Drop dispensing in a viscous outer liquid

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Abstract: The formation and detachment of Newtonian drops in viscous external liquids is investigated. A global analysis of two necking processes is presented in order to highlight the behavior of such thinning phenomena, when controlled either by inertia or by viscous effects. Moving detached droplets in an immiscible outer liquid were studied in terms of velocity and drop-travel distance. Theoretical predictions are proposed and compared with experimental data for the volume of the drop and for the subsequent dynamics that follow after detachment. Our investigations point out that the drop rapidly achieves constant velocity, the value of it being in a satisfactory agreement with the model. Both the influence of the flow rate and that of the material properties on drop volume are pursued.

Key Words: capillarity, drop formation, filament thinning, drop detachment

1. INTRODUCTION

Controlled drop generation is a subject of high practical impact with direct applications in pharmaceutical industry, ink-jet printing and food processing. In all cases precise dosing is required. This results in a need of knowledge regarding drop volumes and drop breakup times. Furthermore, drop generation in a viscous outer liquid is a subject that needs both theoretical and experimental attention.

Depending on the relative balance between inertia and interfacial tension, dispensing a liquid through a nozzle can take place in the form of a dripping or a jetting regime [1]. In both cases drops are created, the drop size distributions having different theoretical backgrounds. We can physically describe a pendant drop (Fig.1-a) by the magnitude of the buoyancy reduced force of gravity relative to interfacial tension [2]. These are incorporated into a non-dimensional parameter called Worthington number, which can be seen as the well-known Bond number, having as characteristic length scale $l_c$:

\[
Wo = (\rho_i - \rho_e) g V_D / 2\pi R_0 \sigma, \quad Bo = (\rho_i - \rho_e) g l_c^2 / \sigma, \text{ with } l_c = \sqrt{V_D / 2\pi R_0} \quad (1)
\]

where the symbols represent: the density of the inner and outer liquid $\rho_i, \rho_e$, the volume of the drop $V_D$, the inner radius of the nozzle $R_0$, and $\sigma$ the interfacial tension. When the buoyancy reduced force of gravity overcomes the interfacial tension that holds the drop to the capillary tip, the drop will start to detach (Fig. 1-b). The breakup process is driven by
capillarity, which acts to minimize the surface energy of the filament. The local stresses that act against capillary action can be inertial, viscous or elastic in nature. The thinning process is therefore governed by the magnitude of these stresses \cite{3}. When an external liquid is present, the characteristic time scale of the breakup process increases, opening the possibility of seeing more of the thinning dynamics, thus emphasizing the importance of the viscosity ratio $\beta$.

![Diagram of forces acting on a pendant drop](image)

**Fig. 1** a) Schematic illustration of the forces that act on a pendant drop formed in a viscous outer medium; b) The detachment of a droplet of water and 10% glycerol when the buoyancy reduced force of gravity exceeds the tension force; c) Series of images showing the necking process that takes place at drop detachment, in the case of a mixture of water and 75% glycerol.

## 2. EXPERIMENTAL DETAILS

The experimental set-up consists of a capillary needle with an inner diameter of 0.84 mm held vertically by a mechanical mechanism. A glass tank was used to hold the external liquid. The injector was connected to a Harvard Apparatus 33 syringe pump for a controlled flow rate. Snapshots of drop formation and filament thinning were taken with a Nikon 1 J5 camera recording at 400 fps.

We used two types of liquids as an external medium, mineral oil and silicon oil, and various water-glycerol mixtures as injected liquids. All configurations, injected liquid-external liquid are immiscible with one another. Viscosity measurements were performed using a rotational Anton Paar Physica MCR301 rheometer.

The density was obtained via a mass per volume method, the mass of a controlled volume of liquid being measured with a Radwag scale. The volume was injected with a Hamilton microsyringe Gastight #1725 of 0.25 ml. A pendant drop technique was used in order to get an estimate of the interfacial tension \cite{4}. Table 1 and Fig. 2 provide the values of the material properties for the liquids in question. As an observation, the interfacial tension coefficient presents a linear decrease in value as the concentration of glycerol increases.
Table 1. Material properties for the injected liquids and the external mediums at 25°C.

