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Fast built and designed microdevices for early-stage liquid-liquid systems studies

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Abstract

This paper presents a comparison of two different microdevices able to produce oil in water droplets without wall treatment or surfactant. A fast technique to build capillaries assembled devices and a process for silicon-glass microdevices are exposed. The aim of this paper is to compare those devices in terms of droplets generation and to show that the capillary assembled device can be used for preliminary tests in droplet generation as a precursor for silicon-glass microdevices.

1. Introduction

For chemical engineering, controlling liquid-liquid flows in a microchannel is a major challenge. One main application for this kind of systems is to acquire data to identify kinetic [1], thermal [2] or mass transfer [3] models. Furthermore, the microscale is also directly used to generate objects at a controlled dimension. Applications can be found in crystallization [4] or polymerization [5].

For the experimentation, first stage is to develop a microfluidic device design which often presents different parts in terms of operations (mixing, droplets generation, residence time for the reaction activation, phases separation).

A lot of iterations between design and manufacturing could be needed to obtain a complete operating device. It is then of interest to be able to generate fast built and cheap systems to explore the early-stage designs.

Liquid-liquid flows in microchannel are governed by many parameters such as the nature of the fluids, the injection mode, the geometry and the surface characteristics of the channels. As interfacial effects are a key factor, lots of works have been made on injection geometry and different materials have been used for microfluidic devices.

Many regimes of droplet generation have been studied by using microfluidic flow focusing in two-dimension [6]. Regular flows are obtained with simple cross-junction [7]. T-junction is also used to form droplets [8]. Water-in-oil emulsions are more often studied than oil-in-water in these devices except in microchannel emulsions [9]. These different geometries are developed to increase the shear rate on the dispersed phase. But, the interactions between liquids and walls are also an important parameter for the droplet generation. In particular, the continuous phase should be wetting the surface and the dispersed one non-wetting [10].

Intrinsic fluid properties such as viscosity, density or interfacial tension and flow conditions are responsible of the droplet break-up. To explain this phenomenon, investigations have been made on dimensionless numbers: Capillary, Reynolds or Weber numbers [11-13].

It is possible to modify these fluids properties and help droplet generation and stabilization with the use of surfactants or viscous additives [14,15]. Different techniques of wall treatment can also be applied on surfaces to make them hydrophilic or hydrophobic: siliconization, silanization or polymer coating for glass or silicon [16]; UV/ozone treatment for elastomers [17]. But the simpler is to adapt material to the kind of emulsions needed. Consequently, manufacturing techniques are often adapted to the material used.

Plastic (PDMS, PU, PMMA...) devices are made by photolithography [18]. A silicon mask with the desired design is first created. Then polymer is moulded on this mask and cured with temperature; a

flat polymer layer is then sealed to the polymer replica by plasma oxidation. PDMS is a cheap material and allows rapid prototyping by replica moulding but it is not suitable for oil-in-water droplets. Although PDMS can be treated to become hydrophilic, it is not long-term available [19,20]. Silicon or glass devices are fabricated in two main steps: channels formation by dry [21] or wet [22] etching, device capping by anodic or diffusion bonding [23]. Glass or silicon microstructuring is a reproducible and reliable technique but it requires specific equipment and use restrictive operating conditions. Glass and silicon devices are suitable for oil-in water droplets generation but are very expensive.

Based on the work of Hisamoto [24] that embedded functionalized capillaries in a PDMS network for analytical purposes, we propose to use untreated fused silica capillaries embedded in Plexiglas plates to generate oil-in-water droplets. Nisisako [16] used PMMA milled plates to generate water in oil droplets and Xu [14] added a quartzose capillary in the PMMA network to provide aqueous phase. In order to approach silicon-glass devices, the droplets flow directly in the fused silica capillaries, Plexiglas plates are just a support for capillaries.

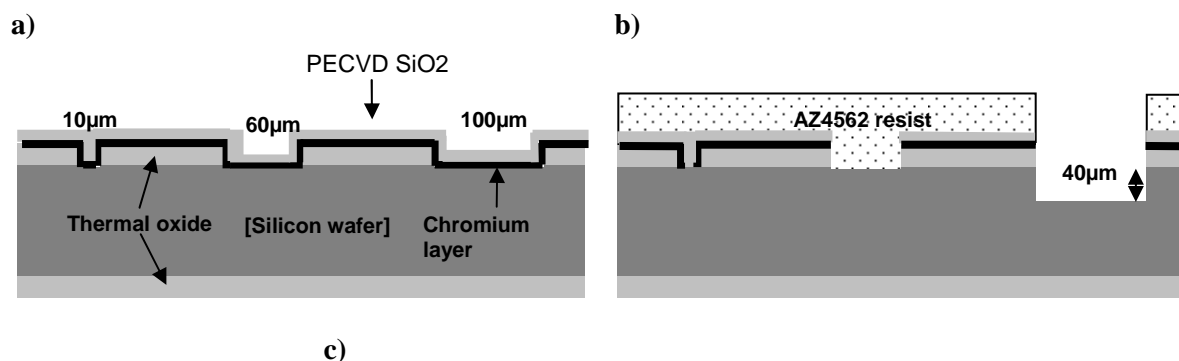
This paper proposes a comparison between two different microfluidic devices: a silicon-glass microchip and a device based on fused silica capillaries assembly. The objectives of this paper are (i) to describe a fast technique to build capillaries based microsystems; (ii) to present results obtained with this device in term of droplets generation, (iii) to show that such a device can be used as a precursor of silicon-glass devices.

