

Microfluidic platform for reproducible self-assembly of chemically communicating droplet networks with predefined number and type of the communicating compartments

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Supporting Information

I. Bilayers and propagation of BZ waves in a quasi-2D system of droplets

In a control experiment we dispersed BZ-droplets directly in the oil phase which we used as the shell phase (5mg/mL solution of lipids in hexadecane). The emulsion was poured onto a Petri dish in which droplets sedimented to the bottom forming a quasi-2D system of sessile BZ droplets. Fig. S1 shows a sequence of snapshots showing propagation of BZ wave fronts between the droplets. Connections between the droplets are visible as bright lines, typical signature of a bilayer [1]. One can also distinguish contact angle θ between the drops being around 45 degrees, a value characteristic for inter-droplet lipid bilayers [2]. We can suspect that in this setup, due to presence of the substrate the bilayers form only sections of a circle (in contrast to full circles in the case of free droplets).

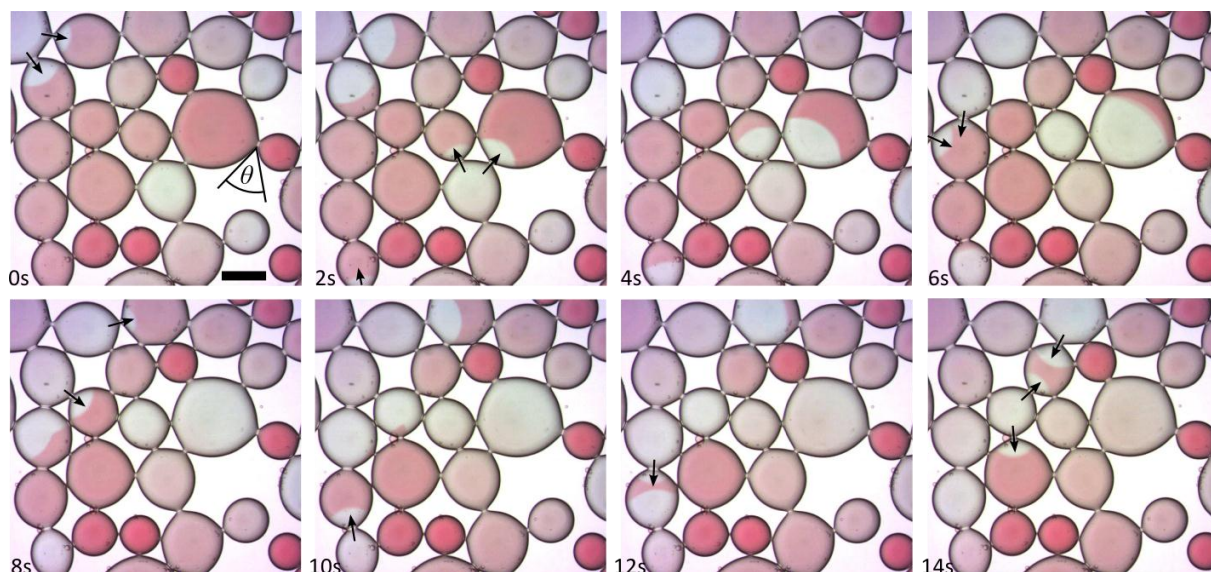


Fig. S1: Snapshots showing propagation of BZ wave-fronts between sessile droplets connected by lipid bilayers. The characteristic contact angle θ is indicated in the first snapshot (0s). The arrows indicate events of passing of the reaction front through a bilayer. Note that the smallest droplets do not oscillate, consistent with our on-chip results, therefore keeping their deep red color. Scale bar is 0.5 mm.

II. Periods of oscillation from single runs of experiment (for ‘singlets’)

In Fig. S2 we show periods of oscillations observed in droplets generated in four different runs of the experiment (representative out of total several tens of runs). The periods tend to be nearly constant for large droplets ($V \geq 0.4 \mu\text{L}$), but the corresponding values differ for different experiments. This indicates importance of physico-chemical conditions on-chip such as changes in temperature, small deviations in concentrations of reagents, interaction with polycarbonate walls or with the hydrophobic coating.

