Chapter 14

A BRIEF HISTORY OF DROP FORMATION

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- **Abstract** Surface-tension-related phenomena have fascinated researchers for a long time, and the mathematical description pioneered by Young and Laplace opened the door to their systematic study. The time scale on which surface-tension-driven motion takes place is usually quite short, making experimental investigation quite demanding. Accordingly, most theoretical and experimental work has focused on static phenomena, and in particular the measurement of surface tension, by physicists like Eötvös, Lenard, and Bohr. Here we will review some of the work that has eventually lead to a closer scrutiny of time-dependent flows, highly non-linear in nature. Often this motion is self-similar in nature, such that it can in fact be mapped onto a pseudo-stationary problem, amenable to mathematical analysis.
- Keywords: drop formation, hydrodynamics, non-linear fluid mechanics, stroboscopic method

Introduction

Flows involving free surfaces lend themselves to observation, and thus have been scrutinized for hundreds of years. The earliest theoretical work was concerned almost exclusively with the equilibrium shapes of fluid bodies, and with the stability of the motion around those shapes. Experimentalists, always being confronted with physical reality, were much less able to ignore the strongly non-linear nature of hydrodynamics. Thus many of the non-linear phenomena, that are the focus of attention today, had already been reported 170 years ago. However, with no theory in place to put these observations into perspective, non-linear phenomena took the back seat to other issues, and were soon forgotten.



Figure 14.1. A figure from Savart's original paper (Savart, 1833), showing the breakup of a liquid jet 6 mm in diameter. It clearly shows the succession of main and satellite drops as well as drop oscillations.

Here we report on the periodic rediscovery of certain non-linear features of drop formation, by retracing some of the history of experimental observation of surface tension driven flow. Recently there has been some progress on the theoretical side, which relies on the self-similar nature of the dynamics close to pinching.

1. Savart and Plateau

Modern research on drop formation begins with the seminal contribution of (Savart, 1833). He was the first to recognize that the breakup of liquid jets is governed by laws independent of the circumstance under which the jet is produced, and concentrated on the simplest possible case of a circular jet. Without photography at one's disposal, experimental observation of drop breakup is very difficult, since the timescale on which it is taking place is very short.

Yet Savart was able to extract a remarkably accurate and complete picture of the actual breakup process using his naked eye alone. To this end he used a black belt, interrupted by narrow white stripes, which moved in a direction parallel to the jet. This effectively permitted a stroboscopic observation of the jet. To confirm beyond doubt the fact that the jet breaks up into drops and thus becomes discontinuous, Savart moved a "slender object" swiftly across the jet, and found that it stayed dry most of the time. Being an experienced swordsman, he undoubtedly used this weapon for his purpose (Clanet, 2003). Savart's insight into the *dynamics* of breakup is best summarized by Fig.14.1 taken from his paper (Savart, 1833).

To the left one sees the continuous jet as it leaves the nozzle. Perturbations grow on the jet, until it breaks up into drops, at a point labeled "a". Near a an elongated neck has formed between two bulges which later become drops. After breakup, in between two such drops, a much smaller "satellite" drop is always visible. Owing to perturbations received when they were formed, the drops continue to oscillate around a spherical shape. Only the very last moments leading to drop formation are not quite resolved in Fig.14.1.



Figure 14.2. Breakup of a liquid column of oil, suspended in a mixture of alcohol and water (Plateau, 1849). First small perturbations grow, leading to the formation of fine threads. The threads each break up leaving three satellites.

From a theoretical point of view, what is missing is the realization that surface tension is the driving force behind drop breakup, the groundwork for the description of which was laid by (Young, 1804) and (Laplace, 1805). Savart however makes reference to mutual attraction between molecules, which make a sphere the preferred shape, around which oscillations take place. The crucial role of surface tension was recognized by (Plateau, 1849), who confined himself mostly to the study of equilibrium shapes. From this insight it follows as well whether a given perturbation imposed on a fluid cylinder will grow or not. Namely, any perturbation that will lead to a reduction of surface area is favored by surface tension, and will thus grow. This makes all sinusoidal perturbations with wavelength longer than 2π unstable. At the same time as Plateau, Hagen published very similar investigations, without quite mastering the mathematics behind them (Hagen, 1849). The ensuing quarrel between the two authors, published as letters to Annalen der Physik, is quite reminiscent of similar debates over priority today.

