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Oil-in-water emulsification via cross-junction microchannel at high throughput

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Abstract. Liquid-liquid dispersion process is experimentally investigated to manufacture oil-in-water emulsion working at high flowrates in the cross-slot type of microfluidics. Two types of the cross layouts, namely symmetric and asymmetric configuration, are compared via characterizing the droplet size and size distribution. Because of the higher shear stress and more intensified interaction, the symmetric geometry of the cross-slot is beneficial to produce less amounts of large droplets and dispersing the oil phase more uniformly at the same hydrodynamic conditions, especially in the low range of Re tested in this study. As the flowrates is enhanced, the disparity between them is diminished due to the fact that the instability inside the channel reaches a high level. The detrimental effect of the symmetric configuration is that energy required to burst the dispersed streams is relatively a little higher than with asymmetric one.

1. Introduction
Great amounts of work have been carried out to study different methods for emulsification, including agitating the mixture of two-phase liquids in a tank using impellers [1], static mixers [2] and high-pressure homogenizers [3]. Microfluidics, attracting plenty of focus in recent decades, is another paramount technical pathway to handle two-phase liquids dispersion issues. Basically, it is a droplet-based technology and well known for its superiority in manipulating droplet dimensions, so that the created emulsion from the microfluidics is closer to a mono-dispersed system. Three conventional types of configurations, namely, flow focusing [4], cross flowing (Y or T junctions) [5] and co-flowing [6], are most widely applied and studied. Considering the fact that the possible flow rate can be treated in these microfluidic facilities is far away from the demand of industrial applications, Belkadi et al. proposed the cross-junction microchannel model to fabricate emulsified biofuel by impinging continuous and dispersed phase fluids directly [7]. It is designed to work at flow rates two or three orders of magnitudes higher (i.e. ~10^2 mL/min) than with a traditional microfluidic device. This work is aimed to characterize droplet size and size distribution in cross-slot microsystems employing head-on impingement of oil and water jets for emulsifying. Except for the hydrodynamic conditions, the effect of the cross-junction layout on the emulsion quality is the main focus of the present study.
2. Methodology

Continuous phase (water) and dispersed phase (rapeseed oil) are supplied into the device by two Armenian pumps (AP TRIX 500). Fluids flow at a flowrate up to 500 mL/min is possible. Two weighing scales (Sartorius-MSE 2203) with an accuracy of 1 mg are used to measure flowrates of water and oil. To assess the pressure drop through the microchannels, two Gems 31IS series pressure transducers (0-250 bar) at the accuracy of 0.25% are assembled at the inlet channels for both phases. The emulsion issues from the two outlets are collected to perform sampling microscopy analysis with assistance of automated granulometry based on images processing by in-house Matlab coding.

2.1. Microchannel

The microsystems designed for highly efficient oil-in-water emulsification utilizing impingement configuration of two streams are shown in Fig.1; one of them, depicted in figure 1(b), has been investigated previously [7] for emulsified biofuel (water-in-oil emulsion) production. Figure 1(a) depicts the general 3D layout of the devices. One slab is mechanically grooved with square micro channels for the aqueous phase, oil phase and emulsions. The microchannels are arranged in a crossroad format. The other slab covers over it with several bolts and nuts tightened. The details of the microchannels layout and dimensions are shown in figure 1(b) and (c). The water channel and emulsion outlet channels have the same square cross section dimensions, which is 600 μm×600 μm. And the inlet channel cross section for the dispersed phase is 300 μm×300 μm. Based on the relative positions in the YZ plane for the two inlet axes, two configurations will be compared here; they are respectively designated as, asymmetric and symmetric. The asymmetric configuration (figure 1(b)) is characterized by an offset of water and oil feeding channels axis in the depth direction (Z) with the magnitude of 150 μm. As for the symmetric one (figure 1(c)), oil and water channels are simply coaxial. In addition, 90° bends in the middle way of outlet channels turns it to flow following the negative X direction, as illustrated in figure 1(d). These bends are intended to enhance breaking-up of droplets [8].

![Figure 1. Microchannel System: (a) microfluidic emulsification device; (b) Asymmetric cross-slot configuration; (c) Symmetric cross-slot configuration; (d) XY plane](image)

2.2. Test conditions

In order to compare the two configurations at different flow conditions, flowrates of water and oil are preset and input into the pump control systems based on targeted Reynolds number and oil to water volume ratio \( f = \frac{V_d}{V_c} \) in the emulsion, as summarized in table 1.

