

# Thermal Modulation and Instability of Newtonian Liquid Microjets

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## ABSTRACT

Liquid microjets are inherently unstable and can be broken into droplets by various means including modulation of pressure, velocity, and/or fluid properties. In this presentation, we discuss the controlled breakup of viscous microjets via thermal modulation of surface tension. Such modulation has been implemented using CMOS/MEMS technology by integrating resistive heating elements around each orifice of a manifold as depicted in Fig. 1. When the heating elements are electrically pulsed, the thermal energy they produce penetrates the surface of the microjet, and is carried downstream to produce a spatial variation of surface tension along the length of the jet, which ultimately causes breakup and drop formation (Figs. 1 and 2). Using this process, microfluidic devices have been fabricated with thousands of individually modulated microjets that can produce steady streams of picoliter-sized droplets at kilohertz frequency rates [1]. In this presentation we review methods for analyzing such devices.

**Keywords:** microjet instability, Marangoni flow, surface tension modulation, thermo-capillary instability, microfluidic drop generator, slender jet analysis.

## 1 INTRODUCTION

The analysis of jet instability has a long history with initial investigations dating back to Lord Rayleigh [2]. However, despite the duration of this research and the substantial interest in this topic, relatively few results exist on the instability of thermally-modulated orifice-driven microjets. This occurs when a microjet is heated in a time-dependent fashion as it passes through an orifice. This pulsed heating modulates the surface tension  $\sigma$  at a wavelength  $\lambda = v_0\tau$ , where  $v_0$  is the jet velocity and  $\tau$  is the period of the heat pulse as shown in Fig. 1. To first order, the temperature dependence of  $\sigma$  is given by

$$\sigma(T) = \sigma_0 - \beta(T - T_0) \quad (1)$$

where  $\sigma(T)$  and  $\sigma_0$  are the values at temperatures  $T$  and  $T_0$ , respectively. Since  $\sigma$  is temperature dependent, the down-stream advection of thermal energy gives rise to a spatial variation (gradient) of surface tension along the jet.

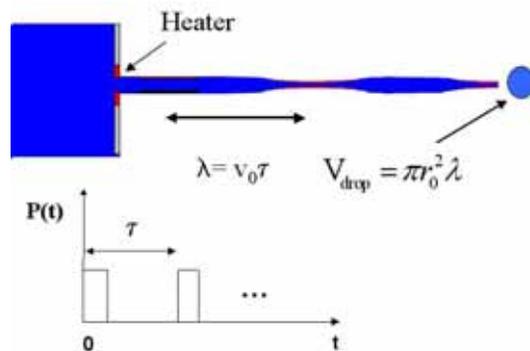


Figure 1: Physical system.

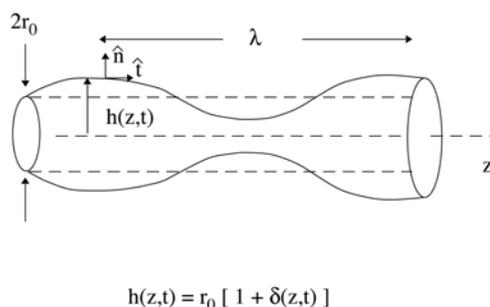


Figure 2: Free-surface geometry and reference frame.

This produces a shear stress at the free-surface, which is balanced by inertial forces in the fluid, thereby inducing a Marangoni flow towards regions higher surface tension (from warmer regions towards cooler regions). The Marangoni flow causes a deformation of the free-surface (slight necking in the warmer regions and ballooning in the cooler regions) that ultimately leads to instability and drop formation.

A rigorous analysis of this phenomenon requires the solution of a fully coupled thermal/fluid/free-surface initial/boundary value problem that takes into account (i) the temperature dependence of all fluid properties (viscosity, density, and surface tension), (ii) the related temperature dependent changes in velocity and pressure at the orifice, (iii) thermal diffusion within the orifice manifold and reservoir, (iv) thermal diffusion and advection within the microjet, (v) the variation of surface tension and induced Marangoni flow along the jet, and (vi) the complex free-surface dynamics. Such an analysis typically requires a

sophisticated numerical program and the use of a relatively large number of computational nodes that are spaced (preferably adaptively) with sufficient density to enable accurate predictions of the free-surface (filaments, drops, and satellites) before, during, and after pinch-off. Even when such a program is available, the time required to perform practical parametric studies of a proposed concept can be prohibitive, and such simulations do not always provide the level of understanding that is often desired to guide the rapid development of new prototype systems.

