

# Analysis, Computations, and Experiments on Pinch-Off in Liquid Jets (05frg060)

March 12 – 26, 2005

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## Introduction

This Focussed Research Group brought together a critical mass of researchers to work on fundamental problems that involve the breakup of liquid jets and on fluid and fluid jet problems that are motivated by industrial applications. Recent theoretical advances in the understanding of the breakup of single fluid jets are ripe to translate the control of breakup of jets. We also used concrete mathematical models to investigate utilizing liquid jet phenomena in the manufacture of micro- and nano-scale structures. Significant work remains to be done in the modeling and analysis of jets with more complicated geometries (e.g., compound jets) and involving complex fluids, which are typically found in industrial applications.

The FRG included applied mathematicians involved in modelling and asymptotic analysis in fundamental problems (Papageorgiou, Siegel, Howell, Young) as well as more applied problems motivated by industrial applications (Huang, Miura, Wylie), and a physicist with expertise in modelling and numerical simulation (Zhang). Many of the program participants are internationally known for their contributions to interfacial fluid dynamics.

## Microdroplet Formation in a Patterned Hele-Shaw Cell

Parallel submicroliter polymerase chain reactions (pcr) have been utilized for DNA diagnostic applications [14]. A lattice of wetting (hydrophilic) patches is patterned on the interior faces of two (hydrophobic) glass plates of a Hele-Shaw cell and the patterns are aligned. A liquid first fills up the cell, and then a second, immiscible fluid is used to displace the excess liquid between the wetting patches to form multiple microdroplet liquid bridges between the plates. The droplets of liquid have a thickness which is usually much smaller than the characteristic lengths of the plates.

Preliminary studies that focused on the steady configuration indicate that the dynamic aspects of the filling process may be important. For example, droplets would not form if the filling speed is too fast. Furthermore, the viscous forces between the displacing and the droplet fluids may be important in the filling process. Motivated by these important issues, during the BIRS FRG, we modelled the dynamic filling process as a pressure-driven, two-dimensional Hele-Shaw flow.

We started a preliminary investigation of solving the model equations numerically using moving boundary methods. The standard boundary integral method has been used to simulate drop dynamics due to electro-wetting in a Hele-Shaw cell [6]. However, this method cannot handle topological changes of the interface, such as during droplet formation as the interface is pushed through a wetting patch. Consequently, level set methods are used to accurately capture droplet formation with little artificial manipulation of the interface. The problem also has been reformulated using a phase field approach where the sharp interface is replaced by a thin layer characterized by an order parameter.

## Influence of Surfactant on Contact Line Stability for Coating Flows

The coating of a surface is a process of obvious industrial importance and provides strength to the surface or achieves some desired physical properties [12]. We consider the two-dimensional coating flow of a moving substrate in contact with a liquid bath (e.g., see Figure 1). Experiments show that

at sufficiently high coating speed, there is an instability of the fluid-substrate contact line, whereby a filament of air is ejected downstream into the liquid bath. This instability, which has been referred to as ‘tip-streaming’, is detrimental to the coating process.

At the BIRS FRG, we investigated the role of surfactant on tip-streaming and air entrainment during coating flows. The presence of surfactant has been shown to be important during tip-streaming for the related problem of a bubble in an imposed extensional flow, see Figure 1. Surfactant transport at a contact line between a liquid and a moving solid substrate is a fundamental problem that has received scant attention.

Unfortunately, mathematical modelling is complicated by the presence of the contact line. It is well known that imposing a dynamic contact angle other than  $\pi$  gives rise to a discontinuous velocity field at the contact line. This is accompanied by a nonintegrable stress singularity at that point, which is physically unrealistic. To avoid this difficulty, we can assume that the interface is tangential to the solid at the attachment point, i.e., the contact angle is  $\pi$ . This has the advantage that, for single fluid systems, there exist local solutions which are devoid of nonintegrable stress singularities.

In our analysis, we therefore assume that the (microscopic) contact angle equals  $\pi$ . Material points on the free surface are prescribed to have speed  $U$ , the speed of the solid, and the surface velocity is continuous at the contact point. The interface then rolls onto the solid, similar to the rolling motion of a tank tread.

