Presented at the COMSOL Conference 2008 Hannover

Mixing and Residence Time Distribution Studies in Microchannels with Floor Herringbone Structures

Alberto Cantu-Perez
Asterios Gavriilidis
Chemical Engineering Department
University College London

Overview

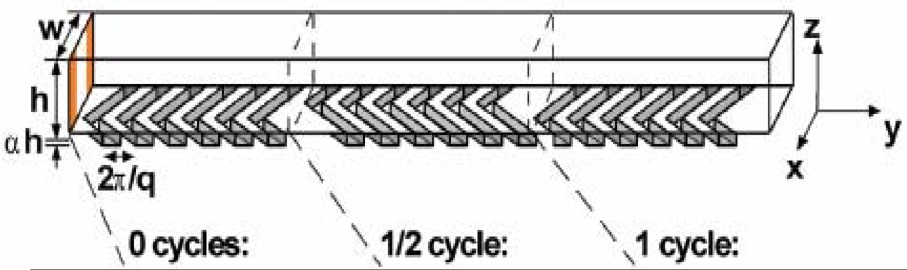
- Introduction
- Geometry of devices
- Use of COMSOL Multiphysics
- Quantification of mixing via:
  - Nearest Neighbour Analysis
  - Evolution of the striation thickness
  - Stretching calculations
- Residence time distributions as a function of Pe
- Discussion
- Summary

Introduction

- Microprocess technology offers advantages compared to macroscopic equipment in terms of high heat and mass transfer.

- Conversion and selectivity in chemical reactions is strongly dependent on the degree of mixing and the residence time distribution (RTD) of the reactor

Geometry studied



Staggered Herringbone Micromixer (Stroock et al 2002).

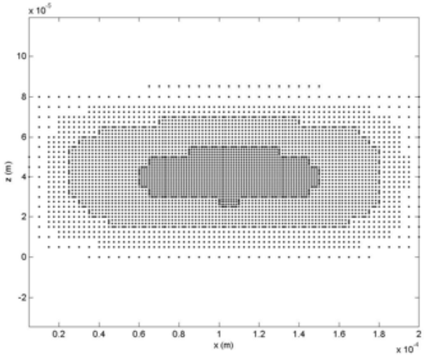
* These properties were selected for modelling to compare with experimental results in Stroock et al. 2002)

	Staggered Herringbones	**Symmetric Herringbones**	**Rectangular channel**
Width (m)	200×10^{-6}	200×10^{-6}	200×10^{-6}
Height (m)	85×10^{-6}	85×10^{-6}	85×10^{-6}
Length per cycle (m)	1.516×10^{-3}	1.516×10^{-3}	1.516×10^{-3}
Volume per cycle (m^3)	2.94×10^{-11} Including grooves	2.94×10^{-11} Including grooves	-
Number of grooves per cycle	12	12	-
Relative groove depth (α)	0.18	0.18	-
Wave vector q (m)	$2\pi/100 \times 10^{-6}$	$2\pi/100 \times 10^{-6}$	-
Groove asymmetry	2/3	1/2	-
θ	45°	45°	-
Groove depth $2\alpha h$ (m)	30.6×10^{-6}	30.6×10^{-6}	-
Fluid Properties			
Density (kg/m^3)*	1200	1200	1200
Viscosity (Pa.s)*	0.067	0.067	0.067
Mean Velocity (m/s)*	0.002	0.002	0.002

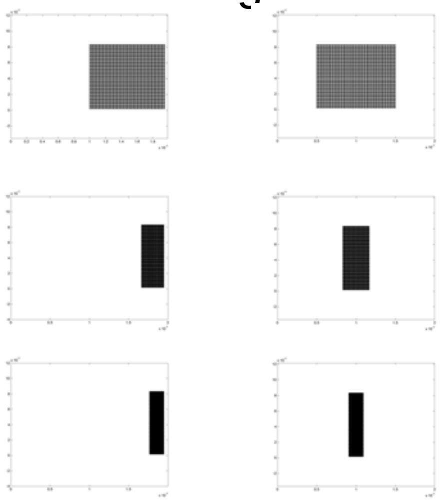
Use of COMSOL Multiphysics

Initial positions of particles

RTD



Mixing



Navier-Stokes Equation

$$\rho \frac{\partial v}{\partial t} - \nabla \cdot \left[\eta \left(\nabla v + (\nabla v)^T \right) \right] + \rho (v \cdot \nabla) v + \nabla p = F$$

Continuity Equation

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho v) = 0$$

COMSOL Multiphysics 3.3

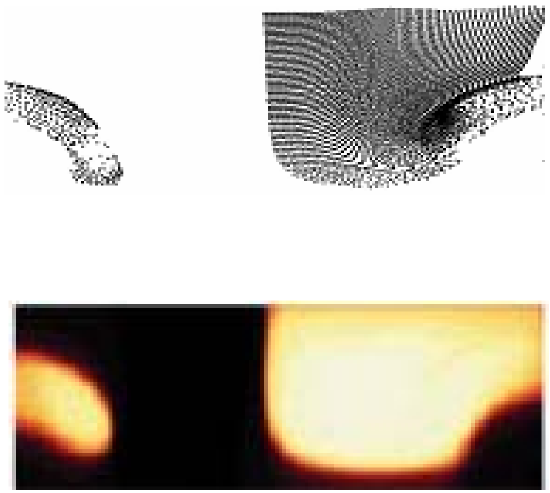
Particle Tracking $$\frac{d\vec{x}}{dt} = v(\vec{x})$$

