$, and $z = x + iy$. Note that $w_0$ satisfies the Laplace equation, and all the solid boundaries are impermeable.
Figure 6.5: The physical \(z\)-plane is mapped to \((a)\) the potential \(w_0\)-plane, and then to \((b)\) the upper-half \(\zeta\)-plane. In the potential plane, streamlines (gray) run parallel to the \(p\)-axis, while in the \(\zeta\)-plane, streamlines (gray) are generated by a sink at \(\zeta = 0\).

to leading order. In our method, the physical \(z\)-plane (figure 6.4) is first mapped to the potential \(w_0\)-plane (figure 6.5\(a\), maintaing labels \(A\)-\(G\)). In particular, the tip of the barrier \((z = i c)\), is mapped to \(w_0 = 0\) because of condition (6.23), while the two stagnation points are located at \(w_0 = \pm k\). The value \(k\) is currently unknown, and corresponds to the barrier ‘length’ in the potential plane.

Next, the \(w_0\)-plane is mapped to the upper-half \(\zeta\)-plane (figure 6.5\(b\)). To simplify calculations, we choose to map point \(F\) to \(\zeta = 1\), and solve for \(\zeta_E\) and \(\zeta_D\) later; the transformation is then

\[
\zeta = \zeta_E e^{\pi w_0}.
\] (6.32)

Now, the goal is to provide a map from the \(z\)-plane to the \(\zeta\)-plane, after which the pressures and velocities follow from (6.32). The Schwarz-Christoffel formula provides this connection [134], and for this particular problem it is given by

\[
z = f(\zeta) \equiv K \int_1^\zeta \frac{(\zeta' - \zeta_E)}{\zeta' \sqrt{(\zeta' - \zeta_D)(\zeta' - 1)}} \, d\zeta'.
\] (6.33)

The symmetry in the potential plane implies that \(\zeta_D = \zeta_E^2\), and the constant \(K\) can be derived by requiring that the velocity upstream satisfies \(d w_0/dz = -1\), or simply that

\[
\pi \zeta \frac{dz}{d\zeta} = \pi \zeta \frac{K}{\zeta} = -1,
\] (6.34)
giving \( K = -1/\pi \). The integral \((6.33)\) can now be computed in closed form and yields

\[
z = f(\zeta) = \frac{1}{\pi} \log \left( \frac{\zeta (-1 + \zeta_E)^2}{1 - 2\zeta + \zeta_E^2 + 2\sqrt{(\zeta - 1)(\zeta - \zeta_E^2)}} \right)^{-1},
\]

(6.35)

where we have selected the principle branch of the square roots and logarithm.

In order to solve for \( \zeta_E \), we now impose the barrier condition, \( f(\zeta_E) = ic \). Due to the branch structure of \((6.35)\), two solutions are given, but the relevant one is

\[
\zeta_E = \left( \frac{1 + \sin(c\pi/2)}{1 - \sin(c\pi/2)} \right).
\]

(6.36)

Notice that the barrier sizes in the physical and potential planes are now related through \( \zeta_E = e^{-\pi k} \). It remains to invert \((6.35)\), which can be simplified to

\[
\zeta = f^{-1}(z) = A(z) - B(z)\sqrt{\Delta(z)},
\]

(6.37a)

while taking the principal branch of the square root, and with components given by

\[
\Delta(z) = \left(1 - e^{\pi(z+ic)}\right)\left(1 - e^{\pi(z-ic)}\right)
\]

(6.37b)

\[
A(z) = \frac{1 - \cos(c\pi) + 2\cosh(c\pi z)}{2\left[1 - \sin(c\pi/2)\right]^2}
\]

(6.37c)

\[
B(z) = -\frac{\left(e^{-\pi z} + 1\right)}{2\left[1 - \sin(c\pi/2)\right]^2}
\]

(6.37d)

The complex potential can now be determined from \((6.32)\), or

\[
w_0(z) = \frac{1}{\pi} \log \left( \frac{\zeta}{\zeta_E} \right) = \frac{1}{\pi} \log \left[ f^{-1}(z) \left( \frac{1 - \sin(c\pi/2)}{1 + \sin(c\pi/2)} \right) \right],
\]

(6.38)

with the pressure \( p_0(x, y) = \Re(w_0) \). Curves of constant pressure and streamlines are shown in figure 6.4 for a fibre of length \( c = 0.5 \).
In computing (6.37), care must be exercised in regards to the branch structure. In particular, the square root of (6.37b), which appears in (6.37a), contains branch points at $z = \pm i(c + 2\pi m)$ for $m \in \mathbb{Z}$. These are manifestations of the discontinuity caused by the barrier, and the easiest way to compute (6.37) is to take the branch cut at $z = ic$ directly down the imaginary axis, and all other branch cuts parallel to the horizontal axis. This step allows the use of (6.37) throughout the physical channel without changing branches.

Possessing a closed-form solution for the leading-order problem is valuable for several reasons. Firstly, the asymptotics in the two limits of small and large fibres are derived easily. Deriving higher-order approximations for flows with asymptotically small geometries is not a trivial problem (see, for example, [136]), and an explicit solution can be used to work backwards to derive the subdominant contributions. Second, the numerical computations in section 6.5 are less intensive computationally once the most singular impermeable behaviour has been subtracted out.

