SUPPLEMENTARY INFORMATION

INTEGRATION OF CAPILLARY-HYDRODYNAMIC LOGIC CIRCUITRIES FOR BUILT-IN CONTROL OVER MULTIPLE DROPLETS IN MICROFLUIDIC NETWORKS

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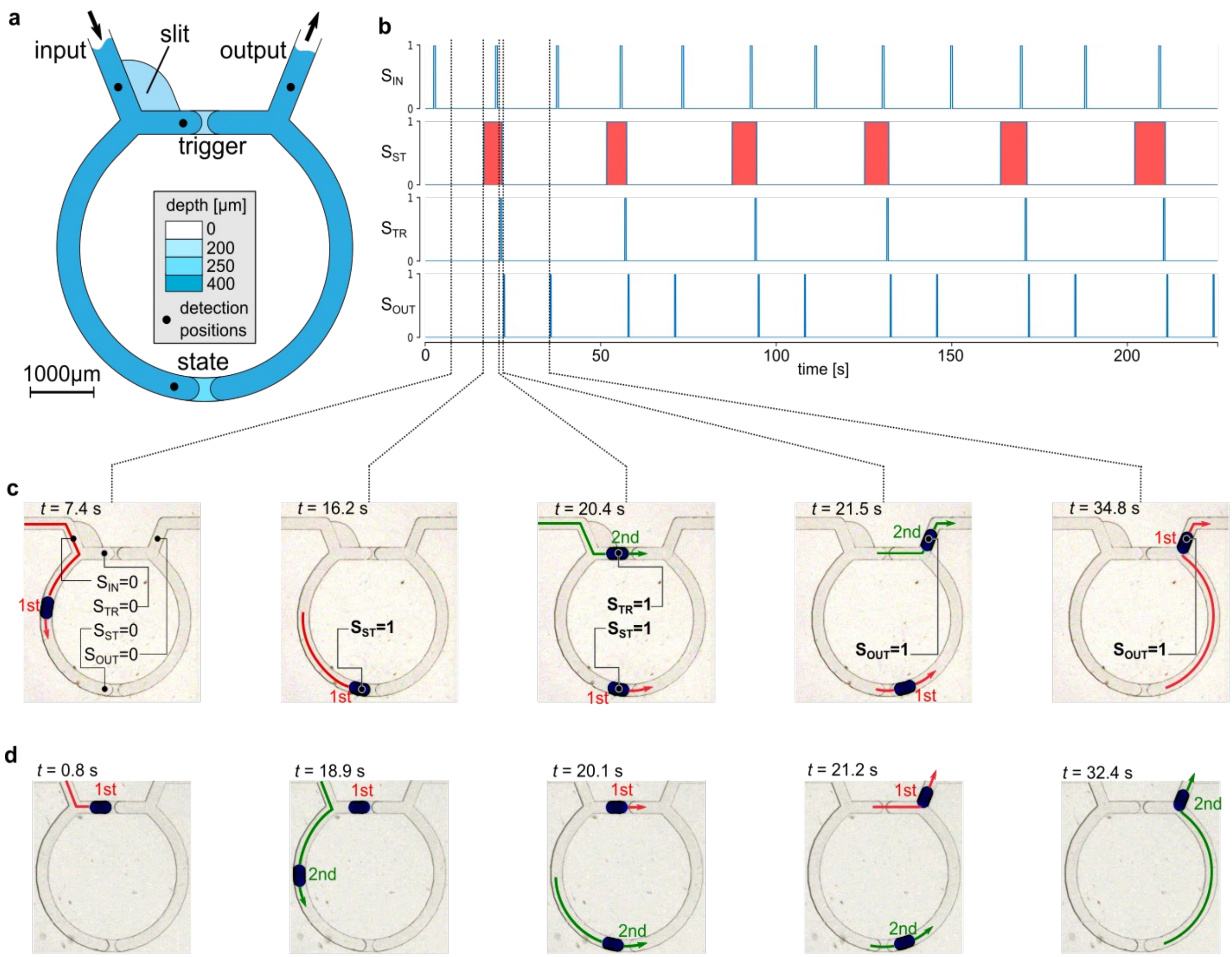
Supplementary Note 1
Bimodal loop with the modified junction

Here we present the bimodal loop with the modified junction to show how we can use the modification for the enhanced control on droplets motion.

