

Fragmentation of viscous compound liquid ligaments

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Liquid is deposited between two rods, one of which is held by a spring and the other is kept fixed. When the spring is released, the liquid stretches into a filament, which eventually breaks up into droplets [1]. If the liquid viscosity is low, the first breakup occurs shortly after the spring is released; the filament pinches off at both ends and immediately fragments into droplets of various sizes. If the liquid is viscous, the first breakup is delayed and is eventually caused by the Rayleigh-Plateau instability. We describe the formation and destabilization of a compound filament, obtained by stretching a capillary bridge containing two immiscible liquids with a viscosity contrast.

Figure 1 shows pictures from a typical filament stretching experiment, in which the core represents 10% of the total volume. A drop of a water-glycerol mixture, dyed in blue, is injected in a capillary bridge of silicone oil, which is 50 times more viscous. Then this capillary bridge is stretched into a compound filament: a water-glycerol core inside an oil shell. Eventually, this filament breaks up into compound droplets: droplets of oil containing one or several droplets of the water-glycerol mixture. Depending on the relative volumes of the inner and outer liquid and also depending on their viscosity difference, the size distribution and the content of the compound drops are different.

Plateau realized that a small regular ripple at the surface of a cylinder reduces its surface area [2]. From there he deduced that surface tension makes a cylinder of liquid unstable, because surface energy tends to a minimum. This minimum is reached when the liquid is broken into spherical droplets. In the same period Lord Rayleigh derived the optimal wavelength of the instability, notably under the effect of viscosity [3]. This capillary mechanism is known as the Rayleigh-Plateau instability.

The wavelength is the distance between two consecutive ripples at the onset of destabilization. To each possible wavelength corresponds a mode of the instability, each of which grows at a specific pace. The optimal wavelength, corresponding to the fastest growing mode, sets the size of the droplets. For an inviscid fluid, the wavelength λ_{RP} of the fastest growing mode is proportional to the radius of the filament a : $\lambda_{RP} = 2\pi\sqrt{2}a$. If the fluid has a finite viscosity, the wavelength is

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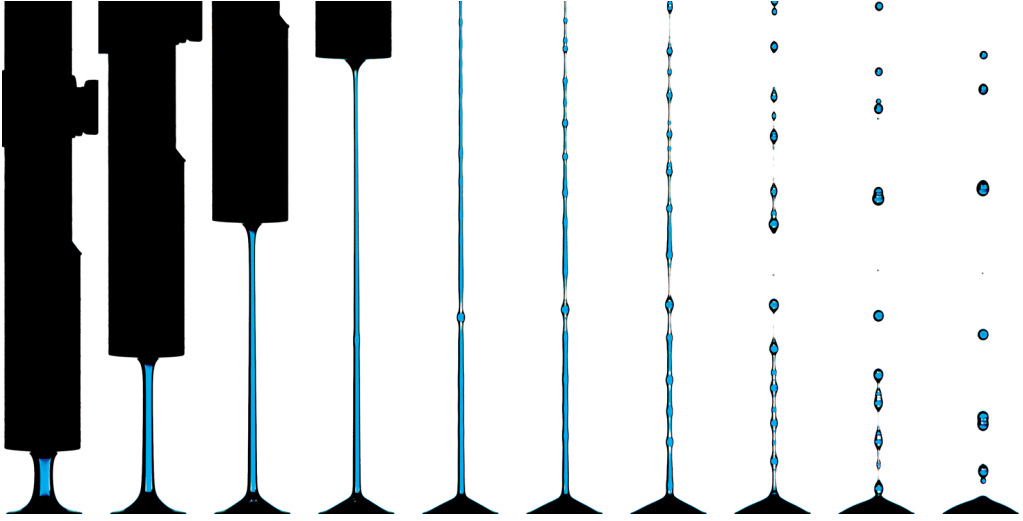


FIG. 1. Stretching and fragmentation of a capillary bridge composed of silicone oil containing a drop of a mixture of water (75% wt.) and glycerol (25% wt.), dyed in blue. The inner drop volume is 10% of the capillary bridge. The rod width is 9.4 mm; the time span between each image is 10 ms.

longer than λ_{RP} ; its expression can be approached by a long-wave approximation of the viscous Rayleigh-Plateau instability $\lambda_{VRP} = 2\pi a\sqrt{2 + 3\sqrt{2}Oh}$, where $Oh = \eta/\sqrt{\rho\gamma a}$ is the Ohnesorge number [4]. The wavelength increases with the viscosity; therefore the effect of viscosity is to make bigger droplets.

