Experimental Investigation of Mixing in a rectangular cross-section Micromixer

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μ - Laser-induced Fluorescence (μ-LIF)

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Summary

In this paper we explain experimental investigations that analyze local velocity fields and concentration fields. We show that steep gradients occur which are dependant on micromixer geometry and entry conditions. These concentration gradients are used to determine the phenomenology of mixing, mass transfer and reaction. Additionally we show that under laminar conditions a complete mixing of two liquids is possible as long as the diffusion distance of the species is shortened sufficiently through diverse convolutions. The results obtained from these experiments allow a more profound understanding of the mass transfer processes in micro-mixing and a improvement in the design rules for static micromixers.

Introduction

The constantly growing demand for efficient, resource saving and environmentally friendly processes has led to a fast development of new methods in terms of intensifying of processes in recent years. Microreaction technology has gained special importance through the easy transferability from laboratory scale to industrial applications because of shortened development periods. Especially areas of great potential for innovation, e.g. nanotechnology and biotechnology, could benefit from microreaction technology. Micromixers are central components of microfluidic systems for chemical and biochemical applications. The fluid flow in tubes and channels on a microscale has emerged as an important area of research. This has been motivated by their use in such diverse applications as micromixers for controlled mixing of reactants in pharmacy or microreactors for unique process regimes and for optimizing conventional processes.
Common passive micromixers work in a laminar flow regime by splitting the fluid into small flow filaments to mix them by recombination and diffusion. This multilamination typically requires complicated three dimensional channel geometries. In our research we use a T-shaped micromixer with a rectangular cross section. This kind of mixer uses the flow-induced creation of secondary vortices inside the micro channels to enhance the mixing performance. Up until now investigations in micromixing have consisted largely of bulk measurements, such as volume flow rates, overall mixing qualities and pressure losses.
For modern technical applications e.g. reactions with high selectivity, a precise analysis of local mass transfer and hydrodynamics for different flow regimes (strictly laminar flow, vortex flow) requires microscope-based measurements of the flow and concentration fields. In spite of microreaction technology offering diverse possibilities to optimize processes with highly selective reactions this capability remains largely unused.
Because of a lack of fundamental knowledge, microreactors are mostly laid out according to “trial-and-error”. In the context of the priority program 1141 of the German Research Foundation (DFG) “Analysis, modeling and calculation of mixing processes with and without chemical reaction” within the scope of a joint research project intensive experimental, analytical and numerical investigations are being performed over the last two years. Part of this joint research project are notably the Institute of Microsystem Technology, Albert-Ludwigs-University of Freiburg, chair of “Design of Microsystems” and the ”Department of Chemistry and Chemical Engineering”, University of Paderborn, Germany. These investigations aim at understanding the phenomenology of micromixing using T-shaped micromixers as an example.

In the last two decades Laser induced fluorescence has established itself as the most important measuring technology for the non-invasive determination of local concentration field

in order to understand mixing phenomenology [1, 2, 3]. A common dye used is Rhodamine B [4]. For examination of micromixing an epi-fluorescent-microscope is used. Because of volume illumination the two dimensional picture contains multiple layers. In order to resolve three dimensional structures that are formed in the mixing process a confocal Laser Scanning Microscopy has proven to be useful [5, 6].

Because the measuring of concentration fields alone does not allow a statement concerning the convective mass flux, a detailed knowledge of the velocity field for the modeling of the mixing process is essential. In recent years Particle Image Velocimetry (PIV) has become a standard procedure to visualize velocity fields. Lately PIV was widely used in microchannels [7, 8]. Besides experimental analysis of velocity fields additional information concerning the shear stress can be extracted from the PIV data.

Experimental setup

The experimental investigation of velocity fields and concentration in micromixers is performed using the experimental setup in figure 1.
While an epifluorescent microscope is used for µ-PIV, a confocal LSM is used for µ-LIF.
The liquids (tempered at 20°C) are delivered out of the pressure containers without pulsation. The mass flow is measured by mass flow meter (Bronkhorst High-Tech BV.).
The T-mixers with different geometries are provided by our partner, the Institute of Microsystem Technology. The notation for the T-shaped micromixers is as follows: width of the mixing channel x width of the inlet channel x height of all channels.

