Pinch-off of a stretching viscous filament and drop transport

Christina Weickgenannt, Ilia V Roisman and Cameron Tropea
Institute for Fluid Mechanics and Aerodynamics, Center of Smart Interfaces, Technische Universität Darmstadt, D-64287, Germany
E-mail: roisman@sla.tu-darmstadt.de
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Abstract
This study is focused on the stretching and breakup of a Newtonian liquid bridge between two parallel plates, one of which moves with a constant acceleration. The experimental device is designed to achieve high acceleration (up to 200 m s⁻²) with high precision. The shape evolution of the liquid bridge and its pinch-off are captured using a high-speed video system. At high enough acceleration the evolution of the midpoint diameter of the bridge is universal; it depends neither on the acceleration nor on the liquid viscosity. Moreover, at high acceleration rates even the breakup time does not depend on the acceleration, but is determined solely by the liquid viscosity. A model is proposed which predicts the instant of the liquid bridge pinch-off. Finally, the volumes of the residual liquid on both fixed and moving plates (which do not include the total volume of the residual secondary drops) is measured. It is shown that the volume on the fixed plate remains almost constant, the volume on the accelerating plate reduces as the acceleration increases.

1. Introduction

The dynamics of liquid bridges have been studied extensively for over one hundred years, starting with Plateau [1] and Lord Rayleigh [2]. Current studies are motivated by a wide variety of practical applications, including floating-zone crystallization, oil recovery, rheological measurements, high-purity crystallization, electrospinning of nanofibers, as well as industrial printing processes such as gravure, flexography, lithography or roll coating [3, 4].

A comprehensive review of the modeling approaches and experimental results on the problems related to jet dynamics can be found in [5, 6], while [7] is focused on the review of studies of the breakup and atomization of jets and films. An experimental study on the deformation of Newtonian liquid bridges has been performed in [8]. The corresponding phenomena are especially complicated in the case of non-Newtonian, rheologically complex fluids. Examples in which non-Newtonian and Newtonian liquids undergo significant elongational stretching are fiber-spinning, extrusion, wet particle collisions in granular flows, and injection molding. Moreover, stretching of a liquid bridge is often used for characterization of the rheological response of polymer solutions and other complex liquids to uniaxial extension [9, 10, 11].

A quasi one-dimensional theory of jet dynamics [12] is a powerful tool for modeling of the flow in a jet and analysis of its instabilities. A similarity solution for pinch-off [13] predicts the rate of the diameter evolution depending on surface tension and viscosity. Solutions for the flow in viscous threads [14] are complemented by finite element computations using a quasi one-dimensional slender jet approximation [15]. These solutions provide insight into the breakup dynamics by analyzing the contour, pressure and velocity distribution simultaneously. Study [16] demonstrated that a one-dimensional long-wave model, as used in several of the above cited studies, exhibits good agreement with 2D axisymmetric computations. Therefore such computations have become a common approach when studying liquid jets and bridges [17].
In a recent study of an extremely fast liquid bridge stretching of low viscosity liquid bridges [18] the break-up length is determined only by the initial bridge geometry. The question still remains on how viscous liquid bridges behave at such conditions.

In many applications and hydrodynamic problems a model is required for the maximum elongation of a stretching bridge, the number and the typical size of the drops produced by breakup, and the typical forces applied by the bridge to the moving end plates. For all these cases key scales are the typical time of bridge stretching until breakup and typical bridge diameter at this instant. The main goal of the present study is to obtain a better understanding of the mechanisms of liquid bridge breakup caused by fast accelerated stretching and to develop a model able to predict the moment of pinch-off.

2. Experimental method

2.1. Experimental setup

An experimental facility, shown schematically in figure 1, induces liquid bridge stretching under controllable and highly reproducible conditions. The apparatus consists of two parallel glass plates, a linear drive system, and a high-speed video system. The upper plate is mounted at a fixed position while the lower one is connected to the linear drive system in order to achieve a downward acceleration. The camera is positioned horizontally and records the stretching and break-up process as well as the motion of the contact line on the end plates during the stretching. The camera resolution at the object plane is 6 μm per pixel. It can also be positioned above the plates to visualize the drop remaining on the stationary plate. The glass plates are covered by a polymer foil. The linear drive operates with a positioning accuracy of 5 μm, while the speed is adjustable in the range of $U = 0.1 \text{ mm s}^{-1}$ up to 4 m s$^{-1}$ with a maximal achievable acceleration of $a_{\text{max}} = 200 \text{ m s}^{-2}$.

