SUPPLEMENTARY INFORMATION

SUPERCELLS: a novel microfluidic reactor architecture for ultra-fast sequential delivery of chemical reagents

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† Electronic Supplementary Information (ESI) available: Includes more details about the simulations, fabrication, set-up and results as well as a movie of an array of a supercells. See DOI: 10.1039/x0xx00000x

Supplementary Information S1

The transient 3D numerical simulations were conducted using Ansys Fluent 18.0. The simulated geometry domain is one supercell with a structured, hexahedral mesh of 1.7 million elements and a uniform, finite volume cell size of 0.5 x 0.5 x 0.5 um^3 (x, y, z). The advection-diffusion, scalar transport equation is used to model transport of the reagent species within the flow domain. The density and viscosity of water at 20 ºC is 998 kg/m^3 and 1.00 x 10^{-3} Pa·s, respectively, and reagent species mass diffusion coefficient is 10^{-9} m^2/s. A spatially-uniform, time-varying velocity boundary condition was applied to inlets 1 and 2 according to equations s1 and s2. A uniform, time-invariant normalized reagent species concentration of 0 and 1 is applied to inlet 1 and 2, respectively. A uniform outlet pressure of zero is assumed. A pressure-based coupled algorithm using a Rhie-Chow pressure dissipation correction with distance-weighted interpolation scheme is employed. For spatial discretization, a second order scheme is used for pressure and second order upwind schemes for momentum terms and species transport. A first order implicit scheme is used for time advancement with a uniform time step of 0.001 s. The flow domain was initialized to zero for velocity (x, y, z-directions), pressure and reagent species concentration.

Simulations of supercell geometry

Figure S1 shows the difference of a non-optimized and an optimized design where the hydraulic resistances of the outlet channels connected to the main cavity are modulated to equalize the advection times for all the supercell corners.

(a) (b)

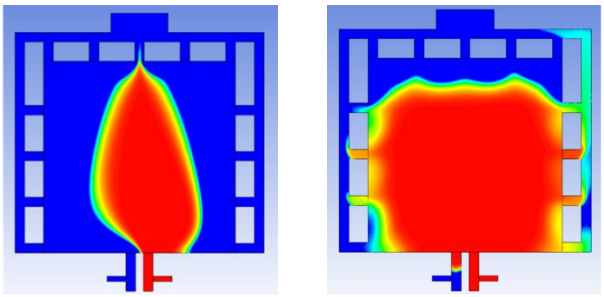


Figure S1: Comparison between (a) a non-optimized supercell with a outlet busbar where all the outlet channels have the same resistance and (b) an optimized design of a supercell where the widths of the outlet channels to the main cavity are adapted to modulate their resistances.

Simulations of experimental results

To simulate the concentration of the two reagents used in the specific design (see Figure 3), velocity inlet boundary conditions were used for both reagent inlets with time dependent velocity as given:

Inlet 1: eq (s1)

$$v_1 = 0.0002 + 2 * t \qquad (if\ t < 0.1)$$

$$v_1 = 0.2 \qquad (if\ t \geq 0.1)$$

Inlet 2: eq (s2)

$$v_2 = 0.2 - 2 * t \qquad (if\ t < 0.1)$$

$$v_2 = 0.0002 \qquad (if\ t \geq 0.1)$$

where v_1 is the inlet velocity of the first reagent in m/s, v_2 is the second reagent in m/s and t is the flowtime in s. For the outlet, a pressure outlet boundary condition with zero pressure was used.

Supplementary Information S2

Device Fabrication

For fabrication of the different fluidic layers, three silicon wafers were bonded together (see Figure S2). For each Si wafer either a channel or via was etched. After bonding, one of the wafers is thinned down and the next channel or via layer was etched to connect to the existing cavities. Figure S3a shows the cross section of Channel2 - Via1 - Channel1. Infrared bonding alignment was used for higher dimensions and allowed overlay error such as between Channel2 and Via1 (accuracy of <5µm). Figure S3b shows a scanning acoustic microscopy (SAM) image of Channel2 to Via1. The third blank wafer was bonded to the stack without alignment and then thinned down. The next two layers were aligned to the existing layers in the stack via infrared through the third wafer. This resulted in an alignment accuracy increase as steppers have higher alignment accuracy than bonding tools. The stepper that was used in this fabrication has accuracy of <2µm which was required for 2.5µm allowed overlay error in the two top layers. Figure S3c shows the alignment between Via0 and Supercell to Channel1.

