



Mixing Studies with Impinging Jets

PIV/PLIF Experiments and CFD Simulation

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by

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To my family and friends

*Ali vimos a veemência do visível
O aparecer total exposto inteiro
E aquilo que nem ousáramos sonhar
Era o verdadeiro*

Sophia de Mello Breyner Andresen

Navegações, 1977

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Abstract

The present work concerns the identification of mixing mechanisms and flow regimes, and the characterization of the lamellar microstructure of the flow arising from mixing of two impinging jets in a mixing chamber of a Reaction Injection Molding (RIM) machine with typical dimensions. The jets' interaction under balanced and unbalanced flow conditions is also studied.

Mixing length scales and stretching properties of the impinging jets flow are studied from 3-dimensional Computational Fluid Dynamics (CFD) simulations coupled with a Lagrangian tracking of the position and deformation of passive material fluid elements. The lamellar structure of the flow is also studied in a Eulerian-Eulerian approach from a two-dimensional non-diffusive two-phase CFD model of the flow coupled with a dynamic grid adaption algorithm. Mixing patterns in the flow are obtained and studied experimentally with the Planar Laser Induced Fluorescence (PLIF) technique. These studies allow the clear identification of the mixing mechanisms in the impingement jets flow. Operational parameters such as the jets Reynolds number and kinetic energy rate ratio are studied and shown to be critical in impingement mixing.

An elastic analogy of the two impinging jets behavior is used to predict the impingement point position under different flow conditions, showing good agreement with CFD simulations of the flow and results from Particle Image Velocimetry experiments. This study offers deeper insight on the system behavior under unbalanced jets flow conditions in order to offer robustness to polymer production in RIM from monomers or pre-polymers formulations with different physical properties or stoichiometric ratios different from one.

Experimental and numerical results show that advective dynamic mixing in the impingement jets flow occurs above a critical Reynolds number equal to 110.

Resumo

A presente dissertação concentra-se na identificação de mecanismos de mistura e de regimes de escoamento, e na caracterização da microestrutura lamelar do escoamento resultante da mistura por jactos opostos numa câmara de mistura com dimensões típicas de uma máquina de moldagem por injeção com reação química (RIM). A interação dos jatos em condições de equilíbrio e não equilíbrio é também estudada.

As escalas de mistura e as propriedades de estiramento do escoamento de jatos opostos são estudadas através de simulações de Computação em Fluidos Dinâmicos (CFD) tridimensionais em conjunto com a monitorização, numa perspectiva Lagrangiana, da posição e deformação de elementos passivos infinitesimais. A estrutura lamelar do escoamento é também estudada numa perspectiva Euleriana-Euleriana através de um modelo CFD bidimensional do transporte de duas fases imiscíveis sem difusão molecular ao qual é associado um algoritmo dinâmico de adaptação da grelha numérica. Os padrões de mistura são obtidos e estudados experimentalmente a partir da técnica de Fluorescência Induzida por Laser (PLIF). Estes estudos permitem uma clara identificação dos mecanismos de mistura no escoamento de jatos opostos em colisão. São estudados parâmetros operacionais como o número de Reynolds e a razão do débito de energia cinética dos jatos, os quais se mostram serem críticos em mistura por jatos opostos.

Uma analogia do comportamento dos dois jatos opostos em colisão com um sistema elástico é desenvolvida para prever a posição do ponto de impacto em diferentes condições de escoamento, mostrando boa concordância com simulações em CFD do escoamento e resultados experimentais de Velocimetria por Imagem de Partículas (PIV). Este estudo oferece uma melhor compreensão do comportamento do sistema em condições de não equilíbrio de modo a tornar mais robusto o processo de produção de polímeros por RIM a partir de formulações de monómeros ou pré-polímeros com propriedade físicas diferentes ou razões estequiométricas diferentes de um.

Resultados experimentais e numéricos mostram que a mistura dinâmica por mecanismos convectivos em escoamentos de jatos opostos ocorre acima de um número Reynolds crítico igual a 110.

Résumé

Ce travail porte sur l'identification des mécanismes de mélange et des régimes d'écoulement, et sur la caractérisation de la microstructure lamellaire de l'écoulement résultant du mélange de deux jets incidents dans une chambre de mélange d'une machine avec des dimensions typiques de moulage par injection avec réaction chimique (RIM). L'interaction des jets dans des conditions de flux symétriques et asymétriques est également étudiée.

Les échelles de mélange et les propriétés d'étirement de l'écoulement de jets opposés sont étudiés à partir des simulations 3-dimensionnelles de dynamique computationnelle des fluides (CFD) couplées avec un suivi Lagrangien de la position et de la déformation d'éléments fluides passifs infinitésimaux. La structure lamellaire de l'écoulement est également étudiée sous une approche Eulérienne-Eulérienne avec un modèle non-diffusif en CFD à deux dimensions et à deux phases de l'écoulement, couplé à un algorithme d'adaptation dynamique de la grille de calcul. Les structures de mélange de l'écoulement sont obtenues et étudiées expérimentalement avec la technique de Fluorescence Induite par Laser (PLIF). Ces études permettent d'identifier clairement les mécanismes de mélange de l'écoulement de jets opposés. Paramètres de fonctionnement comme les nombres de Reynolds et le quotient des débits d'énergie cinétique des jets sont étudiés, lesquels se présentent comme critiques en mélanges avec des jets opposés.

Une analogie élastique du comportement des deux jets incidents est utilisée pour prédire la position du point d'impact dans des conditions d'écoulement différentes, montrant un bon accord avec les simulations CFD de l'écoulement et les résultats d'expériences de Vélocimétrie par Images de Particules (PIV). Cette étude permet de mieux comprendre le comportement du système dans des conditions de non-équilibre afin d'offrir robustesse à la production de polymères en RIM à partir de formulations de monomères ou pré-polymères avec des propriétés physiques différentes ou des rapports stœchiométriques autres qu'un.

Les résultats expérimentaux et numériques montrent que le mélange par convection dynamique dans l'écoulement de jets opposés se produit au-dessus d'un nombre de Reynolds critique égal à 110.

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Notation

a	Amplitude of the perturbation parameter
b	Duration of the perturbation parameter
$\mathbf{C}$	Right Cauchy-Green deformation tensor
C	Courant number
$C_{\text{Rhodamine6G}}$	Rhodamine 6G concentration [kg/m^3]
d	Injector diameter [m]
D	Chamber diameter [m]
$\mathbf{D}$	Stretching tensor [s^{-1}]
$\mathcal{D}$	Molecular diffusivity [m^2/s]
d_p	Seeding particles mean diameter [m]
dR	Initial distance separating two particles [m]
dr	Distance separating two particles [m]
$\dot{E}_K$	Kinetic energy rate [W]
$\dot{E}_{K,in}$	Kinetic energy rate at the injectors [W]
ΔE_p	Stored potential energy [J]
$\mathbf{F}$	Deformation gradient tensor
f	Perturbation function
F	Cumulative distribution function
$F_{i \rightarrow j}$	Force applied by spring i to spring j [N]
f_{laser}	Laser pulsing frequency [Hz]
$f_{\text{laser,max}}$	Maximum laser pulsing frequency [Hz]
H	Chamber height [m]
H	Heaviside function
$\mathbf{I}$	Identity tensor
i	Imaginary unit
I	Pixel grey scale intensity
I_s	Intensity of segregation
$I_{s,0}$	Reference intensity of segregation

k	Spring constant [N/m]
k	Kinetic energy of the velocity fluctuations [m^2/s^2]
l	Injector length [m]
l_i	Length of spring or jet i [m]
l'	Distance from the momentum source point to the chamber inlet [m]
$\dot{m}$	Mass flow rate [kg/s]
$\mathbf{n}$	Outside normal unitary vector to a control volume
n	Wave number [m^{-1}]
n_r	Number levels of grid refinement
N_{elements}	Number of elements in the numerical grid
$N_{\text{elements},0}$	Initial number of elements in the numerical grid
$N_{\text{particles}}$	Number of particles
nD	Mixing zone length from the top of the chamber [m]
$\mathcal{O}(i)$	Order of magnitude of i
p	Pressure [Pa]
P	Power spectrum
p_0	Reference pressure
Q	Flow rate [m^3/s]
$\mathbf{R}$	Orthogonal tensor
r	Radial position at the injectors' circular section [m]
$R(r)$	Coefficient of correlation function
R	Injector radius [m]
r_s	Volumetric flow rate ratio
Re	Reynolds number
s	Striation thickness [m]
$\bar{s}$	Average striation thickness [m]
s_0	Initial striation thickness [m]
s_L	Scale of segregation [m]
$s_{L,0}$	Initial scale of segregation [m]
St	Stokes number
t	Flow time [s]
T	Temperature [$^{\circ}\text{C}$]

Δt	Integration time step size [s]
t_0	Initial time [s]
Δt_{beams}	Interval between consecutive beams in the PIV experiments [s]
$t_{\text{simulation}}$	Simulation calculation time [s]
$\mathbf{u}$	Velocity vector field [m/s]
$\mathbf{U}$	Positive symmetric tensor
u	Axial velocity [m/s]
u_{chamber}	Average velocity in the mixing chamber [m/s]
u_{inj}	Average velocity at the injectors [m/s]
v	Radial velocity [m/s]
$V_{\text{dissipation}}$	Volume where dynamic impingement mixing occurs [m ³]
$\mathbf{x}$	Position vector [m]
$\mathbf{X}$	Initial position vector [m]
Δx	Numerical grid element length [m]
x_i	Component of $\mathbf{x}$ in the i -direction [m]
$\Delta x_{\text{int. region}}$	Length of the interrogation region in the PIV experiments [m]
x_{IP}	Impingement point position [m]

Greek letters

α_i	Volume fraction of phase i
$\bar{\alpha}$	Spatial volume fraction average
$\dot{\gamma}$	Strain rate [s ⁻¹]
δ	Local thickness of a layer [m]
ε	Energy dissipation rate [W]
$\bar{\varepsilon}_{2D}$	2-dimensional energy dissipation rate [W/m]
$\hat{\varepsilon}$	Relative energy dissipation rate [m ⁻¹]
ζ	Non-dimensional rate of strain
λ_i	Stretching/compression in the i -th principal direction of deformation
λ	Stretching function
$\dot{\lambda}$	Stretching rate [s ⁻¹]
Λ	Lyapunov exponent

λ_K	Kolmogorov length scale [m]
μ	Fluid viscosity [Pa · s]
ν	Kinematic viscosity [m ² /s]
ξ	Non-dimensional impingement point position
ξ	Distance from which no correlation exists in the concentration field [m]
ρ	Fluid density [kg/m ³]
$\bar{\rho}$	Average mixture density weighted by the flow rate [kg/m ³]
ρ_p	Seeding particles density [kg/m ³]
σ_α^2	Spatial variance of the volume fraction
σ_{I_s}	Temporal standard deviation of I_s
τ	Chamber mean residence time [s]
Φ_t	Flow or motion
ϕ_K	Kinetic energy rate ratio
ϕ_M	Momentum rate ratio
ϕ_{FR}	Flow rate ratio
Ω	Volume or area domain
$\partial\Omega$	Domain boundary surface or perimeter
$\hat{\omega}$	Non-dimensional vorticity

Mathematical operations

$\langle \rangle$	Spatial average
$\partial_i(\cdot)$	Partial derivative with respect to i
$\hat{\mathbf{e}}_i$	Unit vector in the i -coordinate direction
$\nabla(\cdot)$	Gradient operator
$\nabla_{\mathbf{x}}(\cdot)$	Differentiation with respect to $\mathbf{X}$, $\partial_{x_i}(\cdot)\hat{\mathbf{e}}_i$
$\nabla^2(\cdot)$	Laplacian operator
$(\cdot)^T$	Transpose

Subscripts

1	Left injector
2	Right injector

Abbreviations

2D	Two-dimensional
3D	Three-dimensional
CFD	Computational Fluid Dynamics
CIJ	Confined Impinging Jets
DNS	Direct Numerical Simulation
FFT	Fast Fourier Transform
LDA	Laser Doppler Anemometry
NAJ	Narrow Axisymmetric Jet
PIV	Particle Image Velocimetry
PLIF	Planar Laser Induced Fluorescence
RAM	Random Access Memory
VOF	Volume-of-Fluid

1 Introduction

Mixing is a unit operation present in physical or chemical industrial processes that require the contact of reactants for chemical reactions or homogenization of solutions or solid particles. Over the last decades, mixing by opposed impinging jets appeared as an alternative to conventional mixing in stirred tanks and has revealed to be well suited for continuous processes that require a rapid homogenization of streams (Siddiqui et al., 2009). In this technology, two or more reactants are injected into a confined space, the mixing chamber, through opposed jets. The rapid deceleration and energy dissipation of the two jets, after impingement in the confined space, allows for mixing to be achieved rapidly and without movable mechanical parts, such as stirring devices. Opposed impinging jets mixers/reactors are generally divided into two types, depending on its geometry: Confined impinging jets mixers (CIJ) with cylindrical chamber and injectors (Mahajan and Kirwan, 1996; Johnson and Prud'homme, 2003; Sohrabi and Zareikar, 2005; Marchisio, 2009; Siddiqui et al., 2009; Icardi et al., 2011) and T-jets mixers with rectangular cross-section chamber and injectors (Engler et al., 2004; Bothe et al., 2006; Gradl et al., 2006; Bothe et al., 2008; Adeosun and Lawal, 2009; Gradl and Peukert, 2009). These devices have a wide range of applications in different industrial processes such as precipitators for

nanoparticle production or in reactive polymer injection (RIM). Although the results of the present study are relevant for the different applications of impinging jets mixing, Reaction Injection Molding technology is the focus of this dissertation as the case study for the application of this mixing method. This is due to its economic relevance and growing interest in applications for the automotive and aeronautical industries.

In Section 1.1 the RIM process is presented, namely its applications, challenges and economic relevance. In Section 1.2 the previous works that are the starting point to this dissertation are described. In Section 1.3 the thesis objectives and layout are presented.

1.1 RIM Process: Applications, Challenges and Economic Impact

Reaction Injection Molding (RIM) is an industrial process for the production of plastic parts conceived in the laboratories of Bayer AG in Leverkusen in 1964 (Berins, 1991). In this process, two or more viscous liquid monomers or pre-polymers are injected and mixed in a cylindrical mixing chamber by opposed impinging jets. The mixture that leaves the mixing chamber is discharged into a mold where the polymerization occurs and the plastic part is formed. Efficient mixing of the two monomers, during the residence time inside the mixing chamber before entering the mold, is crucial in order to avoid zones of unreacted monomers that may lead to structural defects and the eventual rejection of the plastic parts (Kolodziej et al., 1986).

The injected fluids have viscosities in the range of 10 to 1000 mPa·s and are introduced in the cylindrical mixing chamber through injectors with 1–3 mm of diameter at high pressure (100 to 200 bar) and velocities in the range of 10 to 150 m/s. The mixing chamber is typically a cylinder with 3–15 mm diameter and length around 5 times the diameter (Oertel, 1985). The mean residence time in the mixing chamber is in the range of 10 – 100 ms.

The RIM process and equipment consist of the following major steps and components as schematized in Figure 1.1:

- Monomers or pre-polymers are initially recirculated in a circuit consisting of storage tanks (7) with low pressure recirculation pumps (4) and heat exchangers (5). The reactants' recirculation step insures low viscosities and temperature uniformity at the beginning and between of each injection step. Most of the machine time is spent in the recirculation step (Macosko, 1989).
- For the injection in the mixing chamber, high pressure injection pistons (6) are filled with the reacting fluids and actuated for the desired flow rates and stoichiometric proportions.
- The reactants are injected in the cylindrical mixing chamber (1) from the injectors (8) where they are mixed by impingement and discharged to a mold (2). After the injection step and when the mold is filled, a cleanout piston (3) is used to remove any remaining material from the mixing chamber.
- Depending on the size of the formed part, the mold is usually filled in 1 to 2 s. Polymerization occurring inside the mold turns the polymeric material into a solid material that is then removed from the mold. Demolding times depend on the used formulation and catalyst; some tested formulations allow for the demolding to be made 30 s after injection (Macosko, 1989).

The complex design of RIM machines (see Figure 1.2) and respective control systems places its minimum cost around €60 000. The mixing head, by itself, has a minimum value around €15 000. If the machine is made for the production of composite material, its price increases almost one order of magnitude, to values around €400 000. (private quote from a leading company).

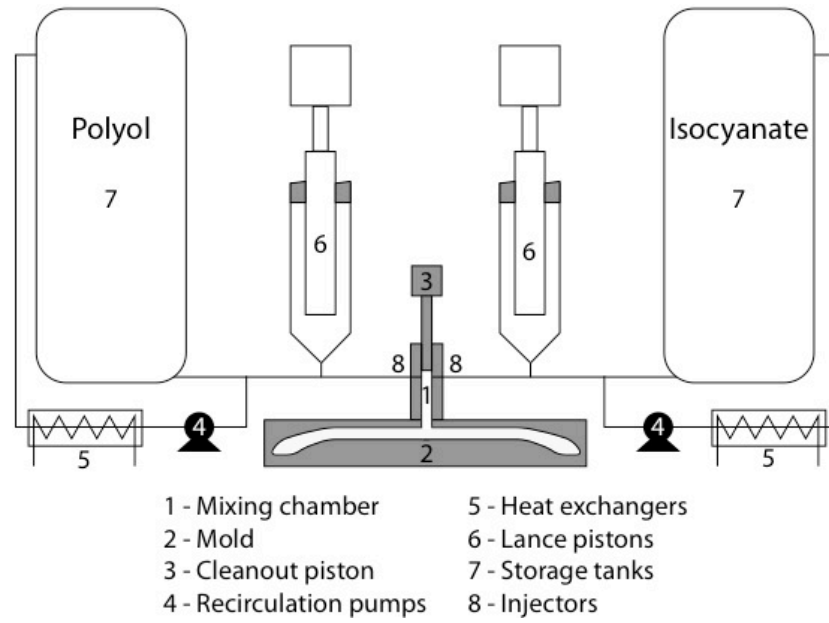


Figure 1.1. Scheme of a typical RIM machine for the production of polyurethanes (adapted from Macosko (1989)).



Figure 1.2. Two RIM machine models commercialized by KraussMaffei.

RIM was originally developed as a technology for the production of polyurethanes, but has also been used to produce other polymers such as polyureas, polyamides, polyesters and polyepoxides. RIM has its main application in the production of large plastic parts for the automotive and aeronautical industry (Macosko, 1989).

Although it is possible to produce a wide range of polymer materials using RIM and combine them with different loadings, from nanoparticles to fibbers, this technology has failed to live up to its full potential since its development in the 60's. Per part costs are more expensive when compared with injection molding of extruded polymers, and this process is only attractive for the production of small runs of a few hundred parts for which the lower investments costs outweighs the produced part cost (Rosato et al., 2004). The main limitations of standard RIM systems are:

- The process is not robust and RIM machines need frequent tuning due to high sensitivity to environmental conditions and operational variables. It is important to adapt the process thermal conditioning system to the location of plant.
- The manufacturer must develop expertise to modify the process variables and formulations of the many available reactants and catalysts, in order to obtain plastic parts with the desired mechanical properties.
- The lack of expertise in the designing of molds for reactive injection affects negatively operations after demolding and the finishing of the produced parts. Most knowledge comes from the technology of injection molding of extruded polymers, which has different requirements.

The process can be optimized to benefit from its advantages (Rosato et al., 2004):

- Lower mold costs due to low injection pressures; an in-mold pressure of 4 to 10 bar that is usually suited for RIM contrasts with the 300 to 2000 bar required for thermoplastic injection molding.

- Design flexibility of the produced parts since the mold is filled with low viscosity fluids; the versatility in processing options and chemical systems used can be applied to produce high-quality and highly styled plastic products.
- Low operating temperatures.
- Since polymerization occurs inside the mold, no molded-in stresses are formed of in the produced part, which can cause warping or cracking of the part after demolding.

Polyurethanes still represent nowadays the main product obtained from RIM, which also represent a significant share of the annual revenue of some of the largest chemical industries such as Bayer AG (Bayer (2008), (2009), (2010) and (2011)), the BASF Group (BASF (2008), (2009), (2010) and (2011)) and the Huntsman Company (Huntsman (2008), (2009), (2010) and (2011)). Table 1.1 resumes the annual revenue of the mentioned chemical companies, with the indication of the share relative to the polymer and polyurethane sales. For the three companies, the polymer sector is among the sectors with the highest sales. Apart from the results for the year 2009, a consequence of the late-2000's global economic crisis, Table 1.1 and Figure 1.3 show that polymers and polyurethanes are growing industries that represent a parcel of billions of Euros in the chemicals market. The values reported by Huntsman were converted from Dollars to Euros at an average exchange rate over the last 5 years of 0.73 €/£.

Table 1.1. Annual revenue in billions of Euros of some of the major chemical industries.

	Total				Polymers				Polyurethanes			
	2008	2009	2010	2011	2008	2009	2010	2011	2008	2009	2010	2011
BASF	62.3	50.7	63.9	73.5	9.7	7.1	9.8	10.0	5.1	4.1	5.4	5.8
Huntsman	7.4	5.5	6.7	8.2	-	-	-	-	2.8	2.1	2.6	3.2
Bayer AG	32.9	31.2	35.1	36.5	-	-	-	-	5.1	3.8	5.0	5.4

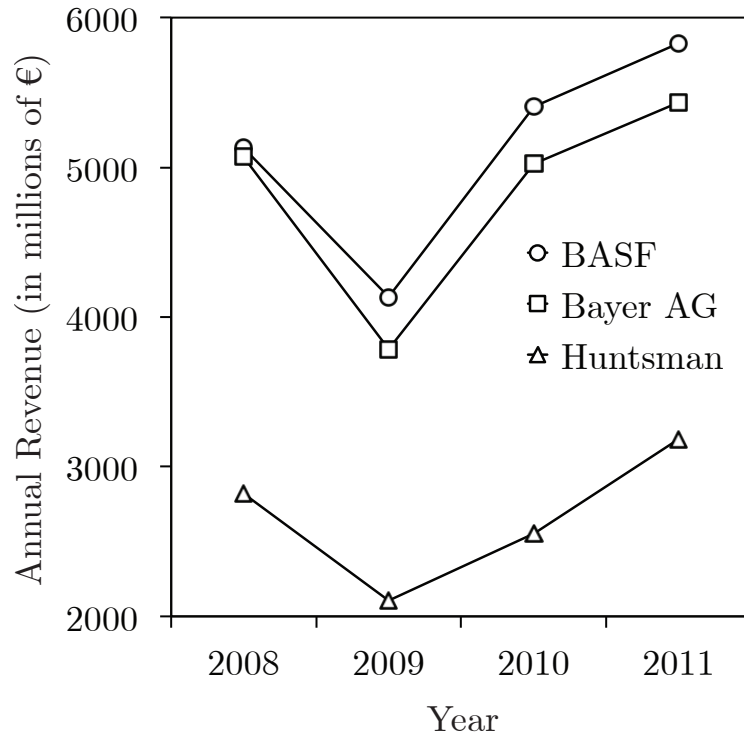


Figure 1.3. Annual revenue in polyurethanes sales for some of the major chemical industries.

The high impact that RIM technology can have on the chemical and automotive industries and the need of solutions that turn the process more robust are the major driving force for this work that was initiated by Teixeira (2000) and Santos (2003) and continues in this dissertation.

1.2 Previous Work

This work is the follow up of the studies done by Teixeira (2000) and Santos (2003) for their Ph.D. thesis. That being so, before the presentation of the objectives and layout of this thesis, it is imperative to situate its departure point.

Teixeira (2000) studied experimentally and computationally from dynamic simulations using CFD tools the flow in the mixing head of a RIM machine, with the purpose of better understanding the mixing mechanisms involved in the process and checking the capacity of the available means at the time for the simulation of these

kind of mixing problems. The study was limited to the case where a Newtonian fluid is fed to both injectors with the same flow rate.

Teixeira (2000) measured the horizontal components of the fluid's velocity in the mixing chamber built at laboratorial scale using a Laser Doppler Anemometry (LDA) system. The Reynolds number was varied between 50 and 600, covering the usual range in industrial practice. Experimental results have allowed the view of the spatial distribution of the flow properties in the chamber, namely average velocities, turbulence intensities and Reynolds stresses. The existence of structures with well defined shapes and similar to all the studied Reynolds numbers was verified. It is shown that the phenomenon reported as the existence of a critical Reynolds number has its origin in the behavior of the flow. The existence of characteristic frequencies in the flow was confirmed.

Teixeira (2000) also performed Direct Numerical Simulations (DNS) of the flow in the mixing chamber considering a 2D geometric model. The DNS strategy was used to study the dynamic behavior of the flow. A parametric sensibility study was made to the simulations' results for the parameters involved in the calculations. The hydrodynamic simulations' results show the mixing mechanisms present in the flow, which are in agreement with the experimentally obtained data and with other published works. This work has shown that the dynamic simulations of the flow in a RIM machine can be a useful tool in the understanding and optimization of the mixing process.

Following Teixeira (2000), the work of Santos (2003) studied the mixing mechanisms in the mixing chamber of a high pressure RIM machine from both from LDA and PIV experiments and CFD simulations.

In the thesis of Santos (2003), typical oscillation frequencies were detected from the LDA data for Reynolds numbers ranging from 250 to 600. The CFD simulations were based on 2D geometric domains and reproduce correctly the dynamic behavior observed experimentally and provide immediate insight into the oscillations base

mechanisms. Operational parameters such as the Reynolds number, the Froude number and the jets' momentum ratio were studied and proven to be critical for mixing in the RIM process. The flow field structures were imaged experimentally using the PIV technique. The effect of Reynolds number was studied between 100 and 500, and detailed measurements were made around the region of the critical Reynolds number that has been shown to be set at 120. Dynamic 2D CFD simulations of mass transfer in the RIM process were performed at Reynolds numbers ranging from 100 to 600. Chemical reaction is simulated dynamically for Reynolds number 300.

1.3 Thesis Objectives and Layout

The main goals of this work are the identification of mixing mechanisms and flow regimes, and the characterization of the lamellar microstructure of the flow arising from mixing of two impinging jets in a mixing chamber of a RIM machine with typical dimensions. The jets' interaction under balanced and unbalanced flow conditions are also studied to set rules for a more robust operation of RIM machines under different operating conditions. The flow in the mixing chamber was studied numerically from Computational Fluid Dynamics (CFD) tools and experimentally from Particle Image Velocimetry (PIV). Mixing patterns in the flow were obtained and studied with the Planar Laser Induced Fluorescence (PLIF) technique.

This work adds further understanding about the RIM process to the one already gained from the work of Teixeira (2000) and Santos (2003) by the use of a 3D geometrical domain for the hydrodynamic simulations, which could not be practically simulated during the periods of the previous works. Numerical problems and computational limitations also led Santos (2003) to consider unrealistic diffusional values for his Dynamic 2D CFD simulations of mass transfer, 10^3 higher than the actual values. Consequently, the fluids at the outlet of the mixing chamber were too homogeneous and so it was not possible to identify any lamellar structures. To

overcome this limitation and study the formation of lamellae, a non-diffusive mass transfer model has been developed.

The state of the art on impinging jets mixing is presented according to its relevance in the beginning of each chapter. The thesis is divided as follows:

- Chapter 2 focuses on the study of the mixing length scales and stretching properties of the flow in a confined impinging opposed jets mixer from 3D CFD simulations coupled with a Lagrangian tracking of the position and deformation of passive material fluid elements. A distribution of striation thicknesses is predicted from the numerical simulations.
- In Chapter 3 a model to describe the lamellar structure of the impinging jets' flow is studied from an Eulerian-Eulerian approach using a 2-dimensional CFD model. The model describes a purely advective mixing of two immiscible feeding fluids using the VOF model, without the effect of diffusion and surface tension. A dynamic adaptive grid algorithm in the region of higher gradients of volume fraction is included in the simulation model to describe smaller mixing length scales without the need to refine equally the entire domain.
- In Chapter 4, PLIF flow visualization technique is used to obtain 2D concentrations fields at the top and bottom of the mixing chamber. One of the injected streams was marked with rhodamine 6G which emits fluorescence when excited by laser light, allowing to detect mixing patterns in the flow. The obtained fields are used to analyze qualitatively and quantitatively the effect of the Reynolds number and the impinging jets' unbalancing on mixing.
- Finally, in Chapter 5, the impinging jets' interaction in the mixing chamber is studied theoretically, experimentally and numerically. An elastic analogy of the two impinging jets behavior is used to predict the impingement point position under different flow conditions, showing good agreement with CFD simulations of the flow and results from PIV experiments. The study offers deeper insight on the system behavior under unbalanced jets flow conditions in

order to offer robustness and rules set to set polymer production in RIM from monomers or pre-polymers formulations with different physical properties or volumetric stoichiometric ratios different from 1.

