Liquid Disintegration: Investigation of Physics by Simulation

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ABSTRACT

Liquid disintegration includes the processes of formation of droplets and droplet fission. It is important in inkjet printing and in the miniaturization of biochemical analytical devices. This paper reports on an investigation of the mechanism of liquid disintegration via numerical simulation. The paper points out that the process is inherently multi-directional; droplet formation occurs only when the effect of the centripetal flow overcomes that of the axial flow.

A reduced-order model for droplet pinch-off, based on this insight, is derived and validated by simulations. Reduced-order models for droplet production have been derived previously from a phenomenological approach. This study shows the inherent limitation of this approach compared to that presented here, based on tracing the evolution of the multi-directional momentums.

Keywords: simulation, reduced-order modeling, droplet, liquid disintegration, inkjet

1 INTRODUCTION

Liquid disintegration includes the process of formation of droplets, or separating a liquid stream into discrete droplets; and the process of droplet fission, or dividing one droplet into smaller ones. Both processes share the same fundamental physics. From the energy perspective, both processes are nothing but energy transfer between surface potential and energy of other forms (thermal, electrical, kinetic, etc.). Disintegration is initiated by distortion of the fluid surface. When the surface area is increased, energy of other forms must be transduced into surface potential to make up the difference; when the surface area is reduced, the difference in surface potential is released.

The scientific study of liquid disintegration can be traced back to the early nineteenth century [1]. It has intensified since the 1970's, driven by the commercial success of the inkjet printing industry, whose core technology is a microelectromechanical (MEMS) device that produces discrete droplets or a stream of droplets [2].

Liquid disintegration is also important in the biotechnology sector. Droplets are the most natural vehicles to carry biochemical agents. Therefore, the control over their production and manipulation has attracted significant attention in the laboratory-automation community. Recently, a droplet-based programmable fluid processor, or PFP, has been developed [3]. The PFP is a centimeter-sized biochip with which general-purpose biochemical analyses may be conducted. In the operation of this chip, minute amounts (picoliter to nanoliter) of chemical sample are drawn from individual reservoirs in the form of droplets. These droplets are then delivered to a reaction chamber where multiple droplets may reside simultaneously. An individually addressable electrode array, embedded underneath the reaction chamber, generates dielectrophoretic forces that can translate droplets to pre-determined locations at pre-determined times. When droplets containing different chemical samples arrive at the same location, the droplets will merge into one droplet and a chemical reaction can occur. Chemical reactions can be detected, categorized, and reported. Hierarchical reactions can be achieved by merging droplets of intermediate reactions. A larger droplet may be split into smaller ones for more efficient manipulation or for detection (Figure 1). Other example applications include a droplet-based cytometer [4].

Understanding the mechanism of liquid disintegration is essential to the design of biochips. Methods that accomplish

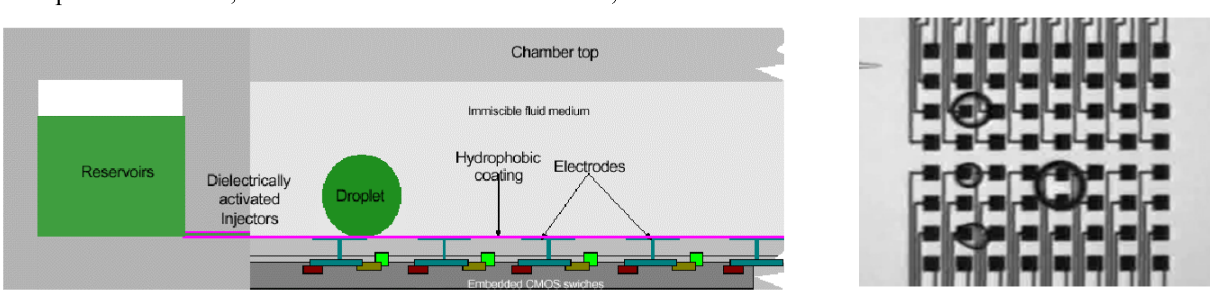

Figure 1. The Programmable Fluid Processor (PFP). Left shows an elevation view of the device. Right shows an experimental image of the reaction chamber with multiple droplets sitting on the top of electrodes in the array. Images courtesy of Professor P. R. C. Gascoyne of M. D. Anderson Cancer Center.

