

pH-Sensitive Planar Laser-Induced Fluorescence Based Characterization Of Turbulent Mixing Induced By Fractal Oscillating Grids

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ABSTRACT

This study examines passive scalar mixing in a tank stirred by fractal oscillating grids. Injection experiments are performed and pH-sensitive planar laser-induced fluorescence measurements are used to evaluate the influence of grid geometry on mixing time and local turbulent scalar behavior. Fractal grids are found to perform better than traditional Cartesian grids, with the I-Fractal configuration showing the highest efficiency. The mean flow generated by grid oscillations, generally more intense for fractal grids, has a major influence on the mixing mechanism, enhancing scalar patch transport toward the highly turbulent region swept by the grid. These results open the way for the development of efficient fractal oscillating grid mixers.

1. Introduction

Turbulence is widely recognized as a fundamentally efficient mechanism for the enhancement of mixing and transport in fluid systems (Dimotakis, 2005), making it an essential tool in various industrial processes, particularly in process and bio-process engineering (San et al., 2017; Luo et al., 2021). A wide range of turbulence-generating devices have been developed to harness this intensification effect. In batch processes, the most famous one is probably the stirred tank reactor, where turbulence is induced by the shear imposed from a rotating impeller (Delbridge et al., 2023; Gebouský et al., 2025). In continuous systems, turbulence can be generated via steady wall shear (Avila et al., 2023), curvature and direction changes (Li et al., 2022), or obstacle-induced perturbations and vortex shedding (Morton & Yarusyevych, 2014). Reactors can also utilize centrifugal instabilities to induce intense turbulence for solid-liquid, gas liquid, or liquid-liquid mixing (Yang et al., 2023; Nemri et al., 2013; Carré et al., 2026)

All those seemingly quite different devices yet share a common feature: a significant mean shear is required to trigger and sustain turbulence, and this poses an issue in several in bio-process applications where the mixing medium may contain shear-sensitive biological material (Wang & Lan, 2018). Providing the right amount of shear-stress is thus a critical constraint in bioreactor development, and options to generate turbulence with low imposed mean shear are required (Michels et al., 2016).

A promising option emerges with the use of oscillating grids (Cheng & Law, 2001), which unlike traditional impeller-based systems, can create a rather homogeneous and isotropic turbulent flow field at limited imposed shear at the wall of the moving device. The principle of turbulence generation indeed resides in the oscillatory behavior and periodic interactions of the grid wakes and jets less than on the inertia in the wake itself (Yan et al., 2007).

Oscillating grid turbulence has been a subject of fundamental interest in fluid mechanics for more than 50 years (Thompson & Turner, 1975; Hopfinger & Toly, 1976; Mcdougall, 1979; Long, 1978; Mcdougall, 1979; Dickinson & Long, 1983; De Silva & Fernando, 1994; Matsunaga et al., 1999; Cheng & Law, 2001; Wan mohtar, 2016), owing to its potential to generate homogeneous isotropic turbulence with supposedly zero mean flow, decouple turbulence and mean flow effects, and study the interaction of turbulence with other phenomena, e.g. stratified (Thompson & Turner, 1975) or viscoelastic fluids (Liberzon et al., 2009), particle and bubble dynamics, gas-liquid (Lacassagne et al., 2019) or gas-solid interfaces (McCorquodale & Munro, 2018). It is also used as a test device for examining sediment re-suspension, microplastics transport, and the behavior of microorganisms in turbulent flows (Traugott et al., 2011; Poulain-Zarcos et al., 2022; Hondzo & Lyn, 1999), and has practical applications as flotation cells (Safari et al., 2017).

