Supporting Information

Evaporation-Driven Water-in-Water Droplet Formation

Byeong-Ui Moon,*,† Lidija Malic,† Keith Morton,† Morteza Jeyhani,‡,§,‖ Abdelrahman Elmanzalawy,† Scott S. H. Tsai,‡,§,‖ and Teodor Veres,*,†

† Life Sciences Division, National Research Council of Canada, Boucherville, Quebec, Canada
‡ Departments of Mechanical and Industrial Engineering, Ryerson University, Toronto, Canada
§ Keenan Research Centre for Biomedical Science, St. Michael's Hospital, Toronto, Canada
‖ Institute for Biomedical Engineering, Science and Technology (iBEST), a partnership between Ryerson University and St. Michael's Hospital, Toronto, Canada

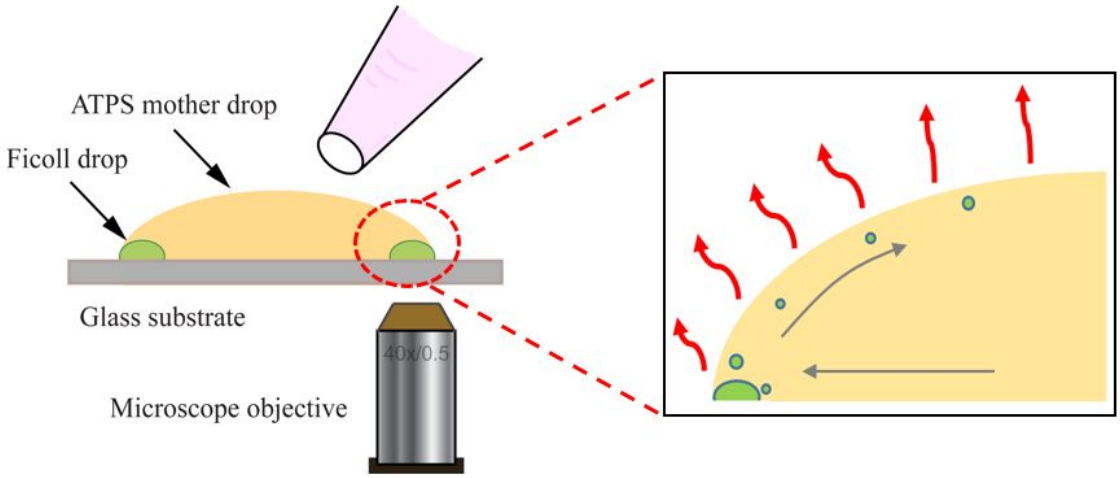

Figure S1. Experimental set-up for W/W sessile droplet formation. The zoomed-in image shows a schematic drawing of ATPS secondary droplet formation at the contact line.

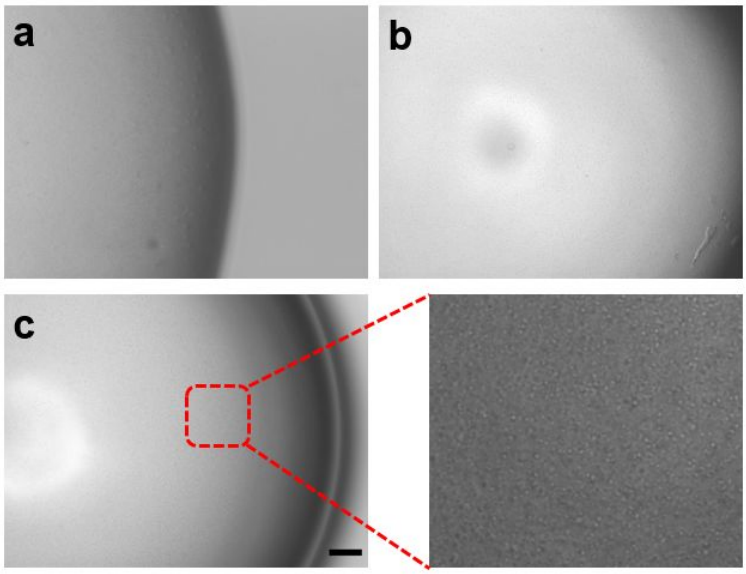

Figure S2. Evaporation based ATPS droplet formation on different materials: (a) TPE (b) PDMS and (c) Teflon. Dashed line indicates a zoomed-in image. We experimentally observe that the ATPS secondary droplets are not coalesced and trapped along the contact line on the high contact angle substrates of TPE, PDMS and Teflon. Instead, small ATPS droplets are formed throughout the bulk mother drop near the water-air interface. Scale bar represents 50 μm.