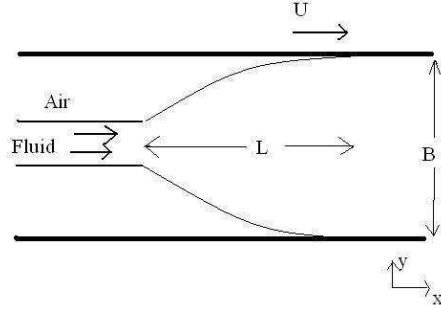


Figure 1: Geometry for the coating problem. The fluid coats a substrate moving to the right with speed  $U$ .

## Influence of Soluble Surfactant on the Breakup of Two-Fluid Viscous Jets

Bubble and drop breakup is a fundamental process in fluid dynamics. At this FRG workshop, our investigation was to determine the influence of surfactant on the breakup of an extended bubble immersed in a much more viscous fluid.

Earlier studies [7] have shown that insoluble surfactant can dramatically retard the pinch-off of the interface. Instead, the interface develops a thin, quasi-stable cylindrical thread connected to nearly spherical regions (i.e., a dumbbell shape). The local surfactant concentration in the thread is large, owing to the relatively small surface area. We therefore expect that in the soluble case, there will be considerable surfactant transport from the interface to the bulk, which will have a significant effect on the pinching dynamics.

A simple model was developed at the workshop to examine the transport of soluble surfactant for a cylindrical interface separating an inviscid inner fluid from a viscous surrounding fluid. The interface location  $r = R(t)$  and velocity  $\dot{R}$  are prescribed functions of time. After a series of transformations, the bulk surfactant concentration was found to satisfy an initial/boundary value problem for the heat equation. This was further transformed into a one-dimensional integral equation for the bulk surfactant concentration. The solution of this equation was left for future work.

## Drawing of Microstructured Optical Fibres

Microstructured optical fibres, consisting of a lattice of air holes in a glass fibre, have many desirable optical properties and offer exciting possibilities for novel applications. The first step in their manufacture is the production of a preform, a few centimetres in diameter, containing the desired distribution of holes. This may be achieved, for example, by sintering together glass capillary tubes. The preform then is heated and drawn down to a typical diameter of  $100 \mu\text{m}$ . A drawn microstructured fibre is shown schematically in figure 2, which is not to scale and has fewer holes than in

practice (say 200, not necessarily circular).

Much empirical progress has been made in constructing fibres with increasingly complex microstructure. However, attempts to model the process mathematically have been limited to an axisymmetric fibre containing a single circular hole [4, 5, 16], which discards some of its most important characteristic features. To improve the flexibility and reliability of the process, several effects contributing to the evolution of the hole require study, including the shrinking of the fibre cross-section during drawing and the flow exerted on each hole by the other neighbouring holes. Surface tension may cause the holes to shrink, potentially closing altogether, thus pressurising the holes may need to be considered mathematically.

At the FRG workshop, significant progress was made on formulating the problem in a mathematically tractable way, yet retaining some physical reality. Using perturbation methods, we transform the slender three-dimensional geometry to a sequence of weakly-coupled two-dimensional problems for each fibre cross-section. This was achieved previously for a simply-connected fibre [2] and for a single bubble in an infinite fluid [8]. Further analysis is required to apply these methods to fibres containing many holes which may be pressurised.

A numerical method was devised by Kropinski [11], who developed an integral-equation approach that is spectrally accurate. However, it is necessary to discretise each fibre cross-section in this way, which is no more efficient than a boundary-integral formulation of the full three-dimensional Stokes flow problem.

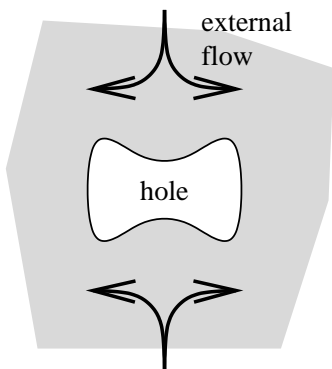


Figure 3: Schematic of a single hole in an external flow.

We have begun to formulate an alternative approach to the problem, similar to Crowdy's [1] model for two-dimensional elliptical pores. Each hole is considered in isolation, subject to an external flow, as indicated in figure 3. For linear and certain nonlinear external flows [13], exact solutions for the interface evolution may be found in the form of a time-dependent conformal map from the unit disc. We propose to describe the interface using a truncated polynomial conformal map.

