Residence Time Distributions in Laminated Microstructured Plate Reactors

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Abstract Residence time distributions (RTDs) have been investigated experimentally for systems with straight and zig-zag channels. The channels are formed by microstructured plates placed on top of each other and containing obstacles and holes to allow flow in 3 dimensions. Experimental RTD measurements were performed by monitoring the concentration of a tracer dye by means of a LED-photodiode system. The RTD was obtained for five different flowrates for both geometries. It was found that the zig-zag channel configuration gives a narrower distribution as compared to the straight channel one. Furthermore, as the flowrate increased the standard deviation of the distribution increased.
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Keywords: Residence time distribution, microchannel, flow inversion

1. Introduction

Residence time distributions (RTDs) are a useful tool for characterising the hydrodynamic behaviour of reactors. Since reaction conversion and selectivity are strongly affected by this distribution it must be accurately measured. Residence time distributions have been measured in different microchannel configurations. Adeosun and Lawal (2005) showed theoretically that microstructured packed bed configurations exhibit a narrower RTD as compared to a simple microchannel. Boškovic and Loebbecke (2008) investigated the RTD of three different split-and-recombine micromixers by fitting an empirical model to deconvoluted experimental data. It was found that for all cases the RTD became narrower by increasing the flowrate due to the formation of secondary flows causing chaotic advection. Stroock *et al.* (2002) proposed a chaotic mixer which consisted of staggered herringbone structures patterned on the floor of the microchannel. It was shown that this staggered herringbone mixer minimised dispersion at high Peclet numbers compared to a rectangular channel.

Residence time distributions are usually obtained by injecting an inert tracer at the channel inlet and measuring its concentration at the outlet. Different approaches for the tracer introduction and recording of the outlet concentration have been presented for microchannels. Günther *et al.* (2004) used a T-junction along with computer controlled syringe pump switching for the introduction of the tracer and an LED-photodiode system for the measurement of tracer concentration. Trachsel *et al.* (2005) injected the tracer as a Dirac-delta pulse by a piezoelectrically actuated membrane and recorded its concentration by fluorescence microscopy. Boškovic and Loebbecke (2008) injected the tracer with an HPLC valve and recorded its concentration by an in-house made UV-vis flow-through cell. Lohse *et al.* (2008) described a novel method for the determination of the RTD based on the optical activation of a caged fluorescence dye. Tracer concentration was determined by fluorescence microscopy. This method allows for the determination of RTD without the need of measuring the inlet signal because the inlet is ensured to be a Dirac-delta pulse.

In this work, the RTD for two different laminated plate reactor configurations are considered containing straight or zig-zag channels. The residence time distributions are obtained by means of a LED-photodiode array system for five different flow rates. Standard deviations as a function of residence time are obtained to compare the level of dispersion encountered.

2. Experimental Details

The reactors used in this study were provided by Chart Energy & Chemicals based on the Shimtec® technology. The reactor is comprised of a stack of microstructured stainless steel and copper sheets with etch-through features. The sheets have dimensions of 150x60mm. Channel depth and total volume of the reactor can be varied by stacking a different number of sheets. Closed channels are obtained by clamping the metal sheets with an acrylic plate with inlet and outlet holes. The copper sheets are placed alternating between the stainless steel ones. The acrylic plates come in contact only with copper sheets. A picture of the assembled and disassembled reactor is shown in figure 1, while in figure 2 sheets of straight channel and zig-zag configurations employed for the corresponding reactors are shown. The inlet and outlet holes in the acrylic plates were moved to the sides so as to allow optical access for the RTD measurement.

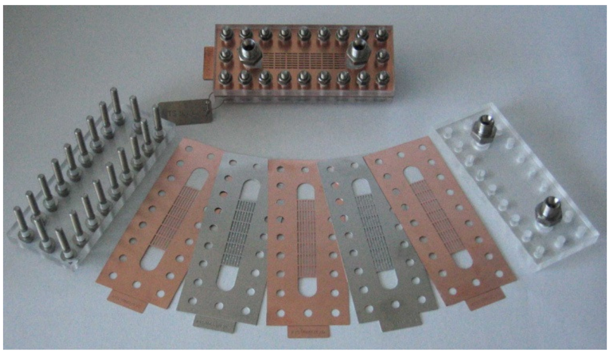


Fig. 1. Assembled and disassembled laminated plate reactor with sheets of straight channel configuration.