2. Device Manufacturing

2.1. Silicon-glass device

Silicon-glass devices are fabricated with two major steps: plasma etching to define both the microchannel and the inlet and outlet holes through the silicon wafer and anodic bonding to seal the channel with the glass wafer. Deep Reactive Ion Etching (DRIE) is commonly used for the fabrication of silicon microstructures when high aspect ratios up to 30:1 and vertical sidewalls are required. For the fabrication of a microchannel with a precise depth, a single photoresist layer is usually necessary to mask the silicon wafer during etching. For the microstructuring of microchannels with various depths on a same silicon wafer, it is necessary to use more complex masking layers. For our application, we needed to etch channels of 10 μm , 60 μm and 100 μm depth, the two smallest emerging into the widest one. To do that we used a technological process developed at IMM [21] (Huber 2003 *et al*).

This process necessitates to deposit four masking layers (thermal and PECVD silicon oxides SiO_2 , a chromium layer and AZ4562 photoresist layer), to perform three photolithographies, three dry etching of the oxides, two wet etching and three deep silicon etching. A schematized description is reported on figure 1.



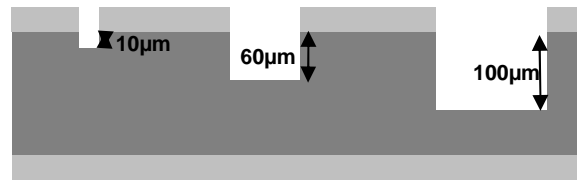


Figure 1 Cross-sectional schematic view of the multilevel silicon depths and widths process. (a) Patterning of the 10µm, 60µm and 100µm-wide channels. (b) First 40µm-deep etching for the 100µm-wide channel. (c) DRIE etching of 50µm and 10µm-deep respectively to obtain the three desired channel depths.

The different steps are described here: after deposit of a 0.5µm thermal oxide on the silicon wafer, the three channels are patterned; deposit of 0.1µm of evaporated chromium layer followed by 1µm PECVD silicon oxide (figure 1a); after the 60µm and 100µm channels opening by PECVD SiO₂ dry etching, a first DRIE of 40µm-depth is performed (figure 1b); after AZ4562 resist removing, a second DRIE of 50µm-depth is made; after PECVD SiO₂ and chromium removing, the last DRIE of 10µm-depth is performed (figure 1c). As a result all the channels are structured.

Figure 2 shows a MEB view of a 60µm wide and deep silicon channel emerging into a 100µm wide and deep channel.

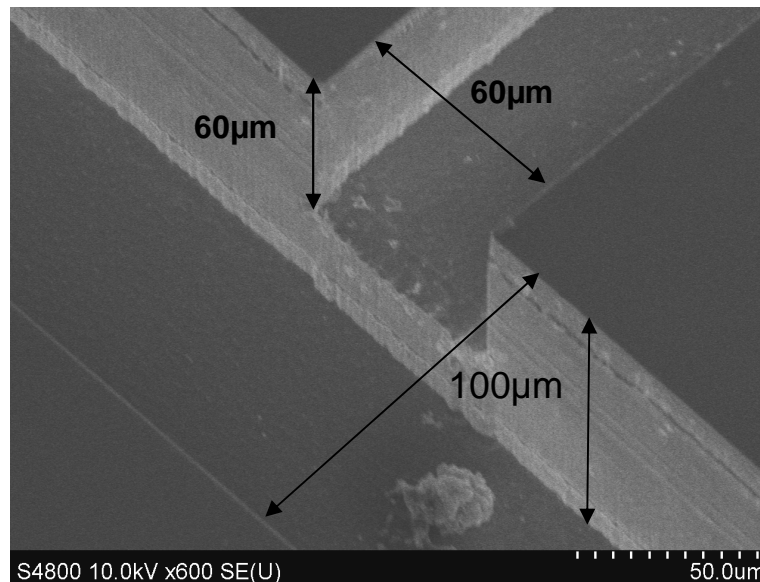


Figure 2 MEB view of a 60µm wide and deep silicon channel emerging into a 100µm wide and deep channel.