On a different note, several studies have demonstrated how grid geometry significantly affects flow topology (Fernando & De Silva, 1993; Xuequan & Hopfinger, 1986; Cheng & Law, 2001; McCorquodale & Munro, 2017). At the same time, fractal grid geometries have been introduced in the early 2000's in wind tunnel experiments (Seoud & Vassilicos, 2007; Hurst & Vassilicos, 2007), with the aim, among others, to enhance turbulent scalar fluxes as compared to classical cartesian grids (Laizet & Vassilicos, 2013). Fractal grids geometries have only recently been used in oscillating conditions in Musy et al. (2024), where the hydrodynamics around fractal oscillating grids was studied using particle image velocimetry (PIV). The overall turbulence intensity was found to be increased by the fractal nature of the grid oscillating, and its spatial distribution strongly affected by the mean flow topology. As a matter of fact, a notable observation was that fractal geometries induced a significant and well structured mean flow. While the later reduces the potential for use in fundamental homogeneous and isotropic turbulence studies, it may be seen as an asset for a batch process mixing application, when combined with preserved efficient turbulence generation in the grid swept region, and propagation inside the tank.

In the present contribution, we move away from the original zero-mean-flow purpose of oscillating grids and instead focus on characterizing the mixing behavior of fractal oscillating grid turbulence systems. More specifically, the purpose of this study is to clarify the influence of grid geometry and flow features on mixing efficiency and dynamics. To this end, pH-sensitive planar laser-induced fluorescence is employed to analyze mixing experiments in which a dye is injected into an oscillating grid turbulent flow. The broader aim is to assess the suitability of these systems for future applications where conventional mixing approaches prove inadequate.

2. Material and Methods

The experimental setup consists of a prismatic glass tank of $260 \times 450 \times 260 \text{ mm}^3$ with a wall thickness of 6 mm (see Figure 1) with a $W=24.8 \text{ cm}$ square base. The agitation system is made of an AKM32E Kollmorgen motor, a speed reducer (reduction ratio of five), a crankshaft system and the grid itself. The oscillation amplitude A_{osc} and the grid oscillation frequency f_{osc} are controlled by an eccentric

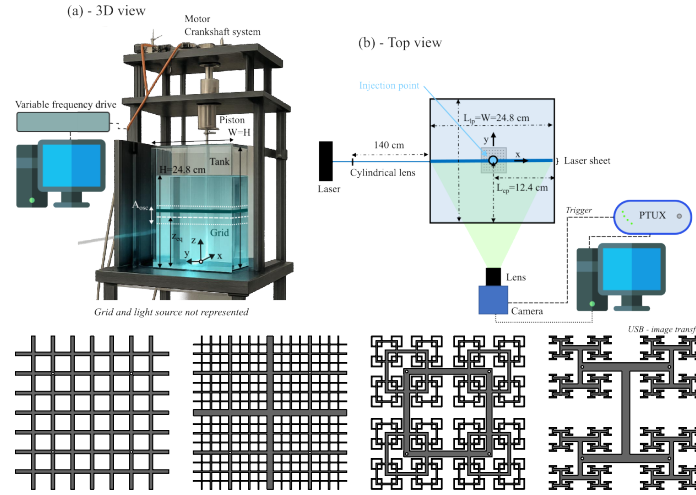


Figure 1. Picture (left) and sketch (right) of the experimental setup and implementation. The bottom row depicts the four different geometries used to generate turbulence (left to right): simple cartesian grid (SC), fractal cartesian grid (FC), fractal square grid (FS) and fractal I-shaped grid (FI) (Musy et al., 2024)

wheel and the motor rotation speed (set using Kollmorgen's AKD-x00306 software), respectively. Four different grid geometries are used (see Figure 1) and the design of which was proposed in Musy et al. (2024). The grid equilibrium position is located in the middle of the fluid volume such that $z_{eq} = H/2$ where H is the height of fluid in the tank.

A pH-sensitive version of planar laser-induced fluorescence (PLIF) is used to visualize concentration fields in the central vertical plane of the tank (Walker, 1987; Lacassagne et al., 2018). Fluorescein sodium is chosen as the pH-sensitive dye, and excited by a continuous 488 nm laser. Fluoresced light images are recorded with an M-lite 5M CMOS camera equipped with a 35 mm lens and monitored by the DaVis software. The acquisition frequency is $f_{acq} = 50 \text{ Hz}$ and the shutter time is set at $dt = 19 \text{ ms}$.