The tracer pulse (Parker Blue dye) was introduced by a 6-port sample injection valve (Rheodyne 7725(i)) equipped with 5 µl sample loop and an internal position signal switch that indicates the time of injection. Tracer detection was performed by light absorption. Illumination was provided by two square LEDs (Kingbright L-1553IDT).

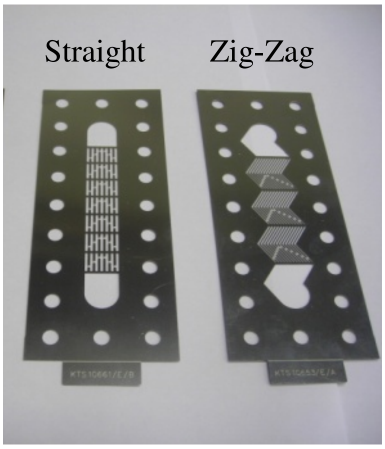


Fig. 2 Sheet geometries employed in the plate reactors. “Straight” have channels with a hydraulic diameter of 1.07 mm and “Zig-Zag” have channels with 0.75 mm.

To make sure that only light going through the desired channel area was collected, black tape was used to mask the neighbouring areas. To isolate the system from ambient light it was placed in a dark box. The detection system was based on a linear diode array detector (TSL, 1401R-LF) which had 128. This was driven using the manufacturer's recommended circuit. A scan of all diodes would take 1.28 ms and the interval between successive scans was 5.12 ms. Data from the sensor was collected using a National instruments PCI-6010 data acquisition card before being analysed and displayed on a computer using a program written in Labview. Every 100 ms the computer would average the previous two scans, calculate the absorbance for each diode and display the result. The absorbance of the tracer dye was found to be in accordance with the Beer-Lambert law. A picture of the experimental set up is shown in figure 3.

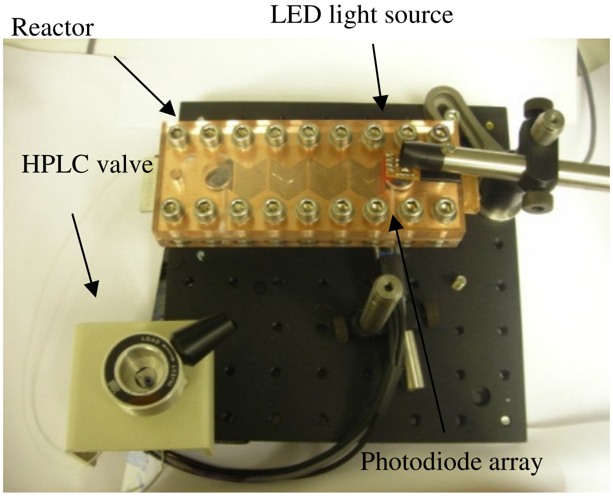


Fig. 2. Picture of the experimental set up used for RTD studies.

2.1 Data Analysis

The mean residence time is calculated according to:

$$t_m = \frac{\sum_{i=1}^{n} t_i I(t_i)\Delta t_i}{\sum_{i=1}^{n} I(t_i)\Delta t_i} \quad (1)$$

where I is the light intensity obtained from the LED-photodiode system and $\Delta t = t_{i+1} - t_i$ is the time interval. The residence time distribution is obtained from:

$$E(t_i) = \frac{I(t_i)}{\sum_{i=1}^{n} I(t_i)\Delta t_i} \quad (2)$$

The RTD in dimensionless form is obtained from:

$$E(\theta) = t_m E(t_i) \quad (3)$$

The variance of the distribution can be calculated as follows:

$$\sigma^2 = \frac{\sum_{i=1}^{n} (t_i - t_m)^2 E(t_i)\Delta t}{\sum_{i=1}^{n} E(t_i)\Delta t} \quad (4)$$

which in dimensionless form changes to:

