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Dry microfoams: Formation and flow in a confined channel

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We present an experimental investigation of the assembly of microbubbles into a 2D foam and its flow in a rectangular microchannel. Using a flow-focusing method, we produce a foam *in situ* on a microfluidic chip for a large range of liquid fractions, down to a few percent in liquid. The channel height is 250 μm , resulting in bubbles whose height to diameter aspect ratio ranges between 0.3 and 1. We can monitor the transition from separated bubbles to the desired microfoam, in which bubbles are closely packed and separated by thin films. We find that bubble formation frequency is limited by the liquid flow rate, and that it creates a modulation of the foam flow, rapidly damped along the channel. The average foam flow rate depends non-linearly on the applied pressure, displaying a threshold pressure due to capillarity. Strong discontinuities in the flow rate appear when the number of bubbles in the channel width changes, reflecting the discrete nature of the foam topology. For a given foam structure the pressure drop scales with the flowrate to the power $2/3$. We compare this dependance with the case of macroscopic foams pushed without a coflowing liquid. We also produce an ultraflat foam (channel height about 8 μm) reducing the bubble aspect ratio to 0.02; we notice a marked change in bubble shape during the flow.

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Microbubbles and microdrops presently attract considerable attention [1, 2, 3, 4, 5]. Their application in a lab-on-a-chip context provides the possibility for individual handling of gas pockets, and allows to create microchemical reactors that are both very rapid and highly parallelized. Specifically, the gas-liquid interface of microbubbles provides a transport location for amphiphilic molecules, with a hydrophilic head and a hydrophobic tail. A decrease in size increases the surface to volume ratio; hence, microbubbles could be used as an efficient carrier for proteins or lipids at high concentration.

Microbubbles can be produced using a microfluidic flow-focusing device: a flowing gas thread is forced, by the co-flowing surrounding liquid, into a small orifice, where the gas thread breaks up at regular time intervals [3, 4, 6]. A set of basic operations necessary for individual bubble manipulation, termed “discrete microfluidics”, has been demonstrated at the millimeter scale [5] in a “dry” foam, where the liquid content is low compared to the gas content. To down-scale these operations and adapt them for microfluidics requires producing a *microfoam*. One of the interesting properties of foams is that they can exist over a large range of sizes [7]. We know the size-dependency of their main physical properties, including structure (governed by their surface tension) [7] and dissipation (dominated, in what follows, by friction on the channel walls) [8]. We would like to determine how these foam properties extrapolate to the micrometer range.

Here, we investigate the continuous production of bub-

bles in a flow-focusing device, and the transitions between different regimes of bubble formation, so as to reach microfoams, thereby extending the studies of Ref. [3] to low liquid fractions. The absence of vertical drainage on the small length scales of a microfluidic system creates constant and stable liquid profiles in the foam films. At the low Reynolds numbers involved, the amounts of gas (bubble volume distribution) and of liquid (liquid fraction) can be reproducibly controlled. We measure the pressure drop and non-linear dissipation along the microfluidic set-up: an orifice, followed by a long channel ending with a free exit. We finally open perspectives for the study of structure and dissipation of ultra-flat microfoams.

We produce the microfluidic device by soft lithography. We first create a mold in a negative photosensitive material (SU-8 2100, MicroChem), with a channel height $h = 250 \mu\text{m}$, width $w = 700 \mu\text{m}$, orifice width $w_{or} = 100 \mu\text{m}$. We then imprint this pattern in a polymer (polydimethyl-siloxane, PDMS) to create the actual channel, and glue it to a glass cover slide using a home-built ozone-cleaner. We also use a sample where, at a distance of 6 mm after the entrance orifice, the foam has to flow through a second constriction, of width $w = 125 \mu\text{m}$. Finally, we will present the use of a positive photo-resist (ma-P 100, Micro Resist Technology) to produce an ultra-flat channel, height $h = 8 \mu\text{m}$ and width $w = 400 \mu\text{m}$.

