Experimental study of turbulent mixing process in simultaneously reacting and non-reacting confined flow

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Abstract Dual-tracer planar laser-induced fluorescence (PLIF) and Particle image Velocimetry (PIV) techniques are used to quantify instantaneous distributions of macro and micro-mixing. The investigated flow is a Partially Stirred Reactor (PaSR) composed of 16 pairs of sheared jets. The typical pattern consists in a jet surrounded by 4 jets in counter flow, arranged on a cartesian grid. The first scalar concentration measurement is performed by PLIF of nitric oxide (NO) seeded in a nitrogen jet, to mark the unmixed jet fluid fraction. PLIF on acetone, seeded in the counter-flowing air jets, marks the total fluid fraction. By combining data from these two simultaneous images, quantitative measurements of micro-mixing between counter-flowing jet fluids can be made on a pixel-by-pixel basis. Simultaneously, two velocity components measurements (by PIV) are used to get information about the velocity field which mixes.

The first objective of this experimental work is to provide a reliable database for different flow geometries and Reynolds numbers. This database is further used to study the influence of such parameters on the micro-mixing properties and in particular on its efficiency. It is shown that micromixing is favored by either increasing injection Reynolds numbers, or injection by small diameters jets in comparison with the flow dimensions (small-scale injection).

1. Introduction

Quantitative characterization and prediction of micromixing are of significant interest in numerous applications, such as chemical processing, combustion, aerospace propulsion, etc. 'Micro-mixing' is generally used when the mixture of two substances A and B occurs at small scales, or at molecular scales. In this case, the contact surface between the two substances is maximal, and the chemical reaction can occur on a maximum surface to volume ratio. This is why chemical reactions are very good candidates to characterize micro-mixing [1]. PLIF has been used extensively to image the scalar field in both reacting and non-reacting flows, and a variety of laser based techniques have been implemented. In high Reynolds number flows, however, the ratio between the largest and the smallest scales is important and the challenge is therefore to visualize both large and small scales. To overcome these difficulties, various strategies have been proposed. In reactive configurations, mixing statistics can be obtained by using a 'flip' experiment [2]. The obvious limitation of this technique is that it cannot be used to obtain instantaneous images of molecular mixing. As an alternative to methods based on fast chemistry, several studies have utilized quenching of fluorescence of tracer species as the product marker, also known as 'cold chemistry' [3]. Hu and Koochesfahani [4] used the quenching of acetone phosphorescence as a marker of mixed fluid, as a variant of cold chemistry technique. The first technique for measuring both instantaneous molecularly mixed fluid quantities and the large-scale structures in high turbulent flow was implemented by [5] in a shear layer. This technique uses acetone PLIF as a passive-scalar measurement of the fluid fractions from each fluid stream of a shear layer, and uses NO PLIF simultaneously to obtain the molecularly unmixed fluid fraction. Thus, the instantaneous molecularly mixed fluid fraction, mixing efficiency, and relevant statistics can be obtained in post-processing. Several studies in (high-speed) turbulent jets, planar shear layers etc. have been performed using this approach [6],[7].
However, the existing studies are mainly devoted to free shear flows that entrain and mix freestream (unmixed) fluids. This work is aimed at a quantitative understanding and the development of models for molecular mixing in turbulence. Mixing in a Partially Stirred Reactor (PaSR) is examined over a range of flow conditions. The proposed PaSR geometry has well-characterized boundary conditions facilitating comparisons with modeling, numerical simulations. The turbulent flow is composed of multiple shear jets \[8\], with a typical pattern consisting in a jet surrounded by 4 jets in counter flows, arranged on a cartesian grid.

First, we provide a reliable experimental data set, for variable flow geometries and Reynolds numbers. Measurements rely on laser diagnostics that simultaneously measure the following:
- The scalar concentration field in cold reacting flow, in which NO is used as a tracer (PLIF on NO). Its fluorescence is strongly quenched by oxygen from the counter-flow jets. This concentration will mark the unmixed scalar.
- The concentration field of a non-reacting scalar (e.g., acetone) transported as is NO (PLIF on Acetone). This latter quantity marks the mixture fraction.
- Two components of the velocity field using PIV.