Seemingly, those conditions differed more between different experiments than between individual droplets in each of the experiments.

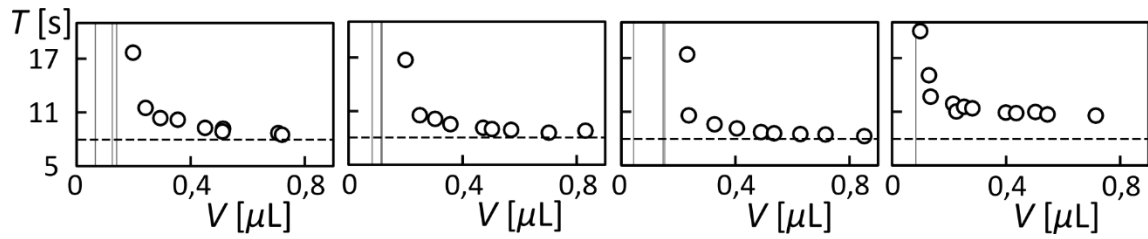


Fig. S2: Periods of oscillations of BZ droplets encapsulated each in an oil shell depending on the volume of the droplet, in four representative independent runs of the experiment. Vertical lines correspond to non-oscillating droplets and the horizontal dashed line indicates the period 8s of a reference droplet suspended directly in oil.

III. Microfluidic chip.

In Fig. S3 we show technical drawing showing the dimensions of the channels and a photograph of the microfluidic chip used in the experiments.