Firstly, the tank is filled with a total of 16.25 L of distilled water: 15.25 L of which will be used for the mixing experiments, and 1 L sampled to prepare the injection solutions. A volume of concentrated fluorescein sodium solution is then mixed into the tank, for a final dye concentration of $5 \times 10^{-8} \text{ mol/L}$. Laser light attenuation and fluoresced light reabsorption can thus be neglected. 1 L of the solution is sampled from the tank to prepare acid (HCl) and base (NaOH) injection solutions with strictly identical dye concentration. Calibration and mixing experiments are then performed in turn: injection of low or high pH fluorescein sodium solutions are performed, the mixing process is recorded, and the final mixed state is used as a known homogeneous pH calibration point (where the pH is controlled by an Avantor pH meter). The injection is performed with a single 4 mm diameter needle, at the free surface and in the center of the tank. It is checked that neither the injection volume nor the injection duration have a significant influence on the measure mixing times and scalar dynamics. The grid is oscillated each time for 10 minutes at $f_{osc} = 1 \text{ Hz}$ with a $A_{osc} = 35 \text{ mm}$.

In order to construct the $I = f(pH)$ calibration curve, fluoresced light intensity is spatially averaged (after masking out the grid) for each calibration image, and the resulting values are plotted as a function of the measured pH in figure 2. The intensity is normalized such that $I_n(pH) =$

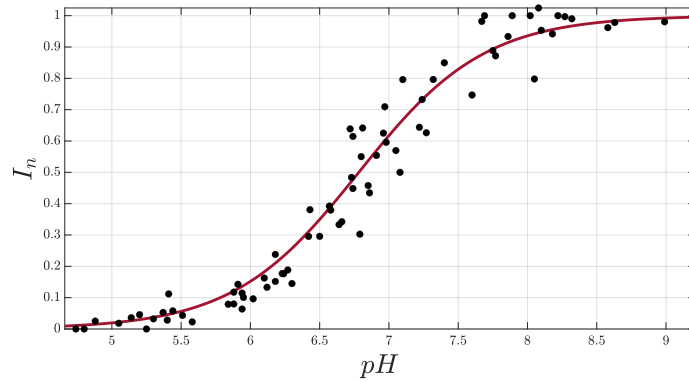


Figure 2. Calibration curve

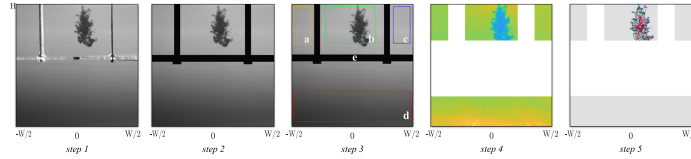


Figure 3. Illustration of the post processing procedure: original image, grid masking, regions of interest, pH field, concentration field

$\frac{I_c(pH) - I_{min}(pH)}{I_{max}(pH) - I_{min}(pH)}$ where I_c is the mean pixel intensity of the post-processed calibration image, I_n is the normalized average image intensity and I_{max} , and I_{min} represents averaged maximum intensity achieved for $I(pH \rightarrow +\infty)$, and minimum intensity for $I(pH \rightarrow 0)$, respectively.

The data is fitted by an hyperbolic tangent curve $I_n(pH) = 0.5 \cdot (1 + \tanh(\alpha \cdot pH + \beta))$, as proposed by Coppeta & Rogers (1998) and Lacassagne et al. (2018). The fitting yields $\alpha = 1.098$ and $\beta = -7.450$, and an expected high sensitivity region between pH 5 and 8 is retrieved. With this calibration curve, mixing images can be processed as follows (see figure 3). The grid and grid supporting bars are masked from the recorded raw images, which are then split into sub-regions. At this stage, the standard deviation of the pixel intensities can be computed in order to estimate the mixing index and mixing time as detailed later. Alternatively, the pH values for each pixel can be deduced from the calibration curve, applying a min-max normalization, and inverting the hyperbolic tan equation. pH values are then converted into instantaneous concentration values such that $C(x, y, t) = 10^{-pH}$.