2. Experimental set-up and diagnostics

2.1 The reactor

The reactor is presented in Fig 1. The basic design allows for a modular and flexible experiment, generating flows in which velocity and scalar fields can be simultaneously investigated. The reactor interior is a rectangular parallelepiped (110 x110x60 mm$^3$) equipped with quartz Suprasil windows (100x80 mm²). The top/bottom porous boundary plates are backed by plena connected to an exhaust piping network through 8 exhaust ports on each cap [8]. Each plenum provides 8 tubes that supply the top/bottom jets individually. Two types of tubes are employed. These are 200 mm long, with an inner diameter of either $D = 10$ mm or $D = 6$ mm, respectively, arranged on a 24 mm spacing Cartesian mesh, Fig. 1. The horizontal distance between two neighboring jet axes is $2L = 24$ mm. Hence, the geometry of the confined flow is fixed through the scales: $L = 12$ mm, $H = 60$ mm and the injection jets diameter $d$. To document internal conditions, one wall is equipped with a 1 mm diameter pressure tap, connected to a digital manometer and a platinum probe for temperature measurement. All the experiments are performed at $P = 1.6$ bar and at room temperature. In this work, the Reynolds number $Re_d = U_{inj} d / \nu$, based on $U_{inj}$, the fluid injection velocity, varies between 6400 up to 16000. We focus on the influence of the lateral confinement and of the Reynolds number on statistics, thanks to the fluid injection by either 10 or 6 mm tubes diameter.

Since the injection jets diameter takes two values while the lateral distance among each two consecutive opposed jets is fixed, the flow configuration is double: CSJ close sheared jets ($d = 10$) and FSJ far sheared jets ($d = 6$).

2.2 Experimental measurements

The ensemble-averaged displacement of Di-Ethyl-Hexyl-Sebacat (DEHS, C26H50O4) particles ($\rho_p = 918$ kg.m$^{-3}$) between pairs of images is estimated using a PIV cross-correlation technique, 2D-2C velocity field. The particles diameter (calibrated with a Malvern difractometer) is between 1 and 1.15 $\mu$m. Due to the periodic pattern of the fluid injection, we have restricted the investigated fields to a couple of counter-flowing jet, (see Fig. 2).
The light source is a Nd-Yag laser (Big Sky laser, 120 mJ/pulse) with a second-harmonic-generating crystal that produces a Q-switched laser output in the green ($\lambda_{P=532}$ nm). Light scattered from the particles is collected on a CCD camera (R&D Vision, 12 bits, 1600 $\times$ 1200 pix2) with a 50 mm f/1.2 Nikkor lens, yielding a magnification of 21.76 pix.mm$^{-1}$. The initial size of the PIV interrogation window is 64 pix$^2$. Six iterations are used to obtain a final interrogation window size of 16 pix$^2$, with a 50% overlap. The spatial resolution of the measurements is determined according to the PIV transfer function [9]. This analysis yields a cut-off spatial frequency for the PIV system of $f_c=2.8.10^{-2}$ pix$^{-1}$ corresponding to a scale of $\approx 1.12$ mm. These spatial resolutions are fixed in a given experimental configuration and do not depend on the flow. The size of velocity field is 55.2$\times$73.5 mm$^2$.

The passive scalar (air+acetone 3%) is injected by the lower side jets (illustrated by white jets in Fig. 3a, while the reactive scalar (N2+NO 300 ppm) by upper jets (red jets in Fig. 3b). Passive
Scalar measurement is investigated by Planar Laser Induced-Fluorescence (PLIF) on acetone molecule. A laser sheet is created by the same optical system as PIV measurement. The light source is a Nd:YAG laser (Spectra Physics) with a fourth-harmonic generating crystal that produces a Q-switched laser output in the UV ($\lambda_{Ac}=266$ nm, 170 mJ). Fluorescence signal is collected on a "Princeton Instrument PhotonMax EMCCD" camera, 16 bits, 512×512 pixels coupled to a visible Nikkor 50 mm focal length, f/1.2 lens. The exposure time on is 2 ms, that is why parasites lights have to be rejected by color glass, by using a dichroic mirror and a BG12 filter.

**Fig. 3:** Simultaneous measurements data sets for $d = 10$ mm. Distributions of the conserved scalar $Z$ (left), the pure fluid mixing fraction $Z_{\text{reactive}}$ (middle) and instantaneous velocity field (right). Close shear jets, $Re_d = 10700$.

**Fig. 4:** Simultaneous measurements data sets for $d = 6$ mm. Distributions of the conserved scalar $Z$ (left), the pure fluid mixing fraction $Z_{\text{reactive}}$ (middle) and instantaneous velocity field (right). Close shear jets, $Re_d = 10700$.

