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# CFD Modelling of Micromixing in a T-mixer with Square Bends

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**Abstract:** CFD simulations are performed to study the micro-mixing in a T-mixer with square bends by using the Villersmaux-Dushman reaction. The calculated segregation indices are compared with previously published data and it is shown that whilst the trend is captured the simulation overpredicts the extent of micromixing. This paper provides guidance on the computational requirements needed to obtain accurate results in such laminar flows and shows the complex species fields that develop within the reactor.

**Keywords:** micromixing, microchannel, CFD, segregation

## Introduction

Micromixing, i.e. mixing at the molecular-scale, controls the outcome of many chemically-reacting systems and crystallization applications [1]. In turbulent flow simulations there are now well-established approaches to take this into account via modelling of the effects of small-scale turbulence probabilistically rather than directly simulating it [2]. However, in laminar conditions the flow must be fully resolved down to the smallest scales.

Many experimental studies have been performed to characterize micromixing in various micro-scale devices using the Villersmaux-Dushman competing reactions [3,4] but the status of computational modelling of micromixing in laminar flows has been less well studied and no coherent advice is available on the best methodology to follow. Here we use the experimental data of Commenge and Falk [5] to validate a CFD model of micromixing in a T-junction with square bends and to demonstrate the complexity of this very simple system.

## Computational Model

In the following sections, we describe the reactions and reaction kinetics, the methodology for quantification of micromixing via the degree of segregation and the CFD model.

## Reactions and reaction rates

The Villersmaux-Dushman reaction system comprises parallel competing reactions composed of a neutralisation reaction (R1) and a redox reaction (R2). It was used initially to study micromixing in a stirred tank [6] and was subsequently extended to the study of continuous flow in microreactors by Falk and Commenge [7]. Details of the experimental method are given in Guichardon and Falk [8] and a detailed study of the reaction kinetics is given in Guichardon et al. [9].

Reaction (R1) is almost instantaneous whilst Reaction (R2) is very fast with a rate of the same order of magnitude as the micromixing process. The reactions used are:



It is the competition between these two reactions that allows the degree of micromixing to be determined. In addition to the above reactions, the iodine formed in reaction (R2) can react with iodide ions as follows:



where reaction (R3) is very fast compared with reaction (R2) and is therefore in equilibrium.

The literature contains a variety of reaction rates with many CFD articles giving incorrect and/or inconsistent values. Here we have adopted the following reaction set based on its consistency with the literature cited above. The rates of reaction (R1) and (R2) are given by

$$r_1 = k_1[\text{H}^+][\text{H}_2\text{BO}_3^-] \quad \text{with} \quad k_1 = 10^{11} \text{ L mol}^{-1}\text{s}^{-1} \quad (1)$$

$$r_2 = k_2[\text{H}^+]^2[\text{I}^-]^2[\text{IO}_3^-] \quad (2)$$

for which Guichardon et al. [9] determined:

$$\log_{10}k_2 = 9.28105 - 3.664\sqrt{I} \quad \text{for} \quad I < 0.166 \text{ M} \quad (3a)$$

$$\log_{10}k_2 = 8.383 - 1.5112\sqrt{I} + 0.237I \quad \text{for} \quad I > 0.166 \text{ M} \quad (3b)$$

where  $I$  is the ionic strength of the mixture given by  $I = \frac{1}{2}\sum_{i=1}^n C_i z_i^2$ , where  $z_i$  is the charge number of species  $i$ ,  $C_i$  is the molar concentration of ion  $i$  and the sum is taken over all ions in the solution [7].

For reaction R3, the equilibrium condition is expressed in terms of the equilibrium constant which is given by:

$$K_B = \frac{k_{3f}}{k_{3b}} = \frac{[\text{I}_3^-]}{[\text{I}_2][\text{I}^-]} \quad (4)$$

where Palmer et al. [10] determined the equilibrium constant as:

$$\log_{10}K_B = \frac{555}{T} + 7.355 - 2.575\log_{10}T \quad (5)$$

The value of  $K_B$  for a temperature of 20°C is 787 l mol<sup>-1</sup>, giving the following reaction rates:

$$k_{3f} = 5.9 \times 10^9 \text{ l mol}^{-1} \text{ s}^{-1} \quad \text{and} \quad k_{3b} = 7.5 \times 10^6 \text{ s}^{-1}. \quad (6)$$

