LIQUID-LIQUID MIXING USING MICRO-FLUIDISED BEDS

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Abstract. This study experimentally investigates the application of a solid-liquid micro-fluidised bed as a micro-mixing device. The experiments were performed in a borosilicate capillary tube with an internal diameter of 1.2 mm (i.e. near the upper-limit dimension of a micro-fluidic system) using borosilicate particles with a mean diameter of 98 μm. Refractive index matching technique using sodium iodide solution was employed to achieve a transparent fluidised bed. Mixing performance of the micro-fluidised bed in terms of mixing time was investigated using dye dilution technique. The preliminary results showed that the mixing time in a micro-fluidised bed can be reduced by 1/8th of the time required for liquid-liquid mixing in a capillary tube. The rise in the mixing rate was considered to be due to secondary transversal mass transport caused by passive chaotic advection.

Keywords: micro-fluidised bed; micro-mixer; liquid-liquid mixing; mixing time; refractive index matching technique; dye dilution technique.

1. INTRODUCTION

Application of micro-fluidic devices for processing of multiphase flows in areas such as medical diagnostics, chemical analysis, power generation and fuel processing, invariably relies upon the physical or chemical interaction between at least two fluid phases. Such interaction is achieved through effective mixing. However, at length-scales associated with micro-fluidic devices where systems operate in a laminar regime with Reynolds numbers typically less than 1, the mixing process is rather poor as it is principally governed by molecular diffusion. Generally, active or passive techniques are employed to achieve effective mixing at micro-scales [1-2]. In active mixing external forces including ultrasound, acoustic, electrokinetic, and magneto-hydrodynamic forces are used to induce mixing. Passive mixing on the other hand is accomplished by maximising the mixing contact area and/or reducing the mixing path through “Streaming Techniques” where the flow path is restructured using geometrical based methods. The existing techniques however suffer from fabrication complexity which limits their mass productions, high fabrication and maintenance costs, and high energy dissipation rates [1-2]. To overcome the shortcomings of the above techniques, a fluidising-based micro-mixer system is considered here.

It is well-established that fluidisation provides efficient mixing and intensification of mass and heat transfer. In recent years, a number of studies focusing on miniaturised fluidised beds have been emerged [3-4]. The major focus of these studies was to establish the fluidisation hydrodynamic characteristics of the fluid beds both experimentally and theoretically. The focus of this work however is to experimentally investigate the mixing performance of two miscible fluids in a miniaturised solid-liquid fluidised bed in terms of mixing time. It should be noted that the work presented here is not intended as a systematic study of two-fluid mixing in a micro-fluidised bed but rather as an experimental proof of concept.
2. EXPERIMENTAL

A schematic diagram of the experimental setup is presented in Figure 1. The setup consisted of (i) a 30 cm glass capillary tube with an internal diameter of 1.2 mm (i.e. a dimension near the upper-limit dimension of 1 mm for micro-fluidic systems), (ii) a fluid reservoir, (iii) two syringe pumps, (iv) a LED light source and focusing lens, (v) a CCD camera, and (vi) a data acquisition system. The bed material was clear borosilicate glass sphere from Cospheric LLC with a size range of 90-106 µm and mean diameter of 98 µm, particle density of 2230 kg/m³, and refractive index of 1.47-1.48. A 52 µm wire mesh was used as a distributor.

![Figure 1. Schematic representation of the experimental setup.](image)

For optical diagnostics, the refractive index of the fluidising medium (i.e. sodium iodide solution) and bed material were matched (forming a transparent fluidised bed) minimising the noise generated by the light reflections from the surface of particles. Dye dilution technique [2] was used to determine the mixing performance at various operating conditions given in Table 1. In this work, the tracer dye stream (i.e. dye + sodium iodide solution) was fed in the centre from the bottom of the capillary tube through a 30G½ needle whilst the sodium iodide solution (56 wt%) was fed annularly passing through the fluid reservoir. The two streams have similar density (1800 kg/m³) and viscosity (0.0018 Pa.s). A LED lamp combined with a biconvex focusing lens was used to illuminate the region of interest using a back lighting technique. A cuvette filled with the sodium iodide solution was placed around the capillary tube to reduce the light refraction at the curved surface of the tube. An IDT XS3 high speed camera was then used to capture the dye dilution profile across the channel cross section at a rate of 50 Hz. 2000-3000 images were captured using IDT Motion Studio software whilst MATLAB software was employed for image analysis. Background images were also obtained at dye flow rates of zero at the start of each experiment. For benchmark purposes, the same experiments were carried out in the empty capillary tube (i.e. fluidised bed with no particles).