### 6.4.2 The small fibre limit ($c \to 0$)

In the limit that $c \to 0$, the flow in the outer region, with $x, y = \mathcal{O}(1)$ fixed and independent of $c$, simply tends to uniform flow. From the exact solution (6.38), we can expand for an outer solution

$$w_{0,\text{out}} = -z - \left(\frac{\pi}{4} \coth \left(\frac{\pi z}{2}\right)\right) c^2 + \mathcal{O}(c^4).$$

(6.39)

The outer solution does not satisfy the fibre permeability condition given by (6.27) to leading order, $\frac{\partial p_0}{\partial x} = 0$, so we search for an inner region where the appropriate scaling is

$$z = cZ \quad \text{and} \quad w_0 = c W_0.$$  

(6.40a, b)
Within the inner region, the problem for $W_0$ consists of solving for potential flow in the upper-half $Z$-plane, with an impermeable slit along $0 < Y < 1$. From the exact solution we have

$$W_0 = -\sqrt{Z^2 + 1} - \left(\frac{\pi^2}{24}\sqrt{Z^2 + 1}\right)c^2 + \mathcal{O}(c^4), \quad (6.41)$$

for which the leading-order term can be seen to match the uniform flow as $|Z| \to \infty$.

In fact, this term is simply the solution for flow past a barrier of length 1 in a semi-infinite plane, and can be derived in the usual way using conformal mapping (c.f. p. 355 in [135]). We may combine (6.39)–(6.41) in order to form a uniformly valid solution $w_{0,\text{unif}} \sim -\sqrt{z^2 + c^2}$, corresponding to a pressure field

$$p_{0,\text{unif}}(x, y) \sim -\Re\left(\sqrt{(x + iy)^2 + c^2}\right). \quad (6.42)$$

**Pressure field for large fibres ($c \to 1$)**

If we write $\gamma = (1 - c)$ and take the limit of large fibres, $c \to 1$, so that $\gamma \to 0$, then at leading order we have flow through a small slit which produces a unit flux. Away from the slit, and in a region where $x$ and $1 - y$ are $\mathcal{O}(1)$, we have from (6.38) an expansion for the potential,

$$w_0 = \frac{2}{\pi} \log \gamma - \frac{1}{\pi} \log \left[\frac{8}{\pi} \left(1 + \cosh(\pi z)\right)\right] + \mathcal{O}(\gamma^4). \quad (6.43)$$

The first term indicates that the pressure is logarithmically divergent everywhere within $\mathcal{O}(1)$ distances in the bulk of the flow, and the second term corresponds to the problem of flow produced in a channel due to a source or sink at a point along the channel wall (c.f. p. 272 in [134]). However, there are two relevant regions where the approximation in (6.43) breaks down, and these regions are marked by locations where the second term becomes of the same order of magnitude as the first.
There is an inner region near the slit where the relevant inner variable, \( Z \), is defined through \( z = i[1 - (1+Z)\gamma/2] \), with an inner solution \( w_{0,\text{in}} = -\frac{1}{\pi} \log[\pi(1+Z)^2] + \mathcal{O}(\gamma^2) \), and a corresponding inner pressure field

\[
p_{0,\text{in}} = -\frac{1}{\pi} \Re \left( \log \left[ \frac{4\pi}{\gamma^2} (1 + ix - y)^2 \right] \right) + \mathcal{O}(\gamma^2). \tag{6.44}
\]

This inner solution indeed satisfies the tip condition \( w_{0,\text{in}} = 0 \) as \( Z \to 0 \), and as \( Z \to \infty \) it re-produces the source term seen in (6.43). In addition to the inner scaling, there is a far-field scaling in (6.43), which is required in order to match the uniform flow. This occurs when \( z = \mathcal{O}(\log \gamma) \), and the second term in (6.43) becomes of leading-order.

### 6.4.3 Predictions of tip deflection

Having derived the leading-order approximation of the potential, \( w_0 \), which yields the leading-order pressure field, we may now integrate the Euler-Bernoulli equation (6.24) four times to obtain the deflection of the beam. Once the boundary conditions at the bottom and top of the fibre are imposed, the expression becomes

\[
u(y) \sim \epsilon \int_0^y \int_c^c \int_c^y \int_0^{y_4} 2p_0(x = 0, y_1) \, dy_1 \, dy_2 \, dy_3 \, dy_4, \tag{6.45}
\]

with the lower limits of integration having been chosen to satisfy the boundary conditions automatically. For small fibres, the pressure along the left-hand side of the fibre is \( p_0 \sim c^2 - y^2 \) from (6.42). Integrating this function in (6.45), we obtain an expression for the maximal deflection for \( c \ll 1 \),

\[
u(c) \sim \epsilon c^5 \left( \frac{\pi}{16} - \frac{2}{45} \right) \simeq 0.15 \epsilon c^5. \tag{6.46}
\]
Figure 6.6: The solid line represents the deflection of the fibre tip, $u_1(c) \sim u(c)/\epsilon$, calculated from (6.45) using the full solution of $p_0$. As $c \to 1$, $u_1(c)$ diverges logarithmically according to (6.47), shown as a dotted line. In the inset the full solution is again plotted as a solid line, and the $c \to 0$ limit from (6.46) is shown dashed.

In the large fibre limit discussed in section 6.40 with $c \to 1$, the pressure is logarithmically large on the scale of the fibre. Using (6.43), we find that the deflection of the fibre tip is governed by

$$u(c) \sim \epsilon \left(-\frac{1}{2\pi} \log(1 - c) + \Lambda\right),$$

where the constant is numerically determined to be $\Lambda \approx -0.176$. The solutions $u/\epsilon \sim u_1$, are shown in figure 6.6. At large and small $c$ the analytical limits, (6.46) and (6.47), closely approximate the full solution (6.45).

Effectively, (6.46) implies that within the small fibre limit, the correct scale of deflection is $\mathcal{O}(\epsilon c^5)$ instead of $\mathcal{O}(\epsilon)$. Consequently, as $c \to 0$, $\epsilon$ is allowed to be quite large (i.e. $\epsilon \ll 1/c^5$), rather than the more strict $\epsilon \ll 1$. Thus, in this limit the fibre can be allowed to be increasingly flexible and the flow rate can be increasingly high without violating the requirements of the linearised theory. Furthermore, because of the $c^5$ dependence, in order to match the observed $\mathcal{O}(1)$ deflections in the experiments (where $c \approx 0.3\text{–}0.8$), $\epsilon$ must in fact be quite large. In the case of large fibres, $c \to 1$, however, the bending parameter must satisfy the rather stringent requirement of
$\epsilon \ll 1/|\log(1 - c)|$. If the fibre is too large, the theory breaks down unless the fibre is kept sufficiently stiff or the flow rate is kept sufficiently low.