There are two possibilities to define the inlet and outlet of the channels: either perform holes through the cap glass wafer by drilling, or perform through wafer holes into the silicon wafer by DRIE. The first method is easier but not appropriate for glass wafers of 500µm-thick. So we opted for dry etching of holes of 1mm-diameter through silicon. At the end of the process the glass wafer (Pyrex 7740[®] from Corning) and the structured silicon wafer are sealed by anodic bonding.

2.2. Capillary device

In this device, capillaries are imprisoned and glued in two Plexiglas support plates. The lower plate is, first etched thanks to a computer numerical controlled milling machine and a micro drill which width is closed to the outer diameter of capillaries. T or cross junctions, as generating areas, are then created on the centre of the plate by computer-aided design. Additional channels of larger dimension are machined far around the generating area: they will receive the excess of glue and prevent the clogging of the junction while capping the device. The upper plate is flat and provides confinement to the device.

Capillary tube lengths (at least 20 centimeters) are adjusted to that of the channels; an additional length is needed to make the connections with the syringes and prevent capillary breaking when bending is important. The cutting has to be straight which is achievable with a simple ceramic cutter;

imperfect cutting can play a negative role on the droplet generation. It is checked with a microscope before using the capillary in the device. For each tube, in order to eliminate the protective coating, the side used for the generating zone is stripped by burning and cleaned with ethanol. This provides a window where phenomena can be observed. The setting of the capillaries in the channels is described on figure 3.

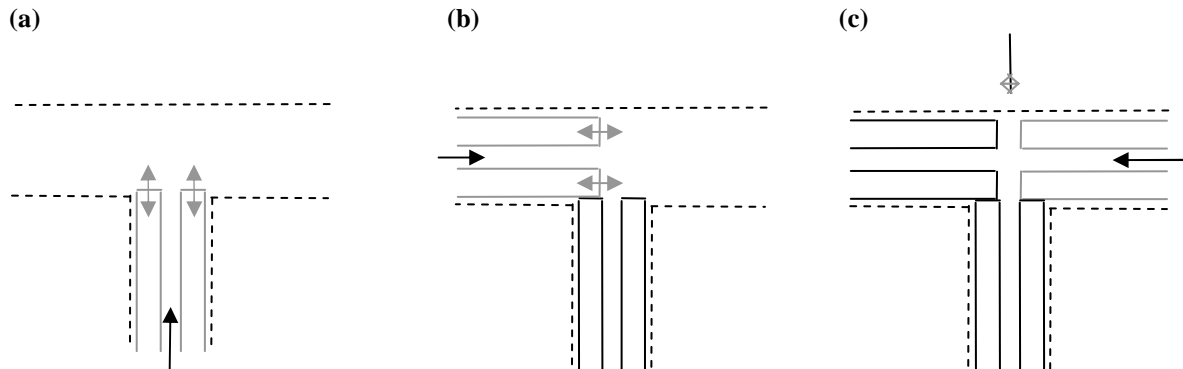


Figure 3 Steps of capillary placement in the etched channels for a T-junction: (a) dispersed phase supply, (b) continuous phase supply, (c) two-phase flow capillary. Capillaries are in full lines and channel walls in dotted lines

It starts with the lateral one (figure 1a). Its outlet reaches the end of the etched channel. Once the positioning is right, quick setting glue is used to fix the tube with an adhesive dot on the edge of the plate.

The first longitudinal tube is then added to the device (figure 1b). The end of the tube coincides with the output of the dispersed-phase tube. The walls of the respective tubes can touch each other. Once the tube is precisely set, another adhesive dot freezes the distance between the two outlets.

The last tube is finally set on the plate (figure 1c). In order to have a symmetrical device, the main and the continuous phase tube are at the same distance from the centre of the dispersed phase one.

As all the capillaries are fixed to the plate, a layer of low setting glue is spread in the channels (except in the drop generating site or the enlargement area to avoid obstructing the tubes) in order to decrease the dead-volumes. The glue should not overpass the channels. During the time the glue is thickening, it is possible to adjust the placement of the capillaries tubes laterally to make them touch each other. Precision of the T is strongly dependant on that of the capillary cutting.

When the glue is hardened, the cap is ready to be fixed. Its dimensions are slightly inferior to the lower plate to favour the bonding. The gluing process is described on figure 4. The same low setting glue is used: a few clusters are set all around the larger channels. The cap is settled and both plates are pressed against each other by hand. The pressure applied has to make the glue spread in the larger channels and around the T but not reach the generating site. The pressure is kept a few minutes until the glue is starting to harden. It is then, possible to add a glue layer all around the junctions of the two plates to increase the tightness of the device.

