3D VISUALISATION OF MIXING IN A TUBULAR REACTOR

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Master's thesis

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Abstract

A laser induced fluorescence technique was used to study the mixing behaviour of a passive scalar in a tubular reactor at Re=2,800. In the experiment, a fluorescent dye was injected in the flow system. The mixing process was visualised by exciting the dye molecules with a laser sheet, meanwhile capturing images with a CCD camera at a frame rate of 1000 fps. By sweeping the laser sheet parallel to itself, a 3D realisation of the concentration field could be reconstructed. Based on many realizations of the scalar concentration field, statistical parameters were derived at three different positions in the reactor. An assessment was done to prove that the setup resolves the micro scale flow structures in both space and time.

Studies on the turbulent structures and the mixing process have been performed. Concentration probability density functions were calculated on basis of the measurements. The PDFs showed that the concentration decreases in the downstream direction. Power spectrum analyses were performed to identify different flow scales. It was found that in the downstream direction the structures become smaller. The scalar energy dissipation has been calculated from the full scalar gradient field vector components. The highest values for the scalar energy dissipation were found at a small distance downstream the injection point. This was a result of sharply edged structures with high concentrations occurring at this position. The largest amount of scalar energy dissipation takes place further downstream in the reactor, as a result of the creation of layer-like structures. Finally, a study of isotropy on the calculated 3D scalar gradient field has been performed. Anisotropy was found at all measured positions.
# List of symbols

## Normal symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unity</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A$</td>
<td>fraction of available light</td>
<td></td>
</tr>
<tr>
<td>$c$</td>
<td>fluorescence concentration</td>
<td>mol/l</td>
</tr>
<tr>
<td>$\langle c \rangle$</td>
<td>averaged concentration</td>
<td>mol/l</td>
</tr>
<tr>
<td>$c_n$</td>
<td>concentration of $n^{th}$ measurement</td>
<td>mol/l</td>
</tr>
<tr>
<td>$C_f$</td>
<td>friction coefficient</td>
<td></td>
</tr>
<tr>
<td>$d$</td>
<td>displacement vector</td>
<td>m</td>
</tr>
<tr>
<td>$D_t$</td>
<td>diameter of tubular reactor</td>
<td>m</td>
</tr>
<tr>
<td>$D$</td>
<td>diffusion coefficient</td>
<td>$m^2/s$</td>
</tr>
<tr>
<td>$f(x)$</td>
<td>image function</td>
<td>grey value</td>
</tr>
<tr>
<td>$F(s)$</td>
<td>Fourier transform of image function $f(x)$</td>
<td></td>
</tr>
<tr>
<td>$g(x)$</td>
<td>image function</td>
<td>grey value</td>
</tr>
<tr>
<td>$G(s)$</td>
<td>Fourier transform of image function $g(x)$</td>
<td></td>
</tr>
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<td>$h(x)$</td>
<td>2D spike function</td>
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</tr>
<tr>
<td>$H(s)$</td>
<td>Fourier transform of 2D spike function</td>
<td></td>
</tr>
<tr>
<td>$i$</td>
<td>number</td>
<td></td>
</tr>
<tr>
<td>$I_0(z)$</td>
<td>laser intensity at point $z$</td>
<td>W</td>
</tr>
<tr>
<td>$I_f$</td>
<td>fluorescence intensity</td>
<td>W</td>
</tr>
<tr>
<td>$I_0$</td>
<td>initial laser intensity</td>
<td>W</td>
</tr>
<tr>
<td>$j$</td>
<td>number</td>
<td></td>
</tr>
<tr>
<td>$k$</td>
<td>number</td>
<td></td>
</tr>
<tr>
<td>$L$</td>
<td>length in the field of detector</td>
<td>m</td>
</tr>
<tr>
<td>$n$</td>
<td>number of measurement</td>
<td></td>
</tr>
<tr>
<td>$n_m$</td>
<td>refractive index</td>
<td></td>
</tr>
<tr>
<td>$N$</td>
<td>total number of measurements</td>
<td></td>
</tr>
<tr>
<td>$P$</td>
<td>point in an image</td>
<td></td>
</tr>
<tr>
<td>$s$</td>
<td>space vector</td>
<td>m</td>
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<tr>
<td>$\langle S \rangle$</td>
<td>averaged skewness</td>
<td></td>
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<tr>
<td>$\langle SD \rangle$</td>
<td>averaged standard deviation</td>
<td>mol/l</td>
</tr>
<tr>
<td>$t$</td>
<td>time</td>
<td>s</td>
</tr>
<tr>
<td>$\Delta T$</td>
<td>measuring time</td>
<td>s</td>
</tr>
<tr>
<td>$u$</td>
<td>velocity</td>
<td>m/s</td>
</tr>
<tr>
<td>$u_*$</td>
<td>friction velocity</td>
<td>m/s</td>
</tr>
<tr>
<td>$U$</td>
<td>mean flow velocity</td>
<td>m/s</td>
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### Symbol Description

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<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unity</th>
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<tbody>
<tr>
<td>$\mathcal{U}$</td>
<td>Kolmogorov velocity</td>
<td>$m/s$</td>
</tr>
<tr>
<td>$W$</td>
<td>minimal spot size laser beam</td>
<td>$m$</td>
</tr>
<tr>
<td>$W_0$</td>
<td>initial minimal spot size</td>
<td>$m$</td>
</tr>
<tr>
<td>$x$</td>
<td>space coordinate</td>
<td>$m$</td>
</tr>
<tr>
<td>$\Delta x$</td>
<td>measurement space resolution</td>
<td>$m$</td>
</tr>
<tr>
<td>$z$</td>
<td>space coordinate</td>
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<td>$z_m$</td>
<td>measurement point</td>
<td>$m$</td>
</tr>
<tr>
<td>$z_w$</td>
<td>position of minimal spot size</td>
<td>$m$</td>
</tr>
<tr>
<td>$z_0$</td>
<td>scale parameter</td>
<td>$m$</td>
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### Greek symbols

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<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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</tr>
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<tbody>
<tr>
<td>$\epsilon$</td>
<td>mean local energy dissipation</td>
<td>$m^2/s^3$</td>
</tr>
<tr>
<td>$\varepsilon$</td>
<td>extinction coefficient</td>
<td>$(mol/lm)^{-1}$</td>
</tr>
<tr>
<td>$\zeta$</td>
<td>scalar concentration</td>
<td>$mol/l$</td>
</tr>
<tr>
<td>$\nabla \zeta$</td>
<td>scalar concentration gradient</td>
<td>$mol/(lm)^{-1}$</td>
</tr>
<tr>
<td>$\nabla \zeta^*$</td>
<td>scalar concentration gradient normalisation constant</td>
<td>$mol/(lm)^{-1}$</td>
</tr>
<tr>
<td>$\eta_b$</td>
<td>Batchelor scale</td>
<td>$m$</td>
</tr>
<tr>
<td>$\eta_k$</td>
<td>Kolmogorov scale</td>
<td>$m$</td>
</tr>
<tr>
<td>$\theta$</td>
<td>polar coordinate</td>
<td>$rad$</td>
</tr>
<tr>
<td>$\phi$</td>
<td>angle</td>
<td>$rad$</td>
</tr>
<tr>
<td>$\lambda$</td>
<td>wave length</td>
<td>$m$</td>
</tr>
<tr>
<td>$\nu$</td>
<td>kinematic viscosity</td>
<td>$m^2/s$</td>
</tr>
<tr>
<td>$\tau_a$</td>
<td>advection time scale</td>
<td>$s$</td>
</tr>
<tr>
<td>$\tau_k$</td>
<td>Kolmogorov time scale</td>
<td>$s$</td>
</tr>
<tr>
<td>$\phi$</td>
<td>polar coordinate</td>
<td>$rad$</td>
</tr>
<tr>
<td>$\varphi$</td>
<td>quantum yield</td>
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### Dimensionless numbers

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<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unity</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Re$</td>
<td>Reynolds number</td>
<td>$-$</td>
</tr>
<tr>
<td>$Re_*$</td>
<td>Reynolds number based on friction velocity</td>
<td>$-$</td>
</tr>
<tr>
<td>$Sc$</td>
<td>Schmidt number</td>
<td>$-$</td>
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### Abbreviations

<table>
<thead>
<tr>
<th>Abbr.</th>
<th>Description</th>
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<tbody>
<tr>
<td>FFT</td>
<td>fast Fourier transform</td>
</tr>
<tr>
<td>LIF</td>
<td>laser induced fluorescence</td>
</tr>
<tr>
<td>PDF</td>
<td>probability density function</td>
</tr>
<tr>
<td>RFFT</td>
<td>reverse fast Fourier transform</td>
</tr>
</tbody>
</table>
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Chapter 1

Introduction

1.1 Framework

Chemical reactions are widely used in industrial production processes. It is not unusual to have several chemical reactions taking place simultaneously. In that case the yield of the process is difficult to predict, though of great importance for the industrial process. Well known as an influential factor to product yield is mixing. In case of competing, diffusion limited reactions a high mixing rate favours the fastest chemical reaction. Contrary, low mixing rate enables the slower chemical reactions to take place. Mixing in flows is mainly driven by turbulence, which is a chaotic phenomenon. Predictions concerning mixing are therefore difficult to do.

Therefore mixing is of great interest for scientists and industries. Several models (e.g. [Bakker, 1996]) have been developed to describe mixing. These models mainly concern mixing at the smallest scales of the turbulent flow since at or below these dynamical scales chemical reactions take place. Experiments have to be done to verify the modelling assumptions. An assumption generally made in the models is that the small scale vortices stretch in time. A proper validation of this assumption can only be done by a 3D measurement of the scalar concentration field.

Another important parameter often mentioned in these mixing models is the scalar energy dissipation, which is proportional to the modulus of the scalar gradient vector field. A determination of this mixing parameters also requires a 3D measurement. Therefore, an experimental setup, based on Laser Induced Fluorescence, has been designed to obtain a 3D visualisation of the scalar concentration field at the Kolmogorov scale, i.e. the smallest scale. Until recently such experiments could only be done in a stagnant flow with extremely large length and time scales ([Dahm et al., 1991], [Su and Clemens, 1999]). However, with the development of new high speed digital camera's it has become feasible to perform such an experiment in an industrial geometry.
1.2 Objectives of the project and thesis

The subject of this thesis is the design and critical validation of an experimental setup that measures a three dimensional concentration field of a conservative scalar mixing into a turbulent main flow of a tubular reactor. In the framework previously described the first step is to obtain a 3D visualisation of the mixing process. This must be done sufficiently accurate to detect properly the smallest occurring structures and scalar gradients in the flow.

After the visualisation, the second step is to qualitatively study the visualised mixing process. Micro mixing models are generally based on the assumption that multiple layers of alternating fluids are formed on the Kolmogorov scale and that these layers shrink in time. This can be validated by the visualised micro scale structures.

Besides qualitative information also quantitative information can be obtained out of the visualised mixing process. The moments of the concentration field are calculated and studied, followed by the scalar energy dissipation calculated on basis of the three components of the scalar concentration gradient. The square root of this scalar energy dissipation is defined as the mixing ratio and represents mixing intensity.