### 6.4.4 Predictions of leakage

We can also develop an approximation for the amount of fluid flowing through the fibre, which can be measured using the horizontal velocity at $x = 0$. The velocity is proportional to the pressure gradient, and as we showed in (6.28) and (6.30), the pressure gradient depends on the relative balances of permeability and fibre bending, and can be described in the two limits as

$$v_x/\beta \sim \mp 2p_0 \bigg|_{x=0^\pm} \quad \text{for } \beta \gg \epsilon \quad (6.48)$$

$$v_x/\epsilon \sim -\left( \frac{du_1}{dy} \frac{\partial p_0}{\partial y} - u_1 \frac{\partial^2 p_0}{\partial x^2} \right)_{x=0^\pm} \quad \text{for } \epsilon \gg \beta, \quad (6.49)$$

where $v_x$ is scaled by the average velocity in the channel, $Q/DH$. From (6.48), we see that when the leakage at the fibre is primarily due to permeability, i.e. $\beta \gg \epsilon$, the largest leakage occurs near the origin at the base of the fibre where the pressure is maximal. The least leakage is near the fibre tip where the pressure is zero. In contrast, when fibre flexion governs $v_x$, i.e. $\epsilon \gg \beta$ in (6.49), then the local leakage is zero at the fibre base and large at the fibre tip. At the tip, the second derivative of the leading-order pressure is infinite, and the fibre is bent, allowing the fluid to pass. Both results are plotted in figure 6.7.

There is one unusual element of the horizontal velocities. Recall that $p_0$ is anti-symmetric, so for $\beta \gg \epsilon$, we see from (6.48) that the velocity is both continuous and positive at the fibre as expected. However, for $\epsilon \gg \beta$, we can verify that the velocity in (6.49) is positive on the left, but negative on the right. Thus, the asymptotic approximations predict a flow rightwards on the left of the fibre, but leftwards on the right. This is counterintuitive because for any non-zero amount of deflection (and
Figure 6.7: The leading-order horizontal velocity, \( v_x/\beta \), at \( x = 0_+ \) for permeability dominant flows, \( \beta \gg \epsilon \) (shown thin). Also plotted is the horizontal velocity, \( v_x/\epsilon \), at \( x = 0_- \) for a flexion dominated flow, \( \epsilon \gg \beta \), with fibre length \( c = 0.54 \) (shown bold). The horizontal velocity at \( x = 0_+ \), for \( \epsilon \gg \beta \), is the negative of the plotted value.

hence any \( \epsilon > 0 \), we would expect the flow at the origin to be both continuous and positive.

In fact, this peculiarity in our asymptotics is due to a further distinguished limit in the process of taking both \( x \to 0_+ \) and \( \epsilon \to 0 \). Notice that in our methodology [c.f. in particular (6.30)], we first took \( \epsilon \to 0 \), projected the fibre onto the axis \( x = 0 \), and only then took the limit of \( x \to 0 \). This, however, is different from taking \( x \to 0_+ \) first (to where the fluid is continuous and has positive horizontal velocity), followed by taking \( \epsilon \to 0 \).

We clarify this issue in the second Appendix, 6.9. There, the conclusion is that there exists a curved region of \( O(\epsilon) \) near the fibre where the asymptotic approximation (6.30) is invalid. The asymptotic series of (6.21) should then be interpreted as an outer approximation to be matched to an inner solution near the fibre. However, because the inner limit of the outer solution is known to all orders by (6.26), the solution in the inner region is unimportant as far as the general macroscopic flow is concerned;
Figure 6.8: Streamlines (black), and constant-pressure curves (gray), both given in increments of 0.125, of $p = p_0 + \beta p_1$ for the permeability-dominated case $\beta \gg \epsilon$ where $\beta = 0.3$. The fibre length is $c = 0.5$, and the inset plot shows the leading-order deflection of the fibre, $u \sim \epsilon u_1$.

it only serves to explain why the velocity on the right of the fibre predicted by \((6.30)\) is negative rather than positive.

6.5 Numerical results

In this section we present numerical computations for the first-order pressure, $p_1$, of section 6.4. In addition, we present a numerical method for solving the system of equations \((6.19)\) with arbitrary degrees of permeability. An eigenvalue decomposition at $x = 0$ is used to calculate $p_1$ in the permeability dominated case ($1 \gg \beta \gg \epsilon$), and for the case of arbitrary permeability ($\beta = \mathcal{O}(1), \epsilon \ll 1$). However, when flexion dominates ($1 \gg \epsilon \gg \beta$), the $p_1$ calculations necessitate a global scheme, and the difficulties of such a scheme are discussed. A coupled finite-element package may be appropriate for this case.

6.5.1 Permeability dominated flows \((\epsilon \ll \beta \ll 1)\)

In flows where the permeability dominates, the majority of the flux past the fibre in the three-dimensional problem can be attributed to the flow of fluid going through
the gap above and below the fibre, rather than due to the fibre bending. The object is to calculate $p_1(x, y)$ subject to satisfying the Laplace equation within the channel, no-flux conditions at the upper and lower walls, with $p_1 \to 0$ as $|x| \to \infty$, and the conditions $\text{(6.28)}$ and $\text{(6.29)}$ at $x = 0$. We expand $p_1$ into a truncated Fourier series with $M$ modes

$$p_1(x, y) = \frac{b_0}{2} + \sum_{m=1}^{M-1} b_m e^{-m\pi|x|} \cos(m\pi y), \quad (6.50)$$

with eigenfunctions chosen so as to satisfy all conditions on $p_1$ except for those at $x = 0$. The unknown coefficients $b_m$ are then obtained by imposing the two boundary conditions along $x = 0$,

$$\sum_{m=1}^{M-1} (m\pi)b_m \cos(m\pi y) = -2p_0(x = 0, y) \quad \text{for } 0 \leq y < c, \quad (6.51a)$$

$$\frac{b_0}{2} + \sum_{m=1}^{M-1} b_m \cos(m\pi y) = 0 \quad \text{for } c \leq y < 1, \quad (6.51b)$$

where $p_0$ is computed using $\text{(6.38)}$.