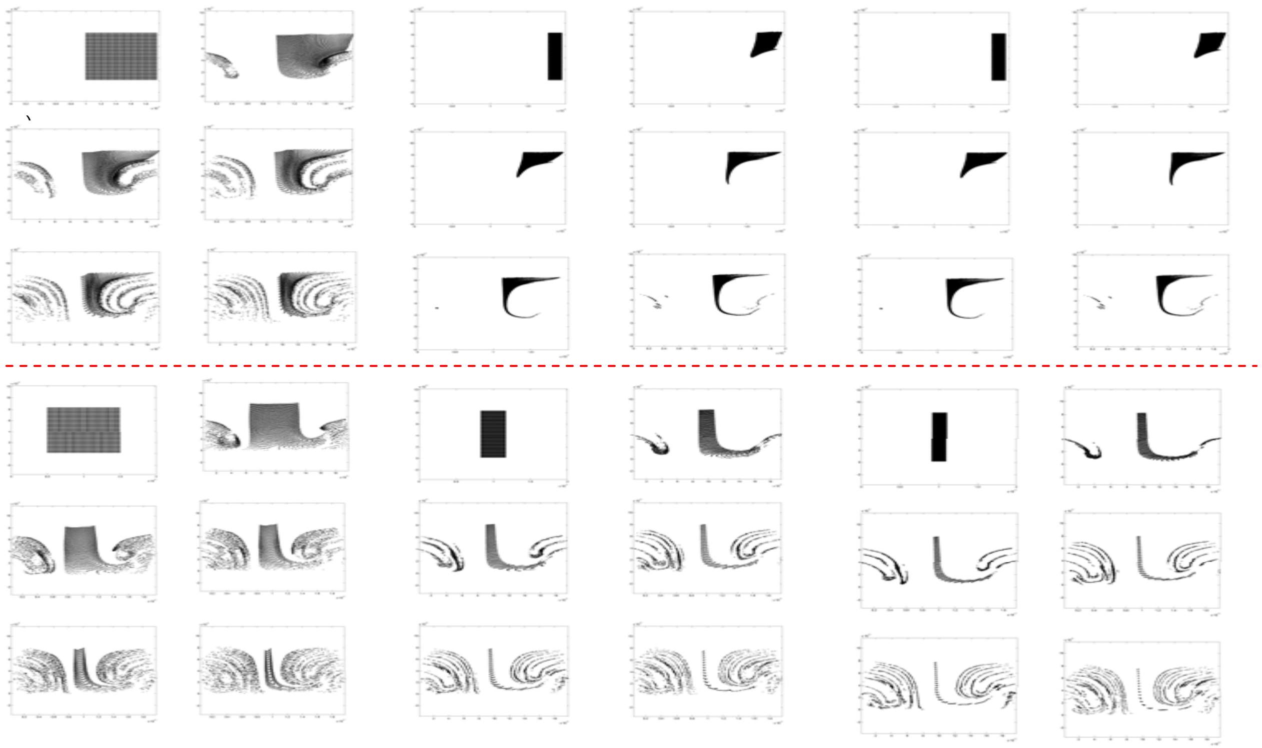
Random Walk $$d\vec{x} = Udt + \sqrt{2Ddt}\,\xi$$

MATLAB 7.0

Mixing visualisation

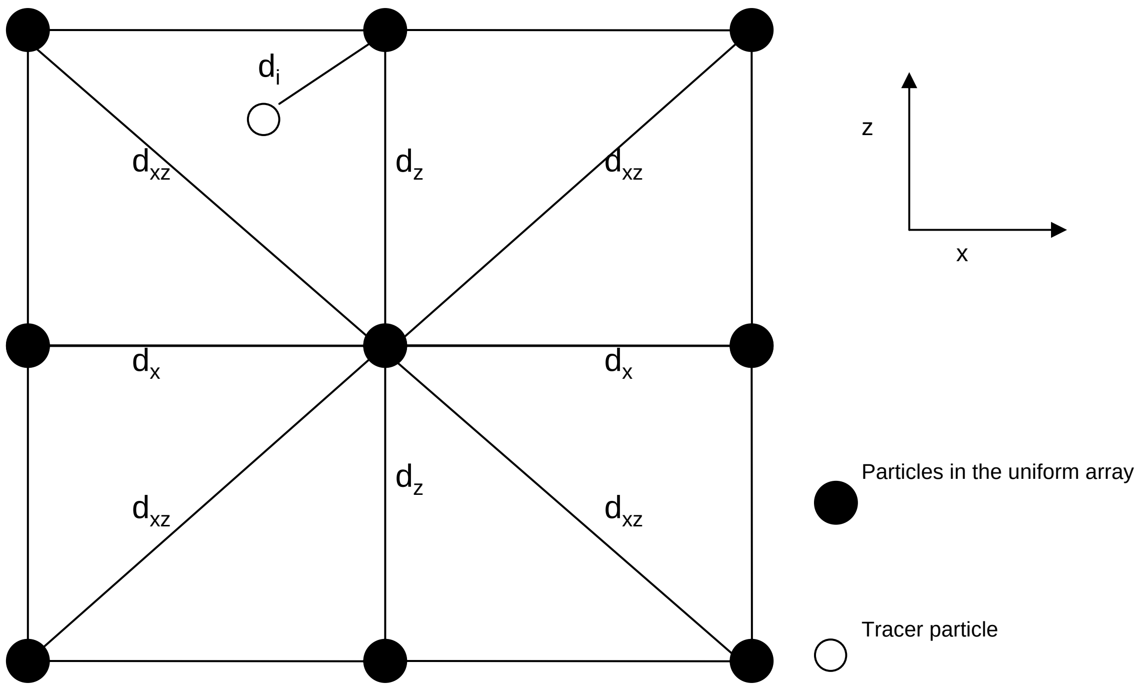
Model Validation | **Mix ratio 1:1** | **Mix ratio 1:5** | **Mix ratio 1:10**





Experimental results from
Stroock et al. 2002

Quantification of mixing via Nearest Neighbour Analysis



$$d_i = \sqrt{\left(x_i - x_{np}\right)^2 + \left(z_i - z_{np}\right)^2}$$

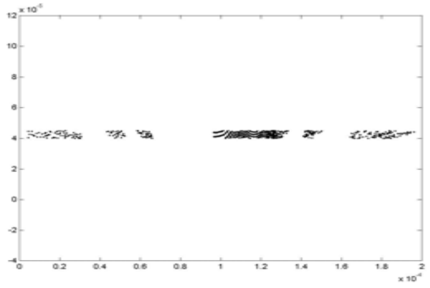
$$d_{mean} = \frac{2d_x + 2d_z + 4d_{xz}}{8}$$