In the µ-PIV process (Co. ILA GmbH, Jülich) fluids are marked with fluorescent nano particles coated with Rhodamine B (polystryrol, particle diameter 500 nm, micro Particles GmbH, Berlin) that follow the liquid stream virtually without inertia. Through two consecutive laser pulses (wavelength 532 nm; pulse width 5 ns, New Wave Research, Inc.) with a defined pulse distance (1 µs) it is possible to determine the local velocity and direction. As a PIV-camera we use a CCD camera (PCO Sensicam QE).
The depth of the measurement plane is defined by the focusing characteristics of the recording optics and is approximately 13 µm.

For measuring the concentration field the experimental setup is extended with a confocal Laser Scanning Microscope (LSM 410, Co. Carl Zeiss) which enables the measurement of single slices (calculated optical slice thickness approx.: 9,6 µm) along the depth of the micromixer. The beam path in the confocal laser scanning microscope is pictured in figure 2.

The enormous advantage of confocal microscopy is the possibility to collect light solely from a single plane. A pinhole sitting conjugated to the focal plane (i.e.confocal) keeps light from the detector that is reflected/emitted from others than the focal plane. The laser scanning microscope scans the sample sequentially point by point and line by line and assembles the pixel information in one image. That way optical slices of the sample are imaged with high contrast and high resolution in all three dimensions. By moving the focus plane single images (i.e. optical slices) can be put together to build up a three dimensional stack that can be digitally postprocessed [9].

For this purpose one inlet stream contains the fluorescent dye Rhodamine B (dissolved in de-ionized water). For small concentrations, i.e. 1 µM, the intensity of the emitted light by the fluorescent dye Rhodamine B is linearly dependent on the concentration of the dye.