A series of experiments with various Newtonian liquids is performed at a constant rate of acceleration ranging between $a = 11.5$ and 170 m s$^{-2}$. The liquid viscosity was varied between $\mu = 10^{-3} \text{ N s m}^{-2}$ and $\mu = 0.96 \text{ N s m}^{-2}$ using water and aqueous glycerin solutions of different concentrations.

2.2. Observations of bridge stretching

One of the great advantages of the setup is that a comprehensive investigation on the influence of stretching dynamics on bridge deformation and breakup is possible. The shadowgraphy images obtained in the experiments visualize the shape evolution of the liquid bridges during the stretching process. Sample results are shown in the image sequence in figure 2, where a water liquid bridge of volume $V = 3 \mu l$ is stretched and the process is recorded until complete breakup of the bridge. Four sequences are shown, which correspond to stretching accelerations of $a = 11.5, 65, 125, 170 \text{ m s}^{-2}$.

The first image sequence shows the deformation of a liquid bridge that is stretched with a low plate acceleration of $a = 11.5 \text{ m s}^{-2}$. The sequence demonstrates the typical phenomena that are reported by numerous authors for the case of slow stretching. A tiny thread forms between the two volumes at the plates, which finally breaks up.

With increasing stretching acceleration the liquid bridge can be stretched longer and the developing shape becomes more and more columnar like. The breakup location is no longer in the middle region as for the case of slow stretching but moves toward the edges.

Apart from the known effects of increasing stretching dynamics, additional peculiarities are ascertained that are only observable due to the systematic variation of the acceleration. With increasing plate acceleration the
ligament heads deform unsymmetrical to each other. The volume part that is connected to the moved plate deforms more than the counter part at the fixed plate. Also the deformation of the middle part is influenced by acceleration. Comparing the third image of every sequence, where the height of the bridge is the same, one can see that the diameter of the middle region increased with increasing plate acceleration. In addition a delay between the breakup at the top and bottom plate develops for higher plate accelerations. The resulting ligament is much thicker, than for the case of slow stretching.

At some instant the jet is pinched at the menisci regions, leading to the detachment of the cylindrical part of the jet. The length of this free jet quickly reduces due to the formation of the globes at its ends. The formation of the globes is similar to the rim formation at the edge of a free liquid sheet [19, 20]. Finally, if the initial jet length is large enough the jet breaks up into several drops due to the Rayleigh capillary instability.

The same experiments are performed with ethylene glycol, which has a lower surface tension and a much higher viscosity than water. Results are shown in figure 3. It is clearly visible that the described phenomena are the same for the different liquid properties. The smaller contact angles of the initial shape of the liquid bridge demonstrates the influence of the lower surface tension. Since the adhesive forces are stronger, the contact line at the moveable plate is fixed even for the case of the highest acceleration. In contrast the contact angle becomes much smaller than for water in this case. In the following process significant differences are observable for ethylene glycol. The ligament is stretched much longer for all accelerations, while the time until the first breakup also increases.

In order to characterize the kinematics of the bridge stretching, the diameter $D$ of the bridge midpoint is measured at various instances. The results of the measurements are shown in figure 4. At relatively low plate accelerations, $a = 10 \text{ m s}^{-2}$, the rate of the jet thinning is reduced with increasing viscosity, as shown in figure 4(a). The diameter evolutions are similar for low viscosity water and for highly viscous glycerin solution, figure 4(b).

In figure 5 the evolution for the midpoint diameter is shown for the relatively low acceleration, $a = 10 \text{ m s}^{-2}$ (in figure 5(a)) and for high acceleration, $a = 160 \text{ m s}^{-2}$ (in figure 5(b)).

It is interesting, that at higher acceleration, shown in figure 5(b) ($a = 160 \text{ m s}^{-2}$), the influence of the liquid viscosity on the evolution of the jet diameter is negligibly small. The experiments also show that this diameter evolution only weakly depends on the plate acceleration, if it is high enough (higher than 100 m s$^{-2}$ in our experiments). This means that at such high accelerations the velocity of the plate, $at$, is higher than the velocity

**Figure 2.** Stretching of water liquid bridge at different plate accelerations. (a) $a = 11.5 \text{ m s}^{-2}$, (b) $a = 65 \text{ m s}^{-2}$, (c) $a = 125 \text{ m s}^{-2}$, (d) $a = 170 \text{ m s}^{-2}$.
of the capillary wave, \( \sim \sqrt{\sigma/(\rho D)} \), propagating along the jet. The information about the plate velocity does not propagate to the jet midpoint. The jet stretching is governed mainly by inertial effects. This assumption is confirmed by the linear dependence of the value of \((D_0/D)^2\) at relatively large times \((t > 10 \text{ ms in figure 6})\) as predicted in (3).