Finally, information about the turbulent flow is obtained through a study of isotropy of the scalar gradient field.

1.3 Outline of thesis

Chapter 2 handles the theory concerning the turbulent flow in which the experiments are performed. The experiment has to resolve the smallest flow structures in both space and time. The most important length and time scales are discussed in chapter 2. It is followed by an introduction of the scalar energy dissipation and its value to micromixing will be clarified. Due to the chaotic nature of the turbulent flow it will be necessary to obtain certain quantities on a statistical basis. Therefore chapter 2 will end with an introduction of statistical validation methods.

Chapter 3 concerns the experiment itself. First the experiment and the Laser Induced Fluorescence technique are explained. Next, the experimental setup is given in detail. As mentioned before this chapter ends with the assessment of the feasibility of the experiment solving the smallest length and time scales. Therefore a comparison is made between the spatial and temporal resolution of the experiment and the turbulent scales, introduced in chapter 2.

Chapter 4 concerns data processing techniques. After visualising the mixing process certain image processing operations are needed before any quantitative information can be obtained from the data. The necessity of these corrections as well as the way they are implemented is discussed in chapter 4. Here also the used algorithm to calculate the scalar concentration gradient field is explained, as well as an explanation how power spectrum analyses are performed on the data.

After visualisation (chapter 3) and application of corrections (chapter 4), the data
processing can actually be performed. The results are presented in chapter 5. This chapter contains the statistic validation of the results, a study of the measured structures and the mean concentration field. It ends with the calculations of the 3D energy dissipation and a study of isotropy of the scalar gradient field.

In chapter 6 conclusions concerning the present work are set forth. These conclusions enclose all of the experimental and theoretical work done.

Finally some recommendations for possible future experimental work, concerning 3D visualisation of mixing in a tubular reactor, are given in chapter 7.

This research was carried out as the final part of the study program for the degree of engineer (M.Sc.) in applied physics. It is performed at the Kramers Laboratorium voor Fysische Technologie at the Delft University of Technology.
Chapter 2

The Turbulent Flow

This chapter provides the theory of turbulent flows. First a short introduction into mixing and turbulence is given. In mixing research in a turbulent flow several length and time scales are of interest. Here, the relevant parameters will be introduced and discussed. Paragraph 2.2 concerns turbulence related parameters and paragraph 2.3 explains the scalar energy dissipation, important parameter in micromixing. The chapter ends with the introduction of some statistical validation tools, as in chapter 5 all quantities are obtained on a statistical basis.

2.1 Mixing and turbulence

Mixing is the process of bringing initially separated fluids into close contact, usually with the purpose of achieving a certain mass exchange of reactants dissolved in the fluids. In the case of competing, diffusion limited, chemical reactions it is known that mixing strongly influences the yield of products. Two different stages of mixing exist. These two stages of mixing are strongly related to turbulence and the behaviour of turbulent structures.

In a turbulent flow energy of the main flow is used to form large eddies (turbulent structures). A dye that is injected into the flow system is advected and dispersed by these large structures. Mixing at this stage is called macromixing. The size of these turbulent structures is determined by the geometry of the reactor. During one eddy lifetime a macro scale eddy cascades into smaller eddies, which in

![Figure 2.1: Illustration of the cascade process in a turbulent flow](image-url)
Chapter 2. The Turbulent Flow

their turn cascade in even smaller eddies. This process continues until the eddies become so small that viscosity is able to dissipate them into heat (see figure 2.1). These smallest eddies are known as the Kolmogorov structures and the scale at which they appear is called the Kolmogorov scale. Mixing occurring at and beyond this scale is denoted as micromixing. These micro scales are characterised by large interfacial area and small length scales over which scalar differences exist. This results in molecular diffusion which is the driving force of micromixing. Especially micromixing is important, given the fact that at this scale chemical reactions take place.

The micro and macro scales of a turbulent flow are essentially different. While the flow on the macro scale is turbulent, the micro scale flow is defined to be laminar. Mechanistic micromixing models therefore solve the micro scale mass balance of the reactants deterministically. All these models include assumptions on the behaviour of the smallest structures. Vortices on the micro scale tend to mix the fluids by rolling up, creating layers of alternating fluids. Due to strain of the macro scale flow these vortices stretch in time, thereby tightening the layers. As a result the scalar gradients increase in these layers. These scalar concentration gradients are mentioned before as the driving force of molecular diffusion, hence vortex stretching influences micromixing.

2.2 Characteristic length and time scales

Characterising scales are useful to describe a flow in which experiments are done. An important phenomenon closely related to mixing is turbulence. Undebatable it can be posed that turbulence enhances mixing. For this reason millions of people stir their coffee after adding milk. The Reynolds number is an appropriate measure for the degree of turbulence. The Reynolds number based on the mean flow velocity $U$ and reactor diameter $D_t$ is defined as:

$$Re = \frac{UD_t}{\nu},$$

where $\nu$ represents the kinematic viscosity. The Reynolds number is a measure for the ratio between inertial and viscous forces. Generally, tubular flows are considered to be turbulent when the Reynolds number exceeds 2,000. Several different length and time scales depending on the Reynolds number, can be recognised (paragraph 2.1). At the macro scale two characteristic parameters are of interest, that characterise the size and velocity of the largest occurring structures in the flow system. This macro length and velocity scale equal respectively the width of the flow system, which in the case of a tubular reactor is the diameter of the tube, and the mean average velocity of the flow. In the framework of micromixing research also several characterising parameters of the micro scale are needed. The important parameters in this thesis can be sorted in length and time scales. The length scales represent the characteristic length of the smallest structures (Kolmogorov structures), referred to
2.2. Characteristic length and time scales

as Kolmogorov length scale $\eta_k$, and the diffusion depth of a scalar within one Kolmogorov life time, referred to as Batchelor length scale $\eta_b$. The former is defined as:

$$\eta_k = \left( \frac{\nu^3}{\epsilon} \right)^{1/4},$$  \hspace{1cm} (2.2)

where $\epsilon$ is the mean local energy dissipation. In this thesis, the Kolmogorov scales are estimated in a fully developed turbulent flow at the centre line of the tubular reactor. [Hinze, 1959] poses that the dimension-less energy dissipation $\epsilon D_t / (2u_s^2)$ (based on the friction velocity $u_s$) at a centre line of a tubular reactor is measured to be 2.2 for a 1D pipe flow. This results in the following expression for $\epsilon$.

$$\epsilon = 4.4 \frac{\nu^3}{D_t^2} Re_k^3.$$  \hspace{1cm} (2.3)

The friction velocity is related to the mean velocity of the flow system $U$ according to ([Bradshaw, 1976]):

$$\left( \frac{u_s}{U} \right)^2 = C_f,$$  \hspace{1cm} (2.4)

where the friction coefficient $C_f$ is given by the Blasius-law applied on pipe flows:

$$C_f = 0.079 Re^{-\frac{1}{4}}.$$  \hspace{1cm} (2.5)

Combining equation (2.1), equation(2.4) and equation(2.5) results in the following relation between $Re_*$ and $Re$.

$$Re_* = 0.28 Re_k^{\frac{2}{3}}.$$  \hspace{1cm} (2.6)

Using the Reynolds number given by equation (2.1), the mean local energy dissipation can be calculated using equation (2.6) and (2.3). Subsequently the Kolmogorov length scale $\eta_k$ at the centre line of a fully developed, turbulent pipe flow can be calculated using equation (2.2). Besides the characteristic length of the Kolmogorov structures it is also relevant to characterise the diffusion depth of such structures during their life time. This Batchelor length scale $\eta_b$ is, according to the penetration theory, defined as:

$$\eta_b = \sqrt{\mathcal{D} \tau_k} = \eta_k \cdot Sc^{-\frac{1}{2}},$$  \hspace{1cm} (2.7)

where $\mathcal{D}$ is the scalar diffusivity, $\tau_k$ the characteristic Kolmogorov life time and the Schmidt number $Sc$ defined as:
Figure 2.2: Figure to clarify the different length scales. A sharp edged Kolmogorov structures diffuse during its lifetime $\tau_k$ into the surrounding flow. The characteristic diffusion depth is defined as the Batchelor scale and in liquids ($Sc \gg 1$) it is much smaller than the characteristic size of the structure.

$$Sc \equiv \frac{\nu}{\mu D}.$$  \hspace{1cm} (2.8)

Figure 2.2 shows the relation between the Kolmogorov scale and the Batchelor scale. In order to measure these space parameters in an experiment they should be compared to the measurement space resolutions. The spatial resolution of the measurement system should be significantly smaller than the smallest length scales if the latter have to be solved.

The time scales characterise the life time of the Kolmogorov structures as well as the time over which a structure can be considered standing still for measurement. These time scales are called the Kolmogorov time scale and the advection time scale respectively. The Kolmogorov time scale is defined as:

$$\tau_k = \left(\frac{\nu}{\epsilon}\right)^{\frac{1}{2}}.$$  \hspace{1cm} (2.9)

The Kolmogorov time scale can be interpreted to be the eddy life time of a Kolmogorov structure before it is dissipated into heat. The advection time scale is defined as:

$$\tau_a = \frac{\eta_k}{U},$$  \hspace{1cm} (2.10)

which can be interpreted as the time for a Kolmogorov structure to pass a fixed point when advected by the main flow velocity. The feasibility of measuring the smallest structures in the turbulent flow without this structures being dissipated or advected during the measurement, should be checked by comparing these characteristic time scales with the measuring time. The measuring time should be significantly smaller than the two characteristic time scales. With the design of the experiment this must be taken into account.
2.3. Scalar energy dissipation rate

Having defined the Kolmogorov length and time scale, it is also possible to define a characteristic velocity of a Kolmogorov structure $U_k$:

\[ U_k = \frac{\eta_k}{\tau_k}, \tag{2.11} \]

It represents the velocity difference within a Kolmogorov eddy. It is related to the deformation speed of a micro scale structure. To ensure that such a structure is not deforming, its deformation due to the internal velocity differences should be significantly smaller than the displacement due to the advection speed.