<table>
<thead>
<tr>
<th>Substance (-)</th>
<th>Density (kg/m³)</th>
<th>Viscosity (Pas)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water</td>
<td>1000</td>
<td>0.001</td>
</tr>
<tr>
<td>W+10% Glycerol</td>
<td>1031</td>
<td>0.002</td>
</tr>
<tr>
<td>W+50% Glycerol</td>
<td>1144</td>
<td>0.007</td>
</tr>
<tr>
<td>W+75% Glycerol</td>
<td>1204</td>
<td>0.053</td>
</tr>
<tr>
<td>Mineral oil</td>
<td>926</td>
<td>0.055</td>
</tr>
<tr>
<td>Silicon oil</td>
<td>951</td>
<td>1.04</td>
</tr>
</tbody>
</table>

Fig. 2 Interfacial tension dependence on concentration

This is seen for both mineral and silicon oil as external mediums, slightly higher values being recorded when for configurations having silicon oil as the surrounding liquid.

The following paragraph describes the procedure for drop generation: a moderate and constant flow rate was set in order to avoid the transition to a jetting regime. As the drop accumulates mass, the buoyancy reduced force of gravity will overcome the tension holding the drop attached to the injector. At that instance a necking process takes place which produces detachment (Fig.1-c). This process was recorded and further analyzed. We also recorded the motion of the drop through the surrounding liquid after detachment until it attained constant velocity. The effect of modifying the flow rate and the material properties, e.g. density, on drop diameter was also emphasized.

3. DROP GENERATION AND DROP VOLUME

We start first by dispensing a liquid such that a dripping state is achieved. As shown in Fig.1, there are three forces that act on the pendant drop. There is a mass force due to gravity, a buoyancy effect due the immiscibility and a tension that holds the drop attached to the injector caused by interfacial tension.

When the mass force overcomes the latter two, the drop detaches from the capillary tip. One can consider that in the vicinity of drop detachment the drop is completely formed, therefore the volume of the drop is simply the product between the flow rate and the time between two consecutive detachment events. That results in a relation that offers the drop generation time:

\[ F_G - F_A \geq F_\sigma \Rightarrow \Delta \rho g Q_0 t_b \geq 2 \pi R_0 \sigma \Rightarrow t_b \approx \frac{2 \pi R_0 \sigma}{\Delta \rho g Q_0} \]  

(2)
where: $\Delta \rho$ is the density difference between the injected fluid and the outer one, $\sigma$ is the interfacial tension, $g$ is the gravitational acceleration, $Q_0$ is the flow rate of injection, $R_0$ is the nozzle inner radius and $t_b$ is the drop generation time. This time was measured and compared with the predictions of Eq. 2 for several flow rates. Fig. 3-a shows the dependence of the drop generation time on the flow rate. As the flow rate is being increased the drop generation time decreases in a hyperbolic manner, which is represented by a linear decrease in a log-log plot.

The experimental points follow the same pattern, with the observation that all values are slightly higher than those predicted. This is generally attributed to the fact that the pendant drop is not a perfect sphere. A pre-factor for the tension force is needed [5], which is found to be a function of the radius of the capillary tip and $\sigma/\rho g$. This present paper does not treat the problem of nonsphericity.

One way to obtain the volume of a drop is by measuring the drop diameter. We found that for flow rates below 6 ml/min, the diameter of the drop does not depend on the feeding flow rate. Taking an average value of this diameter and calculating an average drop volume, one can compare it with the products between individual flow rates and the measured drop generation times (Fig. 3-b). In this manner one can observe the limit in flow rate above which the volume of the drop will be influenced by the flow rate. A flow rate above this limit will add momentum and increases recirculation inside the drop, making Eq. 2 not suitable for describing drop detachment.

Another aspect of drop detachment is seen in relation with changing material properties of a liquid-liquid system. By keeping the external medium fixed and increasing the concentration of glycerol of the injected liquid, the diameter of the drop presented variations. Eq. 2 can be used to give a prediction of the diameter of the drop as follows:

$$
\Delta \rho \frac{\pi D_D^3}{6} \approx 2\pi R_0 \sigma \Rightarrow D_D \approx \sqrt[3]{\frac{12R_0 \sigma}{\Delta \rho g}}.
$$

(3)

Increasing the concentration of glycerol results in a decrease in drop diameter due to a decrease in interfacial tension (see Fig. 2) and due to the increase in density. The values predicted by Eq. 3, in comparison with measured values, are presented in Table 2. The relative errors in determining the diameters were found to be less than 10%.
Table 2. Theoretical prediction for the diameter of the drop, $D_D$, in comparison with measured values, $D_m$, when increasing the concentration of glycerol.