The continuous phase is water with 10 % commercial dishwashing detergent (Dreft, Procter & Gamble); its surface tension is $\sigma = 38 \pm 1 \text{ mN/m}$, measured by the Wilhelmy balance method. The use of this surfactant resulted in an increased wettability of the solution to the PDMS surfaces [9]. Two different syringe pumps were used to push the liquid (11 Pico Plus, Harvard Apparatus, and KDS 100, KD Scientific) at flowrates Q_l ranging

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from 4 to 167 $\mu\text{l}/\text{min}$, constant with $\pm 0.5\%$ accuracy. The dispersed phase is nitrogen. We drive it either at constant flow rate Q_g , using a similar Pico Plus syringe pump; or at constant pressure P_g , ranging from 1 to 21 kPa, using a pressurized tank and a pressure-reduction valve (precision ± 0.15 kPa).

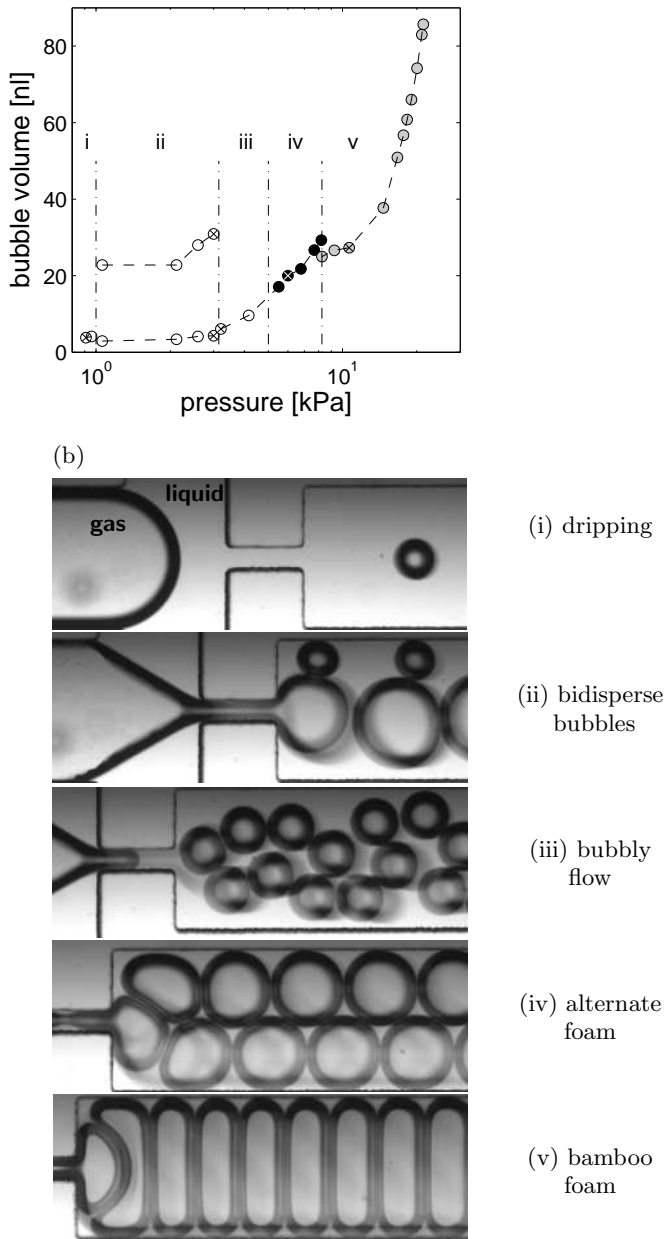


FIG. 1: From wet to dry microfoams. (a) Bubble volume V_b versus gas pressure P_g . Liquid flow rate is kept constant at $Q_l = 167 \mu\text{l min}^{-1}$. Numbers identify the different regimes: (i) dripping flow; (ii) bidisperse bubbles (two symbols are plotted for each pressure); (iii) bubbly flow; (iv) alternate foam (2 rows) with filled symbols; (v) bamboo foam (1 row) with grey symbols. (b) Photographs of these regimes. Crossed symbols in (a) correspond to pictures in (b).