2.3 Scalar energy dissipation rate

Besides the several flow describing parameters, there also exist several mixing describing parameters. Examples of these parameters are interfacial area and scalar energy dissipation rate. The first is already mentioned in paragraph 2.1, therefore here the latter will be discussed. While mixing a conserved scalar quantity $C$, the conserved scalar quantity field $C(x, t)$ should satisfy the advection-diffusion equation

\[ \frac{\partial}{\partial t} + u \cdot \nabla - D \nabla^2 \zeta(x, t) = 0, \tag{2.12} \]

where $u$ represents the local velocity of the flow. The first term on the left-hand side is a change of the scalar in time, the middle term is the change due to advection by the local velocity of the flow and the last term on the left-hand side is the change of the scalar due to diffusion. Since $\zeta(x, t)$ is defined to be a conserved scalar (no chemical reaction is taking place), the right-hand side equals zero. A scalar energy per unit mass $\frac{1}{2} \zeta^2(x, t)$ can be defined analogous to the kinetic energy per unit mass. The transport equation for the scalar energy per unit mass then becomes

\[ \frac{\partial}{\partial t} + u \cdot \nabla - D \nabla^2 \frac{1}{2} \zeta^2(x, t) = -D \nabla \zeta \cdot \nabla \zeta(x, t), \tag{2.13} \]

where a similar term as in equation (2.12) appears on the left-hand side. The term on the right-hand side represents the rate at which scalar variance is being reduced by molecular diffusion. It is an appropriate measure for the local molecular mixing rate. Therefore the scalar energy dissipation rate is defined as $\nabla \zeta \cdot \nabla \zeta$ and it is often used as a meaningful parameter in the area of micromixing. Where the scalar energy dissipation is high, micromixing is considered to be strong. Consequently, from regions where the scalar energy dissipation is low can be deducted that at these regions no micromixing takes place. Alternatively, the scalar gradient magnitude $|\nabla \zeta(x, t)|$ is defined the mixing ratio. The concept of scalar energy and its dissipation rate play a central role in many approaches for the understanding and modelling of micromixing in turbulent flows. In order to determine experimental values for the scalar energy and its dissipation rate a determination of the scalar field $\zeta(x, t)$ in all three dimensions is required.
2.4 Statistical validation methods

A turbulent flow is a chaotic phenomenon. This means that individual realisations are unpredictable. For this reason, parameters are often obtained on a statistical basis. To properly calculate statistical values a large number of independent measurements should be used. This condition contains two aspects that should be checked: sufficient amount of data and independency of data.

The question whether sufficient data is gathered, can be answered in two different ways. The first method is to do several measurements at a position in the reactor where within the measuring volume in time an uniformly distributed scalar field is expected. If the number of measurements performed is sufficient, the time averaged scalar field will be smooth (i.e. uniformly distributed). If the time averaged scalar field is not smooth, the number of measurements should be increased.

Several scalar field parameters can be used to validate the amount of measurements performed. The first parameter to be calculated is the time averaged scalar concentration field \( \langle c \rangle \). It is assumed that the process is stationary. Therefore according the ergodic hypotheses time averaging equals ensemble averaging.

\[
\langle c \rangle = \frac{1}{N} \sum_{n} c_n
\]  
(2.14)

where \( N \) is the number of measurements and \( c_n \) is the concentration of the \( n^{th} \) measurement. Stricter criteria for a sufficient number of measurements are the time averaged standard deviation \( \langle SD \rangle \) and the time averaged skewness \( \langle S \rangle \), estimated by

\[
\langle SD \rangle^2 = \frac{1}{N-1} \sum_{n} (c_n - \langle c \rangle)^2 
\]  
(2.15)
2.4. Statistical validation methods

(2.16) \[
\langle S \rangle = \frac{1}{N} \sum_{n} (c_n - \langle c \rangle)^3
\]

In case of the time average skewness field the criterion of smoothness is the hardest to fulfill. Why

This method cannot be applied on measurements performed at positions where due to flow phenomena not an uniformly distributed mean concentration field is expected. Then, a second method can be applied. It is assumed that sufficient data is gathered if the non uniformities in the mean concentration field have stabilised. The used parameter to study this is the standard deviation of the mean concentration field. This means that the number of measurements should be increased until the standard deviation of the calculated field has reached a constant value.

Independency of the data is checked by studying the auto correlation function of the data. Assumed is that if the used data is independent, the auto correlation function approaches the delta function. Contrary, when the data is correlated, its peak around zero is very wide.

Besides validating whether statistics has converged, the three parameters introduced before, also have qualitative meaning. To illustrate this figure 2.3 is drawn. The figure shows a estimation of the probability density function of the measured concentrations. It is the expectation that the majority of the concentrations measured represent the background, which results in the peak at the lowest concentrations. The other peak represents the concentrations of the structures passing by. The standard deviation represents the width of the concentration distribution and the skewness level of asymmetry, which means that in case of a high skewness value the PDF is highly asymmetric.

It may be concluded that all three parameters increase in value when the number or the concentration of the structures increase. The higher moments are more influenced by a higher concentration per structure than an increase in the number of structures. Combining the information of the three parameters supplies even more information concerning the turbulent flow.
Chapter 3

The 4D Laser Induced Fluorescence Experiment

This chapter deals with the experiment itself. After a short introduction into the experimental method and the Laser Induced Fluorescence (LIF) technique, the experimental setup is described in detail. Values for the length and time scales introduced in the previous chapter are given, as well as the resolutions at which the measurements are executed. This chapter ends with a discussion on the feasibility of the objective of the thesis (i.e. to visualise the micro scale flow) by comparing the experimental resolutions with the turbulent flow scales.

3.1 About the experiment

In this thesis LIF experiments are done to visualise the mixing process in a tubular reactor. LIF consists of a fluorescent dye mixing into a non fluorescent main flow. During the mixing process a laser sheet is put through the flow system, resulting in an excitation of the fluorescent dye, while the main flow remains unexcited. Meanwhile images are captured by means of a digital camera, resulting in images as shown in figure 3.1 in which the grey values are proportional to the local dye concentration. In these images the mixing process is visualised and it is possible to study the structures occurring in the mixing process. If the laser sheet is traversed up or down, parallel

![Figure 3.1: Examples of captured images using LIF](image_url)
3.2 The Laser Induced Fluorescence technique

In this thesis a LIF technique is used to do non intrusive measurements of a scalar concentration field. In this paragraph the principles of this technique will be explained. LIF is based on the fluorescent property of the injected dye (disodium fluorescein). This fluorescein molecule has broad absorption and fluorescence (emission) spectra. The (outer) electrons can be exited from the ground state towards the singlet state by means of light with a specific wave length (see figure 3.2). The excited electron has three pathways to return to the ground state. One is through radiationless collision, second is through transition to a triplet state from where the electron returns to the ground state through phosphorescence. The third way is called fluorescence and involves the return to the ground state after a shift to the lowest vibrational level of the singlet state. Due to the shift to the lowest vibrational level, the wavelengths associated with fluorescence are larger than those of the excitation. This has the advantage that the fluorescent light can easily be separated from all kinds of unwanted reflections of the original light source. The time required for an electron to return through fluorescence to its ground level is in the order of 5 ns (see [Walker, 1993]). Considering a molecule of the size of fluorescein, this period of time is long enough for the emission orientation to be completely independent of the absorption orientation. In other words, although excited by plane-polarised laser light, the fluorescence is randomly polarised and the intensity is independent of direction.

Traversing through a fluorescent solution, meanwhile exciting the dye molecules, the laser beam is attenuated (see figure 3.3). For a beam path, $dz$, the laser beam...
3.2. The Laser Induced Fluorescence technique

Figure 3.3: Absorption and fluorescence along the laser beam path.

attenuation is given by Beer's law:

$$dI_e(z) = -\varepsilon c(z)I_e(z)dz. \quad (3.1)$$

Here $I_e$ is the intensity of the laser beam at point $z$ along the beam path, $c(z)$ is the dye concentration at $z$ and $\varepsilon$ is the extinction coefficient. At high laser intensity two phenomena can occur: saturation, i.e. nearly all molecules in the beam path are excited and fluorescence intensity becomes independent of the laser intensity, or photo bleaching, i.e. the dye molecules are ionised by the laser, so no fluorescence can occur. Photo bleaching results in a fluorescence intensity that is decreasing in time. However, the time scales at which photo bleaching occurs are significant larger then the residence time of the dye molecules in the laser beam. It is assumed that no photo bleaching occurred during the measurements (see appendix B).

As described before, the wavelength of the fluorescent light is larger than the wavelength of the laser beam, so attenuation can be neglected along the receiving path. The emitted fluorescence light depends on the local dye concentration and the incident laser light as follows:

$$I_f(z) = A\varphi L c(z)I_e(z), \quad (3.2)$$

where $A$ is the fraction of available light collected by the camera, $\varphi$ the quantum yield defined as the ratio between emitted and absorbed light, $L$ the length that is in the field of view of the detector (centred around position $z_m$). The absolute intensity of the fluorescence at $z_m$ can be described by

$$I_f(z_m) = A\varphi L \varepsilon c(z_m)I_0e^{-\varepsilon \int_0^{z_m} c dz}. \quad (3.3)$$

Thus, the intensity of the collected fluorescent light depends on the integral of the concentration field along the beam path before the measured area. However, at low enough concentration, the integral from equation (3.3) becomes
Figure 3.4: Impression of the used experimental setup

\[ \varepsilon \int_0^{s_m} c dz \ll 1. \]  

Then, the attenuation of the incident beam may be neglected, resulting in a linear relation between \( I_f \) and \( c(z) \). This is a preferable condition, as correction for the laser beam attenuation is a difficult task, which hardly can be done without introducing errors to the results. Therefore, it has been taken care of that concentrations were low enough to neglect laser beam attenuation during all the measurements (see appendix A.1).

3.3 The experimental setup

Figure 3.4 shows an impression of the setup used for the 4D LIF experiments. At the back the tubular reactor is shown, in which a moderately turbulent flow of non fluorescent tap water is established. This tubular reactor is placed inside a water filled perspex box to prevent the laser beam from refracting too much while entering the tubular reactor. At a certain point a fluorescent dye is injected in the main flow. The jet velocity of the dye is larger than the main stream velocity. Also shown in figure 3.4 is the used Argon ion laser (Spectra Physics, 488 nm), capable of continuously emitting a beam with an intensity up to 8W. For the LIF experiment the laser beam needs to be converted into a sheet. Therefore a cylindrical and spherical lens are inserted in the setup (see figure 3.4 and 3.5). The cylindrical lens is used to create a diverging beam (figure 3.5(b)). The spherical lens has several functions. First it is used to stop the sheet diverging when the required sheet width is reached (figure 3.5(b)). Further, the spherical lens makes the minimal thickness (waist) of the laser sheet to be placed in the measured area of the tubular reactor (figure 3.5(a)). The laser sheet, with a minimal thickness of 120\( \mu m \), is put through
3.3. The experimental setup

Figure 3.5: Side and top view of the optical part of the experimental setup. In the side view the use of the Galvano mirror can be seen. The Galvano mirror is placed in the focus of the spherical lens in order to let the sheet enter the reactor horizontally. Besides, the spherical lens should be positioned that its focus also coincides with the centreline of the reactor in order to have the minimal sheet thickness in the measured area. The laser sheet is shown at the uppermost and the lowermost positions. The top view shows how the cylindrical lens and the spherical lens convert the laser beam into the required sheet. Therefore the Galvano mirror should be placed into the focus of the two lenses. The exact distances between the different components of the setup is given in appendix C. GM=Galvano mirror, CL=cylindrical lens (focal length=-12.0mm), SL=spherical lens (focal length=500mm), GW=glass walls of the tubular reactor and the surrounding water filled box.
Chapter 3. The 4D Laser Induced Fluorescence Experiment

Figure 3.6: Schematic right and front view of the tubular reactor

the tubular reactor at certain distance downstream the inlet of the fluorescent dye. A CCD camera positioned on top of the reactor is used to capture images at a frame rate of 1000 frames per second. An optical filter is used to provoke (Rayleigh) scattered laser light from entering the camera. An NT workstation is used to store the images.