The temporal evolution of the mixing state within the tanks is measured using the intensity of segregation technique (Danckwerts, 1952). The spatial standard deviation $\sigma_{px}(t)$ of the normalized pixel intensity for experiment images I_{nm} and the intensity of segregation (Hoffmann et al., 2006) defined as $\sqrt{I_s(t)} = \frac{\sigma_{px}(t)}{\sigma_{px,max}}$, with $\sigma_{px,max}$ the maximum standard deviation during the experi-

ment are calculated. The mixing index value is then obtained $M(t) = 1 - \sqrt{I_s(t)} = 1 - \frac{\sigma_{px}(t)}{\sigma_{px,max}}$

and normalized as $M_n(t) = \frac{M(t)}{M(\rightarrow +\infty)}$ such that M_n ranges from 0 in a completely segregated medium to 1 in a fully mixed medium. In a typical mixing experiment, M_n starts at 1, decreases to

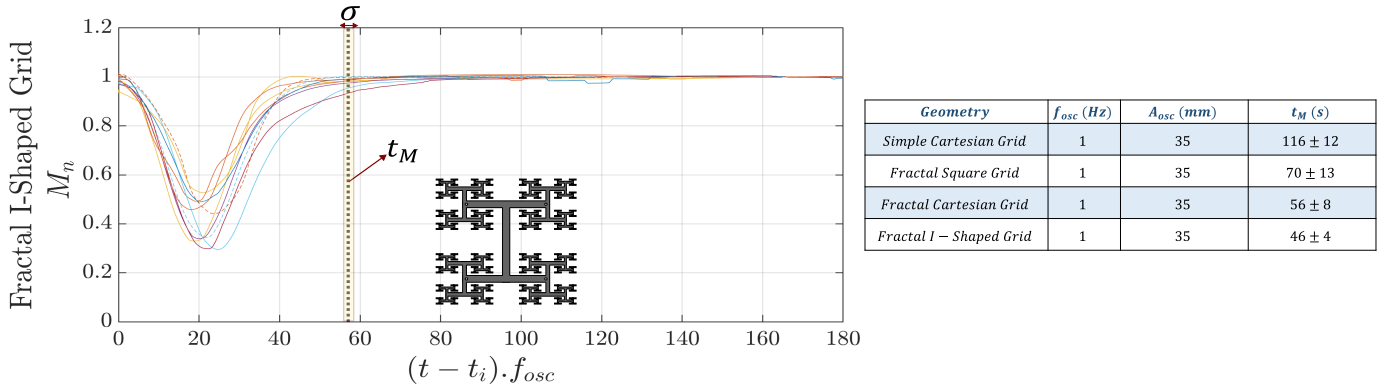


Figure 4. Left: Mixing index as a function of time for the I-fractal grid at $f_{osc} = 1$ Hz, where either acid (full lines, decreasing pH) or base (dashed line increasing pH) is injected. The vertical dashed line shows the average mixing time. Right: summary table of all mixing times measured

almost zero as injection is performed, and increases back to 1 upon mixing. A mixing time t_M can thus be defined as the time it takes for M_n to converge back to 95% its final value.

3. Results

3.1. Mixing time

Figure 4 (left) illustrates mixing index curves for several runs performed with the I fractal grid, with $t = 0$ at injection. It should be noted that the mixing dynamics and mixing times are found to be independent on whether acid or base is injected, demonstrating that the method tracks reaction products as passive scalars. The mixing times and confidence intervals are reported for all four grids in the right part of figure 4. All fractal grids are found to mix faster than the simple cartesian one, with the I-fractal grid being the quickest but also the most repeatable across runs. The lowest repeatability and largest uncertainty is reported for the simple Cartesian grid. This can be interpreted as an effect of the mean flow: well structured for the I grid, it induces a good repeatability of the transition from the injection jet dynamics and the long time grid induced, while this is not the case for the Cartesian grid (Musy et al., 2024).

