Measurement of micro mixing in a tubular reactor using a four-dimensional Laser Induced Fluorescence technique

E. van Vliet  J.J. Derksen  H.E.A. van den Akker

Kramers Laboratorium voor Fysische Technologie, Delft University of Technology, Prins Bernhardlaan 6, 2628 BW Delft, The Netherlands

Abstract

The mixing of a passive scalar into the turbulent flow of a tubular reactor is studied at Re=4,000. In the experiment a fluorescent dye is injected into the flow system. The mixing process is visualised by exciting the dye molecules with a sheet of laser light. The resulting fluorescence, being proportional to the local dye concentration, is imaged with a high speed digital camera, yielding two-dimensional images of the concentration field. By sweeping the laser sheet in the depth direction, parallel to itself, a three-dimensional realization of the concentration field can be reconstructed. As such 3D concentration fields can be captured successively in time, the technique will be denoted as four-dimensional laser induced fluorescence (4D-LIF). Using the 4D-LIF technique, the time evolution of the three-dimensional scalar field is measured. From the measurement data, all three scalar gradient vector components could be obtained, resulting in a true estimate of the scalar energy dissipation field.

Fig. 1. The concentration (a) and square root of the scalar energy dissipation (b) at the faces of a three dimensional data volume consisting of 256×256×50 data points.
1 Introduction

Mixing of initially separated fluids is usually done with the purpose to bring about a certain mass exchange of reactants dissolved in the fluids. In case of competing, diffusion limited reactions, it is well known that mixing strongly influences the yield of products. Mixing at the smallest dynamical scales of a turbulent flow is especially important since at these scales molecular diffusion and hence chemical reactions take place. This stage of mixing is denoted as micro mixing. At the micro mixing scales the flow can be considered to be laminar, consisting of vortices that are being stretched due to the action of strain of the large scale flow.

The behaviour of any conserved scalar quantity $\zeta(x,t)$ can be exactly described by the advection-diffusion equation. Moreover, the scalar energy can be considered, defined in analogy to the kinetic energy as $\frac{1}{2}\zeta^2(x,t)$. The scalar energy satisfies the transport equation

$$\left(\frac{\partial}{\partial t} + u \cdot \nabla - ID\nabla^2\right)\frac{1}{2}\zeta^2 = - ID\nabla\zeta \cdot \nabla\zeta,$$

where $ID$ is the scalar diffusivity. The left-hand side describes the advection and diffusion of the quantity $\frac{1}{2}\zeta^2$. Since the right-hand side is always negative, it can only diminish the scalar energy and hence should be interpreted as a scalar energy dissipation, defined as

$$\chi \equiv ID\nabla\zeta \cdot \nabla\zeta.$$

The scalar energy dissipation is an appropriate measure for the local molecular mixing rate. Determination of the true scalar gradient field $\nabla\zeta(x,t)$ and the associated scalar energy dissipation $\chi$ requires measurement of the scalar quantity in all three spatial dimensions.

Simplification of the exact scalar transport process is usually required for yield predictions of chemical reactions following molecular mixing. Mechanistic micro-mixing models (e.g. Baldyga and Bourne, 1984; Bakker, 1996) use a deterministic approach to solve the small scale mass balances of reactants over a stack of layers of alternating fluids. Vortex stretching is accounted for by assuming the thickness of these layers to shrink in time. Yield predictions strongly depend on the way these ideas of shrinking layers are incorporated into the model. As a result, assessment of micro-mixing models calls for visualisation experiments. Since a stack of layers can be orientated into any direction, the visualisation technique is preferably three dimensional.

In this paper we present the design and first results of an experimental set up that measures the time evolution of a three dimensional concentration field of a dye mixing into the turbulent flow of a tubular reactor (TR). We use a Laser Induced Fluorescence (LIF) technique for non-intrusive measurement of the concentration of the dye. The fluorescent dye is excited by means of a
Fig. 2. Overview of the key elements of the experimental setup, consisting of a laser illumination source, sheet forming optics, a high speed digital CCD camera, a PC to capture the measurement data and some electronics to slave the galvano mirror used to sweep the laser sheet in the depth direction.