Fig. (1) shows examples of the observations in the

250 μm high channel, near the orifice at the channel entrance. We capture still images and movies with a camera (Marlin F131B, Allied Vision Technologies) connected to an inverted optical microscope (IX70, Olympus). By analysing these images we extract the bubble formation rate (break-up frequency f), with a precision of a few percent. We estimate the bubble volume V_b by measuring the apparent area A occupied by the gas in the images, multiplied by h . The edge of the measured area (measured with a precision $\pm 1\%$) was taken in the middle of the dark line around the bubble, systematically underestimating (with up to 10% for non-touching bubbles) the actual gas volume. We estimate *simultaneously* the gas flow rate as $Q_g = V_b f$.

The proportion $\alpha_l = Q_l/Q$ of liquid injected in the system is measured with a precision of $\pm 10\%$, where $Q = Q_g + Q_l$ is the total two-phase flow. It can differ from the foam volume liquid fraction Φ_l .

The minimum pressure for which bubble formation was observed at the liquid flow rate is $P_c = 0.9 \pm 0.15$ kPa (Fig. 1a). This is probably due to the capillary pressure. For a curved interface in the orifice at the limit of bubble detachment, the Laplace pressure is $\sigma(1/r_1 + 1/r_2) = 1.1 \pm 0.2$ kPa, where $r_1 = h/2$ and $r_2 = w_{or}/2$ are the principal radii of curvature.

For pressures P_g above P_c , bubbles form. The pressure drop across the orifice creates a gas-liquid coflow. A gas thread is forced into the orifice and fills a bubble after the orifice. This thread pinches and releases the bubble. After break-up, the gas-liquid interface retracts, as reported in [4]. The interface returns completely into the upstream part (dripping flow (i), Fig. 1b). At higher P_g , there is a coexistence, probably indicating a first-order transition, with a second mechanism, where the interface remains in the orifice instead of retracting. For given P_g and Q_l , both mechanisms give different volumes V_1, V_2 . This results in a flow of period $T_1 + T_2$ [3], with bidisperse bubbles (ii, Fig. 1b).

Increasing P_g , we observe microfoams, with only the second pinch-off mechanism, without rapid interface retraction, hence no inertial non-linearities, period doubling or chaotic bubbling as in [6]. This results in a monodisperse foam, with three possible structures according to the flow rate: bubbly flow (iii), alternate foam with two rows (iv), or bamboo foam with one row only (v). The bubble volume correlates with the fluid fraction (except for the lowest Q_l): $V_b/w_{or}^3 \sim \alpha_l^{-0.95 \pm 0.02}$ (Fig. 2). This correlation is similar to the one observed in axisymmetric conditions by [10, 11] who measured $V_b/w_{or}^3 \sim (Q_l/Q_g)^{-1.11 \pm 0.02}$, in the opposite case of high liquid fraction with Q_l/Q_g higher than 5. It differs from the $V_b \sim P_g/Q_l$ scaling observed with flat bubbles in channels by [3]: we will see further that the gas flow rate and applied pressure are not proportionnal within a low fluid fraction microfoam. To increase the number of bubble rows would require to decrease V_b at given α_l , for instance with an orifice aspect ratio w_{or}/h closer to 1 to restrict liquid flow, or a lower ratio of orifice to chan-

nel width w_{or}/w to enlarge the space available for newly formed bubbles.