Spatial resolution is estimated to 490 $\mu$m thanks to the FTM of the optical acquisition system. Camera’s FTM is determined by taking an image of "infinite" gradient simulated by a razor blade placed on the target is acquired. FFT is applied on the image gradient. The cut frequency is got when the contrast is less than 10%.

Despite a small part of fluorescence signal available, because of high energy laser, the signal to noise ratio is more than 10. Fluorescence signal is normalized by the mean sheet and the maximum grey level is adjusted by a zone in which mixing fraction of passive scalar is one (jet potential core). The reactive scalar is simulated by the oxygen and acetone quenching on NO molecule \cite{3,10} call "Cold chemistry Method". The laser was tuned to $Q_1(1)\rightarrow Q_1(4)$ line of the $A^3\Sigma^+ \leftarrow X^2\Pi(0,0)$ NO
electronic transition ($\lambda_{NO}$=226.18 nm), using a frequency doubled BBO crystal of a dye laser pump by Nd:YAG laser at 355 nm. The global energy laser output is 3 mJ during 10 ns. PLIF images were acquired using an ICCD camera (Princeton Instrument, 16 bits, 512×512 pixels) coupled to a UV Nikkor 48 mm focal length, f/1.2 lens. A UG 5 glass filter 3 mm thickness is used to remove Mie diffusion scattering at 226 nm. The peak signal to noise ratio is about 40. The scalar field is 55×55 mm$^2$ as acetone PLIF field. A huge attention has been devoted to the calibration of the shot-to-shot energy fluctuations in order to correct the spatio-temporal inhomogeneities energy laser distribution. A small part of laser sheet is split using a beamsplitter (see Fig. 2, B#1) and projected on the image edge. The aim of this procedure is to provide accurate quantitative evaluation of the reactive scalar concentration. The normalized fluorescence signal $S_{t,\text{Norm}}$ can be written as:

$$S_{t,\text{Norm}}(x, y, t) = \frac{S_{\text{Norm}}(x, y, t) - \langle \text{Noise}(x, y) \rangle}{\langle \text{Noise}(x, y) \rangle},$$

with $R(x, t) = \frac{\langle U_{\text{split}}(x) \rangle}{U_e(x, t)}$ and $\langle U_{\text{split}}(x) \rangle$ is the mean split energy profile acquired with homogeneous flow fields and $U_e(x, t)$ is the instantaneous energy profile acquired on the image edge of the reactive scalar.

The spatial resolution of NO diagnostic is equal to 640 μm. All scalar fields are filtered using a Wiener filter type [11] which removes high frequency fluctuations. After normalization and filtering processes, residuals fluctuations of passive scalar measurement are ±1% and for reactive scalar measurement ±5%. A post processing is applied to scalar fields to get the same spatial physical mesh for the three measured quantities: the 'reactive' scalar NO, the conserved scalar and the velocity field. A linear data interpolation is applied to the scalars at the level of the velocity field mesh, the latter being the least accurate field. The post processing yields an experimental data set containing at one mesh point: the conserved scalar mixing fraction Z, the reactive scalar mixing fraction $Z_{\text{reactive}}$, and two velocity components (u and v) presented on Figs. 3 and 4.

3. Results

While the axisymmetric jet mixing layer has been investigated using many fluid diagnostic techniques, quantitative measurements of the extent of molecular mixing have been elusive. Chemical reaction in fluid flows is not initiated until the fuel and oxidant streams are mixed at the molecular level, a state distinct from that of macroscopically stirred, but unmixed, reactants. As far as the reactive scalar is concerned, measurements provided simultaneously

- The scalar concentration field for NO, using NO PLIF, that reacts with O$_2$. This scalar is called $Z_{\text{reactive}}$.
- The concentration field of a non-reacting scalar (e.g., acetone) transported as is NO. This latter quantity marks the mixture fraction, Z, and can be considered as inert. The acetone mole fraction field is measured by PLIF on acetone. Therefore, a double species map is produced. A first map, with NO, which will indicate places where NO is pure and unmixed. A second map, simultaneous to the first, with acetone, indicating places where NO would have been present, if it had not been reacted. By subtracting one will then obtain an instantaneous image of molecular mixing $Z_m$, and the reacting zone [6].