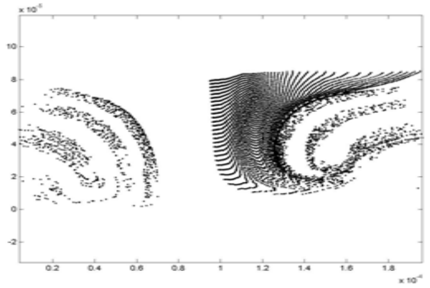
$$\mathrm{var} = \frac{\sum_{i=1}^{N}\left(d_i - d_{mean}\right)^2}{N-1}$$

$$COV = \frac{\sqrt{\mathrm{var}}}{d_{mean}}$$

$$\%mixing = \frac{n_{uniform}}{N}$$

Striation Thickness Procedure





$$w_i = \frac{st_i}{W}$$

$$Avgst_j = \sum_{i=1}^{k} w_i st_i$$

$$Avgst = \sum_{j=1}^{10} Avgst_j$$

$$s(t) = \frac{s(0)}{\lambda} = \frac{s(0)}{e^{(\alpha t)}} = s(0)e^{(-\alpha t)}$$

The thickness of the striations are measured by identifying the initial and final particle of the striation, if the particles are separated within a distance of 2.5 μm then is considered that they belong to the same striation. In addition the thickness of the spaces without particles has also been measured since it represents the other fluid been mixed

Mixing Length via Stretching histories

$$\frac{dx}{dt} = v(x)$$

$$\frac{d(l)}{dt} = (\nabla v)^T . l \qquad l_{t=0} = l_0$$

$$\lambda = \frac{|l|}{|l_0|}$$

$$\lambda_m = \left(\prod_{i=1}^{n} \lambda_i \right)^{\frac{1}{n}}$$

$$a = \lim_{t \to \infty} \left[\frac{1}{t} \ln(\lambda_m) \right]$$

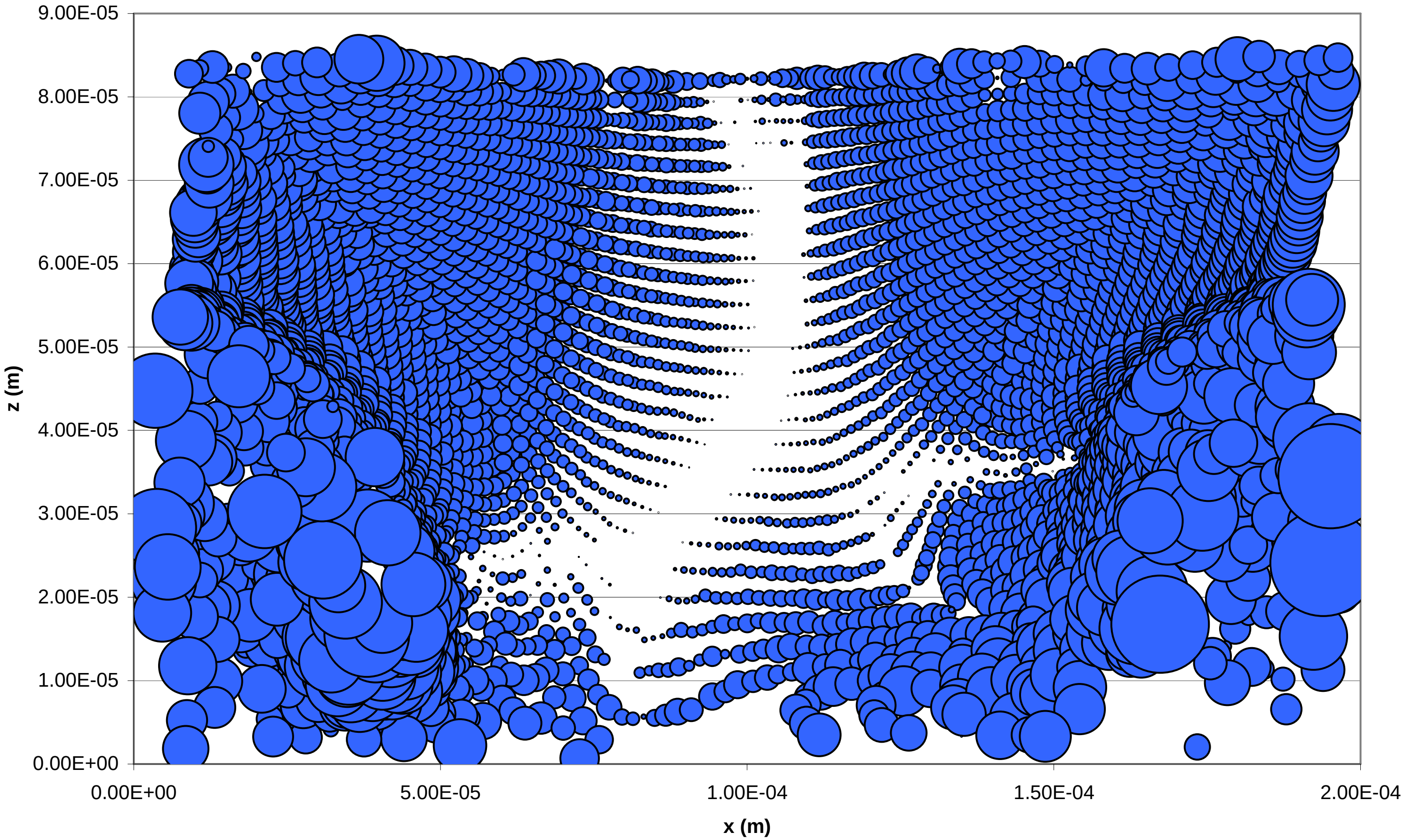
$$s(t) = \frac{s(0)}{\lambda} = \frac{s(0)}{e^{(\alpha t)}} = s(0) e^{(-a t)}$$

