Lab on a Chip

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Electronic Supplementary Information

Microfluidic step-emulsifier that stabilizes the volume of droplets against variation in the rate of in-flow of liquid.

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Device fabrication and characterization

Polycarbonate devices: Microfluidic chips comprise quadratic cross section straight inlet channel and outlet chamber of much larger width than that of the inlet channel. Chips with inlet channel of w_c=200 µm were fabricated in polycarbonate (Macroclear, Bayer, Germany) using a CNC milling machine (MSG4025, Ergwind, Poland). The milled pieces of polycarbonate were cleaned with distilled water, and put in sonic bath with isopropanol (30 min, 40°C). After drying with compressed air polycarbonate chip were aligned and thermally bonded by compressing them together (30 min, 130°C).

Glass-PDMS devices: Microfluidic devices with inlet channel of width w_a=62.7µm were fabricated by standard soft lithography method. The mask was designed in AutoCAD (Autodesk, USA) and then printed on film. SU-8 2025 and 2150 (Microchem, USA) layers were spin-coated on a silicon wafer, baked, exposed to UV light through the mask and developed. Liquid PDMS with crosslinking agent (10:1, Sylgard, USA) was poured on the SU-8 Silicon master and baked in 75°C for 2 hours. PDMS was peeled off the master and after oxygen plasma treatment it was bonded to clean piece of glass.

Surface modification: For all devices hydrophobic modification with Aculon E (Aculon, USA) was performed. Channels were filled with the reagent and left to evaporate overnight, three times to ensure total coverage of the area by the modifier**.** The quality of the coverage was tested during experiments by visual inspection – no wetting of the aqueous droplets was observed.

Imaging: 3D scans of the devices were taken using the optical profilometer Brücker ContourGT-K (Bruker, USA). Images were processed using native client for the microscope as well as free open source program for data visualisation (Gwyddion, http://gwyddion.net/). Fig. S1 shows scans of the master (SU-8 on silicon wafer).

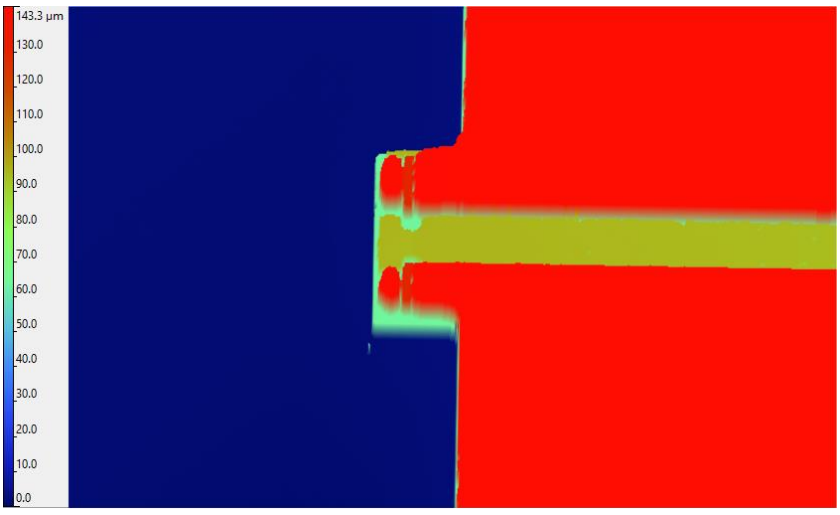

Fig. S1. 3D image of the SU-8 master on silicon wafer for fabrication of PDMS device with constriction and bypasses.

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Dependence of the droplet sizes on the volumetric flow rate

Dependence of the droplet sizes on constriction location: In order to determine the optimal location for the constriction we prepared five microfluidic devices with different locations of the constriction: (a) Δz=0 μm, (b) Δz =50 μm, (c) Δz =100 μm, (d) Δz =200 μm and(e) Δz =300 μm from the outlet chamber, Fig. S2. For further investigations we used system (c) with Δz =100 μm, because it renders smallest droplet volumes in the investigated range of volumetric flow rates. As continuous phase we used fluorinated FC40 oil with viscosity μ_1=4.1 cP and density ρ_1=1855 kg/m^3.

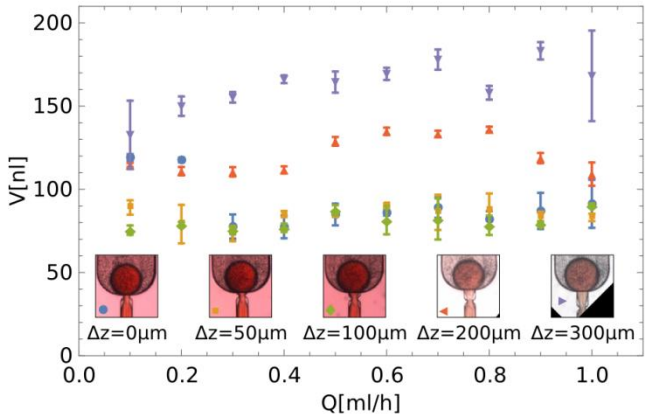

Fig. S 2 Dependence of the droplet sizes on the volumetric flow rate for different positions of constrictions in inlet channels.

