Computational Fluid Dynamics Model of Viscous Droplet Breakup

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Many high-viscosity mixing / blending processes result in the formation of droplets of one material inside a matrix of a second material. The final application often requires that the droplet size be reduced to an acceptable range - properties or characteristics may depend on DSD, and reactions may be induced in one of the two phases. There may be further process requirements such as energy efficiency (low \( \Delta P \)), low capital cost, and high throughput.
**Historic Droplet Breakup Literature:**

- Breakup is impossible at high droplet to matrix viscosity ratios in simple shear flows

- However, breakup is possible in elongational flows

**Capillary Number:**

\[ Ca = \frac{\mu_m G a}{\sigma} \]

- \( \mu_m \): matrix viscosity
- \( G \): deformation rate (shear or extensional)
- \( a \): droplet radius
- \( \sigma \): interfacial tension

*Critical Capillary number versus viscosity ratio \( p = \eta_d/\eta_c \), in simple shear and in elongational flow (after Grace 1971).*
Goals

- Develop a validated model that can predict viscous droplet breakup in complex flow geometries
- Provide insights into the fundamental breakup mechanisms
- Design optimal flow geometries for effective droplet breakup with efficient energy input
Modeling Methodology

### COMPUTATIONAL FLUID DYNAMICS SET-UP

- Code: FLUENT
- Interface Tracking: “Volume of Fluids” Method (VOF)
- Laminar / Transient
- Geometry: SINGLE droplet flowing past obstacles
  - Symmetry planes – captures effect of droplet number density
  - Tight mesh to capture interface

### PROPERTIES

- Rheology: Carreau model
  - Shear Thinning
    - Droplet has higher viscosity than the matrix over the shear rates of interest
- Density (droplet and matrix)
- Interfacial tension (*No observed effect*)
2D Modeling

- **Vary**
  - Obstacle Layout
  - Initial droplet radius (circular / cylindrical)
  - DROPLET Number density
  - Mass flux ($V_{in}$)
  - Rheology
  - Droplet – Obstacle Alignment
Follow droplet interface with time as it flows past obstacles

Colors represent volume fraction of droplet phase
Multiple Mechanisms

1) "Stick-and-Pull":
Higher droplet-to-matrix viscosity ratios cause the droplet to "stick" to the obstacles more tightly so there is more stretching as it is pulled off.

2) "Wire-Spreading":
Lower droplet to matrix viscosity ratios are more easily spread apart by the obstacles and less likely to flow back together.

"Stick-and-Stay"
Compare Breakup at Different Flow Rates
(different shear rates $\rightarrow$ different viscosity ratios)

Inlet velocity: $V_1 < V_2 < V_3 < V_4$

Experimental Observation: break-up is more efficient at lower and higher rates
**Compare Two Different Droplet/Matrix Systems**

<table>
<thead>
<tr>
<th>System A</th>
<th>System B</th>
</tr>
</thead>
<tbody>
<tr>
<td>Same Flow Rate</td>
<td></td>
</tr>
</tbody>
</table>

**Similar Droplet-to-Matrix Viscosity Ratios**
2D Model Validation

- Compare the droplet breakup predicted by the model with experimental data:
  - Experimental Data is in the form of droplet size distributions (DSD) for both the upstream and downstream mixtures
  - Model needs to account for initial DSD and determine final DSD
    - use ImageJ software

- Fluent Plot for droplet volume fraction from 0.5 to 1

- Use obstacle diameter to calibrate

- Equivalent ellipses

- Obtain Predicted DSD and Aspect Ratios
<table>
<thead>
<tr>
<th>Time 1</th>
<th>Time 2</th>
<th>Time 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>•A</td>
<td>•B</td>
<td>•C</td>
</tr>
</tbody>
</table>

**Effect of Obstacle Position**

Identified 3rd MECHANISM: “split and stretch”
Overall DSD Methodology

- Run the CFD droplet breakup simulation for
  - 5 different obstacle alignment cases (A-E)
  - 8 different starting droplet diameters (80-1200 µm)
- Use the Image analysis tool to get the DSD for each case
- Account for possible elongational breakage
  - Droplet Aspect Ratio > “Break-up Aspect Ratio” (BAR)
    → droplet will break up into the minimal number of equal parts so that
    the aspect ratio of each part is less than the BAR value
- Determine the overall DSD for each initial droplet size using
  probabilities of droplet-obstacle contact for each alignment
- Determine the combined DSD for all of the droplet sizes
  using the experimental values for the number of starting
  droplets in each size bin
- Compare to the experimental DSD values
**Droplet-Wire Contact Probabilities**

Determine probabilities for droplet “hitting” large wires in the two rows

- Define “hitting” to mean the droplet edge crosses the wire center
- Event R = droplet “hits” first wire
- Event S = droplet “hits” last wire
- Assume R and S are independent
- \( P(R) = P(S) = \frac{G}{L} \)
  
  where \( G \) = droplet diameter
  
  \( L \) = distance between wire centers

- B: \( P(R \cap S) = P(R) \times P(S) \)
- C: \( P(R \cap S^C) = P(R) \times (1 - P(S)) \)
- D: \( P(R^C \cap S) = (1 - P(R)) \times P(S) \)
- A: \( P(R^C \cap S^C) = (1 - P(R)) \times (1 - P(S)) \)