This approach would allow each hole, and the fibre as a whole, to be characterised by a small number of scalar coefficients in a conformal map. The asymptotic analysis of the slender geometry then would allow us to construct a system of partial differential equations for all these parameters as functions of time and axial position (as in [2]). Work has started on analysing the hole-scale and fibre-scale problems depicted in figures 3 and 2. We anticipate that a working numerical code will be completed and written up within one year.

## Thermal Instability in (Viscous) Glass Threads

Viscosity of glass varies rapidly with temperature. In the drawing of glass threads, heat transfer will play an important role in the dynamics. A thread which cools too quickly will become viscous and require large forces to stretch it, so it is natural to heat the thread as it is being pulled. An important factor in the design of glass pulling devices is that they easily achieve stable and robust operating conditions.

The group has considered a thread that is heated while being pulled with a constant force, following a model proposed in [9]. Physically relevant simplifications then lead to a set of coupled nonlinear hyperbolic equations. Analytical solutions to the steady state equations for both uniformly and non-uniformly heated threads are obtained. We show the surprising result that steady states exist in which an increase in the pulling force actually causes a decrease in the exit speed of the

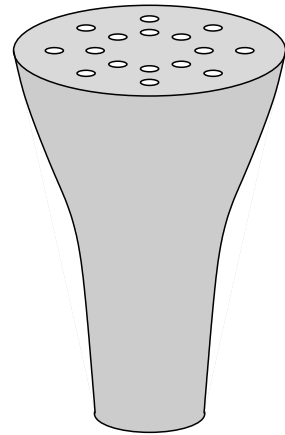


Figure 2: Schematic of a microstructured optical fibre.

thread at the end of the device. This situation can occur if the viscosity varies very abruptly with temperature and the heating rate is large enough. Assuming that the viscosity varies exponentially with temperature, if the heating is uniform, then such behavior does not occur because changes in the viscosity are not fast enough. However, if the heating is non-uniform, then the device can exhibit this behaviour. By considering an initial value problem, we show that these types of solutions are unstable, and if one operates the device in this parameter regime, the thread will pinch.

We also show devices with fixed pulling speed can exhibit hysteretic behavior that leads to rapid changes in the pulling force as the pulling speed is slowly varied.

## Pulling Glass Microelectrodes

From an applied point of the view, the group studied a glass fibre drawing problem related to the pulling of glass microelectrodes. Glass microelectrodes play an essential role in cell electrophysiology, where they are used to inject electric current and dyes into cells and measure membrane electrical potentials. Laboratories using these microelectrodes usually make them using commercially available glass tubes and pullers that use coil heaters to soften the glass during pulling. In [9], a detailed mathematical model was developed to predict the stretching and breakup of the glass tube using a vertical puller. The model is highly nonlinear and was solved numerically. Useful insights were given, e.g., the effect of heater temperature on the formation of electrodes.

During the BIRS FRG, we simplified this model so that an analytical solution can be obtained for a simple case. It is desirable to identify the main factors that have direct influence on the electrode shape, which is of critical importance. We concluded that the source of radiation energy from the coil heater can be approximated by a piecewise constant function. This simplifies the model and allows an analytical solution under a constant pulling force, a feature of more advanced horizontal pullers. Even for the vertical puller, a semi-analytical solution can be obtained.

For an arbitrary heater strength variation, the simplified model allows the implementation of a more efficient Lagrangian-based numerical method. After the BIRS workshop, we have carried out detailed parameter studies on the break-up of the glass tubes to form glass microelectrodes. A paper [10] has been submitted for publication.

One problem that we have not investigated is the detailed breakup mechanism. In the current work, we used a phenomenological breaking stress formula. The breakup of the viscous thread here is different from the surface tension induced instability. Instead, the breakup is most likely caused by spontaneous fracture due to surface damage during the extension process.

## Core-Annular Flows

Core-annular flows are two-fluid flows in circular tubes and consist of a core flow occupying the central region of the vessel surrounded by a lubricating annular fluid. The ability of the annular fluid to ‘lubricate’ the core fluid has potential applications in the oil and food industries, e.g., a highly viscous fluid can be made to flow efficiently with a given pressure gradient due to the slippage that the annular fluid provides. In applications, an interfacial instability can cause a breakup of the core fluid to produce drops or slugs of the higher viscosity fluid suspended in the lower viscosity one, or an emulsion at high flow rates.

