Performance of Different “Lab-on-Chip” Geometries for Making Double Emulsions to Form Polystyrene Shells

Polystyrene in fluorobenzene

Polyacrylic acid in water

Water

Polystyrene in fluorobenzene

Polyacrylic acid in water

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22nd Target Fabrication Meeting
Las Vegas, NV
12–16 March 2017
“Focus-flow” microfluidic geometries were used to successfully form a polystyrene and oil-water-oil double emulsion

- Adding a surfactant in a T-junction device increases the range of fluid velocities that form single emulsions
  - $Ca = 0.00006$ to $0.005$, $\varphi = 0.1$ to 8 for oil/water droplets
  - $Ca = 0.0007$ to 0.13, $\varphi = 0.03$ to 5 for water/oil droplets

- Forming double emulsions using T-junctions in series with a single material is not possible; however, a double-cross geometry formed oil-water-oil double emulsions at high velocity and a low interfacial tension (IFT) value ($0.8 \text{ mN/m}$)

- A focused-flow device formed polystyrene double emulsions over a narrow range of flow-rate ratios ($\varphi = 0.03$ to 0.9)
  - shell wall diam: 2.3 to 4.3 mm; thickness: 30 to 160 $\mu$m
  - future work is to see if reducing the IFT reduces the vacuole content

Ca: capillary number
Hydrodynamic forces that control the droplet’s size depend on the fluids’ velocity ($v$), viscosity ($\mu$), volumetric flow rate ($Q$), and interfacial tension ($\gamma$)

Three forces form the droplet in the desirable “squeezing” regime:

1. Shear—when $v_c \gg v_d$ and/or $\mu$ is large
2. $\Delta$Pressure ($P_U - P_D$)—when $\phi$ is high (droplet fills the channel) and $\gamma_{liq-liq}$ is low
3. Laplace pressure—when $\phi$ is low (droplet is smaller than the channel)

Performance is characterized by dimensionless numbers: $Ca = (v_c \cdot \mu_c)/\gamma$ and $\phi = Q_d/Q_c$
We determined the fluid properties ($\gamma, \mu$) and flow parameters ($V, \varphi$) that are needed to form ICF-size droplets in a single T-junction.

**First T:** oil-in-(water + surfactant) droplet  
(*establishes the inner diameter of the target*)

**Second T:** water-in-(oil + surfactant) droplet  
(*establishes the targets’ wall thickness*)

- High $\varphi$ ($Q_d/Q_c$) allows the oil phase to fill the water channel

- The “squeezing regime” exists when $v_d$ is low and $v_c \gg v_d$
The size of the droplet increases with the flow-rate ratio \((\phi = Q_d/Q_c)\)

- \(Q_d\) and \(Q_c\) are the volumetric flow rates of the disperse and continuous phases
- Acrylic device

![Graph showing the relationship between droplet length and flow-rate ratio for Oil-in-(water + T80) and Water-in-(oil + S80) systems.](image)
The size of the droplet decreases with increasing capillary number

- \( \text{Ca} = \left( \frac{v_c \cdot \mu_c}{\gamma} \right) \), combines the competing effects of the fluid velocity, viscosity, and interfacial energy

- Acrylic device

*Each curve is the squeezing-dripping transition for the maximum \( v_d \) value*
Changing the *device material* to glass changes the chemistries available for making targets and provides a wider range of parameters for making emulsions.

- Fluorobenzene-in-water emulsion to make polystyrene shells

The higher surface energy and higher interfacial surface tension allows emulsions to form at higher fluid velocities than is possible with an acrylic material.
Changing the device material altered the liquid–solid surface energy, allowing water-oil and oil-water emulsions to be formed without needing a surfactant.

**First T: water-oil emulsion**

\[ \mu \sim 26 \text{ centipoise} \]

- Acrylic (IFT = 3.3 mN/m; \( \gamma = 41 \text{ mN/m} \))
- PVC (IFT = 4.8 mN/m; \( \gamma = 42 \text{ mN/m} \))
- Acrylic (IFT = 5 mN/m; \( \gamma = 41 \text{ mN/m} \))

**Second T: oil-water emulsion**

\[ \mu \sim 1 \text{ centipoise} \]

- Glass (IFT = 30 mN/m; \( \gamma = 83 \text{ mN/m} \))
- Glass (IFT = 25 mN/m; \( \gamma = 83 \text{ mN/m} \))
- Acrylic (IFT = 4.8 mN/m; \( \gamma = 41 \text{ mN/m} \))
- Glass (IFT = 48 mN/m; \( \gamma = 83 \text{ mN/m} \))
“Double-Cross” Junction

Positioning the channels to form a “focused-flow” geometry formed oil-water-oil emulsions at very low IFT values (0.8 mN/m)

Oil droplet forming inside a water stream at first cross

<table>
<thead>
<tr>
<th>First cross</th>
<th>Secnd cross</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water + 1% tween</td>
<td>Oil + 0.2% S80</td>
</tr>
</tbody>
</table>

Oil

Water + 1% tween

Oil-in-water droplet formed inside an oil stream at the second cross

<table>
<thead>
<tr>
<th>Water + 1% tween</th>
<th>Oil + 0.2% span</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water + 1% T80</td>
<td>Oil + 0.2% span</td>
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</tbody>
</table>

Oil

Water + 1% tween
A Teflon-coated glass device produced OMEGA-size emulsions for making polystyrene targets.

18 wt% polystyrene in fluorobenzene

0.05 wt% polyacrylic acid in water

Water

18 wt% polystyrene in fluorobenzene

0.05 wt% polyacrylic acid in water

Oil
There is a narrow range of flow velocities where double emulsions form 100% of the time

- The flow rate ratio at the second cross primarily controls the amount of fluid in both phases of the emulsion (inner and outer diameter of the target)
- The target diameter was controlled over a range from $2.3\pm0.07$ to $4.3\pm0.23$ mm with a wall thickness between 30 and 160 $\mu$m (for a 2.4-mm channel diam)
There is a narrow range of flow velocities where double emulsions form 100% of the time (continued)

100% double emulsion formation

\( V_c \) at first cross = 2 mm/s

\( V_c \) at second cross = 20 mm/s

\( V_c \) at first cross = 3 mm/s
Summary/Conclusions

“Focus-flow” microfluidic geometries were used to successfully form a polystyrene and oil-water-oil double emulsion

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\( \text{Ca} \): capillary number
The size of the droplet is determined primarily by the velocity of the continuous phase once that velocity is above 5 mm/s.

- Water-in-(oil + surfactant) emulsion ($\gamma = 3.3$ mN/m)