A little earlier Plateau had developed his own experimental technique to study drop breakup (Plateau, 1843), by suspending a liquid bridge in another liquid of the same density in a so-called "Plateau tank", thus eliminating the effects of gravity. Yet this research was focused on predicting whether a particular configuration would be stable or not. However Plateau also included some experimental sketches (cf. Fig.14.2) that offer interesting insight into the nonlinear dynamics of breakup for a



Figure 14.3. Two photographs of water jets taken by (Rayleigh, 1891), using a short-duration electrical spark.

viscous fluid: first a very thin and elongated thread forms, which has its minimum in the middle. However, the observed final state of a satellite drop in the center, with even smaller satellite drops to the right and left indicates that the final stages of breakup are more complicated: the thread apparently broke at 4 different places, instead of in the middle.

Following up on Plateau's insight, (Rayleigh, 1879) added the flow dynamics to the description of the breakup process. At low viscosities, the time scale τ of the motion is set by a balance of inertia and surface tension:

$$\tau = \sqrt{\frac{r^3\rho}{\gamma}}.$$
(14.1)

Here r is the radius of the (water) jet, ρ the density, and γ the surface tension. For the jet shown in Fig.14.1, this amounts to $\tau = 0.02 \ s$, a time scale quite difficult to observe with the naked eye. Rayleigh's linear stability calculation of a fluid cylinder only permits to describe the initial growth of instabilities as they initiate near the nozzle. It certainly fails to describe the details of drop breakup leading to, among others, the formation of satellite drops. Linear stability analysis is however quite a good predictor of important quantities like the continuous length of the jet.

2. Photography

Rayleigh was well aware of the intricacies of the last stages of breakup, and published some experimental pictures himself (Rayleigh, 1891). Unfortunately, these pictures were produced by a single short spark, so they only transmit a rough idea of the *dynamics* of the process. However, it is again clear that satellite drops, or entire sequences of them, are pro-



Figure 14.4. A sequence of pictures of a drop of water falling from a pipette (Lenard, 1887). For the first time, the sequence of events leading to satellite formation can be appreciated.

duced by elongated necks between two main drops. Clearly, what is needed for a more complete understanding is a sequence of photographs showing one to evolve into the other.

The second half of the 19th century is an era that saw a great resurgence of the interest in surface tension related phenomena, both from a theoretical and experimental point of view. The driving force was the central role it plays in the quest to understand the cohesive force between fluid particles (Rowlinson, 2002), for example by making precise measurements of the surface tension of a liquid. Many of the most wellknown physicists of the day contributed to this research effort, some of whom are known today for their later contributions to other fields (Eötvös, 1886, Quincke, 1877, Lenard, 1887, Bohr, 1909). A particular example is the paper by (Lenard, 1887), who observed the drop oscillations that remain after break-up, already noted by Savart. By measuring their frequency, the value of the surface tension can be deduced.

To record the drop oscillations, Lenard used a stroboscopic method, which enabled him to take an entire sequence with a time resolution that would otherwise be impossible to achieve. As more of an aside, Lenard also records a sequence showing the dynamics close to breakup, leading to the separation of a drop. It shows for the first time the origin of the satellite drop: first the neck breaks close to the main drop, but before it is able to snap back, it also pinches on the side toward the nozzle. The presence of a slender neck is intimately linked to the profile near the pinch point being very asymmetric: on one side it is very steep, fitting well to the shape of the drop. On the other side it is very flat, forcing the neck to be flat and elongated.