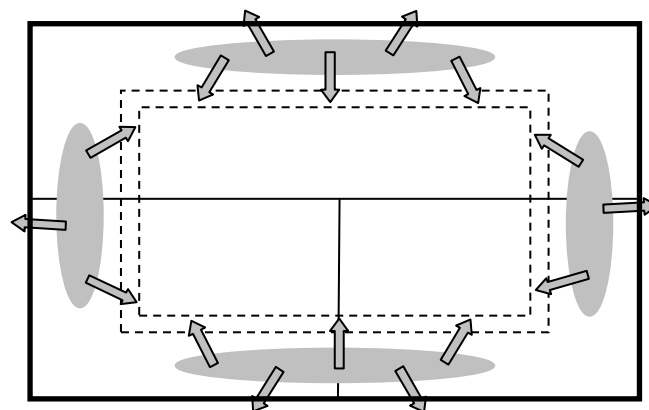


Figure 4 Schematic view of the sealing process: when pressing both plates, the glue (grey) spreads out of the cap (bold square) and into the larger channel (dotted lines) preserving the T-Junction.

The complete manufacturing takes less than a day before starting experiments. The key step of the fabrication is the cap bonding: there must be enough glue to bring tightness to the device but glue has to be cautiously spread to avoid capillary clogging. Figure 4 presents two examples of devices built following this protocol with different generating junctions: T (a) or Cross (b)

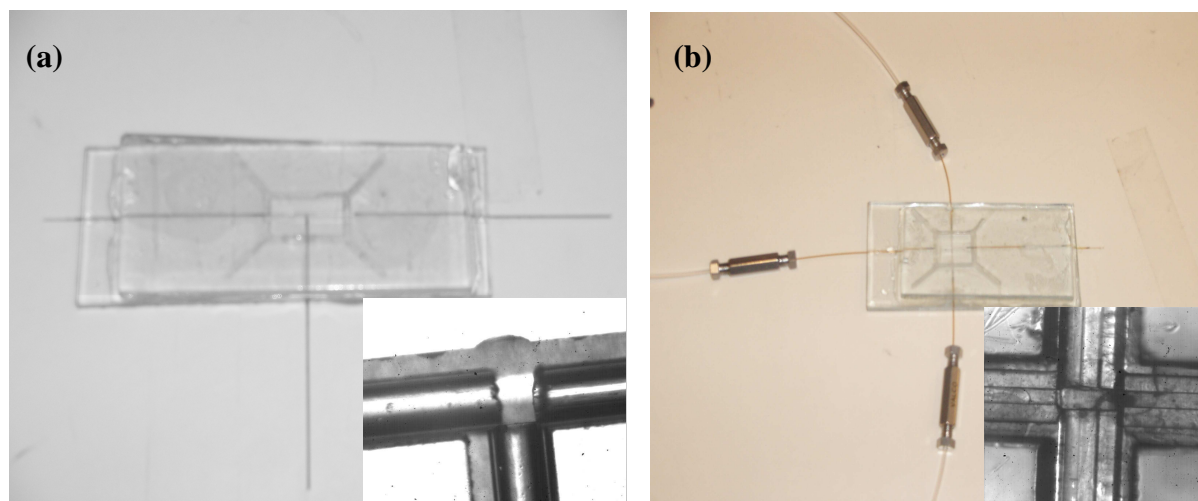


Figure 5 Examples of devices built: (a) T junction, (b) cross junction. On the bottom right, there is a zoom on the generation area

By using the same gluing technique, it is possible to connect capillaries. An enlargement (respectively a reduction) of the main tube diameter can be made by adding a second capillary end to end with it (figure 5a). This junction requires a good precision of capillary cutting and also needs confinement so it has to locate between the two Plexiglas plates. As it is difficult to handle with short-length capillaries, the contact is then made between the larger channel and the edge of the plate. The enlargement can be useful to transform a plug flow to a droplet flow

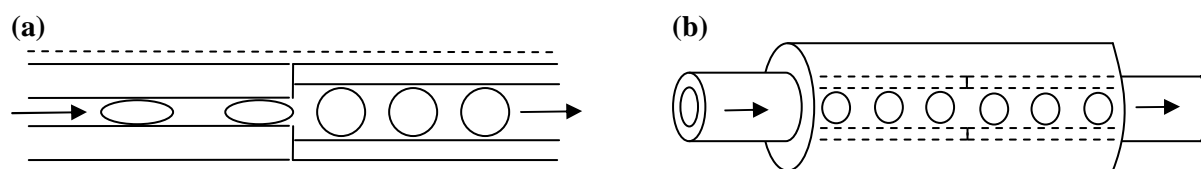


Figure 6 Examples of connections between capillaries: (a) diameter enlargement, (b) 2-block connection

In order to connect different devices made of capillaries, a larger capillary can be used. Standard capillary is 360 microns outer diameter. If 360 microns capillaries are stripped of their coating, they can be inserted into a larger capillary with an inner diameter of 320 microns. Only a few centimetres of larger capillary are needed and it is better if the both small capillaries are inserted symmetrically as shown on figure 5b. Once both small capillaries are put in contact they are fixed with the larger one with two dots of quick setting glue.

The capillary technique presented here does not pretend to be completely reproducible. Capillaries are indeed cut and placed in the channels by hand, it is thus nearly impossible to reproduce exactly the same device twice.

