Supplementary Material (ESI) for Lab on a Chip

Effects of unsteadiness of the rates of flow on the dynamics of formation of droplets in microfluidic systems

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Supplementary information

1. Measurements of oscillations of rate of flow with the use of an analytical balance.

We measured (with an analytical balance Mettler Toledo) time series of mass of liquid flowing out from PHD2000 Harvard Apparatus. Differentiating this series with respect to time yields the rate of flow as a function of time. Fourier transform of this signal yields the period of oscillations. In the table below we present periods measured for a set of syringes and variety of rates of flow and compare them to periods in time series for drop's sizes produced in the T junction fed from the same model of syringe pump.

Syringe diameter [mm]	Rate of flow [mL/h]	Material	Period of oscillations for rate of flow [s]	Period of oscillations for drops' sizes [s]	Predicted period of oscillations [s]
4.73	4	polypropylene / polyethylene	-	16.6	16.0
12.1	2	polypropylene / polyethylene	203.5	-	209.0
12.1	4	polypropylene / polyethylene	104.9	107.6	104.5
12.1	8	polypropylene / polyethylene	53.1	-	52.3
15.6	4	polypropylene / polyethylene	169.4	163	173.73
15.6	8	polypropylene / polyethylene	84.5	84.3	86.9
26.95	8	all glass	271.0	271	259.2
26.95	16	all glass	128.6	-	129.6
26.95	32	all glass	61.9	-	64.8

Table 1: Measurements of rate of flow and droplet's size oscillations for a system supplied by PHD2000 Harvard Apparatus syringe pump. Predicted periods of oscillations were evaluated according to equation: $T = a \pi D^2 / (4 Q)$ where D is the diameter of the piston, Q – the rate of flow and α is the pitch of the thread on the lead screw of the pump.

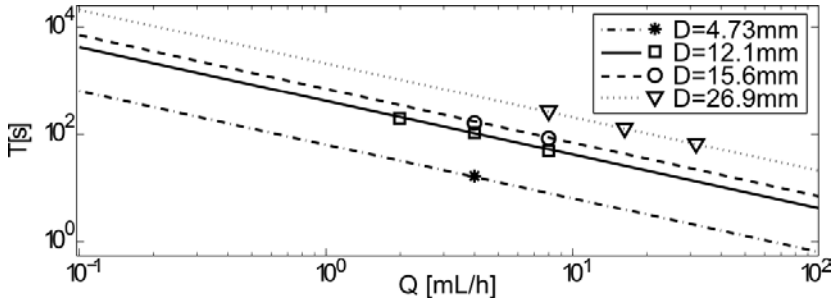

Fig. 1: Period of oscillations of the rate of outflow from a PHD2000 syringe pump. Symbols show measured values, lines are given by equation $T = a\,\pi\,D^2 / (4\,Q)$ *where D is the diameter of the piston, Q – the rate of flow and* α *is the pitch of the thread on the lead screw of the pump. The pitch of the thread on the lead screw for syringe pump PHD2000 is equal 1.058 mm. Coefficient* a *evaluated from experimental data is equal* 1.01 ± 0.04 *mm.*

2. Velocity of tracer droplets

We measured velocity of tracer droplets carried by the fluid flowing out from the syringe pump. Single drops were added to the flow inside microchannel with an external droplet-on-demand system. We observed the motion of drops with a digital camera coupled with magnifying lenses. The velocity was evaluated from positions of a droplet on consecutive frames of the video, during the passage of the droplet through a given, straight segment of the microchannel. As we can see on the Fig. 2, the velocity of the droplet fluctuates periodically for pump feeding and retains almost constant when the continuous liquid was supplied from the pressurised containers.