A high speed digital CCD camera is used to capture two dimensional images of mixing structures. Scanning the laser sheet into the depth direction yields a series of subsequent closely spaced images from which the three dimensional concentration field can be reconstructed. Repeating the process yields the time evolution of the 3D scalar field, hence the technique will be denoted as four-dimensional laser induced fluorescence (4D-LIF).

The optical setup resembles the one developed by Dahm et al. (1990), who successfully studied the mixing driven by a jet in a stagnant liquid. Compared to the latter case, the flow in the tubular reactor is characterised by smaller flow structures, whereas the advection of the bulk velocity puts higher demands to the temporal resolution of the experimental setup. By using recently developed, high speed CCD cameras, we are able to achieve a sufficiently high temporal resolution, combined with an adequate dynamic range of length scales.

2 The optical arrangement

A schematic representation of the optical arrangement is given in figure 2. The blue line of an argon ion laser (continuously emitting 1.4 W) is used as a light source. The laser beam is converted into a horizontal sheet by means of a negative cylindrical lens \( f = -25.4 \) mm. A galvano mirror positioned in the focal point in front of this cylindrical lens is used to scan the laser beam in the depth direction. The galvano mirror is slaved by the CCD camera in a way that every \( N_z \) frames taken by the CCD camera, the laser sheet scans the flow from its uppermost position downwards. After each scan, the sheet is
Fig. 3. Schematic representation (a) and artists’ impression (b) of the tubular reactor at the position of the injector. The internal reactor diameter D_i, the external injector diameter D_f and injector feed opening D_i are 0.10 m, 0.02 m and 2 mm, respectively. To avoid image deformations due to the cylindrically shaped tube wall, a square, transparent box filled with water has been installed around the tube.

instantaneously stepped back to its initial position. A positive spherical lens (f = 300 mm) has been positioned at its focal distance from the galvano mirror. This way, the scanning laser sheet moves parallel to itself after being directed through the lens. Moreover, as the focal points of the cylindrical and the spherical lens coincide, the sheet’s divergence in horizontal direction is compensated for. Finally, the spherical lens has been positioned at such a distance from the flow facility, that the thickness of the laser sheet reaches its minimum of about 100 μm at the measuring position in the flow.

The dye in the tubular reactor that is excited by the sweeping laser sheet is imaged by means of a high speed digital CCD camera positioned above the TR with its optical axis perpendicular to the laser sheet. The CCD camera captures 1000 frames per second on a light sensitive area of 256 times 256 pixels. Each single pixel represents the fluorescence intensity in 256 levels of grey values (1 byte/pixel). The scalar concentration can be obtained from the fluorescence intensity as the two are proportional in the low concentration limit. The image data flow of about 62 Mb/s was stored in RAM of an NT workstation. As one Gb of memory was available, measurement series up to 15 seconds could be performed.

3 The flow facility

The reactor consists of a perspex tube, D_i=0.10 m in diameter, in which a stationary, moderately turbulent flow of water is established at Re=4,000 (see figure 3). The injector, having an external diameter D_f = 0.02 m, was used to add the fluorescent dye to be mixed with the main flow. In this paper, results of measurements that were done at 0.15 m downstream the injector, 1.5 cm
Fig. 4. The instantaneous velocity field of the turbulent wake downstream the injector. The result has been obtained with a large eddy simulation based on a lattice Boltzmann scheme (see Derksen and Van den Akker, 1999). The rectangle indicates the size and position of the measurement volume with respect to the injector. In this paper, the $x-$, $y-$, and $z-$axis point to the stream wise, depth (into the paper) and upwards direction, respectively.

above the centre line of the reactor, will be presented.