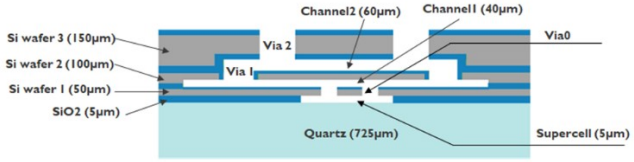
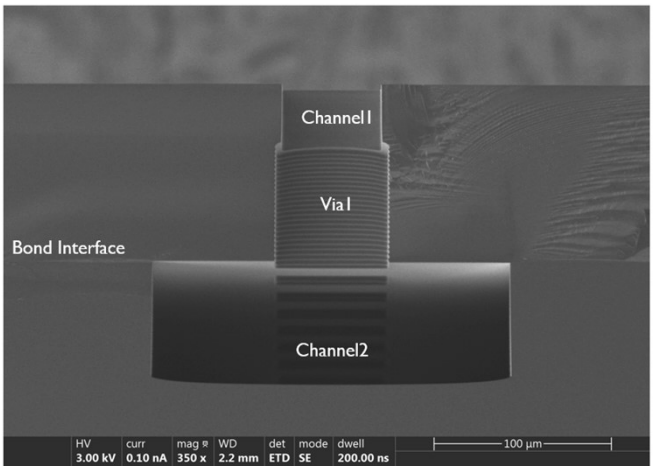
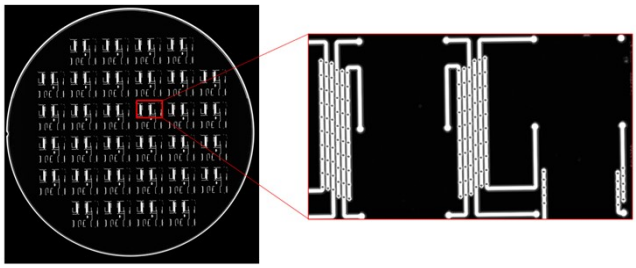


Figure S2: Schematic cross section (not to scale) of the fabricated fluidic device with the heights of the individual layers indicated.

(a)



(b)



(c)

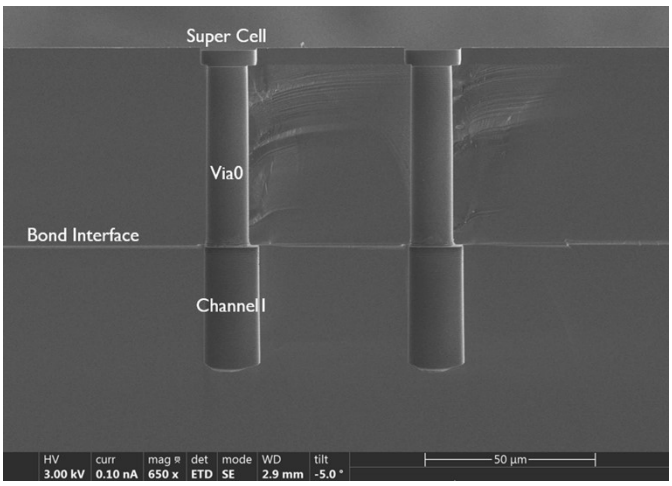


Figure S3: (a) Scanning electron microscopy (SEM) image of Channel2/Via1/Channle1 cross section; (b) Wafer-level sound acoustic microscopy (SAM) image after bonding of the first two Si wafers; (c) Scanning electron microscopy (SEM) image of Channel1/Via0/Supercell cross section.

Supplementary Information S3

Experimental set-up

The concept of high and low resistances is used for fast switching of the reagents. Each inlet was connected to a high resistance line (tubing with a small internal diameter) and a low resistance line (tubing with larger diameter). One of the reagents was connected to the low resistance line allowing it to flow into the supercell, while at the same time the other reagent was switched to the high resistance line. High and low resistance lines allowed low and high flow rates, respectively. For switching between the high and low resistance lines, external valves were connected to a switch board (Fluigent ESS Switchboard) and switching was done using a signal generator device. The resistance of these high and low resistances are described in eq (s4).