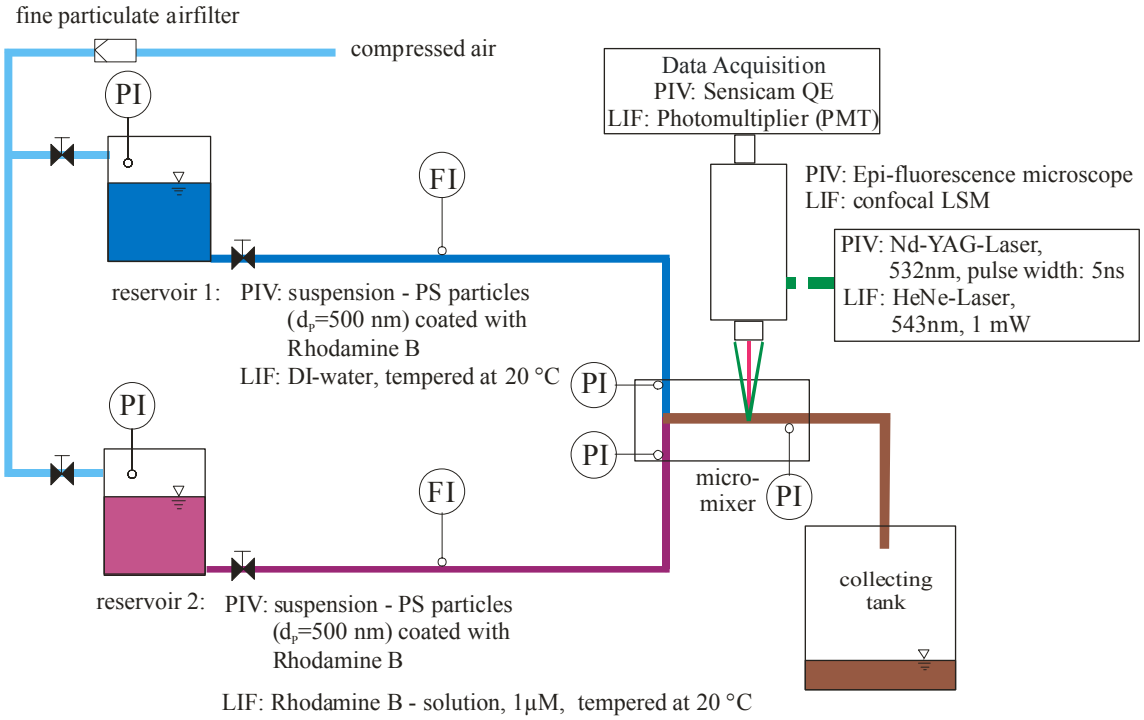

Fig. 1: Experimental setup for µ-PIV and µ-LIF measurements

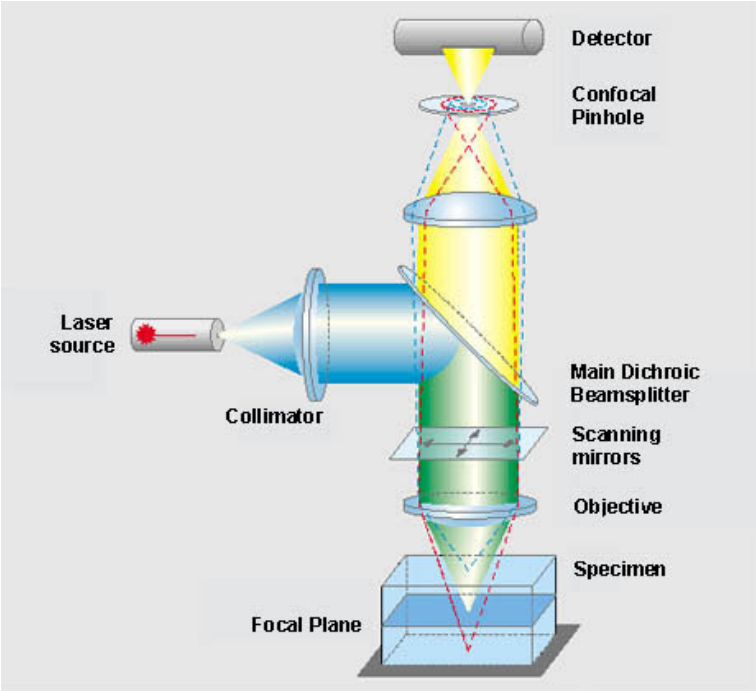

Fig. 2: Beam path in the confocal laser scanning microscope [9]

Experimental method

The point of interest is to collect new precise criteria for characterization of the performance of static mixers at the microscale. Conventional criteria of mixing quality contain only integral and time-averaged data. Due to a lack of measurement technology the essential micromixing and contact time, especially for highly selective reactions, cannot be determined.
Bothe et al. show the influence of local structures on contact area between the species which has an impact on the final step in the mixing process, i.e. diffusion.
For an experimental analysis and classification of these local structures mixer geometries and mass flow rates are systematically varied and concentration and velocity fields at defined positions are measured.
The µ-PIV data (velocity field/shearstress) is processed by means of the Image Processing Toolbox of Matlab® 7 (The MathWorks, Inc.). The concentration, the standard deviation and the local concentration gradient are derived from the grey scale values.

Results and discussion

Figure 3 exemplifies the results of a µ-PIV investigation of the mixing zone in the depth of 50 µm (geometry 400x200x100 µm). The different shear stresses emphasise the existence of different velocity gradients in the micromixer.
Furthermore the asymmetrical velocity distribution along the z-axis indicates the three dimensional velocity structure.