The Kolmogorov length and time scale depend on the local energy dissipation $\epsilon$ according to $\eta = (\nu^3/\epsilon)^{1/4}$ and $\tau = (\nu/\epsilon)^{1/2}$, respectively. The dimensionless energy dissipation $\epsilon D_t/(2u_*^2) = \tau D_t^4/(2\nu^3 Re^3)$ (with the Reynolds number now based on the friction velocity) is about 2.2 on the centre line of a 1-D pipe flow (Hinze, 1959). The Reynolds number $Re_s$ is related with $Re$ according to $Re_s = 0.28Re^{7/8}$ (Bradshaw, 1976), yielding the energy dissipation to be about $3 \times 10^{-6} \text{m}^2\text{s}^{-3}$ for $Re=4,000$. This results in a Kolmogorov length scale of about $\eta=0.8 \text{ mm}$ and a Kolmogorov time scale of about $\tau=0.6 \text{ s}$.

The spatial resolution of the measurement system is put by the Bachelor scale $\eta_B$, i.e. the smallest length scale on which scalar gradients can be sustained in the flow. The Bachelor length scale can be interpreted as the diffusion length $\sqrt{ID\tau}$ of a scalar within one Kolmogorov eddy life time and is therefore related to the Kolmogorov length scale according to $\eta_B = Sc^{-1/2}/\eta$. The Schmidt number is introduced as the ratio of vorticity diffusivity $\nu$ and the scalar diffusivity $ID$: $Sc=\nu/ID$. The Schmidt number of the fluorescent dye used in the experiment (a disodium fluorescein solution) is given to be 1,930, yielding a Batchelor scale of the order of 20 $\mu m$. The temporal resolution of the measurement system is put by the the Kolmogorov advection time scale $\tau_a = \eta/\overline{U}$, interpreted as the time required for a Kolmogorov eddy to pass a fixed point when advected by the mean velocity. Since the mean velocity is 0.04 m s$^{-1}$, the Kolmogorov advection time is of the order of 20 ms.

The radially inserted injector will complicate the flow field significantly. Large scale flow structures are formed in its wake, and mixing will be enhanced. A large eddy simulation has been carried out to study the flow in the reactor in more detail. An impression of the instantaneous velocity field of the wake downstream the injector is given in figure 4. From the simulation it appears that the energy dissipation at undisturbed flow regions agrees reasonably with
a 1-D pipe flow. In the near regions downstream the injector the dissipation is increased significantly, but at some distance away where the measurement were carried out, the dissipation recovers almost completely. For estimating the length and time scales based on the local energy dissipation it is hence reasonable to assume that at this position a 1-D pipe flow can be considered.

4 Spatial and temporal resolution

The 4D-LIF experimental set up has been designed to resolve the smallest flow structures in both space and time. This means that the spatial and temporal resolution had to be small compared to the Kolmogorov length scale $\eta$ and the Kolmogorov advection time scale $\tau_a$, which were estimated to be of the order of 0.8 mm and 20 ms, respectively.

Figure 5 shows a schematic of the 4D-LIF data space. The spatial resolution in the depth direction ($\Delta z$) is determined by the distance between two successive data planes within one data volume, which is set to be 160 $\mu$m. The thickness of the laser sheet should be smaller than this distance. Therefore the average laser sheet thickness over a single data plane has been set to be about 100 $\mu$m. The pixel size of the array sensor of the CCD camera is $l_p \times l_p = 10 \times 10 \mu$m$^2$. The magnification $m$ of the collection optics (defined as the ratio $l_p / \Delta x$) equals 0.091, hence each pixel corresponds to an area of $\Delta x \times \Delta y = 91 \times 91 \mu$m$^2$ in a data plane in the flow. The voxel size of $\Delta x \times \Delta y \times \Delta z = 91 \times 91 \times 160 \mu$m$^3$ is sufficiently small to resolve the Kolmogorov scales. The Batchelor scale, estimated to be of the order 20 $\mu$m, is not fully resolved though.