By means of a galvano mirror placed before in the focus of the spherical lens, it is possible to traverse the laser sheet, parallel to itself, up and down through the reactor (see figure 3.5(a)). This is also illustrated in figure 3.4 where the laser sheet is shown at several positions in the reactor.

During the experiment the measuring volume consisted of ten frames, where a frame is an image captured with the CCD camera. The camera frame rate of 1000 fps corresponds with an exposure time of 1 ms. The pause between the images shot is significantly larger than the time scales at which fluorescence takes place (see paragraph 3.2), hence after glowing does not contaminate the measurement. The trigger electronics, also shown in figure 3.4, are needed to synchronise the movement of the laser sheet with the frame rate of the CCD camera.

In figure 3.6 a schematic front and right side view of the tubular reactor is shown. In the right side view of the tubular reactor the main flow is coming from the left. The sizes of the inlet and the reactor are shown as well to give an impression of the size of the measured area.

3.4 Flow description

The mixing process is studied in a turbulent flow. To characterise the degree of turbulence of the flow the Reynolds number based on the reactor diameter is calculated using equation (2.1. The flow rate of the mean stream is put to 14 l/min. Together with the diameter of the reactor this results in a mean bulk velocity $U$ of about 3 cm/s. This results in a Reynolds number of 2,800, implicating that the main flow can be considered in the lower turbulent region. Note that here the effect of the feedpipe is not taken into account. Lattice-Boltzmann simulations of the velocity
3.5. **Measurement resolutions**

Field (see appendix D) downstream the inlet show that higher velocities occur as a result of the feed pipe. A higher turbulent flow as a result of the feedpipe is found at 15 cm downstream the inlet. Also the fact that the fluorescent dye is injected at larger velocity than the main flow velocity results is not taken into account, although it results in more turbulence.

In the framework of micro mixing research it is necessary to characterise the flow on micro scale. As explained in paragraph 2.2 the micro scale flow can be characterised with two length scales, the Kolmogorov length scale $\eta_k$ and the Batchelor length scale $\eta_b$, and with two time scales, the Kolmogorov time scale $\tau_k$ and the advection time scale $\tau_a$. With the viscosity of water, the energy dissipation $\epsilon$ estimated at the centreline of the tubular reactor, the characteristic macro length scale and the diffusion coefficient of the fluorescent dye in water, values for the characteristic micro scales can be calculated. The values for the two length scales in a fully developed tube flow are $\eta_k = 980 \mu m$ and $\eta_b = 22 \mu m$. As can be seen the Batchelor scale is significantly smaller than the Kolmogorov scale. This as a result of the high Schmidt number of fluids (see paragraph 2.2, i.e. 2000).

The two micro time scales, calculated as described in paragraph 2.2 using the spatial flow scales and medium properties are $\tau_k = 0.96 s$ and $\tau_a = 35 ms$.

### 3.5 Measurement resolutions

The objective to solve the micro scales in both space and time, imposes certain demands on the measurement resolutions. To measure the scalar mixing into the main flow, a CCD camera (trade DALSA) is used. It contains a CCD chip consisting of 256x256 pixels, each at 10 $\mu m$ distance. The camera measures 8-bit grey values with an accuracy of 1 grey value. During the measurement the magnification was put to be 10, resulting in a measuring plane of 2.56 cm by 2.56 cm, measured at a resolution of 100 $\mu m$. By adjusting the voltage on the galvano mirror the distance between each image in the measuring volume can be put also to 100 $\mu m$. Hence, the measurement resolutions are equal in all directions.

Each measuring volume consisted of 10 sequential frames, taken with a capture rate of 1000 frames per second. This results in a measuring time of 10 ms.

The height of the measuring volume was 1 mm. The depth-of-field of the camera is around $3 mm$, hence all images are sharp. All measurements were performed the diafragma number of the camera put to 1.6.

### 3.6 Conclusions about experimental setup

In this chapter the experiment is explained in detail and values for the turbulent flow scales as well as the measurement resolutions are given. Now a validation of the objective to measure a 3D scalar concentration field can be done by comparing the spatial and temporal flow scales and the measurement resolutions. Mentioned before is that in order to solve the micro scale parameters the measurement resolutions must be significantly smaller than the occurring length and time scales in the flow.
To judge what is significantly smaller, the Nyquist criterion is used. The Nyquist criterion postulates that a critical sampling of a sine wave is sampling the signal using two sample points a cycle. Translated to this experiment it poses that a Kolmogorov structure can only be properly detected when it covers at least two sample points. This means that the spatial resolution of the measurement should be half the length scales of the turbulent flow at maximum in order to detect Kolmogorov micro scale structures properly.

Using the values of paragraph 2.2 it can be calculated that the ratio of spatial resolution $\Delta x$ to the two length scales is as follows:

$$\Delta x = 0.1\eta_k,$$  \hspace{1cm} (3.5)

$$\Delta x = 4.5\eta_B.$$  \hspace{1cm} (3.6)

In conclusion this means that, using the Nyquist criterion, the smallest occurring structures can be detected using this setup. However, the occurring gradients at the borders of these structures are not detected. Therefore, the prediction can be made that with this setup sharply edged Kolmogorov structures will be detected.

Similar, the micro time scales and the time resolution are compared. The following relations between the different time scales and measuring time $\Delta T$ can be calculated:

$$\Delta T = 0.01\tau_k,$$  \hspace{1cm} (3.7)

$$\Delta T = 0.3\tau_a.$$  \hspace{1cm} (3.8)

The ratios imply that the Kolmogorov structures are not being dissipated into heat nor are being advected significantly by the mean flow during the time span of one measurement.

A last critical issue according to the feasibility of the objective concerns the deformation of structures in time. The eddies are not allowed to deform during the measurement. The Taylor-hypotheses provides a way of judging deformation velocity by comparing it to the advection velocity $|\overline{\overline{U}}|$. It calculates the ratio $U/|\overline{\overline{U}}|$, where $U$ is the characteristic velocity of a Kolmogorov structure as introduced in paragraph 2.2. This ratio is denoted as turbulence-intensity. The hypotheses postulates that if this ratio is smaller than 10%, the eddy can be considered frozen while passing a measurement point. Using the values from paragraph 3.4 it can be calculated that the turbulence-intensity equals 3.3%. According to the Taylor-hypotheses this means that the micro scale structures can be considered frozen during the measurement. The structures are advected that fast through the measurement volume that the deformation due to the velocity gradient within the Kolmogorov structure is neglectable. The advection velocity was previously determined as satisfying the Nyquist-criterion.

In conclusion it may be posed that the thesis objective of visualising the mixing process at micro scale can be fulfilled. All occurring structures, including the Kolmogorov structures, can be detected with the described setup. However, the gradients on the borders at the Kolmogorov structures cannot be detected. The visualised
structures are not being dissipated nor deformed nor advected during the measurement.

One remark should be made concerning this assessment. The dimensionless numbers are calculated based on the energy dissipation on the centre line of the flow. This is a rough estimation, because the influence of the feedpipe is not taken into account. Furthermore, the dimensionless numbers are order estimates, therefore all conclusions derived here should be checked experimentally. Especially ratios which were close to the Nyquist critical ratio, i.e. the advection time scale and the Batchelor scale, require an experimental study. When calculating the scalar energy dissipation field it is of great importance to check whether the scalar gradient vector field can be detected.
Chapter 3. The 4D Laser Induced Fluorescence Experiment
Chapter 4

Data Processing

In this chapter the image processing operation applied on the raw 8-bit images are discussed. To process the images in order to obtain experimental information about the mixing process and the flow, the software package Scilimage 1.4.1 ([Scilimage, 1998]) is used. In this chapter it will be discussed what operations are applied on the raw data. First the several corrections that are needed will be discussed, followed by an introduction in the applied power spectrum analyses. Then the used algorithm to calculate the scalar gradient field is explained. Finally a threshold applied in the data processing, will be clarified.

4.1 Corrections

The objective of the experiment is to deduce meaningful information about the mixing process out of images shot using 4D LIF. Unfortunately the experiment is far from ideal, so this is not as simple as posed in paragraph 3.1. Due to certain aspects of the experiment which cannot be prevented, corrections are required before any meaningful quantity can be deduced from the data. Aspects for which corrections have been developed are:

- The background and dark current noise. These two badly affect the signal-to-noise ratio.
- The Gaussian profile of the laser sheet. This implies a nonuniform illumination of the scalar field.
- Advection of structures by the mean flow velocity during one measurement. The possibility of this occurring was discussed in paragraph 2.2.

In this paragraph it will be explained why a certain correction is needed and how it is done.
Chapter 4. Data Processing

4.1.1 Noise corrections

The first correction applied to the data is the subtraction of a background image. This is an image shot while no fluorescent dye was present in the measuring volume. Such image theoretically must be fully dark, as no scalar is present. However, due to pollution of the main flow with fluorescent dye by previous experiments and dark signal due to several camera noise contributions, this image is never completely dark. By subtracting this background image of all images taken, this offset light can be removed from the data, thereby improving the signal-to-noise ratio.

4.1.2 Correction for the Gaussian intensity profile of the laser sheet

The second correction concerns the laser sheet. One of the properties of the laser is that its beam has a Gaussian intensity profile. This is undesired as uniform illumination is preferred. It can be handled in two different ways.

One way is to measure the laser beam intensity profile by leading it through uniformly concentrated solution (see figure 4.1). The measured fluorescence field will be proportional to the intensity profile of the laser sheet. The low frequency variation in the span wise sheet direction can be attributed to the Gaussian profile of the laser beam. The high frequency oscillations in the measured intensity field can be attributed to irregularities in the used lenses and mirrors in the experimental setup. When normalising all data with this measured intensity profile, the non uniform illumination due to both aspects mentioned before, is corrected.

Another way of handling the problem of non uniform illumination is a more easy one. By making use of a cylindrical lens with a smaller focal length the laser sheet can be widened until the effect of the Gaussian profile becomes neglectable within the width scale of the measuring volume. Disadvantage of this method is that the
power of the illumination in the measuring volume is decreasing. A high power laser is required to maintain the intensity of illumination. Besides, using this method the high frequency fluctuations in the laser sheet due to irregularities in the used lenses and mirrors are not corrected. Both correction methods are applied in this experiment.

### 4.1.3 Correction for the displacement of the structures during measurement

The last correction applied concerns the deformation of the visualised structures while advected by the mean flow during measurement. In paragraph 3.4 it was calculated using dimensionless numbers and the Nyquist criterion that deformation of structures was not taking place. However, during the measurement it was noticed that a small translation of the structures by the mean flow was occurring during the measurement of one data volume. Therefore a procedure was designed to correct the data for this unwanted effect.