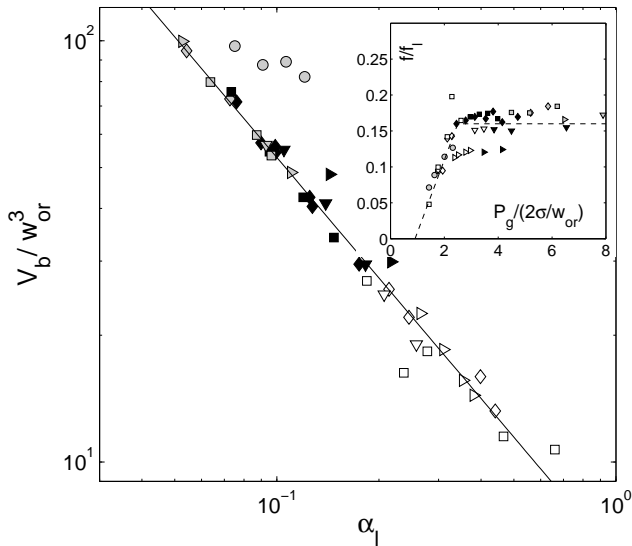


FIG. 2: The bubble volume V_b , in units of $w_{or}^3 = 1$ nl, depends only on the injected liquid fraction α_l : the solid line is a linear fit to all data except the lowest Q_l , $\log(V/w_{or}^3) = (-0.95 \pm 0.02) \log \alpha_l + (1.78 \pm 0.05)$. *Insert*: the bubble formation frequency f in units of $f_l = Q_l/w_{or}^3$ vs. gas pressure P_g in units of $2\sigma/w_{or}$ showing a plateau for high P_g . The dotted line is a piecewise linear fit. Symbols correspond to different values for the liquid flow rate Q_l : (o) 4, (◇) 15, (□) 20, (▷) 30 and (▽) 40 $\mu\text{l min}^{-1}$, for bubbly flows (open symbols), alternate foams (filled symbols), bamboo foams (grey symbols).

The frequency f of bubble formation, determined by a liquid mediated pinch-off time that scales like $1/Q_l$ [4], also depends on the gas pressure. We can express the frequency through the non-dimensional Strouhal number, $St = f/f_l$ where $1/f_l = \tau_l = w_{or}^3/Q_l$ (insert of Fig. 3). All data collapse on a single curve where St is a function of gas pressure only. Two regimes appear: St is first proportional to gas pressure above the threshold $St \simeq 0.1(P_g - P_c)/(2\sigma/w_{or})$ then saturates to a constant value $St \simeq 0.16$. This suggests that the only limiting time at high P_g is τ_l , the time to pinch off the gaseous thread when the liquid flow is blocked by the bubble at the outlet [4]. At low P_g the limiting time varies as $\tau_{fill} \sim [Q_l(P_g - P_c)]^{-1}$. Fast framing sequences confirm that liquid flow is restricted by the gas bubble only after the filling of the orifice. This filling time τ_{fill} decreases with increasing Q_l , consistently with the observation that flow focusing contracts the available space for the gas thread; it also decreases with increasing P_g since the gas pushes the fluid with a velocity increasing with P_g . Note that the transition from a τ_{fill} to a τ_l dominated break-up frequency is accompanied by the regime change from bubbly flow to foam. In the foam state, the liquid flow restriction seems more efficient (see figure 1b, iv and v).

We now turn to the gas pressure drop across the whole

set-up: orifice and channel (Fig. (3)). The threshold in pressure for bubbly flow (found to be 1.0 ± 0.1 kPa, if this parameter is left free in the fit) is compatible with the above explanation by a capillary effect (1.1 ± 0.2 kPa) at the orifice. On the other hand, the slope originates from dissipative effects in the channel: in presence of a second constriction (data not shown), we obtain for regimes (iii) and (iv) the same result as in Fig. (3), translated by about 0.45 kPa along the P -axis (compatible with the expected Laplace pressure necessary to pass the second constriction, 0.6 kPa). We thus write the total pressure drop as the sum of two contributions, $P_g = P_c + \Delta P_{channel}$, where P_c is the static orifice contribution and $\Delta P_{channel}$ is due to dynamic dissipation in the channel.