- Allow cases A and E to be weighted evenly
- Note: Probability of droplet hitting smaller wires is always 100%

<table>
<thead>
<tr>
<th>Droplet Size</th>
<th>( P(R) = \frac{G}{L} ) %</th>
<th>B %</th>
<th>C and D %</th>
<th>A (split w/ E) %</th>
</tr>
</thead>
<tbody>
<tr>
<td>1200</td>
<td>100</td>
<td>60</td>
<td>20</td>
<td>0</td>
</tr>
<tr>
<td>700</td>
<td>55.12</td>
<td>30.38</td>
<td>24.74</td>
<td>20.14/2</td>
</tr>
<tr>
<td>550</td>
<td>43.31</td>
<td>18.76</td>
<td>24.55</td>
<td>32.14/2</td>
</tr>
<tr>
<td>450</td>
<td>35.43</td>
<td>12.56</td>
<td>22.88</td>
<td>41.69/2</td>
</tr>
<tr>
<td>350</td>
<td>27.56</td>
<td>7.60</td>
<td>19.96</td>
<td>52.48/2</td>
</tr>
<tr>
<td>250</td>
<td>19.69</td>
<td>3.88</td>
<td>15.81</td>
<td>64.50/2</td>
</tr>
<tr>
<td>150</td>
<td>11.81</td>
<td>1.40</td>
<td>10.42</td>
<td>77.77/2</td>
</tr>
<tr>
<td>78</td>
<td>6.14</td>
<td>0.38</td>
<td>5.76</td>
<td>88.09/2</td>
</tr>
</tbody>
</table>
# Prediction vs. Experiment

## Droplet Breakup Simulation DSD Predictions

<table>
<thead>
<tr>
<th>Droplet Size Bins (um)</th>
<th>Ave.</th>
<th>Breakup Aspect Ratio (BAR)</th>
<th>Exp DSD</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td>7 to 54</td>
<td>30</td>
<td>21157.66</td>
<td><strong>18739.04</strong></td>
</tr>
<tr>
<td>55 to 100</td>
<td>78</td>
<td>13493.22</td>
<td>5578.46</td>
</tr>
<tr>
<td>101 to 200</td>
<td>150</td>
<td><strong>1525.10</strong></td>
<td>11814.66</td>
</tr>
<tr>
<td>201 to 300</td>
<td>250</td>
<td>33.84</td>
<td><strong>43.83</strong></td>
</tr>
<tr>
<td>301 to 400</td>
<td>350</td>
<td>0.00</td>
<td><strong>33.84</strong></td>
</tr>
<tr>
<td>401 to 500</td>
<td>450</td>
<td>0.94</td>
<td>0.00</td>
</tr>
<tr>
<td>501 to 600</td>
<td>550</td>
<td>0.03</td>
<td>0.00</td>
</tr>
<tr>
<td>601 to 800</td>
<td>700</td>
<td>0.03</td>
<td>0.94</td>
</tr>
<tr>
<td>801 to 1600</td>
<td>1200</td>
<td>0.00</td>
<td>0.05</td>
</tr>
</tbody>
</table>
3D Modeling

hex mesh

tet mesh (around obstacles)

NOTE: Mesh is denser in the middle along the most likely droplet path
Potential Model Enhancements / Challenges

- Multiple starting droplets in single model
  - degree of interaction depends on droplet number density
  - requires large mesh and computational resources

- Miscibility / Diffusion effects
  - VOF method treats the phases as immiscible

- Elongational and/or viscoelastic viscosity models
  - Variations in Literature Observations concerning the Effect of Viscoelasticity on Droplet Breakup
  - We do not currently have a code that has both viscoelastic modeling capability and a VOF method
Viscoelasticity Effect - Literature Observations

➢ Trends:
  ▪ Droplet elasticity → stabilizing
  ▪ Matrix elasticity → destabilizing

➢ Break-up Mechanisms:
  ▪ stretching along flow axis
  ▪ erosion
  ▪ tip streaming
  ▪ elongation in vorticity direction

e.g., PS droplet in PE matrix in simple shear (Couette) flow*:
  • at low shear rates, viscous shearing forces dominate → drop elongates a small amount in the flow direction
  • as shear rates increase, normal stresses induce secondary flow perpendicular to flow direction → droplet takes on a diamond shape
  • at high shear rates, get competition between normal stresses and shear stresses → drops align perpendicular to flow direction
  • at high enough shear rates → droplet elongates and breaks along vorticity axis

*Frej Mighri, Michel Huneault; In Situ Visualization of Drop Deformation, Erosion, and Breakup in High Viscosity Ratio Polymeric Systems under High Shearing Stress Conditions; J. Applied Polymer Science, 100, 2006, pp 2582-91
Summary

- The model has provided insights into some of the mechanisms that can cause droplet breakup
  - Helps to explain some puzzling observed phenomena
- We have generated ideas for new obstacle geometries
  - Some have been implemented
- Although the 2D model results do not perfectly match the experimental data, they do seem to capture the trends
- The 3D model results provide further understanding of the complex interactions between the droplet and obstacles
- **Next Step:** Develop ability to evaluate the effect of viscoelasticity on the droplet breakup in complex geometries
  - Requires new modeling capabilities