How advection during the measurement can lead to deformation is illustrated with figure 4.2. Obviously, if the local velocity is known it is possible to correct for this effect. To be as accurate as possible the mean velocity in one measuring volume is calculated at every captured measuring volume. Instead of capturing one volume at a time, three sequential measuring volumes are measured (see figure 4.3(a)). One image from the first volume and the last volume are lift out to deduce the displacement vector (figure 4.3(b)).

This displacement vector is calculated as follows. The two images are considered to be identical from which one is translated in comparison to the other. This can be assumed as turbulent structures are considered frozen (see paragraph 3.6), even in the time span of two measuring volumes. If the two images are represented by two different function \( f(x) \) and \( g(x) \), where \( x \) represents the distance to the centre of the images, they should satisfy
Chapter 4. Data Processing

Figure 4.3: Determination of the advection velocity. (a) shows three measured sequential volumes consisting of ten frames. The darker frame in the first and last volume are lifted out to determine the advection vector (b). Then the advection per frame is calculated (see text), which enables to correct the middle volume (resulting in (c))

\[ f(x) = g(x) * h(x), \]  
\[ F(s) = G(s) \cdot H(s). \]  
\[ H(s) = \frac{F(s)}{G(s)}, \] 
\[ h(x) = RFFT\{H(s)\}. \]

where \( h(x) \) is a translation function with a spike at translation \( d \). This translation \( d \) is the quantity of interest. The convolution from equation 4.1 becomes a multiplication in the Fourier domain:

\[ f(x) = g(x) * h(x), \]  
\[ F(s) = G(s) \cdot H(s). \]  
\[ H(s) = \frac{F(s)}{G(s)}, \] 
\[ h(x) = RFFT\{H(s)\}. \]

So if the fast Fourier transform is calculated from the two images and one is divided by the other (equation 4.3), then the translation vector between the images can
Figure 4.4: (a) An example of a 2D spike image. (b) and (c) Horizontal and vertical line out of a 2D spike function. Here, the maximum of the 2D spike function is found at pixel (129,138). The peak is sharp, hence the flow velocity is uniform over the entire images.
be deduced out of the reverse fast Fourier transform of this partition (see equation 4.4). A 2D spike function is obtained (see figure 4.4). The location of the spike in relation to the centre gives the translation vector of second image compared to the first. The width of the spike reflects the size of uniformity of the velocity over the images. A sharp peak indicates that the flow velocity is uniform over the entire image. Contrary, a wide peak indicates that the flow velocity difference within the image.

As can been seen in figure 4.3 this translation takes place over a time of 20 frames. So dividing this translation vector by 20 results in the advection per frame. Correction of the middle volume takes place by shifting each frame flow backwards over the advection distance. That is the advection in respect to the first frame of the volume. The pixels of the shifted image are filled with the value of the corresponding point in the original image. If the corresponding point is situated between the pixels of the original image (i.e. when the advection distance does not equal an discrete number of pixels), its value is calculated through linear interpolation of the four surrounding pixels, using the following algorithm:

\[ P(i+dx,j+dy) = \frac{(i+1,j) - (i,j))dx + ((i,j+1) - (i,j))dy + ((i+1,j+1) + (i,j) - (i,j+1) - (1+1,j))dx \cdot dy + (i,j). \]

\[ (4.5) \]

\( P(i+dx,j+dy) \) represents the point in the original data that corresponds with a pixel in the corrected data set. The four surrounding points are visualised in figure 4.5. This advection correction procedure results in a measuring volume as shown in figure 4.3 (c). From this volume the inclining sides are removed so a rectangle volume remains from where at all points the 3D concentration gradient can be calculated.

### 4.2 Power spectrum analysis

Power spectrum analysis are useful when studying the visualised concentration field.
4.3 Gradient algorithm

In this research power spectrum analysis consisted of calculating a probability density function of the space frequencies found in the images. It is assumed that if the visualised concentration field was characterised by small flow structures, a lot of high frequencies are found. Comparing the probability density functions calculated at different positions, a relation between the position in the reactor and the size of the flow structures can be deduced.

The following procedure was used to calculate the frequency probability density function. First one 2D image is taken out of each measuring volume (see figure 4.6(a)). It is multiplied with a window (figure 4.6(b)) in order to smoothen the borders. This prevents from finding high frequencies at the borders of the image. Then its 2D power spectrum is calculated by calculating the forward Fourier transform and multiply it with its complex conjugate. An example of an image and its power spectrum is shown in figure 4.6(c). The high values at the angle of 90 degrees are a result of systematic fluctuations in the span wise direction of the laser beam. Besides this line, the 2D power spectrum is not perfectly round, but it shows higher values around the angle of 35 degrees. This means that more, larger wave vectors are found in this direction. This can be validated in the original image, where it can been seen that most layers are found perpendicular to this direction. The frequency energy spectrum is obtained by integrating the 2D power spectrum over $|\mathbf{k}|$ to a 1D power spectrum. Normalising this spectrum with its total energy the frequency probability density function is obtained.

This procedure is applied on every volume measured. Then can be studied whether a relation exists between the size of the structures and the distance to the inlet of the scalar.

4.3 Gradient algorithm

After the application of the corrections the data can be studied. One of the aspects of interest in micromixing is the scalar gradient field. In this paragraph it will be
Figure 4.7: The calculation of the x-component of the scalar energy dissipation is based on rotations in the xy-plane. This rotation equals averaging over the three shown gradient calculations.

explained which algorithm is used to calculate the scalar energy dissipation field, which is closely related to the scalar gradient field (see paragraph 2.3). The scalar energy dissipation field $(\nabla \zeta \cdot \nabla \zeta)_{ijkl}$ is calculated out of the discrete scalar field $\zeta_{ijkl} \equiv \zeta(x_i, y_j, z_k, t_l)$. It is calculated by direct differentiation of the data using linear central difference approximations. These linear approximations can be based on a different number of data points, going from six up to twenty six surrounding points. Here, the scalar energy dissipation at each point $ijkl$ is calculated using the algorithm of [Dahm et al., 1991], based on 18 surrounding data points:

$$
(\nabla \zeta \cdot \nabla \zeta)_{ijkl} = \frac{1}{16} \left\{ \begin{array}{l}
\frac{1}{\Delta x} (\zeta_{i+1,j,k} - \zeta_{i-1,j,k}) \\
+ \frac{1}{\Delta x} (\zeta_{i+1,j+1,k} - \zeta_{i-1,j-1,k}) + (\zeta_{i+1,j-1,k} - \zeta_{i-1,j+1,k}) \\
+ \frac{1}{\Delta y} (\zeta_{i,j+1,k} - \zeta_{i,j-1,k}) \\
+ \frac{1}{\Delta y} (\zeta_{i+1,j+1,k} - \zeta_{i-1,j-1,k}) + (\zeta_{i-1,j,1+k} - \zeta_{i+1,j,1-k}) \\
+ \frac{1}{\Delta z} (\zeta_{i,j,k+1} - \zeta_{i,j,k-1}) \\
+ \frac{1}{\Delta z} (\zeta_{i+1,j,k+1} - \zeta_{i-1,j,k-1}) + (\zeta_{i-1,j,k+1} - \zeta_{i+1,j,k-1})
\end{array} \right\}^2
$$

(4.6)

The value for the scalar energy dissipation is obtained by rotations in the (x-y) and (x-z) plane. To illustrate this figure 4.7 is drawn. It shows a rotation in the xy-plane in order to calculate the x-component of the scalar energy dissipation. The y- and z-components of the scalar energy dissipation are calculated similar by applying rotations in the xy- and xz-plane respectively. These rotations are introduced to improve the signal-to-noise ratio.

4.4 Threshold

When studying the orientation of scalar gradient field a threshold is preferred. As can be seen in figure 3.1 images mainly contain background. This background represents the non-fluorescent main flow and the grey values in these areas should equal
4.4. Threshold

zero. However, due to experimental uncertainties this is not the case. Generally, after correction for the background and dark current noise, these areas will have low fluctuating values. When studying the scalar concentration field this values do not influence the result because of their low values. However, in the case of study of the isotropy of the scalar gradient field these low valued areas do influence the outcome. The orientation of the gradient vectors will be analysed and each background point will deliver a gradient vector. Due to their significant number, it can be easily seen that the background points influence the result. In this situation it is preferable to separate the background from the data. A threshold value is introduced to do so. Because this negative effect of the background only concerns the study of the orientation of the gradient vectors, the threshold is only applied on the scalar gradient field. If a histogram of the scalar energy dissipation is calculated (see figure 4.8, where this histogram is shown on logarithmic basis), it represent the scalar gradients calculated in the data (see paragraph 2.3). The distribution suggests that it is build up out of two Gaussian distributions. These distributions can be identified as one for the background data and one for the scalar marked regions. The best way to separate background from scalar regions is to insert a threshold at the local minimum of figure 4.8. Only the gradients that exceed this threshold value will be taken into account when the isotropy of turbulence is studied.

Figure 4.8: Probability density function of the natural logarithm of the scalar energy dissipation.
Chapter 5

Results and Discussion

In this chapter the results obtained with the 4D LIF experiment described in chapter 3 are presented and discussed. The results have been corrected with the procedures described in chapter 4.

In paragraph 5.1, the statistical significance and reproducibility of the 4D LIF experiment is tested by taking six independent measurements under the same conditions. In paragraph 5.2, measurements performed at three different distances downstream the injector are compared. Also a comparison is made between two measurements performed at similar positions but in different turbulent flows.

In paragraph 5.3, the mean concentration field and its variance at the different positions in the reactor is studied.

Finally in paragraph 5.4 and 5.5 the announced quantitative information is extracted from the data. The scalar energy dissipation field, introduced in paragraph 2.3, is calculated from the 3D concentration field at three distances from the feedpipe. Furthermore a study of isotropy of the turbulence field is performed.

5.1 Statistical validation of the measurements

Any statistically obtained quantity should be validated on its statistical significance. Sufficient independent measurements should be gathered before any value can be submitted to the result. In chaotic processes as turbulent flows, a large amount of data is required before significant statistics can be calculated. Therefore, first test measurements are performed to validate whether statistical quantities could be obtained properly.

This test measurement consisted of 950 realisations of 3D data volumes acquired at 15 cm downstream the feedpipe. At this position an uniformly distributed scalar field is expected. The injected dye had a concentration of $1 \cdot 10^{-6}$ mol/l. This means that beam attenuation is neglectable (see appendix A.1). Laser power during the test measurements was adjusted to 1.4W. This power level is insufficient to widen the laser sheet until the Gaussian profile becomes neglect, therefore the data is corrected by normalising it by the measured laser sheet profile (see paragraph 4.1.2).
Each measuring volume was separated by a half second pause in order to ensure that all realisations were independent. This independency of data was validated afterwards by calculating the auto correlation function of this measurement and of a measurement consisting of successively captured volumes (no pause). These two correlation functions are shown in figure 5.1. It can be seen in the auto correlation function of the successive volumes that over 15 volumes there is a significant coherence. This equals a time about 0.5 seconds. It can be seen in the auto correlation function of the measuring volumes separated by a half second do not show any correlation.