In the following, the focus is on the molecular mixing via the scalar $Z_m$ and the influence of the geometry and of the Reynolds number on instantaneous distribution and of statistics of the
molecularly mixed fluid fraction $Z_m$. We choose to represent only the field corresponding to one jet, with the vertical $x$ axis and the horizontal $r$ axis normalized with respect to the jet diameter $d$.

3.1 Instantaneous and statistical aspect of molecularly mixed fluid fraction for the close sheared jets geometry

We first extensively pay attention to the CSJ geometry. For CSG flow, Fig. 5 represents instantaneous images of the passive scalar $Z$ (left) and of the molecularly mixed fluid fraction $Z_m$ (right). At the jet injection, a potential core is clearly visible on both figures, where fluid is pure ($Z=1$) and therefore there is no molecular mixing $Z_m=0$. For regions where $Z<1$, it is not a priori obvious to state which quantity of this fluid concentration is pure and which one is molecularly mixed with the other scalar. In this case, the complementary measurements of pure fluid are necessary. The result is the $Z_m$ map (right) in which mixing region is represented. It is to be noted the (approximatively) "V" shapes of the mixing regions. The boundaries of this mixing region are marked by the mixing vortical structures (streamlines obtained from PIV are also represented), which are much more pronounced than those (Kelvin-Helmholtz like) present in a free axisymmetric jet (see Fig. 5 of [6]). Here, both the lateral confinement and the short distance over which jets develop, do influence the micromixing region.

Fig. 5: Instantaneous image of passive tracer $Z$, of fluid mixed at molecular level $Z_m$, of mixing efficiency $\eta_m$ and vorticity. Close sheared jets (Re = 6400).

It is of interest to represent instantaneous values of micromixing efficiency $\eta_m$, Fig. 5 which is equal to zero in regions where molecular mixing is completely absent, and rapidly grows to high values close to 1 on the boundaries of the potential core. In most of the flow volume, $\eta_m=1$, which signifies that the entire scalar present at that point is completely molecularly mixed. Regions where the mixing efficiency rapidly grows are strongly correlated with azimuthal vorticity (large-scale circulation, as solved by PIV) represented on Fig. 5.

Quantitative characterization of molecular mixing is readily done by using statistics, among which the simplest are mean values and variance of $Z_m$ fluctuations (Fig. 7). On the jet axis ($r/d=0$), $\langle Z_m \rangle$ monotonically grows from 0 at $x/d=0$ till values close to 0.8 (80% of molecular mixing) near the exit of the reactor. Between $x/d=3$ and $x/d=6$, i.e. over approximately half of the reactor height, molecular mixing is quite good from a statistical viewpoint. Most of the fluctuations (i.e. time
fluctuations between unmixed and molecularly mixed fluid) occur on the top half of the reactor, near the injection, as it is illustrated by the $Z_m$ variance distribution. Near the injection, fluctuations are strong over radial distances going up to $r/d=0.5$.

![Image](image.png)

**Fig. 6:** Average of fluid molecularly mixed (left) and variance of its fluctuations (right). Close sheared jets ($Re = 6400$).

A more detailed insight in micromixing fluctuations is possible by investigating the probability density functions (Pdfs) of the molecularly mixed fluid fraction, at different axial and radial positions. Figure 7 represents these Pdfs for axial positions varying between $x/d=1$ (in the potential core) till the bottom part of the reactor, $x/d=4$. Pdfs are calculated:

- on the jet axis (red), for which very important values of $Pdf(Z_m)$ are noted for $Z_m=0$ (unmixed fluid) at $x/d=1$ (top left figure). For further downstream positions, molecular mixing progressively improves and the Pdfs peak more and more towards values of $Z_m=1$ (nice aspect of micromixing).
- at a further radial position (violet), for which the shape of the Pdf is approximately the same whatever the axial position,
- out of the potential core (blue) which reflects an improved mixing, better and better towards high values of $x/d$. Finally, while near the injection the Pdfs at different radial positions are strongly different as a sign of strong radial inhomogeneity, at $x/d=4$ the three Pdfs are much more similar, thus reflecting the good homogenization of the molecular mixing.

