Experimental investigation of macro- and micro-mixing in a reactive turbulent channel flow

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Abstract Turbulent mixing plays an important role in a huge number of applications, such as combustion and chemical engineering. It is becoming increasingly important to adjust much more precisely the mixing properties in such flows. For this purpose, a better understanding is needed concerning the relative influence of flow-induced macro-mixing at large scale, and of micro-mixing at small scale due to molecular diffusion. In order to quantify both mixing processes, an experimental procedure has been developed in our group that is based on simultaneous two-tracer-PLIF measurements (Lehwald et al. 2010b). This method has been first tested for laminar flow conditions in static mixers and is here used for the first time with a turbulent channel flow. The objective is to characterize and analyze flows with growing complexity, from a single static mixer element, over several turned mixer elements in a laminar flow, to the mixing behavior of a centrally injected turbulent jet in a quadratic channel. Raw and post-processed results are presented, as well as derived values like the segregation coefficient for turbulent conditions and the segregation index for laminar flow. The relationship between macro-mixing and micro-mixing has been also evaluated using joint probability density functions. These results demonstrate the applicability of the method for large-scale mixing and turbulent flows relevant for industrial applications.

1. Introduction

As laminar and turbulent mixing play an important role for a large variety of process applications, it is becoming increasingly important to adjust much more precisely the properties of mixing devices. One distinguishes micro-mixing, which is associated with molecular diffusion processes at small scales (Hjertager Osenbroch 2004), from macro-mixing, which refers to the flow mixing processes at larger scales (up to the size of the reactor cross section). For turbulent flows, “micro-mixing” would mean a typical length scale below the characteristic Batchelor scale, with “meso-mixing” between Batchelor and Kolmogorov scale and “macro-mixing” above the Kolmogorov scale (Baladyga and Bourne 1999). Micro-mixing is thus an important mechanism that directly influences fast chemical reactions and can be the limiting factor in combustion reactions or biological initiation processes, since the reactants locally come in contact with each other only through micro-mixing (Öncül et al. 2009). Moreover, in microfluidic devices and microreactors, microscale mixing is gaining more attention. In all laminar systems, as also considered in the present work, mixing is strongly associated with diffusion and thus with micro-mixing (Wheat and Posner 2009).

For numerical calculations mixing at large scales (macro-mixing) can be relatively easily described on the employed discretization grid (Lang et al. 1995), while mixing at small scales (micro-mixing) usually has to be modeled. It is therefore important to quantify both effects separately in order to develop and improve corresponding models (Schwertfirm and Manhart 2007).

In laminar flows, mixing is mostly realized thanks to static mixers. Those are important mixing tools especially for highly viscous flows (as found, e.g., in pharmaceutical products, biotechnology, food engineering, and polymer production). Related applications and methods are broadly found in
fluid mechanics and chemical engineering, but also in biotechnology and classical physics. Therefore, the literature in this field is rather broad. Recent studies using optical measurements relied in particular on Particle Image Velocimetry (PIV) or Planar Laser-Induced Fluorescence (PLIF) (Pust et al. 2006, Wadley and Dawson 2005). Mixing in gas-liquid flows has been extensively investigated using Two-Tracer LIF methods similar to that employed in Moghaddas et al. (2002, 2004), Yousefi Amiri and Moghaddas (2010), Zadghaffari et al. (2009). Different methods for a simultaneous visualization of macro-mixing and micro-mixing by planar laser-induced fluorescence (PLIF) have been tested by the authors (see Lehwald et al. 2010b). A suitable method for large flow rates and installations of industrially relevant size is based on simultaneous Two-Tracer-PLIF (2T-PLIF) measurements of a neutralization reaction. This involves the fluorescent tracer Uranine as pH-indicator for quantifying micro-mixing and simultaneously another fluorescent tracer, Pyridine 2, which is independent from the pH and characterizes macro-mixing. Details are given hereafter.

In this paper we show that this measurement technique is generally applicable to any configuration involving liquid-liquid mixing, for single-stage and multistage static mixers, from laminar to turbulent flow conditions. It can be broadly employed for a variety of further configurations, as done presently in our group.