In the present experiment, two successive instabilities occur: that of the core and then that of the shell. Figure 2 shows the stretching of a filament containing an initial inclusion that accounts for

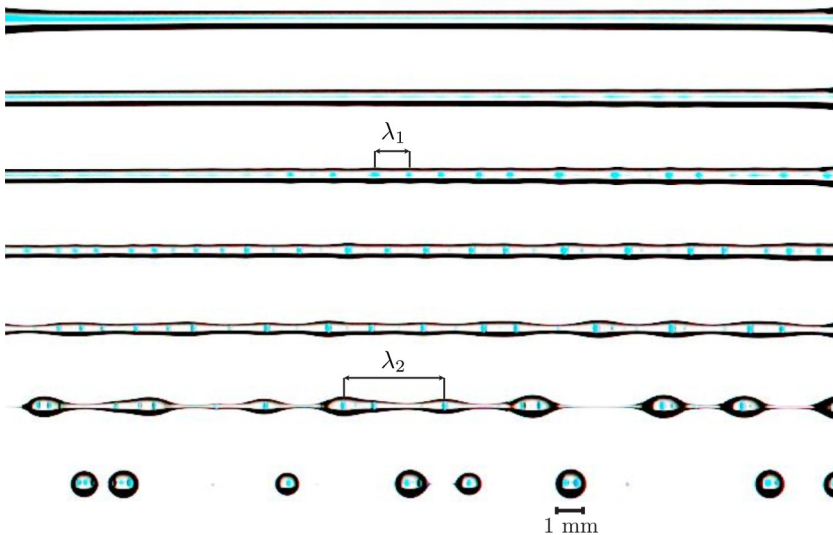


FIG. 2. Close-up view of the successive instabilities in the limit case of a very thin core. The inner water drop volume represents 1% of the capillary bridge. The core destabilizes with a wavelength λ_1 and then the shell destabilizes with a different wavelength λ_2 . The diameter of the biggest droplets is about 1 mm.

1% of the volume of the capillary bridge. In the limit case where the volume of the inner liquid is small compared to the volume of the capillary bridge, one observes a clear separation between the inner and the outer instability. The core destabilizes first because it is thinner and less viscous than the whole filament, and the resulting droplets force the destabilization of the filament. We can define two wavelengths λ_1 and λ_2 that correspond to the core and shell instability, respectively. Interestingly, we observe that the shell wavelength λ_2 (3.6 mm on average for the experiment shown in Fig. 2) is longer than the inviscid Rayleigh-Plateau instability ($\lambda_{RP} = 2.7$ mm), but shorter than the viscous Rayleigh-Plateau wavelength ($\lambda_{VRP} = 5$ mm). This means that the inner droplets that result from the core fragmentation force a shorter wavelength onto the shell. Moreover, as shown in the video, the destabilization of the compound filament is much faster than that of a filament of oil alone, even with as little as a 1%-volume inclusion.

The description of these coupled instabilities bears two main applications: the fragmentation of viscous liquids and the production of compound drops. As mentioned earlier, a filament of viscous liquid destabilizes through the Rayleigh-Plateau instability. We show that adding a nonviscous core enables us to tune the instability. This core could be miscible after some time, for example, two oils of different viscosity, to generate drops with a chosen size distribution. In the case where the liquids are completely immiscible, like oil and water, we obtain compound drops. Compound drops are liquid drops containing at least one other phase, for example, solid particles, bubbles, or droplets of another liquid [5]. They have great potential for industrial applications. For example, in-air microfluidics [6] consists in generating compound drops in flight to prepare controlled emulsions, at a rate ten times faster than classical chip-based microfluidics.

Experimental methods. A droplet of silicone oil is deposited on top of the bottom aluminum rod, using a micropipette. Then a droplet of the dyed water-glycerol mixture is injected inside the silicone oil, using another, smaller micropipette. The capillary bridge is formed by bringing the top rod in contact. The whole device is set against an LED panel for back lighting, and the dynamics of stretching and breakup are recorded using a high-speed camera (Phantom VEO 640) and a macro lens (Nikon 200 mm).

About the video. The video leads the viewer from simple to complex fragmentation. It begins by showing the effect of viscosity, which is to suppress end pinching and therefore to foster the Rayleigh-Plateau instability. It then describes the destabilization of several compound filaments, varying the volume of the nonviscous inclusion. The last sequence shows the complex yet beautiful destabilization of a double compound filament, in which a green and a blue drop interact in their stretching and breakup. The slow fragmentation, upon which the tiniest perturbations act, is sublimated by Satie's *Gymnopédie No. 1*, with its calm and soft and slightly irregular rhythm.

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