Characteristic flow rate: The systems under investigation are positioned in direction of gravitational field, and formed droplets flow upwards due to buoyancy. Terminal velocity of droplet flowing in unlimited system filled with continuous phase equals[1]

$$v_0 = \frac{2}{3}\frac{\rho_o - \rho_d}{\mu_o}\frac{\mu_o + \mu_d}{2\mu_o + 3\mu_d} R_0^2 \, g \,,$$

where R_0 is radius of the droplet, and g=9.81 m/s^2 . As droplet phase we use water: ρ_d=998 kg/m^3, μ_d=1 cP, and as continuous phase oil HFE7500: ρ_o=1614 kg/m^3, μ_o=1.24 cP [2]. For small flow rates the step-emulsification process is capillary driven and for square in cross-section inlet channel of width w formed droplets have radius R_0=w.

We define characteristic flow rate for this process as

$$Q_0 = v_0 w^2 \,.$$

This characteristic flow rate is maximal flow rate for large enough outlet chamber which can be applied in the process. For higher flow rates the droplets are not able to flow from the step before the next droplet comes, so they collide and coalesce. In real system with finite height and width of outlet chamber the maximal flow rate is smaller than Q_0.

In our square in cross-section geometries characteristic flow rates equal for w = 200 *μm*, Q_w = 7.6 *ml/h*, and in PDMS system for a = 62.7 *μm*, Q_a = 0.074 *ml/h*.

Localization of the instability with a constriction

To determine whether the geometrical constriction forces localization of the droplet breakup we observed behavior of the neck in different devices (fig. S3).

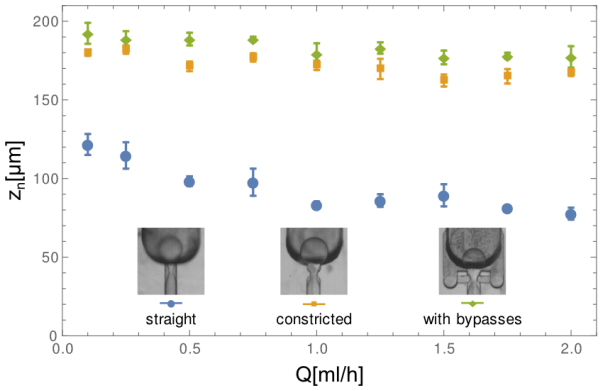

Fig. S 3 Dependence of the position of the neck z_n upstream from the step on the volumetric flow rate Q for three different geometrical designs.

We observed that the breaking point in the systems with bypasses and constrictions is steadily located within constrictions (constriction starts 100 μm from the step and ends 200 μm upstream). In a straight channel, however, the neck visibly changes position. This suggests that indeed the constriction introduced into the channel localizes the instability and point at which the droplet breaks-off.

Viscosity of aqueous solutions

Glycerin (POCH, Poland) was mixed with miliQ water, the viscosity was calculated according to literature[3] for 20°C - the temperature in

which the experiment was conducted. Coefficient α and Q0 values were calculated from data shown in Fig. 3.

Table TS 1 Properties of tested glycerin solutions[3] and calculated values characterizing the system.

Glycerin [w/w]	*ρ[kg/m^3]*	*μ[mPas]*	*Q_0[ml/h]*	*V_0[nl]*	*α [-]*
0%	998.0	1.00	7.63	30.2	1.01
10%	1024.7	1.30	7.11	22.0	4.43
20%	1051.3	1.74	6.60	26.3	2.65
25%	1064.6	2.01	6.36	28.5	1.35
30%	1077.9	2.46	6.10	28.8	1.08
40%	1104.1	3.69	5.62	26.8	1.08
50%	1131.1	6.02	5.17	23.5	1.69
60%	1157.7	9.46	4.80	20.1	2.78

References

1 S. Kim, J. Karrila, *Microhydrodynamics: Principles and Selected Applications*, Butterworth-Heineman, Stoneham 1991

2 3M™ Novec™ 7500 Engineered Fluid, manufacturer's brochure (http://multimedia.3m.com/mws/media/65496O/3mtm-novectm-7500-engineered-fluid.pdf)

3 N. S. Cheng, *Industrial & Engineering Chemistry Research*, 2008, **47**, 3285-3288; on-line calculator can be found here: http://www.met.reading.ac.uk/~sws04cdw/viscosity_calc.html