Laser optical Measurements and numerical Investigations of Macro- and Micromixing in stirred Vessels

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Introduction

Today, increasing energy costs require effective processes, especially in chemical and pharmaceutical industries. Particularly, energy-intensive mixing processes offer high potential for optimization. Despite great efforts of research mixing of liquid-liquid components is not completely predictable. To construct mixing systems, empiricism is used rather than basic knowledge. Local mixing quality is a crucial parameter influencing the economics of such processes. Because of this interaction the interest of investigations concentrates on the measure of local mixing quality in regard to the macroscopic energy inserted mostly by stirrers. In case of a superimposed chemical reaction, complete mixing on molecular scale is required. The prediction of the local composition on molecular scale of a multicomponent system is not possible for complex geometries in mixing processes. A review of available models for mixing processes is given by Baldyga and Bourne (1999). The interaction between stretching, diffusion and reaction at small scales is described by Ottino (1994) in a lamellar model. Muzzio et al. (2003, 2002) extended a one-dimensional model to the complex flow in stirred tanks. Anderson et al. (2002) presented an extended mapping technique for chaotic flows in a journal bearing flow. Gollub et al. (2003) experimentally determined the mixing for a two-dimensional time-periodic flow exhibiting chaotic mixing. Despite the progress in predicting reactive mixing there is still a need for experiments, visualizing the local distribution of inert and reactive tracers with high spatial and temporal resolution. Laser induced fluorescence has proven to be a suitable measurement technique by Kling and Mewes (2003). The visualization of passive scalars indicates the convective mixing process (Villermaux et al. (1996), Distelhoff and Marquis (2000), Guillard et al. (2000)). In viscous mixing applications stretching and folding occur simultaneously at different rates in each portion of the flow, creating complex, layered structures. A seminal paper concerning the concept of chaotic advection has been published by Aref (1984). Reactive tracers which are pH-sensitive are used for measuring the micromixing indirectly by Bellerose and Rogers (1994) and Hong et al. (2002). In T-shaped micro-mixers the transport phenomena on different scales are investigated by means of pH-sensitive tracers using scanning methods by Hoffman et al. (2006). Van Vliet et al. (2000) used also a laser scanning method for investigating three dimensional concentration fields.

In order to visualize local compositions and dissipations the described research project comprises two laser optical measurement techniques with high spatial and temporal resolution. These are published by Faes and Glasmacher (2007, 2008). An investigation of local mixing quality, local energy dissipation and their correlation is performed. Therefore, the 4D-LIF technique is developed for analyzing the concentration field in dependence of time and a PIV
Experimental Set-Up

The laser optical experiments are performed in a cylindrical glass vessel, which has an inner diameter of $d_e = 90$ mm. For the investigations a multi-stage pitched blade stirrer is used. A rectangular viewbox filled with pure glycerol and positioned round the mixing system allows an iso-optical study and minimizes optical distortions. The working fluid is a mixture consisting of pure glycerol and a solution of calcium chloride. It is possible to measure velocity and concentration fields throughout the whole mixing system. The viscosity of the glycerol mixture is $0.93$ Pas measured with Rheometrics RFS II at room temperature. The experimental set-up, consisting of the two optical measurement techniques, is schematically depicted in figure 1.

The particle image velocimetry, a LaVision system, is used to measure local velocity vectors. The laser is a double pulsed NewWave® Nd:YAG laser ($\lambda = 532$ nm) with a frequency of $10$ Hz. The illuminated laser plane can be adjusted freely by an automated linear positioning system. The illuminated horizontal laser plane is projected to the CCD-camera by means of a mirror under $45^\circ$, which is placed below the viewbox. The camera and laser control, data acquisition and processing is performed with a software package DaVis® from LaVision. For measuring the velocity field (PIV), the particle paths are visualized by seeding the working fluid with hollow glass spheres of mean diameter of $10$ µm. The reflected signals of the glass spheres are captured temporally with a CCD-camera, Imager Intense 1376 x 1040 pixel. For the image processing a cross-correlation algorithm with an interrogation window of $32 \times 32$ pixel with $50\%$ overlap is selected. In each case 200-500 double images are captured. The second non-intrusive optical laser measurement technique is the 4D two-color laser induced fluorescence technique. It is used to measure the concentration fields of two fluorescent dyes simultaneously as described by Kling (2004). The laser is a NewWave® Nd:YAG ($\lambda = 495$ nm), Tempest 30 with GWU OPO® VisIr. The CCD-camera, Imager 3 (640 x 480 pixel) and Image Intensifier delivered by LaVision, is equipped with a so-called Double-Image Optics (DIO), which permits to capture the same display window twice at the same time. The optical DIO consists of two apertures, which are equipped with a set of adjustable and fixed mirrors, and two optical filters (BP523/10 and RG645) for separating the fluorescent lights of the dyes. For the multi-stage pitched blade stirrer a volume of 2 ml is injected within 10 s. The illuminated laser planes have a thickness of $500$ µm. For receiving three-dimensional images in dependence of time (4D) the cameras, the mixing vessel and the light sheet optics are mounted onto a linear positioning system. In order to measure the concentration field, the stirrer is stopped. The three-dimensional images are reconstructed out of 170 two-dimensional plane images, captured in a distance of $500$ µm, using Imaris®-Software from Bitplane. Each two-dimensional plane image is an averaged plane image out of 16 single plane images captured in one second. The scanning procedure from the bottom of the mixing vessel to the free surface of the working fluid takes 10 minutes including post processing. The laser measurement technique of PIV and 4D-LIF are used sequentially.
Macro- and Micromixing