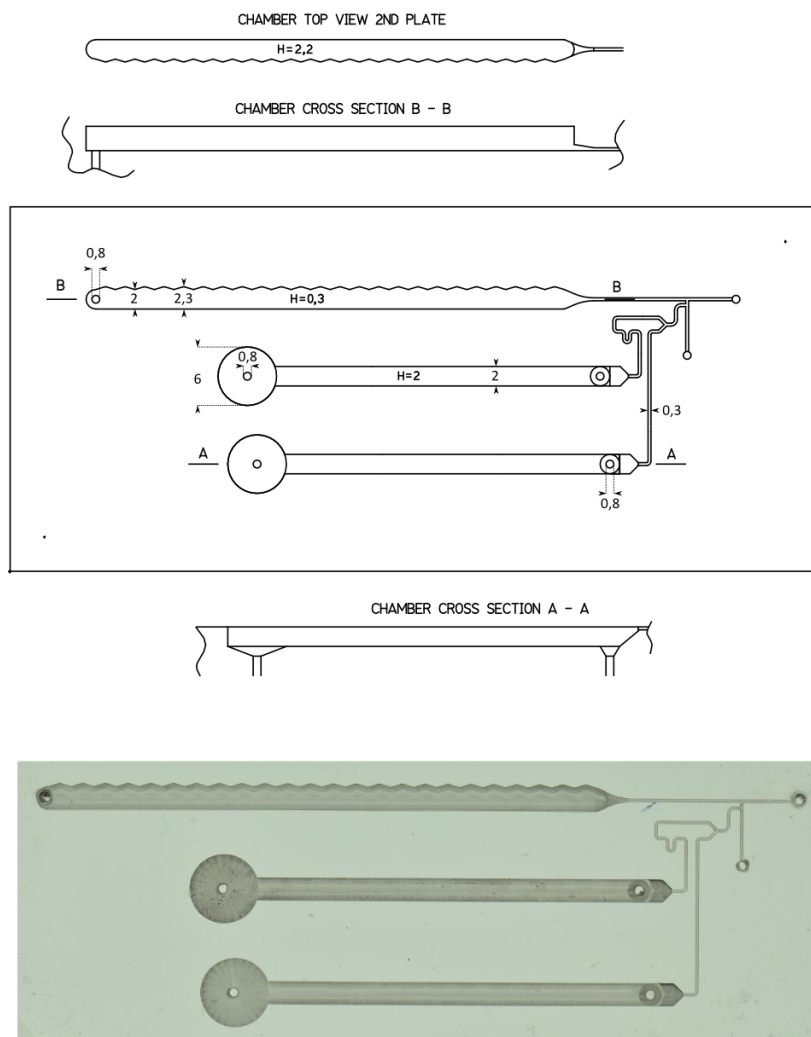


Fig. S3: Technical drawing with all dimensions in millimeters and with H denoting the depth of the milling. The chip was made of three polycarbonate plates, of which the first contained all the channels milled to various depths and circular inlet/outlet ports, as shown in the drawing; the second plate represented the chamber (as shown at the top; this plate was milled through as the thickness of the plate was 2.2 mm, the same as H); the third plate (not shown) was not milled at all and served as a cover for the channels.

IV. Evolution of the dynamic patterns in ‘doublets’ containing two almost identical droplets

In Fig. S4 we show time-evolution of the modes of propagation of the chemical wave-fronts between BZ-droplets in 3 representative ‘doublets’ (out of total 12 generated in the experiment) composed each of two droplets of nearly equal volume. One can notice that the modes change with time, finally approaching the ‘X’, ‘Y’, or ‘XY’ state (see main text for details).

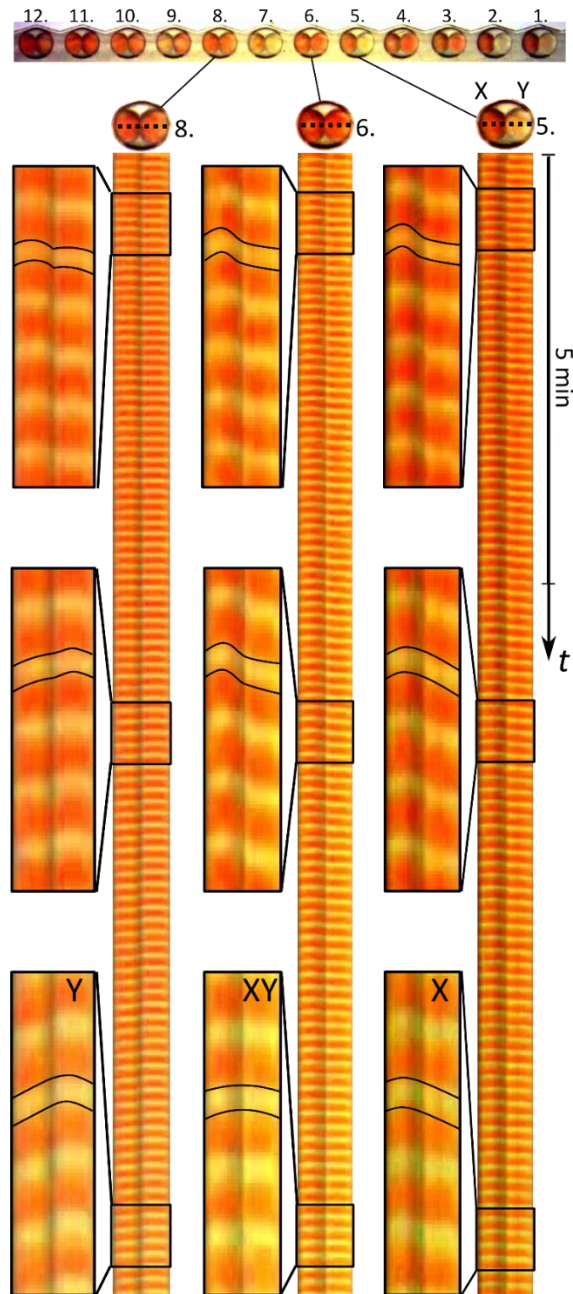


Fig. S4: Time-evolution of the dynamics patterns in doublets containing two almost identical droplets. The three cases are shown in which the final mode was 'X', 'Y' or 'XY'. The droplets are numbered according to the order of generation.

References:

- [1] S. Thutupalli, S. Herminghaus, R. Seeman, *Soft Matter*, 2011, **7**, 1312-1320.
- [2] J. N. Vargas, R. Seeman, J-B. Fleury, *Soft Matter*, 2014, **10**, 9293-9299.