Hesitant Nature

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The addition of even minute amounts of a polymer to a liquid can modify some of its properties drastically, like the ways in which jets of the liquid break into droplets or the dynamics of dripping droplets at an orifice. The elasticity conferred by the polymers can delay the separation of the drops considerably, which hesitate between falling or running back to the orifice, 'gobbling' along their way the thread to which they are attached. Other free-surface flows are also of a hesitant nature.

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1. Introduction

In his memoirs, François Jacob, the co-laureate with André Wolf and Jacques Monod of the 1965 Nobel Prize in Physiology or Medicine, evoked the relationship he had with Monod. He explains the different conceptions they had about the way Nature works (Jacob 1987, translation from french by the author)

> Nature, I used to consider her as a fairly good girl. Generous, but somewhat sloppy. Somewhat muddled. Working bit by bit. Doing her best with what she had.

By contrast, Monod had a rigid view inherited from Cartesian reductionism of how Nature operates, obeying strict, permanent, elegant and universal rules.

The wealth of phenomena characteristic of free-surface flows offers a brilliant illustration of those many exceptions which prove the rule. Sometimes, Nature gives the impression of 'hesitating' between one state and another, between one rule and another. This is particularly true when capillarity, a fundamental force acting at the liquid/gas interface, is a relevant partner. This is even truer when additional ingredients come into play, as in the work of Clasen *et al.* (2009, this issue, vol. 636, pp. 5–40) who have considered the classical problem of liquid dripping from the orifice of a tap, when the liquid carries a minute amount of polymer. The polymer molecules of high molecular weight are long flexible chains, which stretch with the flow and resist the deformation from their equilibrium coiled state with a nontrivial relaxation time. These chains are thus able to store energy, resulting in elastic behaviour. Their addition to a liquid, even with concentrations as low as 10 p.p.m. have dramatic effects, since they completely alter the character of final jet breakup (Amarouchene *et al.* 2001). Polymers also alter drop dripping dynamics, the drop now hesitating between detaching and falling freely or remaining attached and running back to the orifice, 'gobbling' along the way the thread to which it is attached, as explained by Clasen *et al.* (2009).

FIGURE 1. The 'gobbling' phenomenon: a large terminal drop periodically develops at the end of a thin jet of a viscoelastic fluid (100 p.p.m. of polyacrylamide in aqueous solution). The drop initially moves upward, 'gobbling' the thread, before detaching. The time interval between the images is 6 ms (figure from Clasen *et al.* 2009).

2. Overview

For a jet to be formed with a substantial continuous portion along its axis when issuing from a tap at velocity V, the transit time λ/V of the fluid particles over a distance given by an unstable wavelength λ should be shorter than the instability time scale. The instability at play here has a capillary origin, and was understood following Laplace's initial work by Plateau and Rayleigh (see e.g. Pomeau & Villermaux 2006; Eggers & Villermaux 2008). The most amplified wavelength is proportional to the jet radius R, giving birth to the periodic blobs along the jet visible in figure 1. Liquid viscoelasticity does not affect λ much, nor the capillary time it takes for amplification $T = \sqrt{\rho R^3/\gamma}$ if γ is the liquid surface tension and when viscous corrections can be disregarded (see also Sauter & Buggisch 2005). The condition for jet formation is found by balancing R/V with T, i.e. by requiring the Weber number

$$We = \frac{\rho V^2 R}{\gamma} = O(1),$$

as Clasen *et al.* (2009) have found. Although this condition corresponds to the detachment of the drops from the orifice in a normal Newtonian fluid, it does not capture the subtle dynamics associated with the transition in a viscoelastic liquid stream. The breakup time, which is comparable to the instability time scale T in normal fluids, is now much longer. It is severely delayed by elastic resistance, allowing the formation of long thin threads linking the blobs (figure 1). A big terminal drop now hangs, connected to the orifice by a spider's thread, which both feeds it, making the drop heavier, and pulls it back by surface tension. When $We \lesssim 1$, the trade-off between these opposing effects results in hesitant, oscillatory dynamics, which can be quantitatively described according to a phenomenology by Clanet & Lasheras (1999). When the terminal drop is heavy enough to overcome all the restoring forces, it detaches, and a new cycle begins. This is the 'gobbling' phenomenon.