liquid disintegration may be classified into three categories: continuous jetting, drop-on-demand and those that utilize the Marangoni effect. In continuous jetting, a pressurized liquid flow from a nozzle breaks into a stream of droplets owing to the exponential growth of surface oscillations; a process that has been successfully explained by the linear stability analysis originated by Rayleigh [5]. This disintegration technique does not offer control over the event of droplet production. The Marangoni effect [6][7] creates an interfacial flow, which in turn induces liquid movement in the bulk due to viscosity. The movement in the bulk determines whether droplet pinch-off (fission) occurs, similar to the drop-on-demand method. The drop-on-demand method is the focus of this paper.

Numerical simulation is used throughout this paper and simulations of a drop-on-demand model problem are analyzed in detail. The model problem simulations show that, even though the axial flow along the injection direction is primarily responsible for the jet growth, the droplet formation process is largely determined by a secondary centripetal flow inside the jet. Therefore, the production of a droplet is a multi-directional rather than unidirectional process. This is the fundamental reason why the descriptive or phenomenological reduced-order modeling approaches [8] that treat droplet production as unidirectional can achieve only limited reusability. That is, a model designed for one device under one operating condition is usually not useful for other cases.

The model-problem simulations also reveal that the centripetal and axial flows work at cross-purposes. If the centripetal flow is sufficiently strong, droplet pinch-off will occur. Based on this understanding, a reduced-order model for droplet pinch-off is derived. Over a hundred simulations are carried out to adjust and validate the model. A discussion of these simulations provides a conclusion to the paper.

2 METHODS AND TOOLS

All of the simulation results presented in this paper were obtained by using the multi-physics simulation package CoventorWare™ (Coventor, Inc., Cambridge, Massachusetts).

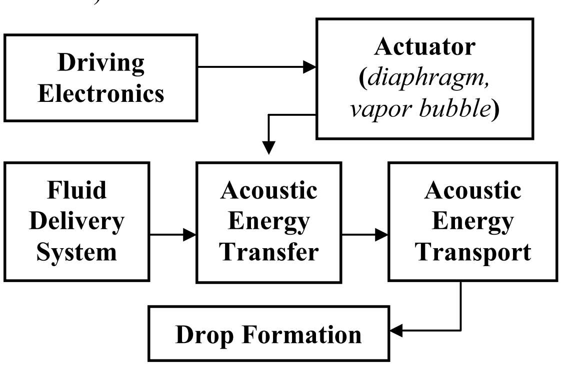

Figure 2. A schematic of the drop-on-demand process.

CoventorWare™ integrates FLOW-3D® (Flow Science Inc., Santa Fe, New Mexico) as its free-surface flow simulation engine. FLOW-3D® uses Volume-of-Fluid (VOF) methodology to handle problems with surfaces that undergo large-amplitude deformation and/or break-up. In contrast to other VOF software, FLOW-3D® applies true stress boundary conditions at free surface and eliminates the diffuse boundary layer between liquid and air that is commonly found in other VOF software. The dynamic contact angle is recovered from calculations accounting for effects including that of surface tension, wall adhesion, air pressure, liquid viscosity, and gravity.

3 MODEL PROBLEM

A typical drop-on-demand process may be described as in Figure 2. An electronically controlled actuator generates a large-amplitude fluid displacement. This disturbance travels in the fluid as acoustic energy and at the ejection nozzle generates one droplet. The model problem for this process is illustrated in Figure 3. A large-amplitude pressure pulse represents the acoustic signal delivered to the nozzle (distance l away in Figure 3). Without loss of generality, this model problem encapsulates all of the complex aspects of the actuation and acoustic propagation in the pressure pulse so that the numerical investigation can concentrate on the mechanism of droplet production.