$$\boxed{t = \frac{\ln\left(\frac{(s(0))^2 2a}{D} + 1 \right)}{2a}}$$

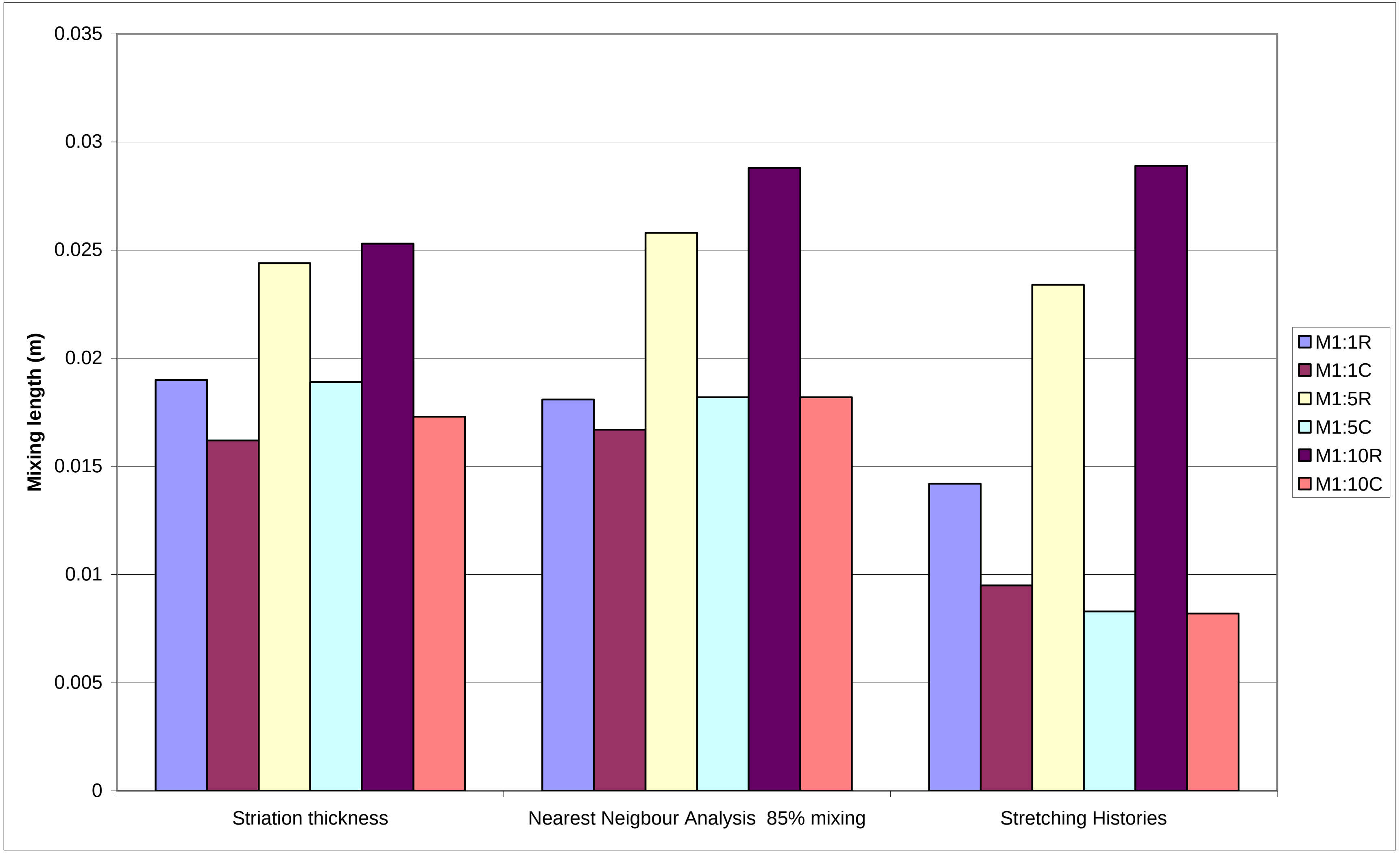
- Mixing time can be estimated with the specific stretch calculated with the equations on the left.

- Mixing is achieved when the penetration distance equals the striation thickness.

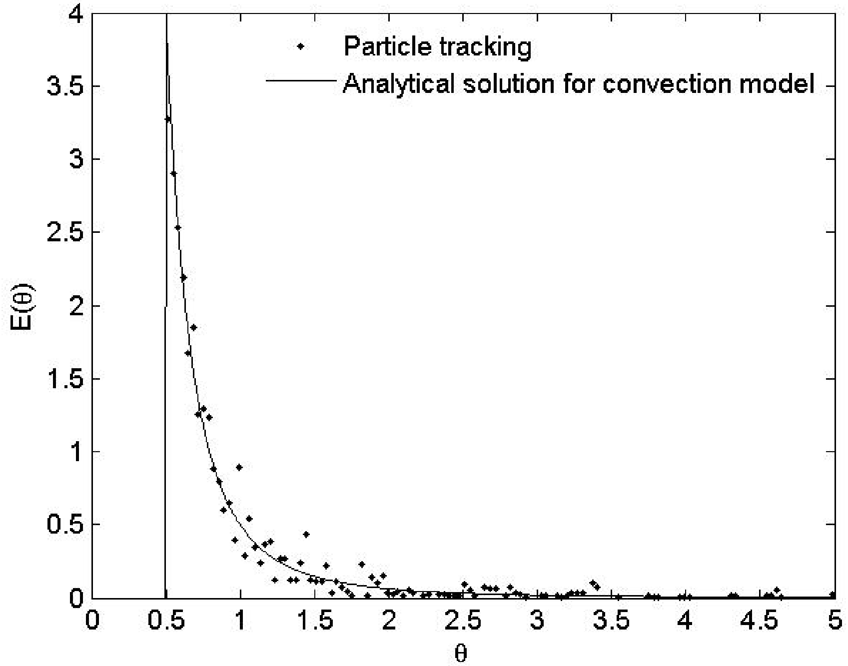
Distribution of stretching values on the cross section of the channel.