This problem involves modelling and mathematical analysis based on the Navier-Stokes equations in a three-dimensional axisymmetric geometry with a free boundary separating the two fluids. Some attempts have been made to attack this formidable problem with direct simulations. When the annular fluid layer is thin compared with the core fluid radius, rational asymptotic expansions lead to an evolution equation for the interface, which includes long wave instability and nonlinearity. Of particular interest is the behaviour of solutions with long wave periods and which become chaotic via a Feigenbaum period doubling cascade.

The group considered the problem when the core fluid has a small radius compared to the pipe radius and has a viscosity that is small compared to that of the surrounding fluid. This fits nicely with the holey fiber work considered by the group, since it has a finite geometry due to the presence of the walls. Pressure-driven flow also is different and the two problems are complementary. The group considered the problem asymptotically in the case of a highly viscous annulus and an inviscid

core. A nonlinear evolution equation was derived and was studied for nonlinear features, e.g., traveling waves. The equations need to be solved numerically, which should suggest some more analysis, in order to produce a publication. All these aspects are currently being investigated.

## Surface-Tension-Driven Breakup of an Air Bubble in a Viscous Liquid

If you invert a nearly-full jar of maple syrup, you will see an air bubble form and rise upwards. If the air bubble becomes sufficiently elongated during the rise, it will break up into smaller bubbles. Recently, it has been shown that this phenomenon exhibits exceptional breakup dynamics [3], i.e., one which retains the effects of boundary and initial conditions to the final point of breakup. Previous examples of surface-tension driven breakup have shown that the interface shape collapses onto a single, unique form after appropriate dynamic rescaling of the coordinate axes. Such scale-invariant dynamics is obtained when the behavior is governed solely by the proximity of the breakup, with no dependence on boundary and initial conditions. The memory-preserving breakup dynamics was identified as a result of surface-tension driven breakup with an essentially static interior, with evidence provided from experiments, simulations, and theory [3]. Recent numerical simulations [15] of a surface-tension driven breakup of a cylindrical hollow inside a viscous jet provided further confirmation of this unusual property associated with static-interior breakup.

A long-wavelength model for the time-evolution of the bubble surface [3] and static-interior breakup process was derived to describe the breakup dynamics. Three common breakup scenarios are analysed: the detachment of a large bubble from a nozzle, the breakup of an infinitely long cylinder (see [15]), and the breakup of a finite-sized bubble. Exact expressions for the bubble shape and interior pressure are derived for the simpler limiting situations of infinite cylinder breakup and nozzle detachment. Our analytical results show that the shape at breakup retains an imprint of boundary and initial conditions. They also show that the long-wavelength dynamics associated with a static-interior breakup cannot give rise to new minima in the bubble shape.

As bubble breakup is approached, the solution of the long-wavelength equation for surface evolution approaches the same form regardless of initial and boundary conditions. Since the collapse does not distort the neck shape, this shape retains an imprint of initial and boundary conditions, as noted in [3].

For an air bubble, the interior flow speed is always significantly larger than the exterior flow speed, and therefore, the breakup corresponds to surface-tension driven breakup with a static interior. Near breakup, the bubble neck simply collapses inward at a uniform rate, see Figure 4, in contrast to other situations where the interior flow is significant and the breakup dynamics evolves towards a scale-invariant form. The analysis shows, in the long-wavelength limit, that the static-interior breakup has the unusual property that all unstable modes grow at the same rate, i.e., there is no fastest growing mode. As a consequence, the breakup dynamics is highly sensitive to details of the initial shape.

In the long-wavelength limit, an initial shape with a minimum, however small, breaks up into two bubbles. An initial shape which is everywhere convex, however extended, rounds into a sphere.

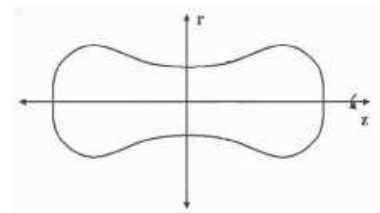


Figure 4: A bubble immersed in a viscous exterior liquid.

## List of Participants

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