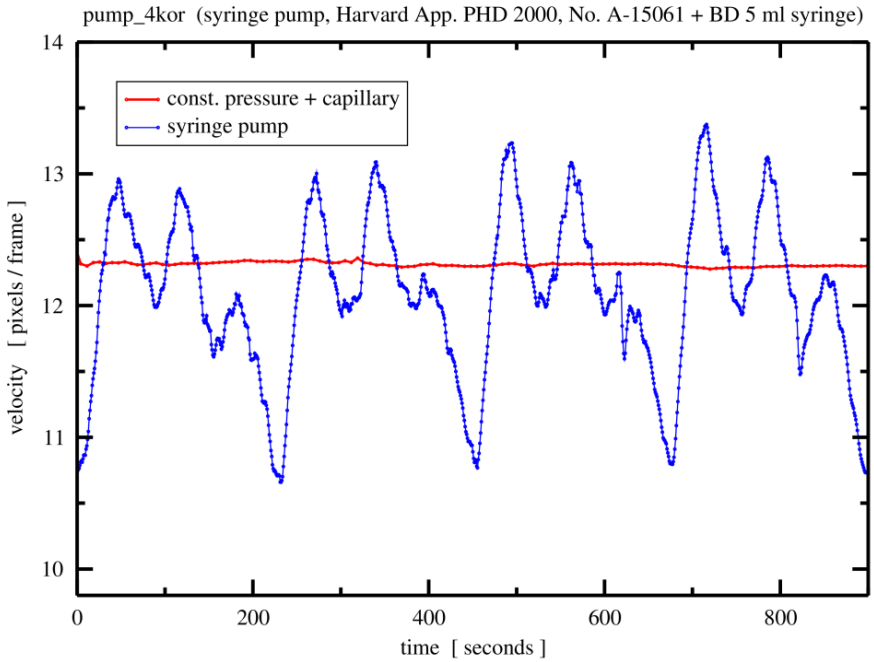

Fig. 2: Time series of velocity of tracer drops flowing through microchannel supplied by use of pressurised capillaries (red) and syringe pump (blue).

3. Calibration of capillaries

Using the resistive capillaries for delivering stable rates of flow requires calibration of the hydraulic resistance of the capillary. First estimate the Reynolds number for the flow on the basis of the dimensions of the capillary and the range of required rates of flow. If the flow is laminar, the relation between the pressure P applied to the container with liquid and the rate of flow Q through the capillary is linear: P=RQ where R is the hydrodynamic resistance.

In order to calibrate for R place the capillary in the bath and apply a known pressure to the container. Then evaluate the rate of flow by measuring the rate of change of mass on an analytical balance collecting the liquid flowing out from the capillary. The ratio of P/Q yields R. Alternatively, for more precise calibration collect a number of values of Q for different values of P and fit a line: P = RQ to retrieve R (Fig. 3).

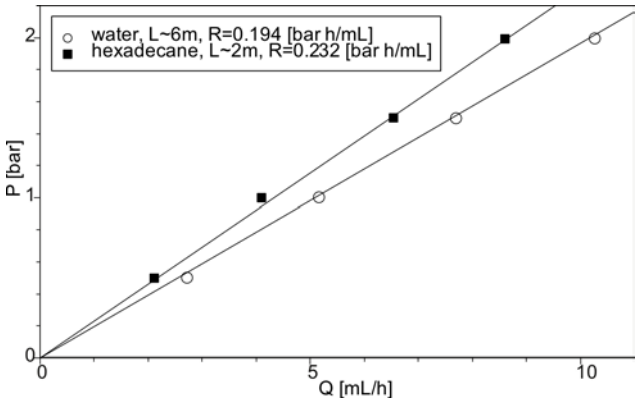

Fig. 3: Calibration of capillaries: L ~ 2m for hexadecane and L ~ 6m for water.

The resistance of the capillary changes with viscosity, that in turn depends on temperature. Thus, a given calibration is valid only for the temperature at which it was conducted. It is important to avoid changes of temperature during the experiment. We propose to place capillary into a container filled with water – this stabilized the temperature to the ambient. We made calibration before and after each series of measurements in order to verify that the resistance of capillary was stable during the experiment.

4. Details of the capillary feeding system

Fig. 4 illustrates schematically the capillary system for feeding constant rate of flow into microfluidic chips. We used compressed air (8 bar) applied via pressure regulators (MNR0821302447, Rexroth, Germany) to a custom made reservoir for fluids. The container interfaced with the chip via a steel capillary of inner diameter of 0.21 mm (Mifam, Poland) and of length varied from 1 m to 8 m. For connections between the steel capillaries, and between the capillaries and needles (needle of outer diameter of 0.8 mm,), we used elastic Tygon tubing (inner diameter 0.25 mm, outer 2 mm, Ismatec, Switzerland). In order to avoid instability of the connections we always put the end of the capillary into the needle, so that the walls of the elastic tube could not block the lumen of the capillary.