Additionally, animations of experimental and numerical results are included in a DVD attached to this dissertation. The included animations were prepared to be played with Apple QuickTimeTM Player for Microsoft Windows or Mac OS X. Animations were organized in the following folders, where “~” indicates the root of the DVD:

- Folder “~/Chapter2/” contains animations from the particle tracking in the dynamic 3D CFD simulations at different Reynolds numbers.
- Folder “~/Chapter3/” contains animations from dynamic the 2D CFD simulations with the non-diffusive two-phase model. Variables shown are the volume fraction of each phase and the vorticity of the flow.
- Folder “~/Chapter4/” contains animations from the PLIF experimental results at the top and bottom of the transparent mixing chamber.

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2 Stretching Properties of Impinging Jets Flow

2.1 Introduction and State of the Art

Mixing from impinging jets occurs due to the stretching and folding of the two streams in the mixing chamber promoted by a hydrodynamic instability. The result is the formation of a lamellar structure out of the fed streams. Figure 2.1 shows an image of the lamellar structure of the flow resulting from impingement mixing obtained from Planar Laser Induced Fluorescence experiments, described later in Chapter 4. In the particular case of Reaction Injection Molding (RIM), due to the short residence times (10 to 100 ms) inside the mixing chamber, molecular diffusion is not expected to occur significantly. Instead, distances between the two initially segregated monomers or pre-polymers must be reduced rapidly in the mixing chamber by advection mechanisms. Later in the mold, diffusion must homogenize the mixture at a molecular level in a time scale smaller than the reaction time scale. For the rate of polymerization reaction to become chemically controlled, the formed lamellar structure must have striation thicknesses, s , of the order of $10\text{ }\mu\text{m}$ (Lee et al., 1980). The striation thickness in a binary layered mixture of components A and B is defined as

$$s = \frac{\delta_A + \delta_B}{2}, \quad (2.1)$$

that is, half the sum of the local laminae thickness of each component, δ_A and δ_B .

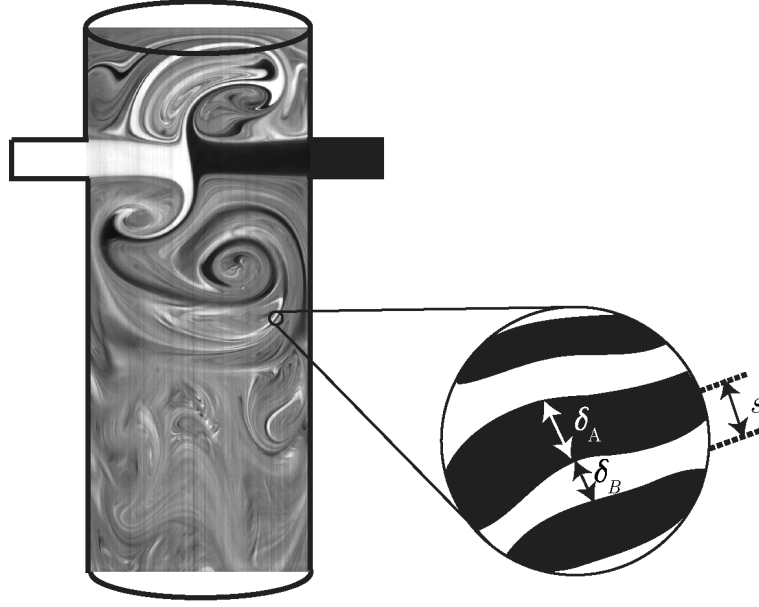


Figure 2.1. PLIF image of impingement mixing of a binary mixture.

Lee et al. (1980) proposed a model to estimate an average striation thickness, $\bar{s}$, in RIM's lamellar structure, considering that the impingement process is, on a local scale, a rapid, two dimensional stagnation flow, and assuming that the stretching remains in the plane of each lamina. From these considerations, they obtained

$$\bar{s} = \left(\frac{2(1 + r_s^{-1})d^5}{nD^3} \right)^{1/2} \text{Re}^{-1/2} \quad (2.2)$$

where d and D are the injectors and chamber diameters, respectively, r_s volumetric flow rate ratio between the two jets, and nD is the distance from the top of the mixing chamber until the point where dynamic impingement mixing occurs, observed to be $nD \approx 2D$ to $3D$ (Kolodziej et al., 1982). The Reynolds number is defined as

$$\text{Re} = \frac{\rho d u_{\text{inj}}}{\mu}. \quad (2.3)$$

where ρ and μ are the fluids density and viscosity, respectively, and u_{inj} the average velocity at the injector.

Tucker III and Suh (1980) proposed, using isotropic turbulence theory considerations, that an average striation thickness in RIM is determined instead by the smallest hydrodynamic length scale of the flow, the Kolmogorov scale. Tucker III and Suh (1980) state that mixture clumps smaller than smallest hydrodynamic length scale are completely contained in a single eddy motion, thus not experiencing shear deformation. From dimensional considerations, Tucker III and Suh (1980) obtained

$$\bar{s} \propto \frac{d}{\text{Re}^{3/4}}, \quad (2.4)$$

carried out further by (Lee et al., 1980) to obtain

$$\bar{s} = \left(\frac{ndD^3}{1 + r_s^{-1}} \right)^{1/4} \text{Re}^{-3/4}. \quad (2.5)$$

The models of Lee et al. (1980) and Tucker III and Suh (1980) for the mean striation thickness show some similarities, such as a dependency on the geometrical mixing chamber parameters and the formation of smaller lamellae thicknesses as Re is increased. However, each model predicts substantially different values for the striation thickness. For typical RIM dimensions at $\text{Re} = 300$, Equation (2.2) estimates an average striation scale around $6 \mu\text{m}$, while Equation (2.5) estimates a value around $100 \mu\text{m}$. The two models main limitation resides in the fact that flow heterogeneities and different residence times inside the mixing chamber will produce a distribution of striation scales, which both fail to provide. In fact, the quality of the final plastic part will be mostly influenced by the largest lamellae. Large striation structures may not react in the required time due to diffusional limitations and lead to structural or aesthetical defects. Studies on the effect of a distribution of striation thicknesses in the polymerization reaction can be found in Fields and Ottino (1987a) and (1987b), and Sokolov and Blumen (1991a) and (1991b).

Kolodziej et al. (1982) measured experimentally a distribution of striation thickness in the actual formed plastic and its dependence on the Reynolds number by mixing and letting react a polyurethane formulation in a RIM machine with typical dimensions, marking the polyol reactant with carbon black. With microscope visualizations of the lamellar structure in the formed plastic, a striation thickness histogram was determined for each Reynolds number. Due to the use of the carbon black marker, the polyol reactant laminae appear dark in the captured images. The striation thickness distribution was measured in a polymer sample extracted from the center of the tube mold to avoid the effect of further striation thinning in the shear flow in the tube. Large striations ($500\text{ }\mu\text{m}$) were not recorded since they correspond to unreacted polymer, and, due to optical limitations, striation thickness less than $10\text{ }\mu\text{m}$ were not detectable. The authors concluded that the obtained average striation thickness is not quantitatively accurate to validate either Equation (2.2) or (2.5). Stoichiometry seems to have been the only concern for setting the two feeding streams flow rates, resulting in $r_s = 1.5$, which, as it will be discussed in Chapter 4 and 5, presents some concerns since the jets' imbalance can affect strongly mixing quality.

Baldyga and Bourne (1983) developed a model that estimates a distribution of striation thickness. The proposed model also predicts a reduction of scales in the light of statistical theory of turbulent diffusion combined with an exponential residence time distribution of the fluid inside the mixing zone of the chamber to provide a distribution of lamellae thickness. Further deformation of the striation scales below the Kolmogorov scale, where viscous forces and a dependence on Re would be significant, was not considered. Most certainly due to this assumption, results from this model show good agreement with the experimental results of Kolodziej et al. (1982), where scales smaller than $10\text{ }\mu\text{m}$ could not be detected.

Later, Bourne and Garcia-Rosas (1985) included in the model of Baldyga and Bourne (1983) the further striation thinning due to laminar parallel flow in the remain mixing chamber. The model shows that the average striation thickness is reduced at

least one order of magnitude in the length from the dynamic mixing zone to the outlet. The striation thickness probability distribution below or equal to $10\text{ }\mu\text{m}$ also increases significantly from zero to more than half. These results seemed to explain, only from isotropic and homogeneous turbulence theory considerations of Tucker III and Suh (1980) and Baldyga and Bourne (1983), how mixing length scales are reduced in the RIM machine's mixing chamber to values that allow complete chemical reaction in the mold, despite of non turbulent flow in the mixing chamber in the range of industrial practice.

Later, May (1996) obtained a striation thickness distribution in RIM from a computational fluid dynamics point of view. A field equation was derived from statistical theory of turbulent diffusion in order to describe the striation thickness reduction. May (1996) presents similar results to the ones obtained by Baldyga and Bourne (1983). The difference between them reside in that no assumption for the chamber residence time distribution is necessary by solving directly the flow field and turbulent properties equations.

Since May (1996), no known further developments have been published on the characterization of the lamellar structure in RIM. In the computational study of mixing in RIM made by Santos et al. (2005) the diffusivity value was set to a value too high to be able to identify any lamellar structures from the results. In the CFD study of Fonte et al. (2011), despite avoiding the effect of diffusion, the spatial discretization was not sufficiently refined to include all scales. Even with the evolution of the available computational power, the description of the reduction of scales in RIM by direct simulation of material surfaces is still extremely resource consuming. Even for a simplified 2D model of the flow, as it will be seen in Chapter 3, the exponential generation of interfacial area demands the use of an increasingly dense spatial discretization in time to properly describe the interfaces and avoid numerical problems. Up to this date, the description of a lamellar structure in RIM from simulation of fluid interfaces is yet not feasible for 3D flows. Instead,

the length scales generated in RIM can be predicted by the stretching distributions in the flow since their statistical properties are identical (Paul et al., 2003).

In this chapter a Lagrangian approach is used for the study of stretching in RIM. The deformation undergone by material elements is tracked in the actual flow from direct CFD simulations. The main goals of this work are to resume the challenge of characterization of the lamellar structure and offer further understanding on the mechanisms for mixing scales reduction in RIM.

This chapter is organized into five sections. In Section 2.2 the mathematical foundations of the kinematics of flow deformation and mixing are described. The 3D computational model of the flow in a RIM machine's mixing chamber and the numerical characterization of its stretching properties are described in Section 2.3. Results are shown and discussed in Section 2.4. Conclusions are presented in Section 2.5.

2.2 Kinematics of Deformation

The stretching properties of the flow were studied in this work from a Lagrangian point of view by tracking the position and deformation of spherical material elements in the flow. As material spheres with initial infinitesimal radius $|\mathrm{d}\mathbf{X}|$ are advected, they rotate and deform continuously into ellipsoids. Figure 2.2 illustrates the deformation of a spherical fluid element along a pathline.

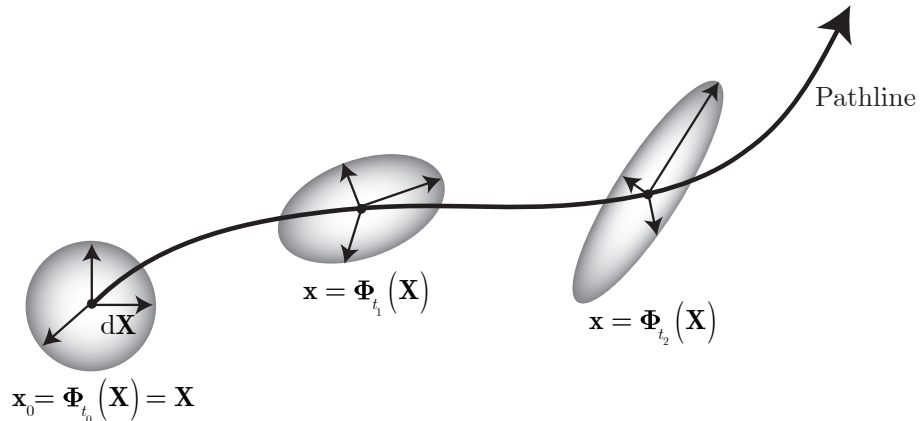


Figure 2.2. Deformation of an infinitesimal spherical element with time.

The mathematical description of the elements' motion can be seen, in the language of dynamical systems, as a *mapping* or a *point transformation* that returns the position, $\mathbf{x}$, of a particle $\mathbf{X}$, i.e., the particle initially at position $\mathbf{X}$, at a given time,

$$\mathbf{x} = \Phi_t(\mathbf{X}) \text{ with } \mathbf{x}_0 = \Phi_{t_0}(\mathbf{X}). \quad (2.6)$$

The function Φ_t is called the *flow* or *motion*. Equation (2.6) results from the integration of the advection equation for each element

$$\frac{d\mathbf{x}}{dt} = \mathbf{u}(\mathbf{x}, t) \quad (2.7)$$

from the initial position $\mathbf{x}_0 = \mathbf{X}$, where $\mathbf{u}$ is the velocity vector and t the flow time.

The distance between the elements centroid and any point on its surface at a given instant is related to the initial radius by

$$d\mathbf{x}(\mathbf{X}, t) = \mathbf{F}(\mathbf{X}, t) \cdot d\mathbf{X} \quad (2.8)$$

where $\mathbf{F}$ is the deformation gradient tensor, and can be obtained directly from the flow as

$$\mathbf{F} = \left(\nabla_{\mathbf{x}} \Phi_t(\mathbf{X}) \right)^T \quad (2.9)$$

where $\nabla_{\mathbf{x}}(\cdot) = \partial_{x_i}(\cdot) \hat{\mathbf{e}}_i$ represents differentiation with respect to $\mathbf{X}$. Even for simple systems, $\Phi_t(\mathbf{X})$ can be difficult or impossible to obtain analytically, but $\mathbf{F}$ can be obtained numerically from the Eulerian velocity field as

$$\frac{d\mathbf{F}}{dt} = \left(\nabla \mathbf{u} \right)^T \cdot \mathbf{F}, \quad (2.10)$$

with $\mathbf{F}(t_0) = \mathbf{I}$ as the initial condition (Ottino, 1990) where $\mathbf{I}$ represents the identity tensor. Locally, the deformation of the ellipsoid surface caused by $\mathbf{F}$ can be

decomposed, using the tensor polar decomposition theorem, into the product of a positive symmetric tensor, $\mathbf{U}$, and an orthogonal tensor, $\mathbf{R}$, (Han, 2007)

$$\mathbf{F}(t) = \mathbf{R}(t) \cdot \mathbf{U}(t). \quad (2.11)$$

Geometrically, the decomposition of the deformation transformation of the element can be perceived as a stretching transformation $\mathbf{U}$ followed by a pure rotation transformation $\mathbf{R}$ (Figure 2.3). Since pure rotation does not produce deformation of the material elements, applying a transformation $\mathbf{R}$ to an element followed by the opposite transformation $\mathbf{R}^T$, i.e., rotation in the opposite direction, leads to no change. Using the orthogonal property of the tensor $\mathbf{R}$,

$$\mathbf{R} \cdot \mathbf{R}^T = \mathbf{R}^T \cdot \mathbf{R} = \mathbf{I}, \quad (2.12)$$

an independent operation of rotation, will be obtained multiplying $\mathbf{F}$ by its transpose,

$$\mathbf{F}^T(t) \cdot \mathbf{F}(t) = \mathbf{U}(t)^2 = \mathbf{C}(t) \quad (2.13)$$

where $\mathbf{C}$ is called the right Cauchy-Green deformation tensor and returns the square of local change in distances from the points $\mathbf{x}$ to $\mathbf{x} + d\mathbf{x}$ due to deformation. If a new coordinate system is chosen to be coincident with the principal axes of deformation, the stretching/compression of the material element in the i -th principal direction of deformation, λ_i , can then be obtained from the eigenvalues of $\mathbf{C}$,

$$\mathbf{C}(t) \cdot d\mathbf{X}_{(i)} = \lambda_i(t)^2 \mathbf{I} \cdot d\mathbf{X}_{(i)} \quad (2.14)$$

where $d\mathbf{X}_{(i)}$ represents the direction vector of the i -th principal axis and λ_i is given by

$$\lambda_i = \frac{|d\mathbf{x}_{(i)}|}{|d\mathbf{X}_{(i)}|}. \quad (2.15)$$

A system has as many principal directions as dimensions and an axis is being stretched if $\lambda_i > 1$ or compressed if $0 < \lambda_i < 1$.

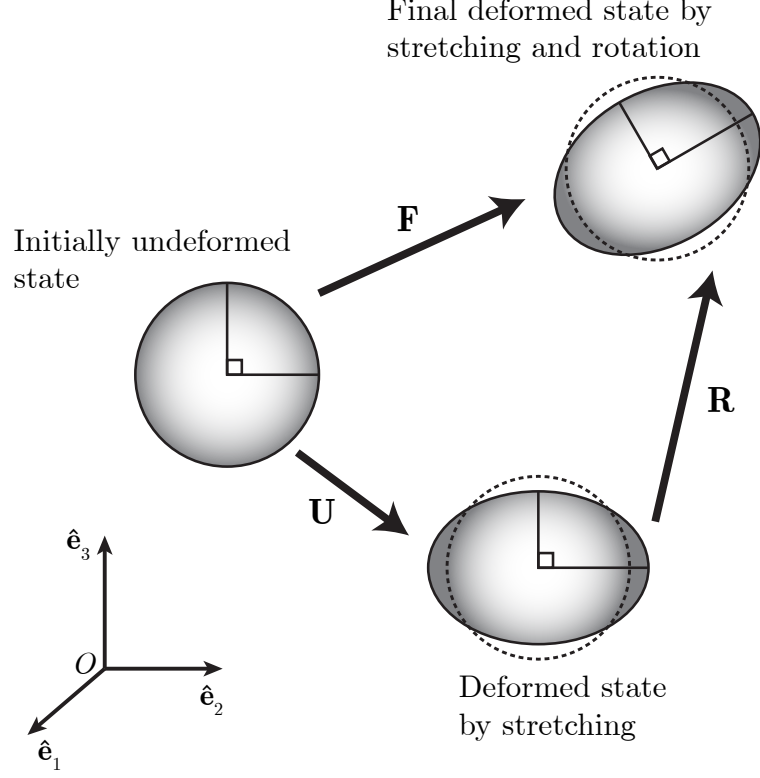


Figure 2.3. Geometrical interpretation of the deformation gradient tensor polar decomposition.

The deformation of materials in the flow by increasing interfacial area and by thinning striation thicknesses accelerates interdiffusion of components leading to mixture at a molecular level. The distance between striations is inversely proportional to the surface area: the thinner the striations, the faster the diffusion (Ottino and Wiggins, 2004). For that reason, the stretching, $\lambda = \max(\lambda_i)$, is introduced here as a measure of mixing.

The stretching and the stretching rate, $\dot{\lambda}$, are related in a fundamental way to the dynamical systems concept of *Lyapunov exponents*. The system's Lyapunov exponents, Λ_i , are defined as

$$\Lambda_i(\mathbf{X}) = \lim_{t \rightarrow \infty} \frac{1}{t} \int_0^t \frac{\dot{\lambda}_i}{\lambda_i}(\mathbf{X}, t') dt' = \lim_{t \rightarrow \infty} \frac{1}{t} \ln \lambda_i(\mathbf{X}, t) \quad (2.16)$$

and can be seen as a long time average of the specific rate of stretching/compression, $\dot{\lambda}_i/\lambda_i$ (Ottino, 1990). For a system to be chaotic, it must have at least one positive Lyapunov exponent $\Lambda = \max(\Lambda_i) > 0$. In a chaotic flow, an advected material element will stretch exponentially with time as

$$|\mathbf{dx}| \approx |\mathbf{dX}| e^{\Lambda t}. \quad (2.17)$$

In *pure shear flow*, an element will grow linearly with time ($\lambda \sim t$) as the axis of stretching gets aligned with the streamlines. Better mixing efficiency can be obtained by random reorientation of the material elements relative to the flow streamlines. The effect of the reorientation will result on an exponential stretching of material elements (Ottino and Wiggins, 2004).

2.3 Stretching Histories from CFD Simulations

In this section the strategy for the determination of the striation thickness distribution in the impingement jets flow from a Eulerian-Lagrangian approach is described. The flow is studied for $Re = 100, 110, 200$ and 300 .

First, in Section 2.3.1, a 3-dimensional CFD model of the flow in a confined impinging jet mixer of a RIM machine with typical dimensions is presented. In Section 2.3.2, the algorithm for the tracking of the position and deformation of passive material elements in the flow from the CFD simulations is presented with the objective to determine the flow stretching distribution. A methodology to estimate the distribution of striation thickness from the stretching distribution function is described in Section 2.3.3.

2.3.1 3D CFD Dynamic Simulations

2.3.1.1 Physical Domain

For the flow simulations a 3D geometrical domain was considered. The domain consists of a cylindrical chamber with typical dimensions of a confined impinging jets

reactor in RIM with a diameter $D = 10$ mm and a height $H = 50$ mm. Cylindrical injectors are placed normally to the cylindrical chamber at 5 mm from the top, making an angle of 180° between each other. The injectors have a diameter $d = 1.5$ mm and a height $l = 2.5$ mm. The mixing chamber was divided in half by a plane normal to the injection direction. The plane has the possibility to be *activated* and *deactivated* for the reasons explained below in Section 2.3.1.3. Figure 2.4 shows a schematic representation of the geometrical domain and its dimensions.

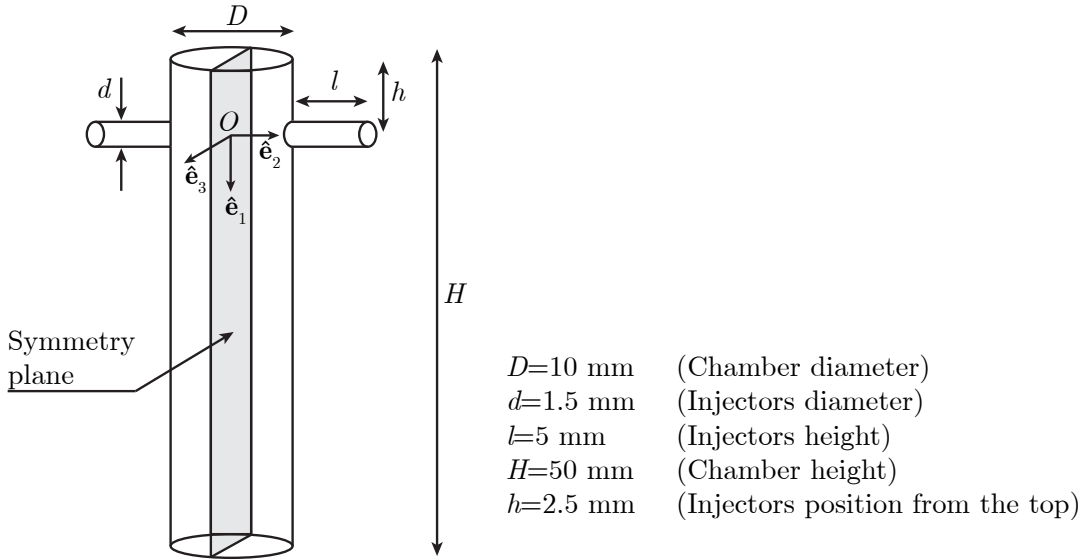


Figure 2.4. Schematic representation and dimensions of the simulation domain.

2.3.1.2 Governing Equations and Boundary Conditions

The flow of an incompressible fluid inside the RIM machine's mixing chamber was simulated by solving the transient Navier-Stokes equations, for mass conservation

$$\nabla \cdot \mathbf{u} = 0, \quad (2.18)$$

and for momentum conservation

$$\rho \left(\partial_t \mathbf{u} + \mathbf{u} \cdot \nabla \mathbf{u} \right) = -\nabla p + \mu \nabla^2 \mathbf{u}. \quad (2.19)$$

The conservation equations are to be solved for the following boundary conditions. The no-slip condition was assumed at the chamber walls, $\mathbf{u} = \mathbf{0}$. A symmetry or shear-free condition at the inside symmetry plane, when enabled, setting $u_2 = 0$ and

$\partial_{x_2} u_1 = \partial_{x_2} u_3 = 0$. This symmetry condition was used to force an initial symmetric solution of the flow in steady state conditions, which is difficult to obtain at higher Reynolds numbers as previously observed by Santos et al. (2010). A fully developed parabolic velocity profile was set at the inlets. A uniform and constant reference pressure was set at the outlet, $p = p_0$, equal to the atmospheric pressure. A density of $\rho = 10^3 \text{ kg/m}^3$ and a viscosity of $\mu = 2 \times 10^{-2} \text{ Pa} \cdot \text{s}$ were assumed for the working fluid.

2.3.1.3 Domain Discretization

The simulation domain was discretized with 1.3 million uniformly sized hexahedral finite volumes with maximum edge length of $\Delta x = 150 \text{ } \mu\text{m}$. Adjustments to the elements shape were done automatically by the mesh generation software to accommodate the hexahedra to the curved surfaces of the mixing chamber, ensuring the maximum edge length of $150 \text{ } \mu\text{m}$. The geometrical domain and simulation grid for the flow simulations were generated with the software packages *DesignModeler* and *Meshing* included in Ansys® 14 suite.

Despite of the flow being laminar for the studied range of Re, the grid element size was chosen accordingly to the Kolmogorov scale, the smallest structure possible in a turbulent flow, to ensure that all the hydrodynamic length scales were being correctly simulated without the need for sub-grid turbulence modeling. This approach for turbulent flows simulation where the Navier-Stokes equations are solved directly is known as Direct Numerical Simulation (DNS). The Kolmogorov scale, λ_K , in isotropic and homogeneous turbulent flows can be estimated from

$$\lambda_K = \left(\frac{\nu^3}{\varepsilon} \right)^{\frac{1}{4}} \quad (2.20)$$

where ν is the kinematic viscosity and ε is the turbulent energy dissipation rate of the flow into heat and can be estimated from a balance to the kinetic energy rate

over the simulation domain boundary surface, $\partial\Omega$, neglecting the contribution of friction at the walls. With this assumption, the energy dissipation rate is given by

$$\varepsilon \approx \frac{\iint_{\partial\Omega} \frac{|\mathbf{u}|^2}{2} \rho (\mathbf{u} \cdot \mathbf{n}) dS}{\rho V_{\text{dissipation}}} \quad (2.21)$$

where $V_{\text{dissipation}} = \left(\frac{\pi}{4} D^2 \right) (nD)$ is the volume of the cylindrical mixing chamber where dynamic impingement mixing occurs and nD , the distance from the top of the mixing chamber until the point where dynamic mixing still occurs, has been observed to be $nD \approx 2D$ to $3D$ (Kolodziej et al., 1982). The vector $\mathbf{n}$ represents here the normal unitary vector to the outside normal unitary vector to the boundaries of the domain.