<table>
<thead>
<tr>
<th>$c$ (%)</th>
<th>0</th>
<th>50</th>
<th>75</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\beta$ (-)</td>
<td>0.02</td>
<td>0.12</td>
<td>0.97</td>
</tr>
<tr>
<td>$D_D$ (mm)</td>
<td>5.8</td>
<td>3.8</td>
<td>3.4</td>
</tr>
<tr>
<td>$D_m$ (mm)</td>
<td>6</td>
<td>4.2</td>
<td>3.6</td>
</tr>
<tr>
<td>Relative error (%)</td>
<td>3.3</td>
<td>9.5</td>
<td>5.5</td>
</tr>
</tbody>
</table>

The effect of increasing the viscosity of the inner fluid, or that of the outer medium, can be seen not only in an increase or decrease in drop diameter, but also on the detachment process itself. Fig. 4 presents three cases of drop detachment, one of which highlights the transition state between the dripping regime and the jetting regime. The transition from dripping to jetting is characterized by a critical Weber number, which in turn is a function of the Bond number [2]. A highly viscous outer or/and inner fluid can produce the migration of the detachment point further away from the capillary tip as seen in Fig. 4-b. The transition mentioned above is a transient state, usually difficult to model due to its chaotic nature.

Fig. 4 Drop detachment in three cases: a) dripping regime: 10% glycerol in water immersed in mineral oil, b) dripping regime: 75% glycerol in water immersed in silicon oil, c) :transition: 10% glycerol in water immersed in silicon oil

4. THINNING DYNAMICS OF NEWTONIAN FILAMENTS IN THE PRESENCE OF ANOTHER LIQUID

The actual pinch-off of the fully formed drop is a matter of material properties. As the drop detaches from the tip, a thinning filament appears connecting the tip and the drop. Filament breakup is driven by capillarity. For Newtonian filaments, the resistive force can be either inertia or viscous in nature. The non-dimensional parameter that quantifies the relative balance between viscous stresses and inertia-capillary effects is the Ohnesorge number, defined as:
Oh = \eta_i/\sqrt{\rho_i\sigma D(t)} ,

where: \eta_i and \rho_i are the viscosity and the density of the injected liquid, D(t) being the diameter of the filament. When Oh numbers are below 0.2, inertia is the resistive force that acts against capillarity. Above this value the viscous effects will dominate. As the filament thins, it can undergo different stages. For a given set of material properties, it can first go through an inertia-capillary process, and as the characteristic length scale decreases, i.e. the filament diameter, the thread enters a capillary-viscous regime. The process is quite fast when the external fluid is in a gaseous state, due to high values of superficial tension and low external viscous stresses. The process can be slowed down if the external fluid is a liquid. The effect of an external medium is a recent research topic because of the complexity added by doubling the parameters needed to fully describe the phenomenon [6].

This present paper is concerned with behaviour of two such filaments in the presence of mineral oil as an external liquid. Both substances are Newtonian liquids, the first one is a mixture of water and 50% glycerol and the second water and 75% glycerol. The liquids were chosen such that they would present Oh numbers, for the filament thinning process, below and above the critical value of 0.2. In order to characterise globally the necking phenomenon we propose a relation for the evolution of the diameter based on Tomotika’s linear stability analysis of a viscous thread surrounded by another viscous liquid [7].

Fig. 5 a) Global thinning behavior of two Newtonian filaments in mineral oil, representative cases for inertia controlled thinning, Oh < 2, and viscous controlled thinning, Oh > 2; b) Dispersion relations for the growth rate as a function of the non-dimensional wave number for the mixtures used to exemplify global thinning behavior

We consider that the thinning process is induced by a perturbation and that the initial diameter of the filament is decreasing in time by an amount proportional to the maximum growth rate of disturbance predicted by [8]. The equation that describes the evolution of the filament can be written in the following form: D(t) = D_0 - \xi_0 e^{\omega t}, where: D_0 is the initial diameter of the filament, \xi_0 is the initial amplitude of the disturbance and \omega is the maximum growth rate (see Fig. 5-b). For both fluids the thinning behaviour is similar, but the time to actual pinch-off, for the mixture with Oh < 2, is approximately two times smaller than that of the second liquid with Oh > 2. Taking the logarithm of Eq. 4, one can employ a linear regression method to find the optimal value for the initial amplitude of disturbance:

\xi_0 = n \prod_{i=1}^n (D_0 - D_i) \exp\left(\omega \sum_{i=1}^n t_i \right).
The values for the initial amplitude in both cases are: 0.2% and 1.3% of the inner diameter of the injector.

Fig. 5-a shows that the prediction for the mixture with a concentration of 50% glycerol is in good agreement over the entire time interval. For the second substance the prediction overestimates the breakup time.

A possible cause for this behaviour could be the nonlinear nature of the dynamics close to pinch-off.