As each data plane consists of 256 $\times$ 256 pixels, its size $L_x \times L_y$ in the flow is $23.3 \times 23.3$ mm$^2$. The size of the data volume in the depth direction depends on the number of data planes $N_z$ according to $L_z = N_z \Delta z$. Choosing a large number of data planes improves the dynamical range of length scales resolved in the depth direction. A trade-off exist, however, between this dynamical range and the temporal resolution $\Delta T$ to acquire the data volumes. Increasing $N_z$ degrades the temporal resolution according to $\Delta T = N_z / f_{\text{CCD}}$ ($f_{\text{CCD}}$ being the camera frame rate of about 950 fps). For visualisation purposes the number of data planes per volume is chosen to be as large as 50, giving $L_z = 8$ mm combined with a temporal resolution of $\Delta T = 52$ ms. For determining the scalar energy dissipation only 3 data planes would be required to measure the component of the scalar gradient vector in the depth direction. In our case, $N_z$ was chosen to be 8, giving a depth of the data volume of $L_z = 1.3$ mm combined with a temporal resolution of 8.4 ms.

The spatial resolution of the measurement system estimated so far really is an upper limit. As the image plane moves with respect to the camera, care has to be taken to capture sharp images. The highest detectable frequency at the sensor array, denoted as the array limited cut off frequency, depends on the pixel to pixel distance $l_p$ according to $f_{\text{c.array}} = 1/(2l_p)$, which corre-
Fig. 5. (a) Schematic representation of a data volume consisting of $N_z$ closely spaced data planes of $N_p$ times $N_p$ pixels each. (b) The (4D) data space consisting of (3D) data volumes captured successively in time, mapping the conserved scalar concentration field in the $L_x \times L_y \times L_z$ volume in object space. The data volumes are sampled at a temporal resolution $\Delta T$ on a $N_p \times N_p \times N_z$ grid of $\Delta x \times \Delta y \times \Delta z$ sample volumes.

Responds to $m/(2l_p)$ in the flow field. Next to the sensor array, however, also the lenses of the collection optics impose a constrained to the maximum detectable frequency, which is known as the diffraction limited cut of frequency $f_{c,lens}$. The cut off frequency of the entire system, $f_{c,\text{total}}$, is then determined by $f_{c,\text{total}} = f_{c,\text{array}}^{-1} + f_{c,lens}^{-1}$. For an in focus object, $f_{c,lens}$ is much larger then $f_{c,\text{array}}$, hence in this case the cut off frequency of the system is totally determined by the latter. Focussing errors, however, reduce the diffraction limited cut off frequency severely. For a lens with finite aperture and focussing error $\delta$ (the absolute value of the difference between the ”in-focus” object distance and the actual distance), the diffraction limited cut off frequency can be written as (Hopkins, 1955)

$$f_{c,lens} = 1.22\frac{f^2(1 + m)}{m^2\delta},$$

where $f$ is the lens f-number defined as the ratio of the focal length and the aperture diameter of the lens, $f/d$. The maximum focussing error $\delta$ is typically as large as half the the depth of the measuring volume $L_z/2$. Hence, increasing $L_z/2$ at some point starts to affect the cut off frequency of the entire system due the decreasing diffraction limited cut off frequency. The decrease of $f_{c,lens}$ can be compensated by choosing a high f-number (i.e. choosing a smaller diaphragm), however this also limits the light gathered by the optical system proportional to $\sim 1/f^2$ and hence degrades the signal to noise ratio. A good balance between spatial resolution and the signal quality occurs when the array limited and diffraction limited cut off frequency are approximately
equal (Paul et al., 1990): \( f_{c,\text{lens}} \approx f_{c,\text{array}} \), hence

\[
\frac{1}{2l_p} = 1.22 \frac{f_z(1 + m)}{m^2 L_z/2},
\]

yielding an expression for the f-number in terms of magnification \( m \), pixel to pixel distance \( l_p \) and the depth of the measuring volume \( L_z \):

\[
f_z = 0.20 \frac{m^2 L_z}{(1 + m)l_p}.
\]