In the bi-modal loop considered in the main article, the first droplet entered the branch of the smaller resistance (the shorter one). Here we use the modified junction with the slit to fix the custom direction of the first droplet regardless of the resistances of branches. Such a solution provides a higher degree of control, allowing to direct the first droplet to the long branch and swap functionalities between loop-branches.

Supplementary Fig. 1 shows the analysis of the motion of the pair of droplets through the modified junction. Thanks to the modification, unlike the case of the regular bimodal loop, the first droplet enters the long branch (compare Supplementary Fig. 1c and Supplementary Fig. 1d), where it stops at the obstacle setting the excited state. As a result, the functionalities of obstacles have been swapped: trigger-obstacle is placed now in the short branch while the state-obstacle in the long branch.



Supplementary Fig. 1 The bimodal loop with a slit in the input junction. **a** schematic view of the loop. Time diagram of the droplet-presence-signals. **c** experimental micrographs - the flow of two consecutive droplets through the bimodal loop with a modified junction, and **d** – micrographs from the regular junction for the reference.

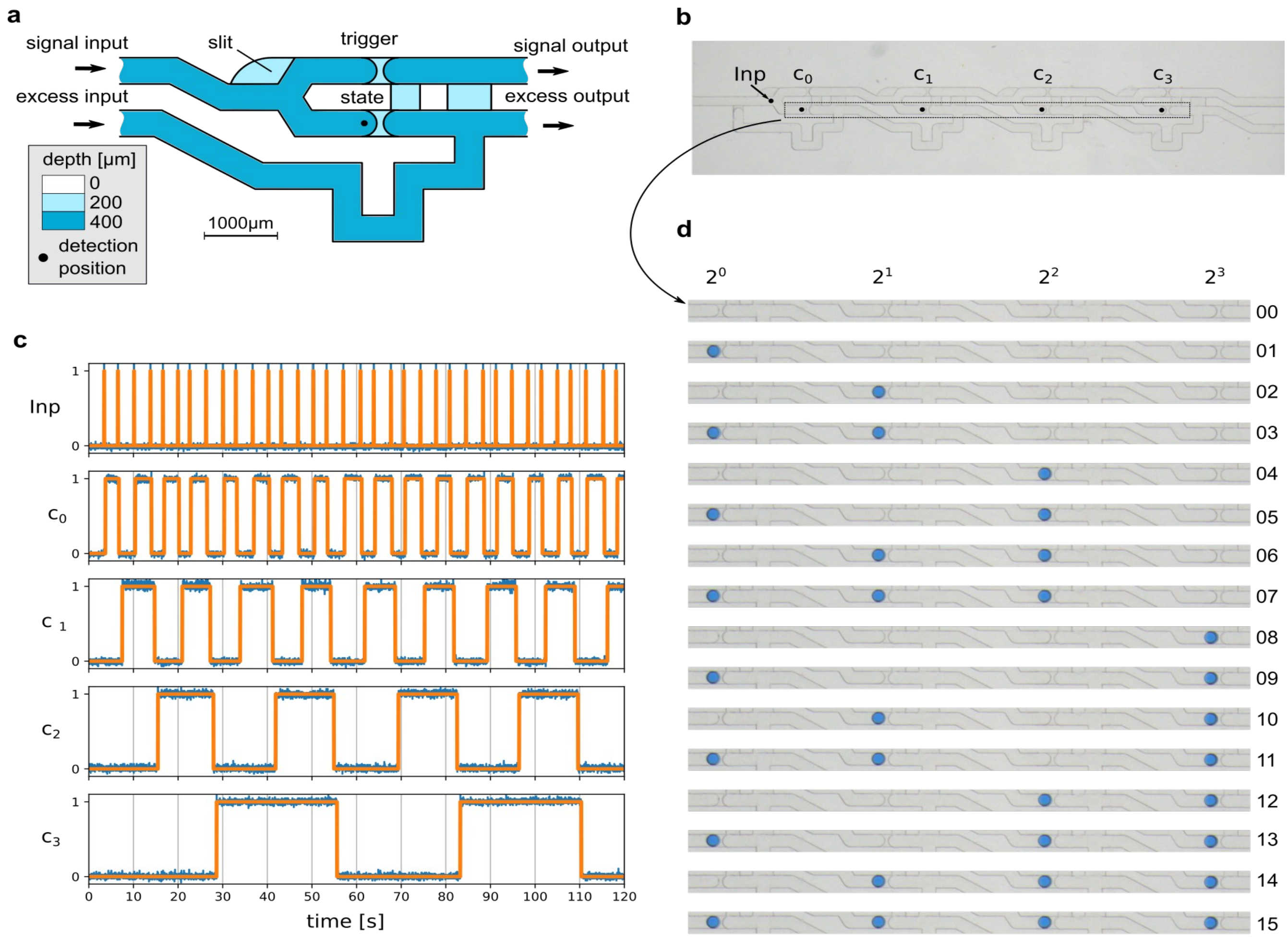
As we showed in this example, a modified junction provides more flexibility for encoding the motion of droplets in a parallel system of channels.