### Quantification of micromixing

The above system is widely used to quantify micromixing via the use of the segregation index,  $X_S$ , which is 0 when the flow is perfectly mixed and 1 when it is completely segregated [11]. When micromixing is poor reaction (R2) is favoured, whereas when micromixing is fast almost all the  $\text{H}^+$  ions are consumed by reaction (R1), so there is little or no  $\text{I}_2$  formed. From reaction (R2), two moles of  $\text{H}^+$  are required for every mole of  $\text{I}_2$  generated. Therefore, for continuous flow mixers, the selectivity of the iodide reaction,  $Y_I$ , is defined via:

$$Y_I = \frac{2(\dot{n}_{\text{I}_2} + \dot{n}_{\text{I}_3^-})_{\text{out}}}{(\dot{n}_{\text{H}^+})_{\text{in}}} = 2 \frac{\dot{q}_{\text{out}}([\text{I}_2] + [\text{I}_3^-])_{\text{out}}}{\dot{q}_{\text{acid, in}}[\text{H}^+]_{\text{in}}} \quad (7)$$

where  $\dot{n}$  denotes the molar flow rate and  $\dot{q}$  denotes the volumetric flowrate. Note it is often implicitly assumed that the inlet flow rates of both streams are the same, in which case equation (7) reduces to:

$$Y_I = 4 \frac{([\text{I}_2] + [\text{I}_3^-])_{\text{out}}}{[\text{H}^+]_{\text{in}}} \quad (8)$$

The segregation index is given by

$$X_{ST} = Y_I/Y_{TS} \quad (9)$$

where  $Y_{TS}$  is the selectivity of iodide when there is total segregation. In this case the mixing time is very long, and the two reactions (R1) and (R2) can be assumed to be infinitely fast and the selectivity is controlled only by the relative concentrations of  $[\text{IO}_3^-]_{\text{in}}$  and  $[\text{H}_2\text{BO}_3^-]_{\text{in}}$  so that:

$$Y_{TS} = \frac{6[\text{IO}_3^-]_{\text{in}}}{6[\text{IO}_3^-]_{\text{in}} + [\text{H}_2\text{BO}_3^-]_{\text{in}}} \quad (10)$$

### Geometry and Operating Conditions

The geometry and operating conditions are chosen to replicate the study of Falk and Commenge [7]. The geometry of the mixer and operating conditions are given in Figure 1. The concentration of  $\text{I}_3^-$  is measured by spectrophotometry at 353 nm following the Beer-Lambert law:

$$[\text{I}_3^-] = \frac{OD}{\epsilon l} \quad (11)$$

where  $OD$  is the measured optical density (or light absorption),  $\varepsilon$  is the molar extinction coefficient of  $I_3^-$  ions at 353 nm and  $l$  is the optical path length, taking values of  $26047 \text{ L mol}^{-1} \text{ cm}^{-1}$  and 5mm, respectively.

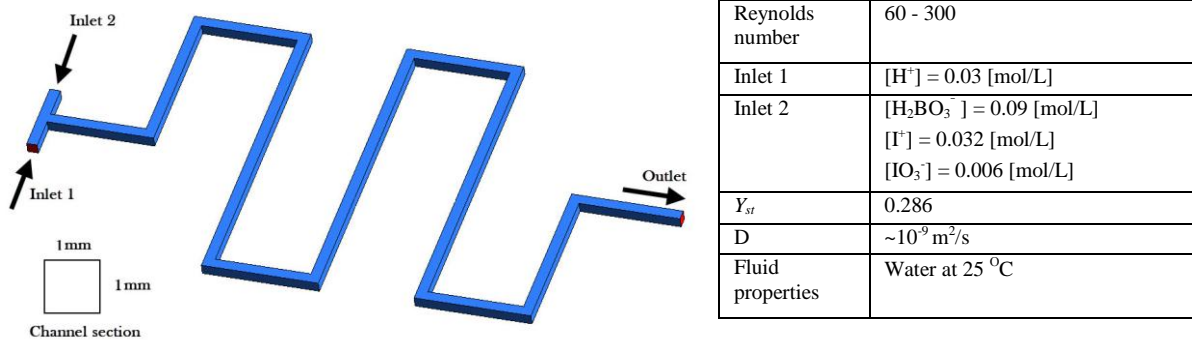


Figure 1: Geometry and operating conditions used in the study.

## CFD Model

Simulations were performed for both steady and transient, three-dimensional flow using the following equations for conservation of mass, momentum and species:

$$\nabla \cdot (\rho \mathbf{u}) = 0 \quad (11)$$

$$\frac{\partial(\rho \mathbf{u})}{\partial t} + \nabla \cdot (\rho \mathbf{u} \otimes \mathbf{u}) = -\nabla p + \nabla \cdot (\mu(\nabla \mathbf{u} + \nabla \mathbf{u}^T)) \quad (12)$$

$$\frac{\partial(\rho Y_i)}{\partial t} + \nabla \cdot (\rho \mathbf{u} - \rho D_i \nabla Y_i) = M_{w,i} \sum_{r=1}^{r=3} R_{i,r} \quad (13)$$

These equations were solved using ANSYS Fluent 19. Initial simulations were performed in unsteady mode but the flow was found to be steady and so this approach was abandoned as it offered no advantage. The equations are very stiff, so the in-built Stiff Chemistry solver with Direct Integration was employed together with the pseudo-transient solver for the mass, momentum and species equations. Typically, the pseudo-timestep used for the species equations was 1000 times larger than that used for the momentum equation. However, this approach still required more than 50,000 iterations to obtain an acceptable atomic balance over the system, running on 72 cores for the finest mesh. Second order upwind differencing was applied to the convective terms of all equations.