In our case \( m = 0.091, \ l_p = 10 \ \mu \text{m} \) and \( L_z = 8 \ \text{mm} \), it implies that the f-number had to be about unity. The fairly large depth of view of the camera lens operated at its maximum f number is due to the small size of the CCD chip \((2.56 \ \text{mm} \times 2.56 \ \text{mm})\) relative to the sizes of the images formed by the 50 mm optics.

5 Results

In this section we present some results obtained with the 4D-LIF technique. First, we consider the applied image processing techniques for obtaining the scalar quantity (concentration) and the three dimensional gradient vector from the raw 8 bit depth images captured by the CCD camera. Next we deal with the measurements consisting of 50 data planes for visualisation purpose. Finally, the measurement data is analysed more quantitatively using data volumes consisting of 8 data planes.

5.1 Image processing

Next to the fluorescent signal, each image consists of a background signal due to (Raleigh) scattered laser light and a dark signal due to several camera noise contributions. The combined background and dark image can be measured separately in the tubular reactor operating without fluorescent dye being injected. To correct for this combined dark and background image, it is subtracted from each data plane afterwards.

The Gaussian shaped intensity profile of the laser sheet results in a non-uniform illumination in span wise sheet direction over the measuring volume. It is assumed that the time averaged scalar concentration is constant over the data volume, which is reasonable in central regions of the reactor at some distance downstream the injector. The variation in span wise sheet direction of the average image obtained from the measurements thus can be fully attributed to the non-uniform illumination. All concentrations presented are normalised with the mean local concentration \( \zeta^* \).
Fig. 6. The time evolution of the concentration field (a) and square root of scalar energy dissipation field (b) at a central data plane extracted from successive realisations of three dimensional data volumes consisting of 8 layers. The $x$-axis indicates the stream wise direction (see figure 4).

The scalar gradient field has been calculated using a central difference scheme which does not only take into account the scalar differences in the axis direction, but also the scalar differences in the diagonal axis directions (see Dahm et al., 1990). The associated scalar energy dissipation $\chi = \nabla \cdot \nabla \zeta$ has been normalised with a characteristic scalar energy dissipation $\chi^* = ID(\zeta^*/\eta_B)^2$. 
5.2 Qualitative analyses of the transient three dimensional data

Figure 1(a) shows the concentration at the faces of data volume consisting of 50 data planes. The three dimensional nature of the data volume is clearly illustrated by the continuity of the scalar structures over the edges of the data volume. The side planes, constructed from the subsequent closely spaced images, and upper plane show similar vortical structures, giving a qualitative indication that both the spatial resolution and the dynamical range of length scales in depth direction is adequate. The square root of the normalised scalar dissipation \((\chi/\chi^*)^{1/2}\) is shown at the faces of the data volume in figure 1(b). This quantity, equal to the modulus of the scalar gradient vector \(|\nabla \zeta|/(\zeta^*/\eta_B)|\), is shown rather than the scalar energy dissipation \(\chi/\chi^*\) for the reason that a larger range of mixing rates can be visualised with the scale of 255 colours. Both the concentration and the scalar dissipation appear to occur in layer like structures being randomly orientated with respect to the laser sheet. The three dimensional character of the measurement data allows the orientation of layers to be taken into account when determining quantities such as layer thickness and interfacial area. For critical assessment of micro mixing models the 4D-LIF technique therefore seems to be a promising technique.