Another key point is the reliability of such devices; a clear attention has to be paid to which solvents are used in these devices: for example, acetone will damage Plexiglas plates. Some solvents may dissolve the glue used to seal the device or create block connections. The commercial glues used for the fabrication are resistant to temperature in the range of -30 to 70°C and Plexiglas can afford up to 80°C. If extended temperature range (up to 350°C) is needed, it is possible to thermostat the capillary at the outside of the device. In terms of mechanical resistance, the capillary device can be fed at 10

ml.h⁻¹ with 100 microns square section capillaries on a length of 50 centimeters, which corresponds roughly to 4,5 bars, the devices may then work without leakage up to 5 bars.

The fabrication of such devices is pretty fast and it only uses cheap materials. As a wide collection of capillaries is available through the market, the capillary device is then modular: with the same milled plates, it is possible to produce many different devices with varying inner diameters; it is also possible to change the diameter of the mill to adapt it to the capillary outer diameter. The connection technique presented above extends further the possibilities of such devices beyond the simple droplet generation.

3. Experimental material

The capillary tubes used have either a square section (side 100 μm) or a round section: standard 360 microns outer diameter and larger ones. They are made of fused silica and coated with a protective polyimide layer.

The quick setting glue is cyanolite glue. The low setting one is a bi-component araldite. The plates are made of Lexan Plexiglas or polycarbonate.

The fluids are stocked into 1 to 20 ml syringes placed on syringe pumps. They are injected at constant flow through polyethylene tubing. The connection with the capillaries is then assured with fittings without dead-volumes (figure 4b).

To protect Si-Glass microreactors and make the connection with syringes, a case has been designed. The case is composed of two Plexiglas round plates maintained by screws. The inferior plate is bored and threaded: each hole corresponds to each input or output of the microreactor. The same polyethylene tubing is then used to connect syringes to headless nanoports mounted with viton joints. These nanoports are screwed in the inferior plates until the microreactor touches the upper plate. Openings are made on the upper plate in order to check if the microreactor is well-centred on the case. Viton joints bring the tightness to the system.

The dispersed phase is silicone oil (20 cSt viscosity) and the continuous phase is demineralised water. As the channels have a small width and consequently, the droplets velocities are very high, the observation is made with a fast CCD Camera coupled with a microscope (Nikon SMZ-10). Images acquisition rate is 1800 frames per second. Exposition time (0.55 ms) is chosen in order to have enough light for contrast distinction and short exposition for the droplets edges sharpness. A 100W spot lamp is used to provide necessary light.

The images are acquired with transmission light at the exit of the capillary device whereas they are acquired just after droplet generation with reflexion light with Si-Glass microreactors.

4. Results and discussion

This part describes results obtained with silicon-glass microreactors and capillary devices while generating oil droplets in water. Both devices are compared and an empirical model of droplet size is proposed.

4.1. Silicon-glass device

First fluid admittance in the Si-Glass microreactors can be difficult, so before first use, they are kept in water for a few hours. Because of microscope limitations and difficulty to introduce fluids, it was difficult to handle with 10 microns microchannels, then only reactors with 60 microns and 100 microns microchannels are presented in this article.

Before starting droplets generation, it was important to determine the range of flow rates in each device. In order to find these operating conditions, the oil flow rate (F_o) is set at 100 μL.h⁻¹, while water flow (F_w) rate is changed.

For 60-microns microchannels, oil flow rate varies from 10 to 200 μL.h⁻¹ and water flow rate from 100 to 1500 μL.h⁻¹. For 100-microns microchannels, oil flow rate is set between 50 to 500 μL.h⁻¹ while water flow rate between 500 to 4000 μL.h⁻¹. More important water flow rates can be delivered through

the channels but there is a strong observation limitation due to reflection light. At high flow rates, the interface between water and oil becomes less and less sharp up to having difficulty to distinguish oil droplets.

At first use, flow becomes stable after 15-20 minutes and slug regime is observed. The time of flow stabilization is lower when just a parameter is changed. Frames are acquired just after the generation site. Slugs appear to be monodisperse (slug length variation < 5%). As expected, when the water flow rate increases, the droplet size decreases and the distance between droplets increases.