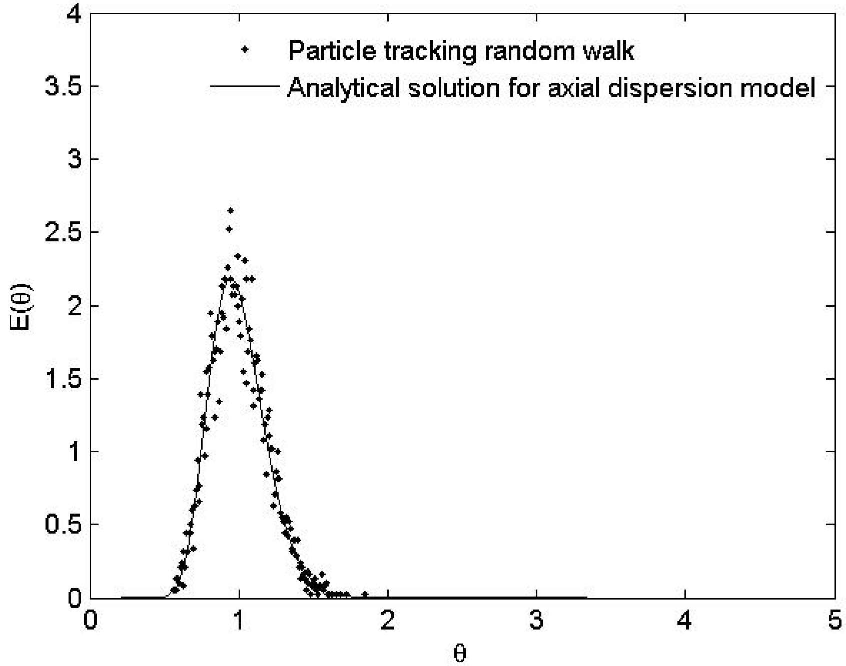


Mixing lengths for all mixing ratios and injection locations obtained with three different methods



Validation of numerical method for RTD





•The RTD obtained from the particle tracking algorithm (no diffusion) agrees well with the convective model for a cylinder with $E(t)=\frac{t_m^2}{2t^3}$ $t \geq \frac{t_m}{2}$ as shown on the figure on the left side. This is only valid in the limit of Pe~∞

•For Pe~10^2 the RTD should be obtained by the particle tracking with random walk method. The figure on the right shows the agreement between the RTD from the random walk procedure and the analytical solution to the axial dispersion model for a cylinder with Pe=153 $Pe=\frac{U_m d}{D}$

Comparison SHM and Rectangular channel

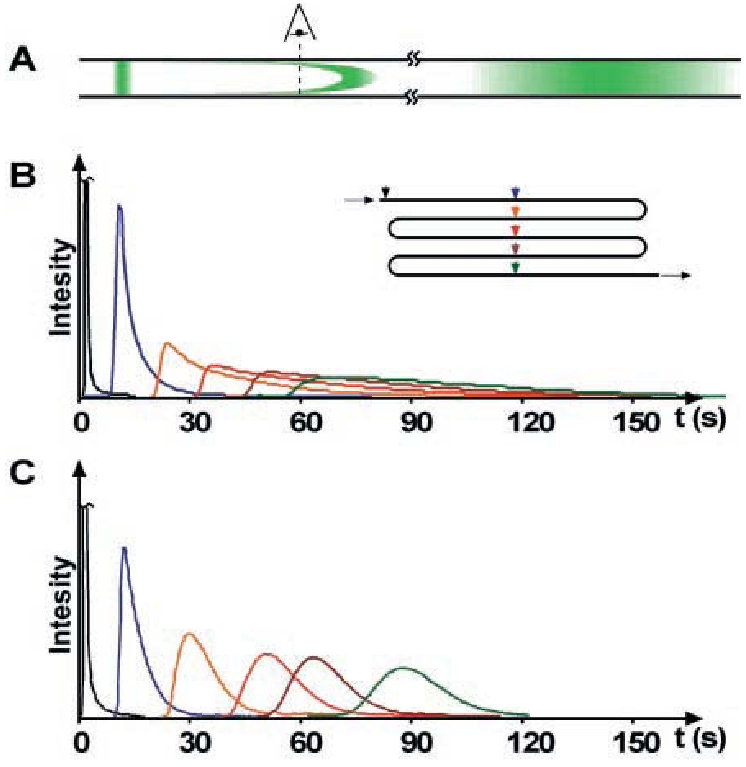
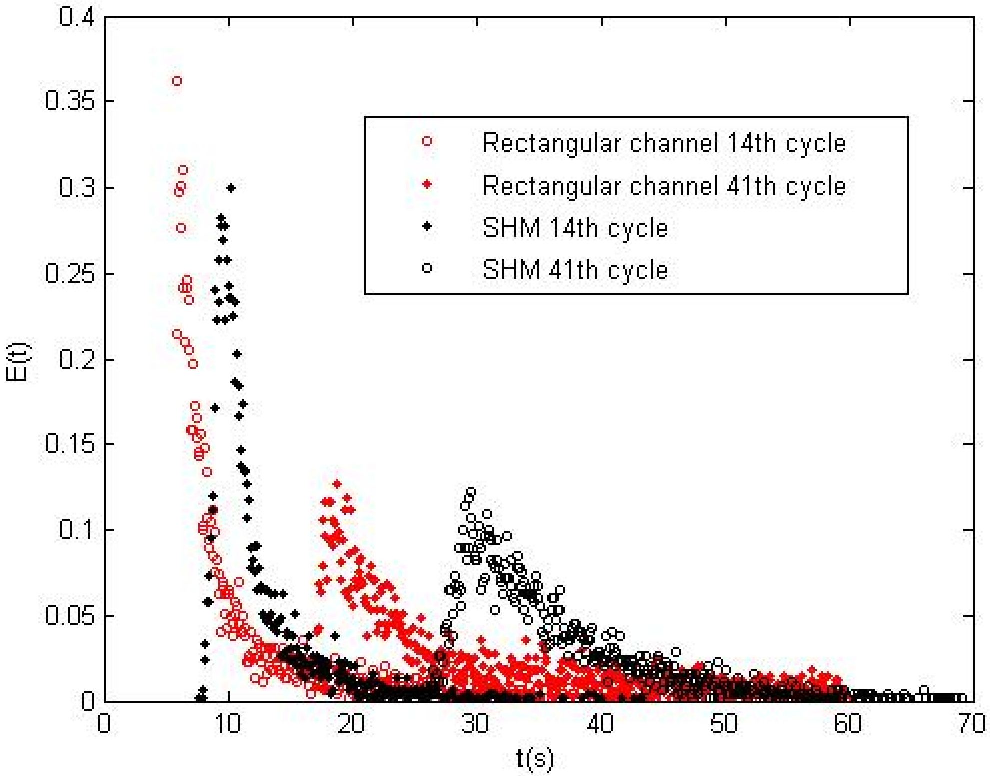