Since the outlet velocity is about 5% of the inlet velocity due to the difference of flow areas, the kinetic energy rate of the exit fluid was also neglected. Assuming fully developed parabolic velocity profiles at the inlets, Equation (2.21) results in

$$\varepsilon \approx \frac{\int_0^{d_1/2} \left(2u_{\text{inj},1} \frac{(d_1/2)^2 - r^2}{(d_1/2)^2} \right)^3 \rho_1 \pi r dr + \int_0^{d_2/2} \left(2u_{\text{inj},2} \frac{(d_2/2)^2 - r^2}{(d_2/2)^2} \right)^3 \rho_2 \pi r dr}{\bar{\rho} V_{\text{dissipation}}} \quad (2.22)$$

where the indices 1 and 2 represent the left and right injector, respectively, $r \in [0, d/2]$ is the radial position at the injectors' circular section area and $\bar{\rho}$ is the average density weighted by the volumetric flow rate, Q , as

$$\bar{\rho} = \frac{Q_1 \rho_1 + Q_2 \rho_2}{Q_1 + Q_2}. \quad (2.23)$$

Assuming equal density and viscosity at both injectors, the condition of this computational study, the Kolmogorov scale of the flow can be estimated from

$$\lambda_K = \left(\frac{D^3 n d}{1 + 1/\phi_K} \right)^{\frac{1}{4}} \text{Re}^{-\frac{3}{4}} \quad (2.24)$$

where the feeding kinetic energy rate ratio is given by

$$\phi_K = \frac{\rho_1 d_1^2 u_{\text{inj},1}^3}{\rho_2 d_2^2 u_{\text{inj},2}^3}. \quad (2.25)$$

For the geometry in this work and the highest value of Reynolds studied, assuming $\phi_K = 1$, the Kolmogorov scale of the flow, can be estimated from Equation (2.24) with values between $120 \mu\text{m}$ and $150 \mu\text{m}$, for $n = 2$ and $n = 3$, respectively.

This estimated flow scale range shows that, even if one were dealing with a turbulent flow, all the hydrodynamic flow scales are correctly simulated with the selected grid spacing, without the need for further refinement or turbulence models. According to Libby (1996), acceptable accuracy in DNS is obtained if grid spacing is between four to six times greater than the Kolmogorov scale. For the present work the grid scale is of the same order of λ_K .

2.3.1.4 Numerical Solution at Steady State

The flow equations were solved with the finite-volume commercial CFD solver ANSYS Fluent® 14 in an Intel® Xeon® 3 GHz 8-core PC with Windows 7 64-bits.

The CFD simulations were initially performed at steady state, setting the transient term of Equation (2.19) equal to zero. For the steady state simulations the symmetry plane (Figure 2.4) was enabled in the chamber. This procedure was used to ensure a symmetric initial state for transient simulations of the flow. The advective terms of Equations (2.18) and (2.19) were discretized with the *Third-Order MUSCL* scheme for momentum and the *Standard* scheme for pressure. The pressure-velocity coupling scheme was set to *PISO* and gradients were computed with the *Least Squares Cell*

Based scheme. All flow variables were initialized with a value equal to zero and steady-state symmetric solutions were accepted for residues lower than 10^{-6} .

2.3.1.5 Numerical Solution at Transient State

The dynamic flow simulations were started from the symmetric steady-state simulations by removing the symmetry plane boundary condition and imposing a non-symmetric finite perturbation. The perturbation was set by varying the velocity of the left injector with time through a Fluent User Defined Function (UDF). The boundary condition at the left inlet was replaced by

$$\mathbf{u} = -2u_{inj} \frac{\left(\frac{d}{2}\right)^2 - r^2}{\left(\frac{d}{2}\right)^2} f(t/\tau) \mathbf{n} \quad (2.26)$$

where f is the perturbation time function affecting the left inlet velocity and τ the chamber mean residence time defined as

$$\tau = \frac{D^2 H}{2u_{inj} d^2}. \quad (2.27)$$

This methodology has been previously used by Teixeira (2000) and Santos (2003), and care was taken not to induce any discontinuity in the flow from the perturbation. For that, a smooth time function with a continuous first derivative was chosen,

$$f(t/\tau) = 1 + \frac{a}{2} \left(1 - H(t/\tau - b)\right) \left(1 - \cos\left(\frac{2\pi}{b} t/\tau\right)\right) \quad (2.28)$$

where $H(t)$ is the Heaviside function. The parameter a is the maximum amplitude of the perturbation, which was set to $a = 0.1$. The parameter b is the time relative to chamber mean residence time over which the perturbation is applied, chosen to be $b = 0.1$. Figure 2.5 shows a plot of the evolution of the perturbation time function in time.

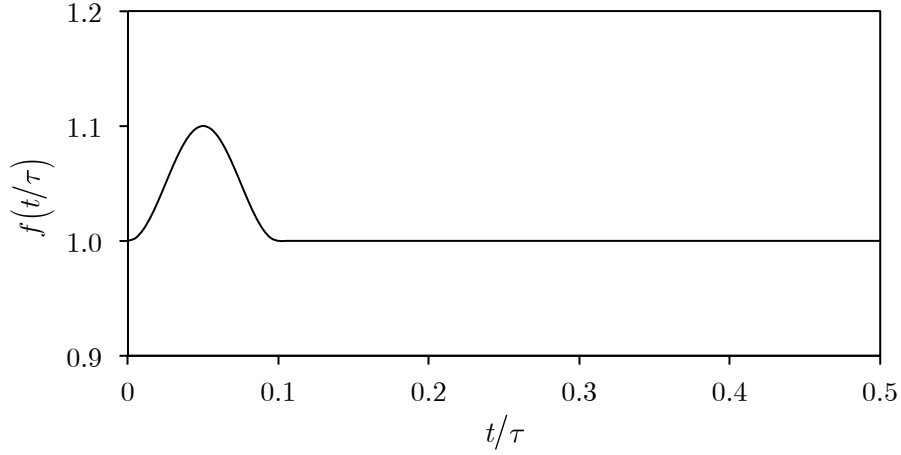


Figure 2.5. Perturbation function applied to the left injector velocity.

The perturbation has a limited impact in time and small amplitude (10% of the inlet velocity). After a time equal to 0.1τ no further perturbation was imposed to the system's variables and the boundary conditions remained constant in time. The influence of the perturbation parameters on the flow dynamics was evaluated by Santos et al. (2010) who have shown that the perturbation has no impact on the flow dynamics after the transient period where the system evolves from the initial state. Before the particle tracking step, transient simulations were done for a flow time equal to 3τ to ensure that the initial perturbation no longer had an impact in the flow.

The transient term of Equation (2.19) was discretized with a *Second Order Implicit* formulation and the time step between iterations was chosen according to

$$\Delta t = \frac{\Delta x}{2u_{\text{inj}}}, \quad (2.29)$$

corresponding to a Courant number, C , equal to 1. The Courant number is here defined as the ratio of the time step to the characteristics advection time at the inlet, where the velocity is higher, $\Delta x/u_{\text{inj}}$ (Ferziger and Peric, 2002)

$$C = \frac{u_{\text{inj}} \Delta t}{\Delta x} \quad (2.30)$$

For the numerical resolution of partial differential equations in time dependent problems where the advective terms are dominant, $C \leq 1$ ensures stability in the solutions. The system state for $t = 3\tau$ was assumed as the starting point for the particles' tracking. The methodologies used in the tracking step are described in Section 2.3.2.

2.3.2 Particle Tracking and Stretching Histories

Particles were tracked numerically in the transient impinging jets flow after a time equal to $t = 3\tau$ after the perturbation by integration of the advection (Equation (2.7)) up to the time when the first injected particle leaves the chamber. The tracked particles were considered to be passive, having the same physical properties as the fluid, thus, not perturbing the flow. A cluster of 172 particles was uniformly injected at each mixing chamber inlet surface. From this point forth, for simplicity, the injection time will be considered as $t = 0\tau$ for the results that concern the Lagrangian analysis.

Each particle position was calculated simultaneously with the flow simulation in ANSYS Fluent® 14 by integrating the advection equation (Equation (2.7)) with a sixth order Runge-Kutta scheme. The particles' position, velocity components and velocity gradient were exported from Ansys Fluent® and stored in ASCII files for each time step. Afterwards, results were imported and processed with MATLAB® 2011b for Mac OS X. Since $\nabla \mathbf{u}$ could not be evaluated at any point of the domain from the stored particle data but only at the particles' position, the integration of Equation (2.10) to obtain the deformation gradient tensor was performed with the first order Euler method. The use of a higher order integration scheme was not considered imperative since the time step was small, $\Delta t = \mathcal{O}(10^{-5}\tau)$, for the acquired time series of around 6000 points for each particle. The stretching experienced by each particle with time was calculated from Equations (2.13) and (2.14).

2.3.3 Prediction of Striation Thickness Distribution

The striation thickness distribution in the flow can be predicted from the stretching distribution of the material elements in the mixing chamber. An approach similar to the one of Baldyga and Bourne (1983) was adopted, but while Baldyga and Bourne (1983) assumed an isotropic and homogenous turbulent flow at the top of the mixing chamber, this study is based in the actual stretching distribution in the laminar chaotic flow from the 3D CFD simulations.

Take two randomly oriented passive particles, α and β , placed at the inlet, initially separated by an infinitesimal distance dR . As the particles are transported by the flow, impingement mixing tends on average to separate the two particles by a distance dr . If a fluid element $dX_1 dX_2 dX_3$ is considered (Figure 2.6) with the two points α and β in opposed diagonal vertices, for $t = t_0$ the initial distance between the two particles is given by

$$dR^2 = dX_1^2 + dX_2^2 + dX_3^2. \quad (2.31)$$

If initial dimensions are assumed equal, $s_0 = dX_1 = dX_2 = dX_3$, Equation (2.31) becomes

$$dR = \sqrt{3}s_0. \quad (2.32)$$

As the two particles are transported in the flow, the fluid element will be stretched in the principal direction of deformation (in this case, $\hat{\mathbf{e}}_3$). As schematized in Figure 2.6, for $t > t_0$ the distance between a particular pair of particles α and β will be given by

$$dr^2 = dx_1^2 + dx_2^2 + dx_3^2. \quad (2.33)$$

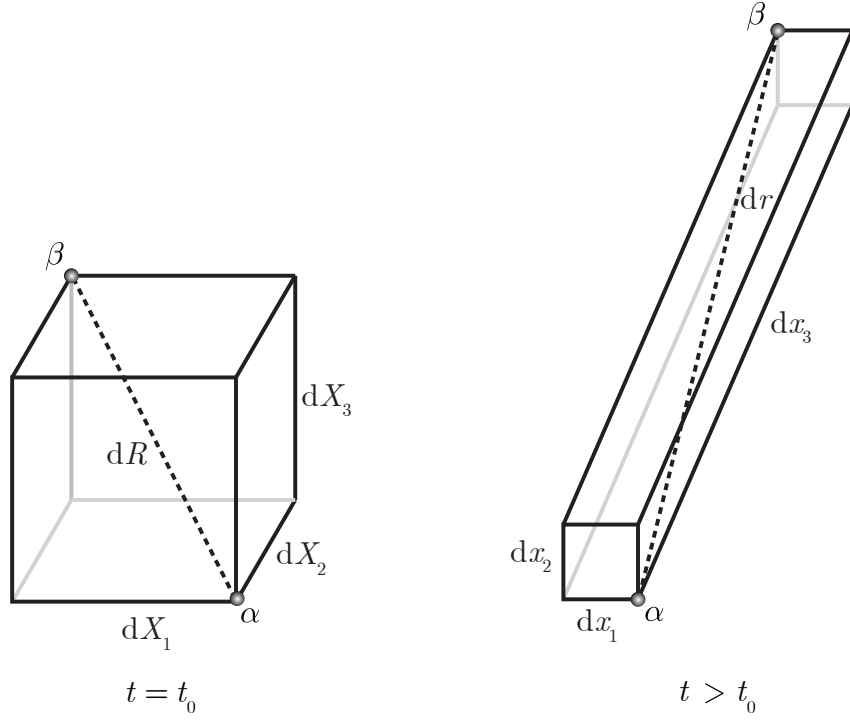


Figure 2.6. Separation of two particles in the flow as a result of fluid deformation.

The fluid element will be continuously stretched by the flow and for $t \gg t_0$ it will no longer resemble a prism but instead it will resemble a thin filament. In that moment, since $dx_3 \gg s_0 > (dx_1, dx_2)$, it can be assumed that

$$dr^2 \approx dx_3^2. \quad (2.34)$$

For an incompressible fluid flow, $\nabla \cdot \mathbf{u} = 0$, the element's volume at the initial instant and for long deformation times are related by

$$dx_1 dx_2 dx_3 = s^2 dx_3 = s_0^3 \quad (2.35)$$

where dx_1 and dx_2 were replaced by the fluid filament thickness, or striation thickness, s . The evolution of the distance of between the two particles will be given by the stretching function, λ , of the flow as

$$dr(t) \approx \lambda(t) dR = \lambda(t) \sqrt{3} s_0 \quad (2.36)$$

by Equation (2.32). Replacing Equations (2.34) and (2.36) in Equation (2.35) gives

$$\sqrt{3}\lambda s_0 s^2 = s_0^3. \quad (2.37)$$

The striation thickness for a particular particle can then be estimated from

$$s(\mathbf{X}, t) = \left(\sqrt{3}\lambda(\mathbf{X}, t) \right)^{-\frac{1}{2}} s_0 \quad (2.38)$$

were the initial striation thickness s_0 is close to the injectors' diameter,

$$s_0 \approx d. \quad (2.39)$$

In the present work, s_0 was considered equal to d .

Knowing the stretching distribution in the flow, $\lambda(\mathbf{X}, t)$, from the CFD simulations, as described in Section 2.3.2, a striation thickness distribution can be estimated with the methodology described in this section.

It should be pointed out that this methodology allows calculating an upper limit for the mixing length scales in the flow since further reduction of laminae from the effect of diffusion and reaction is not included in the calculation. Striation thicknesses are allowed to decrease down to any value, assuming the fluid as a *continuum* at all scales.

2.4 Results and Discussion

2.4.1 Flow Field

The flow field at equilibrium for $t = 0\tau$ and for a time equal to $t = 3\tau$ after the perturbation in the inlet velocity are compared here, specifically the strain rate field and streamlines. The comparison of the flows at these two instants aims to show its evolution after the perturbation for different Reynolds numbers.

Instantaneous strain rate fields do not directly offer the information about the distribution of striation scales observed in the mixture of the two feeding streams.

Furthermore, striation scales also depend on the Lagrangian trajectories in the flow. However, the instantaneous strain rate maps are presented here as indicative of the spatial distribution of the stretching potential in the flow. A non-dimensional rate of strain, ζ , was defined, as

$$\zeta = \frac{1}{2} \frac{(\mathbf{D} : \mathbf{D})^{1/2}}{\dot{\gamma}_{\text{injector}}}. \quad (2.40)$$

where $1/2(\mathbf{D} : \mathbf{D})^{1/2}$ is the norm of the rate of strain tensor and $\dot{\gamma}_{\text{injector}}$ is the strain rate experienced by the fluid at the injectors' walls and given by

$$\dot{\gamma}_{\text{injector}} = \frac{8\mu}{\rho d^2} \text{Re}. \quad (2.41)$$

Figure 2.7 to Figure 2.10 show the non-dimensional rate of strain field for several Re in the planes $x_1 = 0$ and $x_3 = 0$ at the instants $t = 0\tau$ and $t = 3\tau$. Due to different orders of magnitude of the strain rate in the flow, logarithmic scales were chosen for the maps. From all the simulations it is observed that the rate of strain decays rapidly several orders of magnitude from the inlets until the outlet. Three main regions are identified based on the order of magnitude of ζ :

- $0.1 < \zeta < 1$ in the region of the jets and the manifold resultant of the jets impingement;
- $0.01 < \zeta < 0.1$ in the remaining volume of the top of the chamber until a distance $\sim 2D$ from the top;
- $\zeta < 0.01$ from a distance $\sim 2D$ from the top until the outlet, with higher deformation close to the chamber walls and practically no deformation in the center.

For $Re = 100$ (Figure 2.7) the flow presents the same strain field after 3τ of flow time, apart from an asymmetry observed in the impinging jets' surface. An increase of the strain rate is observed at the top of the mixing chamber as Re is increased for $Re \geq 110$.

Despite of constant boundary conditions after the time limited initial perturbation, it is observed that for $t = 3\tau$ and $Re = 300$ (Figure 2.10), the jets' impingement point position is not in the center of the chamber. This behavior is in agreement with experimental observations in Chapter 4 where for constant inlet conditions an oscillation in time of the impingement point around the center of the chamber is observed.

Streamlines at the two flow instants, $t = 0\tau$ and $t = 3\tau$, are also presented here not as an indication of the material trajectories inside the chamber but to indicate the evolution of the zones in the chamber where dynamic impingement mixing occurs and the zones where the flow becomes parallel. Figure 2.11 to Figure 2.14 show front and top views of the streamlines in the mixing chamber for times $t = 0\tau$ and $t = 3\tau$ as a function of Re .

For all the symmetric steady initial states the two fed streams are segregated. Fluid injected from the right injector remains on the right side of the chamber, and in a similar way, fluid injected from the left injector remains on the left side of the chamber. The only interfacial area generated between the two fluids is equivalent to the area of the symmetry plane. Flow is mainly characterized by the presence of two recirculation vortices at the top of the chamber occupying the volume up to a height of $\sim 2D$ from the top wall, which increases as Re is increased. A zone of parallel flow is observed in the remaining chamber.

For a flow time $t = 3\tau$ after the perturbation, the following flow patterns are observed, depending on the Reynolds number:

- For $\text{Re} = 100$ (Figure 2.11) the system recovers from the initial perturbation for a state close to the symmetric flow state.
- For $\text{Re} = 110$ (Figure 2.12) the flow does not recover from the perturbation and an unsteady state is achieved. The flow is similar to the symmetric state but starts to exhibit some oscillation of the jets impingement surface. Two recirculation vortices at the chamber top are still identifiable. Additional energy dissipation from the oscillations makes the flow to become parallel after a distance from the top chamber wall smaller to the one at $\text{Re} = 100$.
- For $\text{Re} > 110$ (Figure 2.13 and Figure 2.14) the deformation of the jets' impingement surface is increased and complex shaped streamlines are observed at the top of the chamber. Coherent structures of two recirculation vortices are no longer observed in the instantaneous flow. The increase of energy dissipation due to the strong dynamic behavior in the top of the chamber makes the flow to become parallel sooner as Re is increased, i.e. after a smaller distance from the top.

This behavior of the flow agrees with the previously observed by Teixeira (2000) from Laser Doppler Anemometry experiments, where the velocity fluctuations detected signal in the mixing chamber decreases one order of magnitude for distances higher than $\sim 1.5D$ to $2D$ from the top.

As also observed in Chapter 5, $\text{Re} > 110$ is the critical Reynolds number that marks the transition from a laminar steady flow regime to a self-sustained laminar chaotic flow regime.

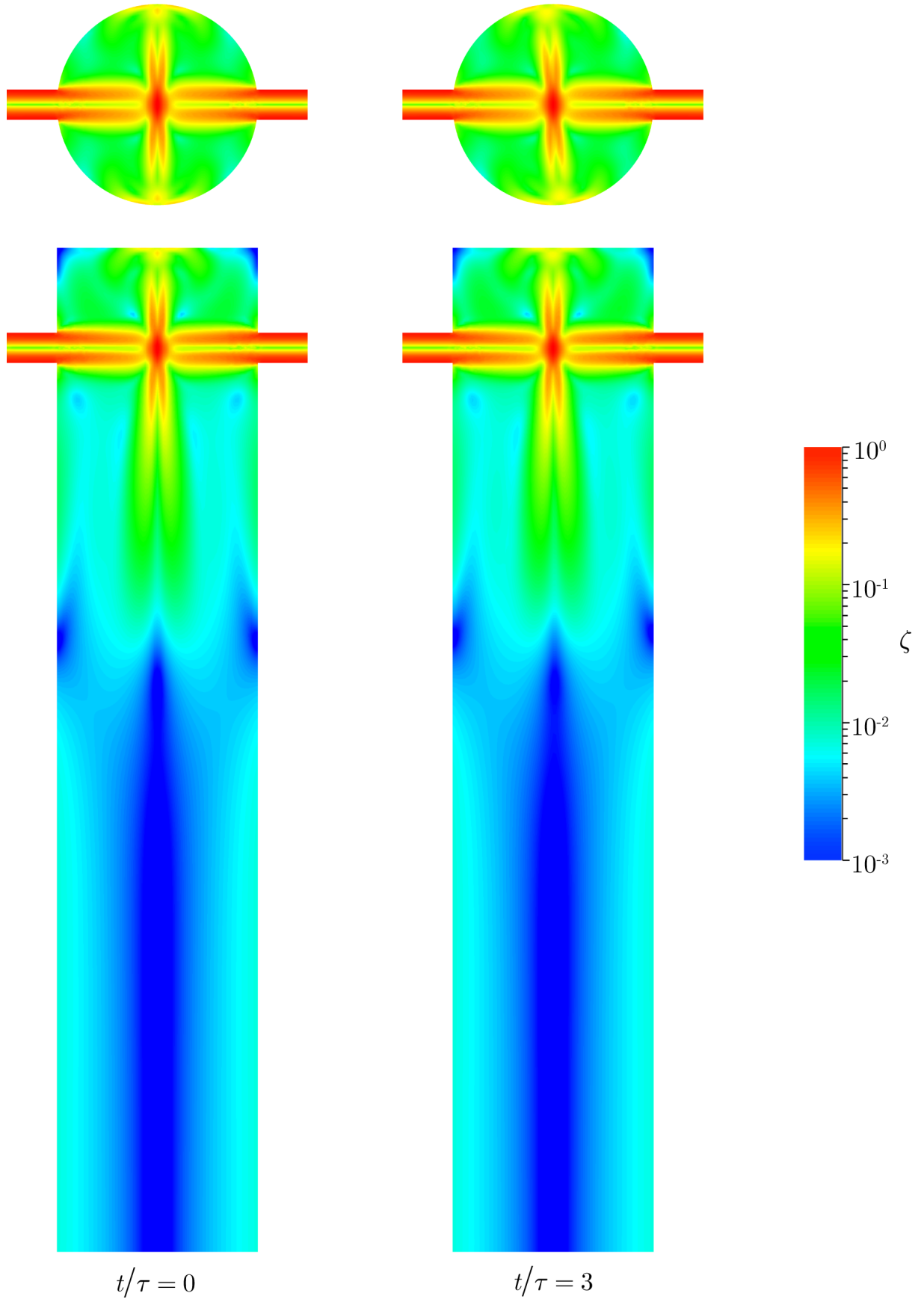


Figure 2.7. Instantaneous non-dimensional strain rate field in plane $x_1=0$ (top) and $x_3=0$ (bottom) for $\text{Re}=100$ from CFD simulations.

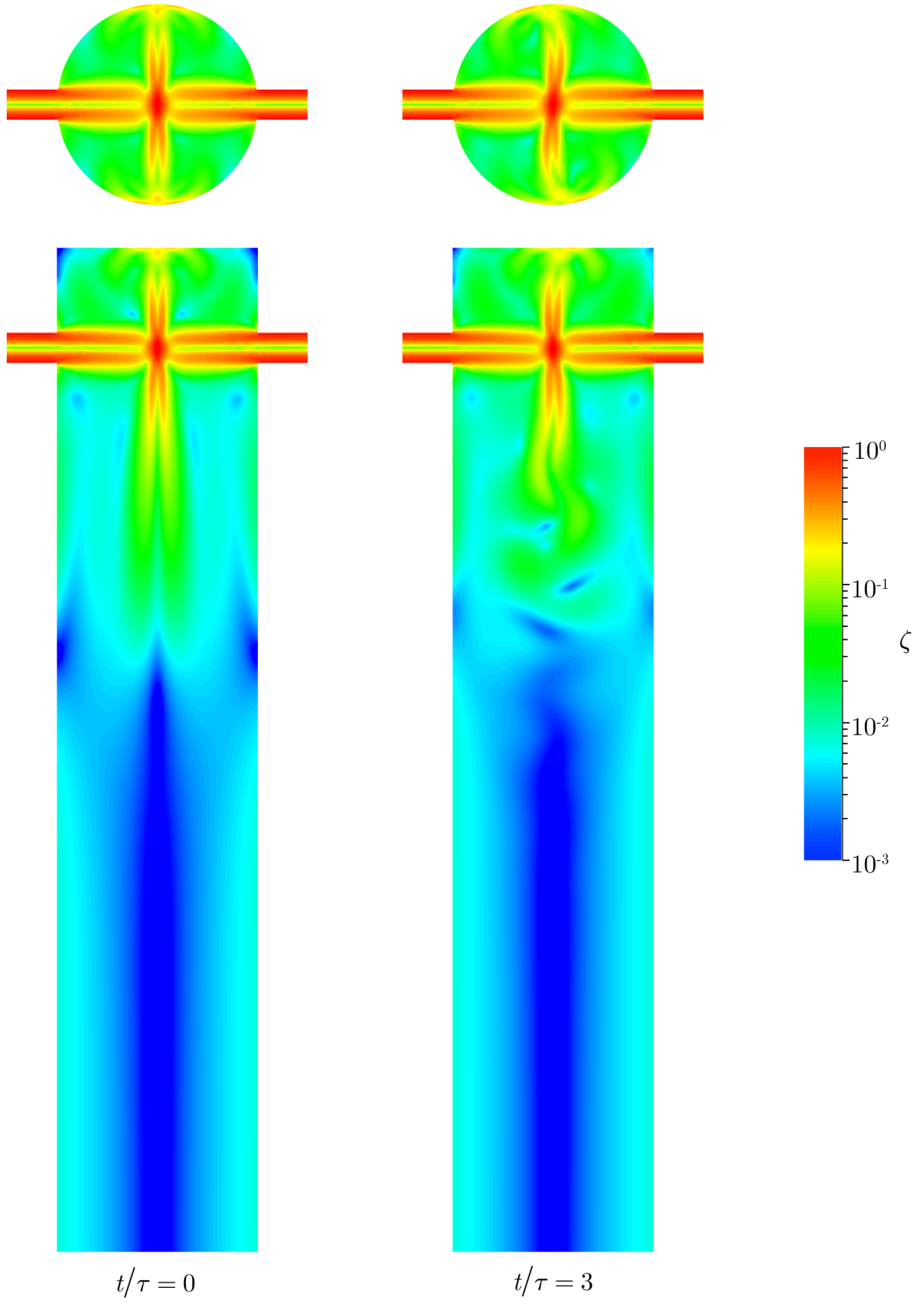


Figure 2.8. Instantaneous non-dimensional strain rate field in plane $x_1=0$ (top) and $x_3=0$ (bottom) for $Re=110$ from CFD simulations.