$$\sigma_\theta^2 = \frac{\sigma^2}{t_m^2} \quad (5)$$

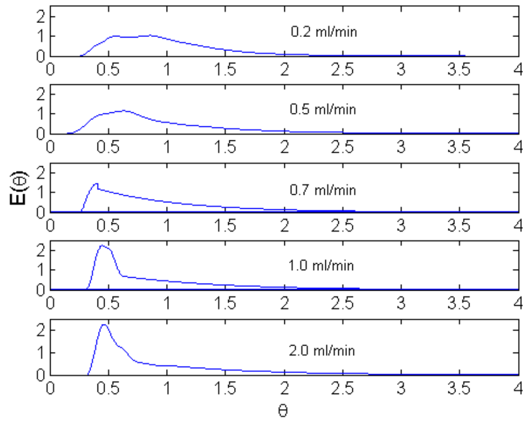
4. Results and Discussion

The residence time distributions for the two different geometries at five flowrates were obtained experimentally. An average of three measurements was taken with an approximate error of 8%. The error was calculated by calculating the standard error $\left(SE = \frac{stdev}{\sqrt{n}}\right)$ and divide it by the average value for every data point. An average over all the data points gave a value of 8%.

Figure 4 shows the dimensionless RTD obtained from equation (3) for all flowrates and reactor configurations. For both geometries as flowrate increases the peak of the RTD is encountered at earlier times. However the peak height increases monotonically for the straight channel configuration, but shows a minimum for the zig-zag configuration as flowrate increases. It can be seen that for all cases the zig-zag configuration shows a narrower distribution compared to the straight one.

Although the RTD graphs have the advantage of giving a qualitative idea of how narrow the distribution is, sometimes is hard to tell which distribution is actually narrower. For this reason the dimensionless standard deviation of the RTD, obtained from the square root of equation 5, is plotted against residence time in figure 5. The figure confirms that the zig-zag geometry has a narrower RTD (given by its lower standard deviation) than the straight geometry at all times. The lower hydraulic diameter of the zig-zag configuration partly explains the lower dispersion obtained for this geometry when compared to the straight one.

A



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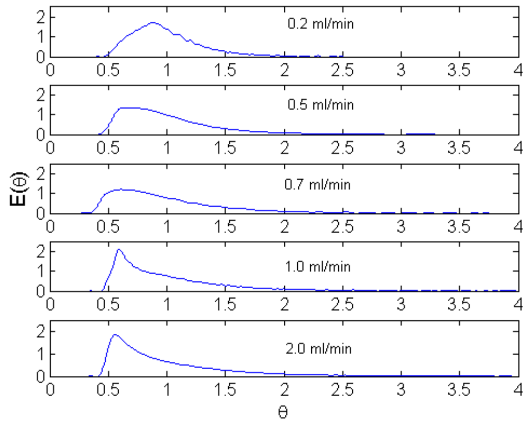

Figure 4. Dimensionless RTDs for the two reactor geometries at different flowrates. A) Straight, B) Zig-Zag.

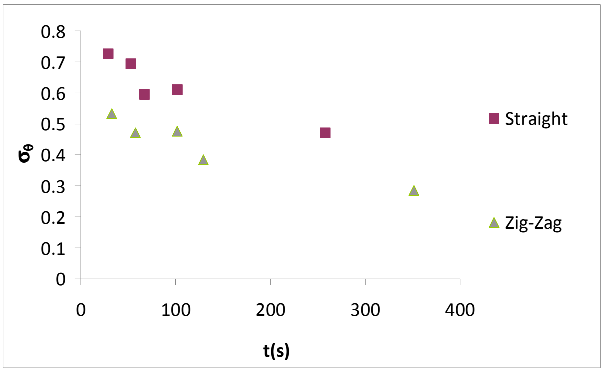

Fig 5. Dimensionless standard deviation of the RTDs for the two geometries at 5 different residence times (flowrates).

5. Conclusions

Residence time distributions for two different reactor configurations with straight and zig-zag channel geometries were investigated experimentally. The results indicated that using channels in a zig-zag configuration results in a narrower RTD than for straight ones. Therefore reaction conversion and selectivity can be improved by using a zig-zag configuration. For both geometries it was found that the peak of the RTD is found at earlier times when flowrate increases. The peak height increased monotonically for the straight configuration when flowrate increased, while in the zig-zag configuration it passed through a minimum. In this study only plates with the same geometry have been considered (straight or zig-zag), however a combination of plates may give an optimised result.

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