Figures 4 and 5 show simulation results for nozzle radius $a=5\mu m$, and nozzle length $l=40\mu m$. The liquid has surface tension $\gamma=7.4e\text{-}2Kg/s^2$ and viscosity $\mu=1e\text{-}3Kg/m/s$. The actuation is represented by pressure pulse amplitude $P_0=0.7Mpa$ and pressure pulse period $T=4\mu sec$. For the first half of the period, the pressure is positive and this half-period is called the "acceleration stage". The second half-period, with negative pressure, is called the "deceleration stage".

Figure 4 shows the free surface profile. The necking, or reduction of the cross-section of the liquid jet at one point (the "pinch-off" point) occurs at the beginning of the deceleration stage. The necking continues throughout the deceleration stage until the cross-section converges to one point and the droplet breaks from the jet (t=18).

Simulation results show that the flow inside the nozzle

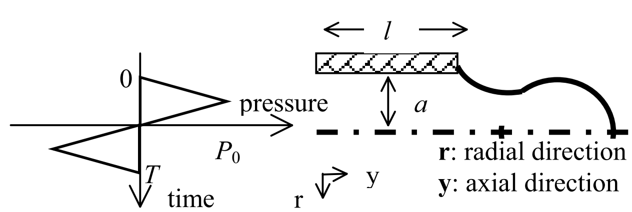

Figure 3. The model problem. The input pressure pulse is of triangular profile and is initiated in the liquid at $y=-l$. The nozzle is at $y=0$.

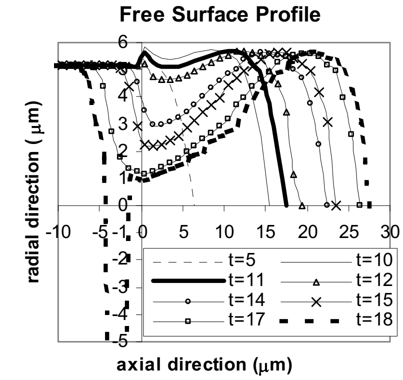

Figure 4. The evolution of the free surface position. Negative radius indicates the profile of the cavity formed after droplet breakup. The time increment is 0.05T.

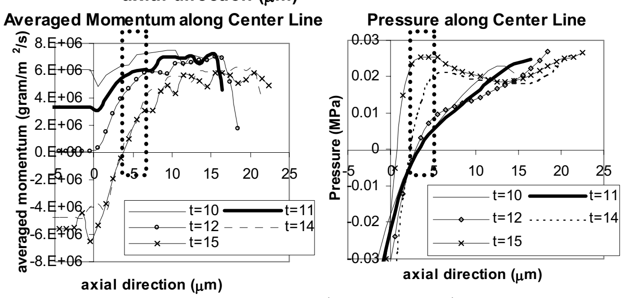

Figure 5. The evolution of the averaged momentum and pressure along the axial direction.

behaves as Poiseuille flow: the momentum has no spatial variation and the pressure varies linearly. In the acceleration stage, the liquid flow accelerates and the jet grows. In the deceleration stage, the flow inside the nozzle and that inside the jet are distinct from each other. The flow inside the nozzle slows down and eventually switches direction, following the negative gauge pressure at the actuation end. However, the flow inside the jet maintains more or less unchanged momentum (for instance, t>=11 in Figure 5). The disparity in momentum at both sides of the pinch-off point drains liquid out of its vicinity and eventually breaks the droplet from the jet body.