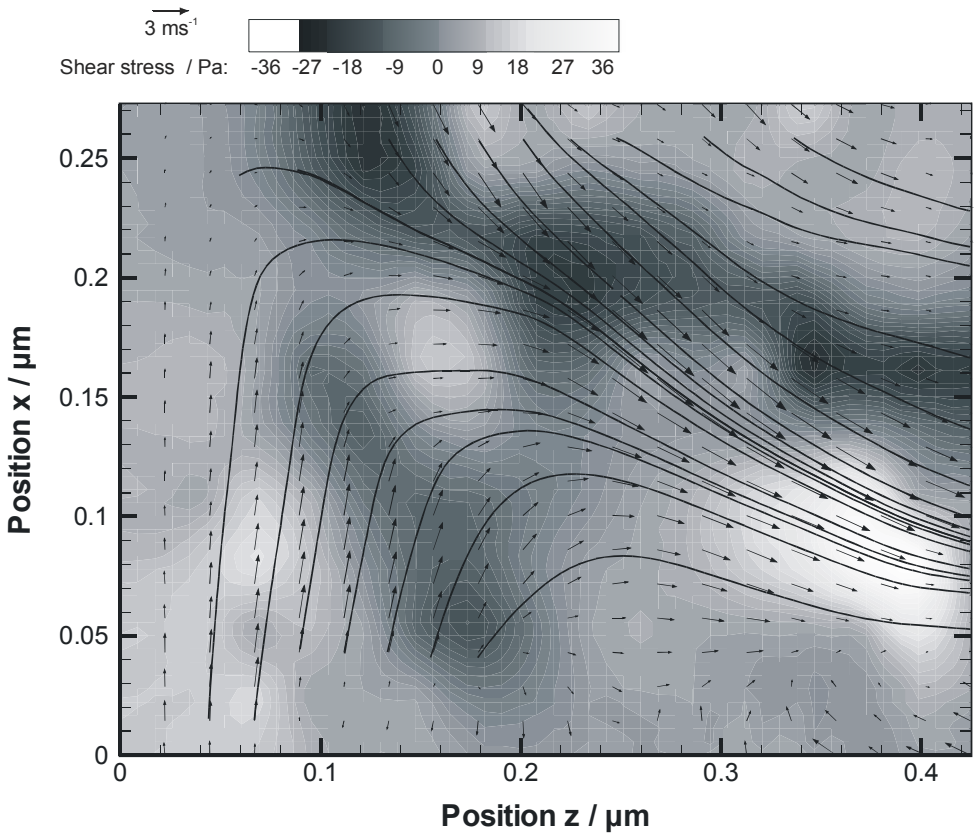

Fig. 3: Velocity field and shear stress in a micromixer - geometry 400x200x100 µm

Figure 4 shows three layers along the depth of the mixing channel which contain the velocity information. This is the basis for the “Residence Time Distribution” (RTD).

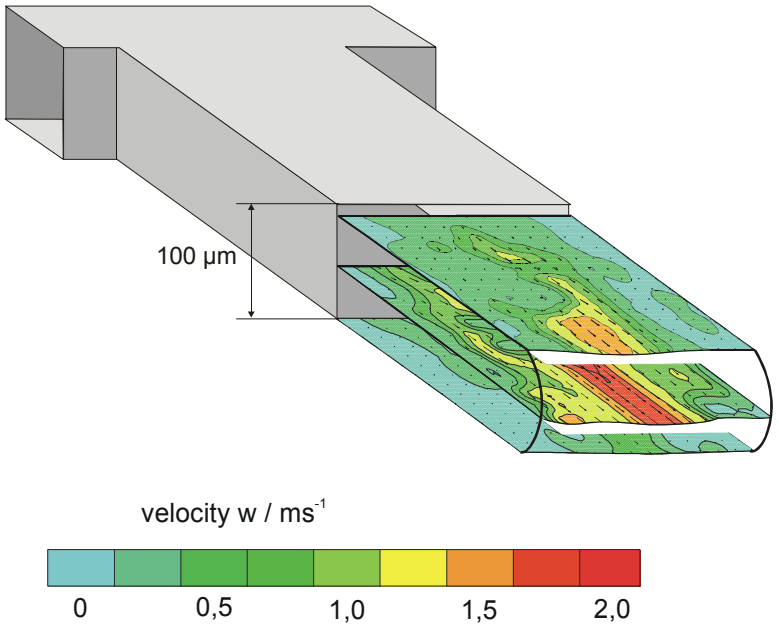

Fig. 4: Velocity field in 3 different depths - micromixer geometry 400x200x100 µm

The three dimensional structures clarify the concentration fields in figure 5 which are visualized by means of the confocal Laser Scanning Microscope. In this case the micromixer geometry is 200x100x100 µm. The Re number, calculated according to equation 1, is 186.

$$\mathrm{Re} = \frac{u \cdot d_h}{\nu} \qquad (1)$$

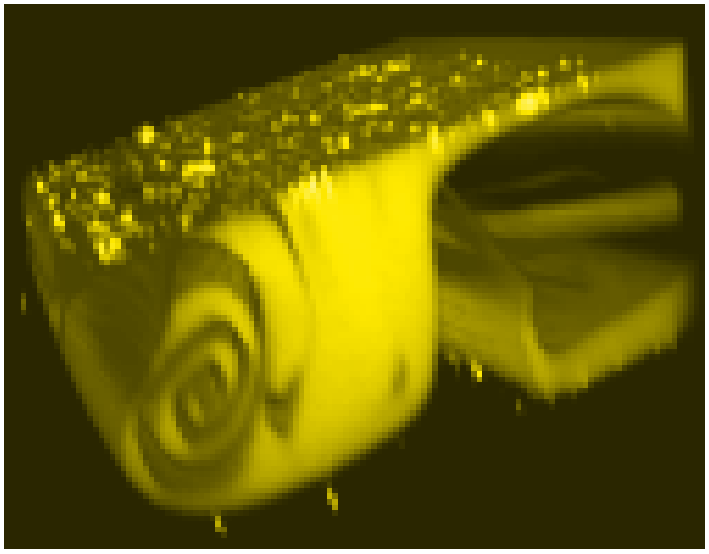

Fig. 5: Three dimensional concentration field inside a micromixer

Figure 6 shows the concentration field inside a T-shaped micromixer at a depth of 50 µm for two different Re-numbers, Re=120 (left side) and Re=240 (right side), view from above.