The convection, namely the macromixing, is visualized by an inert fluorescent dye (carboxy-SNARF). The transport on molecular scale is visualized by a fluorescent dye (fluoro-4) reacting with calcium ions dissolved in the working fluid. The fluorescent emission characteristic of this dye changes due to a chemical reaction with calcium ions. The progress of the chemical reaction shows indirectly the micromixing, because mixing on molecular scale is required for the progress of chemical reactions. After a mixture of both dyes is injected into the mixing system, the concentration fields of the inert and reactive dyes are measured. The fluorescence intensity of the dyes are calculated by Lambert-Beer’s law. The quantum yield $\Phi$ describes the effectiveness of the fluorescent emission $I_F$:

$$I_F = \Phi I_0 \exp(-\varepsilon sc)$$  \hspace{1cm} (1)

$I_0$ is the intensity of the exciting light, $\varepsilon$ is the molar extinction coefficient and $s$ is the length of the measurement volume. A simplification by series expansion can be done for small concentrations so that $I_F$ depends linearly on the concentration $c$ of the dye:

$$I_F = \Phi I_0 K \varepsilon sc = mc$$  \hspace{1cm} (2)

$K$ is a parameter depending on the measurement system, considering for example the viewing angle of the detector. For constant parameters, $m$ is a calibration factor. This allows a correlation of local dye concentrations and measured intensities. More details, especially the calibration of the measurement system and the correction of the shot-to-shot noise of the laser, are described in Kling and Mewes (2004).

The local degree of deviation is a quantitative measure for the quality of micromixing. It is defined as
\[
\Delta(\bar{x},t) = 1 - \frac{c_{1,\text{react}}(\bar{x},t)}{c_1(\bar{x},t)}
\] (3)

and can be calculated by comparing the local concentration of the reaction product \(c_{1,\text{react}}\) with the virtual concentration \(c_1\) of the reacting dye locally appearing without any reaction. Due to the injection as a mixture and transportation in the same manner the local concentration of the reactive dye \(c_1\) without chemical reaction can be calculated by the knowledge of concentration \(c_2\) of the inert dye and the initial concentration ratio to

\[
c_1(\bar{x},t) = c_2(\bar{x},t) \frac{c_{1,0}}{c_{2,0}}.
\] (4)

The local degree of deviation is the fraction of the reacting dye which has not reacted yet. For a value of the local degree of deviation \(\Delta = 1\) the mixture is completely segregated on macroscopic scale. During the mixing process a homogenous fluid on microscopic scale is achieved and the local degree of deviation decreases to \(\Delta = 0\) for a completely micromixed fluid.

**Numerical Methods**

For mixing processes it is desirable to have local information about concentration and velocity fields to optimize the mixing geometry or the position of the feeding point. For a theoretical prediction of local mixing quality it is necessary to choose the correct physical models and boundary conditions. In this work, the data of the numerical calculations are validated by the experimental investigations. The numerical calculations are performed with a commercial CFD code by ANSYS. The geometrical models are equivalent to the glass mixing vessel equipped with the full-glass stirrer in the experimental set-up. The meshes of the stirrer configuration consist of 6.0 million finite volumes for the multi-stage pitched blade stirrer. The validated results serve for the reduction of experiments and their high costs.

**Results and Discussion**

**Multi-stage pitched blade stirrer**

The speed of each stirrer is adjusted due to the chosen Reynolds numbers for laminar conditions. The mixing vessel is filled with 500 ml of the working fluid. Due to a complete field of viewing of the uniform secondary flow field, the distance from the bottom of the mixing vessel to the centre of the lower blade of multi-stage pitched blade stirrer is set to 25 mm as depicted in figure 2. The distance between the centers of the two stages is 30 mm. The multi-stage pitched blade stirrer induces a complex three-dimensional flow field with three torus-shaped eddies. The structure of the eddies depends of the rotational speed of the stirrer. Here, three different injection positions are analyzed, which are depicted in figure 2 as A, B, and C. The progress of macro- and micromixing is depicted in figure 3 for injection position B.
Figure 2: Schematic of the multi-stage pitched blade stirrer (a), and of the three injection positions A, B, and C (b)

Figure 3: Progress of macro- and micromixing for a multi-stage pitched blade stirrer, Re = 3, injection position B
After the injection of the dye mixture, lamellar structures are formed and transported upwards before they reach the two stages of the downward-downward pumping stirrer. The striation thickness of the lamellas decreases due to the convective transport of the volume. The progress of the micromixing proceeds simultaneously. The local degree of deviation decreases from the starting value of 1, which is indicated by red color. The dye volume transported into high regions of dissipation near the blades of the stirrer is micromixed well. The local degree of deviation decreases. A quantitative analysis is given during the oral presentation.

**Conclusion**

Macro- and micromixing in laminar flow regime is analyzed by experimental and numerical methods. The quality of the mixing process is described by the local degree of deviation, calculated by the local concentrations of an inert and a reactive dye. The progress of mixing is observed as a function of time for different injection positions and Reynolds numbers. In a complex flow field, induced by a multi-stage pitched blade stirrer, local concentrations are measured and visualized four-dimensionally. Mixing on molecular scale depends mainly on the flow path through the mixing vessel and the inserted local energy dissipation. Micromixing starts in the boundary layers of the lamellas, indicated by a decreasing local degree of deviation. The developed numerical model provides the prediction of macro- and micromixing. The numerical calculations show good accordance with the experimental investigations. The developed methods and investigations in this project facilitate the understanding of transport phenomena in liquid-liquid mixing systems for laminar conditions.

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**References**

Villermaux, J. et al. (1996), "Planar laser induced fluorescence technique for measurements of concentration fields in continuous stirred tank reactors”, Experiments in Fluids, 22, 95-102