<table>
<thead>
<tr>
<th>Total flow rate ((Q_T)), µL/min</th>
<th>10</th>
<th>20</th>
<th>35</th>
</tr>
</thead>
<tbody>
<tr>
<td>Particle Reynolds number, (Re_p)</td>
<td>0.18</td>
<td>0.35</td>
<td>0.62</td>
</tr>
<tr>
<td>Dye stream flow rate ((Q_d)), µL/min</td>
<td>0</td>
<td>2</td>
<td>0</td>
</tr>
<tr>
<td>Clear stream flow rate ((Q_c)), µL/min</td>
<td>10</td>
<td>8</td>
<td>20</td>
</tr>
<tr>
<td>Flow ratio, (Q_d / Q_T)</td>
<td>0</td>
<td>0.2</td>
<td>0</td>
</tr>
</tbody>
</table>

*Calibration case for background subtraction.
2.1 Quantification Method for Mixing Characterisation

The criterion used to evaluate the performance of the liquid-liquid mixing within micro-mixers was the uniformity of the dye intensity. The concentration of the dye was estimated from the shadow image generated by the diffusing jet of opaque dye stream using the image processing in MATLAB software. The background images obtained at the start of each experiment was averaged and subtracted from individual mixing images. In a shadow image, the pixel intensity is inversely proportional to the concentration of dye at that point. Therefore a negative of each image was taken after the background subtraction. The normalised concentration was then calculated by,

\[
C^* = \frac{I - I_{\text{min}}}{I_{\text{max}} - I_{\text{min}}}
\]  

(1)

where \(I_{\text{max}}\) is the maximum intensity (i.e. the dye intensity at the nozzle exit), \(I_{\text{min}}\) is the minimum intensity and \(I\) is the intensity of each pixel.

The mixing index was calculated to determine the degree of mixing. First the x-variance of the concentration profile, \(\sigma_x\), was determined by fitting a Gaussian curve (Equation 2) through the normalised profile.

\[
f(I) = \exp \left( -\frac{(I - I_{\text{max}})^2}{2\sigma_x^2} \right)
\]

(2)

where \(I\) is the intensity of the pixel, \(I_{\text{max}}\) is the intensity at the nozzle inlet, \(\sigma_x\) is the x-variance of the concentration profile in a horizontal line at a given height. The intensity of the peak was then adjusted for each height whilst maintaining the same area under the curve and x-variance. The y-variance was then calculated from the above concentration profiles. The mixing index, \(M\), is given as,

\[
M = \frac{\sigma_x - \sigma_y}{\sigma_y - \sigma_{y,\infty}}
\]

(3)

where \(\sigma_{y,\infty}\) is zero (i.e. flat concentration profile) and the initial variance, \(\sigma_{y,0}\), is \(p \times (1 - p)\). \(p\) denotes the fraction of tracer dye \(2\).

3. RESULTS AND DISCUSSION

Typical time-averaged normalised concentration profiles of the dye in the capillary tube and the micro-fluidised bed are presented in Figures 2a and 2b, respectively. As Figure 2a shows, in the capillary tube the interface between the two streams remains undisturbed with dye and clear streams following their distinct core and annular flow paths, respectively. Conversely, under the same operating conditions, enhanced mixing was observed when the two fluid streams were introduced into the miniaturised fluidised bed (Figure 2b).