Clasen *et al.* (2009) have emphasized that this effect is specific to polymer solutions and is never observed in their experiments with jets of pure water. But the gobbling phenomenon also occurs (albeit for different reasons) with isolated ligaments of Newtonian fluids, as seen in figure 2. A liquid ligament is formed from the fast
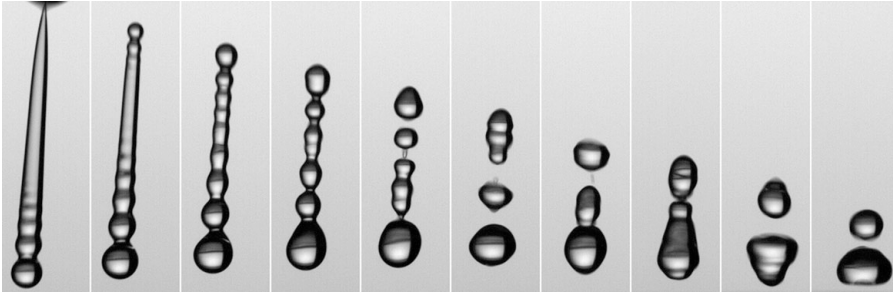
FIGURE 2. A liquid ligament (initial length 2 cm) is produced by the rapid removal of a rod from a water pool. As soon as the liquid ligament disconnects from the liquid bath and the rod, the stretching stops. Then begins a sequence of longitudinal rearrangements and breakups. This interplay results in a coarsening of the globules constituting the ligament, which ultimately sets the final drop size repartition. The time interval between the images is 6 ms.

extension of a bridge connecting a solid rod and a liquid pool. At some point, the ligament detaches, with an aspect ratio depending on both the extension strength and initial bridge radius. Since the ligament cross-section is not uniform, capillary forces induce longitudinal rearrangements, causing regions of the ligament with a small cross-section to empty into larger ones because the Laplace pressure is weaker there. However, these rearrangements occur on a substrate which is, at least locally, cylindrical. As already noted, this geometry favours instability and the formation of globules, or pre-drops visible in figure 2, separated by a distance of the order of the initial ligament average diameter. This is the first stage of drop formation, which would continue up to drop breakup if the ligament were perfectly smooth and infinitely long. But it is not, and this instability develops concomitantly with the longitudinal rearrangements. Nature hesitates between filling large liquid blobs at the expense of adjacent smaller blobs and separating the blobs, resulting in a coarsening of the globule assembly constituting the ligament, which ultimately sets the final drop size repartition. This is how sprays form and their size distribution reflects this mechanism (Villermaux 2007).

Hesitation between breakup and coalescence had been noticed earlier for a drop deposited at the surface of a pool, in which it ultimately coalesces (Charles & Mason 1960). For a given drop radius, the time it takes to complete the expulsion of its own volume into the pool is very similar to the capillary time T it takes to break the neck connecting the drop to the pool. The coincidence of these two time scales causes a very nice 'coalescence cascade'. The drop 'hiccups', emptying partially and then separating before starting a new cycle, shrinking step by step into the pool, the duration of each step being related to the capillary time based on the current drop radius (Thoroddsen & Takehara 2000; Blanchette & Bigioni 2009).

Conversely, the same sequence of iterated instabilities occurs for drop fission. As the drops separate, and if the liquid is very viscous, the thread linking them elongates. At some point, it is locally a cylinder which in turn destabilizes by a capillary instability forming a drop, sometimes called a 'satellite' (Brenner, Shi & Nagel 1994). This sequential hesitation between the drop and the cylindrical shape also occurs in the presence of viscoelastic effects, leading to a 'beads on a string' fractal-like structure of up to four generations (see Oliveira & McKinley 2005; Yildirim & Basaran 2006 and the final threads in figure 15 of Clasen *et al.* 2009).

3. Future

The use of polymers enables the making of very thin yet resistant strands, like synthetic textiles, fibres or spider webs. However, fluid drops produced by the 'gobbling' process in a dilute polymer solution are bigger than they are in Newtonian fluids. Common sense suggests that two disjointed drops will merge provided they are brought sufficiently close to each other. This is only partially true, as detailed observation demonstrates that coalescence occurs when nearby drops are moving *apart* (Bremond, Thiam & Bibette 2008). Squeezing the tiny interstitial lubrication film between the drops down to a width at which thermal noise will break it takes forever. Pulling them apart causes the interstitial pressure to decrease. Since drops are deformable, they respond to this altered pressure field by forming concentrated 'nipples', merging at a point, nucleating a bridge between the drops which soon coalesce. Moving the drops away has caused them to merge, a more than hesitant behaviour! An important question for the future is whether polymers diluted in the drops and/or in the interstitial fluid reinforce or cancel this phenomenon. A key result of the paper by Clasen *et al.* (2009) is that a small but finite unbalanced capillary force acting along the finite-length jet is important in determining the collective dynamics, particularly the 'hesitation' of the pendant drop. As noted by Clasen *et al.* (2009), the standard Rayleigh approach to capillary breakup of an infinitely long jet must be treated with great caution when applied to viscoelastic jets.

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