## Results

Simulations were made using two different hexahedral meshes in which cubic elements were employed with 20 and 40 cells across the duct. Given that the meshes are regular and all the schemes used (convection, diffusion, pressure) are second order Richardson extrapolation can be used to estimate a grid independent result. The results are shown in Figure 2 below. It is evident that mesh independence is much easier at higher Reynolds numbers, where there is more rapid mixing and reaction (R1) dominates. At the lowest Reynolds number of 60, even with the very fine mesh there is still some remaining mesh dependence. Agreement with the experimental data is good in that the trend is well predicted but there is a tendency to predict better micromixing than is observed in the experiments. It is evident that at the high Reynolds number no amount of mesh refinement is going to change this.

Figure 3 shows the exit concentrations of the I species used in the calculation of the segregation index for the extremes of the Reynolds numbers considered. At  $Re = 300$  the concentration is almost constant ( $< 0.8\%$ ), whereas for  $Re = 30$  the pattern is complex and mirrors the Dean vortices responsible for mixing.

## Conclusions

CFD simulations have been successfully performed to study micromixing in a T-mixer with square bends. The simulations are computationally very costly requiring very fine meshes and

>200,000 iterations to obtain convergence. The results capture the experimental trend but over predict the degree of micromixing. The complexity of the concentration distribution is highlighted.

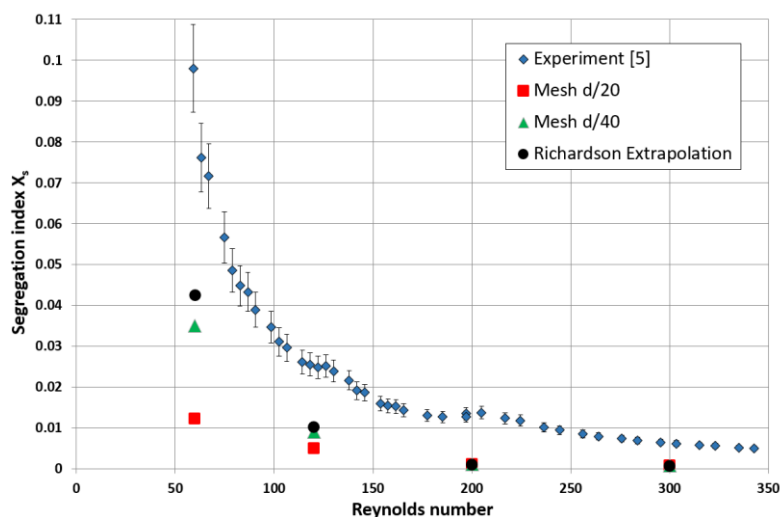


Figure 2: Comparison of the simulated segregation index with the data for concentration set 1 of Commenge and Falk [5].

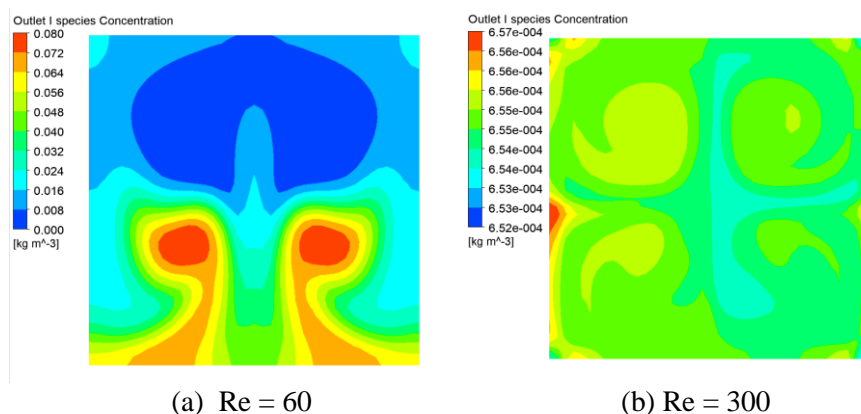


Figure 3: Exit concentrations of the I species used in equ. (8) to determine the segregation index.

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