The theory of mixed boundary value problems, studied using a dual series formulation such as this one, is discussed in [137]. We solve for the coefficients $b_m$ using collocation: we distribute $M$ points along the interval, $y \in [0, 1]$, and solve the resultant $M \times M$ system of equations using Newton’s method. Generally, the solution converges rapidly, and only a modest number of uniformly distributed mesh points are required ($M = 50$ for most computations).

In figure $6.8$, the streamlines of the two-term approximation, $p \sim p_0 + \beta p_1$ are drawn for the case of permeability dominated flows with $\beta = 0.3$. The ability for the model to capture a small degree of leakage past the fibre is evident. We have chosen a relatively large value of $\beta$ simply to emphasise the influence of leakage, and the first-order streamlines ($p_1$-streamlines) clearly show passage through the fibre. As $\beta$ tends to zero, the majority of the streamlines that pass through the fibre are seen to
Figure 6.9: Streamlines (black), and constant pressure lines (gray), both given in increments of 0.125, of the numerical solution for $\beta = \mathcal{O}(1)$ and $\epsilon \ll 1$, from section 6.5.2. The fibre length is $c = 0.5$, and permeability $\beta = 2$. The inset plot shows the leading-order deflection of the fibre, $u \sim \epsilon u_1$.

be localised near the origin. At this point $v_x / \beta$ given in (6.48) is maximised, and the first-order contribution is sufficiently large to divert the flow through the fibre, rather than simply around the structure.

6.5.2 Small deflection of fibres with arbitrary permeability

$$(\epsilon \ll 1, \beta = \mathcal{O}(1))$$

Though up to now our analysis has been focused on the limit of small permeability, our model in (6.19) is also valid for arbitrary permeability. If we let $\beta = \mathcal{O}(1)$ but keep $\epsilon \ll 1$, then we can treat the problem using the boundary perturbation method only, expanding $p$ and $u$ explicitly in terms of $\epsilon$ as $p = p_0 + \mathcal{O}(\epsilon)$ and $u = \epsilon u_1 + \mathcal{O}(\epsilon^2)$.

We emphasise that in this new asymptotic framework the leading-order pressure, $p_0$, and deflection, $u_1$, both include effects of permeability.

Since we are still concerned with small deflections, $p_0$ is again assumed to be anti-symmetric about $x = 0$. This allows us to extend the condition of $p_0 = 0$ throughout the gap $y \in (c, 1)$ at $x = 0$, as in section 6.4. The boundary condition at the fibre
(6.19e) becomes at leading order
\[ \frac{\partial p_0}{\partial x} \bigg|_{x=0} = 2\beta p_0 \bigg|_{x=0^+} = -2\beta p_0 \bigg|_{x=0^-}, \] (6.52)
and to solve for \( p_0 \) we choose to write,
\[ p_0 = \phi + \tilde{p}_0, \] (6.53)
where we have separated \( p_0 \) into an approximate solution \( \phi \) and a correction \( \tilde{p}_0 \).

When \( \beta \) is small and the bulk of the fluid is deflected around the fibre, we take \( \phi \) to be the leading-order impermeable solution in section 6.4.1. However, if \( \beta \) is larger, and particularly in the limit that \( \beta \to \infty \), the boundary condition along the fibre becomes \( p_0 \to 0 \) at \( x = 0 \), and we expect that the fluid completely passes through the fibre; the leading-order approximation suggests that we choose \( \phi = -x \).

In either case, if we use the Fourier series representation of (6.50) for \( \tilde{p}_0 \), the boundary condition at the fibre (6.52) becomes
\[ (-2\beta) \frac{b_0}{2} + \sum_{m=1}^{\infty} (-m\pi - 2\beta) b_m \cos(m\pi y) = 2\beta \phi - \frac{\partial \phi}{\partial x} \quad \text{for } 0 \leq y < c, \] (6.54)
and is combined with (6.51b) for \( c \leq y < 1 \). The right-hand side of (6.54) is evaluated as \( x \to 0^- \), and is equal to 1 when we choose \( \phi = -x \).

The equation for the deflection is the same as (6.24), and the leading-order deflection \( u_1 \) is numerically integrated with (6.45). We emphasise again that in this case \( p_0 \) contains effects of leakage and therefore the prediction \( u_1 \) is based on leakage effects as well. An example of the numerical computation of \( p_0 \) and \( u_1 \) is shown in figure 6.9 with permeability \( \beta = 2 \) and fibre length \( c = 0.5 \). It should be apparent that the large value of \( \beta \) has allowed the fluid to pass nearly unhindered by the fibre, and thus the streamlines closely approximate those for uniform flow.
6.5.3 Flexion dominated effects \((\beta \ll \epsilon \ll 1)\)

Returning to the asymptotic formulation presented in section 6.4, we examine the first-order correction to the pressure field in the flexion dominated regime. The difficulty of numerical computations in this regime is that we no longer know the value of \(p_1\) on \(x = 0\) and \(c < y < 1\) [c.f. for comparison (6.51b)]. Thus, the dual series approach, which effectively reduces the two-dimensional problem in the channel to a one-dimensional problem along \(x = 0\), no longer applies for this case.