2. Experimental technique

The method used for the simultaneous and separate characterization of macro-mixing and micro-mixing has already been described in Lehwald et al. (2010b) and is just briefly summarized here. In order to quantify micro-mixing by PLIF, a neutralization reaction involving the fluorescent tracer Uranine as pH-indicator has been identified as a suitable technique. The local pH-value is changed by the acid-base reaction between the main stream and an injected central jet, and as a consequence, the emission intensity of uranine alters. This local pH modification can be quantified by PLIF, using equal initial concentrations in both streams. It is a direct marker of the very fast acid-base reaction, and therefore indirectly of micro-mixing. Uranine and thus the pH-field are visualized simultaneously with another fluorescent tracer, Pyridine 2, which is independent from the pH-value and characterizes macro-mixing only. Both tracers are excited at the same wavelength (532 nm, doubled Nd:YAG laser) and their fluorescence signals can be clearly separated with two different optical filters (a 550 nm (10 nm FWHM) bandpath filter for Uranine, and a 705 nm (30 nm FWHM) bandpath filter for Pyridine 2). The PLIF images are acquired using two intensified CCD cameras (NanoStar, LaVision). These are placed side by side at an angle of 75° towards the measurement area, imaging the vertical center plane behind the mixer or jet inlet (Fig. 1).

The details of the optical setup, image evaluation and calibration procedure can be found in Lehwald et al. (2010b). The acquired PLIF data have been postprocessed and evaluated using the program DaVis from LaVision. The concentration calibration curves, used to transform the measured fluorescence light intensities to concentration and pH-fields, have been programmed into DaVis as a last step of the post-processing procedure.

This experimental method has been used in several flow configurations, all relying on a channel flow of 91 mm x 91 mm cross section and 300 mm measurement section length. The tracers are injected through a 13 mm x13 mm square tube, with the same velocity as the main flow. A traversing system allows the observation of the flow over a streamwise length of 300 mm, at three different positions. This measurement section is part of a bigger, gravity-driven flow installation (Fig. 1), which has been already described in detail in previous papers (Lehwald et al. 2008, Lehwald et al. 2010b). Water flows down to the test section from an upper tank, which lies roughly 10 m above the axis of the channel. The volume of this tank sets the upper limit on the total acquisition time, since any noticeable change of liquid level in the tank must be avoided in order to keep constant gravity-driven flow conditions.
At the entrance of the measurement section, an unperturbed pipe flow is observed. The test section, made of acrylic glass, has a square shape in order to increase the quality of all optical measurements by limiting refraction, laser beam divergence and image distortion. At the end of the test section, a flow meter is installed and the required flow rates are adjusted using a precision control valve. From there, the fluid goes to a sink, so that tracers do not recirculate in the installation (open loop). The static overpressure in the test section is 0.87 bar. The flowrates used for the experiments are:
- laminar case: \( Q = 185 \, \text{l/h}, v_{\text{mean}} = 6.2 \, \text{mm/s}, \text{Re}=565 \) (based on the channel cross-section);
- turbulent case: \( Q = 1310 \, \text{l/h}, v_{\text{mean}} = 44 \, \text{mm/s}, \text{Re}=4000 \).

The main flow contains the fluorescent tracer Uranine with a concentration of 1.0 mg/l at a pH-value of 8 (Fig. 2). Uranine with the same concentration (uncertainty ±2.5%) and, depending on the experimental run, a pH-value of 5.5-6.0, together with the fluorescent macro-mixing tracer Pyridine 2, at a concentration of 1.5 mg/l (same uncertainty of ±2.5%), are injected along the centerline of the channel (Fig. 2). Both streams are co-current and have the same velocity. The outlet of the L-form, central injection system has a square section of 13x13 mm² and a streamwise length of 100 mm. The temperature difference between the main fluid and the injected water/Uranine/Pyridine 2/HCl mixture is controlled by a thermostat (Haake Phoenix 2 P1-C25P), to keep it below 0.5 K. This accurate temperature control was observed to be absolutely necessary in order to avoid a rapid stratification (induced by buoyancy) between the injected mixture and the main flow.

