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View online: http://dx.doi.org/10.1063/1.4913202
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Passive removal of immiscible spacers from segmented flows in a microfluidic probe

Xander F. van Kooten, Julien Autebert, and Govind V. Kaigala
IBM Research-Zurich, Säumerstrasse 4, CH-8803 Rüschlikon, Switzerland

(Received 3 December 2014; accepted 4 February 2015; published online 18 February 2015)

Microfluidic probes (MFPs) are a class of non-contact, scanning microfluidic devices that hydrodynamically confine nanoliter volumes of a processing liquid on a surface immersed in another liquid. So far only chemical processes using a single processing liquid have been implemented using MFPs. In this letter, we present the design and implementation of a probe head that allows segmented two-phase flows to be used, which will enable different chemical species to be sequentially delivered to a surface in defined volumes and concentrations. Central to this probe head is a spacer-removal module comprising blocking pillars in the injection channel, a bypass and an orifice leading to the aspiration channel. We present a capillarity-based analytical model that provides insight into the functionality of the module based on geometrical parameters. In addition, we study the difference between two- and three-channel modules and predict a 30% reduction in fluctuation of the footprint of the confined liquid for the three-channel module. We show that such a module with a 15 μm pillar spacing, a 30 μm orifice width, and an oblique angle of 30° can remove immiscible spacers (Fluorinert FC-40) from an aqueous flow at a rate of up to 15 spacers per second while maintaining hydrodynamic confinement of processing liquid. © 2015 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution 3.0 Unported License. [http://dx.doi.org/10.1063/1.4913202]
pinning of its interface at the pillars causes it to be directed into the bypass, where it is then trapped due to a capillary force induced by the pinning of its front interface at the orifice.

When a subsequent incoming spacer reaches the blocking pillars, it merges with the spacer in the bypass and blocks the flow in the injection channel. The pressure increase resulting from this blockage causes a volume of IP to be ejected through the orifice into the removal channel (left in Fig. 2(a)) where it is sheared. As the module typically operates with a capillary number Ca ≈ 1, the size of the ejected spacers can be modified by designing a constriction in the removal channel. After the ejection of a fraction of the IP, spacers can be modified by designing a constriction in the removal channel. The pressure in the injection channel must push the front interface pinned at these two points (I and II, respectively, below) before the interface breaks, we determine how the module behaves of the side interface is determined by the pillar spacing da and the oblique angle β. (b) For pIP − pwa ≪ Pmax, no pinning occurs at the side interface. (c) As pIP − pwa approaches Pmax, pinning occurs at pillar B. (d) For Pmax < pIP − pwa < Pmax, pinning occurs at pillar A. (e) Schematic close-up view of the interface (blue line) pinned at pillar B, having minimum curvature radius Rf, (f) Interface pinned at pillar A, with minimum curvature radius Rf.

\[
\Delta P_{f,max} = \gamma \left( \frac{1}{\max \{R_f,2 \} + 1/R_f}\right),
\]

where γ is the interfacial tension of the oil-in-water emulsion, and Rf is the minimum curvature radius of this interface in the x,y plane, given by \( R_{f,i} = \frac{-d_i}{(2\cos(\theta - \psi_i))} \) (i = 1, 2). Through similar geometrical reasoning, for the curvature radius in z-direction, we use \( R_z = -2\cos(\theta)/h \), where h is the channel height and θ is the contact angle. In the broadly applicable case where \( \psi_1 \neq \psi_2 \), both \( R_{f,1} \) and \( R_{f,2} \) must be evaluated as the larger of these two yields a lower maximum pressure drop \( \Delta P_{f,max} \) and therefore determines the pressure drop at which the front interface breaks.

High pressure drop—at the blocking pillars: The side interface of a trapped spacer is in contact with the blocking pillars at all times. The development of a section of the side interface between pillars A and B consists of three stages.

(i) No pinning occurs when the pressure drop across the side interface is small compared with the maximum pressure drop that it can withstand before breaking (Fig. 2(b)).