To study whether a sufficient amount of data is used, the mean concentration field, mean standard deviation field and mean skewness field are calculated. One frame out of each of these three mean volumes is shown in figure 5.2 with its normalised standard deviation (i.e. the standard deviation of the time averaged field normalised with the time and space average). It can be seen that the mean concentration field is smooth, the standard deviation field more intermittent and the skewness field is highly intermittent. This is also represented in the values for the normalised standard deviations. For the mean concentration field, standard deviation field and skewness field this quantity respectively equals 0.11, 0.14 and 0.21.

The three images in figure 5.2 indicate that sufficient data was gathered to calculate the mean concentration field and to a less extend the mean standard deviation field, as these images are fairly smooth. However, in the skewness images individual structures can be recognised, hence not sufficient data was acquired to calculate this 3rd moment field properly.

Note that this result can be meaningful when studying the flow. The fact that in the mean skewness field individual structures can be identified, while the mean concentration field is smooth, indicates that the structures with the highest concentrations are a minority in the flow. It appears that the majority of the structures has had the time to diffuse to lower concentrations. This is remarkable because in paragraph 3.6 it is argued that diffusion should be neglectable in this turbulent flow. The statement that the structures in fact do seem to diffuse is confirmed by figure 5.3 where a probability density function of the occurring concentrations in the flow is shown. There can be seen that a whole range of concentrations appear in the flow. Moreover, the maximum concentration occurring in the flow is half the injected concentration. Measuring such a wide range of concentration at such short distance downstream the inlet points to a more complex flow than expected in advance. It indicates that the residence time in the flow before passing the measuring volume is much larger than in a fully developed flow. This deviation is probably the result of the feedpipe.

Now that a series of 950 measuring volumes seems sufficient to calculate the mean concentration field, it is interesting to determine the minimal required number of measuring volumes. Therefore the mean concentration field is calculated using a variable number of measuring volumes. The standard deviation of these mean concentration fields as a function of the used number of measuring volumes is shown in figure 5.4. The minimal number of blocks required for valid results amounts 300.
5.2 Comparison different measurements

Another important aspect when validating experimental results is reproducibility. If the obtained results have statistical significance they should lead, ceterus paribus, to similar results. Therefore, the mean concentration, standard deviation and skewness fields are determined from six data sets, obtained under identical conditions. The normalised values for the time and space averages of the three fields are shown in figure 5.5. The mean concentration is normalised with the injected concentration, the standard deviation with the average concentration and the skewness is normalised with the standard deviation of the data set. The results appear to be is reproducible. Averaging in time and space also the skewness becomes reproducible. Despite the fact that high concentration structures are a minority, their overall contribution seems constant over several measurements.

Concluding, measuring 950 volumes is sufficient to calculate the mean concentration field and the standard deviation field. To calculate the skewness field properly, more data is required. However, although not statistically validated it still is a meaningful flow quantity. A study of proper calculation of the mean concentration field in relation the number of blocks processed, resulted in a minimal number of blocks required. This number is around 300 blocks consisting out of 10 frames. All time and space averaged results, including the skewness field, were reproducible.

5.2 Comparison different measurements

After the statistical validation through test measurements, the main measurements can be executed. This measurements consisted of 315 measuring volumes measured at three different positions in the tubular reactor. The injected concentration was
Figure 5.2: From left to right: a frame out of the mean concentration field (relative standard deviation=0.11), the mean standard deviation field (relative SD= 0.14) and the mean skewness field (relative SD=0.21).

Figure 5.3: Probability density function of the occurring concentration in the measured data set. The peak close to zero represents the background data. The PDF indicates that a wide range of concentrations appear. The injected fluorescence concentration was $5 \cdot 10^{-7}$ mol/l.
5.2. Comparison different measurements

Figure 5.4: The normalised standard deviation of the time averaged concentration volume as a function of the number of processed volumes.

Figure 5.5: Six measurements of the time and space averaged concentration, standard deviation and skewness, done to check reproducibility of the results.
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Figure 5.6: Two frames out of two different measurements. The left frame is shot at Re=4,200, the right frame is shot at Re=2,800

5.10^{-7} \text{ mol/l}, hence attenuation of the laser beam is considered neglectable (see appendix A.1). The laser power during these measurements was adjusted up to 4.5W. This power level made possible to widthen the laser sheet in order to make the Gaussian laser sheet profile neglectable. Converting grey values into concentrations was done by measuring the (linear) relation between fluorescence and concentration (see appendix A.2). It was found that 1 grey value equalled 9.510^{-10} \text{ mol/l}.

In this paragraph a comparison is made between measurements done under different conditions. First two measurements at different levels of turbulence are studied (i.e. a comparison between the test measurements and main measurements), then measurements at different distances from the inlet are compared (i.e. a comparison within the main measurement).

5.2.1 Different Reynolds numbers

Measurements are done at a Reynolds number of 2,800 and 4,200. These measurements consisted of 315 and 950 measuring volumes respectively. Both should be sufficient in order to determine quantities on a statistical bases (see paragraph 5.1). The acquired images are studied to see whether any systematic flow differences could be recognised. Two representative frames are shown in figure 5.6. Both images show a layer-like pattern at this position in the reactor. Based on the Reynolds numbers smaller structures may be found in the measurement at Reynolds 4,200 (see equations (2.2) and (2.3)). To verify this power spectrum analysis of the concentration field are performed, as described in paragraph 4.2. The resulting frequency PDF for the two different measurements are shown in figure 5.7. In the figure it can be seen that relatively the most high frequencies are found in the turbulent flow with Reynolds 2,800. This is in contradiction to what was expected on basis of the Reynolds numbers. Two remarks can be made concerning this result.

It may be doubted whether the comparison of these two measurements is legal. The
measurements might not be performed under identical conditions. There was a period of four months between the two measurements in which the setup had to be totally rebuild. A new laser and different lenses were used in the latest measurement. Due to these adjustments it may be questioned whether a comparison is allowed. The second remark concerns the calculation of the Reynolds number. The Reynolds number is based on the mean flow velocity. As mentioned before, the feedpipe is not taken into account. However at 15 cm downstream the feedpipe it is most unlikely that the feedpipe does not influence the flow field. The question may arise how representative these Reynolds numbers are for their measurement. The flow field downstream the inlet is inhomogeneous, which means that a small displacement in relation to the feedpipe can lead to a different flow velocity. The measurements were separated by a four months period, so a small displacement is not imaginary. The Reynolds numbers are not very accurate, hence they should be interpreted as an order estimation. Then the obtained result cannot be surprising.

5.2.2 Different distances to feedpipe

Measurements are performed at three distances downstream the inlet of the fluorescent dye. The first measurement was done at only 5 cm of the feedpipe, the second and third at respectively 15 and 37 cm. All measurements were performed successively using an identical setup. An image out of each measurement session is shown in figure 5.8.

The turbulent structures passing the measurement volume at 5 cm of the inlet are characterised by large sharp edged lumps of fluorescent dye. Here no multiple layers of dye and water are formed. The structures measured at such small distance downstream the feedpipe can be identified as the macro scale structures. They are characterised by little interfacial area, which means that little molecular diffusion is taking place.
Figure 5.8: Three frames measured at three distances downstream the feedpipe. The left frame is shot at 5 cm from the inlet, the middle at 15 cm and the right is shot at a distance of 37 cm downstream the feedpipe.

At 15 cm from the inlet multiple layers of alternating fluids appear. The cascade process has taken place, hence the occurring structures can be identified as the micro scale structures. Mixing is taking place, resulting in the appearance of diffusive areas.

At 37 cm the flow pattern has not changed significantly from the measurement at 15 cm. Still multiple layers of alternating fluids can be recognised and mixing is still taking place.

To validate the findings previously described, the power spectrum of the concentration field is calculated. It may be expected that positions which are dominated by small layer-like structures contain more energy on the higher frequencies. Contrary, positions which are characterised by large lump-like structures will contain more energy at the low frequency part of the spectrum. The power spectrum at three positions normalised with the total energy is shown in figure 5.9. The spectrum verifies the findings previously described. The 5 cm measurement contains relatively a lot of energy in the lower part of the power spectrum and relatively few energy in the high frequency part. As expected the 15 cm and 37 cm are very similar. These spectra contain relatively a lot of energy in the high frequency part, which is consistent with the found layer-like structures. As can be expected the most high frequencies, i.e. the smallest structures, are found furthest downstream the inlet.

5.3 Time averaged concentration field

The mean concentration field is calculated at three distances downstream the inlet. A single frame out of each time averaged 3D data volume is shown in figure 5.10. Their space and time averaged concentration are put below. Because of the low mean concentration these images are strongly enhanced (contrast stretched). For comparison also the relative standard deviation, i.e. the standard deviation within the field normalised with the time and space averaged concentration, is given.
First remark is the non uniformities in each mean concentration field. This in contrast to the mean concentration field calculated in paragraph 5.1. The question arises whether these non uniformities are a result of insufficient amount of data or whether this is in fact a flow phenomenon. To distinguish the flow aspects from statistical variance, the number of processed volumes is changed and at every different number of data volumes the calculation of the time average concentration field is redone. This is executed for all three measurements. In figure 5.11 the time and space averaged standard deviation normalised with the time and space averaged concentration is shown as a function of the number of processed data volumes. It is assumed that sufficient data is used when the relative standard deviation does not change with an increment of the number of used volumes. According to the left graph of figure 5.11 the result using measurement closest to the feedpipe is statistically found. A variation in the number of processed block does not change the result at all. The middle and right graph of figure 5.11 are very similar of shape. The relative standard deviation seems to have reached a constant value, but the number of blocks processed seems critical. However, this would point to the statement that all non uniformities in the mean concentration fields are systematic.

Concerning the mean concentration field of the 5 cm measurement another remark can be made. The areas with the highest concentrations are situated in the upper regions of the image. Remarkable is that this concentration is not found downstream the image (lower regions of the image), knowing that no photo bleaching is occurring within the measuring time (appendix B). It indicates a different flow direction than expected in advance. It would be most likely that structures are advected along the mean flow direction, from top to bottom in the images. However, it seems that structures did not pass the measuring volume along the mean flow direction in the tubular reactor, but crossed the measuring volume along the scanning direction. This is verified during the measurements where could be seen that close behind the
feedpipe a flow upwards was formed, perpendicular to the centre line of the tubular reactor. Therefore the non uniformities in this mean concentration field can be identified as a flow phenomenon.

In the case of the 37 cm measurement the unevenesses could also be result of a phenomenon found during the experiment. It was noticed that the fluorescent had a tendency to attach to the reactor wall at large distance from its inlet. At 37 cm downstream the inlet this effect was undoubtly present. The high concentrated area along the sides of the mean concentration image could be a result of this phenomenon.

However, in the case of the 15 cm measurement the non uniformities could not be verified during the measurement. It is noticed that the value of its relative standard deviation is significantly smaller than those of the other two measurements, so this time averaged volume is smoother than the ones calculated at other positions. But the remaining non uniformities cannot be explained.