Numerical computations in this regime require a global method. For example, we may attempt to compute \(p_1\) using a boundary integral approach [138, 139]. However, there is a significant difficulty towards implementing this approach (and indeed, any global solver): from the discussion in section 6.4.3 we know that the normal derivative of \(p_1\), given by (6.30), contains a non-integrable singularity at the point \((0, c)\). In fact, our asymptotic analysis in section 6.4 can be used to simplify these numerical schemes by allowing us to remove the dominant singularity at the fibre tip. However, implementing these numerical methods would take us beyond the scope of this chapter, and we have chosen to leave such a scheme to future work.

6.6 Discussion

To assess the validity of the leading-order model presented in this work, we may compare the predictions of fibre deflection to the experimental observations of section 6.2. The basis of our theoretical treatment is that the small deflection follows \(u \sim \epsilon u_1\), and we showed how to calculate \(u_1\) as a function of the dimensionless fibre height \(c = h/H\) using (6.45), i.e. \(u_1(c)\). Substituting the definitions of dimensionless \(u\) (6.18) and \(\epsilon\) (6.20c) into the asymptotic form, \(u \sim \epsilon u_1\), we can write the dimensional deflection \(u\) as

\[
    u \sim \left[ \frac{6\mu(D + d)H^4Q}{EI_D^3} \right] u_1(c), \quad (6.55)
\]

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Figure 6.10: (a) Experimental deflection data measured from one particular fibre, with \(w = 22 \, \mu m\), \(h = 241 \, \mu m\), \(d = 56 \, \mu m\), and \(D = 66 \, \mu m\). The solid line represents the slope of \(u\) versus \(Q\) as \(Q \to 0\) for this fibre, fitted from the first five data points. (b) Data points show values of \(u_1\) for all experiments (determined from measured \(u/Q\) and average \(E\)), along with theoretical predictions given by solid and dashed lines. In the experiments \(w\) varies from 22 to 34 \(\mu m\), but the form of \(u_1\), (6.56), scales out this dependency. The horizontal error bars are smaller than the data points. The two middle points are crossed to highlight them for discussion in the text. Values of \(\beta\) lie between 0.0179 and 0.0226.

which gives a prediction for the slope of \(u\) versus \(Q\), for small \(Q\). We now rearrange this equation to compare physical quantities on the right to a calculated quantity, \(u_1(h/H)\), on the left, and obtain

\[
u_1(c) \sim \left[ \frac{E (\frac{1}{12} du^3) D^3}{6 \mu (D + d) H^4} \right] \frac{u}{Q}, \tag{6.56}\]

where we have substituted \(I\) from (6.17). Thus, \(u_1\) is essentially a scaling of \(u/Q\), and should collapse the data for different geometries (different \(w\)) so that it is only a function of \(h/H = c\).

6.6.1 Comparison to experiments

We now seek to validate (6.56): first, we determine the initial slope \(u/Q\) for an individual fibre from the recorded deflection data by fitting a least-squares linear
regression to the data points at low flow rates. One data set, with its linear regression, is shown in figure 6.10a.

Now all terms in (6.56) are known except for the Young’s modulus, $E$. Most importantly, we know the ratio $c = h/H$, and therefore we may calculate a prediction for $u_1(c)$ using (6.45). We thus solve for the unknown $E$ in (6.56) for each experiment, using the measured slope $u/Q$ and the prediction $u_1(c)$. We then average $E$ over all experiments to obtain $E = 63 \pm 22$ kPa. An additional benefit of this experimental setup then is that it allows us to measure the elasticity of PEGDA polymerised with these specific cross-linking conditions, which we believe had not been done before.

We may now determine experimental values of $u_1$ from the expression on the right-hand side of (6.56), using the average value of $E = 63$ kPa, and the measured values of $u/Q$, $w$, $d$, $D$, $H$, and $\mu$. Note that, since the average value of $E$ is used for all experiments, the effect of $E$ is only to offset all data points by a multiplicative constant; the shape of the $u_1(c)$ trend is unaffected. Recall that $w$ varies from 22 to 34 µm, and that $h$ varies from 144 to 293 µm; all other parameters are held constant. All experimental values of $u_1$ are plotted as a function of $c$ in figure 6.10b, with the dependency on $w$ scaled out through (6.56).

The leading-order solution $u_1(c)$, computed using (6.45), is shown alongside the data in figure 6.10b for comparison. The analytical solution for small $c$, given by (6.46), is presented as well. Error bars in the plot are determined from estimated variances in the parameters involved in calculating $u_1$, as well as the variance in $E$ reported above, and the error in calculating the regression for the initial slope of $u$ versus $Q$. (Student’s $t$ test, with 95% confidence). The permeability parameter, $\beta$, ranges from 0.0179 to 0.0226, and since $\beta$ is small we expect the experimental values of $u_1$ to lie close to the leading-order solution.

Indeed, the data appears to agree well with the trend $u_1(c)$ calculated from (6.45), though there exists some scatter. We can posit various reasons for the observed
scatter, with perhaps the most likely being variations in fibre elasticity. Great care was taken to ensure constant polymerisation conditions, but this is a relatively new experimental technique, and reproducibility issues have been raised before [122]. The Young’s modulus $E$ could vary from fibre to fibre and we have no direct way of measuring this on a specific fibre; instead we average over the suite of experiments to obtain a value of the Young’s modulus with variances. Variations in Young’s modulus are most apparent in the two “crossed” data points in figure 6.10b. The geometry of these experiments is nearly identical, but they lie on opposite sides of the predicted trend, indicating the presence of some unobservable difference between the two fibres. Another possible reason for the scatter is difficulty in measuring the fibre depth, $d$.

The model is able to predict the deflection within reasonable error bounds for much of the dataset however, and this resolution is likely good enough for one application of our technology: microfluidic flow measurement. Given a microfluidic channel of a certain geometry, and a microscope capable of sensing deflections at a certain resolution, our model can be used to specify the size of the sensing fibre necessary for a range of expected flow rates. Then, as long as the flow remains in the linear regime, only one calibration experiment is necessary to precisely calibrate flow measurement.