3.2. Mixing dynamics

Reynold's decomposition allows to decompose instantaneous concentration fields $C(x, y, t)$ into an averaged $\bar{C}(x, y, t)$ and a fluctuating contribution $c'(x, y, t)$, where c' illustrates the effect of turbulence on mixing. In the present study, the transient nature of the mixing experiments prevent the use of the ergodicity principle and make it impossible to obtain $\bar{C}$ by time averaging, while repeating experiments a sufficient number of times for statistical convergence is virtually impossible.

The "mean" concentration value is thus estimated by smoothing the concentration time series over a span of twice the mean flow typical time scale, thereby removing the large-scale mean flow contribution.

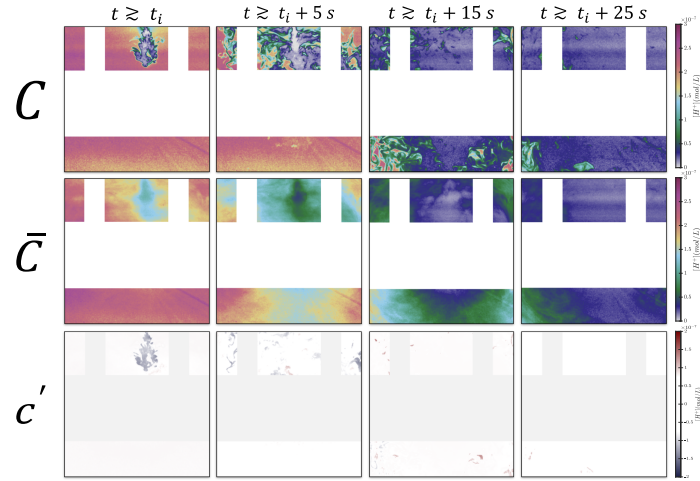


Figure 5. Example of Reynolds Decomposition for a Fractal I-shaped (FI) grid experiment. First line: instantaneous concentration field $[H^+] = C$. Second line: averaged concentration field $[H^+] = \bar{C}$. Third line: fluctuating concentration field $[H^+] = c'$.

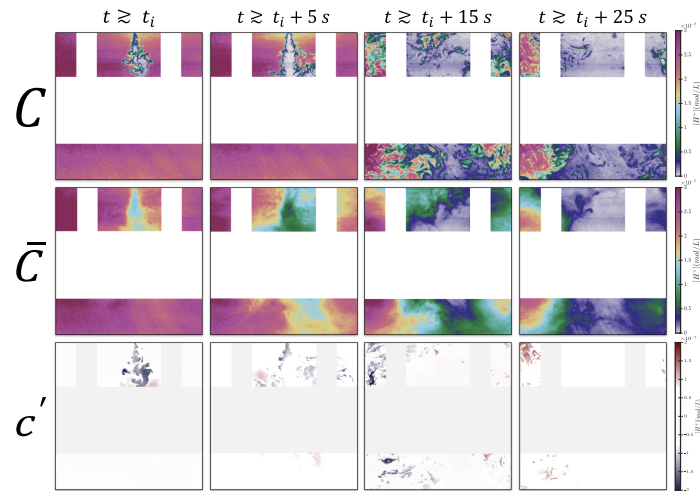


Figure 6. Example of Reynolds Decomposition for a Simple Cartesian (SC) grid experiment. First line: instantaneous concentration field $[H^+] = C$. Second line: averaged concentration field $[H^+] = \bar{C}$. Third line: fluctuating concentration field $[H^+] = c'$.