However, as noted before, few people took note of the fascinating dynamics close to breakup. From a theoretical point of view, tools were limited to Rayleigh's linear stability analysis, which does not include a



Figure 14.5. A drop of water (left) and a glycerol-alcohol mixture (right) falling from a pipette (Edgerton et al., 1937). The drop of viscous fluid pulls out long necks as it falls.

mechanism for satellite formation. Many years later, the preoccupation was still to find simple methods to measure surface tension, one of them being the "drop weight method" (Harkins and Brown, 1919). The idea of the method is to measure surface tension by measuring the weight of a drop falling from a capillary tubes of defined diameter. Harold Edgerton and his colleagues looked at time sequences of a drop of fluid of different viscosities falling from a faucet (Edgerton et al., 1937), rediscovering some of the features observed originally by Lenard, but adding some new insight.

Fig.14.5 shows a water drop falling from a faucet, forming quite an elongated neck, which then decays into *several* satellite drops. The measured quantity of water thus comes from the main drop as well as from some of the satellite drops; some of the satellite drops are projected upward, and thus do not contribute. The total weight thus depends on a very subtle dynamical balance, that can hardly be a reliable measure of surface tension. In addition, as Fig.14.5 demonstrates, a high viscosity fluid like glycerol forms extremely long threads, that break up into myriads of satellite drops. In particular, the drop weight cannot be a function of surface tension alone, but also depends on viscosity, making the furnishing of appropriate normalization curves unrealistically complicated.

3. Modern times

After Edgerton's paper, the next paper that could report significant progress in illuminating *non-linear* aspects of drop breakup was published in 1990 (Peregrine, 1990). Firstly, it contains a detailed sequence of a drop of water falling from a pipette D = 5.2mm in diameter, renewing efforts to understand the underlying dynamics. Secondly, it was proposed that close to pinch-off the dynamics actually becomes quite



Figure 14.6. A high-resolution sequence showing the bifurcation of a drop of water (Peregrine, 1990).



Figure 14.7. A sequence of interface profiles of a jet of glycerol close to the point of breakup (Kowalewski, 1996). The experimental images correspond to $t_0 - t = 350\mu s$, 298 μs , and 46 μs . Corresponding analytical solutions based on self-similarity of the entire profile are superimposed.

simple, since any *external* scale cannot play a role. Namely, if the minimum neck radius h_{min} is the only relevant length scale, and if viscosity does not enter the description, than at a time $t_0 - t$ away from breakup on must have

$$h_{min} \propto \left(\frac{\gamma}{\rho}\right)^{2/3} (t_0 - t)^{2/3}$$
 (14.2)

for dimensional reasons. At some very small scale, one expects viscosity to become important. The only length scale that can be formed from fluid parameters alone is

$$\ell_{\nu} = \frac{\nu^2 \rho}{\gamma}.\tag{14.3}$$

Thus the validity of (14.2) is limited to the range $D \gg h_{min} \gg \ell_{\nu}$ between the external scale and this inner viscous scale.

These simple similarity ideas can in fact be extended to obtain the laws for the entire profile, not just the minimum radius (Eggers, 1993). Namely, one supposes that the profile around the pinch point remains the same throughout, while it is only its radial and axial length scales which change. In accordance with (14.2), these length scales are themselves power laws in the time distance from the singularity. In effect, by making this transformation one has reduced the extremely rapid dynamics close to break-up to a static theory, and simple analytical solutions are possible.

The experimental pictures in Fig.14.7 are again taken using a stroboscopic technique, resulting in a time resolution of about $10\mu s$ (Kowalewski, 1996). Since for each of the pictures the temporal distance away from breakup is known, the form of the profile can be predicted without adjustable parameters. The result of the theory is superimposed on the experimental pictures of a glycerol jet breaking up as black lines. In each picture the drop about to form is seen on the right, a thin thread forms on the left. The neighborhood of the pinch point is described quite well; in particular, theory reproduces the extreme *asymmetry* of the profile. We already singled out this asymmetry as responsible for the formation of satellite drops.

One of the conclusions of this brief overview is that research works in a fashion that is far from straightforward. Times of considerable interest in a subject are separated by relative lulls, and often known results, published in leading journals of the day, had to be rediscovered. However from a broader perspective one observes a development from questions of (linear) stability and the measurement of static quantities, to a focus that is more and more on the (non-linear) dynamics that makes fluid mechanics so fascinating.

Acknowledgments

I have the pleasure to acknowledge very helpful input from Christophe Clanet and David Quéré.

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