Studying the Interfacial Dynamics of Miscible Systems with Microfluidics

John Schwalbe
Chemical and Biomolecular Engineering, Cornell University

NNIN REU Site: NanoTech User Facility, University of Washington, Seattle, WA
NNIN REU Principal Investigator: Dr. Amy Q. Shen, Mechanical Engineering, University of Washington
NNIN REU Mentor: Shilpa Beesabathuni, Mechanical Engineering, University of Washington
Contact: jas699@cornell.edu, amyshen@uw.edu, shilpanb@uw.edu

Abstract:

Various microfluidic features are considered for the study of the interfacial dynamics of miscible systems. We take advantage of upper critical solubility temperature (UCST) behavior of a given miscible system. Fully realizing the potential of our experimental plan will require integrating multiple features in one microdevice. The challenges that arise include varying length scales between features, temperature control, and complex control systems.

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Introduction:

We are interested in the dynamics of interfaces between two miscible phases. While interfaces between equilibrated phases are thermodynamically stable, interfaces between miscible phases are inherently transient. These interfaces exhibit interesting mass transfer properties and effective interfacial tension [1]. Understanding these dynamics is important to modeling various industrial and biological processes. Enhanced oil extraction and mucus spreading in the lungs are two examples. Droplet microfluidics is an ideal platform to study interfaces because the small length scales allow for very controlled velocity profiles.

The devices were fabricated using soft lithography [2], so they could be prototyped quickly.

To observe the dynamics of miscible interfaces, it was important that we have a known initial condition. To do this, we took advantage of the temperature dependence of solubility. Below a critical temperature, the UCST, two immiscible phases form. Above it, the two components are miscible in all proportions. To exploit this experimentally, droplets were formed below the UCST, denoted as phases I and II in Figure 1. Next, a single droplet was trapped above a heater. The droplet was then heated above the UCST, shown with dot-dash lines in Figure 1. Finally, thermodynamics took over and the droplet began to dissolve, shown with dashed lines in Figure 1. Isobutyric acid (IBA) and water have a reasonable UCST, for example. But, there are other interesting systems that could be explored with this technique including ionic liquids and thermoresponsive polymers. Implementing this plan was difficult. Droplet formation, trapping, and heating on a single chip were all challenging.

Experimental Procedure:

We considered a variety of device features. For droplet formation, we used T-junctions and flow-focusers [3]. In a T-junction, a channel carrying the dispersed phase entered a channel carrying the continuous phase at 90°. Depending on the relative sizes of the channels and the flow rates, T-junctions

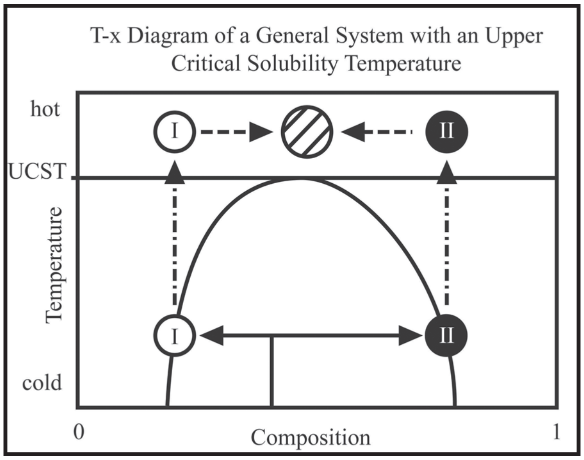

Figure 1: Process schematic.

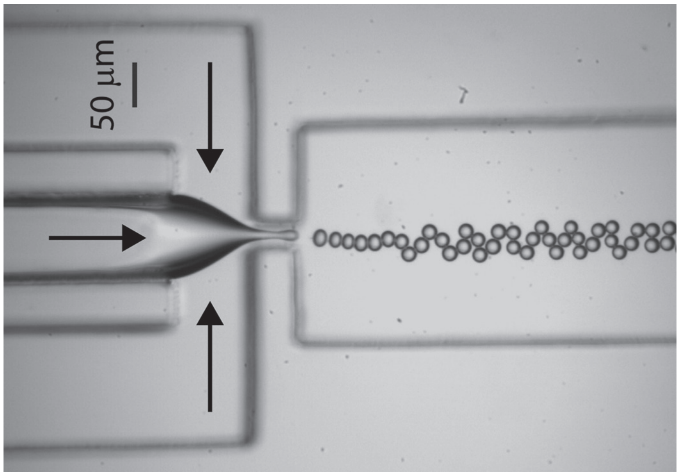

Figure 2: A flow focuser producing droplets of water in oil.