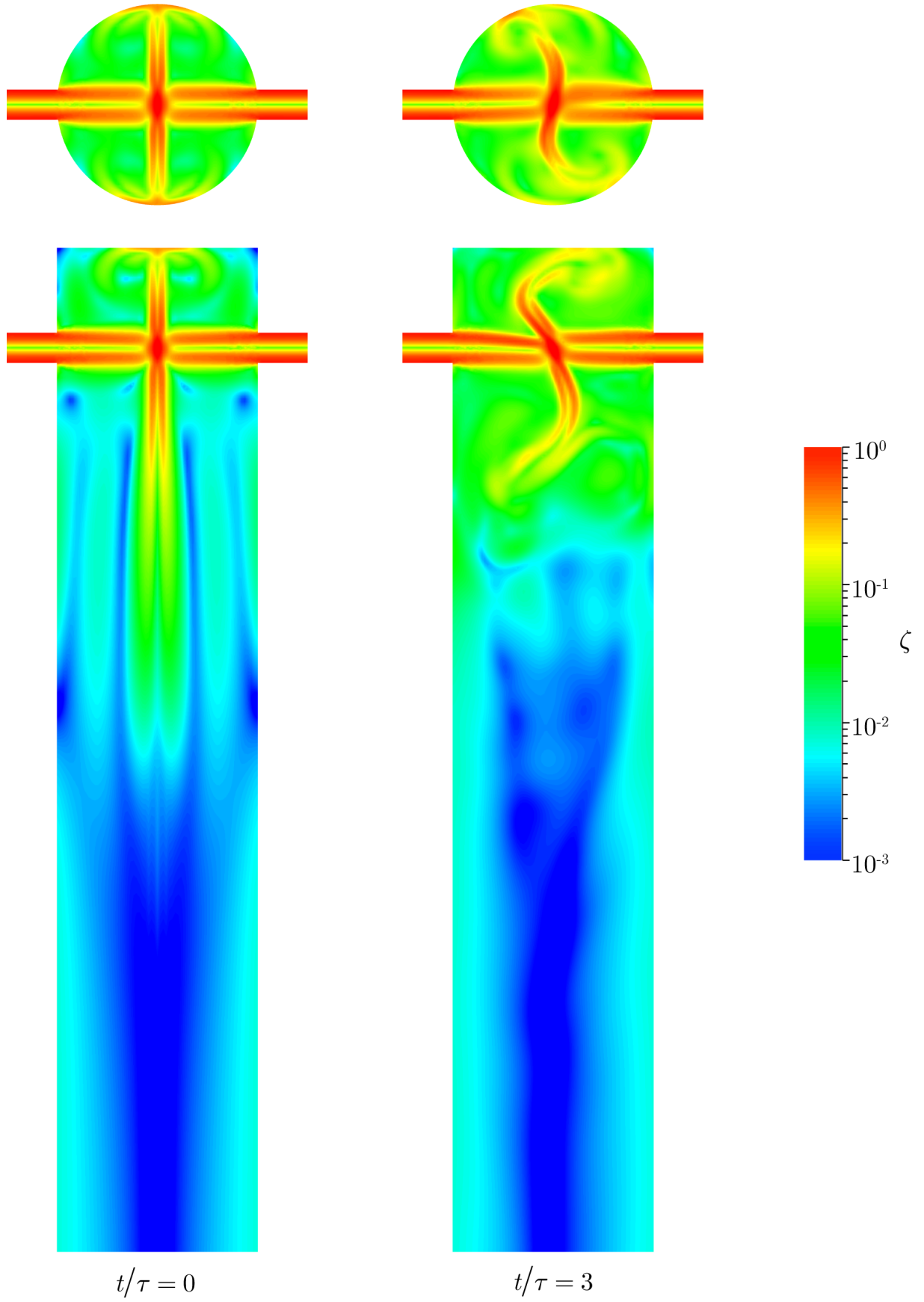


Figure 2.9. Instantaneous non-dimensional strain rate field in plane $x_1=0$ (top) and $x_3=0$ (bottom) for $Re=200$ from CFD simulations.

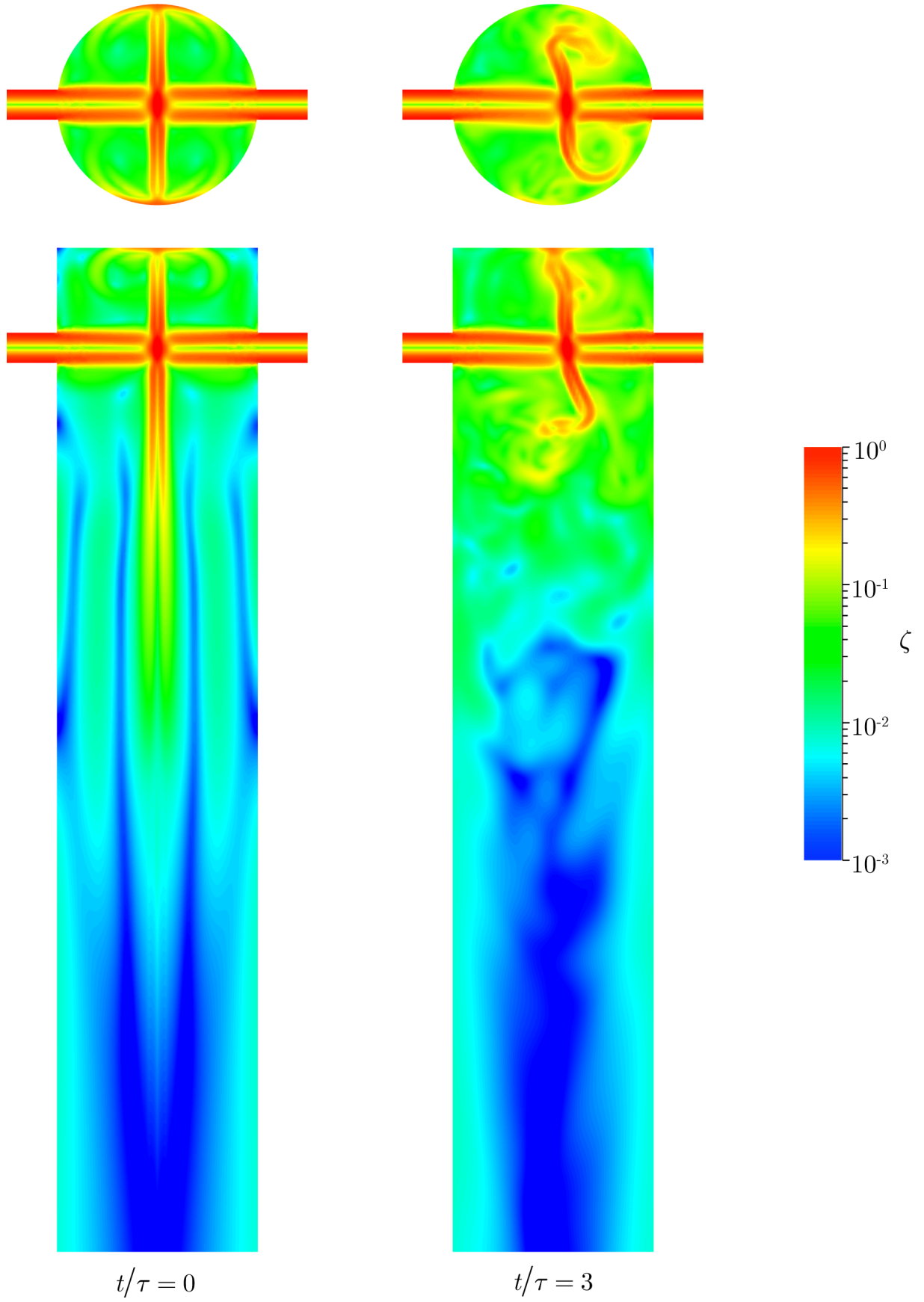


Figure 2.10. Instantaneous non-dimensional strain rate field in plane $x_1=0$ (top) and $x_3=0$ (bottom) for $Re=300$ from CFD simulations.

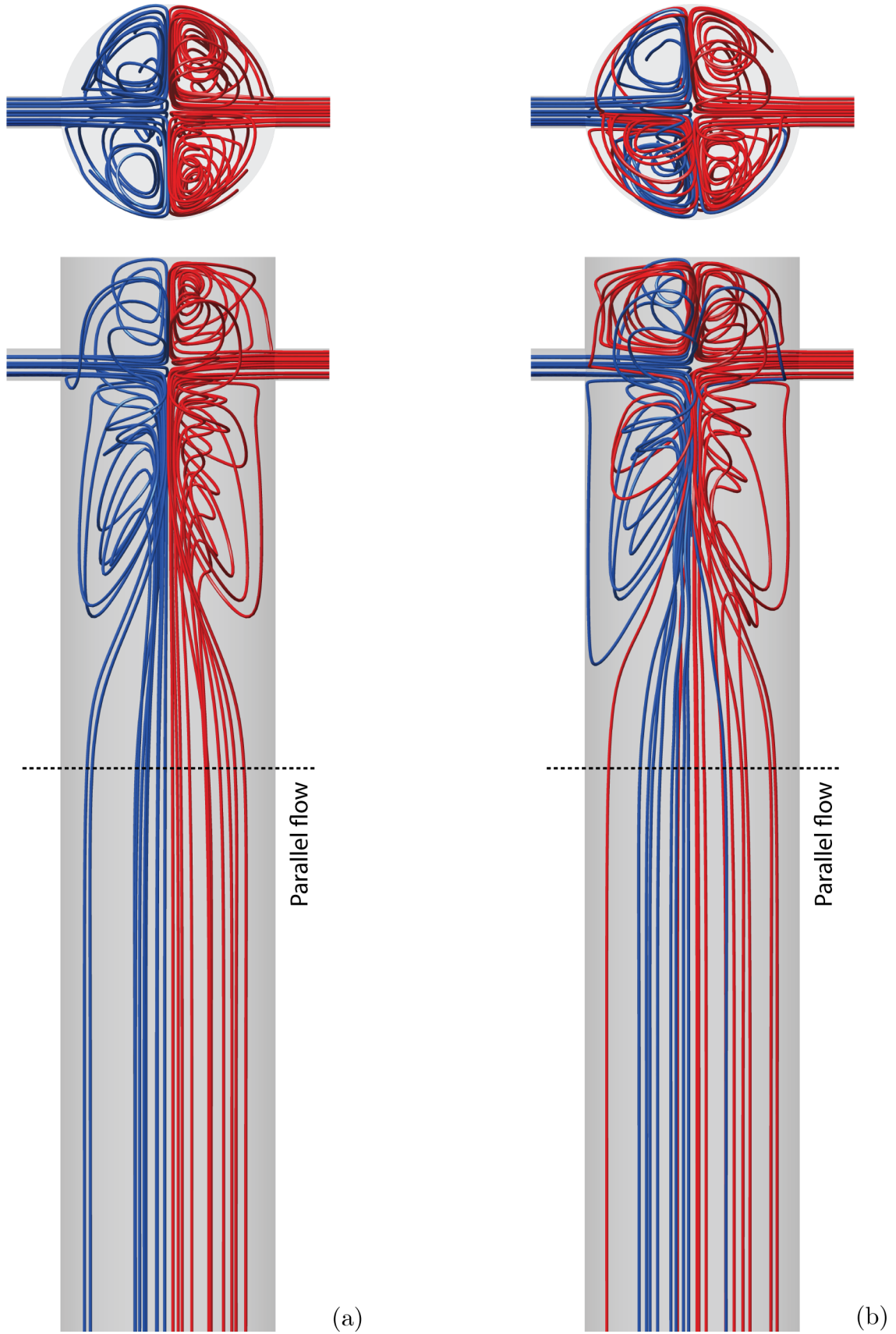


Figure 2.11. Top and side views of the streamlines from the inlets for $Re=100$: a) $t/\tau = 0$; b) $t/\tau = 3$.

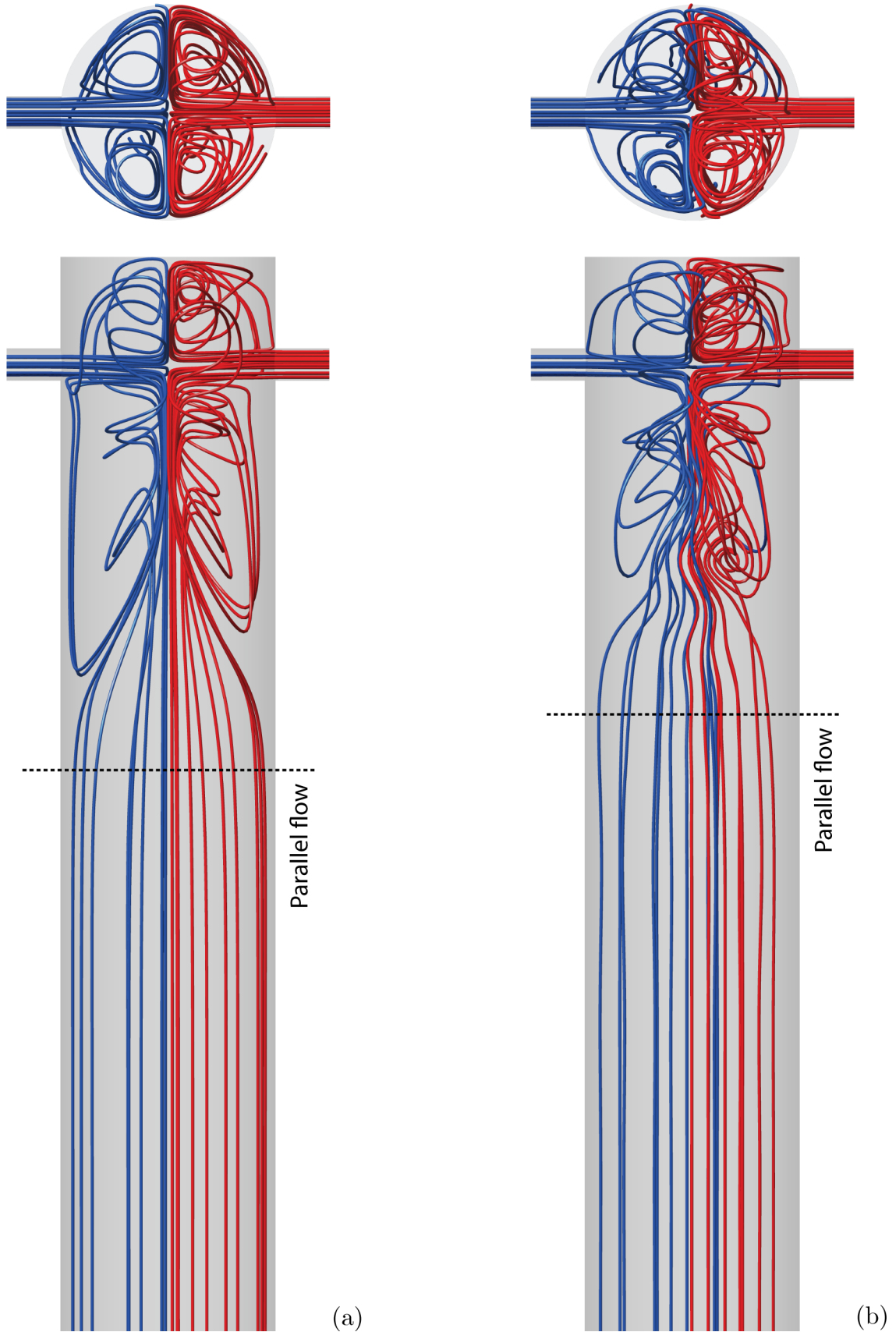


Figure 2.12. Top and side views of the streamlines from the inlets for $Re=110$: a) $t/\tau = 0$; b) $t/\tau = 3$.

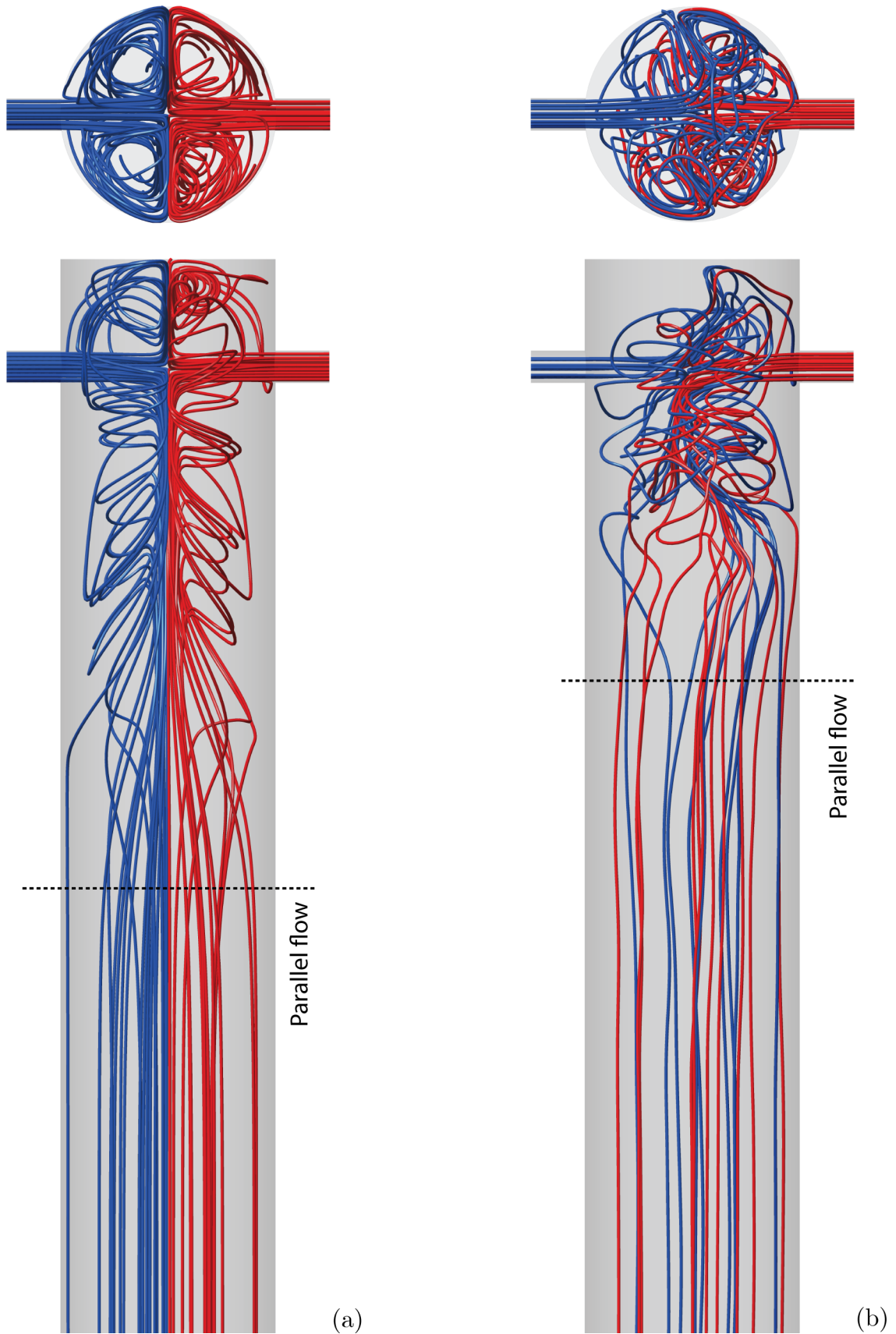


Figure 2.13. Top and side views of the streamlines from the inlets for $Re=200$: a) $t/\tau = 0$; b) $t/\tau = 3$.

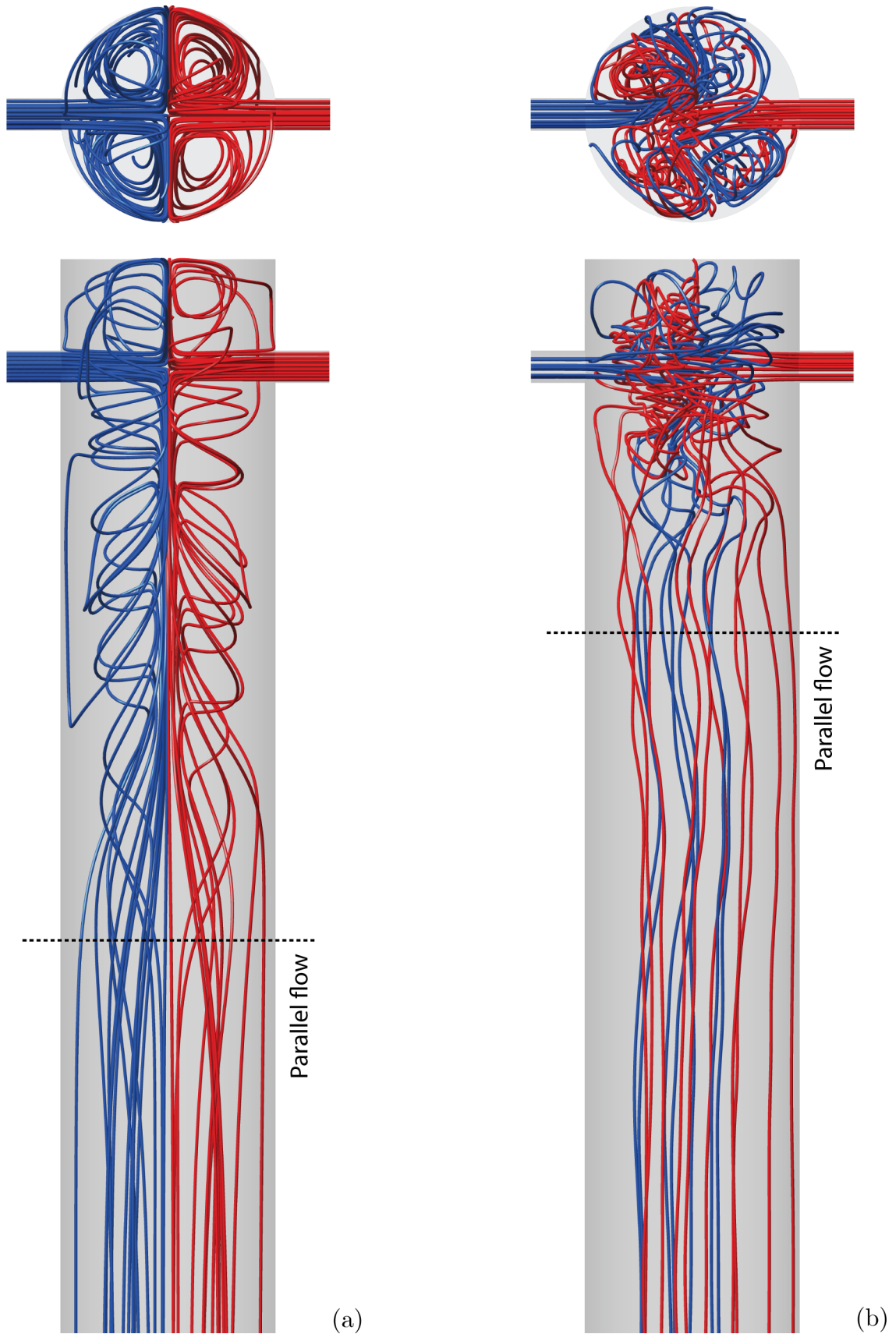


Figure 2.14. Top and side views of the streamlines from the inlets for $Re=300$: a) $t/\tau = 0$; b) $t/\tau = 3$.

2.4.2 Particle Distribution

Passive particles were tracked in the flow for $Re = 100, 110, 200$ e 300 . Results from the particle tracking in the flow are presented in Figure 2.15 to Figure 2.22, composed with the particle distribution inside the mixing chamber at different flow times. Also, for better visualization of the particles' dynamics, animations are included in the DVD that is part of this work. Figure 2.15 to Figure 2.22 show a dynamic sequence of top and front views of the particles being transported in the RIM machine's mixing chamber.

For $Re = 100$ (Figure 2.15 and Figure 2.16) particles enter the chamber and follow the jets until the impingement point. After impingement, the particles spread radially, relatively to the jets axis, at the top of the mixing chamber. Due to the steady flow achieved under these conditions, particles maintain themselves mainly segregated until leaving the chamber, i.e., particles injected from the right injector remain on the right side of the chamber until the outlet, and particles injected from the left injector remain on the left side. Some mixing is achieved because the perturbed flow is no longer symmetric, allowing for some particles to be transferred from one side to the other at the chamber walls. For better visualization, an animation of the particles' trajectories for $Re = 100$ can be found in the DVD (folder “~/Chapter2/Re100”, files “Re100_Front.mov” and “Re100_Top.mov”).

When the flow rate is increased to obtain $Re = 110$ (Figure 2.17 and Figure 2.18), particles enter the chamber and follow the jets until the impingement point, and afterwards they spread radially. As shown in Chapter 5, this value of Re is confirmed experimentally as marking the onset of the dynamic regime. Some oscillation of the impingement surface is observed downstream of the inlets, which improves the particles' distribution in the chamber. Due to the deformation of the two jets' impingement surface, material exchange between the right and left sides of the chamber occurs. An animation of the particles' trajectories for $Re = 110$ can be found in the DVD (folder “~/Chapter2/Re110”, files “Re110_Front.mov” and “Re110_Top.mov”).

For $Re = 200$ (Figure 2.19 and Figure 2.20) and $Re = 300$ (Figure 2.21 and Figure 2.22), particles are also transported by the jets from the inlet until the center of the chamber where they impinge and distribute radially. For these flow conditions, a dynamic state is achieved and the jet's impingement manifold is highly deformed. A homogeneous field of particles is observed at early stages of the particles' trajectories inside the mixing chamber. Animations of the particles' trajectories for $Re = 200$ and 300 can be found in the DVD (folder " $\sim$ /Chapter2/Re200", files "Re200_Front.mov" and "Re200_Top.mov" for $Re = 200$; folder " $\sim$ /Chapter2/Re300", files "Re300_Front.mov" and "Re300_Top.mov" for $Re = 300$).

The results of this section show the importance of achieving a dynamic flow regime in impingement jets mixing. Otherwise, no exchange of material between the two sides of the mixing chamber occurs. The hydrodynamic instability achieved for $Re > 110$ is the mechanism responsible for the spatial distribution of the two fed streams in the chamber.

From the obtained particles trajectories and strain rate experienced by each particle along its trajectory, a stretching rate distribution inside the mixing chamber is obtained in Section 2.4.3.

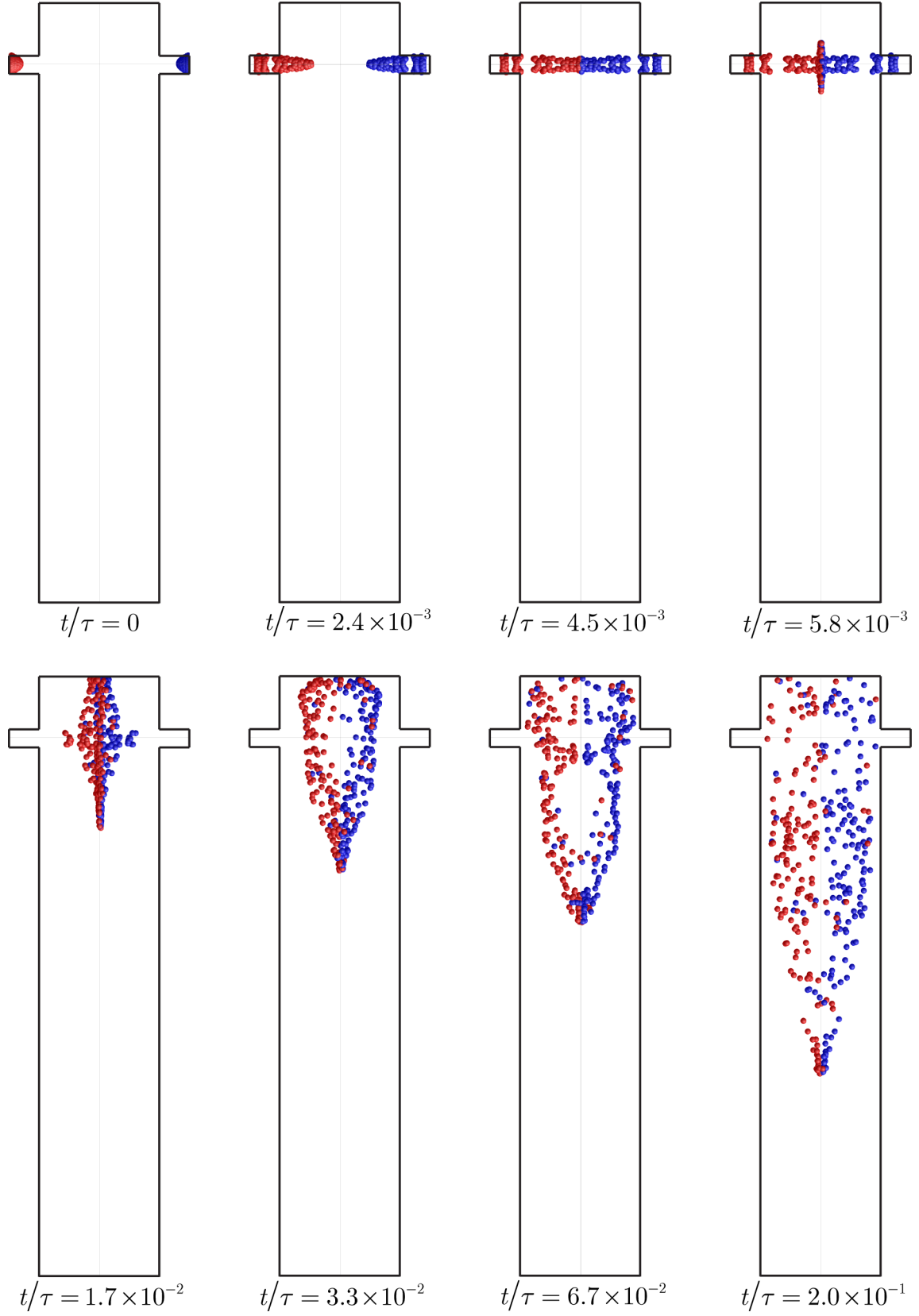


Figure 2.15. Front view of the particles' trajectory for $Re=100$.