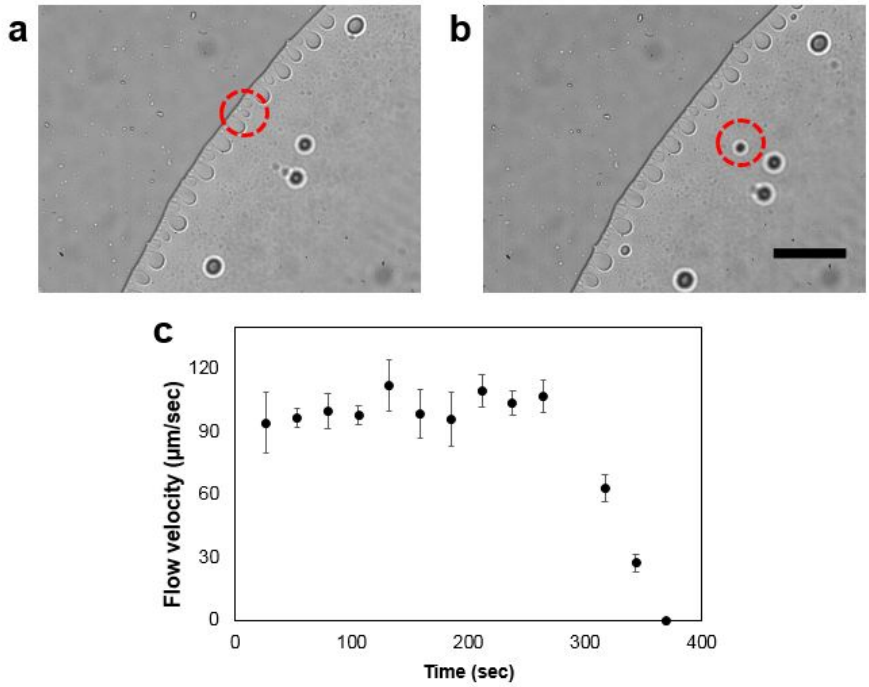

Figure S3. Recirculating flow velocity measurement. Small ATPS satellite droplets are used as tracing markers to measure recirculating flow velocity at the contact line. (a) A satellite secondary droplet is positioned at the edge of the mother droplet as shown with a dashed red circle ($t = 0$). (b) The same secondary droplet leaves the contact line and moves towards the center of the bulk solution. The dashed circle shows the same droplet. Scale bar represents 50 μm. (c) A graph of the measured flow velocity as a function of time. We measure a 0.5 μL mother droplet to analyze the velocity change of the satellite droplets. The resulting data shows that the flow velocity is constant until water evaporation reaches a critical point – this suggests that the contact angle is approximately constant until the critical point.

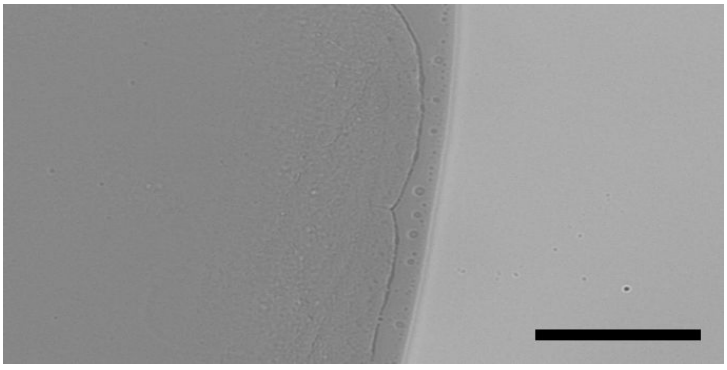

Figure S4. ATPS droplet morphology change at contact line with Collagen Type 1. We prepare the collagen at 400 times dilution in the ATPS. We observe that the presence of collagen significantly changes ATPS-W/W droplet formation. Scale bar represents 50 μm.

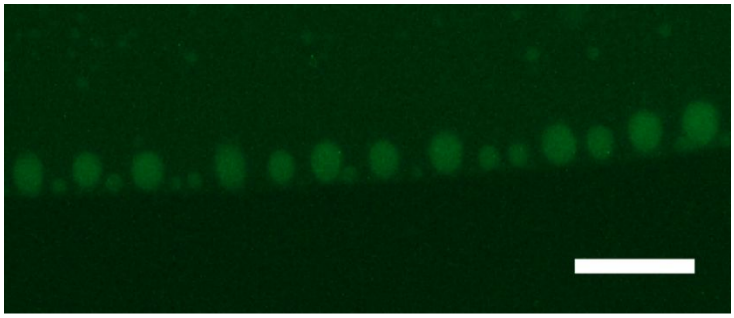

Figure S5. Fluorescence image showing the boundary of the 1 µL of a main ATPS droplet, and secondary droplets. The image shows that the fluorescently-labeled BSA is partitioning into to the secondary droplets. The images are taken 50 sec after ATPS main droplet deposition. Scale bar shows 30 µm.

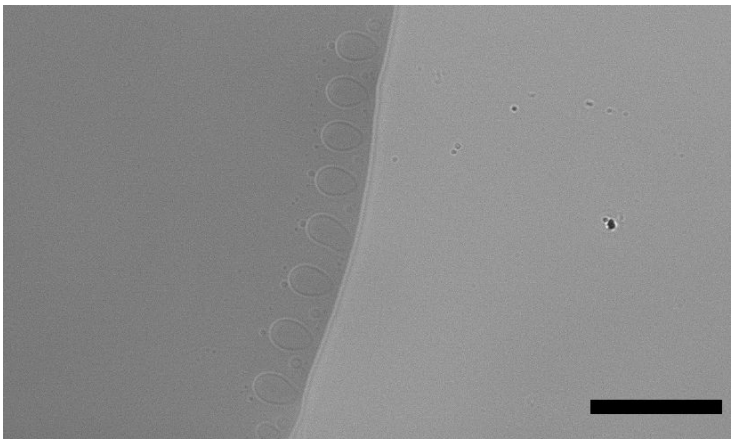

Figure S6. ATPS secondary droplets at contact line using a PEG-dextran system. We prepare a mixture of PEG 5% (w/v, M_w, 10 kDa) and dextran 0.8% (w/v, M_w, 500 kDa). A 1 µL of the mother drop is deposited on the glass slide. Scale bar shows 50 µm.

Supplementary Information Movie 1:

Video shows secondary droplet formation at the contact line of the main sessile droplet. Initially, evaporation at the contact line drives phase separation and the generation of sub-micron size secondary droplets. The sub-micron size secondary droplets coalesce to form larger secondary droplets, which are aligned and pinned along the contact line. Some secondary droplets escape to the bulk of the main sessile droplet as evaporation continues. Here, the main sessile droplet volume is 0.5 μL. This video is recorded at a frame rate of 30 fps, and played back at 10× the actual speed.