The additional important implication is that in such a modified loop, the first droplet is released after the second one. The change of droplets order occurs because the trigger-obstacle lies closer to the outlet than the state-obstacle and, additionally, the flow-through the shorter branch is faster. Such a mechanism can be used for the realization of permutations, rearranging the order of droplet in the sequence.

Supplementary Note 2
Binary counter

We integrated serially four bimodal loops with a slit (see Supplementary Note 1) to obtain a binary counter. Optimizing the device for the increase of its operational speed, we reduced the lengths of both the trigger- and the state-branch in every single counting-module (see Supplementary Fig. 2a). Additionally, similarly to the design of the 3-fold decimal counter (see the Main Article and Supplementary Note 3), we augmented the bimodal loop with two separate outlets: the excess-output and the signal-output channels, respectively (see Supplementary Fig. 2a). Two corresponding inlets ensure the compatibility for connections of subsequent units. Such a construction ensures that after resetting the state in the bimodal loop, only one of two emitted droplets enters the signal-inlet of the following counter. The other droplet is directed to the excess collector channel. We integrated four such modules obtaining a 4-fold binary counter (see Supplementary Fig. 2b). In such a system, the state positions of 4 counter modules form the four-bits binary display (which assumes values from 0 to 15 – see Fig. 2d), where the result of the droplets counting can be readout. The experiments showed that the binary counter works without errors, correctly processing every 16 droplets (Fig. 2d and Supplementary Movie 5).

Unlike in the other systems presented in the paper, in the case of the binary counter we used standard T-junction droplet generator[1].