<table>
<thead>
<tr>
<th>( V_c ) (mL/min)</th>
<th>( V_d ) (mL/min)</th>
<th>( Re_c )</th>
<th>( f )</th>
</tr>
</thead>
<tbody>
<tr>
<td>50</td>
<td>5, 7.5, 10, 12.5</td>
<td>1520</td>
<td>10%, 15%</td>
</tr>
<tr>
<td>100</td>
<td>10, 15, 20, 25</td>
<td>3050</td>
<td>20%, 25%</td>
</tr>
<tr>
<td>150</td>
<td>15, 22.5, 30, 37.5</td>
<td>4570</td>
<td></td>
</tr>
</tbody>
</table>
3. Results and discussion

3.1. Drop size distribution

Figure 2 plots the comparison of cumulative distribution function (c.d.f) at the smaller flow rate conditions with varied dispersed oil fraction. It can be noticed from figure 2(a) that the cumulative probability ramps up rapidly at the range where drops are small, i.e. 1 to 10 μm. Within this range of droplet size, no significant difference between two cross-slot geometries can be distinguished. Disparities between the two configurations show up in the range of relatively bigger drops sizes as marked with green box in figure 2(a) and zoomed-in details is presented in figure 2(b). In general, the flow pattern inside asymmetric cross-slot configuration induces the formation of more large drops than with the fully symmetric one, so the cumulative probability distribution curves for the symmetric configuration converge to 100% faster at all tested oil fraction cases.

![Figure 2. Cumulative distribution function at $Re_c=1520$: (a) [0-80 μm]; (b) [5-60 μm].](image)

As the flow rates of both phases increase further, the geometry of the cross-slot seems to have less and less influence on the drop size distribution. Figure 3 shows cumulative probability evolution at the highest $Re$ condition investigated in the present study ($Re_c=4570$). It is sure that two phases with higher momentum impinging with each other at the compact microchannel allow getting smaller droplet size generally. Besides, the drop size distributions obtained after emulsification with varying oil fraction rate are also very close, the cumulative probability curves being quite coincident.

![Figure 3. Cumulative distribution function at $Re_c=4570$: (a) [0-80 μm]; (b) [5-60 μm].](image)
3.2. Mean drop size & emulsification efficiency

Figure 4(a) depicts the arithmetic mean drop diameter ($D_{10}$) and distribution SPAN ($[d_{90} - d_{10}] / d_{50}$) for tested conditions. Overall, the particular designed microfluidic microsystem tested in this study at the conditions of great amounts of dissipated kinetic energy leads to averaged drop diameters less than 10 μm at all experimental cases. No matter how the cross-slot is arranged geometrically, the averaged drop diameter, as well as the distribution SPAN, decreases with $Re$. The minimum averaged drop diameter measured is presently 3.9 μm. In the same order of ideas, at a given water flowrate, $D_{10}$ reduces as the oil flowrate increases if the water flowrate is kept because of the higher oil flow momentum imposed. It indicates the better performance of the symmetric configuration in terms of fabricating finer and more narrow distributed droplets in the final emulsion, especially at low $Re$ conditions.

Figure 4(b) illustrates the correlation of mean drop diameter to energy dissipation in different systems. It is found that the asymmetric configuration appears to be more efficient considering this criterion. Although symmetric configuration assists to manufacture smaller and more uniform oil droplets than the asymmetric configuration, it requires relatively more energy to fulfil this.

![Graph showing mean drop diameter and emulsification efficiency](image)

**Figure 4.** Mean drop diameter and emulsification efficiency: (a) $D_{10}$ & SPAN vs. $f$; (b) $D_{10}$ vs. $E$.

4. Conclusions

In the present study, experimental work is carried out to characterize the effect of the cross-slot layout on oil-in-water emulsification in the head-on impingement microchannels designed to work at high throughput. The arithmetic mean diameter of created emulsions is less than 10 μm at all experimental cases, and the minimum mean drop diameter, 3.9 μm, is obtained at the highest energy dissipation condition. Generally, the symmetric geometry of the cross-slot works better than the asymmetric one, allowing to produce a smaller number of large droplets and dispersing the oil phase more uniformly at the same hydrodynamic conditions, especially when $Re$ is low. As flowrates of both phase increases, the disparity is diminished. The symmetric configuration shows advantage in manufacturing finer and less poly-dispersed emulsions than the asymmetric one at given flow conditions. However, it demands relatively higher energy cost and its manufacturing is less simple.

References