Figure 3. Stretching of ethylene-glycol liquid bridge at different plate accelerations. (a) \(a = 11.5 \text{ m s}^{-2}\), (b) \(a = 65 \text{ m s}^{-2}\), (c) \(a = 125 \text{ m s}^{-2}\), (d) \(a = 170 \text{ m s}^{-2}\).

Figure 4. Evolution of the midpoint diameter of a liquid bridge: comparison for two various liquid viscosities. The liquid bridge volume \(V = 3 \mu\text{m}\) and the initial gap thickness between the plates \(H_0 = 0.8 \text{ mm}\) are constant for all the experiments. (a) Water bridge; (b) glycerin solution bridge.
Note that the graphs in figures 4 and 5 do not provide information about the breakup time, since the pinch-off occurs near the menisci and this event does not immediately influence the evolution of the midpoint diameter.

3. Inertia dominated flow in a stretching liquid jet

The dynamics of a nearly cylindrical jet stretching is already well understood [12]. The flow in the bridge is governed by the momentum and mass balance equations. If the jet is nearly cylindrical (far from the menisci regions) and the flow is axisymmetric these equations can be linearized and significantly simplified. The expression for the axial force \( F \) applied to a cross-section of a circular jet of the radius \( R \) accounts for the internal viscous stresses and the force associated with surface tension \( \sigma \), linearized in respect to small \( R_z \),

\[
F \approx 3\mu\pi R^2 u_z + \sigma \pi R \left( 1 + RR_{zz} \right),
\]

where \( z \) is the axial coordinate and \( u \) is the axial liquid velocity, assumed to be nearly uniform through the cross-section. The momentum balance equation in the axial direction and the mass balance yield [12]

\[
\rho \pi R^2 \left( u_t + uu_z \right) + F_{,z} = 0, \quad 2R_t + 2uR_{,z} + R u_{,z} = 0.
\]

Figure 5. Evolution of the midpoint diameter of a liquid bridge: comparison for two different accelerations. The liquid bridge volume \( V = 3 \mu m \) and the initial gap thickness between the plates \( H_0 = 0.8 \) mm are constant for all the experiments. (a) Low acceleration, \( a = 10 \) m s \(^{-2}\); (b) high acceleration, \( a = 160 \) m s \(^{-2}\).

Figure 6. Evolution of the midpoint diameter of a liquid bridge at various liquid viscosities at relatively high acceleration, 160 m s \(^{-2}\). Measurements confirm remote asymptotic solution (3).

Note that the graphs in figures 4 and 5 do not provide information about the breakup time, since the pinch-off occurs near the menisci and this event does not immediately influence the evolution of the midpoint diameter.

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4. Pinch-off time

The results of measurements of the pinch-off times $t_{br}$ at the meniscus near the fixed plate are shown in figure 7 for liquids of various viscosities. To demonstrate the repeatability of the experiments the data from three experiments are shown for each set of parameters. It is interesting that for all the liquids the breakup time first reduces with the plate acceleration but then, starting from approximately $a = 40 \text{ ms}^{-2}$, remains almost constant. The dashed lines in figure 7(a) corresponds to the best fit of the data in the form $t_{br} = t_{br}(\mu) + Ba^{-1/2}$, where the fitting parameter $B$ is the same for all the experimental data, shown in figure 7(a), and the parameter $t_{br}$ significantly depends on the liquid bridge viscosity.

There is a very clear increase of $t_{br}$ for higher liquid viscosities, as shown in figure 7(b).

Respectively, for relatively large plate accelerations the breakup length of a liquid viscous bridge increases nearly linearly with $a$. This result is illustrated in figure 8.