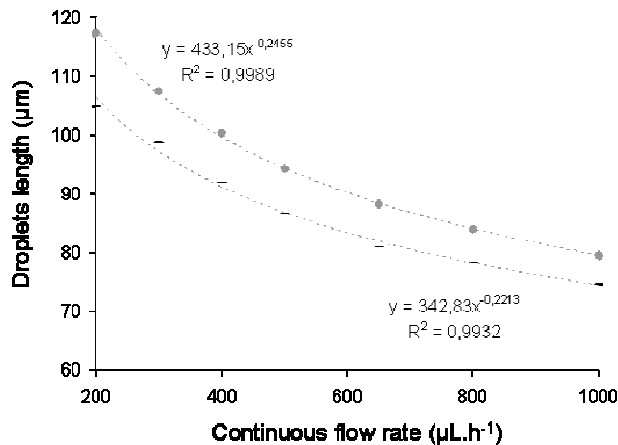


Figure 7 Slug length function of water flow rate at different oil flow rates: 50 $\mu\text{L.h}^{-1}$ (\bullet) and 10 $\mu\text{L.h}^{-1}$ (\triangle)

Dispersed phase flow rate has also an impact on slugs' length. Figure 6 presents data on 60x60 microchannels: reducing oil flow rate from 50 to 10 $\mu\text{L.h}^{-1}$ involves a 10% decrease of slug length. Whatever the size of the channels, a potency law fits well the plug length in function of water flow or either Ca number. The exponent is within the range of 0.15 to 0.25 whatever the conditions or the channels' diameter.

The Si-Glass microreactors have a good reproducibility. Figure 7 shows the data obtained for two different plates at the same water and oil (100 $\mu\text{L.h}^{-1}$) flow rates. The difference on the slug lengths and slug velocities is pretty thin.

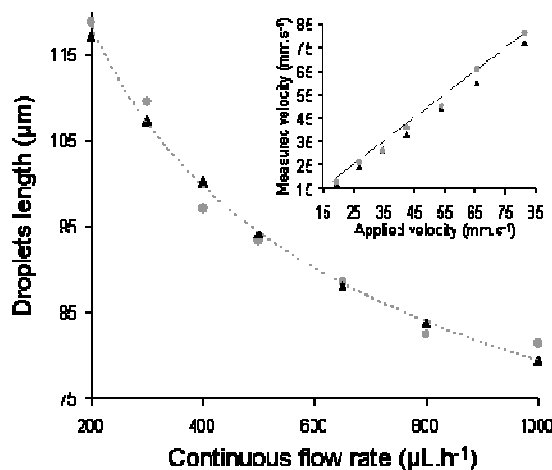


Figure 8 Reproducibility of droplet generation on different microreactors in terms of slug flow and velocity (upper graph)

The upper graph of the figure represents the measured velocity function of the imposed superficial velocity. The different measured velocities are very close to the imposed one (dotted line); it seems then that there is no slippage of the slug in the continuous phase.

On the 100x100 microchannels, it is possible to test the influence of the dispersed phase channel diameter on the slug length. Data are shown on figure 8. Results are pretty interesting because for the

same oil flow rate there is a slug length decrease of 20% by changing a 100 microns channel to a 60 microns. Distance between slugs is also reduced while using a 60 microns channel.

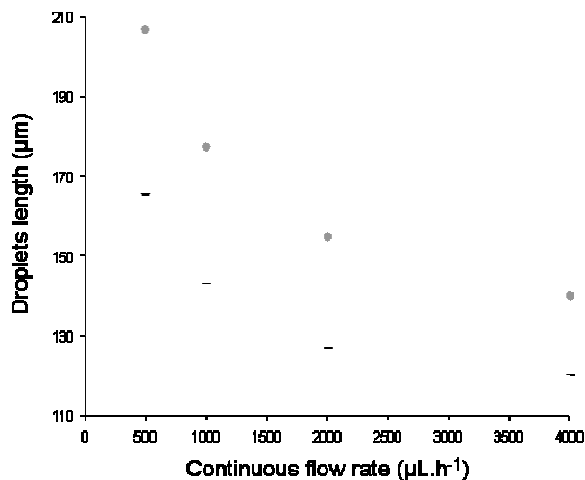


Figure 9 Slug length function of water flow rate at the same oil flow rates ($100 \mu\text{L}\cdot\text{h}^{-1}$) but with different dispersed channel size: 100 microns (●) and 60 microns (-)

Si-Glass microreactors are very simple of use and provide interesting results and good reproducibility. Nevertheless, they are pretty expensive and can be long to fabricate. It can then be interesting to have reactors much simple to produce and with comparable results in order to make preliminary tests and design conception. As fused silica is closed to glass and silicon, capillaries devices have been constructed on this purpose.

4.2. Capillary device

Before starting droplets generation, the air imprisoned in the device while bonding the plates, has to be removed. The presence of air in the device can block the flow or provokes serious turbulences. The aim of this article is not to present a detailed flow chart but to show the feasibility of droplets generation in this kind of device. As capillaries have the same diameter and square section, the same operating conditions are used. Despite the important dead-volumes, oil does not seem to enter in contact with the walls of the plates. A thin oil line follows the water flow line until the inlet of the output tube where the droplet break-up takes place. The water wets well the capillary walls while oil is relatively non-wetting. The flow becomes stable after a few minutes and droplets are monodisperse (variation < 5%).