The temporal resolution of the three dimensional data space can be increased significantly by reducing the number of data planes (see section 4). Figures 6(a) and 6(b) show the concentration and scalar energy dissipation at a central data plane extracted from a sequence of three successive realisations of data volumes consisting of 8 layers. All three spatial components of the scalar gradient vector are used to estimate the scalar energy dissipation. The temporal resolution of 8.4 ms is adequate to resolve the Kolmogorov advection time scale \(\tau_a=20\) ms, which is reflected by the relatively small changes of the successive realisations. Moreover, we can reasonably assume the scalar field to be ‘frozen’ during the time that a data volume is scanned.

5.3 Mixing rate distributions

In this paragraph the scalar gradient measurements will be analysed more quantitatively. The number of data planes is limited to eight in order to achieve a statistically significant number of realisations of data volumes. Moreover, the time interval between the successive realisations was taken to be large in order to obtain statistically uncorrelated measurement data. All 950 realisations were taken into account for the presented probability densities.

The normalised probability densities of the three scalar gradient components are given in figure 7(a). The peak of the probability density near zero is generally broadened by the noise outside the dye regions where the concentration approaches zero and the signal to noise ratio is too low to allow proper differentiating. The tails of the probability densities are formed by the contribution
Fig. 7. (a) The probability density function of all three scalar gradient vector components. (b) The distribution of the mixing rate obtained from one-, two-, and three-dimensional approximations of the true scalar energy dissipation, based on the sum of squared gradient vector components \( (\partial \zeta / \partial x)^2, (\partial \zeta / \partial x)^2 + (\partial \zeta / \partial y)^2 \) and \( (\partial \zeta / \partial x)^2 + (\partial \zeta / \partial y)^2 + (\partial \zeta / \partial z)^2 \), respectively.

of the scalar interface regions. The skewness of all three distribution is close to zero, which means the gradients are distributed symmetrically over positive and negative values. The scalar gradient distribution of \( y \)-component (see figure 4 for the axis definitions) tends to be slightly narrower than the other directions. Although the scalar field appears to be quite isotropic, there exists a slight degree of anisotropy.

The probability density of the logarithm of the scalar mixing rate is considered in figure 7(b). The distribution of the one- and two-dimensional approximations of the scalar energy dissipation field, \( \chi_{1D} \) and \( \chi_{2D} \) respectively, is compared to the result obtained from the full three dimensional measurement of \( \chi \) (indicate as \( \chi_{3D} \) in the figure). The noise of the outside regions which was recognised as the peak around zero gradients in figure 7(a), is now reflected by the bimodality of all distributions in 7(b). The left part should be considered to be noise of the measurement. The scalar dissipations from interface regions are shown by the right part on the high end values. It should be noted that the large noise contribution is due to the low fraction of dye present in most of the measurements. This is inherent to the high advection velocity of the flow in the reactor compared to, for instance, a jet injected into a stagnant liquid. If the scalar field were fully isotropic, then the average values of the one-, two-, and three-dimensional dissipation distributions would be in the ratio
(0.33):(0.67):(1.00). From our distribution the maximum values of the signal peaks are taken to be indicative for the average of the distribution, which is reasonable since the distributions are expected to be symmetric. The maximum values of these signal peaks are found in the ratio (0.33):(0.45):(1.00), again indicating to a slight degree of anisotropy due to the lower contribution of the y-direction. Apart from that, it can be concluded that the scalar gradient component in the depth direction has a significant contribution to the total mixing rate and hence should not be neglected when determining $\chi$.

6 Conclusions and outlook

In this paper, the design and some preliminary results with a 4D-LIF setup for measuring 3D concentration fields are presented. It is shown that the small scale flow structures can be well resolved in both space and time. From the measurements the scalar energy dissipation has been obtained from all the three scalar gradient vector components.

The measurements seems to be promising for critical assessment of micro mixing models. The mixing rate distribution, however, shows a large noise contribution from the outer regions where the low scalar values result in a rather poor signal to noise ratio. In the near future, a large frame 25 W Argon ion laser will be used for the measurements. This will improve the overall signal to noise ratio, resulting in a more proper estimate of the mixing rate.

References