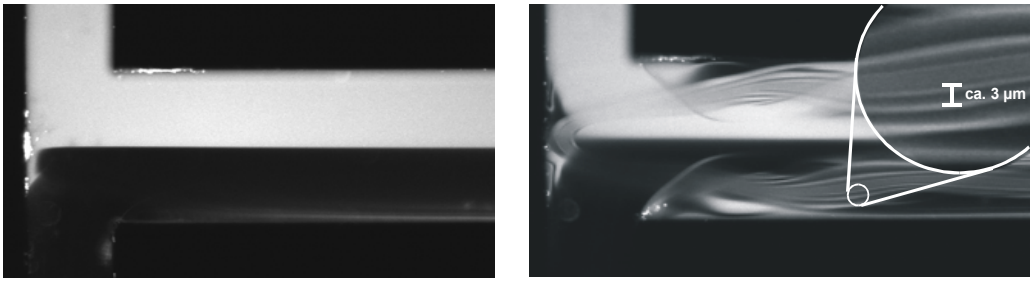

Fig. 6: T-micromixer (200x100x100 µm), system: water/Rhodamine B,
temperature: T=20°C, (left Re=120, right Re=240)

These pictures confirm the different flow regimes that are characterised and calculated by means of CFD in Bothe et al. [10] and Kockmann et al. [11].

In the strictly laminar flow regime (figure 6, left) both streams meet at the mixing zone and flow without convective mass transfer along the mixing channel. For higher Re-numbers the "engulfment regime" occurs in which two vortices develop asymmetrical to the main axis (fig. 6, right). In this flow regime fluid elements from one side reach the opposite side where a "multilamination" takes place through the resulting engulfment (distance of the finest structures approx. 3 µm).
This enlarges the contact area for diffusion so that in this flow regime a good mixing performance can be expected.

For characterization of the mixing results the "potential for diffusive mixing" Φ, equation 3, is used, see Bothe et al. [10]. The mixing quality is defined by eq. 2, where σ^2_{max} is the maximum variance of the mixture and σ^2_M is the variance of the mixture inside the cross sectional plan.

$$\alpha = 1 - \sqrt{\frac{\sigma_M^2}{\sigma_{max}^2}} \tag{2}$$

$$\Phi = \frac{dxdy}{A} \sum_{i=2}^{i\,max-1} \sum_{j=2}^{j\,max-1} \left(\frac{\left| c\left(x_{i+1}, y_j\right) - c\left(x_{i-1}, y_j\right) \right|^2}{4dx^2} + \frac{\left| c\left(x_i, y_{j+1}\right) - c\left(x_i, y_{j-1}\right) \right|^2}{4dy^2} \right)^{1/2} \tag{3}$$

The concentration gradient and mixing quality are calculated by means of the grey value distribution taken from the µ-LIF pictures.

With increasing Re-number the contact area increases as well, as seen in figure 6 (view from above).

Furthermore the meaning of Φ is explained by its dependence on the position z along the mixing channel. The greatest increase occurs in the area of convergence of the two streams, see fig. 7.