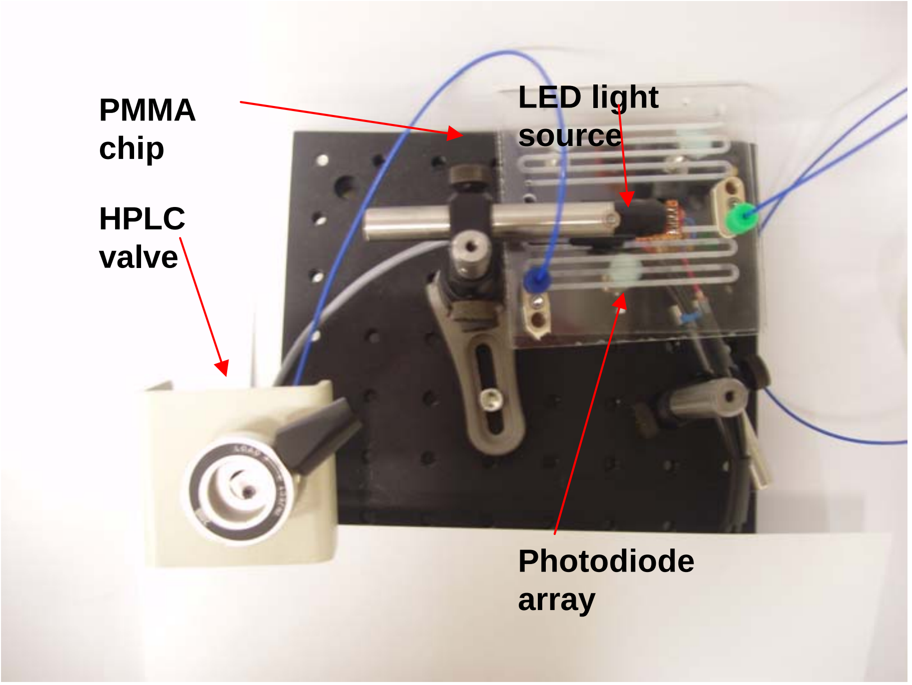
Figure from Stroock et al. 2002



In both experimental (figure on the left from Stroock et al 2002) and modelling results (figure on the right) the SHM shows a narrower RTD than an unstructured rectangular channel. Pe=10^4 Dimensions of the channel: width=200μm height 85μm.

Experimental Set up

- An HPLC pump was used to move the solvent carrier through the capillaries and the fabricated chips.
- An HPLC valve equipped with a 5ml loop was used to inject the tracer (Parker Quink Ink Permanent Blue with *D*~$1\text{x}10^{-10}$ m^2/s)
- The signal of the tracer was measured at the injection and the outlet by means of an in house made LED detector.
- The intensity of the signal was recorded and processed with a program developed in LabVIEW.
- The experimental chips were fabricated in PMMA by conventional micromachining



Experimental set up

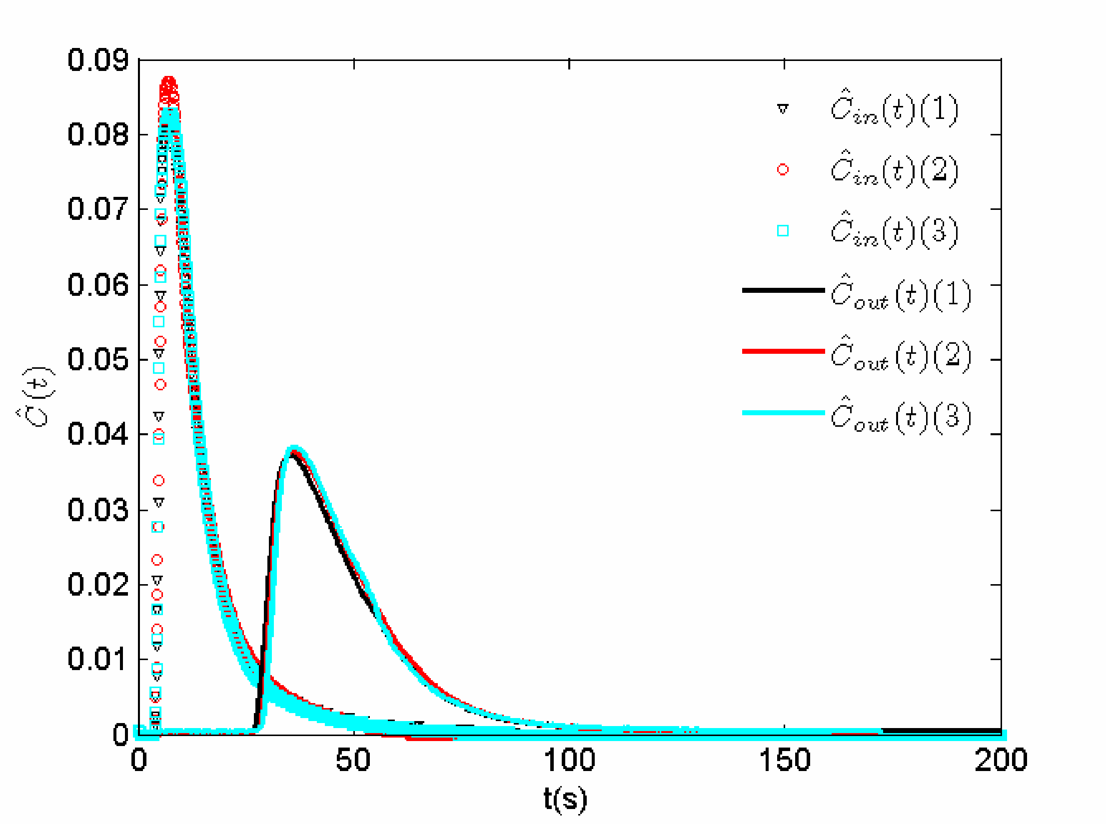
Experimental Results

•An average of 3 measurements is taken on each location.

•For the set up of the LED detector 2 locations have been selected for measurement (Injection and L2=21cm).

