Electronic Supporting Information

Production of monodisperse drops from viscous fluids

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Influence of the nozzle geometry on the size of drops

The triangular nozzles with rectangular cross-sections form drops whose size is independent of the fluid flow rates if nozzles operate in the dripping regime.¹⁸ Similarly, nozzles containing rectangular reservoirs produce drops whose size is independent of the fluid flow rates¹⁹ and straight nozzles produce drops whose size is only weakly dependent on the fluid flow rates.^{18, 1924} This comparison suggests that the mechanism by which drops are formed is similar. However, these data were acquired using fluids with different viscosities and surface tensions and on devices with different heights such that the results are difficult to compare. To test the influence of the nozzle shape on the drop size, we produce drops in triangular nozzles, and in straight channels that have rectangular cross-section. We employ an aqueous solution with a viscosity of 4 mPa.s as an inner phase; the viscosity is increased through the addition of 10 wt% poly(ethylene glycol) (PEG) 6 kDa. We employ a fluorinated oil, HFE7500, containing a triblock copolymer surfactant as an outer phase;^{41, 42} it has a viscosity of 1 mPa.s.

The rectangular channels are 200 μ m wide and 40 μ m tall such that they are only slightly narrower than the cross-section of the triangular nozzles at the step. The outer oil phase is injected at 12 mL/h and the inner phase is injected at rates that vary between 2 mL/h and 12 mL/h. Within the tested regime, the sizes of drops produced in both devices are independent of the flow rate of the outer phase. Interestingly, drops produced in rectangular channels are 30 % larger than those produced with triangular nozzles, as a comparison of the red and blue circles in Figure S1a reveals. Moreover, if drops are formed at flow rates above 6 mL/h, their size distribution is significantly broader if straight rectangular channels are used, as indicated in Figure S1b. Similarly, the size of drops is independent of the flow rate of the outer phase if produced in triangular and rectangular nozzles, as shown in Figure S1c. However, also in this case, drops produced in rectangular nozzles are significantly larger and display a broader size distribution than those produced in triangular counterparts, as shown in Figure S1d.

To test if the larger size distribution of drops produced in rectangular channel is a result of a too low aspect ratio of the nozzle, we produce drops in 400 μ m wide and 40 μ m tall rectangular channels. The size distribution of drops produced in these wide channels is significantly lower than that of drops produced in 200 μ m wide, straight channels, as shown in Figure S1d, in good agreement with previously reported results.⁴³ However, drops produced in these wide channels are approximately 50 % larger than drops produced in triangular nozzles with identical heights, as shown in Figures S1a and S1c.



Figure S1: Influence of the nozzle geometry on the size and size distribution of drops produced from an aqueous solution with a viscosity of 4 mPa.s. The viscosity of the outer phase is 1 mPa.s. The influence of (a, b) the inner and (c, d) outer flow rate on (a, c) the diameter of the drop and (b, d) their coefficient of variation, CV. Drops are generated in triangular nozzles (\bullet) and straight channels that are 200 µm wide (\bullet) and 400 µm wide (\bigcirc). The nozzle height is kept constant at 40 µm. (a, b) The flow rate of the outer phase is kept constant at 12 mL/h and that of the inner phase is kept constant at 2 mL/h.

Modification of the nozzle

Triangular nozzles produce drops that display a narrow size distribution if they operate in the dripping regime, as indicated in Figures S2a and S2b. By contrast, if they operate in the jetting regime, drops display a very broad size distribution, as shown in Figure S2c. To facilitate the backflow of the outer phase into the triangular nozzle, we introduce two triangular insets, as shown in Figure S2d. However, these insets tend to guide the flow of the inner phase towards one of the edges of the nozzle, thereby preventing its proper operation. To facilitate the back-flow of the outer phase without perturbing the flow of the inner phase, we introduce shunt channels that intersect the triangular nozzle 175 μ m apart from the step, as shown in Figure S2e. This simple modification to the nozzle design increases the maximum throughput of the nozzle by up to 100%.



Figure S2. Nozzle geometry. (a) Schematic illustration of a triangular nozzle in operation with the top (left) and side view (right). (b, c) Optical micrographs of a triangular nozzle that is operated in the (b) dripping regime (left) where drops with a narrow size distribution are produced (right) and (c) jetting regime (left) where polydisperse drops are produced (right). (d, e) Schematic illustration of modified triangular nozzles in operation. To facilitate the backflow of the outer phase, the triangular nozzle is modified with (d) two triangular insets and (e) two shunt channels. The modifications are indicated with two arrows.

Movie S1: Formation of aqueous drops in perfluorinated oil using triangular nozzles with 40 μ m wide two dimensional shunt channels. To visualize the flow of the continuous phase during the drop formation, SiO₂-particles that have been modified with perfluorinated silanes are added to the continuous phase. The movie is slowed down 10 times.