(ii) As the pressure drop across the interface increases, the interface expands until the contact line at pillar B pins at point S (Figs. 2(c) and 2(d)). As \( \theta \leq \pi \), the
interface is not affected by the straight left edge of pillar B below point S, and the interface will assume a symmetrical profile between S and T (Fig. 2(d)). We obtain an expression for the minimum curvature radius in the \(x,y\) plane by forcing the crossover angle (which provides a minimum curvature radius) \(\phi_1 = \pi/2 + \beta\) in the expression for pinning on cylindrical pillars:

\[
R_{1,\text{min}}(\phi_1) = \frac{d/2 - dp \sin(\pi/2)}{\sin(\pi/2 - \phi_1)}. \tag{2}
\]

In Eq. (2), \(d\) is the pillar pitch in a rotated reference frame, given by \(d = (d_g + d_p)/\cos(\beta)\). The maximum pressure drop that the interface can sustain before breaking is then \(\Delta P_{1,\text{max}} = \gamma(1/R_{1,\text{min}} + 1/R_z)\).

(iii) When the pressure drop across the side interface exceeds \(\Delta P_{1,\text{max}}\), the contact line on pillar A will proceed from T to U and the contact line on pillar B will slide downwards from S to V (Figs. 2(e) and 2(f)). A new pinning situation is established at U. The radius of the interface in the \(x,y\) plane in this position is derived geometrically

\[
R_{2,\text{min}}(\phi_2) = \frac{1 - \cos(\beta + \phi_2 - \pi/2) + d_g}{\cos(\beta + \phi_2 - \pi/2) \sin(\beta - \phi_2 - \pi/4)}, \tag{3}
\]

where \(\beta\) is the oblique angle of the pillars, \(d_g\) is the spacing between pillars, and the crossover angle \(\phi_2\) was found numerically (Fig. 2(f)). We note that if the crossover angle does not satisfy \(\phi_2 \geq \phi_2 = \pi/2 - \beta\), the point U will be situated on the transition from the circular to the straight edge of the pillar, and we must force \(\phi_2 = \phi_2\) to find the minimum curvature radius. Laplace’s law gives the maximum pressure drop that the interface can withstand in this position before breaking as \(\Delta P_{2,\text{max}} = \gamma(1/R_{2,\text{min}} + 1/R_z)\).

The three stages described above can be translated into design requirements to obtain a functional module. At the side of the interface pinned at the pillars, the smaller of \(R_{1,\text{min}}\) and \(R_{2,\text{min}}\) will indicate whether the strongest pinning occurs in stage (ii) or (iii). We define a pressure ratio \(\kappa\), which provides a comparison between the maximum pressure drop across the interface at the orifice and the blocking pillars. Equations (1)-(3) can be combined with Laplace’s law to form the inequality

\[
\kappa = \frac{1/\max\{R_{f,1}, R_{f,2}\} + 1/R_z}{1/\min\{R_{1,\text{min}}, R_{2,\text{min}}\} + 1/R_z} < 1. \tag{4}
\]

For the front interface of the spacer to break at the orifice before the side interface breaks at the blocking pillars, \(\kappa\) must be smaller than one. Figure 3 shows the pressure ratio \(\kappa\) against the oblique angle of the pillars for various ratios of orifice width to pillar spacing \((d_o/d_g)\).

According to Fig. 3, the design with \(d_o/d_g = 2\) and \(d_g = 15\mu m\) satisfies \(\kappa < 1\) for all oblique angles. Therefore, any angle can be chosen if the gap parameters \(d_o\) and \(d_g\) are kept constant. In doing so, we took into account the two additional consequences of changing the oblique angle \(\beta\): (i) as \(\beta\) increases, the radius \(R_z\) of the rear interface decreases. A smaller radius of the rear interface of the trapped spacer leads to a higher pressure drop across that interface. This leads to more rapid merging with the front interface of an incoming spacer. (ii) The oblique pillars direct the incoming volume into the bypass more strongly as \(\beta\) increases, as pinning of the interface only occurs at the left edge of the pillars, while the section of the interface at the right edge is able to deform towards the bypass in the \(-x\) direction. These two influences of \(\beta\) are related to the dynamics of spacer removal (interface propagation and merging) and are less easily quantified than the static (pinning) situation described by Laplace’s law.

Taking the design considerations above into account, we chose \(\beta = 30^\circ\) as a trade-off between a low pressure ratio \(\kappa\), a small rear interface radius \(R_z\), and an unobstructed path along \(-x\) for rapid merging of spacers.