![Fig. 1: Experimental set-up.](image1)

![Fig. 2: Schematic view of the injection configuration and of the central, vertical laser light sheet (green)](image2)
With this general fundamental setup different flow configurations have been examined. They are, in the order of growing complexity: a) a laminar, unperturbed pipe flow, b) a laminar flow with one static mixer element (SMX-type), c) a laminar flow with several mixing elements, which are turned by 90° each, d) a turbulent pipe flow without mixer. Chronologically speaking, case b) was the first to be examined and has been described in detail in Lehwald et al. (2008, 2010b), Leschka et al. (2007). All specifications concerning the mixer and its position in the above described channel can be found there. The geometry of the employed mixer mimics that of the commercially available SMX mixers, which have been characterized extensively in Zalc et al. (2002). Commercial static mixers use multiple elements in series, to take advantage of the repeated reorientation of material lines that leads to exponential stretching and mixing. That is why for case c) up to four single mixer elements, identical to those used in the previous publications, have been placed one behind the other and turned by 90° each. As it can be seen in the results section, nearly complete mixing has been already attained after three mixer elements; the low signal-to-noise ratio did not allow further useful measurements with four elements or more. Cases a) and d) are the limiting cases in this study: the first represents the simplest configuration of a reference to which all the others can be compared, the second is its complement for turbulent flow conditions, which are very often examined in mixing studies, due to huge application fields. In all cases main and secondary flows are initially injected with the same velocity.

3. Results

The following images (Fig. 3) show the Two-Tracer-LIF measurement results (top: concentration field of Pyridine 2; bottom: pH, determined from Uranine fluorescence intensity) for the laminar case a) (Re=565) over a length of 300 mm in the vertical center plane of the channel. Since the flow is laminar, it is possible to show a composite image built from three separate images in the streamwise direction, with a nearly perfect overlapping of the individual images, taken at different times. The progressive widening of the Pyridine 2 concentration and low-pH jet injected with the secondary flow along the channel centerline can be clearly seen on these images. This indicates the ongoing diffusion and chemical reaction at the interface between the main (yellow arrows) and secondary (green arrows) flow. But, laminar mixing processes are so weak on the considered length, that the Pyridine concentration, as well as the pH of the inner jet, did not change noticeably.

![Fig. 3: Measurement results for laminar flow (Re=565, case a), composed of three separate images](image-url)
The results obtained for a Reynolds number of 4000 (case d) are presented in Figure 4 for the same three measurement positions. Since the flow is now turbulent, it is impossible to combine these three separate images in a single one. The development of the turbulent vortical structures can be clearly seen on these images, as well as fast pH change (lower row) due to increased micro-mixing (and hence reaction), resulting from the much larger contact surface between main and secondary flow. The pH of the structures represented in the third image has increased from the injected pH (with a value of 5.5) to about 6.5. Additionally, these structures are smaller compared to those visible on the Pyridine 2 image, thus showing the progress of chemical reaction.

The images shown above are then postprocessed further, in order to obtain a quantitative measure for micromixing only, since the Uranine intensity (or the derived pH-value) is a result of both, macro-mixing and micro-mixing. After calculation of molecular concentration fields of H⁺-Ions, normalized non-dimensional concentrations of Pyridine 2 and H⁺-Ions are determined (Fig. 5, left and middle). The degree of deviation $\Delta (x,t)$ (Fig. 5, right) is then estimated as an indicator for micro-mixing, following: $\Delta (x,t) = 1 - \frac{\bar{c}_{Py} (x,t) - \bar{c}_{H+} (x,t)}{\bar{c}_{Py} (x,t) + \epsilon_{tol}}$, where $\epsilon_{tol}$ is a very small tolerance ($\epsilon_{tol} = 2.e^{-16}$) used to avoid division by zero (Lehwald et al. 2010a). A value of $\Delta$ equal to one means that micromixing has not started yet. On the other hand, $\Delta$ becomes equal to zero when micro-mixing is completed.

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**Fig. 4:** Measurement results for turbulent jet flow (Re=4000, case d) at the three measurement positions from 0 to 300 mm after secondary flow injection.  
Upper row: Pyridine 2 concentration; lower row: pH, determined from Uranine fluorescence

**Fig. 5:** normalized concentrations of pyridine 2 (left), H⁺-Ions (middle) and as micro-mixing indicator the degree of deviation $\Delta$ (right) for case d) Re=4000 at the third position
For a quantitative analysis of the mixing efficiency, the segregation index proposed by Danckwerts (1952) and based on the normalized concentration variance is calculated for the laminar flow. The segregation coefficient (Warhaf 2000) is used instead for the turbulent case. Figure 6 clearly shows the very slow progress of mixing for the laminar jet conditions (left, case a) compared to the fast mixing obtained for the turbulent jet flow (right, case d). These curves also show the non negligible influence of three dimensional structures, which transport tracer to the measurement plane and may lead to a local increase of the segregation index, visible through the unphysical “hills” in the curves. This effect is naturally more intense in the turbulent case, which is strongly associated with three-dimensional structures.