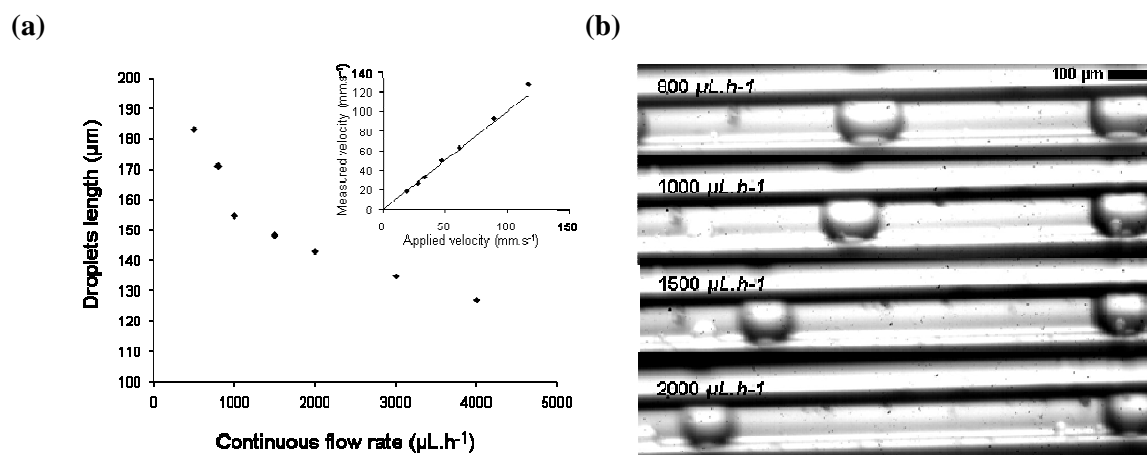


Figure 10 (a) Slug length function of water flow rate at constant oil flow rate ($200 \mu\text{L}\cdot\text{h}^{-1}$), slug velocity function of superficial velocity (upper graph) and flow visualisation at different water flow rates (b).

The measures are at constant oil flow rate ($F_o = 200 \mu\text{L}\cdot\text{h}^{-1}$) and water flow rate is varied from $F_w = 500 \mu\text{L}\cdot\text{h}^{-1}$ to $F_w = 4000 \mu\text{L}\cdot\text{h}^{-1}$. As expected, when the water flow rate increases, the droplet size decreases and the distance between droplets increases. It is noticeable on the frames of figure 9. The range of slug lengths decreases from $185 \mu\text{m}$ to $125 \mu\text{m}$. An increment in water flow rate may further reduce droplet length but it also reduces the sharpness of the droplets edge and makes difficult the length determination. In terms of droplets velocities, the range is from 20 to $120 \text{mm}\cdot\text{s}^{-1}$. In this device also, there does not seem to be a droplet slip velocity by comparison with the applied velocities.

4.3. Devices Comparison

The capillary used for the hand-made devices have a 100 microns square section. The dimensions are thus comparable to a 100 microns microchannel on the silicon-glass microreactor. A comparison of the slug length is presented on figure 11: a T junction is used in both cases for the droplet generation.

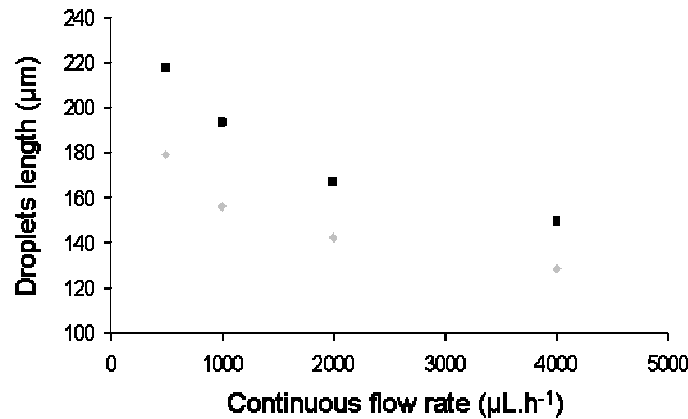


Figure 11 Slug length in function of water flow rate at the same oil flow rates ($200 \mu\text{L}\cdot\text{h}^{-1}$), injection mode (T junction) and channel size (100 microns) for the capillary device (♦) and the silicon-glass chip (■)

A variation of 30 to 40 microns slug length is noticeable between the capillary device and the silicon-glass chip. The difference seems to reduce at higher flow rate. Nevertheless both curves have quite the same shape. As the same flow rates, injection mode and diameter are used, it seems that this difference is due to the material. The other difference between both devices is the presence of dead-volumes around the generation area (figure 5a), but their main effect is to slightly reduce the shear rate of the continuous phase on the dispersed phase. Dead volumes should consequently have a negative impact on rather than helping the droplet generation.