In this presentation we discuss various methods for analyzing thermally-modulated Newtonian liquid microjets. Specifically, we discuss (i) an analytical formula for estimating the time-to-breakup  $T_b$  as a function of the modulation wavelength, jet radius, and fluid properties; (ii) a 1-D numerically-based slender-jet model for predicting the free-surface at pinch-off (drops and filaments); and (iii) a fully-coupled thermal-CFD approach for studying drop formation and satellites during and after the formation of multiple droplets.

## 2 EQUATIONS OF MOTION

The equations governing the behavior of a non-isothermal viscous microjet of an incompressible Newtonian fluid with surface tension  $\sigma$ , viscosity  $\mu$ , density  $\rho$ , specific heat  $c_p$ , and thermal conductivity  $k$ , are as follows:

### Navier-Stokes:

$$\rho \frac{Dv}{Dt} = -\nabla p + \mu \nabla^2 v, \quad (2)$$

$$\text{where } \frac{D}{Dt} = \frac{\partial}{\partial t} + v \cdot \nabla.$$

### Thermal:

$$\rho c_p \frac{DT}{Dt} = k \nabla^2 T. \quad (3)$$

### Continuity:

$$\nabla \cdot v = 0. \quad (4)$$

### Boundary Conditions:

Thermal:

$$-k \hat{n} \cdot \nabla T = h_c (T - T_\infty). \quad (5)$$

Normal Stress:

$$(\mathbb{T} \cdot \hat{n}) \cdot \hat{n} = -2H\sigma. \quad (6)$$

Tangential Stress:

$$(\mathbb{T} \cdot \hat{n}) \cdot \hat{t} = \hat{t} \cdot \nabla_s \sigma. \quad (7)$$

Kinematic (at jet surface):

$$\frac{D}{Dt}(r_s - h(z, t)) = 0. \quad (8)$$

On axis ( $r = 0$ ):

$$v_r = \frac{\partial v_z}{\partial r} = \frac{\partial T}{\partial r} = 0, \quad (9)$$

where  $v$ ,  $p$ , and  $T$  are the velocity, pressure and temperature distributions along the microjet,  $v_r$  and  $v_z$  are the radial and axial velocity components,  $h(z, t)$  defines the free-surface (Fig. 2),  $\hat{n}$  and  $\hat{t}$  are unit vectors normal and tangential to the free-surface,  $\nabla_s$  is the gradient operator along the free-surface,  $\mathbb{T}$  is the stress tensor, and  $h_c$  is the coefficient for thermal convection off the free-surface. The function  $H$  is given by

$$H = \frac{1}{2} \left( \frac{1}{h(1+h'^2)^{1/2}} + \frac{h'}{(1+h'^2)^{3/2}} \right), \quad (10)$$

where  $h' = \partial_z h$ .

Equations (2) - (9) need to be solved subject to an imposed time-dependent thermal modulation applied to the microjet at the orifice.

## 3 ANALYTICAL ANALYSIS

Equations (2)-(9) are too complicated to solve in closed-form. However, an analytical solution exists for a related problem, which can be used to estimate the time-to-breakup  $T_b$  for the process of interest here. Specifically, an analytical solution has been derived for the temporal instability of an infinite viscous microjet with a periodic variation of surface tension imposed along its length [3]. This analysis is based on a linearization of equations (2)-(9) and an uncoupling of the thermal diffusion. The linearized infinite jet analysis provides reasonable estimates of  $T_b$  for orifice-driven modulation as long as the jet velocity  $V_0$  is

much greater than the capillary velocity  $v_c = \sqrt{\sigma/(\rho r_0)}$  [3]. In the linear analysis, the free-surface and surface tension are written as

$$h(z, t) = r_0 [1 + \delta(z, t)], \quad (11)$$

$$\sigma(z, t) = \sigma_0 - \frac{\Delta\sigma}{2} \left( 1 + \cos\left(\frac{2\pi}{\lambda} z\right) \right), \quad (12)$$

where  $r_0$  and  $\sigma_0$  are the unperturbed radius and surface tension of the jet,  $\delta(z, t)$  is the deformation of the free-surface, and  $\Delta\sigma$  is the maximum change (perturbation) in surface tension, which is temperature dependent. To simplify the analysis, a coordinate system  $\eta = z - v_0 t$  is chosen that is at rest with respect to the jet, and the equation of motion for  $\delta(z, t)$  becomes

$$\begin{aligned} \frac{\partial^2 \delta}{\partial t^2} + \frac{\sigma_0}{2\rho r_0} \left[ \frac{\partial^2 \delta}{\partial \eta^2} + r_0^2 \frac{\partial^4 \delta}{\partial \eta^4} \right] - \frac{3\mu}{\rho} \frac{\partial^3 \delta}{\partial \eta^2 \partial t} = \\ + \frac{1}{2\rho r_0} \frac{\partial^2}{\partial \eta^2} \left( \frac{\Delta\sigma}{2} \left( 1 + \cos\left(\frac{2\pi}{\lambda} \eta\right) \right) \right). \end{aligned} \quad (13)$$