Last remark concerns the time and space averaged concentrations of the three different measurements shown in figure 5.10. A decreasing average is expected further downstream the injector. This is confirmed in the measurements.

The time and space average concentrations are compared with the injected concentration and a calculated final concentration in the flow. The injected concentration was $5 \times 10^{-7}$ mol/l, so all measured average concentrations were much smaller. This is a result of the large amount of non scalar marked region which are measured.

From the mean flow velocity and the injection velocity the final concentration can be calculated. The flow rate is 14 l/min and 0.6 grams of fluorescent solution is injected per second, which results in a final concentration of $1.28 \times 10^{-9}$ mol/l. It appears that at 37 cm the average concentration approaches the final concentration.

It is difficult to derive qualitative information about the turbulent structures out of the concentrations averages. The large amount of the background dominate these averages. Moreover, it is not possible to distinguish whether high average concentrations are a result of the appearance of structures with high concentrations or a result of a large amount of structures with average concentrations. Therefore it is useful to study the probability density functions of the concentrations at different positions. Then a qualitative comparison can be made between the different measurement. The PDF for the concentrations measured at three positions in the tubular reactor are shown in 5.12. Now, it can be seen that at all three positions in the flow concentrations over a whole range exist. The measurement closest to the inlet of the fluorescent dye contains relatively the most structures with high concentrations. Contrary, the measurement furthest from the inlet contains the most structures with low concentrations. The measurement executed 15 cm downstream the feedpipe seems to have less high concentrations than the 5 cm measurement, though still more than measurement furthest from the inlet. All these findings are in accordance with the expectations.

### 5.4 Scalar energy dissipation field

After a study of the scalar concentration field, the scalar energy dissipation field can
5.4. Scalar energy dissipation field

Figure 5.10: Three enhanced frames out of the mean concentration fields at different distances to the inlet. The left image is calculated using the measurement at 5 cm (normalised standard deviation = 0.61), the middle uses the measurement at 15 cm (normalised SD = 0.23) and the right the one at 37 cm (normalised SD = 0.42).

Figure 5.11: Validation of the calculated mean concentration fields. The relation of the number of volumes used on the normalised standard deviation is studied. From left to right the results obtained using the 5, 15 and 37 cm measurement are shown.
Chapter 5. Results and Discussion

Figure 5.12: Probability density functions of concentration measured in the flow at different positions downstream the inlet.

Figure 5.13: The grey value over a line in one measurement is presented in the graph. The steepest gradients cover at least three pixels.
5.4. Scalar energy dissipation field

![Image](image_url)

Figure 5.14: Calculation of the scalar energy dissipation out of the measurements. The images show a single frame out of a measured data volume and the calculated 3D scalar energy field.

![Image](image_url)

Figure 5.15: Probability density functions of the scalar energy dissipation calculated from the three components of the scalar gradient field.
be calculated. First the scalar gradient field has to be calculated. This could be
difficult, as in paragraph 3.6 is argued that the occurring gradients are to steep to
detect, because the Batchelor scale is smaller than the space resolution. However,
while studying the data (paragraph 5.2) large diffusive region could be recognised
in the flow. This led to the idea that scalar energy dissipation was determinable.
To found that statement the grey value profile of a sharp edged image was studied
(see figure 5.13). The grey values along the line in the image, are shown in the
graph. It can be seen that even the steepest concentration gradients go over at least
three pixels. This concludes that, despite previous expectations, the gradients can
d be determined with this setup.
As mentioned before in paragraph 2.3 the scalar energy dissipation is a good quan-
tification for molecular mixing rate. Obviously molecular mixing takes place mainly
at the interfacial areas of the scalar marked regions. Calculating the scalar energy
field of one measuring volume should result in a field were the edge of the turbulent
structures are enhanced. Such operation is executed and the result is shown in figure
5.14. The result satisfies the expectations, meaning that the highest values are
located at the the interfacial areas of the two fluids.
Now, the probability density functions of the scalar energy dissipation over an entire
measurement can be calculated. The result obtained using the measurement at dif-
ferent positions in the reactor is shown in figure 5.15. The scalar energy dissipation
is normalised by a constant $\nabla \zeta^*$, which equals a the energy dissipation due to a
scalar gradient of 1 grey value per pixel. The PDF’s turn out to have a large peak

\[ \ln(\frac{\nabla \zeta}{\nabla \zeta^*})^2 \]
5.4. Scalar energy dissipation field

at low concentration gradients and a side lob to the higher gradients. The large peak represents the gradients calculated in the background regions. The side lob at high concentration gradients represents concentration gradients in the scalar marked regions and will be referred to as the signal part of the PDF. Studying the scalar energy dissipation of the scalar field, only the signal part of the PDF is of interest. A remark has to be made concerning the measurement 5 cm downstream the feedpipe. By accident this measurement is executed injecting half the concentration used in the other two measurements. Because there is a linear relation between concentration and fluorescence, it is possible to scale up this measurement. This scale up causes a shift right in the PDF of this measurement. This shift right also affects the noise peak. This is a result of the worse signal-to-noise ratio of this measurement. For a study of micro mixing only the signal part of the PDFs is of interest. Therefore in figure 5.16 the signal part of all three measurements is plotted in one graph. It is the expectation that further from the feedpipe the edges of the structures become less steep and that through the formation of layer-like structures the amount of interfacial area increases. In terms of scalar energy dissipation this would result in a decreasing signal peak shifting to the higher values of the scalar energy dissipation when going further downstream the feedpipe. This expectation is confirmed by figure 5.16. It can be seen that the highest and lowest values for the scalar energy dissipation are measured respectively at 5 and 37 cm from the feedpipe. Besides, the largest and least amount of scalar energy dissipation (i.e. the area of the signal peak) is measured respectively at 37 and 15 cm from the feedpipe. However, this change in amount of scalar energy dissipation is not convincing. The absence of a large increment in the amount of scalar energy dissipation when going further from the feedpipe is a result of a difference in the amount of turbulent structures measured. Due to dispersion of the scalar, less structures are measured further from the inlet. Hence, the increment in scalar energy dissipation by creating layer-like structures further downstream the inlet, is being reduced by the fact that at those positions less structures are measured.

Studying the PDF's of the scalar energy dissipation, the measurement performed 15 cm from the inlet is situated between the other two measurements. This agrees with the expectation.

It is checked whether the outcomes are not a result of statistical variance but a systematic flow aspect. Therefore, again the number of processed volumes is adapted. The PDF's obtained on the basis of a different number of measuring volumes for the 37 cm measurement are presented in figure 5.17. When using few measuring volumes the outcome was unstable. However, when the number of processed blocks exceeded 150 volumes, the outcome became reproducible. This test is performed for all three measurement and the outcomes were similar. Therefore it can be posed that the PDF's calculated using 315 measuring volumes from figure 5.16, were statistically significant.
5.5 Isotropy of the turbulence field

After calculating the scalar gradient field in order to determine the scalar energy dissipation, the next step is to study the isotropy of the turbulent flow. [Nieuwstadt, 1998] considers turbulence on the macro scale anisotropic and on the micro scale isotropic. The anisotropy of the macro scale turbulence is a consequence of the fact that turbulence extracts its energy out of the main flow which obviously has an angle of preference. However, along each step of the cascade process information concerning this anisotropy is lost, therefore the micro scale is considered isotropic.

In the used flow system it is difficult to give any expectations about the isotropy of the turbulent flow. The influence of the feedpipe is unknown and as already concluded before, it is unlikely to suppose that it does not disturb the turbulence field. This makes predictions about the isotropy of the flow a difficult issue. Although measuring the micro scale it is unknown if an isotropic turbulence field can be expected.

The study of isotropy is done through a study the orientations of the gradient vectors. First a histogram is made of the scalar gradients in three directions occurring in the flow. The results of this calculation at three positions in the reactor is shown in figure 5.18. As can be seen a whole range of gradients appear in the flow. In a isotropic flow the contributions of three directions to the scalar energy dissipation should be equal. It can be seen that this is only the case in the measurement at 37 cm. At the other positions the z-gradient slightly dominates.

The next step is to determine the gradient vectors in polar coordinates. The exact
5.5. Isotropy of the turbulence field

Figure 5.18: Probability density functions of the concentration gradients in three directions occurring in the turbulent flow. The peak at the lowest gradients regions is due to the background data.

Figure 5.19: Definition of parameters used in the study of isotropy.
Chapter 5. Results and Discussion

Figure 5.20: The probability density functions of the angles of the concentration gradient vectors in an isotropic system.

Definition of the polar parameters is shown in figure 5.19. When studying the probability density functions of the two spherical angles in the case of isotropic turbulence, the results are evident. In a isotropic turbulent flow the scalar gradient vector is expected to have no angle of preference. This results in a PDF for $\phi$ that shows a constant value of $(2\pi)^{-1}$ and in a PDF for $\theta$ that shows half a period of a sine function with a maximum equal to a half (see figure 5.20).

In paragraph 4.4 is posed that for a correct study of isotropy a threshold should be introduced. To validate this assumption first the PDF’s of $\phi$ and $\theta$ are calculated without making use of a threshold value. The calculated PDF’s are presented in figure 5.21. As expected the results turn out very noisy. This is obviously the case because of the large amount of scalar gradient vectors belonging to the background. Low value gradients are highly influenced by discretisation because then the discretisation steps are relatively large. This introduces the peaks in the phi PDF. A threshold is inserted to separate the data of interest from the background data. After inserting a threshold, as described in paragraph 4.4, the PDF of the dissipation energy result as presented in figure 5.22. It is verified that the threshold works properly.

Consequently, the PDF’s of the angles of the concentration gradient vectors are calculated using a proper threshold and shown in figure 5.23. The result turns out remarkable. The high frequency fluctuating part has been almost removed, so its presence could be attributed to the background noise. One low frequency fluctuation around the predicted value of $(2\pi)^{-1}$ remains in the $\phi$-PDF. The PDF for $\theta$ has changed dramatically. It shows a preference to the $z$-coordinate. This was noticed before in the scalar gradient histogram (figure 5.18). It was checked whether the outcome was not a statistical variance by changing the number of blocks. The outcome appeared to be stable.

The same calculation is performed for the other to positions in the reactor (figure 5.24 and 5.25). Also the measurement at 15 cm shows the preference for the $z$-coordinate. This in accordance with the result obtained in figure 5.18. The measurement done at the furthest position from the feedpipe shows a different PDF. It shows a preference for all three coordinates.
5.5. Isotropy of the turbulence field

Though the results were statistically significant, it is difficult to find a physical explanation for the found PDF's. Therefore it is hard to judge the found anisotropy.

The question may arise whether the data processing is performed correctly or whether the measuring method influences the result. Therefore two tests are performed. First a study of isotropy is done on a generated white noise image. It is assumed that the gradient vectors in this image are isotropic. The result of the PDF for theta is shown in figure 5.26(a). The result satisfies the expectations so it is concluded that the data processing works properly.