The process of the bridge stretching before pinch-off can be subdivided into two stages. The first stage is the initial bridge deformation when the plate velocity is relatively small. The bridge shape is approximated well by static meniscus surfaces, classified by Plateau [22] and determined by the condition of constant curvature. It is known that for a given liquid volume in a bridge a maximum distance between solid bodies exists for which a static shape can still exist [23]. For a liquid bridge volume $V = 3 \mu l$ and initial gap thickness $H_0 = 0.6 \text{ mm}$ the maximum static gap between two plates is $H_{\text{static}} = 1.1 \text{ mm}$. The value of $H_{\text{static}}$ is determined by the position until which all the investigated curves coincide. The dependence of the midpoint diameter on the distance $H$ between the plates is almost identical for all the investigated liquid viscosities and accelerations on the interval $H < H_{\text{static}}$. The duration of this quasi-static bridge stretching is therefore $t_{\text{static}} = \sqrt{2(H_{\text{static}} - H_0)/a}$. The value of $t_{\text{static}}$ decreases with increasing acceleration. This effect is the main reason for the larger pinch-off times for smaller accelerations, shown in figure 7(a). At higher accelerations the time $t_{\text{static}}$ is negligibly small in comparison to $t_{br}$. This means that the value of $t_{br} - t_{\text{static}}$ does not depend on the plate acceleration.
At larger times the flow in the liquid bridge quickly approaches the long-time approximation (3). The constant $\eta$ has dimensions of square root of time. Since it does not depend on the viscosity, because the bridge is initially stretched by surface tension, it can be represented in the form $R_0^3 \sim \eta \rho \sigma$. The radius and the velocity gradient in the jet at large times $t \gg \tau$ are scaled therefore as $R \sim R_0^{1/2} \rho^{1/2} \sigma^{-1/4} t^{-1/2}$ and $u_z \sim t^{-1}$, respectively.

In most of our experiments the breakup occurs first near the static meniscus and then after a very short delay near the moving plate. The inertial effects near the static plate are negligibly small in comparison to the viscous and capillary effects even for high accelerations. Therefore, the shape of the meniscus is determined by the balance of the surface tension applied to the wall and the viscous force applied to the jet cross-section at the jet exit region from the meniscus. The viscous part of the force applied to the jet cross-section are estimated with the help of (1) as

$$F_\mu \sim \frac{\mu \rho^{1/2} R_0^{7/2}}{\sigma^{1/4} t^{1/2}}.$$  

This force applied to the jet cross-section ensures the concave shapes of the menisci, matching the nearly cylindrical jet between them. When this force reaches some critical minimum value $F_\sigma$, the meniscus starts to quickly approach its static shape of truncated sphere. This critical force is determined by the meniscus geometry and surface tension and thus can be scaled as $F_\sigma \sim R_0 \sigma$, since the radius of the wetted part of the plates does not change significantly during fast bridge stretching.

The force $F_\sigma$ reduces in time. A hypothesis that the pinch–off occurs when $F_\sigma \approx F_\mu$ yields the following scaling for the breakup time

$$t_{br} \sim T \equiv \frac{R_0^{5/4} \rho^{1/4} \mu^{1/2}}{\sigma^{3/4}}.$$  

This scaling, which predicts that the breakup time $t_{br}$ is proportional to $\mu^{1/2}$ for high accelerations is qualitatively supported by the measurement results shown in figure 7(b).

Since the breakup time depends on the initial geometry of the liquid bridge, namely on the initial radius $R_0$ and the initial gap thickness between the plates $H_0$, we introduce an additional dimensionless parameter, $k = H_0 / R_0$, and try to present the experimental data in the form

$$t_{br} = \beta k^\alpha T,$$

where $\beta = 5.99$ and $\alpha = 0.5$ are obtained from the best fit to the experimental data.

The dependence of the measured breakup time on $k^{0.5} T$ is shown in figure 9. In the experiments the liquid volume ranges from 1 to 6 $\mu l$, the viscosity ranges from 0.001 to 0.455 N s m$^{-2}$, and the parameter $k$ is varied from 0.26 to 0.9. Only the results for high plate acceleration are presented in figure 9, corresponding to the saturation of the dependence of $t_{br}$ on $a$ (as seen in figure 7). The nearly linear dependence between the terms, shown in figure 9, can be interpreted as evidence supporting the hypothesis that the pinch–off occurs at the instant when the forces applied to the bridge cross-section is comparable to the critical tension ensuring the concave shape of the menisci.

At lower plate accelerations the breakup time is influenced by the initial time of bridge deformation, which can be roughly estimated as $t_{static} \approx (2R_0/a)^{1/2}$, since the maximum static length of a liquid bridge is comparable with its radius. The final proposed correlation for the breakup time, valid for an entire range of plate accelerations (from $a = 5$ to $a = 160$ m s$^{-2}$) is obtained in the form

$$t_{br} = 5.99 k^{0.5} T.$$  

Figure 9. Breakup time of a liquid bridge as a function of the time scale $k^{0.5} T$ at high plate accelerations, for which $t_{br} \gg t_{static}$.
In Figure 10 the experimental data for the breakup time is compared with the predictions obtained using correlation (8). The straight line in the graph corresponds to perfect agreement. The agreement is rather good, which indicates that our scaling analysis is applicable also to the lower magnitudes of plate acceleration.

