The behavior of fluids at the microscale

Bubble Clocks

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Science has made significant progress in handling fluids at the microscale. Perhaps the most fascinating potential lies inherent in precisely-controlled methods for generating bubbles and droplets in miniaturized chemical laboratories

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Washing our hands, swimming, or pouring a cup of tea - these are the sorts of experiences that contribute to our intuitions about fluid flows. Yet do these intuitions apply generally? In fact, it turns out that they do have their limits. Over the past two decades, scientists have turned their attention to the flow of fluids (i.e. liquids or gasses) in miniaturized ducts. Using technology similar to that employed in producing electronic circuits, it is now possible to "print" miniature networks of channels with cross-sections typically on the order of tens of micrometers across - the width of a human hair. The flow of liquids at this scale is strongly dominated by the effects of viscosity, or "friction" within the fluid - effects of a kind we rarely experience in everyday life, perhaps only when sneaking a spoon of honey from the pantry. When viscosity is dominant, fluids flow laminarly (i.e. without turbulence): two streams, when put together, flow together side by side and mix only by molecular diffusion. Unlike

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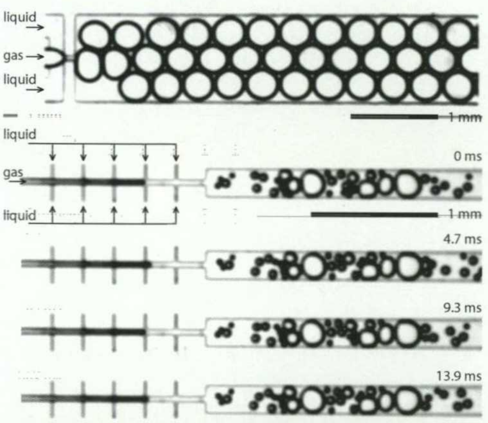
Unlike the disorderly processes of fluid atomization that we know from everyday life, the formation of drops and bubbles in microfluidic systems can be made almost perfect

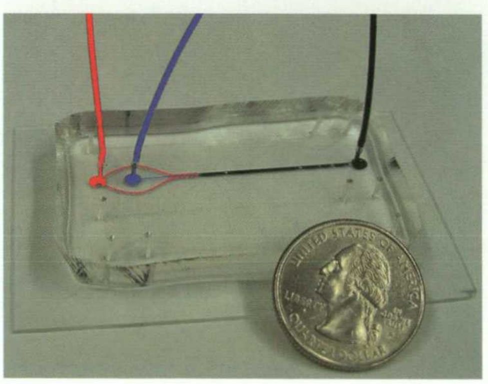
large-scale flows, e.g. those in a mountain stream, microfluidic networks involve no turbulence - all fluctuations in such a flow die out due to the aforementioned "friction," and the flow of liquids and the concentration of chemicals dissolved in them can be precisely controlled in space and time. These features, together with the typically minute consumption of fluids, underpin the intense current interest in microfluidics, as a technology that may one day have the kind of impact on chemical analysis that electronics had for the processing of information. The ultimate goal is to create automated chemical laboratories that would fit on the palm of one's hand.

Laminar flow itself - although practical - is not particularly interesting. Yet a host of new phenomena arise when two immiscible fluids are made to flow through microchannels. The interfacial forces are intrinsically nonlinear and make for fascinating behavior. Microfluidic systems turn out to be ideal venues for the precisely-tuned generation of droplets and bubbles.

The interface between immiscible fluids represents an energy expense. The cost is directly proportional to the area of the interface, and is therefore a function of its



Top panel: a single flow-focusing bubble generator reproducibly forms bubbles of uniform size. Bottom panel: a device comprised of five flow-focusing bubble generators coupled in series. The generators interact and produce complex series of bubbles of different sizes. Surprisingly, this intricate behavior is stable - the system will continue to produce bubbles in this complex manner for minutes and hours on end



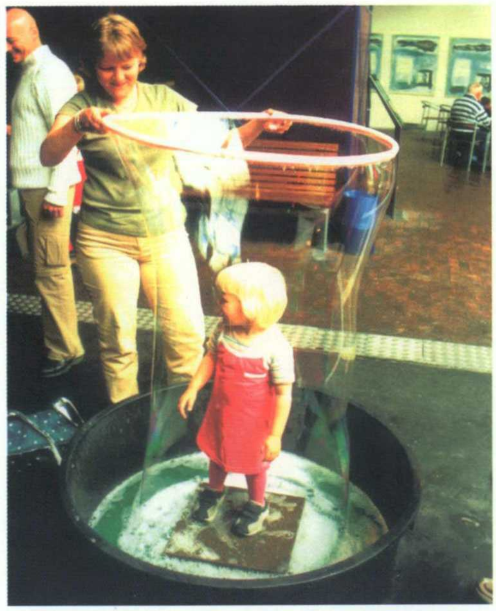
A microfluidic flow-focusing device

shape. For a given volume enclosed by such an interface, the shape that minimizes the energy expense is a sphere; drops, when left alone without any external forces acting on them, thus assume spherical shapes. This "ideal" morphology is not, however, always dynamically accessible. If the volume of an immiscible fluid is deformed too much before it can retreat into a spherical shape, it will break into smaller drops. This instability offers a convenient method of breaking up an immiscible fluid into smaller chunks. All that one needs to do is to deform the volume