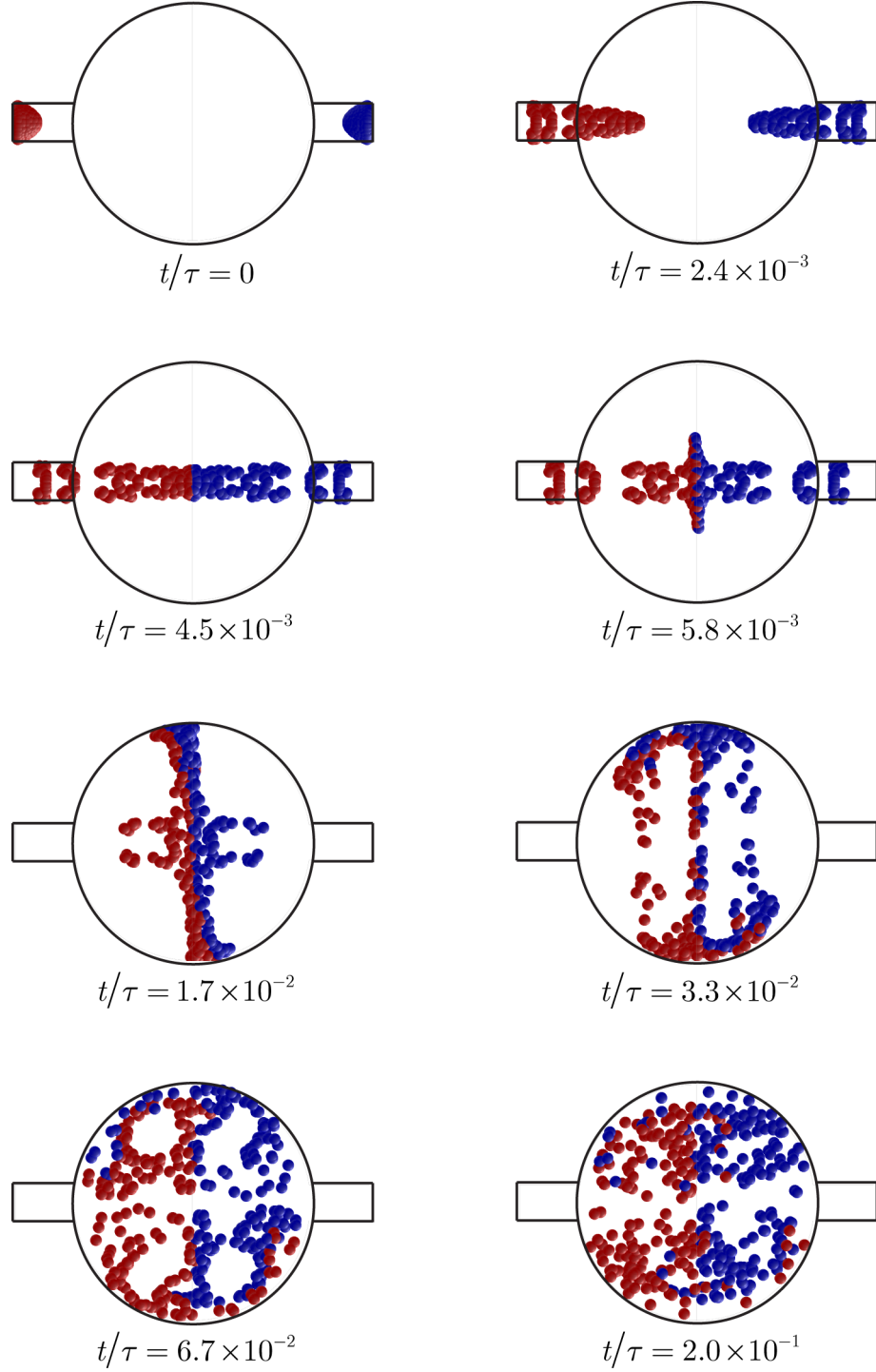


Figure 2.16. Top view of the particles' trajectory for $Re=100$.

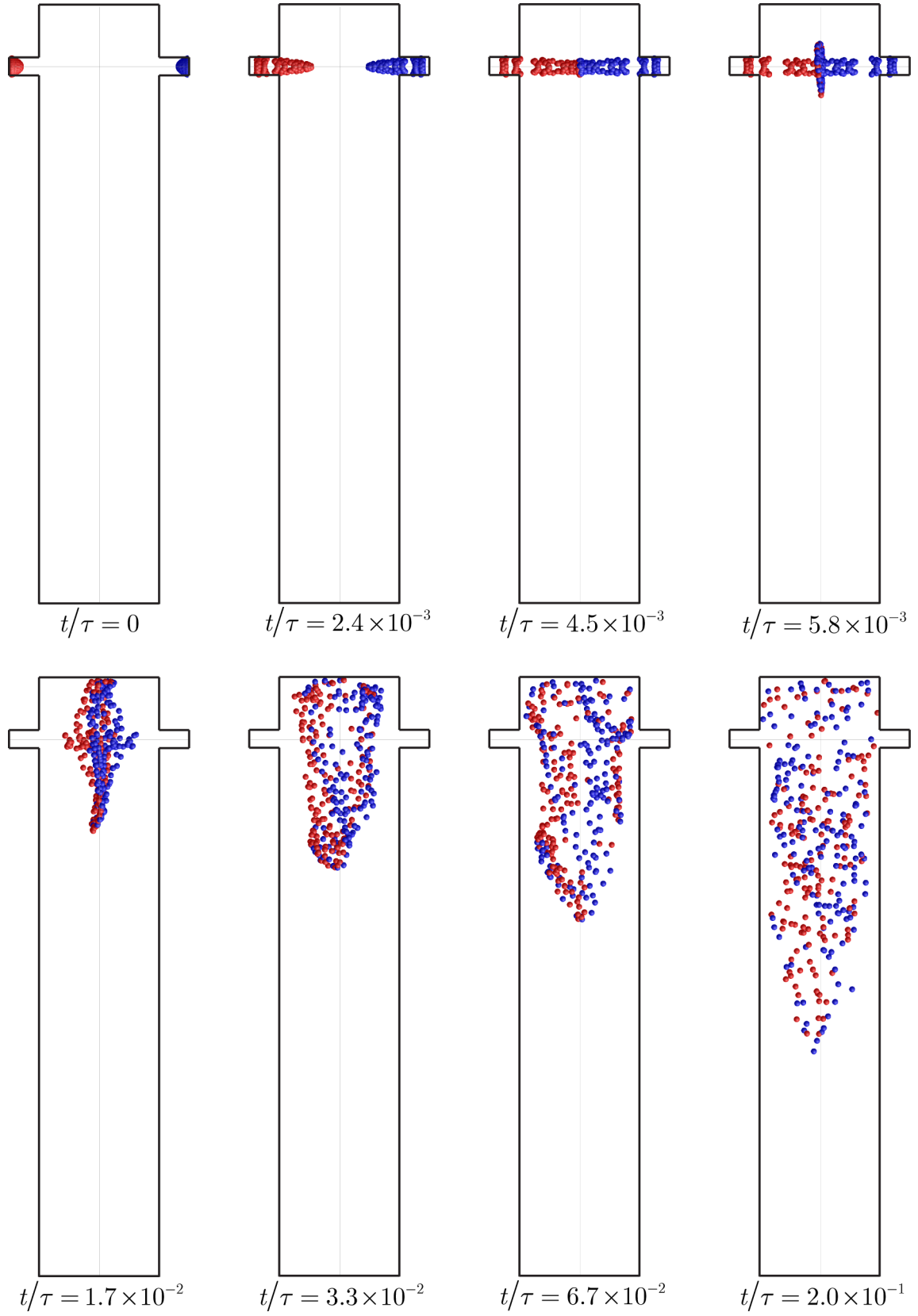


Figure 2.17. Front view of the particles' trajectory for $Re=110$.

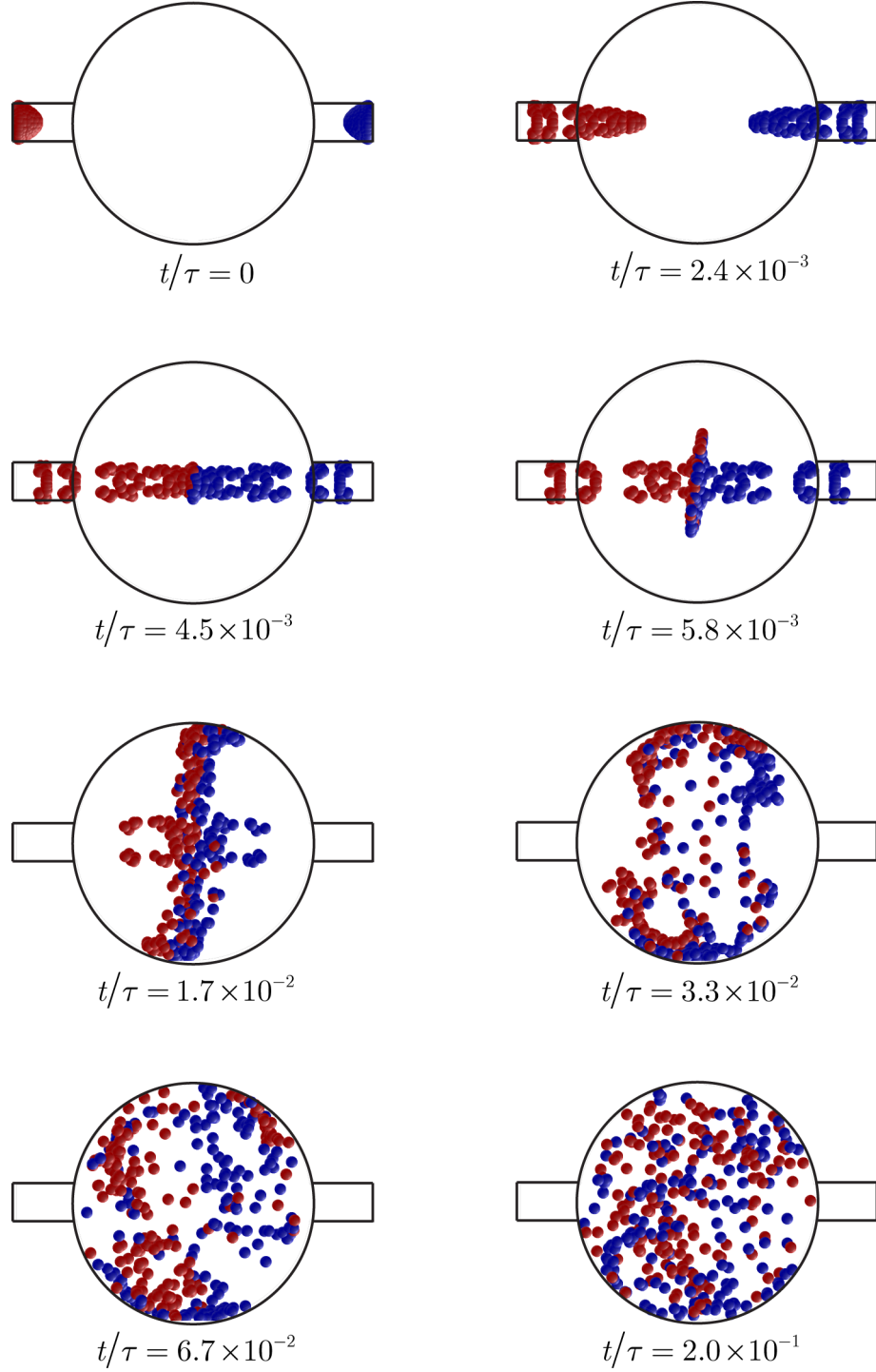


Figure 2.18. Top view of the particles' trajectory for $Re=110$.

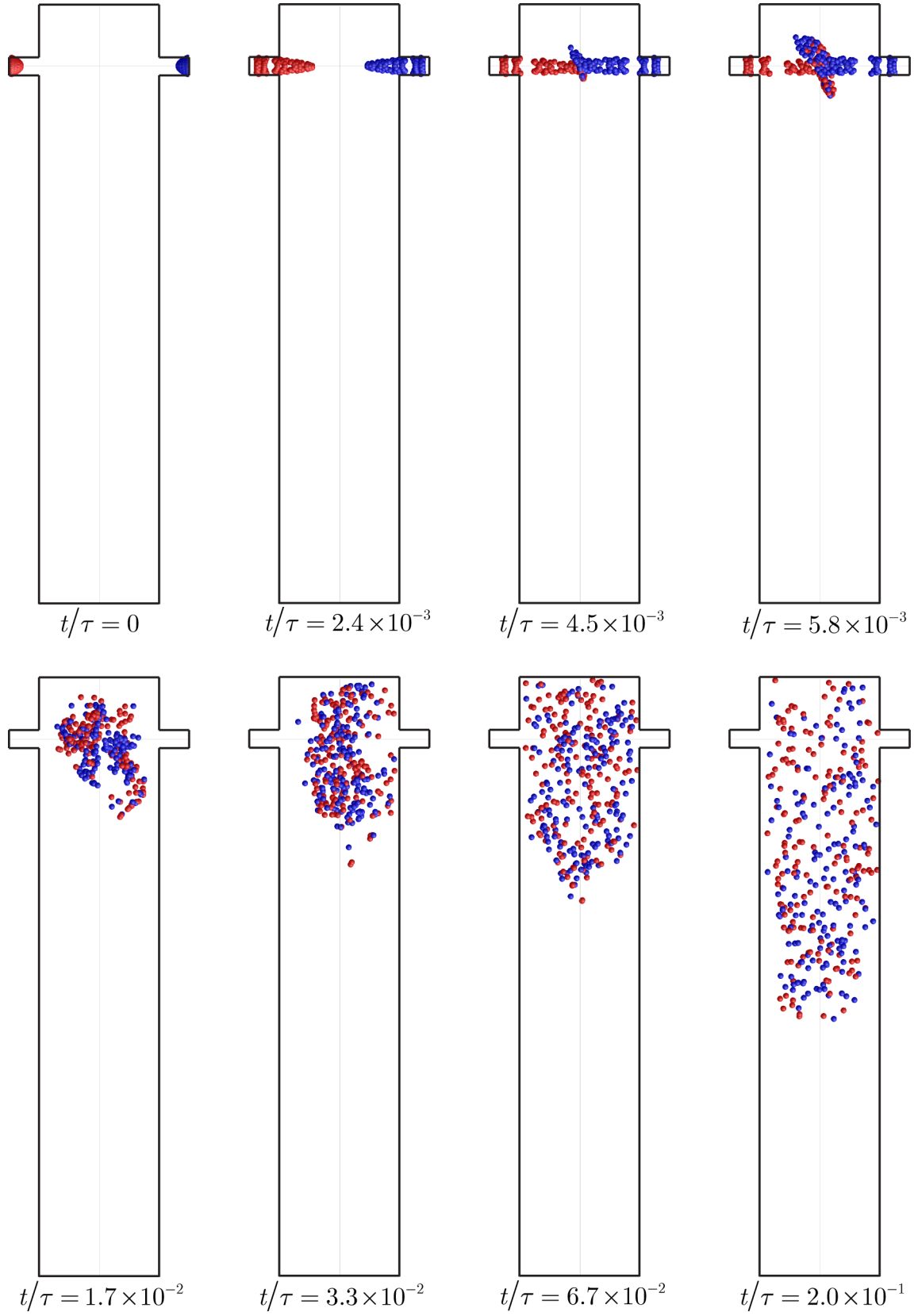


Figure 2.19. Front view of the particles' trajectory for $Re=200$.

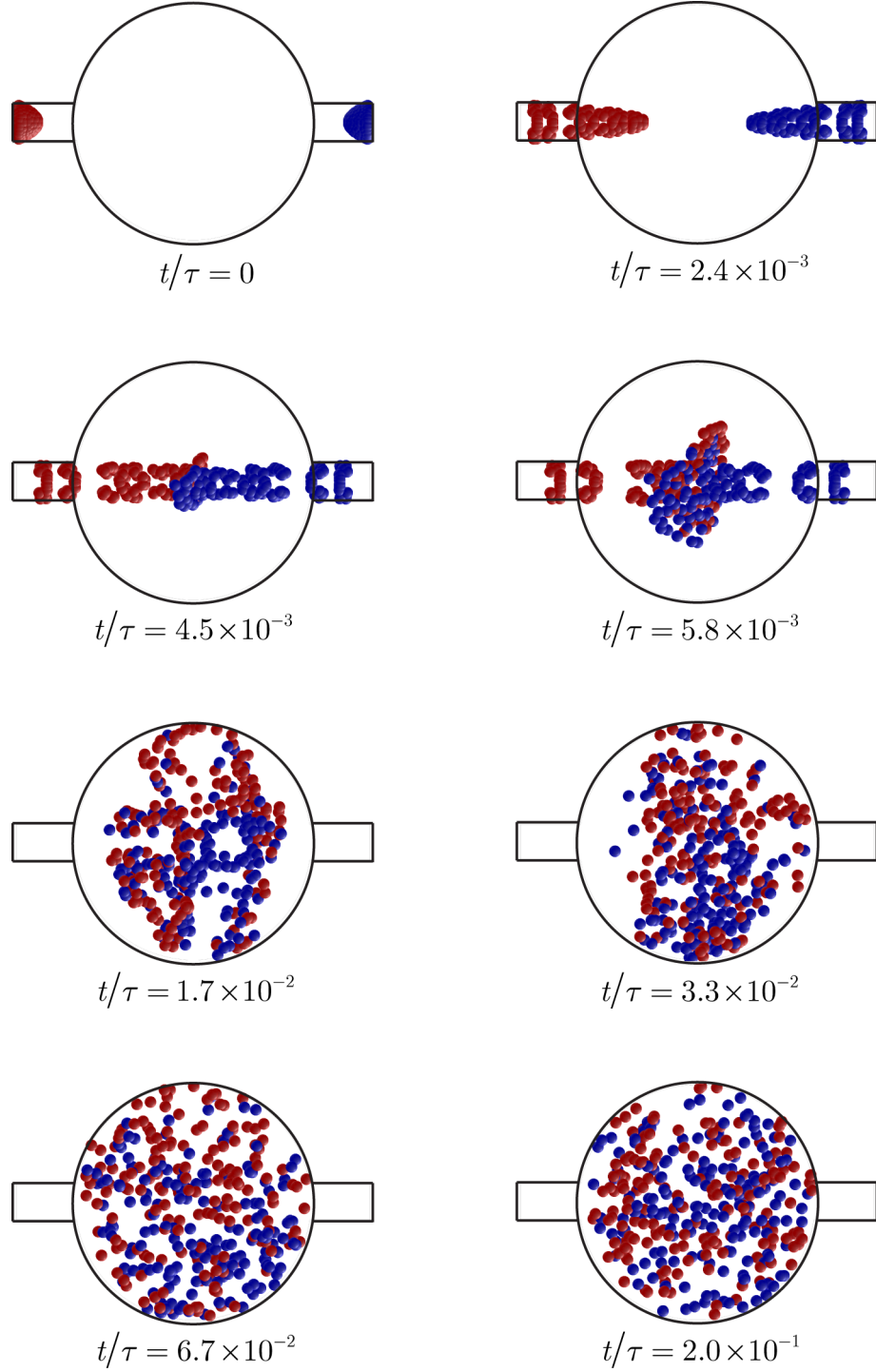


Figure 2.20. Top view of the particles' trajectory for $\text{Re}=200$.

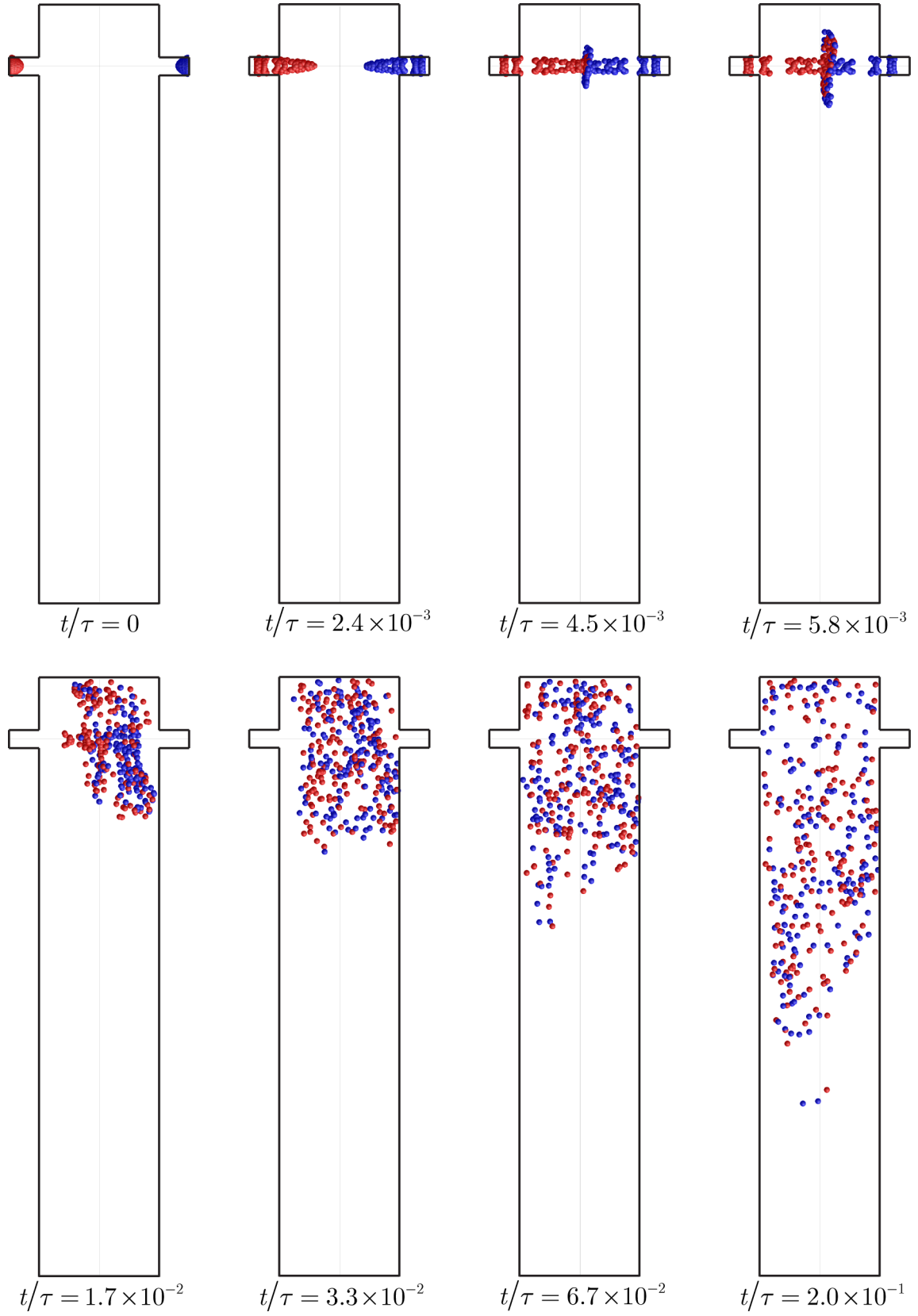


Figure 2.21. Front view of the particles' trajectory for $Re=300$.

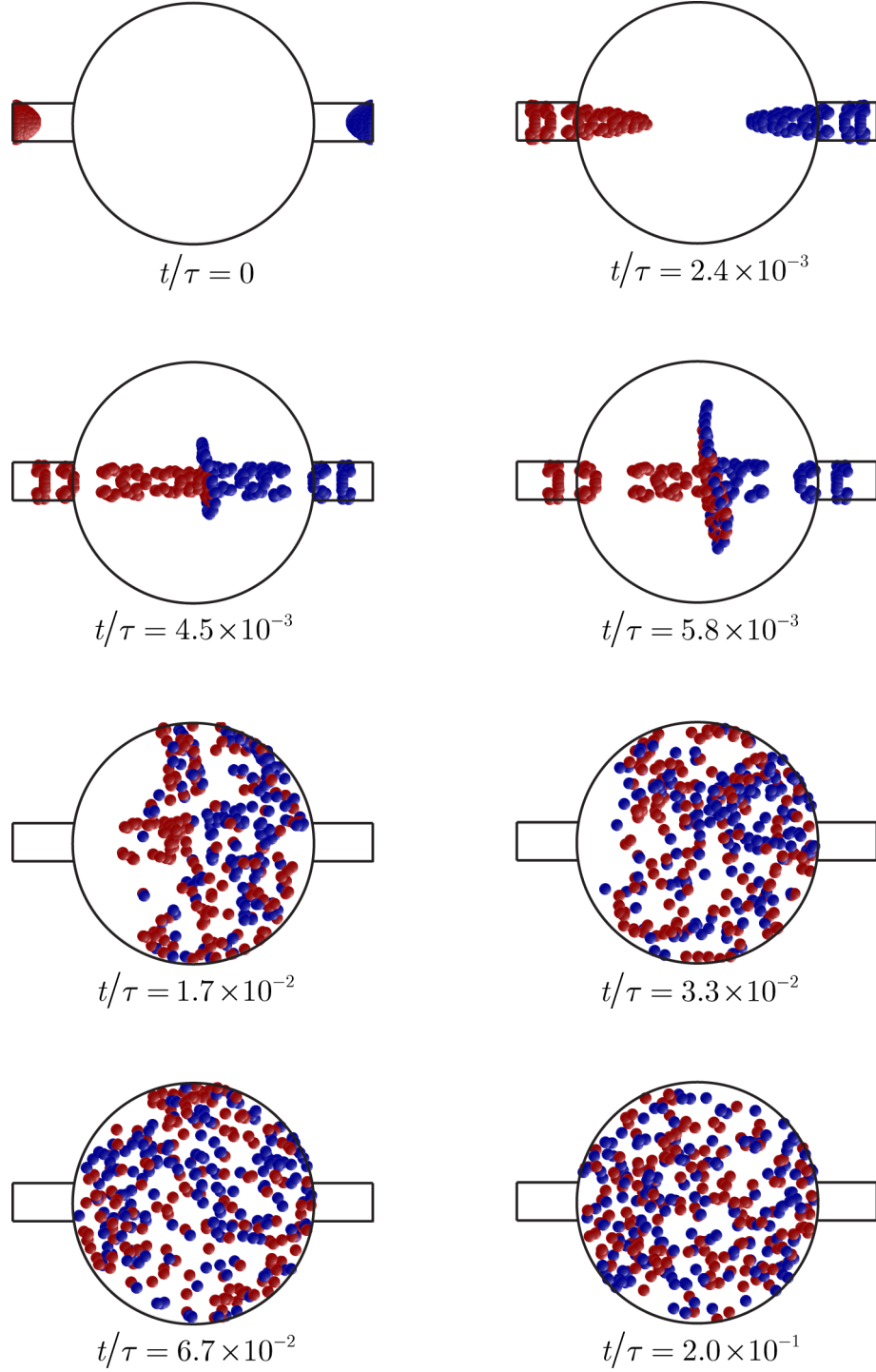


Figure 2.22. Top view of the particles' trajectory for $\text{Re}=300$.

2.4.3 Stretching Histories

From the flow strain properties extracted along the particles' trajectories, the stretching experienced by each element was calculated as described in Section 2.3.2 for $Re = 100, 110, 200, 300$. Figure 2.23 shows the average stretching, $\langle \lambda \rangle$, as a function of time and the Reynolds number. The average stretching is calculated as

$$\langle \lambda(t/\tau) \rangle = \frac{1}{N_{\text{particles}}} \sum_{i=1}^{N_{\text{particles}}} \lambda_i(t/\tau) \quad (2.42)$$

where $N_{\text{particles}}$ is the number of injected particles. Figure 2.23 shows that for $Re = 100$ and 110 , despite of the flow being unsteady for $Re = 110$, the elements undergo, on average and for longer times, a linear stretching with time. As shown in Section 2.4.2, material transfer between chamber sides occurs for $Re = 110$ but trajectories reorientation relatively to the streamlines is not potentiated by the flow.