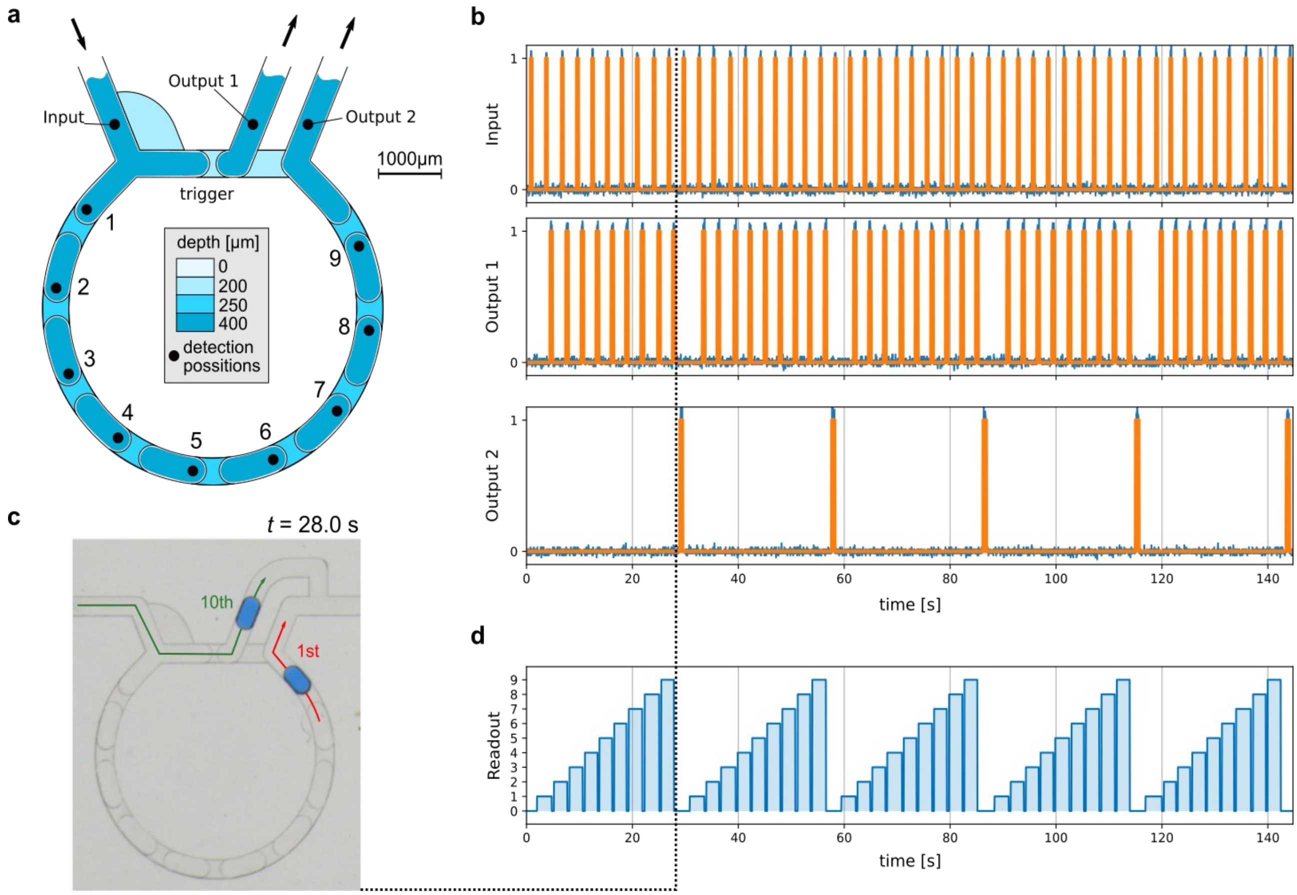


Supplementary Fig. 2 The 4-fold cascade binary counter. **a** schematic top view of the single counting-module. **b** photo of the whole device (taken before the start of droplets generation). Black dots – detection points for the droplet-presence-signals. Inp – the signal from droplets entering the system. C_0, C_1, C_2, C_3 – state-signals from each counting module. The black frame indicates the part of the device comprising of all state positions and can be seen as a binary 'display'. **c** time diagrams for all measured signals. The blue noisy lines – the brightness of pixels at the measurement position as a function of time (normalized and inverted so that the high value corresponds to the presence of a droplet). Orange lines – the digital droplet-presence-signals obtained by thresholding of the brightness signals. We can see that each subsequent module divides the incoming frequency by 2. This frequency division is well known in electronic binary counters. **d** experimental micrographs of the 'display' taken after introducing subsequent droplets. The number of introduced droplets is shown in right column. The readout from the display equals $\sum_{i=0}^{3} c_i \cdot 2^i$ where c_i assumes value 1 for the presence of a droplet at *i*-th position or 0 for its absence. So the 'display' accurately indicates the result of the droplets counting (see Supplementary Movie 5).

Supplementary Note 3
Decimal counter with two outlets

In order to adapt the decimal counter loop to the cascade-integration, we augmented it with two outputs. This solution enables the separation of trigger-droplets from the droplets leaving the state branch. In such a way, 10^th^ droplet, and generally the last droplet per every ten droplets, is directed to separate output and is used as a signal informing about the counting of 10 droplets (see Fig. 3). The additional slit connecting the outputs allows for the flow of continuous liquid while ensures separation of the routs of droplets outgoing from opposite branches of the loop. That element reduces the pressure difference between outlets ensuring similar hydrodynamic conditions as in the case of the decimal loop with a single output.