Equation (13) defines an initial-value problem for  $\delta(\eta, t)$ , which is solved subject to the initial conditions  $\delta(\eta, t) = 0$ , and  $\partial_t \delta(\eta, t) = 0$ . When thermal diffusion is taken into account, the solution is of the form [3]

$$\begin{aligned} \delta_h(\eta, t) = \frac{\Delta\sigma}{2\sigma_0} \frac{\left[ \frac{r_0^2 - r_h^2}{r_0^2} + \frac{2}{r_0^2} \sum_{m=1}^{\infty} c_m e^{-\alpha_m \gamma_m^2 t} J_0(\gamma_m r_0) \right]}{\left[ 1 - r_0^2 \left( \frac{2\pi}{\lambda} \right)^2 \right]} \\ \times \left[ 1 + \frac{\alpha_-}{\alpha_+ - \alpha_-} e^{\alpha_+ t} - \frac{\alpha_+}{\alpha_+ - \alpha_-} e^{\alpha_- t} \right] \cos\left(\frac{2\pi\eta}{\lambda}\right). \end{aligned} \quad (14)$$

The analytical solution (14) describes the behavior of the free-surface  $h(\eta, t) = r_0 [1 + \delta(\eta, t)]$ . The function  $\delta(\eta, t)$  obtains a maximum when  $\eta \equiv \eta_{\max} = \pm \frac{\lambda}{2}, \pm \frac{3\lambda}{2}, \pm \frac{5\lambda}{2}, \dots$ , and a minimum when  $\eta \equiv \eta_{\min} = 0, \pm \lambda, \pm 2\lambda, \pm 3\lambda, \dots$ . Jet breakup (pinch-off) occurs when  $\delta(\eta_{\max}, t) = 1$  or  $\delta(\eta_{\min}, t) = -1$  (when  $h(\eta, t) = 2r_0$  or 0, respectively). Thus, the time to breakup can be computed by solving for the value of  $t = T_b$  that gives  $\delta(\eta = 0, t) = -1$ . Parametric calculations of  $T_b$  vs. percent modulation and reduced wavenumber  $k = 2\pi r_0 / \lambda$  (as a function of jet radius) are shown in Figs. 3

and 4, respectively. This analysis was performed for a microjet of water with a 10  $\mu\text{m}$  diameter [3].

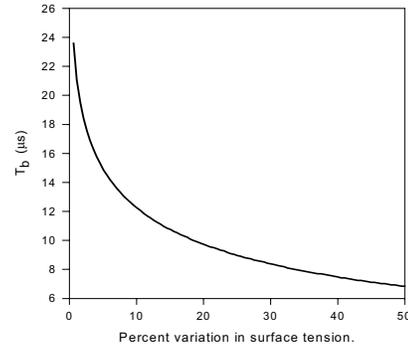


Figure 3:  $T_b$  vs. percent variation in surface tension.

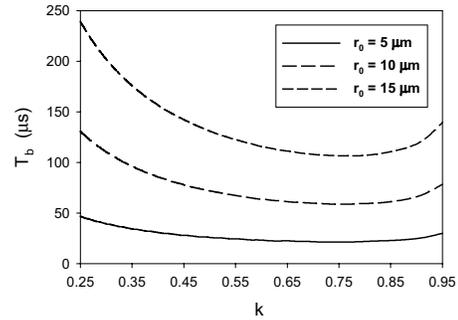


Figure 4:  $T_b$  vs. wavenumber  $k$  and jet radius.