6.7 Conclusion

In this chapter we presented an experimental and theoretical study investigating the dynamics of confined fibres bent by an external viscous flow. Using a novel microfluidic setup, we constructed flexible micron-sized fibres anchored in a channel, and performed experiments measuring tip deflection versus flow rate. We proposed a mathematical model that reduced the three-dimensional geometry of the flow to a two-dimensional Hele-Shaw approximation. Within the two-dimensional approximation,
boundary conditions at the fibre allowed for leakage flow through the small gap above and below the fibre, giving the confined fibre an effectively permeability.

Motivated by the experimental observation of a linear relationship between deflection and flow rate for highly confined fibres at low flow rates, asymptotic solutions were sought in the limits of small flexion ($\epsilon$) and small permeability ($\beta$). The leading-order pressure field, $p_0$, was derived in closed form using complex variable methods. It was shown that there exist two distinguished limits for the first-order pressure correction, $p_1$, and results for these two limits are summarised in table 6.1. A similar problem formulation was then used to numerically solve for the pressure field corresponding to arbitrary degrees of dimensionless permeability, $\beta$.

We use the leading-order pressure field, $p_0$, to predict the leading- and first-order deflection of the fibre, and validate our calculations with analytical limits at small and large fibre heights. The predicted deflection compares favourably to the results of our microfluidic experiments, and allows us to measure the elasticity of PEGDA polymerised with our specific cross-linking conditions. We note that there still exist open questions to be studied with the tools presented in this chapter. Further work could be done investigating the regime of large fibre deflections, the dynamics of a system with multiple fibres, and the effects of channel elasticity.
6.8 Appendix: Derivation of beam bending equation

In this appendix we derive the simplified beam bending equation (6.16). We define the force per unit length acting on a fibre as \( f = f_s e_s + f_n e_n \), and the force and moment transmitted through the fibre as \( T = T_s e_s + T_n e_n \) and \( M \) respectively. The unit vectors \( e_s \) and \( e_n \) are in the tangential and normal directions. We perform a force and moment balance on an infinitesimal section of the fibre, while specifying inextensibility, to obtain

\[
\frac{dT}{ds} + f = 0 \quad \text{and} \quad \frac{dM}{ds} + T_n = 0. \tag{6.57a, b}
\]

Assuming that the moment is tied to the curvature through \( M = EI \kappa \), we arrive at a set of equations to describe the deflection of the fibre through a balance of forces in the two directions,

\begin{align*}
\text{e}_s \text{ component :} & \quad \frac{dT_s}{ds} + EI \kappa \frac{d\kappa}{ds} + f_s = 0 \quad \tag{6.58a} \\
\text{e}_n \text{ component :} & \quad -EI \frac{d^2\kappa}{ds^2} + \kappa T_s + f_n = 0. \quad \tag{6.58b}
\end{align*}

Recall that \( f_n \) is related to the pressure drop across the fibre and is given by (6.15). The tangential force, \( f_s \), can be estimated to be \( (\mu \frac{\partial v_s}{\partial n}) d \), where \( v_s \) is the component of velocity locally parallel to the fibre. Hele-Shaw theory does not allow for the no-slip condition to be satisfied at the face of the fibre, so the gradient \( \frac{\partial v_s}{\partial n} \) exists to decrease the velocity from a bulk value to zero. [133] shows that for simple Hele-Shaw problems there is an inner region the size of the channel depth \( D \) where this transition takes place, but for our problem we estimate that it should depend on the depth of the fibre \( d \) instead. Thus, we chose \( d \) as the size of the inner region,
setting the scale for the denominator of the gradient, and taking the average velocity \( Q/DH \) as a scale for \( v \) we estimate that \( f_s \sim \frac{\mu Q}{DH} \).

To simplify (6.58), we nondimensionalise and drop the smallest terms. The pressure and all lengths (including \( \kappa^{-1} \)) are nondimensionalised according to (6.18). The tangent force \( f_s \) is nondimensionalised by the scale in the previous paragraph. The scale of \( T_s \) is set by a balance between the first and second terms of (6.58a), with \( T_s \sim EI/H^2 \). If instead the scale of \( T_s \) had been chosen to balance the first and third terms, with \( T_s \sim \mu Qd/D^2 \), there would be an inconsistent balance, with the second term being \( \mathcal{O}((\epsilon D/H)^{-1}) \gg 1 \). Keeping the same names for dimensionless quantities, we have

\[
\begin{align*}
\textbf{e}_s \text{ component } : & \quad \frac{dT_s}{ds} + \kappa \frac{d\kappa}{ds} + \frac{D}{6(D + d)} \frac{D}{H} \epsilon f_s = 0 \quad (6.59a) \\
\textbf{e}_n \text{ component } : & \quad -\frac{d^2\kappa}{ds^2} + \kappa T_s - \epsilon [p_+ - p_-] = 0. \quad (6.59b)
\end{align*}
\]

The Hele-Shaw approximation is based on the assumption that \( D/H \ll 1 \), so we drop the last term in (6.59a). We now integrate this equation in \( s \) and set the integration constant equal to zero since the end of the fibre has no applied tension \( (T_s = 0) \), and no applied moment \( (M = \kappa = 0) \). Substituting into (6.59b), we have reduced the system to one ODE,

\[
\frac{d^2\kappa}{ds^2} + \frac{1}{2} \kappa^3 = -\epsilon [p_+ - p_-]. \quad (6.60)
\]

This equation is re-dimensionalised to arrive at (6.16).
Figure 6.11: Streamlines $\Im(w)$, accurate to $O(\epsilon^2)$, and given in increments of 0.1, for a vertical fibre at $x = \epsilon = 0.25$, with $c = 1$ in an unbounded half-plane. The shaded portion denotes the inner region. The inset shows the pressure $p(x, 0.25)$ with the outer solution (thin), and the inner solution (bold gray).