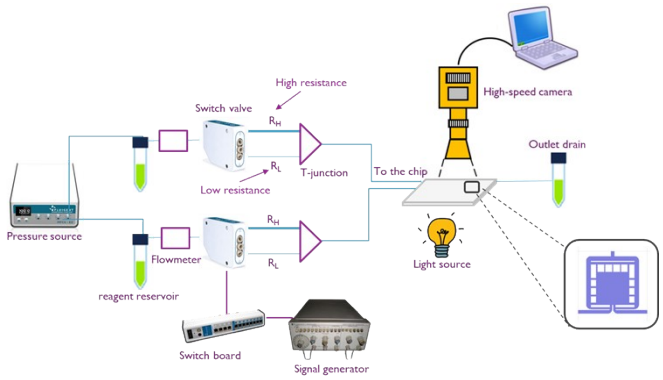
$$\Delta p = R \times Q \qquad \text{eq (s3)}$$

Where Δp is the pressure drop in Pa, Q is reagent flow rate in $m^3\ s^{-1}$ and R is the resistance in $Pa.s.m^{-3}$.

$$R = \frac{128\ \mu L}{\pi d^4} \qquad \text{eq (s4)}$$

Where μ is the reagent viscosity in Pa.s, L is the length of the channel/tubing in m and d is the diameter of the channel in m. This equation is only used to calculate the resistance of circular tubing in the experimental setup. To note, the resistance of supercell, 1.3e+15 $Pa.s.m^{-3}$, was obtained from the numerical simulation.

(a)



(b)

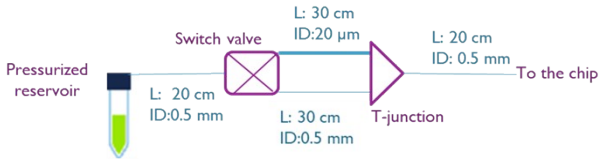


Figure S4: (a) schematic of the experimental set-up to test the fast switching of the proposed supercell design. Images are captured with a high-speed camera; (b) schematic of the low and high resistance lines allowing the fast switching in between the reagents.

Supplementary Information S4

The reagent replacing phenomena in the vertical (z) direction are also studied in the 3D numerical simulation described in Figure 4. Figure S5 shows the cross-sectional plots of reagent mass fraction in 5 selected planes at two moments, corresponding to the timepoints in Figure 4b and d. This gives the details on how one reagent (e.g. red, from inlet 2) interfaces and replaces the other one (e.g. blue, from inlet 1) over the switching time at different locations.

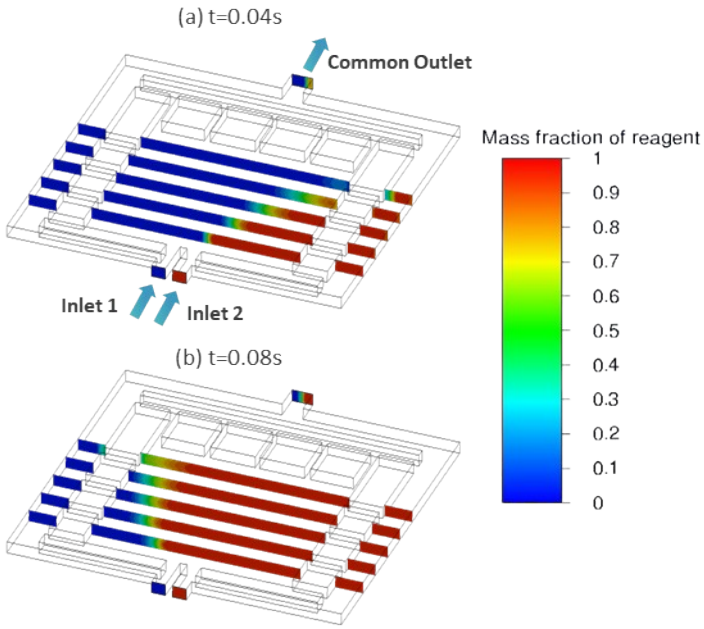


Figure S5: the cross-sectional views of reagent mass fraction in 5 planes at -50, -25, 0, 25, and 50 μm offset from the center of the cavity at t = 0.04 and 0.08 s. The numerical simulation details were the same as described in Figure 4.

Supplementary Information S5

Average switching times for different arrays at a flow rate of 10 nL/s. This switching time indicated in the table is the average of at least 3 supercells within the same 10 x 10 array. The results show slight differences between various arrays.

Table S1: Average switching times for different arrays at a flow rate of 10 nL/s.

	Average switching time [s]	STDV [s]
Array 1 (Figure 4)	0.17	0.07
Array 2	0.31	0.04
Array 3	0.30	0.05
Array 4	0.20	0.05
Array 5	0.22	0.06
Array 6	0.21	0.06
Array 7	0.17	0.07
Array 8 (Figure 5)	0.23	0.02

Electronic Supplementary Information (ESI)

The video shows an example of the switching between 2 reagents in the 10x10 array of supercells for a flow rate of 10 nL/s. Because of the limitation of the limited field-of-view of the microscope used, only 48 supercells are shown.