•The RTD was obtained with:

$$E(t) = FFT^{-1}\left(\frac{FFT(C_{out}(t))}{FFT(C_{in}(t))}\right)$$



Experimental results for a staggered herringbone channel with Pe~10^4.

Comparison Experimental vs Modelling for channels with herringbones

•Experimental data have been corrected for through the wall measurement. (Levenspiel et al. 1970)

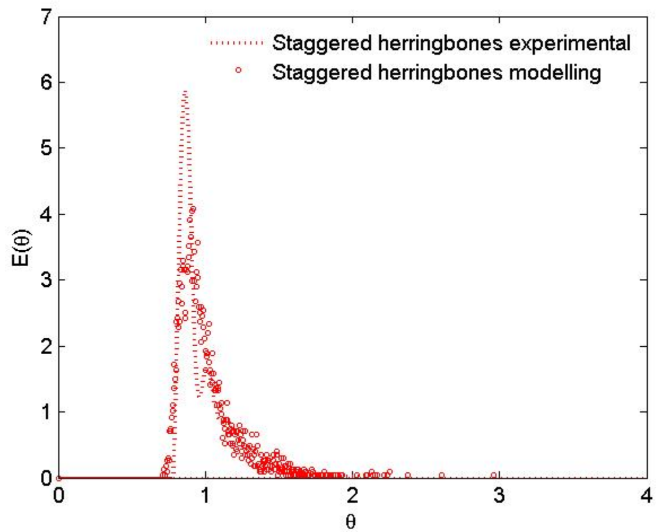
Experimental conditions were:

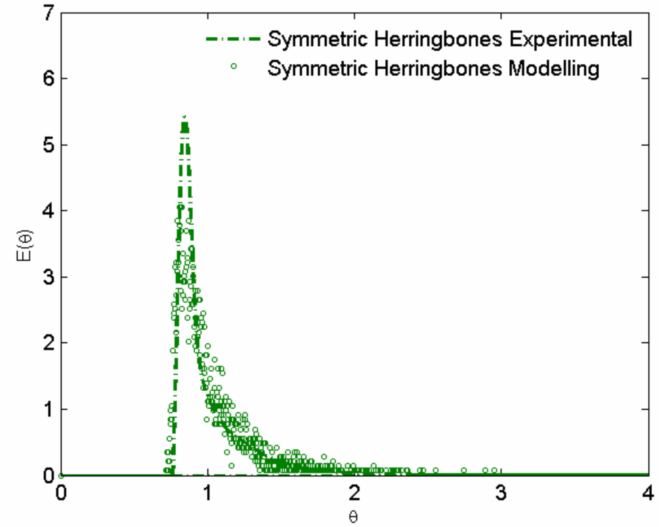
W=2mm

H=0.85mm

Pe~10^4

•The qualitative agreement between the experimental results and the modelling is good.

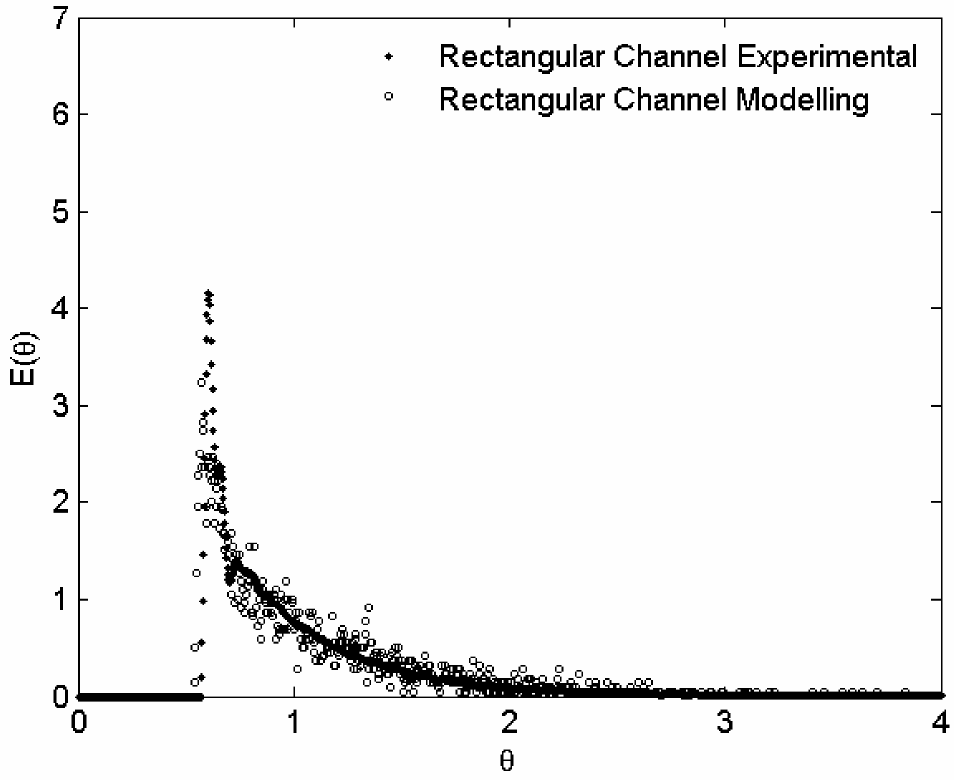




Comparison Experimental vs Modelling for a rectangular channel

•The RTD curves for a rectangular channel with $Pe \sim 10^4$ show a more asymmetric distribution than for channels with herringbone structures.