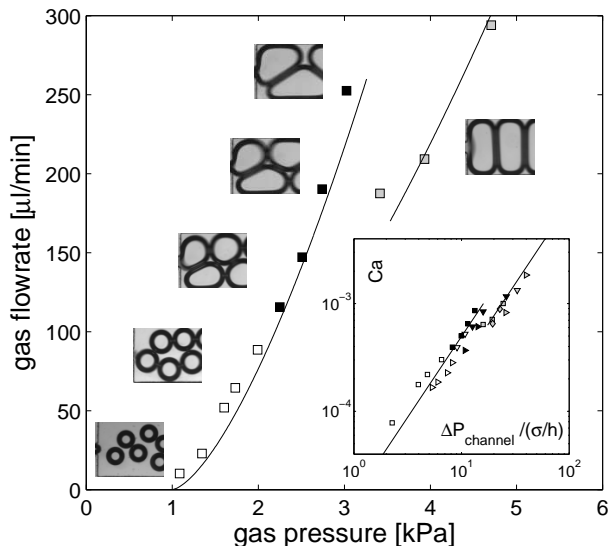


FIG. 3: Gas flow rate Q_g vs. gas pressure P_g showing a capillary threshold and discontinuity at the transition from alternate to bamboo foam ($Q_l = 20 \mu\text{l min}^{-1}$). *Insert*: adimensional axes using $Ca = \mu Q_g/S\sigma$ versus $\Delta P_{channel} = P_g - P_c$ with superimposed data for different liquid flow rates $Q_l =$ (◇) 4, (□) 20, (▷) 30 and (▽) 40 $\mu\text{l min}^{-1}$. The solid lines on both plots are fit on alternate and bamboo foams at $Q_l = 20 \mu\text{l min}^{-1}$ with the power law $\Delta P_{channel}/(\sigma/h) = \beta Ca^{2/3}$, with $\beta = 1.7 \times 10^3$ (alternate) and $\beta = 2.5 \times 10^3$ (bamboo).

The structure transition from an alternate to a bamboo foam induces a discontinuous decrease of the gas flow rate (Fig. 3). This is a signature of the discrete character of the foam. It can be interpreted by assuming that dissipation mainly occurs in the liquid films, close to the walls [8]. Introducing the capillary number $Ca = \mu v/\sigma$ (of order $10^{-4} - 10^{-3}$) containing the bubble velocity v (estimated as $v \simeq Q_g/S$ in the dry foam state) and liquid viscosity μ , the pressure drop writes $\Delta P_{channel} = \bar{\lambda} \frac{n L_{proj}}{S} \sigma Ca^{2/3}$, with n the total number of bubbles, L_{proj} the projection on the cross section of the wetting perimeter per bubble, S the cross section area, and $\bar{\lambda}$ a numerical constant [8]. We find by image analysis that the transition to bamboo structure is associated with an increase in the bubble number n of 15 % (com-

pactification) together with an increase of the projected length L_{proj} of 50 % (this length is measured between vertices centers, for contacts with the walls parallel to the image plane, neglecting side walls), consistent with the observed increase of $\Delta P_{channel}$ by 50 %. The dissipation in the liquid films touching the walls is $\bar{\lambda} = 22 \pm 5$. This value can be compared with the one measured on a millimetric foam in a channel with a comparable aspect ratio: $\bar{\lambda} = 38 \pm 04$ [8]. In our case liquid is injected continuously, possibly explaining the lower value for $\bar{\lambda}$.

The liquid flow, varying by a factor 10, does not influence much the gas pressure drop in our geometry (see insert in fig 3). As we see no change in the thickness of lubricating liquid films between gas and walls, which would change drag forces, we assume that liquid flows mostly in the corners [12], and is not as obstructed by bubbles in the channel as with a cylindrical geometry [13]. The actual liquid content Φ_l can be estimated by the bubble volumes. Another method is to measure simultaneously the time and space averaged bubble velocity $\langle \bar{u}_g \rangle$ together with the gas flowrate; the liquid fraction then follows from $Q_g = (1 - \Phi_l) \langle \bar{u}_g \rangle S$. Both methods provide a liquid fraction $\Phi_l \simeq \alpha_l$: the liquid fraction is controlled by the injection parameters. Similarly, the average liquid velocity can be estimated from $Q_l = \Phi_l \langle \bar{u}_l \rangle S$, which yields $\langle \bar{u}_l \rangle / \langle \bar{u}_g \rangle = \frac{\alpha_l}{(1 - \alpha_l)} \frac{(1 - \Phi_l)}{\Phi_l}$. Here we obtain $\langle \bar{u}_l \rangle / \langle \bar{u}_g \rangle \simeq 1$: there is no substantial relative drainage of the liquid within the foam.