4.4. Empirical model

Based on the precedent considerations, main parameters influencing the droplet generation [14,15,25] are listed: channels geometry, intrinsic fluid properties and flow rates. As it is easier to handle with dimensionless numbers, slug length is divided by the hydraulic diameter. Dimensionless numbers: Capillary number, that measures the magnitude of viscous to surface tension forces and Reynolds number, that measures the magnitude of inertial to viscous forces, are also worth entering the model. A potential law is used as it fits well the experimental data (figure 4). The resulting model is developed on equation 1.

$$\frac{L}{d_1} = A \times \left(\frac{d_1}{d_2} \right)^\alpha \times Ca_1^\beta \times Ca_2^\gamma \times Re_1^\delta \times Re_2^\epsilon \quad (1)$$

L is the length of the plug and d the hydraulic diameter of the channel. The capillary number is written $Ca = \mu Q / S \sigma$ with μ as the viscosity of the fluid, Q the flow rate, S the section of the channel, σ the interfacial tension between continuous and dispersed phases and Reynolds number $Re = \rho Q d / S \mu$ with

ρ the density of the fluid. A , α , β , γ , δ , and ε are constants. 1 refers to the continuous phase, 2 to the dispersed phase.

The model has been applied to the data presented previously on Si-Glass and capillary devices. The model parameters are identified by fitting the experimental values. Results are shown on table 1.

Table 1 Constants of the model obtained for a T junction in capillary and Si-Glass devices.

Models	A	α	β	γ	δ	ε
T junction Capillary	0.55	0.25	-0.11	-0.23	-0.07	0.29
T junction Si-Glass	0.65	0.25	-0.11	-0.23	-0.07	0.29

The comparison of the models and the data is represented on figure 12. Experimental data are placed on abscissa and calculated values on ordinate. The straight line symbolizes the perfect correspondence between models and experimental data; dotted lines a deviation of 10%. Calculated values for Si-glass devices (black squares) and capillary devices (grey dots) are closed to the experimental data. Models can thus be validated.

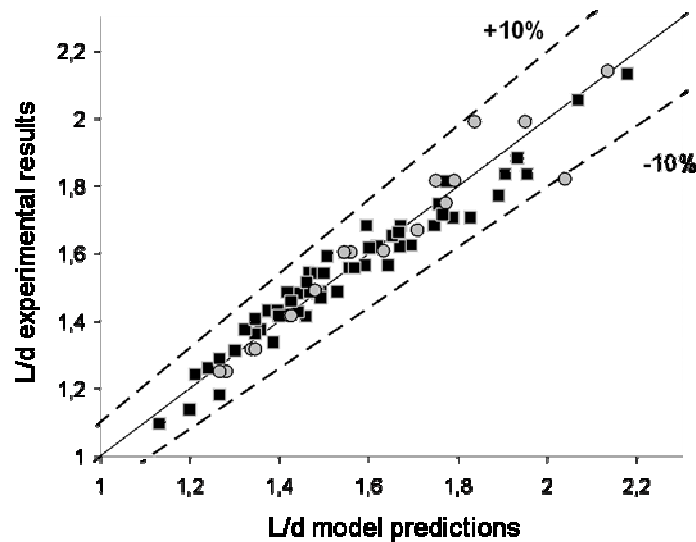


Figure 12 Comparison of the model with the data gathered for the capillary device (\bullet) and the silicon-glass chip (\blacksquare) in the range of slug aspect ratio between 1.1 and to 2.2

The only difference between the two models is on the constant A : it is higher for the Si-Glass device and this constant has approximately the same relative deviation than the variation of slug length in favour of capillary devices noticeable on figure 11. The exponents are common to both devices. This proves that is possible to build a model that could characterize and make the link between two microdevices using the same geometry and same fluids but different materials.

This similarity between the two devices is interesting: it offers the possibility to make preliminary tests on a capillary device before designing a silicon glass microchip. Capillary devices are indeed far less cost effective and faster to build. They can provide for example, data on the design or the residence time needed for special applications. The silicon glass device, more reproducible, will then be directly designed and functional for the desired application.

With the capillary device, it is also possible to connect different capillary blocks in order to apply further actions on droplets or provide more residence time. The connection described on figure 3b has been tested and three different blocks have been connected without leakage or perturbation on the two-phase flow.

5. Conclusion

This paper presents two different techniques to create microfluidic devices. Both devices are used to generate oil-in-water droplets without surfactant or wall treatment and present comparable results in terms of droplet size and velocity. The first device is a silicon glass microchip obtained by dry etching and anodic bonding. Its fabrication is complex and requires specific equipment and is cost effective but is very precise and reproducible. On the other hand, a simple and rather fast technique is used to create a device based on fused silica capillaries, Plexiglas plates and commercial glues. This device has the advantage to be cheap and modular. It is then a practical tool to make a preliminary study on droplet generation before designing a more complex device. As connections are possible between capillary devices, this droplet generator can be considered as the first operating unit in a chemical engineering approach. The connection between blocks will provide further microfluidic units such as reactive chamber, residence time or separation systems. Thanks to the similarity of both devices, the capillary device can be defined as a precursor of silicon-glass chips

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