## 4 SLENDER JET ANALYSIS

For slender microjets, which are of interest here, equations (2)-(9) can be simplified using a perturbation expansion in  $r$  for the unknown variables  $h$ ,  $T$  and  $v$ , and retaining the lowest order terms. This leads to the following 1-D slender jet equations:

$$\frac{\partial v}{\partial t} + v \frac{\partial v}{\partial z} = -\frac{1}{\rho} \frac{\partial(2\sigma H)}{\partial z} + \frac{3\mu}{\rho h^2} \frac{\partial}{\partial z} \left( h^2 \frac{\partial v}{\partial z} \right) + \frac{2}{\rho h} \frac{\partial \sigma}{\partial z}, \quad (15)$$

$$\frac{\partial h^2}{\partial t} = -\frac{\partial(h^2 v)}{\partial z}, \quad (16)$$

$$\frac{\partial T}{\partial t} + v \frac{\partial T}{\partial z} = \alpha \frac{1}{h^2} \frac{\partial}{\partial z} \left( h^2 \frac{\partial T}{\partial z} \right) - \frac{2\alpha h_c}{hk} (T - T_\infty). \quad (17)$$

In this approximation there is thermal diffusion along the axis of the microjet and convection off its surface, but no radial diffusion (1-D approximation).

Equations (15)-(17) can be solved using various numerical methods. A finite difference approach with an implicit  $\theta$ -weighted time-stepping was used for our analysis. Specifically, a uniform (staggered) mesh was used in which  $h$ ,  $p$ , and  $T$  are evaluated on one set of nodes, and the velocity  $v$  is computed on interlaced nodes

midway between the first set. The program used here was developed and implemented in MATLAB by Prof. Michael Brenner. It can predict the structure of drops and filaments as a function of modulation wavelength and fluid properties as shown in Figs. 5 and 6. Parametric analysis of these structures can be completed in a few minutes.

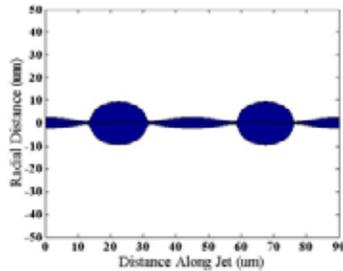


Figure 5: Drop and filament formation for  $\lambda = 9r_0$ .

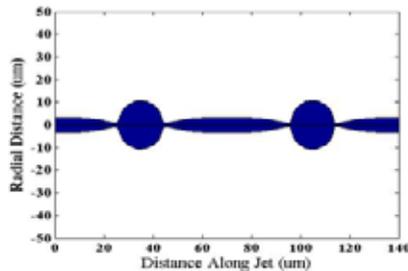


Figure 6: Drop and filament formation for  $\lambda = 14r_0$ .

## 5 COUPLED THERMAL-CFD ANALYSIS

The rigorous solution of equations (2)-(9) requires a fully coupled thermal-CFD analysis. A commercial software package FLOW-3D is used for this in our laboratory [4]. The thermal-CFD simulations take into account detailed models of the orifice (material properties, layering, and dimensions), as well as the applied power level and pulsing scheme. These simulations track the formation of multiple droplets during, and long after, pinch-off as shown in Fig. 7. Each simulation typically requires a few days to complete, and a detailed post-processing is conducted to examine key physical processes such as thermal diffusion along the jet and its impact on surface tension, and induced pressure and velocity variations near the orifice.

## 6 CONCLUSIONS

CMOS/MEMS technology has been used to produce novel microfluidic devices that can generate picoliter-sized droplets with unprecedented speed and versatility. These devices implement an orifice-driven thermal modulation of the microjet. In this presentation, we have discussed various methods for analyzing this process. These include (i) an analytical approach, (ii) a 1-D slender jet analysis, and (iii) a fully-coupled thermal-CFD analysis.

The analytical approach provides insight into the basic physics of jet instability and is useful for parametric

analysis of  $T_b$  as a function of jet radius, wavelength, and fluid properties. However, it does not predict the structure

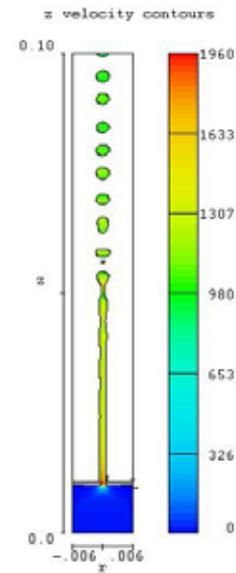


Figure 7: Coupled thermal-CFD simulation.

of droplets or filaments during pinch-off. The 1-D slender jet analysis predicts the structure of drops and filaments as a function of the modulation wavelength and fluid properties, but provides no information beyond pinch-off. Moreover, it does not account for the radial diffusion of thermal energy, which weakens the thermal perturbation and delays breakup. The coupled thermal-CFD analysis provides a complete description of the process, but is time-consuming and awkward for parametric analysis, which is often required for the development of novel prototype drop generation systems. In practice, all three approaches are used to develop novel microfluidic droplet generators.

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