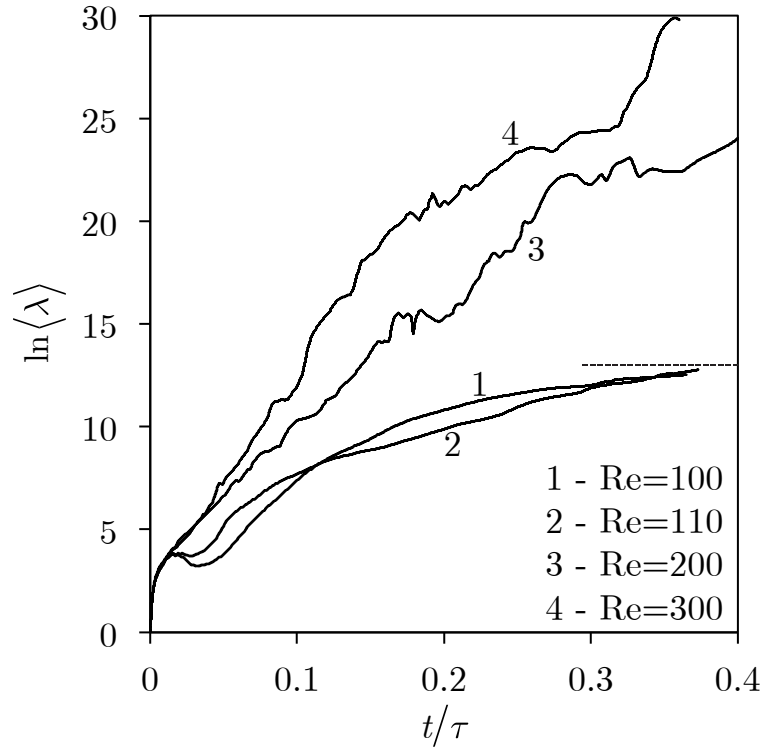


Figure 2.23. Average particle stretching at several Reynolds numbers.

For $Re = 200$ and 300 (Figure 2.23) the average stretching increases nearly exponentially with time. Due to its dynamic behavior, the flow develops the ability to reorient the trajectories relatively to the streamlines and improve mixing. This nearly exponential average stretching of fluid elements increase with time is a feature of chaotic flows (Ottino, 1990). These results clearly show that the flow is laminar and that the nature of mixing in impinging jets, in the studied range of Reynolds number and when the dynamic regime is onset, is chaotic advection.

Not all of the elements experience the same stretching. Stretching is highly dependent on each element's origin and residence time at the top of the chamber, where strain rate is higher and the flow reorients trajectories. The parallel flow in the remaining chamber also contributes to the deformation of the elements but not at an exponential rate. The distribution of stretching experienced by the elements has been determined. Figure 2.24 shows the cumulative distribution function of the stretching up to the moment when the first particle leaves the mixing chamber. Figure 2.24 shows that, in the studied range of Re , the average stretching experienced by the elements increases several orders of magnitude as Re is increased. Also, the fraction of elements with higher deformation increases highly with Re . As an example, while for $Re = 100$ 90% of the particles suffer stretching up to $\lambda = e^8$, the same fraction for $Re = 300$ suffer stretching 6 orders of magnitude higher up to $\lambda = e^{22}$. These results also predict that, as Re is increased, smaller striation scales are achieved in the flow, contrary to the proposed by Baldyga and Bourne (1983).

The stretching distribution function will be used in Section 2.4.4 to determine the distribution of striation scales, following the methodology of Section 2.3.3.

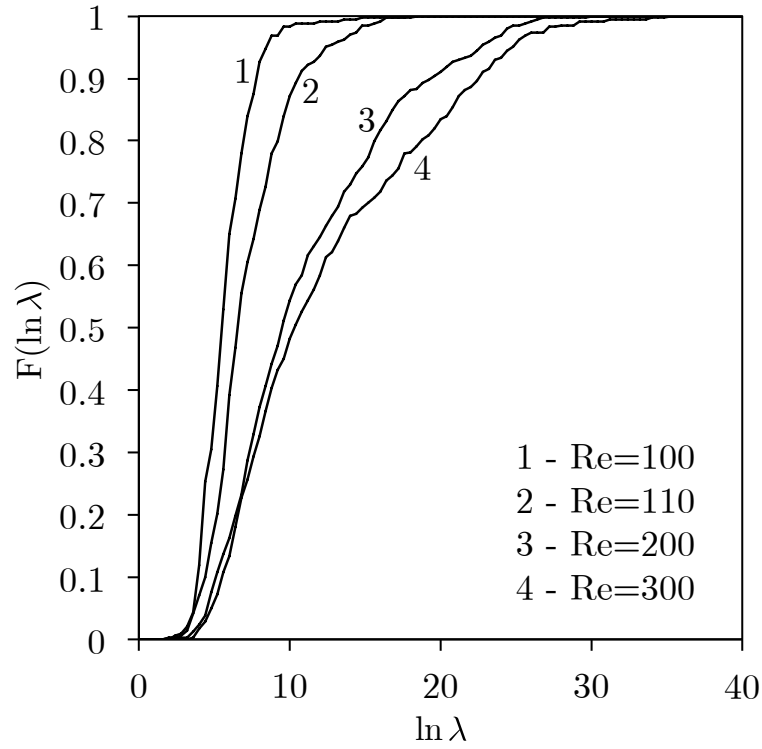


Figure 2.24. Cumulative distribution of the particle stretching.

2.4.4 Striation Thickness Distribution

The striation thickness of the lamellar structures in the flow was estimated with the methodology described in Section 2.3.3. Figure 2.25 shows the cumulative striation thickness distribution function from the CFD simulations for $Re = 100, 110, 200$ and 300 inside the mixing chamber at the moment when the first injected element leaves the mixing chamber. Figure 2.25 also shows the striation thickness distribution predicted by the lamellar model of Baldyga and Bourne (1983), based on the statistical theory of turbulent diffusion, for the mixing chamber dimensions of Figure 2.4, $r_s = 1$ and $n = 2.5$. The distribution for $Re = 100$ is merely indicative and is not expected to be observed in the actual flow since the absence of material transport between sides will maintain the fluids mainly segregated.

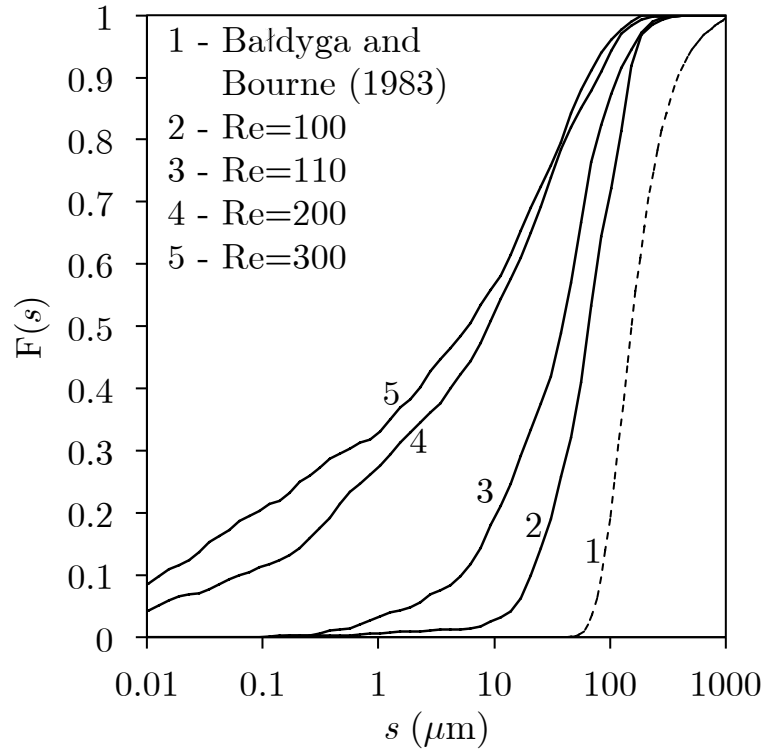


Figure 2.25. Cumulative striation thickness distribution.

For $Re > 110$, and despite that most elements will undergo further deformation until completely leave the chamber, Figure 2.25 shows that more than half of the striations have thicknesses lower than the desired of the order of $10 \mu\text{m}$ to avoid diffusional limitations during the polymerization in the mold (Lee et al., 1980).

As the Reynolds number is increased, Figure 2.25 shows further reduction of mixing scales. This observation is consistent with the observed experimentally in Planar Laser Induced Fluorescence experiments in Chapter 4. The mean striation thickness also decreases with increasing Re . Figure 2.26 shows the mean average striation thickness obtained from the CFD simulation and compares it with the values predicted by the models presented in Section 2.1 for mixing chamber dimensions of Figure 2.4, $r_s = 1$ and $n = 2.5$. For Reynolds numbers, at which the self-sustained laminar chaotic flow regime is fully established, the order of magnitude of the average striation scale is in more agreement with the values predicted by the kinematic models of Lee et al. (1980) than with values predicted from the models based on the statistical theory of turbulent diffusion. These results support the argument that the flow in the studied range of Re is laminar and not turbulent. Considerations of

isotropy and homogeneity of turbulent properties for description of impingement jets flow are not suited physically.

The effect of diffusion is not included in the calculation of the striation thickness distribution, and thus the distributions in Figure 2.25 should be assessed with caution. If the reduction of scales is large enough, molecular diffusion will be able to fade striations with thickness $s \sim \mathcal{O}(\sqrt{\mathcal{D}t})$ for times $t \sim \tau$, where $\mathcal{D}$ is the molecular diffusivity. For molecular diffusivities of monomers or pre-polymers around $\mathcal{D} \sim 10^{-11} \text{m}^2 \text{s}^{-1}$ (Bourne and Garcia-Rosas, 1985), striations with thicknesses $s < 1 \mu\text{m}$ would be completely eliminated by diffusion still inside the mixing chamber. The prediction of mixing length scales less than $1 \mu\text{m}$ in this work suggests a phenomenon of *rupturing* in addition to the mixing mechanisms of folding and stretching in RIM. A blob of material can be imagined to be stretched enough in the flow to form a sharper filament. In the mathematical abstraction of a *continuum* of matter, a thin line of material (infinitesimally small) with sharp interface would permanently stretch and unite the two extremities of the initial blob, independently the degree of stretching undergone by the blob. However, diffusion will eventually fade the thinner unions of the filament, resulting in rupturing of the blob, i.e., the formation of different isolated blobs of material. Rupturing is not used here with the same meaning as the break-up phenomenon due to surface tension in immiscible fluid systems but as a mechanism where thin laminae fade by diffusion. This mechanism is also observed and mentioned in Chapter 3 from an Eulerian approach to the problem of reduction of scales with numerical simulations of the impinging jets flow.

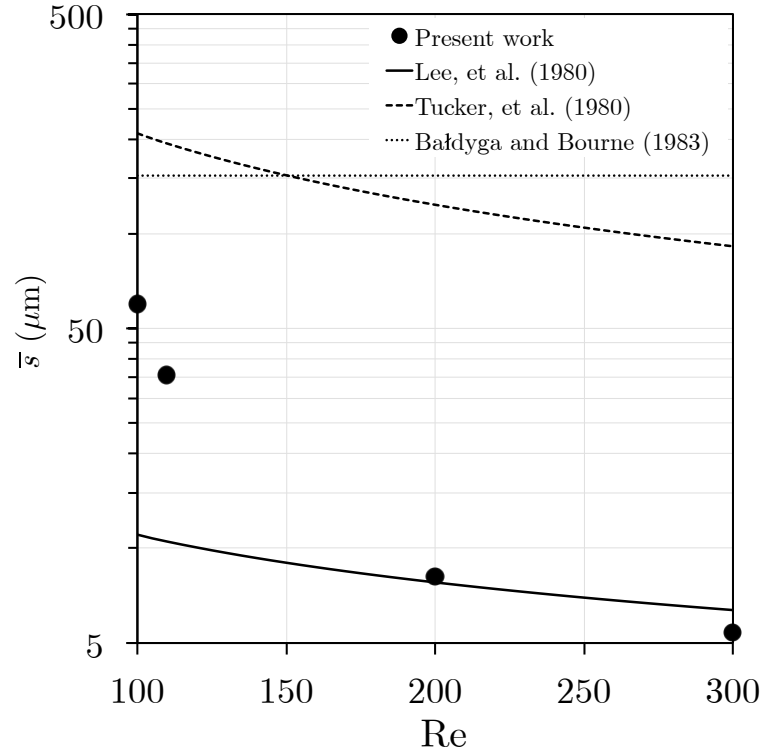


Figure 2.26. Mean striation thickness as a function of Reynolds number.

2.5 Conclusions

In this chapter the lamellar microstructure generated by impinging jets flow, with applications to RIM, is studied with Computational Fluid Dynamics techniques from a Lagrangian approach. Two flow regimes were identified in the considered range of Reynolds numbers:

- For $\text{Re} < 110$ the flow is laminar and steady with poor mixing properties;
- For $\text{Re} > 110$ the flow develops to a self-sustained laminar chaotic regime with strong dynamics and good mixing properties.

For $\text{Re} > 110$, reduction of scales in impingement mixing results from an increase of the stretching rate combined with a strong flow reorientation, both at the top of the mixing chamber. $\text{Re} > 110$ is the critical Reynolds number that marks the transition from a laminar steady flow regime to a self-sustained laminar chaotic flow regime. The combination of the two mechanisms (stretching and reorientation) allows the

flow to exponentially generate interfacial area between the two monomers or pre-polymers in RIM and reduce scales to a value that avoids diffusional limitations during the polymerization reaction in the mold. For $Re > 110$, the flow is shown to produce nearly exponential average stretching of fluid elements with time, a characteristic feature of chaotic flows. Impingement mixing is observed to occur in the top of the chamber until a distance $\sim 2D$ from the top wall. In the remaining chamber the flow develops a nearly parallel profile. The flow in the bottom of the chamber still contributes to further reduction of mixing length scales but linearly due to the lack of reorientation of trajectories. Smaller striation thicknesses in the monomers or pre-polymers mixture are observed as the Reynolds number is increased.

Computational results of this work are compared to theoretical models existent in the literature. Results show to be consistent with the kinematic model of Lee et al. (1980), rather than with models based on the homogeneous and isotropic turbulence considerations (Baldyga and Bourne, 1983; May, 1996; Tucker III and Suh, 1980). These results support the argument that the flow in the studied range of Re is laminar and mixing is achieved by chaotic advection and not by turbulence.

Simulations show formation of striation scales thin enough to be faded by diffusion in times close to the mixing chamber mean residence time. This result suggests a mechanism of *rupturing*, associated with stretching and folding mechanisms, capable of generating isolated blobs of material in the medium.

Results show how impingement mixing in RIM produces striation length scales sufficiently thin for the rate of polymerization reaction to become chemically controlled in RIM.

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3 2D CFD Modeling of Lamina Dynamics in Impinging Jets Flow with VOF

3.1 Introduction and State of the Art

The study of mixing in impinging jet mixers, namely in RIM machines' mixing chambers, has been the subject of study over the past years, where the characterization of mixing was achieved using different methodologies and techniques such as: from the adiabatic temperature rise in the mixing chamber (Lee et al., 1980; Kolodziej et al., 1982; Kolodziej et al., 1986); the study of the final mechanical properties of the polymer (Kolodziej et al., 1982; Kolodziej et al., 1986); a test chemical reaction (Kusch et al., 1989); from mixing of fluids with a tracer or seeded with particles (Malguarnera and Suh, 1977; Lee et al., 1980; Tucker III and Suh, 1980; Sandell et al., 1983; Wood et al., 1991; Johnson et al., 1996; Trautmann and Piesche, 2001; Schütz et al., 2005); and using laser velocimetry methods (Wood et al., 1991; Johnson et al., 1996; Johnson, 2000; Teixeira et al., 2005; Santos et al., 2008; Santos et al., 2009). Many of these studies observed that the main mixing mechanism in RIM is the formation of vortices downstream the opposed jets impingement point, where these vortices engulf the fluids from the opposite jets (Lee et al., 1980; Wood

et al., 1991; Santos et al., 2008). Nevertheless a thorough characterization of the mixing mechanisms in this type of reactors and quantification of the mixing length scales is not yet available. The main reasons for this are probably related to the vortices evolving very rapidly and being formed at high rates, and also to the tracer diffusion that rapidly homogenizes the two fluids (Sandell et al., 1983).

The effect of the striation thickness distribution on the course of unpremixed polymerizations was studied by Fields and Ottino (1987a) and (1987b), particularly for the RIM process. In these studies a lamellar model was used to evaluate the extent of the polymerization reaction in one-dimensional distributions of alternating monomers, from both monodispersed and distributed material striations, and also the effect of the stretching history on monomers layers. The studies have shown that the reaction time scale could be several orders of magnitude underestimated if a monodispersed striation thickness distribution is considered. The striation thickness distribution may have a severe impact on the polymer properties such as the maximum and average molecular weight achieved.

From 1986 until now, the increasing availability of computational resources has allowed solving many scientific and engineering problems, although the issue of striation thickness distribution in RIM has remained without an answer. The latest known attempt to tackle this challenge was made by May (1996) using the statistical theory of turbulent diffusion.

Santos et al. (2005) also studied the impingement mixing in a RIM machine mixing chamber from CFD simulations with species transport. Due to computational limitations, an unrealistically high value for the diffusion, 10^3 times larger than the actual values, was considered in the simulations. Computer memory limitations and high computational times made impracticable the use of the actual molecular diffusion values, even in reactors as small as a mixing chamber of RIM machines. Even so, the 2D CFD simulation took around six months to cover a period of five passage times in the mixing chamber (approximately 50 ms). As a consequence, the

fluids at the outlet of the mixing chamber were too homogeneous and it was not possible to identify any lamellar structures; thus the available data from Santos et al. (2005) could not be used to quantify a distribution of mixing length scales. In the particular case of RIM, the main mixing mechanism is not diffusion, neither turbulent nor molecular; molecular diffusivity in polymer systems is around $\mathcal{D} = \mathcal{O}(10^{-11})\text{m}^2\text{s}^{-1}$ and in the range of industrial interest ($\text{Re} \leq 600$) the flow is laminar. The main mixing mechanism is the striation thinning of the initial laminae, i.e., the monomers laminae that are produced by the opposed jet streams. Here the Reynolds number is defined as

$$\text{Re} = \frac{\rho d u_{\text{inj}}}{\mu} \quad (3.1)$$

where d is the injectors' diameter, u_{inj} the mean velocity at the injector and ρ and μ the fluid's density and viscosity, respectively.

Previous works on the simulation of mixing without diffusion were made using particle tracking (Howes et al., 1991; Aubin et al., 2003). Roberts (2010) used particle tracking to show the need for methods that are not affected by diffusion for the identification of the local microstructure in laminar flows. Another method for the quantification of mixing without diffusion is the conformal mapping method (Galaktionov et al., 2000; Galaktionov et al., 2001; Kruijt et al., 2001; Galaktionov et al., 2002a; Galaktionov et al., 2002b; Singh et al., 2008; Meijer et al., 2009). Galaktionov et al. (2002a) used this method in a 3D geometry with industrial application, the Kenics static mixer.

An alternative computational model is here proposed for the thorough characterization of the advective mixing mechanisms and visualization of the lamellar structure in impinging jets flow. An Eulerian-Eulerian approach is used using Computational Fluid Dynamics (CFD) with the Volume-of-Fluid (VOF) model to simulate the dynamics of mixing of two phases in the flow. Simulations using the VOF model track the interface between both phases and the formation of thinner

laminae of material. Both phases are set to have the same physical properties and the surface tension at the interface is set to zero. This model was chosen over the approach previously used by Santos et al. (2005) due to the fact that with VOF, a value of zero for the diffusion coefficient between the two phases can be set.

Although the study of mixing in a cylindrical mixing chamber of a RIM machine is in mind, a 2-dimensional (2D) model of the flow was chosen for its ability to describe some of the most important flow features in impingement mixing, and because a 3-dimensional (3D) model, as will be demonstrated in this chapter, would not be computationally affordable for a VOF simulation with acceptable spatial discretization to simulate the length scales of the lamellar structure.

The 2D model for the simulation of the mixing chamber has been validated over the last years for the simulation of cylindrical RIM machines mixing chambers (Santos, 2003; Santos et al., 2009; Santos et al., 2010). The main differences between the 2D model and the actual 3D flow field are:

- In the 3D flow field the jets follow a straight line up to the impingement point and afterwards the fluids are spread radially forming a pancake like structure that promotes a large interface for the contact of both fluids (Wood et al., 1991). The stretching due to the fluids spreading can reduce the initial scale of the jets to a tenth of its original value. In the 2D geometry, the jets are bent towards the outlet prior to impinging each other, and immediately after the impingement point the jets maintain the same velocity (Santos et al., 2005).
- On both 2D and 3D geometries, the self-sustainable chaotic flow regime is characterized by the formation of vortices after the jets' impingement point. These vortices engulf fluid from both jets and evolve towards the outlet. The main 2D/3D difference is that in 3D the vortices dissipate faster, typically within a distance equal to one to twice a mixing chamber diameter from the jets' impingement point (Lee et al., 1980; Teixeira et al., 2005), while in 2D the vortices evolve throughout longer distances.

Nevertheless, there are some common features between the 2D and 3D flows, and this has been the basis for using the 2D model on the representation of the 3D flow. The 3D flow features that are simulated by the 2D model are:

- A steady flow regime at lower Reynolds numbers where the two fluid streams flow segregated throughout the mixing chamber.
- A chaotic flow regime where the two fluid streams are engulfed by successive vortices that shed from the jets impingement point towards the mixing chamber outlet.
- The jets' impingement point oscillates with a frequency equal to the vortices formation rate (Santos et al., 2009).

Furthermore, the results from the 2D model have an increasing interest if new RIM machine mixing chamber designs are considered. The fact that the vortices in 2D geometries evolve through longer distances of the mixing chamber was the basis for a new RIM mixing head (European Patent EP 1 732 747 and US Patent 7708918) where the mixing chamber is prismatic. The prismatic mixing chamber for RIM is an extrusion of the 2D geometry in the normal direction. Experimental results obtained on prismatic geometries were published by (Sultan et al., 2010), where it was observed that the 2D model simulates the flow patterns observed in the prismatic chambers.

This work replaces turbulence assumption by the actual visualization of laminar mechanisms, which are more physically sound as far as mixing in RIM is concerned. The results of this work identify the laminar advective mechanisms that enable the decrease on mixing scales in RIM down to the value of $10\ \mu\text{m}$ required for chemical reaction without diffusional limitations in the mold (Lee et al., 1980).

In the following sections is described the process of development of grid-adaptive non-diffusive model for the simulation of the dynamics of the lamellar structures formed

in impingement jets flow. Part of the results presented in this chapter were published by Fonte et al. (2011).

3.2 CFD Model of the Flow

3.2.1 Physical Domain

The 2D physical domain corresponds to an axial cut of a typical industrial RIM machine mixing chamber with the same dimensions of the cylindrical chamber used in the experimental work of Teixeira (2000), Santos (2003), Teixeira et al. (2005), Santos et al. (2008) and in this work (Chapters 2 and 4). The diameter of the injectors and of the chamber are $d = 1.5$ mm and $D = 10$ mm, respectively. The chamber height is $H = 50$ mm and the axis of the injectors is located at a distance $h = 5$ mm from the top of the chamber. A schematic drawing of the simulation domain and respective dimensions is shown in Figure 3.1. For the steady state simulations, a symmetry plane was placed in the middle of the chamber, along the chamber height dividing it into two. This symmetry condition was used to force an initial symmetric solution of the flow in steady state conditions, which is difficult to obtain at higher Reynolds numbers as previously observed by Santos et al. (2010). The symmetry plane is removed from the domain for the dynamic simulations.

Contrary to the observed in the 3D mixing chamber cylindrical geometry where the flow is parallel to the chamber walls at the outlet (see Chapter 2), backflow into the domain occurs in the 2D mixing chamber, as reported by Teixeira (2000) and Santos (2003). To avoid problems when defining the outlet boundary condition, an extended outlet was used by extending the chamber length a distance equal to D , which ensured that swirling fluid reentering from the extended outlet would not reach the real chamber outlet. Simulation results were obtained in the entire extended volume but only considered in the domain within the real dimension of the chamber, marked in gray in Figure 3.1.

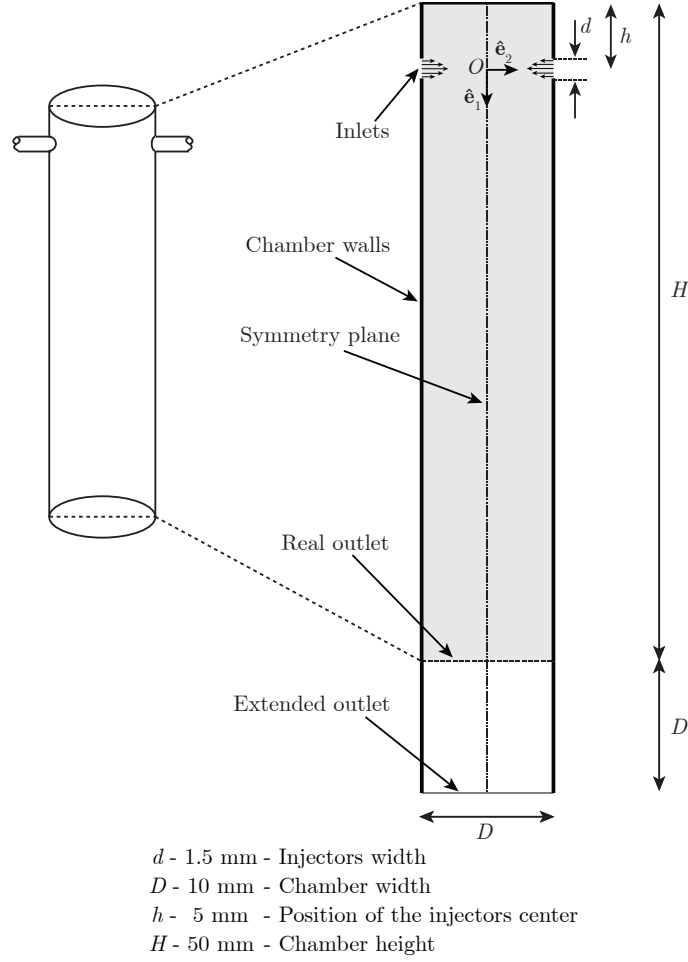


Figure 3.1. Schematic representation of the simulation domain.

3.2.2 Governing Equations and Boundary Conditions

The flow of an incompressible fluid inside the 2D simplified domain of the RIM machine's mixing chamber was simulated by solving the Navier-Stokes equations:

$$\nabla \cdot \mathbf{u} = 0 \quad (3.2)$$

for mass conservation, where $\mathbf{u} = u_1 \hat{\mathbf{e}}_1 + u_2 \hat{\mathbf{e}}_2$ is the 2D velocity field, and

$$\rho(\partial_t \mathbf{u} + \mathbf{u} \cdot \nabla \mathbf{u}) = -\nabla p + \mu \nabla^2 \mathbf{u} \quad (3.3)$$

for momentum conservation, where t is the flow time and p the pressure.

The conservation equations for the flow field were solved for the following boundary conditions: the no-slip condition at the chamber walls, setting $\mathbf{u} = \mathbf{0}$; a symmetry or shear-free condition at the inside wall, when enabled, setting $u_2 = 0$ and $\partial_{x_2} u_1 = 0$, a fully developed parabolic flow at the inlets; a uniform and constant reference pressure at the extended outlet, $p = p_0$, equal to the atmospheric pressure.

3.2.3 Domain Discretization

The computational grids consisted of uniformly spaced quadrilateral cells, with different sizes. Previous works from Teixeira (2000) and Santos (2003) have shown a grid with spatial discretization $\Delta x \leq 100 \mu\text{m}$ to be sufficiently refined to simulate all flow hydrodynamic length scales in the range of the studied Reynolds numbers. All simulations were performed with numerical grids with elements with size below or equal to $100 \mu\text{m}$. The geometrical domain and simulation grid were generated with the software packages *DesignModeler* and *Meshing* included in Ansys® 14 suite.