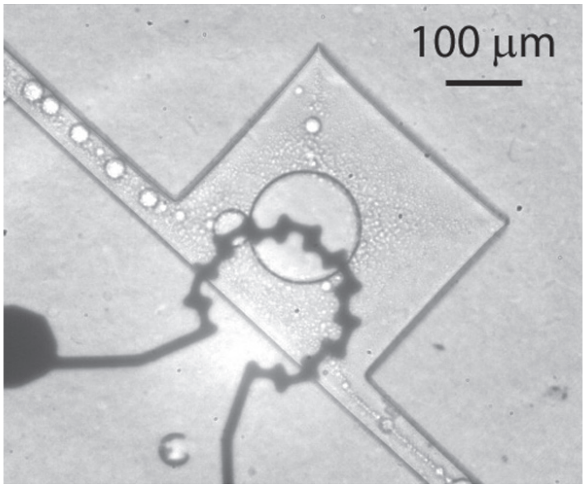

Figure 3: An IBA/water emulsion in a vortex trap.

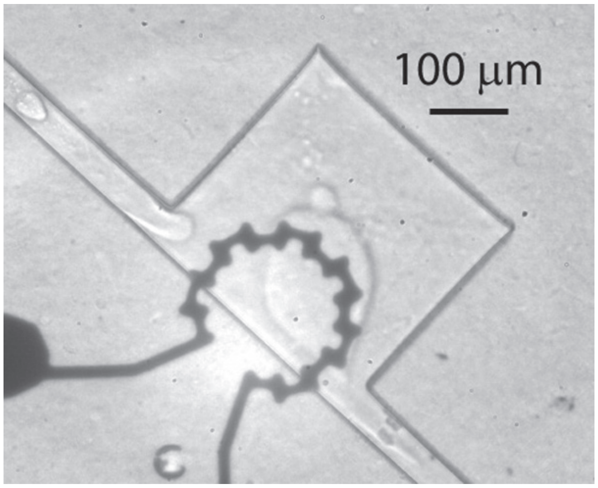

Figure 4: An IBA/water mixture shortly after it was heated above the UCST.

could generate droplets of varying sizes in either a dripping or a jetting mode. In a flow-focuser, the dispersed phase was introduced into the middle of the carrier phase. The resulting two-phase flow was then forced through a constriction. The resulting viscous and capillary forces caused the dispersed phase to break into droplets, as seen in Figure 2 [3].

Trapping the droplet was the next challenge. Cross-slot chambers were a powerful option. They consisted of two channels crossing at 90°. In use, the channels acted as two inlets and two outlets. This created a stagnation point in the middle of the chamber where a droplet could be trapped. When used in conjunction with a feedback control system, droplets could be trapped indefinitely [4]. Another attractive feature was that the droplet deformed in the velocity field, and the effective surface tension could be deduced from the shape of the droplet [5].

A hydrodynamically simpler option was a vortex trap. An emulsion in a vortex trap can be seen in Figure 3. At high enough flow rates, a vortex will form in the expansion. Vortex traps have been used to trap cells [6]. But, it is an open question whether or not they can also be used to trap droplets.

Finally, temperature control was vital to this experiment. Traditional thermal stages cannot ramp the temperature fast enough. We chose resistance heaters made with a lift-off procedure instead. The geometry of the heater could be tweaked to get the desired performance. But, electrolytic gas production was observed in some designs above a threshold voltage. So, it is a constrained optimization problem.

Results and Discussion:

A fully integrated device remains elusive. The most successful feature was the heater. Figure 4 shows the result of heating an emulsion above the UCST of the system, in this case IBA and water. The blurring of the interface clearly shows the dissolution process and the shape of the droplet reflects the effective interfacial tension. However, the variations along the circumference of the droplet show the need for a more homogenous temperature increase. This image also highlights the need for an integrated device. The small droplets surrounding the central droplet are undesirable. They are there because an emulsion was pumped into the trap rather than the uniform droplets that a flow-focuser or T-junction would create.

But, as Figure 3 shows, the vortex traps have potential. Their simplicity is an inherent advantage. They require no complicated feedback control. However, viscous droplets cannot cross streamlines in the sense that particles can. So far vortex traps have worked with emulsions, but this may pose a problem with the orderly, monodisperse droplets produced by a T-Junction or a flow-focuser.

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