Figure 3a and 3b illustrate the normalised concentration distribution profile at different elevations along the height of the capillary tube and the micro-fluidised bed, respectively. The bottom of the capillary is considered as zero elevation. The flatness of the profile corresponds with the degree of mixing (i.e. a flat profile represents a complete mixing). In the capillary tube the concentration profile remained almost unchanged over the height of the tube (Figure 3a) with the shape of the profile representing non-mixed regions along the examined elevations of 0.82, 1.3, 2.3, 3.3 and 4.3 mm. In the miniaturised fluidised bed however the
concentration profile became flatter as the height increased approaching the average concentration profile (i.e. the dashed line in Figure 3b). That in turn indicates an increase in the homogeneity of the mixture and hence significant improvement in mixing.

Figure 2. Time-averaged concentration profiles for (a) capillary tube, (b) micro-fluidised bed [QT = 20 µL/min, Qd / QT = 0.4].

Figure 3. Concentration profiles at different elevations for (a) capillary tube, (b) micro-fluidised bed [QT = 35 µL/min, Qd / QT = 0.2].

Figure 4 shows the mixing index versus residence time for both the capillary tube and the micro-fluidised bed. Generally the mixing index decreases as the time increases indicating an increase in the mixing efficiency. Clearly the rate of mixing achieved in the micro-fluidised bed is much greater than that of the capillary tube. For example at the total flow rate of 35 µL/min and flow ratio of 0.2, a mixing index value of 0.1 (i.e. 90% mixing efficiency) can be achieved in the micro-fluidised bed at 1/8th of the mixing time required for the capillary tube.
Further improvement was achieved as the dye flow rate to total flow rate ratio increased from 0.2 to 0.5. A mixing index of 0.03 (i.e. 97% mixing efficiency) was obtained at less than 8 s.

Figure 4. Mixing index versus residence time \([Q_T = 35 \, \mu L/min]\).

Figure 5 shows the effect of an increase in total flow rate on mixing performance of the micro-fluidised bed. As the flow rate increases from 10 to 35 \(\mu L/min\), the rate of mixing increases reducing the mixing time for achieving mixing efficiency of 92% (i.e. mixing index of 0.08) by more than half.

Figure 5. Mixing index versus residence time \([Q_d / Q_T = 0.2]\).
The results of this preliminary study show the advantage of using a miniaturised fluidised bed as a micro-mixer over a core-annular micro-mixer experimentally. In a core-annular micro-mixer (i.e. the capillary tube), mixing is mainly driven by molecular diffusion which causes transversal mass transport. As a result the mixing is extremely slow and inefficient requiring long residence time and hence great mixing length. In the micro-fluidised bed however, the mixing is considered to be driven by passive chaotic advection with a three dimensional orbit which in turn leads to secondary transversal mass transport. Also, the bed materials (i.e. particles) act as obstacles and hence lead to periodic changes in the flow directions causing splitting and recombining effect with streamlines crossing each other. Such unique characteristics can improve mixing significantly at low Reynolds numbers.

Since the governing mechanisms for mixing in a micro-fluidised bed (i.e. destabilisation of the diffusion layer between the two fluids by fluidised particles) and a core-annular micro-mixer (i.e. diffusion) are fundamentally different, a rigorous examination of the micro-fluidised bed performance is needed to establish the micro-fluidised bed mixing efficiency relative to that of more efficient micro-mixers developed based on flow destabilising principles. Such a study is currently underway at The University of Newcastle and the results will be presented in future publications.

4. CONCLUSIONS
The novel idea of using a fluidised bed with micro-scale dimensions as a micro-mixer for mixing of two miscible liquids was examined experimentally. The results of the study were benchmarked against mixing behaviour in a capillary tube in which the driving mechanism for liquid-liquid mixing is molecular diffusion only. Generally, the fluidisation process found to reduce mixing time dramatically, achieving high mixing efficiencies at less than 1/8th of the time required in a capillary tube. An increase in the flow rate was found to increase the mixing rate within the micro-fluidised bed, and hence reducing the mixing time for high efficiency mixing of two liquids significantly.

Acknowledgement
This work was supported by the Australian Research Council and the University of Newcastle Priority Centre for Advanced Particle Processing and Transport.

5. REFERENCES