3.2.4 Numerical Solution in Steady State

Initially, steady state simulations of the flow were performed, setting the transient term of Equation (3.3) equal to zero. The symmetry interior plane was enabled in the chamber to ensure a symmetric flow field solution. The velocity vector field was set equal to zero and $p = p_0$, in the entire domain as the initial condition. The obtained steady state results of the flow inside the mixing chamber were later used as an initial condition for dynamic simulations.

Simulations were performed in an Intel® Xeon® 3 GHz 8-core machine running the finite-volume commercial CFD software ANSYS Fluent™ 12.1 for 64-bits Windows®.

The COUPLED scheme was used for the pressure-velocity coupling. The PRESTO! and QUICK schemes were used for the numerical discretization of the pressure and momentum advective terms of Equations (3.2) and (3.3), respectively.

Steady state simulations results were accepted when a stable and constant value of the normalized residues of the continuity and momentum conservation equations was reached. In all simulations these values were lower than 10^{-8} . In addition to this criterion, the inlet and outlet mass flow rates were continuously compared to guarantee that the mass conservation principle was being satisfied.

3.2.5 Numerical Solution in Transient State

Dynamic flow simulations were started from the symmetric steady-state simulations by removing the interior symmetry condition and imposing a non-symmetric finite perturbation. The perturbation was set by varying smoothly the velocity of the left injector with time with the same methodology of Teixeira (2000) and Santos (2003). The maximum amplitude of the perturbation was set to 10% of the inlet velocity and for a limited duration equal to 10% of the chamber mean residence time. The chamber mean residence time, τ , is defined as

$$\tau = \frac{DH}{2u_{\text{inj}}d}. \quad (3.4)$$

The influence of the perturbation parameters on the flow dynamics was evaluated by Santos et al. (2010) who have shown that the perturbation has no impact on the flow dynamics after the transient period where the system evolves from the initial state. Any quantitative analyses to the flow from transient simulations were done for flow times superior to 3τ to ensure that the initial perturbation had no longer impact in the flow.

An Explicit First Order scheme was used for the discretization of the transient terms of the momentum equation (Equation (3.3)) in the dynamic simulations. The time step size, Δt , was chosen according to

$$\Delta t = \frac{\Delta x}{3/2u_{\text{inj}}}. \quad (3.5)$$

For the dynamic simulations, a normalized value of 10^{-4} for the residues of all variables was used as a convergence criterion.

3.3 VOF Model for Interface Tracking

To study the dynamics of the lamellar structure in the impingement jets flow the VOF model was used. The VOF model, first introduced by Hirt and Nichols (1981), allows tracking the interface between two immiscible passive phases, A and B, in the flow and its purely advective mixing by solving an equation for a scalar, α_A , representative of the volume fraction of one of phase A. A value of $\alpha_A = 1$ means that a finite volume in the discretized domain is filled with phase A and $\alpha_A = 0$ means that the finite volume is empty of phase A. When a finite volume contains the interface between the two phases, the scalar α_A assumes a value $0 < \alpha_A < 1$, depending on the amount of each phase present in that volume. In the binary mixture, the volume fraction of phase B, α_B , is obtained by using the constraint $\alpha_B = 1 - \alpha_A$, forcing the sum of the volume fractions of each phase in each control volume in the 2D discretized domain to be equal to 1.

The spatial and temporal distribution of α_A is obtained by solving, along with the conservation equations for mass (Equation (3.2)) and momentum (Equation (3.3)), the advection transport equation

$$\partial_t \alpha_A + \mathbf{u} \cdot \nabla \alpha_A = 0. \quad (3.6)$$

Density of $\rho = 10^{-3} \text{ kg/m}^3$ and viscosity of $\mu = 2 \times 10^{-2} \text{ Pa} \cdot \text{s}$ were assumed for both phases, close to the physical properties of the fluid used in the experiments of Chapters 4 and 5. No surface tension was considered at the interface, thus not requiring additional terms to be added to the momentum conservation equation (Equation (3.3)) to take into account forces at the interface.

The scalar advection equation (Equation (3.6)) is solved for the following boundary conditions at the inlets:

$$\begin{cases} \alpha_A = 0 \text{ at the right inlet,} \\ \alpha_A = 1 \text{ at the left inlet.} \end{cases} \quad (3.7)$$

At the extended outlet, when backflow occurs, i.e., $\mathbf{u} \cdot \mathbf{n} < 0$, the following conditions were used:

$$\begin{cases} \alpha_A = 0 \text{ for } x_1 = H + D/2 \text{ and } x_2 \in [-D/2, 0], \\ \alpha_A = 1 \text{ for } x_1 = H + D/2 \text{ and } x_2 \in [0, D/2]. \end{cases} \quad (3.8)$$

Since the flow is non-diffusive, the volume fraction value at the walls and outlet, when $\mathbf{u} \cdot \mathbf{n} > 0$, are defined by the flow field.

As the initial condition, the left side of the chamber, to the interior wall, was set with $\alpha_A = 0$ and the right side with $\alpha_A = 1$ in the initial symmetric flow field. An Explicit First Order scheme was also used for the discretization of the transient terms of the advection equation (Equation (3.6)).

The accurate numerical integration of Equation (3.6) for tracking the discontinuous and sharp moving interfaces between the two phases is the critical task in this method (Hyman, 1984; Ferziger and Peric, 2002). Low order numerical schemes for the discretization of the advective term of Equation (3.6) can produce artificial mixing of the two fluids by inducing numerical diffusion at the interface. Higher resolution discretization schemes, interface reconstruction methods or higher grid refinement near the interface must be used to avoid this numerical problem and keep interfaces sharp.

Different high-resolution discretization schemes, available in Ansys Fluent[®], for the discretization of the advective terms of Equation (3.6) were tested in order to choose the best scheme suited for the description of the lamellar flow in the mixing chamber.

All discretization schemes for an explicit VOF formulation available were tested, namely:

- The Modified High Resolution Interface Capturing (HRIC) scheme, which consists of a non-linear blend of upwind and downwind differencing;
- The QUICK scheme, which is based on a weighted average of second-order upwind and central interpolations schemes of α_A ;
- The Compressive Interface Capturing Scheme for Arbitrary Meshes (CICSAM), which uses a slope limiter of the interface and is particularly suitable for flows with high ratios of viscosities between the phases.
- The Compressive scheme, which is a second order reconstruction scheme based on the slope limiter of the interface;
- The Geometric Reconstruction scheme, which assumes that the interface between two fluids has a linear slope within each cell and uses this linear shape for calculation of the advection of fluid through the cell faces.

Further information about these high-resolution discretization schemes can be found in the Fluent Theory Guide (Ansys, 2010).

The distribution of the two phases inside the mixing chamber for a grid element size of $50\ \mu\text{m}$ and Reynolds number equal to 300 was evaluated in the dynamic simulations of the flow after the perturbation until higher deformation of the initial interface started to be observed. Figure 3.2 shows the distribution of the two phases inside the domain for a time $t = 0.4\tau$ and for the different high-resolution schemes.

Despite no diffusional term is included in Equation (3.6), Figure 3.2 shows that results with the QUICK and Modified HIRC and CICSAM schemes present artificial diffusion at the interface. Other numerical problem was detected, the formation of “waves” at the interface in zones of higher shear when using the Modified HIRC, CICSAM and Compressive Schemes.

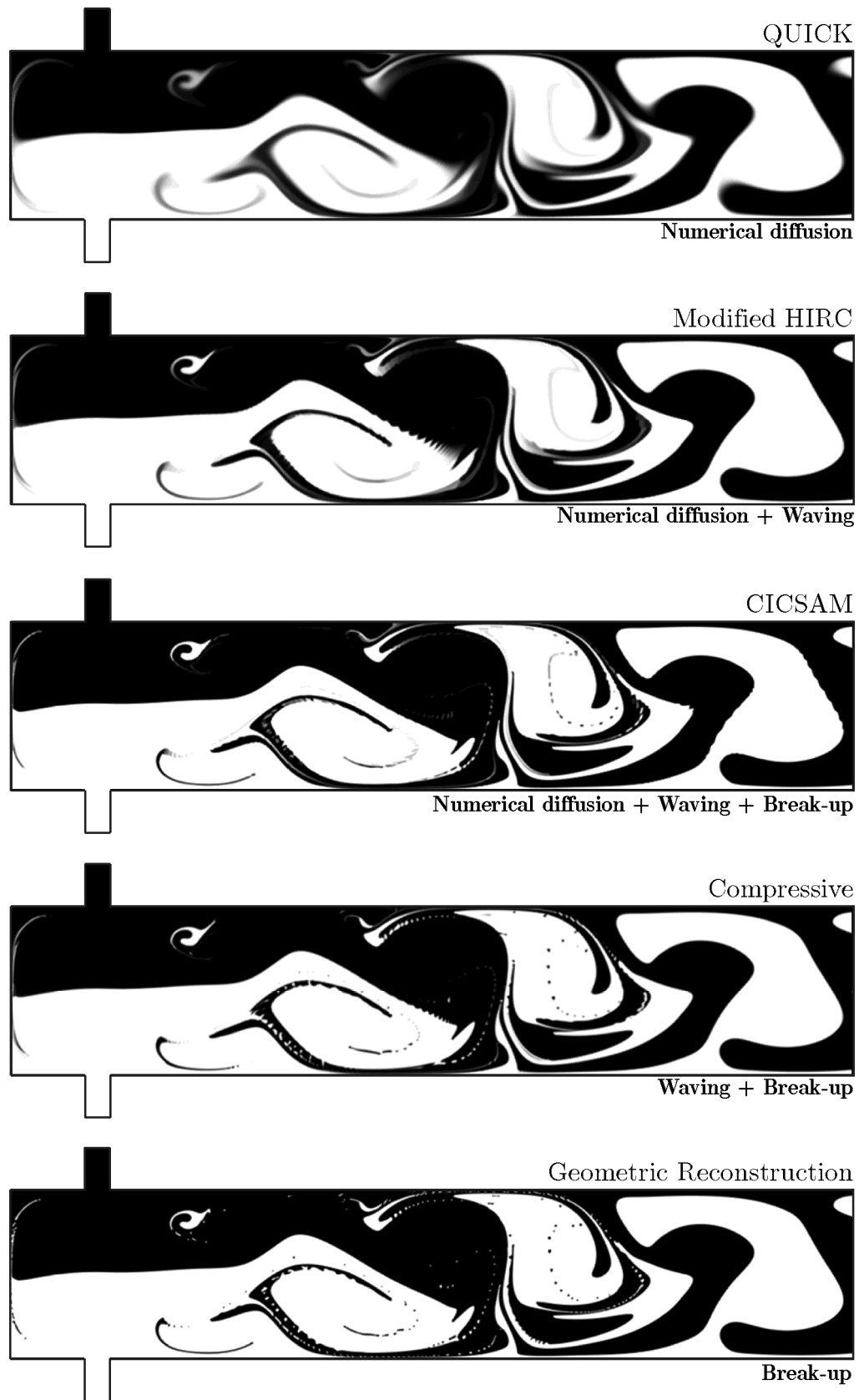


Figure 3.2. Distribution of phase A (black) and phase B (white) in the mixing chamber with different discretization schemes at a time $t=0.4\tau$.

Although no surface tension exists between phases, break-up is observed for smaller laminae when describing filaments of fluid thinner than the grid scale with the CICSAM, Compressive and Geometric Reconstruction schemes. This behavior indicates that the discretization schemes based on slope delimiters or interface reconstruction cannot describe laminae thinner than the grid spacing, and so the grid scale should be set accordingly to the smallest scale expected or physically possible in the system.

The best description of interfaces in the flow in the mixing chamber was considered to be obtained with the Geometric Reconstruction scheme since it avoids numerical diffusion and artificial waving of the interface.

The described methodology will be used in Section 3.4 to evaluate the effect of the Reynolds number in the formation of the lamellar flow out of two immiscible phases in impingement jets mixing.

3.4 Reynolds Number Effect in Mixing

The effect of the Reynolds number in impingement jets mixing in RIM is assessed from the contour plots of the two phases introduced in the mixing chamber separately, one phase from each jet. Two different Reynolds numbers were chosen for the study, below ($Re = 100$) and above ($Re = 300$) the critical value that marks the onset of the dynamic mixing as observed by Teixeira (2000) and Santos (2003). Figure 3.3 and Figure 3.4 show dynamic sequences of the volume fraction distribution maps of the two phases inside the mixing chamber and the maps of the component of the dimensionless vorticity normal to the 2D plane as a function of the dimensionless time t/τ for Reynolds numbers $Re = 100$ and 300 . The dimensionless vorticity, $\hat{\omega}$, is defined as

$$\hat{\omega} = \frac{d}{u_{inj}} (\nabla \times \mathbf{u}). \quad (3.9)$$

For a first approach, a grid with a grid spacing $\Delta x = 50 \mu\text{m}$ was chosen. In Figure 3.3 and Figure 3.4 the right injector is at the top-left corner and the left injector is at the bottom-left corner of each map. Both cases, $\text{Re} = 100$ and 300 , started from a steady state, where the two opposed jets impinge in the mixing chamber axis and are bent towards the outlet. Upon impinging each other, the jets flow towards the outlet in the center of the chamber and parallel to the mixing chamber axis. The right side of the chamber was set to contain phase A and the left side phase B. After being perturbed, the flow evolves differently for $\text{Re} = 100$ and $\text{Re} = 300$.

For $\text{Re} = 100$ after the initial perturbation (see Figure 3.3) the initial interface between the two phases is disturbed by the formation of vortices. As time advances, the system tends to a non-symmetrical steady state (Santos et al., 2010), which differs from the initial symmetric state. The jet that is formed inside the mixing chamber by the combination of the two inlet streams is attracted to one of the chamber walls and does not continue to flow, as initially, in the middle of the chamber, in a phenomenon resembling the Coanda effect (Houghton and Carpenter, 2003). In this flow regime, the fluids from the two opposed jets remain segregated while flowing through the mixing chamber.

Increasing the Reynolds number to 300 (see Figure 3.4) the system tends to a self-sustained chaotic dynamic laminar flow regime after being perturbed. This dynamic state is characterized by the existence of two counter-rotating vortices immediately downstream the inlets, each occupying half of the chamber diameter. These vortices detach to an alternating rotating vortex street along the chamber axis, which extend throughout the whole chamber width. At this chaotic state, strong jet oscillations occur, as observed experimentally by (Teixeira et al., 2005) and (Santos et al., 2009). From the evolution of the volume fraction maps in Figure 3.4, the role of these fully developed vortices is clear: when evolving throughout the chamber the fast counter-rotating vortices promote the formation of thinner laminae of each phase either by the stretching and folding of the material. Due to insufficient grid discretization, the

rupture of the formed laminae is observed when they reach thicknesses inferior to $50 \mu\text{m}$.

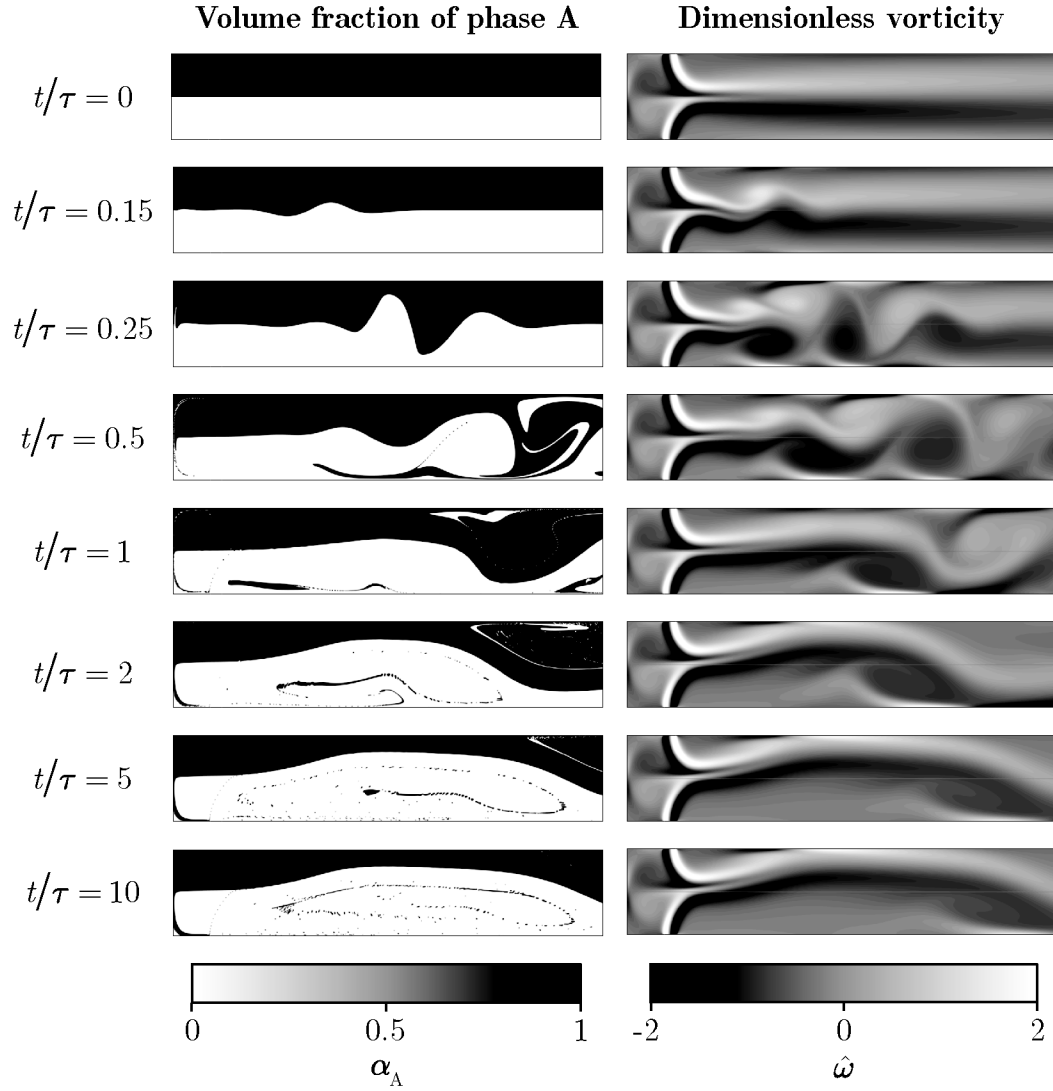


Figure 3.3. Maps of volume fraction of phase A and dimensionless vorticity in the mixing chamber as a function of the dimensionless time for $\text{Re}=100$.

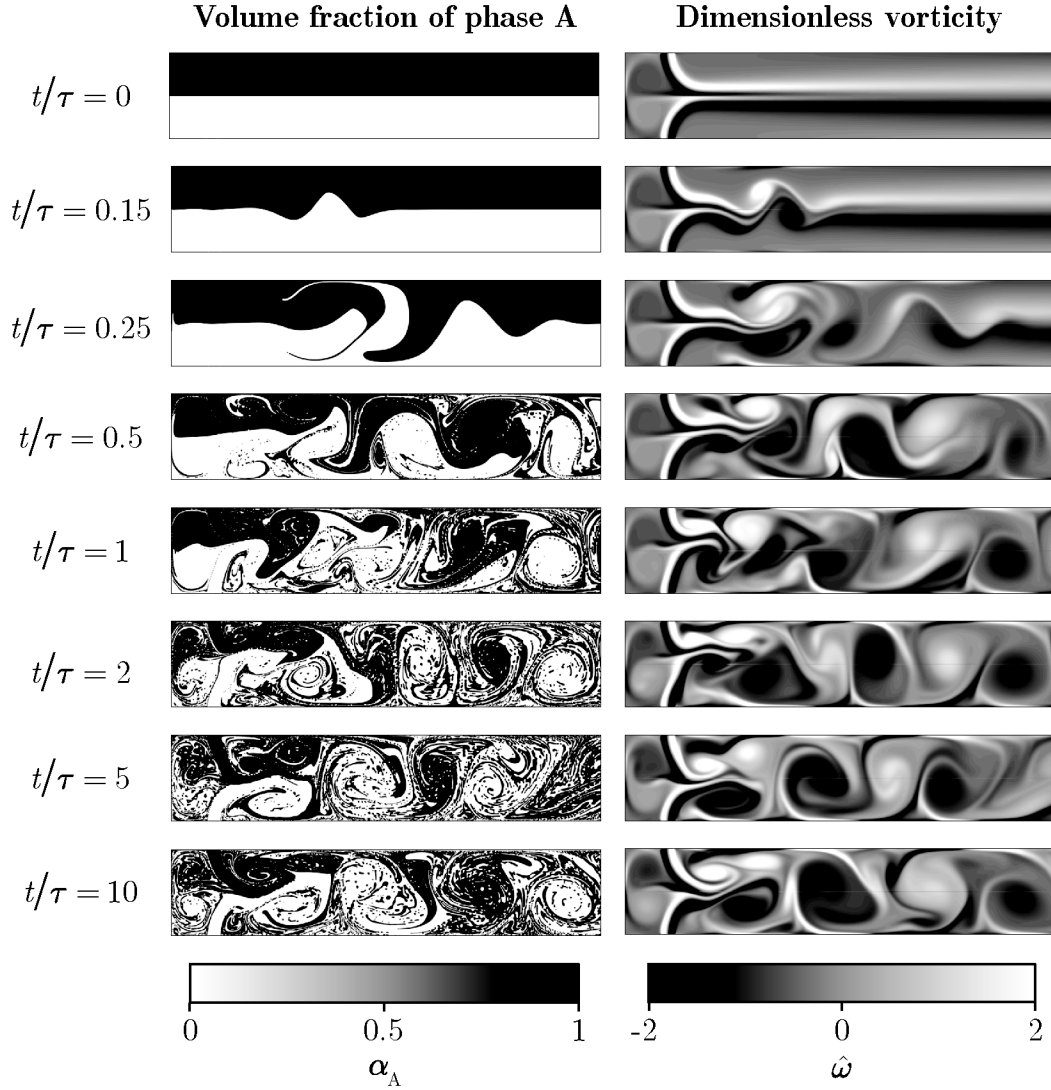


Figure 3.4. Maps of volume fraction of phase A and dimensionless vorticity in the mixing chamber as a function of the dimensionless time for $Re=300$.

Comparing the vorticity and volume fraction maps, it is seen that each vortex is mainly formed with fluid from one of the injectors because volume fraction values in the center of the vortices are always associated to the same sign of the vorticity values. Positive values of vorticity are associated with counter clockwise rotation of the vortex mainly formed by fluid from the right jet where fluid from the left injector engulfs. For negative values of vorticity, the opposite situation occurs.

Animations of the volume fraction and non-dimensional vorticity maps are included in the DVD (folder ~/Chapter3/Section1.4).

Although the analysis of the volume fraction maps from the dynamic simulation provides very good insight into the mixing mechanics, it lacks the quantification of the mixing degree. In Section 3.5 mixing is quantified for the two flow regimes.

3.5 Scale of Segregation Calculation

The mixing of the two phases was quantified in the line that defines the outlet boundary since it will represent the quality of the mixture that is discharged into the mold in RIM. To quantify the purely advective mixing of the two immiscible phases, a linear scale of segregation, s_L , as defined by Danckwerts (1952) was used. This quantity is analogous to the length scale of turbulence of the statistical theory of turbulence and is related to the average size of the clumps of each phase.

The scale of segregation is obtained from the coefficient of correlation function, $R(r)$, between pairs of volume fraction values at points in the outlet separated by a distance r ,

$$R(r) = \frac{\overline{(\alpha(x) - \bar{\alpha})(\alpha(x+r) - \bar{\alpha})}}{\overline{(\alpha(x) - \bar{\alpha})^2}} \quad (3.10)$$

where $\bar{\alpha}$ is the average value of the volume fraction of one of the phases at the outlet boundary, $x \in [0, D]$ is the position at the outlet line, and r is the distance that separates two points with volume fraction $\alpha(x)$ and $\alpha(x+r)$. The obtained curve, $R(r)$, is known as the *correlogram* of the mixture. For values of $r \rightarrow 0$ the probability of finding points in the domain with equal volume fractions is high and $R \rightarrow 1$. As r is increased, there is a value, ξ , for which no correlation between the points exist and $R(\xi) = 0$. The scale of segregation is obtained by integrating the correlogram from $r = 0$ to $r = \xi$ (Danckwerts, 1952; Tadmor and Gogos, 2006):

$$s_L = \int_0^\xi R(r) dr \quad (3.11)$$

For an uniform stripped pattern of volume fractions, the scale of segregation is equal to one quarter of the thickness of one layer (Tadmor and Gogos, 2006) and is related to the striation thickness, s , as

$$s = 4s_L. \quad (3.12)$$

The fastest way to calculate the correlation function of the volume fraction is using the Fast Fourier Transform (FFT) technique as proposed by Tucker III (1991), rather than Equation (3.10). The power spectrum $P(n)$ of the volume fractions is defined as

$$P(n) = \left| \int_{-\infty}^{\infty} (\alpha(x) - \bar{\alpha}) e^{-2\pi i n x} dx \right|^2 \quad (3.13)$$

where n is the wave number with dimensions of reciprocal length and $i = \sqrt{-1}$. The coefficient of correlation function is calculated from the inverse Fourier transform of $P(n)$,

$$R(r) = \int_{-\infty}^{\infty} P(n) e^{2\pi i n r} dn. \quad (3.14)$$

For the calculation of the scale of segregation, the history of the volume fraction profile at the outlet for $Re = 100$ and 300 was recorded and it is shown in Figure 3.5a and Figure 3.6a, respectively. From the observation of Figure 3.5a it is clear that for $Re = 100$, after the perturbation, the flow recovers to a steady state with the complete segregation of both phases. For $Re = 300$, Figure 3.6a shows the passage through the outlet of consecutive counter-rotating vortices: counterclockwise rotating vortices containing mainly phase A and clockwise vortices containing mainly phase B.

For each volume fraction recorded profile the power spectrum and coefficient of correlation function were calculated from the FFT and inverse FFT functions included in MATLAB® 2011b for Mac OS X, according to the technique proposed by Tucker III (1991). Figure 3.5b and Figure 3.6b show the scale of segregation as a

function of the dimensionless time for $Re = 100$ and 300 , respectively. The values of the scale of segregation obtained for $t/\tau = 0$ are in agreement with what should be obtained for a uniform stripped pattern, where the scale of segregation is equal to one quarter of the thickness of one layer $s_{L,0} = D/8 = 1.25$ mm (Tadmor and Gogos, 2006).

The calculated scale of segregation shows the role of the advective patterns present in the self-sustained chaotic regime on the decrease of the mixing length scales. For $Re = 100$ (Figure 3.5b) the scale of segregation tends to the value of the initial symmetric steady state, where the two phases injected by each jet are completely segregated. For $Re = 300$, as shown in Figure 3.6b, the scale of segregation reaches values one order of magnitude smaller than the initial segregated state. The scale of segregation significantly varies with time. These variations are associated to the passage of consecutive counter-rotating vortices leaving the mixing chamber. The vortices centers are zones with mostly fluid rotation and no deformation, which can be seen as traveling elliptical points in the flow presenting mainly one of the phases. The vortices borders can be seen as hyperbolic trajectories in the flow or zones with higher deformation potential and present small portions of each fluid.