As we showed in the Main Article, the decimal counter with two outlets allows for effective cascade integration of multiple decimal counters.



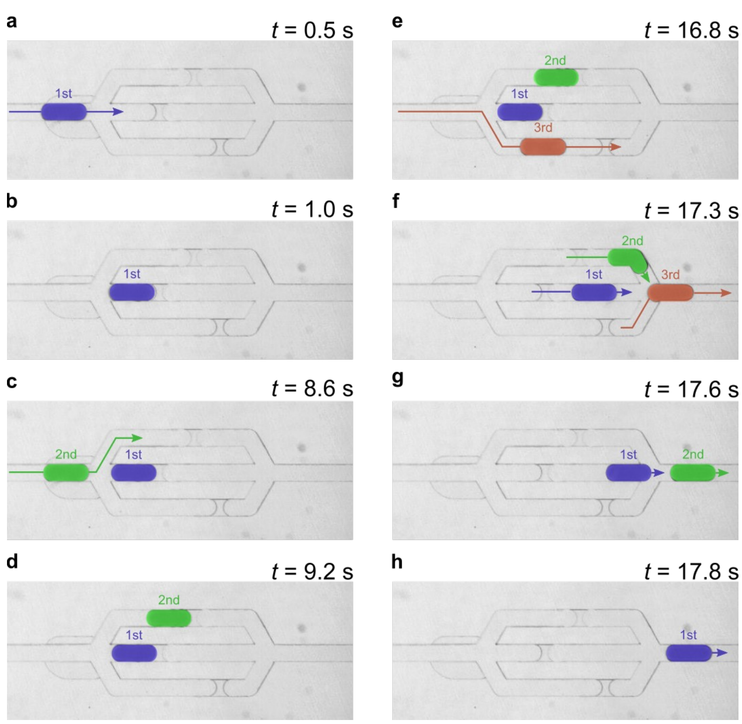
Supplementary Fig. 3 The decimal counter with two outputs. **a** the schematic top view of the counter with a trigger-barrier in the short branch and with 9 state-barriers in the longer branch. Both Output 1 and Output 2 are connected by the slit, which on the one hand allows for the flow of continuous phase and so that reduces the pressures difference, on the other hand, separates the motion of droplets. **b** experimental micrograph taken after the inflow of the 10^th^ droplet. The first droplet is released from the loop and enters the Output 2, while the 10^th^ droplet travels through the Output 1. **c** signals of the presence of a droplet at measurement positions Input, Output 1 and Output 2 as indicated in **a**. The blue noisy lines – the brightness of pixels at the measurement positions as a function of time (normalized and inverted so that the high value corresponds to the presence of a droplet). Orange lines – the digital droplet-presence-signals obtained by thresholding of the brightness signals. **d** the readout obtained by analysis of the presence of droplets in state positions (see the analysis of the decimal counter in the Main Article).

Supplementary Note 4
3 – way

Here we present an additional example of the 3-way device to show that further modifications of the base elements can yield new functionalities. The device consists of a 3-way junction dividing the input channel into three parallel channels, each of them equipped with one obstacle (Supplementary Fig. 4 and Supplementary Movie 8). The construction of a 3-way junction ensures, that 3 subsequent droplets are distributed between outlets in a specific way - the first droplet to the middle channel (Supplementary Fig. 4b), the second one to the top channel (Supplementary Fig. 4d), and the third one to the bottom branch (Supplementary Fig. 4e). Two first droplets are immobilized in the structure while the third one blocks the last bypass-channel and triggers the release of all droplets (Supplementary Fig. 4e-h). Hence we obtain 3 states - Besides the ground state (without any droplet trapped in the device), there are two excited states with immobilized droplets. The first excited state with the first droplet in the middle channel (Supplementary Fig. 4b) and the second excited state - with the additional second droplet immobilized in the top channel (Supplementary Fig. 4d).