5. DROPLET MOTION IN A VISCOUS LIQUID

After pinch-off, the drop starts to move through the surrounding medium. The equation of motion for a non-deforming drop in the limit of low Reynolds numbers is:

\[ F_G - F_A - F_S = \rho V_D \frac{dv}{dt} \]  

where: \( V_D = \frac{\pi D_D^3}{6} \), \( F_G = \rho g V_D \), \( F_A = \rho_e g V_D \), \( F_S = 3\eta \pi D_D v \) are the drop volume, the mass force, Archimedes force and Stokes friction force, respectively, indices “i” and “e” representing a material property of the injected or the exterior fluid.

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Fig. 6  
(a) Comparison between Stokes flow prediction for the velocity of the drop and the corrected form;  
(b) Drop acceleration as a function of time;  
(c) Prediction and experimental values for drop-travel distance;  
(d) Newtonian vs. viscoelastic necking phenomenon and subsequent motion  
(in a), (b), and (c) the drop is a 50% glycerol in water mixture moving in mineral oil; the viscoelastic fluid is 0.06 % polyacrylamide in water)
Introducing these relations in Eq. 6 and considering as initial condition zero velocity at \( t=0 \), yields the following ordinary differential equation and its solution:

\[
\frac{dv}{dt} = b + av \\
v(t) = \frac{b}{a} (e^{at} - 1) \\
v(t) = \frac{\Delta \rho g V_D}{3 \pi \eta_D} (1 - e^{at})
\]  

where: \( a = -3\pi \eta_e D_e/\rho_i V_D \) and \( b = \Delta \rho g/\rho_i \).

The solution is highlighted in Fig. 6-a by the blue line. Because of the fact that the Reynolds number is above the critical value for Stokes flow to hold, a correction is needed for the drag force [9]. Including this, aspect Eq. 6 becomes:

\[
\frac{dv}{dt} = b + av \left(1 + \frac{1}{6} Re^{2/3}\right)
\]

where: \( Re = \rho_e v D_e/\eta_e \).

The equation was solved in Mathematica® and its prediction is represented by the red line in Fig. 6-a. As expected the increase of the drag force reduces considerably the terminal velocity of the drop.

Drop acceleration, Fig. 6-b, and drop-travel distance, Fig. 6-c, reveal that the drop rapidly attains constant velocity, which can also be seen in the linear dependence on time of the travel distance.

The experimental values of the travel distance are found to be in good agreement with theoretical predictions. Although not pursued here, it is worth mentioning that the presence of elasticity causes the delay of the breakup process and therefore modifies the travel distance variation in time (Fig. 6-d).

Longer filaments are seen connecting the drop and the tip of the nozzle, the extra resistive force caused by elastic stresses influencing the motion of the drop.

### 6. CONCLUSIONS AND FUTURE WORK

Dispensing a liquid in an environment other than air changes the time scales associated with drop detachment and drop motion. The interfacial tension presents lower values than its corresponding surface tension, for a specific liquid in question. Decreasing interfacial tension by increasing density produces a decrease in drop diameter.

The frequency of drop generation exhibits a linear dependence on the flow rate of injection, behavior encountered only in the dripping regime. The volume of a detaching drop may or may not vary with the flow rate.

We found that below a certain flow rate the volume of the drop remains constant. Increasing the concentration of glycerol added in water results in an increase of viscosity and density, and a decrease in interfacial tension. This increase has a decreasing effect on the diameter of a detaching drop.

The necking phenomena that is seen when the drop detaches from the capillary tip, is a rapid process, which can be slowed down by considering a viscous outer liquid. A first observation can be made concerning the influence of viscosity on the time needed for the filament to rupture.

Experimental results show that increasing the viscosity of the injected liquid increases the breakup time of the filament. When the thinning process is resisted by inertia, we found that globally, the evolution of the filament can be approximated by an exponential decay...
based on linear stability analysis. If the resistive stresses are predominantly viscous then the predictions overestimate the breakup time.

Moving drops in viscous outer liquids attain constant velocity rapidly after pinch-off. For a non-deforming drop, a correction for the drag force from Stokes theory is needed in order to accurately predict the velocity of the drop.

Further attention will be given to the transition from dripping to jetting (Fig. 7-a), in the presence of viscous outer liquids, identifying the threshold value of characteristic non-dimensional parameters.

Future studies will include the filament thinning process in the case of a liquid-liquid system with a viscosity ratio equal to unity (Fig. 7-b).

Also the influence of elasticity, Fig. 7-c will be investigated in terms of filament breakup time and drop-travel distance.

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