Such momentum disparity is a result of the dynamics of the pressure propagation and the presence of surface tension. During the acceleration stage, the positive portion of the pressure pulse propagates throughout the entire liquid body including the jet, where it linearly decays along the axial direction. However, during the deceleration stage, the negative portion of the pressure pulse appears to stop at the pinch-off section (dotted box in Figure 5) and does not propagate throughout the jet. Creation and maintenance of this signal blockage is the very reason that a successful droplet pinch-off is achieved.

Further understanding requires the Laplace-Young equation, which explains the role of surface tension. According to Laplace-Young, assuming the ambient air pressure is zero, the liquid pressure underneath the free surface may be expressed as $\gamma(1/R_1 + 1/R_2)$ where R_1 and R_2 are free-surface radii of curvature. At the tip of the jet, both R_1 and R_2 are of the order of a, thus the pressure P_t is approximately $2\gamma/a$. At the pinch-off section before necking occurs, R_1 is equal to a', the neck radius, and R_2 is infinite, thus the pressure P_p is equal to γ/a'.

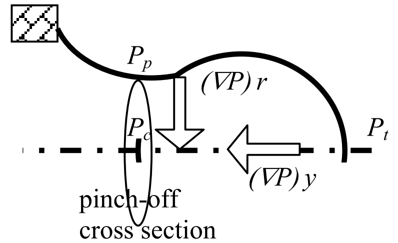

Figure 6. Illustration of the race between the two pressure gradients.

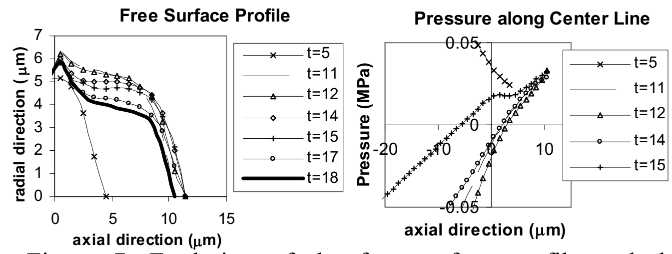

Figure 7. Evolution of the free surface profile and the pressure along the axial direction, for $P_0=0.4Mpa$ and time increment $0.05T$.

During the deceleration stage, the reduction of P_c, the liquid pressure at the center of the pinch-off section, affects two pressure gradients: the axial pressure gradient $(\nabla P)y$ and the centripetal pressure gradient $(\nabla P)r$. $(\nabla P)y$ forces the liquid flow to respond to the pressure pulse and tends to slow down the flow inside the jet and eventually reverse the flow direction back towards the nozzle.

$(\nabla P)r$ induces a secondary centripetal flow. The centripetal flow shrinks the neck a' thus increasing γ/a' and consequently P_c. The rise of P_c can be clearly observed in the pressure plot in Figure 5. With more or less fixed P_t, the rise of P_c means $(\nabla P)y$ is reduced: from positive at the beginning of the deceleration stage, to approximately zero at t=14, and to negative at t=15. At this point, the flow inside the jet is accelerating rather than decelerating until the pinch-off.

It is the axial flow that contributes to the jet growth (and shrinkage). However, the secondary centripetal flow is the key to droplet detachment from the jet body. To prove this, Figure 7 shows another simulation result where P_0 is reduced to $0.4Mpa$ and the other parameters are kept the same. In this case, the secondary centripetal flow is too weak to raise P_c during the deceleration stage. Therefore, the pressure gradient is spatially uniform throughout the process. That is, the pressure pulse propagates throughout the entire liquid body without blockage. Hence, no momentum disparity is created and, of course, no droplet pinch-off occurs.