The instantaneous concentration fields (top row), average concentration fields and $\overline{C}(x, y, t)$ (middle row) and fluctuation concentration fields c' (bottom row) thus defined are represented for the an examples of I fractal grid experiment in figure 5 and simple Cartesian grid experiments in figure 6, at times ranging from t_i to about $t_i + 25$ s. The second row allows to see clearly the mean-mixing process and the global propagation of the scalar patch and tank concentration homogenization, initially jet dominated (first two columns for each figure) and quickly then mean flow dominated (last two columns).

The spatial distribution of concentration fluctuations provides further insight on the mixing process. For the Fractal I grid (figure 5), turbulent scalar fluctuations appear successively in various regions along the mixing process: first in the top corners, then in the bottom corners. In contrast, for the Simple Cartesian (figure 6) grid, fluctuations spread across the fluid domain, first vertically and then horizontally, as an effect of the injection process first and the relatively isotropic turbulence diffusion next.

Mean advection directly impacts the local turbulent mixing process: the mean flow associate to the Fractal I grid, which was extensively described in Musy et al. (2024), defines early turbulent mixing zones, delaying turbulence action in some others. Simple Cartesian grid's weaker mean flow allows for a broader and earlier spatial distribution of turbulent mixing, however not necessarily implying a homogeneous and fast mixing time. Analysis of the the fractal Square and fractal Cartesian grids' dynamics (not shown here) lead to similar conclusions regarding the role of the mean flow on the overall mixing dynamics.

4. Conclusions and perspectives

In this work, the mixing mechanisms generated by fractal oscillating grids were investigated using pH-sensitive PLIF experiments. Four grid geometries were examined: a simple Cartesian grid used as the reference configuration and the I-shaped fractal grid, extensively analyzed in this study, together with fractal Cartesian and fractal Square grids.

Fractal grids were generally observed to lead to faster mixing than the reference Cartesian grid. A Reynolds decomposition adapted to the transient characteristics of the experiments made it possible to analyze the influence of mean flow and turbulent structures on the mixing dynamics. The grids with the highest mixing efficiency (in particular the I-fractal grid) were those in which the organized upward mean flow rapidly dispersed the injection patch near the surface, followed by a downward motion transporting it toward the highly turbulent region swept by the grid, where mixing is most effective.

These observations highlight the two dominant mixing mechanisms in fractal oscillating grid stirred tanks, namely mean flow advection and turbulent small-scale dispersion. Although mean flow effects are traditionally regarded as detrimental in the use of oscillating grid systems for fundamental applications, they are shown here to positively contribute to overall mixing performance. Meanwhile, the local shear stresses produced near the stirring device (the grid) remain low compared with those found in conventional impeller-stirred tanks, opening promising perspectives for bio-process applications involving shear-sensitive materials.

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Nomenclature

PIV	Particle image velocimetry
W	Width [cm]
A_{osc}	Oscillation amplitude [mm]
f_{osc}	Oscillation frequency [Hz]
z_{eq}	Grid equilibrium position [cm]
H	Height of fluid in the tank [cm]
$PLIF$	Planar laser-induced fluorescence
f_{acq}	Acquisition frequency [Hz]
dt	Shutter time [ms]
z_{eq}	Grid equilibrium position [cm]
I	Pixel intensity [gray level]
I_c	Mean pixel intensity of the post-processed calibration image [gray level]
I_n	Normalized mean pixel intensity
I_{max}	Maximum mean pixel intensity [gray level]
I_{min}	Minimum mean pixel intensity [gray level]
α	Fluorescence sensitivity to pH in the region of intense change (fitting coefficient)
β	pH value that the sensitivity is highest (slope of the curve, fitting coefficient)
C	Instantaneous concentration value
I_{nm}	normalized pixel intensities of experiment images
σ_{px}	Standard derivation of pixel intensities over each image
$\sigma_{px,max}$	Maximum standard deviation of pixel intensities in ensemble of experiment's images
I_s	Intensity of segregation
M	Mixing index
M_n	Normalized mixing index
t_M	Mixing time
$\overline{C}$	Averaged concentration field
c'	Fluctuating concentration field

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