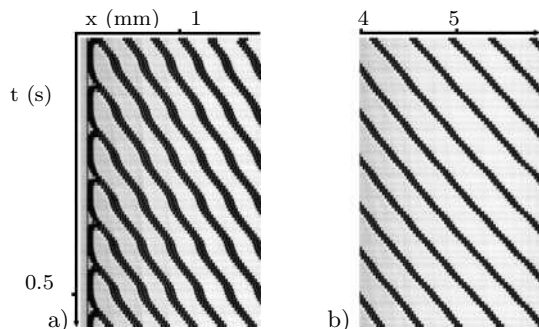


FIG. 4: Space-time diagrams of the foam flow in the constriction sample. The vertical axis is the time, flowing downwards; the horizontal axis is a (small zone of) the axis of the channel, with the foam flowing from left to right, $Q_l = 16.7 \mu\text{l min}^{-1}$ and $P = 4.9 \text{ kPa}$. Dark pixels indicate a bubble edge. (a) The channel entrance. (b) Just before the exit constriction.

The above flow rate measurements are in fact *time-averaged* flows. Due to the periodic creation of bubbles, the pressure in the entrance orifice probably varies at frequency f . Each new bubble has to push the foam to create a place for itself; thus the foam velocity at entrance also oscillates at frequency f (Fig. 4a), between 3.1 and 8.3 cm/s. However, this oscillation is damped over a few mm along the channel (data not shown). Very little oscillation is observed after the entrance (Fig. 4b). Friction, probably mainly at the channels walls, thus homogenizes the flow at the micrometer scale.

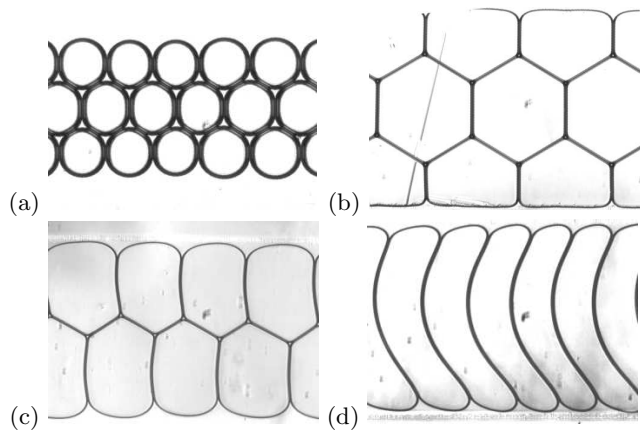


FIG. 5: Ultraflat foams in the $8 \mu\text{m}$ high channel: (a) Flowing, wet and (b) static, dry 3-rows foams. (c) 2-rows and (d) 1-row flowing dry boomerang foams, flowing from left to right.

The ultra-flat channel represents a 30-fold decrease in height, to $8 \mu\text{m}$, and a 18-fold decrease in aspect ratio for the channel section, to 0.02. We can continuously produce various foams in such a channel (Fig. 5), including one with 3 bubble rows (Fig. 5a). We can dry it, using the following batch method. We shut the liquid inlet and pull the syringe at the gas inlet. As long as the underpressure is smaller than the Laplace threshold (here 11 kPa), the bubbles are blocked by the orifice and only liquid flows out of the foam. This forced drainage yields hexagons with a standard deviation in the edge length of only 1.8 % (Fig. 5b). Since the apparent wall thickness on images ($10 \mu\text{m}$) is comparable to the height, the bubble walls are probably very curved, and with *no flat film between bubbles*, contrary to the previous set-up. Thus the actual liquid fraction Φ_l of the central bubble row is probably smaller than, but close to, the apparent one (fraction of black pixels) $\Phi \approx \Phi_l^{image} \simeq 10^{-1}$. This contrasts with usual foams with larger aspect ratio, where the same picture of hexagons with straight walls and small vertices would correspond to much lower fluid fractions, $\Phi_l \simeq 10^{-2} < \Phi_l^{image}$ [7].

Foams flowing in this ultraflat channel undergo an unusual boomerang-like distortion (Fig. 5c,d). We expect a larger friction at the center, where bubble walls along the top and bottom plates are thin, than on the channel sides, where water accumulates. These edges are likely more entrained by the liquid flow than the centers, assuming that for small aspect ratios corner sections are less expandable, providing $\langle \bar{u}_l \rangle / \langle \bar{u}_g \rangle > 1$. Now within the reference frame moving at the liquid velocity, the bubbles move in the opposite direction, from right to left. The distortion of bubble shapes is then similar to the observations of millimetric bubbles pushed without any liquid flow [5].

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