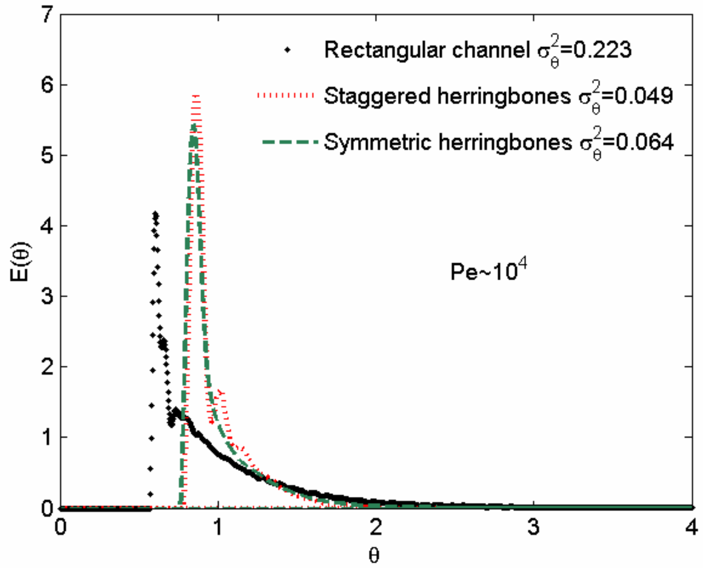
The RTD for a rectangular channels is similar to the one predicted by the convective model with a high peak at half the mean residence time, followed by a long tail.

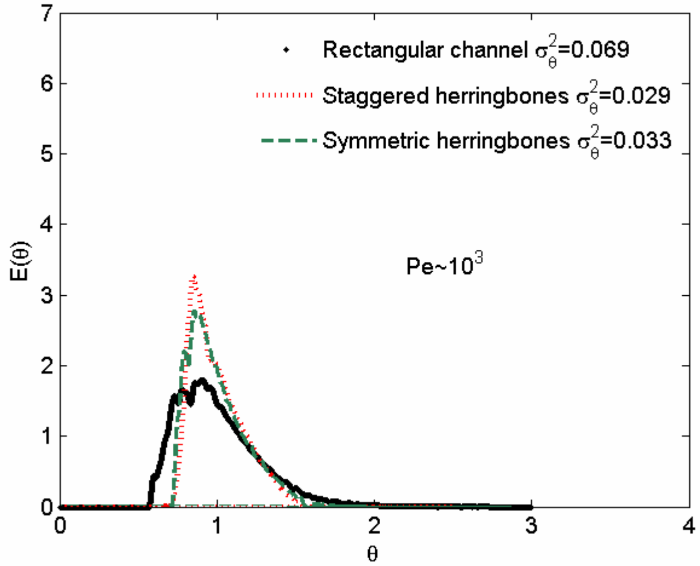


Experimental Comparison of channels with and without herringbone structures for different Pe

• For Pe~10^4 the RTD for the staggered herringbone channel shows a narrower distribution than a rectangular unstructured channel. The channel with the symmetric herringbones also has a better performance than the plain channel and is similar to the one with staggered herringbones.

•For Pe~10^3 the channels with herringbones have nearly the same distribution and they both still outperform the plain channel (although the differences are more subtle).

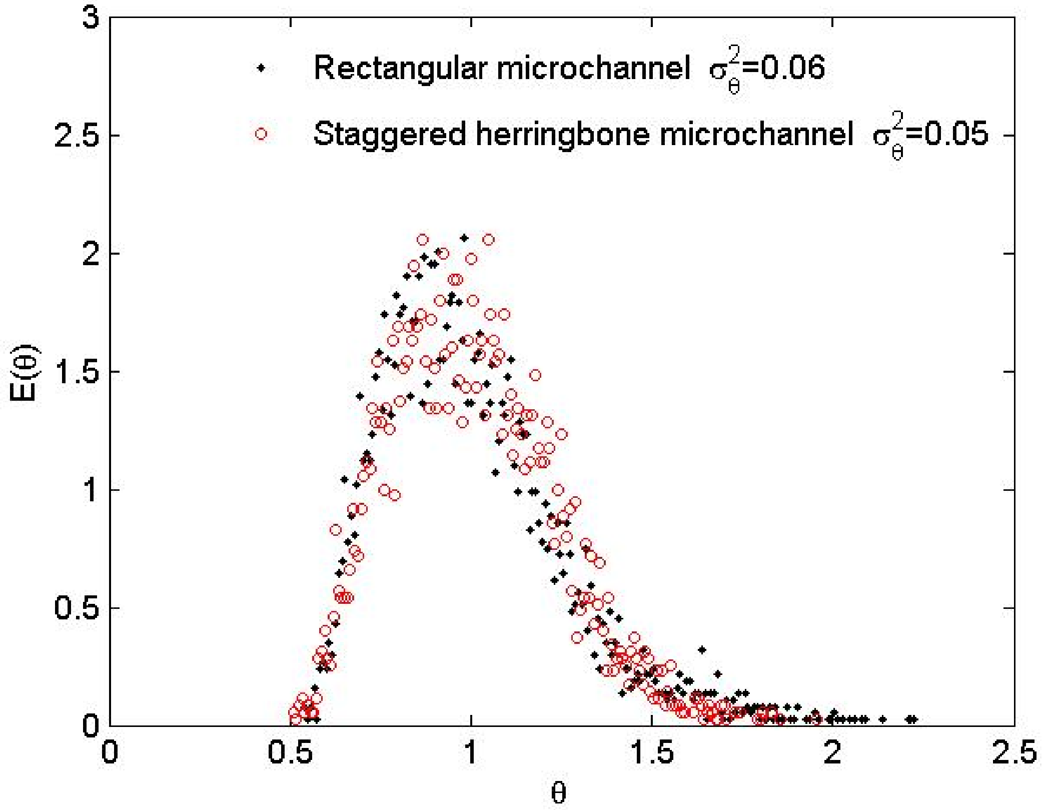




Comparison SHM and Rectangular for small Pe

•The modelling results for Pe~10^2 show that the RTD fro a plain channel and one with staggered herringbones are similar.

•As Pe decreases mass transfer by diffusion becomes important and the herringbones are no longer necessary to enhance mass transfer



Conclusions

- CFD and particle tracking simulations are found suitable to obtain mixing behaviour and RTD in microchannels.
- Staggered herringbone structures may be used to enhance mixing and narrow the RTD in a microchannel.
- For all mixing ratios studied, placing one of the fluids in the centre of the channel resulted in lower mixing lengths
- The dimensions of the channel with the SHS may be increased up to an order of magnitude compared to a standard rectangular channel without losing its performance in terms of narrow RTD. This has significant implications in terms of pressure drop and susceptibility to clogging.
- When Pe is small ($Pe<=10^2$) the use of herringbone structures will not have a strong impact neither on RTD nor on mixing.