Typically, the apex of the MFP head is about 20 \(\mu m\) from the surface, and the volume of confined liquid is on the order of 100 pl. A change in the volume of injected processing liquid from 100 pl to 120 pl then corresponds to a 10% increase of the radius of the footprint. The extent of the footprint variation, caused by hydrodynamic coupling between the removal module and the HFC, is important as it determines the accuracy and reproducibility of local surface processing. To study this coupling, we designed and compared two probe heads with different spacer-removal modules (Figs. 4(a) and 4(b)).

In a dual-aspiration module (Fig. 4(a)), \(Q_{HFC} = Q_i - Q_{oil} = 0\) as the incoming spacer blocks the flow at the blocking pillar. This brief period without injection of processing liquid lasts for \(\Delta t_{idle} = V_i/Q_i\). However, during this time, some processing liquid remains between the apex and the surface. If the time required to aspirate the remaining
volume of processing liquid exceeds $\Delta t_{\text{dual}}$, the hydrodynamic confinement persists. Therefore, by simply setting a higher arrival rate of spacers at the module, fluctuations in the footprint can be reduced.

In the two-channel module (Fig. 4(b)), a single aspiration inlet is used to form a HFC and to remove spacers coming from the removal module. In this case, an additional time $\Delta t_2 = V_a/Q_a$ is considered during which the flow of the immiscible phase from the bypass into the aspiration channel blocks the aspiration of continuous-phase processing and immersion liquid. Typically, the volume of incoming spacers is equal to the volume of ejected spacers. Therefore, the time during which the area of the footprint is not constant is increased by $\Delta t_1 + \Delta t_2 - \Delta t_{\text{dual}} = 1 + Q_r/Q_a$ in the single-aspiration module as compared with the dual-aspiration module. Under standard operating conditions, where $Q_a \approx 3Q_r$, this corresponds to a total fluctuation time that is approximately 30% longer for the single-aspiration module.

We tested a spacer-removal module integrated into a MFP head using an emulsion of Fluorinert FC-40 in water containing 50 μM Rhodamine B for visualization. The fabrication of the silicon/glass MFP head is described elsewhere.

The MFP head had an on-chip T-junction for the insertion of FC-40 spacers upstream of the removal module. We observed a continuous HFC during removal (Fig. 5). It can be seen that the front interface at the orifice breaks before pinning occurs at pillar A, and that pinning of the interface at pillar B occurs with a crossover angle $\varphi_1 \approx 120^\circ$. This indicates that the simple model based on the dimensionless parameter $\kappa$ is sufficient to predict that pinning will first occur at the orifice, followed by pinning at pillars B and A.

We note that the model quantitatively describes only the static pinning behavior of the oil interface. Therefore, the model can be expected to accurately scale to different geometries ($\beta$, $d_o$, $d_g$). However, as discussed above, the model does not quantitatively describe the two dynamic influences on the removal characteristics of the spacer-removal module. The dynamic influences are independent of $d_o$ and $d_g$ but contribute to the removal characteristics more strongly as the oblique angle increases. This contribution is convenient when designing a functional device, as dynamic influences will lower the apparent pressure ratio $\kappa$. Correspondingly, our quantitative model of the static influences is expected to more accurately describe module geometries with low oblique angles than geometries with high angles ($\beta > 45^\circ$).

During typical experiments lasting tens of minutes, no injection of spacers into the immersion liquid was observed. After the flow was initiated, the module showed a consistent, predictable behavior, removing all spacers, and adapting to changes in the flow rates up to an arrival rate of $\sim 15$ spacers per second.

We demonstrated that two-phase segmented flows can be used in a microfluidic probe while maintaining a stable confinement of processing liquid. Such segmented flows open the route for localized sequential chemistry by reducing dispersion. With this, one can envision highly efficient patterning of proteins on surfaces and multiplexed immunohistochemistry on tissue sections.

This work was supported by the European Research Council (ERC) Starting Grant, under the 7th Framework Program (Project No. 311122, BioProbe). We thank Julien Cors, Ute Drechsler, and Yuksel Temiz for their help in fabrication and Andrew deMello and Robert Wootton (ETH Zürich) for valuable discussions. Carlotta Guiducci (EPFL), Robert Lovchik, Emmanuel Delamarthe, Bruno Michel, and Walter Riess are acknowledged for their continuous support.

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