It should be noted that correlation (8) is obtained only for the breakup times corresponding to the bridge pinch-off near menisci. At higher viscosities the measured pinch-off times exceed the times of Rayleigh capillary instability of stretching cylindrical jets, which leads to the jet breakup and formation of several drops, observed before the pinch off time. The influence of this instability is indicated in Figure 10 by some deviation of the measured breakup times from the predicted at higher $t_{br}$, corresponding to higher viscosities.

In the limiting case of zero viscosity the first term in the right hand side of (8), which includes the characteristic viscous time $T$, disappears, leading to the dependence of the breakup time on acceleration in the form $t_{br} = 5.99k^{0.5}T + \sqrt{\frac{2K_0}{a}}$.

In Figure 11 the experimental data for the breakup time is compared with the predictions obtained using correlation (8). The straight line in the graph corresponds to perfect agreement. The agreement is rather good, which indicates that our scaling analysis is applicable also to the lower magnitudes of plate acceleration.

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In the limiting case of zero viscosity the first term in the right hand side of (8), which includes the characteristic viscous time $T$, disappears, leading to the dependence of the breakup time on acceleration in the form $t_{br} = t_{static} = (R_0/a)^{1/2}$, the same as recently obtained in [18]. The time $t_{static}$ is defined only by the initial bridge geometry. For high accelerations and high liquid viscosities, typical for the drop transport processes in printing technology, this term is negligibly small and can be neglected.

5. Drop transport

The amount of liquid that remains on the plates after bridge breakup is investigated in terms of plate acceleration and liquid viscosity. The initial aspect ratio of the bridge does not vary in these experiments. Figure 11 shows that the volume fraction that remains on the fixed plate is more or less constant for increasing acceleration. On the moving plate one measures a decrease in the amount of liquid after breakup. This is due to the fact that the liquid bridge head at the moving plate contracts more for large plate accelerations. Correspondingly more liquid remains inside the ligament after breakup.
The asymmetry of a stretched liquid bridge has been observed before for the capillary driven stretching. For non-Newtonian liquid bridges, observed in [24], the asymmetry is caused by a self-similar pinch off of a liquid droplet. In our case the asymmetry is initiated mainly by the inertial forces associated with the plate acceleration. Liquid viscosity does not have a clear influence on the amount of liquid transfer. It can only be stated that the most liquid is transferred from the moving plate in the case of a water bridge. This is due to the fact, that the contact line of the bridge moves for a low viscous liquid like water, while it is more or less fixed when viscosity becomes larger, [25]. To verify this assumption experiments with Teflon surfaces are performed. The receding contact angle of the different liquids is measured on the surface by deflating a drop with a syringe. When the volume is withdrawn the contact angle recedes, approaching a minimum value referred to as the receding contact angle. This value is between $\theta = 78^\circ$ and $89^\circ$ for the different liquids.

To demonstrate the effect of a large receding contact angle on the bridge stretching experimental images of the breakup moment are shown for water and a 80% glycerin solution in figure 12. In both experiments the bridges are stretched at a high plate acceleration. For the case of water it can be seen that the contact line contracts drastically at the Teflon surface, as one would expect. In contrast, for the case of glycerin, the contact line is more or less pinned, even though the measured receding contact angle was $78^\circ$.

6. Conclusions

We have studied drop transport between two plates, as a result of a fast bridge stretching. The moving plate moves with a constant acceleration, which is accurately controlled. We have demonstrated that the breakup time of a stretching Newtonian filament increases significantly with the liquid viscosity. The breakup length of the filament increases almost linearly on the plate acceleration, if this acceleration is high enough.

A model for the breakup time is proposed, which predicts very well the value of the breakup time for a wide range of the liquid viscosity and plate acceleration.

Finally, the residual volumes on the fixed and moving plates are measured. It is shown that the volume on the fixed plate only slightly depends on the plate acceleration. At high accelerations nearly half of the initial bridge volume remains on the fixed plate. However, the residual volume on the moving plate decreases as the acceleration increases. This means also that at higher accelerations more liquid volume of the bridge atomizes and generates small liquid drops.

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