6.9 Appendix: Non-uniformity of limits and fibre tip divergence

In the asymptotic analysis of section 6.4.3, it was observed that the first order correction predicts that the horizontal fluid velocity is positive on the left of $x = 0$ but negative on the right. We explained that this is an effect of the distinguished nature of the $x \to 0$ and $\epsilon \to 0$ limits. In this section, we clarify this point, and also comment on the divergence of the asymptotic approximations near the fibre tip, which is an important issue for the numerical computations in section 6.5.

It suffices to study a simpler problem: consider flow in the upper half-plane past an infinitesimally thin vertical barrier of height $y = c$ placed at $x = \epsilon$. The exact solution is known through conformal mapping (in fact, this is simply the solution (6.41) shifted in $x$ by an amount $\epsilon$). The complex potential, with $w(z) = p(x, y) + i\psi(x, y)$ is given by

$$w(z) = A\sqrt{(z - \epsilon)^2 + c^2}, \quad (6.61)$$
with $A = 1$ for upstream ($x < \epsilon$) and $A = -1$ for downstream ($x > \epsilon$). Assuming that $z$ is held fixed, we expand (6.61) to obtain an outer expansion for the potential,

$$w_{\text{out}}(z) = A_0 \left( \sqrt{z^2 + c^2} - \epsilon \frac{z}{\sqrt{c^2 + z^2}} + \epsilon^2 \frac{c^2}{2(z^2 + c^2)^{3/2}} + O(\epsilon^3) \right),$$

(6.62)

with now $A_0 = \pm 1$ depending on $x \lesssim 0$.

As mentioned in section 6.4.3, the asymptotic approximation (6.62) predicts a discontinuous velocity at $x = 0$, and this is due to the fact that when we expand (6.61) to obtain (6.62), we fix $z = O(1)$, take $\epsilon \to 0$, and then only afterwards take the limit of $\Re(z) \to 0$ in (6.62) to derive the value along $x = 0$. This is different to taking $x \to 0$ first, and then afterwards expanding for small $\epsilon$.

The key is to introduce a boundary layer of size $\epsilon$ near $x = \epsilon$. Near the barrier, we introduce the re-scaled coordinate $X = (x - \epsilon)/\epsilon$, and from (6.61), we obtain

$$w_{\text{in}}(X, y) = B \left( \sqrt{c^2 - y^2} + \epsilon \frac{iXY}{\sqrt{c^2 - y^2}} + \epsilon^2 \frac{c^2X^2}{2(c^2 - y^2)^{3/2}} + O(\epsilon^3) \right),$$

(6.63)

where $B = \pm 1$ for $X \lesssim 0$. Thus, when the value of $y$ is fixed with $0 < y < c$, we use $\Re[w_{\text{out}}]$ for the pressure when $x < 0$, $\Re[w_{\text{in}}]$ from $0 < x < 2\epsilon$, and again $\Re[w_{\text{out}}]$ from $x > 2\epsilon$. Any multiple of $\epsilon$ is valid since such boundary layers are only defined within orders of $\epsilon$. An example of such an approximation, accurate to $O(\epsilon^2)$ in each region, is shown in figure 6.11. There, we see that the introduction of the inner region is necessary to ensure that the streamlines are continuous across the origin.

Note that the introduction of the inner region on the right of the fibre is more for aesthetic reasons, because at higher orders the outer approximation (6.62) for $x > 0$ will begin to ‘form’ the fibre at $x = \epsilon$. However, for all finite truncations of the series, the velocity at $x = \epsilon$ will continue to be non-zero. The inner region to the right of the fibre provides us with the ability to achieve a zero velocity at $x = \epsilon$ using a finite asymptotic approximation.
Figure 6.11 also makes it apparent that an additional matching region must be imposed along the path of the tip of the fibre, \( z = x + ic \) for \( 0 < x < \epsilon \), and the worst of this behaviour is seen at the point \( z = ic \), where the outer approximation \( w_{\text{out}} \) in (6.62) is clearly divergent. Of course, these are all the inherent caveats of using a (singular) asymptotic methodology that depends on shifting the geometry of the problem.

The full problem of section 6.4 is different in that it requires an upper channel wall at \( y = 1 \), and also the fibre position is no longer fixed at \( x = \epsilon \), but is rather at \( x = u(y) = \epsilon u_1 + \epsilon^2 u_2 + \ldots \). Consequently, the exact solution is unknown, and we must derive the inner and outer solutions term by term, carefully matching at each order.

However, the qualitative conclusions we have reached for the model problem remain valid for the full problem: the outer asymptotic expansion (6.21) is invalid within a boundary layer of \( \mathcal{O}(\epsilon) \) near the barrier, and this explains the odd velocities when taking \( x \to 0 \) in (6.49). The inner solution near the fibre can be determined by re-scaling near the fibre, and matching outwards to the inner limit of the outer solution. However, because the fibre is deflected according to (6.45), we assume that this inner problem would need to be determined numerically.
Chapter 7

Conclusions and Future Work

Due to the disparate nature of the content presented in this thesis, overarching conclusions for the thesis as a whole will not be presented. Rather, this chapter will be devoted to conclusions drawn from the work on liquid-infused surfaces that was presented in Chapters 1-3. The remainder of the chapters contain standalone conclusions.

In Chapter 1 a study was presented on the behavior of liquid-infused surfaces subject to an external flow. It was shown that the shear stress from the flow acts to drain the infused liquid from a portion of the surface, while allowing the remainder of the surface to remain fully wetted. This drainage behavior limits the applications where liquid-infused surfaces may be used, much how the pressure-induced failure mode of conventional gas-cushioned superhydrophobic surfaces limits applications of those surfaces.