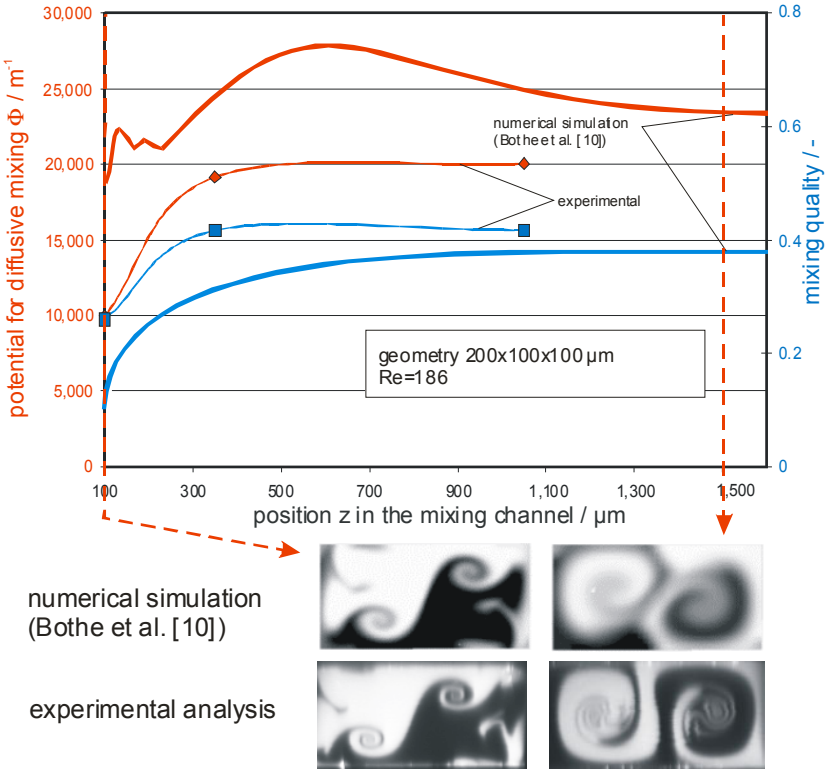

Fig. 7: Potential for diffusive mixing Φ and mixing quality α against position z in the mixing channel, Re = 186, micromixer geometry: 200x100x100 µm; corresponding cross section areas (numerical simulation and experimental analysis)

At a position of 3 mixing channel widths no noticeable increase is seen. The reconstruction of the vertical profiles shows that along the mixing channel there is an increasing engulfment with a negligible increase in contact area.

When comparing these results with commercial CFD-applications, see figure 8 for a comparison between numerical simulation [10] and experimental analysis. It is obvious that fine structures (size approx. 3µm) cannot be displayed with commercially available CFD software. This makes validation of the CFD results indispensable.

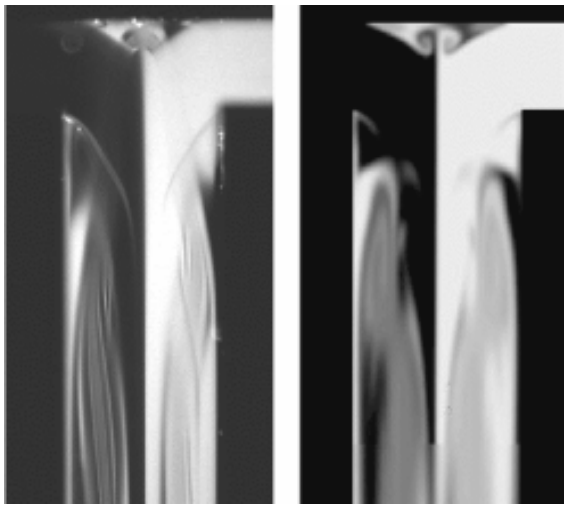

Fig. 8: Experimental (left side) and numerical analysis (right side) of a concentration field (micromixer geometry 200x100x100 µm; Re=186); y=50 µm (height 100 µm),
numerical simulation: from Bothe et al. [10]

The presented results show that in T-shaped micromixers through simple hydrodynamic mixing very fine structures (approx. 3µm) can be achieved even under laminar flow conditions.
Thus large contact areas between the reactants can be reached. With very narrow RTD, these aid the achievement of specified (defined) contact time and reaction time.
Through the knowledge of the specific influence of flow-structures and the specific contact area, parallel-consecutive reactions should be able to be reached in microreactors with greatly increased selectivity.
The prediction of velocity and concentration fields is already possible with acceptable accuracy, as shown by the results of the numerical simulation and the experimental analysis.

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Nomenclature

A	[m²]	cross section
c	[kgkg^{-1}]	dimensionless concentration of a species
d_h	[m]	hydraulic diameter
i,j	[-]	control variables
Re	[-]	Reynolds-number (Re= u d_h / ν)
T	[°C]	temperature
u	[ms^{-1}]	velocity
x, y	[m]	coordinates in the Cartesian coordinate system
α	[-]	mixing quality

η	[Pas]	dynamic viscosity
λ	[nm]	wavelength
ν	[$m^2 s^{-1}$]	kinematic viscosity
σ	[-]	variance of mixture
τ	[Pa]	shear stress
Φ	[m^2/m^3]	potential for diffusive mixing

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