Second noise measured during the experiment is studied. A study of isotropy of the background should have a comparable result as the generated white noise. In this way it is possible to check whether the measuring method introduces some anisotropy. A way of studying measured noise is to use the threshold as an upper limit, thereby separating the background data. The calculated theta-PDF for the background data is shown in figure 5.26(b). The calculated PDF approaches the curve of isotropy, but it has a large peak at $\theta = \pi/2$. This peak is a result of an effect which only occurs in the plane of the images. The laser sheet does not have an homogeneous intensity profile (figure 4.1). Due to the laser and the lenses the intensity profile contains oscillations. This effect only exists in the plane of the laser ($\theta = \pi/2$), as the laser sheet is traversed up and down. Therefore the gradients in the xy-plane are little overestimated. This affects mostly the gradient which are near the xy-plane because their sensitivity for fluctuations is larger than the gradients along the z-coordinate. This effects makes that the PDF is flatter than the isotropic curve. The fluctuations in the laser sheet introduce a very small preference to the z-coordinate.

However despite this effect which provokes the peak in the PDF of theta, the PDF approaches very much the isotropic curve. Therefore the conclusion is drawn that the experiment does not influence the result significantly.

This points again to the conclusion that the PDFs calculated using the experimental data are valid. It indicates that the found anisotropy is a flow phenomenon.
Chapter 5. Results and Discussion

Figure 5.22: The probability density function of the scalar energy dissipation calculates from the 5 cm measurement after inserting a threshold. Concentration gradients below the threshold value are rejected.

Figure 5.23: The probability density functions of the angles of the concentration gradient vectors using the measurement at 5 cm distance to the feedpipe, after inserting a threshold value.
5.5. Isotropy of the turbulence field

Figure 5.24: The probability density functions of the angles of the concentration gradient vectors using the measurement at 15 cm distance to the feedpipe, after inserting a threshold value.

Figure 5.25: The probability density functions of the angles of the concentration gradient vectors using the measurement at 37 cm distance to the feedpipe, after inserting a threshold value.
Figure 5.26: (a) The probability density functions of theta calculated from a generated white noise image. (b) The probability density functions of theta calculated from the background data. The background data is separated from the rest of the data using the threshold as upper limit.
Chapter 6

Conclusions and Recommendations

6.1 Conclusions

• In this thesis, the design of, and results with a LIF setup for measuring 3D concentration fields in a tubular reactor at Re=2,800 were presented. It was shown that small scale flow structures could be resolved in both space and time.

• Turbulent structures were studied at three positions in the tubular reactor. Through concentration probability density functions insight was obtained in the development of the structures in the flow. It was found that the concentrations decrease in the downstream direction. Through power spectrum analysis different flow scales were distinguished. It was found that the structures became smaller in the downstream direction. The results agree with preliminary expectations.

• From the measurements, the scalar energy dissipation has been calculated from all three scalar gradient vector components. The results show a low signal-to-noise ratio. At the macro scale, the highest values for the scalar energy dissipation were found. This was due to sharply edged structures present at this scale. At the micro scale, the largest amount of scalar energy dissipation is expected to take place. This is a result of the formation of layer-like structures at these scale, which cause an large increment of interfacial area. A comparison of the amounts of scalar energy dissipation between micro and macro scale was difficult, as the number of structures measured was not constant and difficult to control. Nevertheless, the tendency seemed to be consistent with the expectation described before.

• A study of isotropy on the calculated scalar gradient field was performed. A threshold was inserted to separate noise from signals. Anisotropy was found at all measured positions in the tubular reactor.

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6.2 Recommendations

• Perform measurements at more positions in the tubular reactor. Three positions is few in order to construct a proper view on mixing as a function of the distance to the injection point.

• Perform measurements along the diameter of reactor to obtain a radial profile. In this way the calculated scalar energy dissipation field at different positions will be better to compare, as a equal amount of scalar will pass.

• Perform measurements at maximum laser intensity. This will increase the signal-to-noise ratio. No measurements were performed at maximum laser intensity, as it was unknown whether the galvano mirror could stand such high intensities.
Appendix A

Laser intensity in relation to concentration

A.1 Laser attenuation

Measurements are performed in a box containing a uniform fluorescence concentration. At a constant laser power of 686 mW, the concentration is increased from $1 \times 10^{-7}$ mol/l to $8 \times 10^{-7}$ mol/l in steps of $5 \times 10^{-8}$ mol/l. The local fluorescence intensity is calculated by averaging the measured fluorescence intensity over twenty-five neighbouring pixels at a position of two centimeters from the wall. This means that at this position the laser beam has passed through two centimeters of the uniform concentration. The result is shown in figure A.1. It can be seen that attenuation of the laser beam starts to be significant when the beam crosses two centimeters in concentration of $4 \times 10^{-7}$ mol/l.

In the experiment measurements are performed using two different concentrations of fluorescent dye. The main measurements are performed injecting a fluorescent dye with concentration of $5 \times 10^{-7}$ mol/l while in the test measurements a concentration of $1 \times 10^{-6}$ mol/l is used. In both experiments the dye is led into the main flow through a two millimeter diameter tube. This imposes the maximum path-length through the initial concentration to be in the order of 2 mm. Therefore, it is acceptable to consider attenuation of the laser beam in all measurements is neglectable.

A.2 Converting grey values into concentration

Using the linear part of figure A.1 it is possible to convert grey values into concentrations. According to the linear function between concentration and fluorescence intensity, a fluorescence intensity of 23.5 grey values is measured at concentration of $1 \times 10^{-7}$ mol/l. The dark current and dye pollution of the tube flow cause a grey value offset of 7.5 grey value. Hence, 16 grey values are a result of fluorescence of the dye molecules. This experiment is executed with a laser intensity of 686 mW, however the main measurements are performed using a 4.5 W laser intensity. Using
Appendix A. Laser intensity in relation to concentration

Figure A.1: Fluorescence intensity in grey values as a function of the concentration in a uniformly concentrated box. The fluorescence intensity is measured 2 cm from the side of the box.

the linear relation between fluorescence and laser power it can be calculated that at laser power of 4.5W this concentration would have led to a fluorescence intensity of 105.6 grey values. From this can be calculated that, at laser power 4.5W, 1 grey value represents a dye concentration of $9.5 \times 10^{-10}$ mol/l.
Appendix B

Photo bleaching

Photo bleaching is the phenomenon of decreasing fluorescence in time when a high powered laser beam is put through a fluorescent concentration. Beside excitation and fluorescence, the laser beam also ionises the fluorescent dye. In time the concentration decreases and therefore also the fluorescence intensity. To quantise this phenomenon a laser beam of 4 Watt is put through a uniform concentration of $1 \cdot 10^{-7}$ mol/l. Attenuation of the laser beam can be considered neglectable (see appendix A.1). The space average concentration is measured in time and the result is shown in figure B.1. The decrement of the fluorescence intensity is in the order of 0.15 grey values per second.

The residence time of the dye in the laser beam can be estimated using the length of the measuring volume and the mean flow velocity. The length of the measured area is 2.5 cm and the mean flow velocity is in the order of 3 cm/s. This result in a residence time of the fluorescent dye in the laser beam in the order of one second. It may be concluded that photo bleaching is taking place at a larger time scale than the measurement time scale. Therefore photo bleaching is considered as not taking place.
Figure B.1: Local averaged fluorescence intensity in grey values as a function of time
Appendix C

Laser sheet waist

When a laser beam is focussed in a point, it has a minimal spot size $W$, given by

$$W = \frac{\lambda}{\pi \theta n_m},$$  \hspace{1cm} (C.1)

where $\theta$ is the angle of the laser beam in a medium with a refractive index $n_m$. The minimal spot size is affected by the lenses, changes of medium and the perspex and glass walls in the setup. After each of these changes, a new minimal spot size (i.e. waist) has to be calculated. This is done using the theory of [Yariv and Yeh, 1984]. It defines a radius of curvature $R$ of the laser beam as a function of the position $x$ along the laser beam direction as,

$$R(x, W_0, z_0, z_w) = (x - z_w) \times (1 + \left(\frac{z_0}{x - z_w}\right)^2), \hspace{1cm} (C.2)$$

where $W_0$ is the initial waist, $z_w$ is the location of the new waist and $z_0$ a scale parameter. Besides a radius of curvature, also the new waist is defined.

$$W(x, W_0, z_0, z_w) = W_0 \times \sqrt{(1 + \left(\frac{x - z_w}{z_0}\right)^2}). \hspace{1cm} (C.3)$$

The two beam parameters are combined in one complex beam parameter. Using the A,B,C,D law, after each lens or change of medium, a new complex beam parameter is calculated. Starting from the initial laser beam parameters, the laser beam parameters in the tubular reactor can be calculated.

To verify the initial laser beam waist and angle, measurements of the beam waist outside the reactor are performed. A cylindrical lens with focal length of 250 mm was used. The results are compared with the outcome of the model of [Yariv and Yeh, 1984], see figure C.1. The model agrees with the measurements.

Now that the initial laser beam parameters are verified experimentally, the minimal spot size of the laser beam in the reactor can be calculated using the model of
Appendix C. Laser sheet waist

Figure C.1: Calculated and measured laser beam in air profile using a spherical lens with a focal length of 250 mm

[Yariv and Yeh, 1984]. The calculated laser beam profile in the tubular reactor with a cylindrical lens with a focal length of 500 mm, is shown in figure C.2. It can be seen that the minimal sheet thickness equals 121 μm at the e⁻²-border. The sheet thickness at the borders of the measuring volume equals 151 μm. The exact distances in between the several components in the setup are given in the table below.

<table>
<thead>
<tr>
<th>Distance</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Distance laser - galvano mirror</td>
<td>2.77 m</td>
</tr>
<tr>
<td>Distance galvano mirror - cylindrical lens</td>
<td>2.5 cm</td>
</tr>
<tr>
<td>Distance cylindrical lens - spherical lens</td>
<td>35 cm</td>
</tr>
<tr>
<td>Distance spherical lens - perspex wall</td>
<td>49.7 cm</td>
</tr>
<tr>
<td>Thickness perspex wall</td>
<td>9.8 mm</td>
</tr>
<tr>
<td>Distance perspex wall - glass wall</td>
<td>5.9 mm</td>
</tr>
<tr>
<td>Thickness glass wall</td>
<td>5.3 mm</td>
</tr>
<tr>
<td>Distance glass wall - centre measuring volume</td>
<td>5 cm</td>
</tr>
</tbody>
</table>
Figure C.2: Calculated laser beam profile in the tubular reactor using a spherical lens with a focal length of 500 mm
Appendix C. Laser sheet waist
Appendix D

Lattice-Boltzmann simulation of the flow field

A Lattice-Boltzmann simulation of the velocity field in the tubular reactor at a Reynolds number of 4,200, has been performed. A cross section is shown in figure D.1. It can be seen that the feedpipe disturbs the velocity field significantly. The fully developed tubular flow returns 15 cm downstream the feedpipe.
Figure D.1: A cross section from the Lattice-Boltzmann simulation of the velocity field in the tubular reactor. The diameter of the reactor is 10 cm.
Bibliography


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