However, since a finite length of any textured surface will remain wetted indefinitely, each wetted surface has a threshold shear stress below which no infused oil will drain. If a surface is to be used above that threshold shear stress, the texture must be modified in order to ensure that oil is retained. One way to modify the texture would be to introduce physical barriers that run in the spanwise direction, separated by a
streamwise distance that is less than the stable length of oil that can be retained at a
design-limited shear stress. However, the drainage behavior of a sectioned surface like
this would be modified from that presented in Chapter 1 since the wetted sections
of such a surface would by necessity end in a solid barrier rather than the reservoir
end-condition that was investigated in Chapter 1.

To understand how barriers affect drainage, in Chapter 2 we present experiments
and theory exploring the drainage behavior of dead-end grooves exposed to an external
flow. The downstream barriers act to increase the stable wetted length by allowing a
positive Laplace pressure to develop within the grooves. It is found that this increased
length is metastable, and the grooves overflow above a certain shear-stress threshold,
causing the wetted length to decrease. After overflowing, however, the wetted length
remains slightly longer than would be expected from the theory in Chapter 1 because
a Marangoni stress develops due to trace surfactants in the flow. Based on the results
from this chapter, physical barriers appear to be a viable option for preventing oil
drainage due to shear.

Creating precise physical topology on a rough surface is difficult and costly, so in
Chapter 3 we explore how chemical patterning can also be used to prevent infused
liquid from draining, both from shear as well as from gravity. By creating stripes of a
chemistry that is preferentially wet by the external fluid rather than the infused fluid,
we disrupt the continuity of the infused fluid and prevent it from draining. These
sacrificial regions perform similarly to the physical barriers investigated in Chapter
2.

Thus, in Chapters 1–3 of this thesis, the shear-driven failure mode of liquid-infused
surfaces is introduced and quantified, and then mitigated through implementation of
physical and chemical patterning. The work presented here will allow liquid-infused
surfaces to be used in scenarios where they would otherwise be unstable, enabling
many more applications that leverage the advantages of these surfaces. However,
questions still remain with regards to the failure of liquid-infused surfaces and similar problems, so I end with a brief list of some directions for future study:

- **Complex surface geometries** – The research presented here focused mainly on longitudinal grooves as a model texture for studying drainage of liquid-infused surfaces. In Chapter 1 it was briefly shown that a surface consisting of randomly placed posts drains in a similar manner, but the differences were not quantified. Hence, a direction for future research would be to develop a theoretical understanding of the length- and time-scales that are relevant to non-groove geometries.

A further step would be to consider surface textures with varying depth, since both the grooves and the randomly placed posts are essentially two-dimensional geometries of constant depth. A random rough surface that is generated with a scalable process such as abrasion or electrodeposition would have features of varying depth, so it would be desirable to gain an understanding of how such features would effect the drainage dynamics. To start with, experiments could be performed on streamwise grooves of triangular cross-section, since this geometry may capture the key features of a non-constant depth but still be conducive to theoretical analysis. These grooves could be fabricated either by etching silicon with a solution of potassium hydroxide, or by molding from commercially available prismatic film. One interesting feature of triangular-grooved surfaces is that the Laplace pressure within the groove approaches infinity as the meniscus retreats further into the corner of the groove. Because of this feature, the capillary rise height on such a surface is predicted to be arbitrarily large [140].

- **Liquid-infused surfaces exposed to turbulence** – This thesis focused on the behavior of liquid-infused surfaces subject to laminar flow. In a variety
of scenarios, where liquid-infused surfaces are used for their drag reducing or anti-biofouling properties, the surfaces may also be exposed to turbulent flow. The pressure and velocity fluctuations that are present in turbulent flow may alter the drainage behavior of a liquid-infused surface from the laminar case. The response of a surface to spatial and temporal fluctuations in pressure and velocity will probably depend on the structure of the surface as well as the viscosity ratio between the two phases. It may be possible to design a surface to be resistant to specific types of fluctuations.

- **Shear-driven failure of superhydrophobic surfaces** – Conventional gas-cushioned superhydrophobic surface have been shown to fail due to applied pressure in a variety of scenarios, from drop impact to turbulent pressure fluctuations. However, a superhydrophobic surface with interconnected gas cavities will probably fail under shear as well, since the gas that is trapped in the surface is still a movable fluid. Since the ratio of viscosities will be reversed from that which is considered here, the analysis will need to be redone. In a theoretical analysis of this problem, the velocities of the two phases will need to be matched at the interface, and matching of shear stress at the interface will be less critical.

- **Surfactant effects on drag reduction** – In Chapter 2 it was shown that a portion of a liquid-infused surface can be rendered immobile due to Marangoni effects that are related to gradients in surfactant concentration. A pressing question, then, is whether this phenomena will adversely effect the ability of a liquid-infused surface to reduce drag. If drag reduction is caused by finite-slip at the liquid-liquid interface, an immobile surface may not reduce drag.

- **Thermal Marangoni flow on liquid-infused surface** – A final direction for future study is to explore the dynamics of thermal-driven Marangoni flow on liquid-infused surfaces. This type of flow would occur if a temperature gradient
is imposed along the length of a liquid-infused surface: the region of the surface that is at a higher temperature will have a lower surface tension, so the infused fluid will tend to flow away from that region.

If hot and cold regions are separated by a distance, $L$, and are held at temperatures that impose a difference in surface tension, $\Delta \gamma$, the resulting Marangoni stress will be $\tau = \Delta \gamma / L$. Now, using the theory that was developed in this thesis, the steady-state length of the surface due to the Marangoni stress is predicted to be $L_\infty \sim \gamma / \tau \sim L \gamma / \Delta \gamma$. Since $\Delta \gamma < \gamma$ by necessity, we predict that $L_\infty > L$, meaning that a liquid-infused surface that is subjected to a thermal Marangoni stress may never dewet. It is likely that this will be true for certain surface textures but not true for others, due to varying values of the numerical prefactor.
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