As all droplets are released from the obstacles simultaneous, the appropriate arrangement of obstacles allows for setting different delays between droplets, thus, the encoding of custom order of droplets leaving the device. In this case, we set the positions of obstacles in such a way, that releases the third droplet as the first one, while the droplet being previously the first one is moved to the third position (Supplementary Fig. 4f-h).

This example shows that modifications of such base geometrical components as slits, obstacles, and junctions allow for achieving novel functionalities, which have the potential for the encoding variety of algorithms on droplets in microfluidic channels.



Supplementary Fig. 4 The system of three parallel channels with 3-way junction – experimental observations. We add artificial colors to droplets for the visualization of the transformation of the initial sequence of droplets (see Supplementary Movie 8).

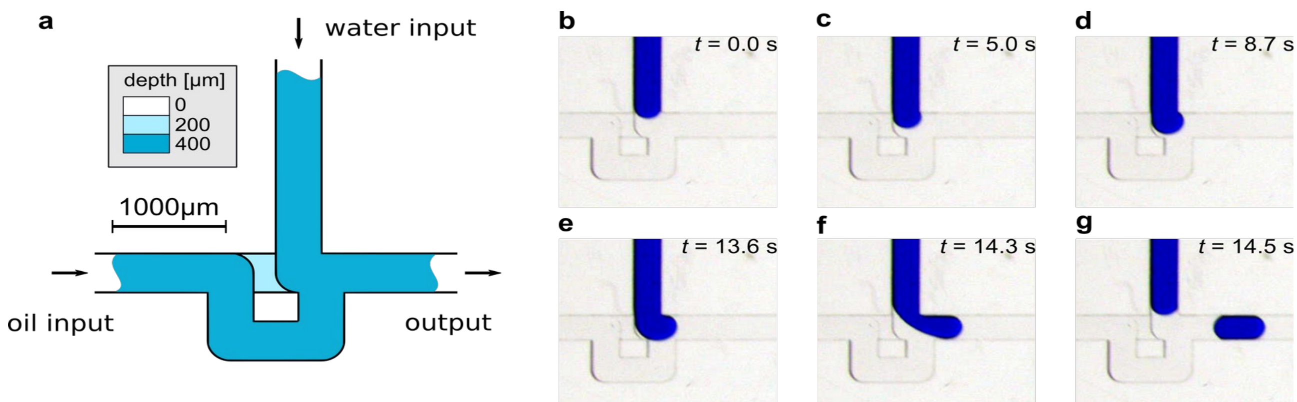
Supplementary Note 5
Droplet Generator Unit

Supplementary Fig. 5 shows the Droplet Generation Unit we used for the fabrication of droplets. It is a modified version of the previous block-and-brake type generator[2]. Such a construction, unlike the regular T-junction generator, produces droplets of similar size regardless of flows of both phases[1].

During the first trials with droplet-logic, we realized that the optimal size of droplets in most cases is about double the channel width. The other crucial parameters were the speed of the flow and frequency of generated droplets. The speed of the flow on the one hand, cannot be too fast (it cannot generate pressure drop exceeding the break-through pressure), but on the other hand, it should not be too slow to ensure a reasonable time of experiments. The frequency of the generation of droplets must be sufficiently low to avoid the interference of numerous droplets at once in the microfluidic units.

As known, both the frequency and the length of droplets generated in the regular T-junction depend on the flows[1]. Hence, those parameters cannot be adjusted independently. In the case of our experiments, droplets would be too small if produced in regular T-junction.

The solution we used provided more flexibility to the control over the crucial parameters, which could be effectively adjusted, thus, ensuring the robust performance of the investigated systems.



Supplementary Fig. 5 Droplet generation unit used in the work. **a** schematic top view. **b**-**g** micrographs showing the complete sequence of the generation of a single droplet.

Supplementary references

1. Korczyk, P. M. *et al.* Accounting for corner flow unifies the understanding of droplet formation in microfluidic channels. *Nat. Commun.* **10**, 2528 (2019).
2. van Steijn, V. *et al.* Block-and-break generation of microdroplets with fixed volume. *Biomicrofluidics* **7**, 024108-024108–8 (2013).