4 A REDUCED-ORDER MODEL FOR DROPLET PINCH-OFF

It is the effect of $(\nabla P)r$ overcoming that of $(\nabla P)y$ at the beginning of the deceleration stage that is the condition for successful droplet pinch-off. Recognizing that at the beginning of the deceleration stage $a'=a$, assuming a

Poiseuille flow profile, and accounting for the pressure jump at the jet tip due to the surface tension, such a condition can be expressed as $F>0$, with F defined as

$$F = \frac{aT}{64\pi\mu l}(p_0 - \frac{4\gamma}{a}) - 1 \quad (1)$$

To validate this condition, a set of simulations has been carried out. The parameters are varied such that the surface tension coefficient $\gamma=0.02Kg/s^2$, $0.074Kg/s^2$ and $0.1Kg/s^2$; the viscosity $\mu=0.001Kg/m/s$, $0.002Kg/m/s$ and $0.005Kg/m/s$; and P_0 covers the range from $0.25MPa$ to $1MPa$ with a $0.05MPa$ increment. 144 data points have been collected and the results shown in Figure 8. Since Equation 1 predicts successful droplet pinch-off only for $F>0$, the differing simulation results indicate that the secondary flow effects not included in Equation 1 are indeed important. These effects are the transduction of acoustic energy to the kinetic energy and viscous dissipation of the secondary flow inside the jet that is induced by the surface tension. To compensate for this, the condition $F>0$ is modified to $G>0$, with G given by

$$G = \frac{aT}{64\pi\mu l}(p_0 - \frac{4A\gamma}{a}) + \frac{B}{\mu} + C \quad (2)$$

where constants $A=5.2$ represents the additional pressure potential loss due to surface tension, $B=-2e-4Kg/m/s$

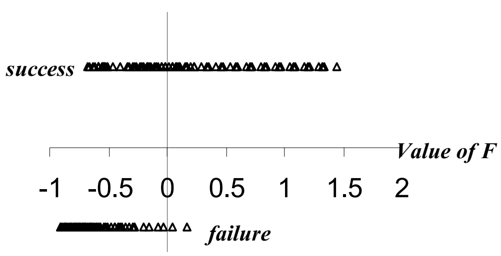

Figure 8. F values for 144 simulations. The two ordinate values are "success" indicating droplet formation is observed and "failure" indicating droplet formation is not observed.

represents additional viscous dissipation, and $C=-0.231$ is to reset the transition value of G to zero. These constants are determined by fitting the $G>0$ condition with the 144 data samples of Figure 8. Figure 9 shows the values of G for the 144 simulations. It is expected that since this model is derived from the underlying physical principals of droplet pinch-off, rather than a phenomenological approach, it is applicable to any drop-on-demand device driven by a large amplitude acoustic signal.

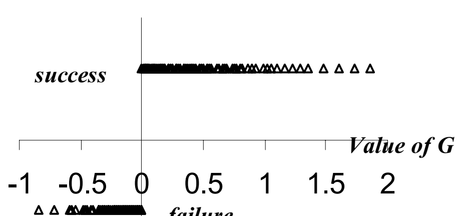

Figure 9. G values for 144 simulations.

5 CONCLUSION AND FUTURE WORK

Numerical simulation is used to investigate the mechanism of drop-on-demand. The study shows that droplet pinch-off results from the centripetal flow effects overcoming those of the axial flow.

It is a rather broadly observed practice to adopt the so-called phenomenological approach to derive a reduced-order model for liquid disintegration (for a representative work see [8]). This method discards the secondary centripetal flow that is primarily responsible for the success or failure of the disintegration process. Hence this approach has achieved limited reusability. That is, a model derived for one device cannot be easily applied to describe the behavior of other devices that work under the same operating principle.

Recognizing the multi-directional nature of the liquid disintegration process, a novel reduced-order model for droplet formation is currently under development. Using the approach established here eliminates the reliance on phenomenological observation. Rather, this approach closely traces the evolution of the momentums of the centripetal flow and of the axial flow inside the jet; and so is expected to lead to a reduced-order model that will deliver accurate droplet pinch-off time and location, as well as the quality of the generated droplet (volume, terminal velocity, and sphericity). This model will be applicable to a broad range of liquid disintegration devices. The model and its performance will be presented in a future publication.

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