### 3.2 The influence of Reynolds number and of the geometry on the molecularly mixed fluid fraction

One question addressed here is the effect of increasing Reynolds number, up to to $Re_d=16000$, on the micromixing. Figure 8 represents three instantaneous images of molecularly mixed fluid fraction $Z_m$ for the lowest Reynolds number $Re_d=6700$ (three top figures) and for the highest Reynolds number $Re_d=16000$ (three bottom figures). Both of them concern the same geometry, that of close shear jets ($d=1cm$). Two features are to be noted: first, at higher Reynolds number, the
potential core (where the jet fluid is pure and micromixing is therefore completely absent) is longer, incursions of pure fluid are present till distances as large as x/d=4. Second, micromixing on edges of potential core is more efficient and takes place at earlier values of x/d. This suggests that the volume of micromixed fluid increases for higher and higher Reynolds numbers (not shown here).

**Fig. 7**: PDF($Z_m$) for x/d = 1 (top left), x/d = 2 (top right), x/d = 3 (bottom left) and x/d = 4 (bottom right). Lines represent Pdf($Z_m$) at r/d = 0 (red), r/d = 0.25 (violet) and r/d = 0.5 (blue). Close sheared jets (Re = 6400).

Better micromixing for larger Reynolds is emphasized by mean values of molecularly mixed fluid fraction $\langle Z_m \rangle$, which are illustrated on Fig. 9 (left), on the jet axis (solid lines) and at r/d=0.25 (dashed lines). The latter radial position corresponds to points within the shear layers, the intensity of the shear $\langle \frac{\partial U}{\partial r} \rangle$ being much stronger for increasing Reynolds numbers. On the jets axis, increasing the Reynolds number corresponds to higher values of $Z_m$, which are reached earlier. As an example, at x/d=1, $Z_m=0.15$ for Re$_d$=6700, while $Z_m=0.2$ for Re$_d$=16000. Moreover, in the shear layers situated on both edges of the potential core at r/d=0.25, for a downstream position x/d=0.5 values of $Z_m$ are equal to 0.25 for the lower Reynolds and as high as 0.5 for the largest Reynolds. Qualitatively, the Pdfs of $Z_m$ are similar to that earlier discussed for Re$_d$=6700, with the clear distinction that higher values of $Z_m$ are clearly privileged, and this is true for downstream positions as small as x/d=0.5, for radial positions placed in the lateral shear layers. The connection between these properties and the local structure of the velocity field is certainly a subject to be presented in a near future. Note that an improved micromixing with increasing Reynolds, in the shear layers of a single round jet, was also emphasized by [6], over a wider range of Reynolds numbers.

Last, we test the influence of a slightly variable geometry on the micromixing efficiency. We
choose to compare, at the same injection Reynolds number $Re_d=10700$, values and characteristics of $Z_m$, for $d=1\text{cm}$ (close shear jets, illustrated on the top three instantaneous images on Fig. 10) and for $d=0.6\text{cm}$ (far shear jets, see the last three instantaneous images on Fig. 8). For far shear jets, the dimensions of the potential core are reduced, leaving more space to the molecularly mixed fluid. Therefore, micromixing is improved when the pure scalar is injected at smaller scales.

**Fig. 8:** Instantaneous images of $Z_m$ for close sheared jets for $Re_d = 6400$ (three top) and $Re_d = 16000$ (three bottom).

**Fig. 9:** Evolution of $<Z_m>$ (left) and of $\text{var}(Z_m)$ (right) as functions of the downstream position $y/d$
on the jet axis (solid lines) and at $r/d = 0.25$ (dashed lines). $Re_d = 6400$ (blue) and $Re_d = 16000$ (red).

Fig. 10: Instantaneous images of $Z_m$ for far sheared jets for $Re_d = 10700$. Injecting jet diameter $d = 1$cm (top three images) and $d = 0.6$cm (three bottom images).

4. Conclusions

We investigated flow and micromixing in a Partially Stirred Reactor (PaSR) composed of 16 pairs of sheared jets. The typical pattern consists in a jet with counterflow. Dual-tracer planar laser-induced fluorescence (PLIF) and Particle image Velocimetry (PIV) techniques are used to quantify instantaneous distributions of macro and micro-mixing. The total fluid fraction $Z$ was quantified by PLIF on acetone, whereas the pure fluid fraction was detected by PLIF on NO, quenched by oxygen (‘cold chemistry’). Instantaneous images of $Z$, $Z_m$ and two-velocity field $u$ and $v$ were used to investigate mixing and in particular micromixing.

It was shown that micromixing is favored by either increasing injection Reynolds numbers, or injection by small diameters jets in comparison with the flow dimensions (small-scale injection).
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