Microfluidic systems can generate precisely-tuned droplets and bubbles

of the fluid; interfacial tension does the rest of the job. A dripping faucet provides an example: gravity pulls the liquid down against the interfacial tension that tries to maintain the pending drop in as compact a shape as possible. When this shape is elongated enough it breaks and a droplet is released. One might also employ the inertia of a fast-flowing liquid, as in a fire-hose sending out a jet of liquid into the air; the jet subsequently breaks into a multitude of drops. Another method involves using shear stresses: mayonnaise is typically prepared this way by stirring yolk and lemon juice into oil. The viscous stresses deform volumes of yolk and juice into shapes that break up into tiny drops. The common features of all these methods include a force driving the break-up, arising from interfacial stresses, and a typically wide distribution of sizes among the resulting segments of fluid.

In the field of microfluidics, however, things work differently. One of the typical micro-devices used in the preparation of drops is called a flow-focusing geometry. Three channels meet at a junction followed by an orifice that leads to an outlet channel. The two outer inlets provide the continuous carrier liquid, while the central inlet delivers the fluid to be dispersed. The pressure gradient along the orifice pulls the stream of immiscible fluid towards the outlet channel and an elongated "neck" forms. Because interfacial forces are typically stronger than the shear stresses in microfluidic systems, the tip of the discontinuous fluid stream fills most of the cross-section of the orifice and confines the carrier liquid to thin films. The interface resting on the walls of the device is stable and - in a sense - analogous to a catenoid formed by a soap film span between two rims. The flow of the liquid around the neck, in the thin films, is subject to increased resistance and, as we maintain the inflow of the liquid into the orifice, the pressure upstream of the orifice rises to a value that exceeds the pressure inside the stream of the discontinuous fluid. As a result, the neck is squeezed at a rate exactly proportional to the rate of inflow of the liquid into the orifice. The volume of the drop (or bubble) is simply given by the rate of flow of the inner fluid through the neck, multiplied by the time it takes to collapse or break it. Importantly, both the squeezing of the interface and the flow of the fluid through the collapsing neck are much slower than the equilibration processes of the shape of the interface, and of the pressure fields in the two fluids. Break-up is thus reminiscent of quasi-static processes in thermodynamics; as a result all random fluctuations are averaged out, each break-up process is virtually the same, and the device generates virtually identical bubbles or drops. Microfluidic methods therefore facilitate the preparation of custom-tailored suspensions of droplets and bubbles, with independent control over their size and volume fraction.



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The walls of a microfluidic device provide stability to the interface, much like two round rims can provide stability to a catenoid-shaped soap film spun between them

A microfluidic clock

We have used this well-characterized process to construct a "microfluidic clock" - an unusual time-keeper that can tick with almost any chosen periodicity. The device consists of a serial array of flow-focusing bubble generators all fed from a common supply of carrier fluid. If a single generator can be viewed as an oscillator, the multi-orifice device is an array of *coupled* oscillators. The stream of gas penetrating the orifices affects the pressure in them; this pressure is transmitted to all the other orifices by the network of channels, and, in turn, it affects the rate of inflow of the liquid - and consequently the rate of collapse of the immiscible thread - into each of the orifices in the array. We have observed that over a finite range of liquid flow rates and pressure applied to the stream of gas, this device formed long sequences of bubbles of different sizes, and these sequences repeated over and over, almost ideally. Surprisingly, in spite of the intricacy of these limit cycles, small deviations in the repetitive behavior (in terms of the time of emission of consecutive bubbles or their sizes) did not become amplified, as they usually do in nonlinear dynamic systems - rather, they decayed, leading the system back towards "ideal" behavior. Stabile oscillations of comparable complexity have never been observed in mechanical systems before. We believe that the separation of time-scales between the slow evolution of the interface, and the fast exchange of information between the orifices and relaxation rates, could serve as a guide for the design of synthetic systems exhibiting stable, complex dynamics without requiring active external control.

Such "made-to-order" droplets indeed already have one use: they can serve as chemical beakers. In microfluidic systems it is possible to form such minute, picoliter-sized beakers in large numbers, from hundreds to tens of thousands per second, each one independently prepared, processed and analyzed. What lies in store for us in the future is a way of doing chemistry inside droplets, as reliably as the operation of a Swiss watch. ■

Further reading:

Garstecki P., Gitlin I., Diluzio W., Kumacheva E., Stone H.A., Whitesides G.M. (2004). Formation of monodisperse bubbles in a microfluidic flow-focusing device, *Appl. Phys. Lett.* 85, 26-49.

Garstecki P., Fuerstman M., Whitesides G.M. (2005). Oscillations with uniquely long periods in a microfluidic bubble generator, *Nature Physics*, 1, 168-171.