Previous similar mixing characteristic measurements with a static mixer (case b) have already shown this effect. It is clearly observed when changing the measurement plane from the vertical center plane (Fig. 7, left) to the horizontal center plane of the channel (Fig. 7, right). The corresponding segregation index (Fig. 8) shows strong local peaks, which are due to the transport of mass from the third dimension into the image plane. This effect becomes even more obvious from the calculated mean concentrations of the tracer, which are represented by dotted curves in figure 8. They show a concentration increase at the same time as the segregation index increases and vice versa. Theoretically, if no 3-d effects would exist, these concentrations should be constant over the 2-d slices of the image.

Fig.6: Segregation index of macro-mixing calculated for the laminar (left, case a) and turbulent (right) case d) in the third position

Fig.7: Pyridine 2 concentration after a static mixer at the same position (case b), left : vertical center plane, right : horizontal center plane.
In real industrial applications, several mixing segments are combined together, thus inducing a repeated reorientation of material lines that leads to exponential stretching and mixing. Usually, these mixing elements are turned by 90° each, which leads to a flow with marked 3-d structures, leading to a fast homogenization of the whole volume. Such flows have been examined in case c). Figure 9 presents the measurement results for the Pyridine 2 concentration (upper row), the pH, derived from the Uranine fluorescence (middle row), and the calculated $\Delta$-values, corresponding to the degree of micro-mixing (lower row). They have been acquired in the vertical center plane of the channel, directly after the mixer outlet, for a static mixer consisting of one, two or three elements (from left to right). The huge increase in mixing efficiency, induced by the addition of mixing elements is obvious from these images (pay attention to the different scales).

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<th>Elements</th>
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<td><img src="image2.png" alt="Image" /></td>
<td><img src="image3.png" alt="Image" /></td>
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<tr>
<td>pH (Uranine)</td>
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<td><img src="image5.png" alt="Image" /></td>
<td><img src="image6.png" alt="Image" /></td>
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<tr>
<td>delta</td>
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**Fig. 9:** Pyridine 2 concentration, pH and $\Delta$-values measured directly after the outlet of a mixer, consisting of one, two or three elements (case c). Different scales have been applied for a stronger contrast of the Pyridine 2 and pH images.
From the Δ-images it can be seen, that micro-mixing is nearly completed directly after two or three elements. Only very few islands remain in the pictures, but even those with a very low Δ-value. Macro-mixing has also diminished strongly from the injection concentration of 1.5 mg/l to maximum values of 0.3 mg/l (5 times less than the initial value) after three mixing elements. This effect can also be seen on figure 10, where the probability density functions of macro-mixing (normalized Pyridine 2 concentration) and micro-mixing (Δ-values) are plotted against each other for the three mixing elements and at three different distances from the mixer outlet (0-100 mm, 100-200 mm and 200-300 mm). Completed mixing is represented by values near 0 for both mixing types (lower left corner). Values near 1 (upper right corner) correspond to the pure secondary flow, before mixing.

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<th>elements position</th>
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Fig. 10: Probability density functions between macro-mixing (normalized Pyridine 2 concentration) and micro-mixing (Δ-values) for one, two and three consecutive mixing elements (case c) at three different distances from the mixer outlet.

The growing number of values found in the lower left corner with increasing distance from the mixer as well as with an increasing number of mixing elements is obvious (please note the log color scale). Blue pixels correspond to 1 or a few realizations; deep red squares correspond to a million of events. Clearly, the red zone observed in the lower left corner grows rapidly from left to right and top to bottom, illustrating the progress of mixing. At the same time, the red points near the lower right corner correspond to the initial state of the main flow (before mixing), which is still observed in the outer regions of the channel.
4. Conclusions

Thanks to two-tracer-PLIF measurements, it is possible to investigate and quantify simultaneously macro-mixing and micro-mixing in large-scale mixing flows with a high geometrical complexity, both for laminar and turbulent conditions. The mixing characteristics can be analyzed in details and the relationship between macro-mixing and micro-mixing can be examined quantitatively using this technique. Mixing efficiency, as needed for industrial applications, can be measured in terms of segregation coefficient/index or mixing length/time. The developed method can be identically applied for laminar as well as for turbulent flows, for static mixers as well as for jet mixing flows. However, three-dimensional flow structures lead to experimental issues and may strongly impact the observations. Thus three-dimensional (stereoscopic or tomographic) measurement techniques become more and more important for a fully quantitative analysis.

5. Acknowledgements

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Literature