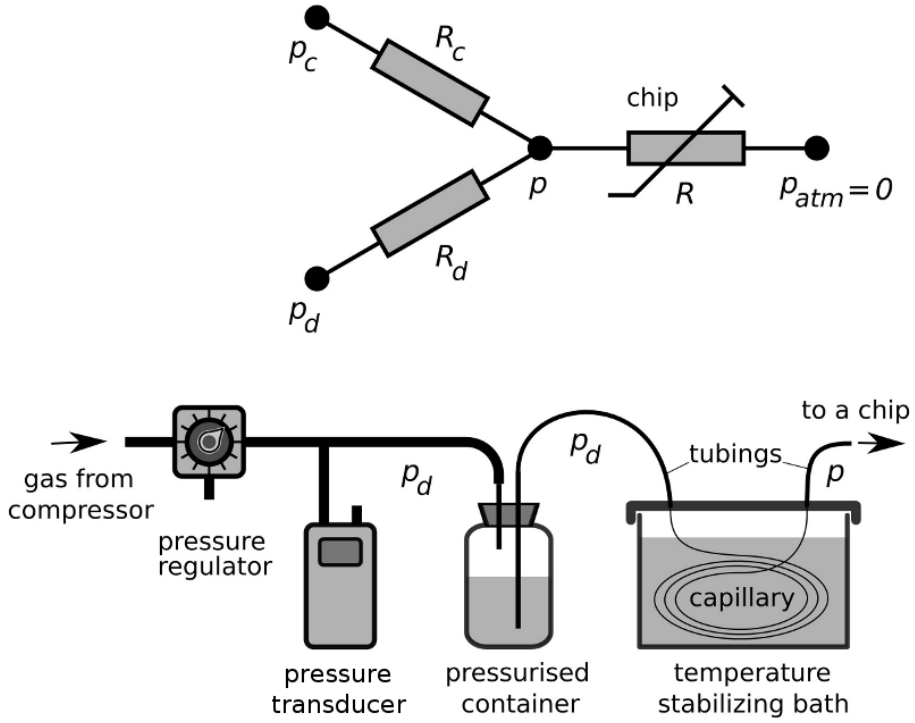

Fig. 4: Top: The equivalent electric circuit of the system consisting of two pressurized containers (continuous phase under pressure p_c, droplet phase under p_d) from which the liquids are delivered by the capillaries of hydrodynamic resistance R_c, R_d into the T-junction (pressure p) then form droplets and flow together through a chip of resistance R (varying, due to the generation and motion of droplets) up to the outlet (pressure 0, since all pressures are relative to the atmospheric pressure). Bottom: detailed view of the feeding line for the droplet phase. The second line is built analogously.

We calibrated the resistance of the capillaries by characterizing the rate of flow of the fluids as a function of the applied pressure for a fixed (ambient) temperature. In general, having capillaries of known resistance (e.g. already calibrated for a desired liquid), one may easily calculate flow rates of continuous phase (Q_c) and droplet phase (Q_d) from Ohm and Kirchhoff circuit laws:

$$Q_c = \frac{p_c}{R_c} \frac{1 + x_d\,(1 - p_d/p_c)}{1 + x_c + x_d} \qquad (1)$$

$$Q_d = \frac{p_d}{R_d} \frac{1 + x_c\,(1 - p_c/p_d)}{1 + x_d + x_c} \qquad (2)$$

$$Q = \frac{p_c\,x_c + p_d\,x_d}{R\ (1 + x_c + x_d)} \qquad (3)$$

where $x_c = R / R_c$ and $x_d = R / R_d$.

It should be noted that although Q_c and Q_d are not mutually independent functions of respective pressures p_c and p_d, the cross-dependency becomes irrelevant for small values of x_c and x_d i.e. when the resistance of the capillaries is much larger than the resistance of the chip. Keeping $R_c >> R$ and $R_d >> R$ also minimized the variation of Q_c and Q_d in response to the variation of R. It is practical to to have p_c and p_d of similar magnitude, hence for a given range of rates of flow the user should tailor resistance of capillaries to meet the following condition: $R_c / R_d \approx Q_d / Q_c$.

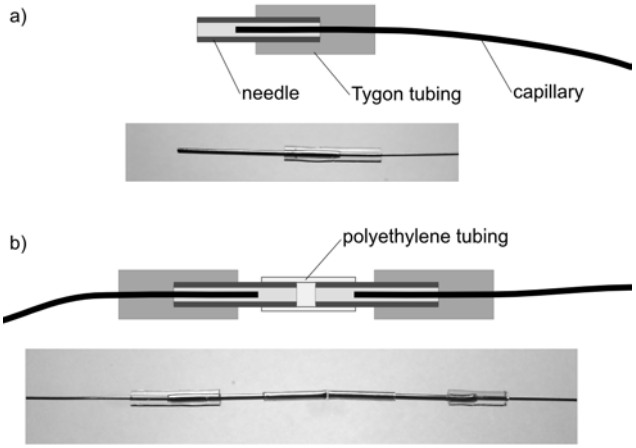

Fig. 5: The end of capillary (a) and connection between capillaries via polyethylene tubing PE 60 Becton Dickinson (b).