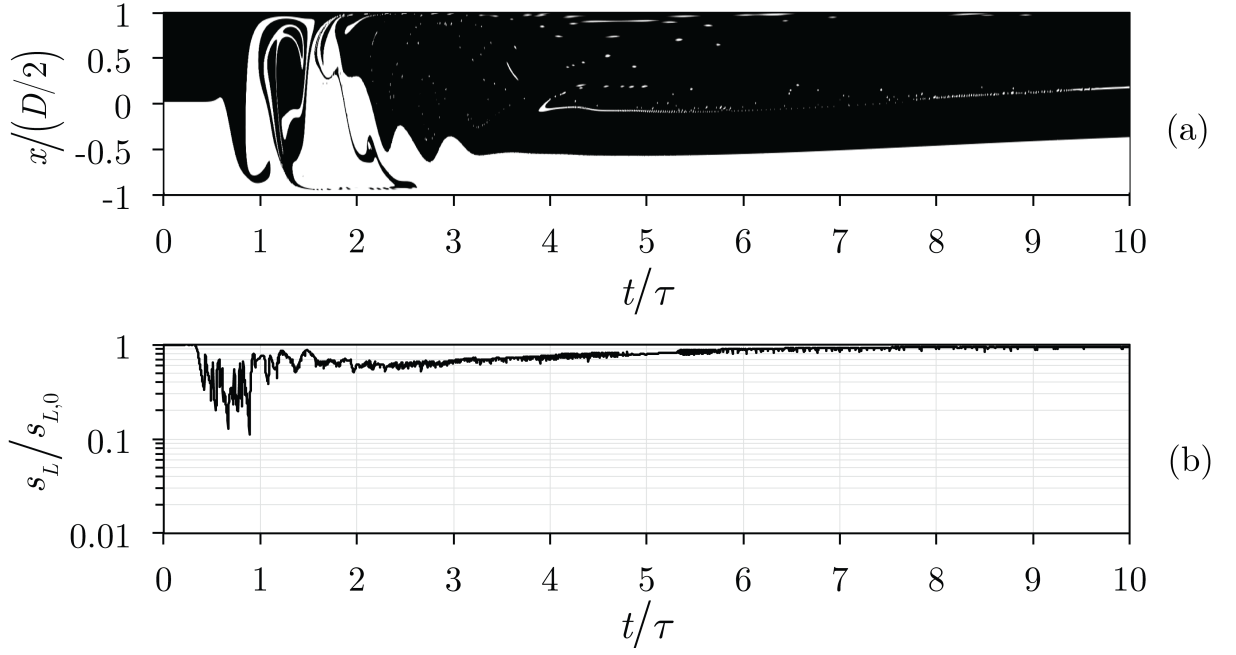


Figure 3.5. History of the volume fractions of phase A (black) and phase B (white) at the outlet of the mixing chamber (a) and the scale of segregation at the outlet of the chamber as a function of the dimensional time (b) for $Re=100$.

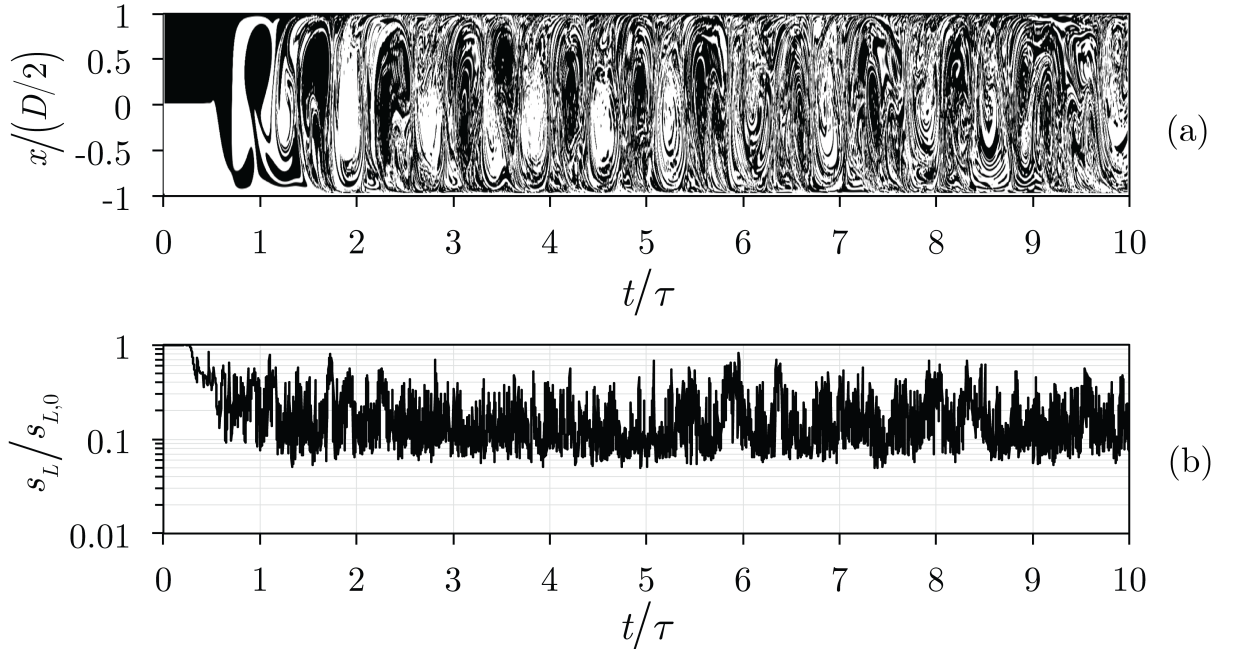


Figure 3.6. History of the volume fractions of phase A (black) and phase B (white) at the outlet of the mixing chamber (a) and the scale of segregation at the outlet of the chamber as a function of the dimensional time (b) for $Re=300$.

The cumulative distribution of the segregation scale was calculated for the flow regime at $Re = 300$ between $t = 3\tau$ and $t = 10\tau$ (see Figure 3.7). The segregation scale is non-dimensionalized by the segregation scale in the initial steady symmetric state, $s_{L,0} = 1.25 \text{ mm}$. Figure 3.7 shows that the segregation length scale in the mixture can vary of one order of magnitude at the outlet, which is explained by the different striation thinning present at the traveling vortices centers and at its borders. The minimum measured segregation length scale is of the order of the grid element size, which can be an indication, as already observed from the results in Figure 3.3 and Figure 3.4, that the chosen grid spacing of $50 \mu\text{m}$ is insufficient to describe completely the lamellar structure of the flow in the mixing regime for $Re = 300$. The influence of the grid element size on the description of the mixing scales is explored in Section 3.6.

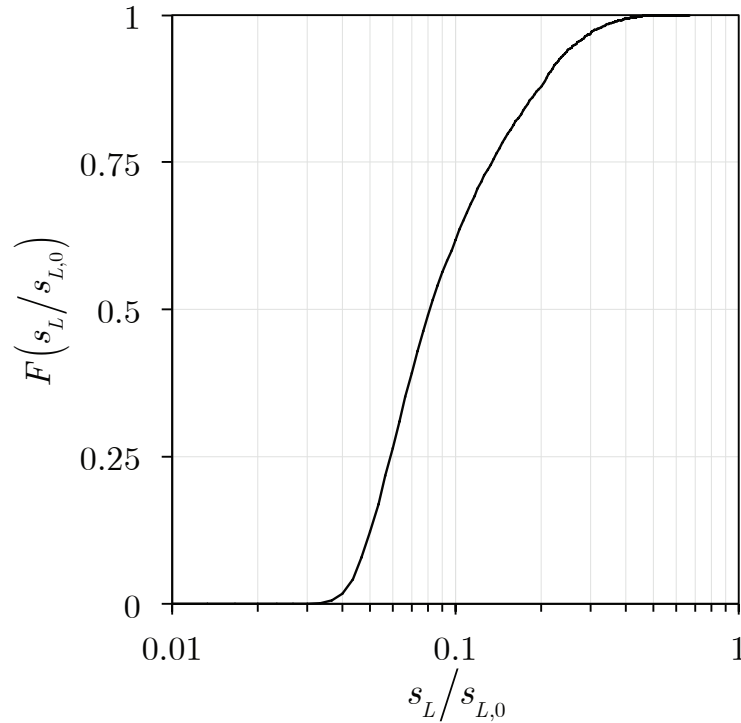


Figure 3.7. Cumulative distribution of the relative scale of segregation for a grid element size of $50 \mu\text{m}$.

3.6 Grid Element Size Study

In Section 3.5 the used grid element size was shown to be insufficient to describe all the lamellae thicknesses in the flow in the dynamic regime for $Re = 300$. To evaluate the influence of spatial discretization in the results, grids with different element sizes were tested: $100\ \mu\text{m}$, $50\ \mu\text{m}$ and $25\ \mu\text{m}$. Each time the grid element size is reduced to half, the number of elements in the grid increases 4 times: from 5×10^4 for $100\ \mu\text{m}$ to 8×10^5 elements for $25\ \mu\text{m}$. Figure 3.8 shows the distribution of the two phases (phase A in black, phase B in white) inside the mixing chamber for $Re = 300$ and a flow time $t/\tau = 10$, where it is clear that as spatial discretization increases, portions of the flow with smaller dimensions are observed and a more accurate description of lamellar structures is obtained.

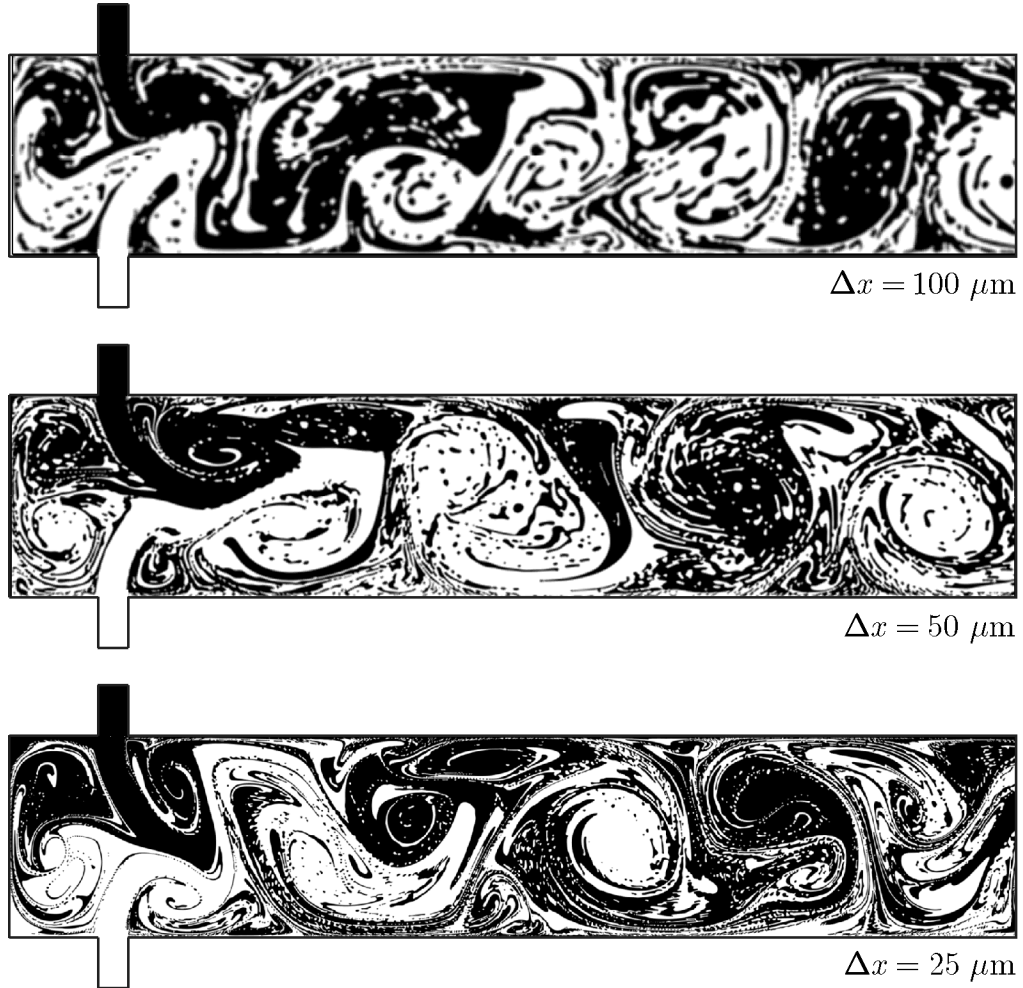


Figure 3.8. Distribution of phase A (black) and phase B (white) in the mixing chamber for $Re=300$ and a flow time equal to 10τ .

Figure 3.9 shows the cumulative distribution of the length segregation scale obtained for the different grid element sizes and measured at the outlet for every time step between $t = 3\tau$ and $t = 10\tau$. The segregation scale is non-dimensionalized by the segregation scale in the initial steady symmetric state, $s_{L,0} = 1.25$ mm. Figure 3.9 shows clearly the influence of the spatial discretization in the obtained mixing scales. For a computational grid with $\Delta x = 100$ μm , only segregation scales higher than $0.05s_{L,0}$ are observed, while with a grid 16 times denser ($\Delta x = 25$ μm) in half of the measured time interval, segregation scales lower than $0.05s_{L,0}$ are now measured. Figure 3.9 shows that even with a grid spacing of 25 μm the minimum detected scale of segregation in the mixture is still of the order of the grid element size. A spatial discretization $\Delta x = 25$ μm can still insufficient to simulate all the scales in the flow for $\text{Re} = 300$. Trying to overcome this limitation without further refinement of the entire simulation domain, a dynamically adaptive grid simulation model was developed as described in Section 3.7.

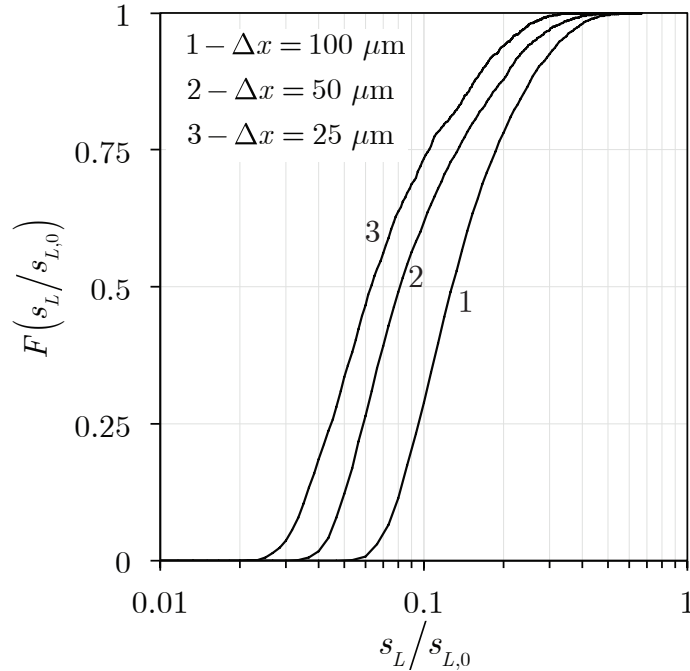


Figure 3.9. Cumulative distribution of the relative scale of segregation for different grid element sizes.

3.7 Adaptive Grid Scheme

The use of 2D simulation grids with uniform and sufficient discretization to simulate all length scales present in the lamellar flow in RIM would be extremely resource consuming. For illustration, the simulation of fluid laminae down to $1\text{ }\mu\text{m}$ of thickness from the 2D model would require a computational grid with 5×10^8 elements, corresponding to an extrapolated need of 2.5 TB of RAM during calculations. Additionally, to comply with the criterion of Equation (3.5), the time step size would have to be substantially decreased. As the grid refinement increases, the calculation time between iterations will be higher due to the increase of finite volumes in the mesh and a higher number of time iterations will be required to simulate the same flow time, increasing the total simulation time to unaffordable values.

Trying to overcome computational limitations, a dynamically adaptive mesh algorithm as described by Wang et al. (2004) and available in Ansys Fluent[®] was explored. The simulation started from a base grid with a spatial discretization of $50\text{ }\mu\text{m}$. The software was set to adapt the grid at each time step in the two phases' interface, refining it by levels in the region with higher volume fraction gradients. When locally a higher discretization was no longer needed, because no interface exists, the software was set to coarse the grid for the original element size. Figure 3.10 shows a schematic representation of the adaptive refinement algorithm for two levels of refinement where higher density of elements is achieved to better define the interface.

The incremental grid adaption by levels in regions with higher volume fraction gradients minimizes numerical problems due to interpolation of quantities between different sized elements during calculations. For an adaptive scheme with n_r levels of refinement, the minimum element size is equal to

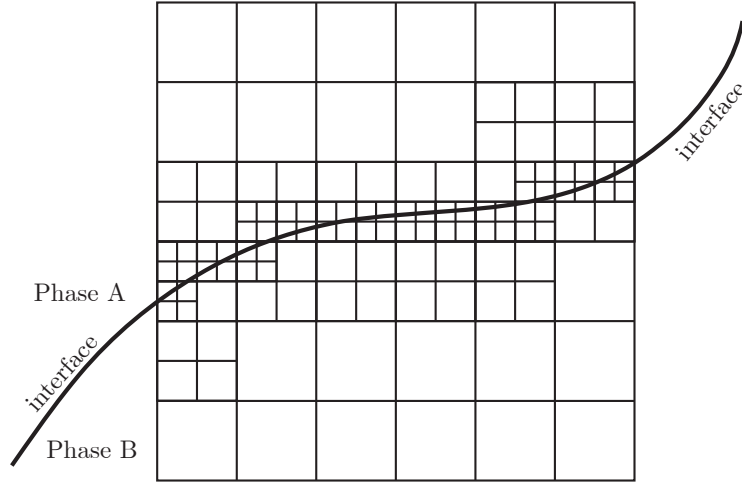


Figure 3.10. Representation of the grid refinement scheme in the interface between the two phases for 2 levels of refinement.

$$\left(\Delta x_{n_r}\right)_{\min} = \frac{\Delta x_0}{2^{n_r}}, \quad (3.15)$$

where Δx_0 is the element's size in the original grid. The adaptive refinement avoids the need of a denser uniform grid but does not bypass the need to use a lower time step in the simulations since it should be set accordingly to the smallest element size to avoid numerical instabilities in advection controlled flows. For an adaptive scheme with n_r levels of refinement, the time step is given by

$$\Delta t = \frac{\Delta t_0}{2^{n_r}} \quad (3.16)$$

where Δt_0 is the time step of the base grid, calculated from Equation (3.5).

The adaptive grid algorithm was applied to the simulation for $Re = 300$ with 3 levels of refinement, describing lamellar structures up to $6.25 \mu\text{m}$ of thickness. Figure 3.11 shows an illustrative detail of formed laminae in the flow with the adapted grid overlapped (in green) where it is possible to observe a greater density of elements in the zones where interface exists. Phase A is represented in black, while phase B is represented in white.

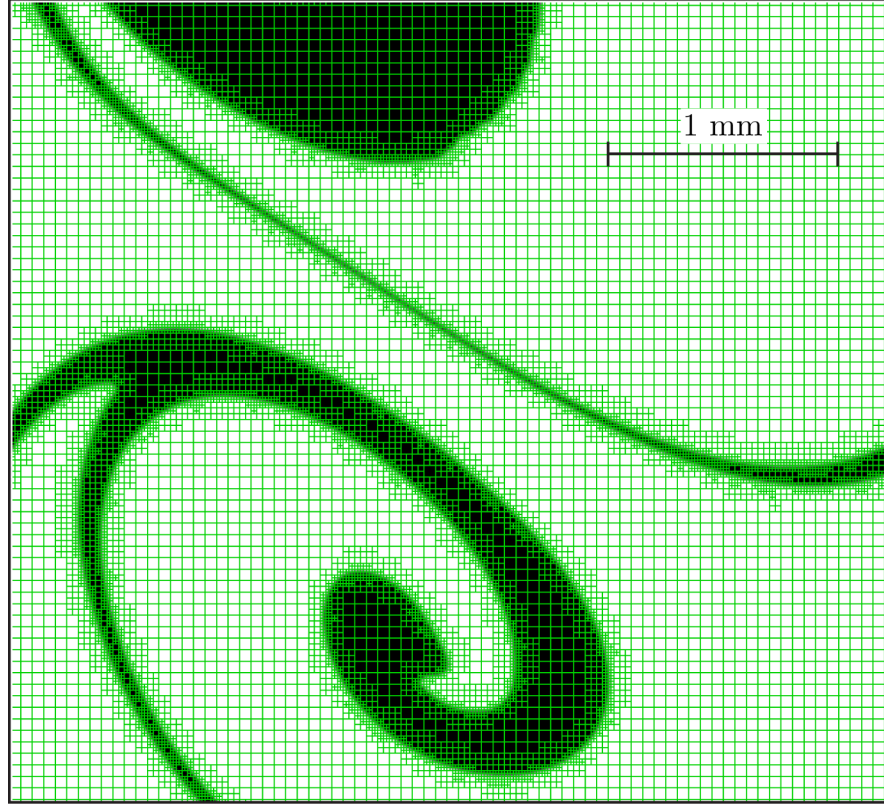


Figure 3.11. Illustrative detail of formed laminae of phase A (black) in the simulation domain with the adapted grid overlapped.

Figure 3.12 shows the distribution of the two phases inside the mixing chamber with time for $Re = 300$ with the adaptive grid algorithm. Comparing with the results of Section 3.6 with grid spacing of $100\ \mu\text{m}$, $50\ \mu\text{m}$ and $25\ \mu\text{m}$ for the same Reynolds number, Figure 3.12 shows a further reduction of striation scales in the flow and a better description of the lamellar structure of the flow in the mixing chamber. Break-up is still observed when laminae reach a thickness of $6.25\ \mu\text{m}$, implying that not all scales in the flow are yet described due to grid limitations and further reduction still occurs in the actual flow. Figure 3.13 shows a detail of the mixture for $t/\tau = 1.2$ in a square of the domain next to the exit for better visualization of the lamellar structure and the break-up of the laminae. An animation of the mixing of the two phases with the adaptive algorithm is included in the DVD from $t/\tau = 0$ to $t/\tau = 1$ (folder ~/Chapter3/Section1.7a).

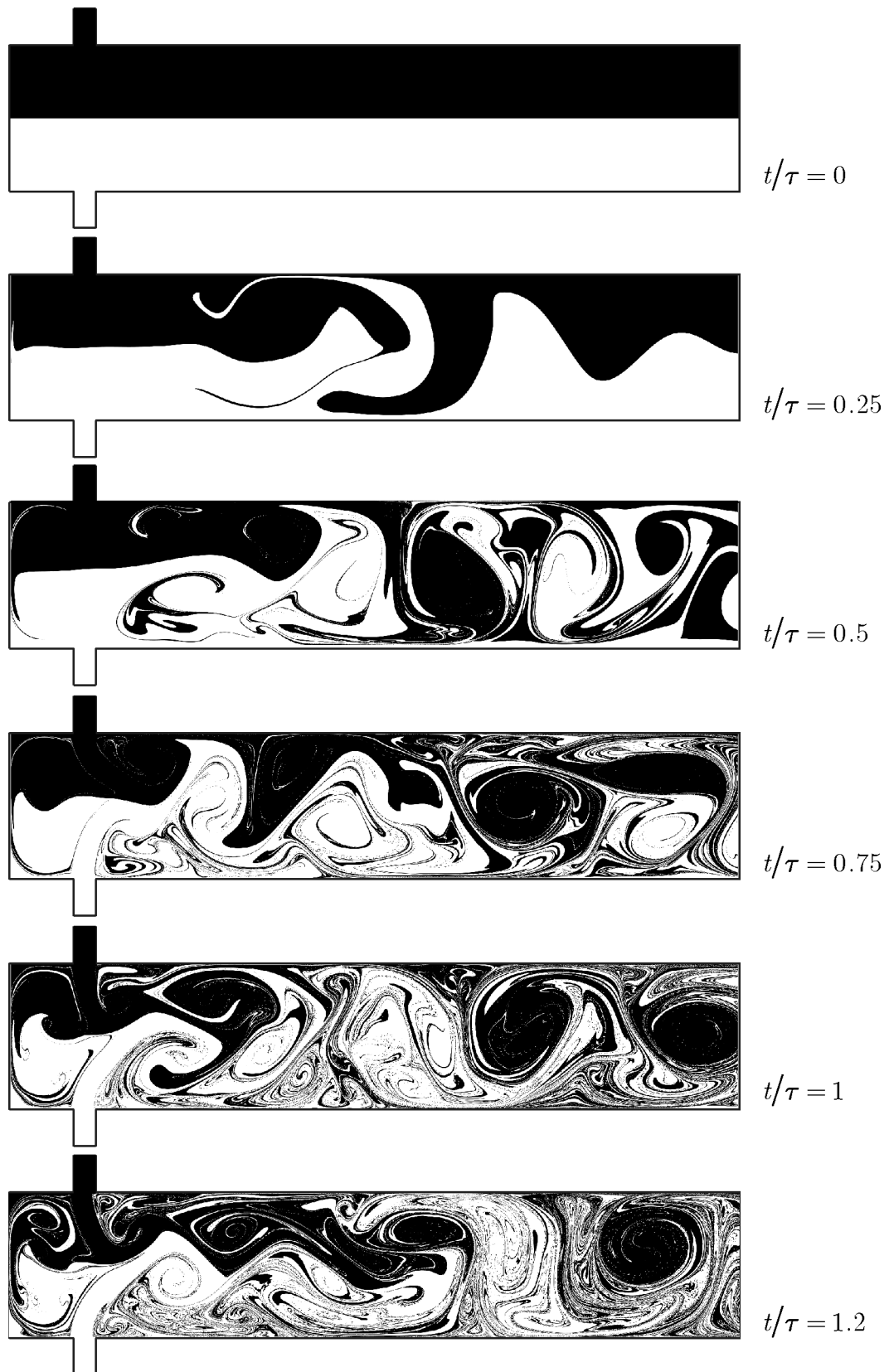


Figure 3.12. Distribution of the two phases inside the mixing chamber with time for $Re=300$ and an adaptive mesh algorithm with 3 levels of refinement.

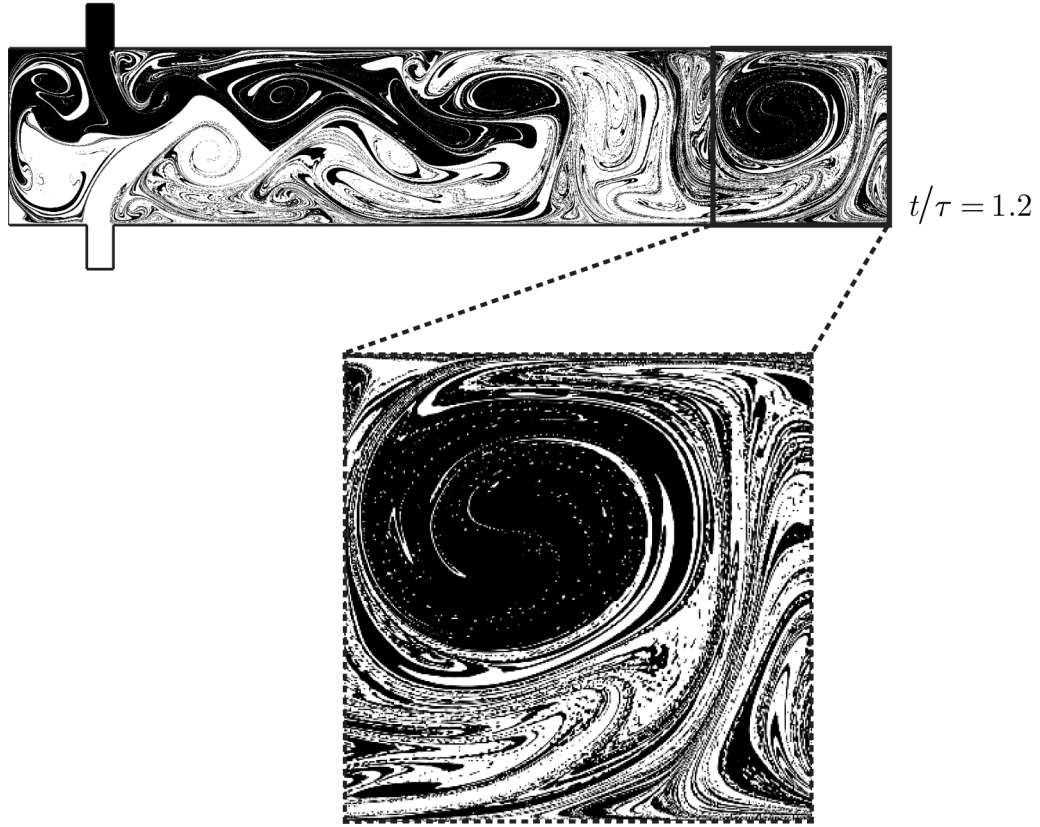


Figure 3.13. Detail of the distribution of the two phases inside the mixing chamber at the outlet for $Re=300$ with the adaptive mesh algorithm with 3 levels of refinement and for a flow time equal to $t/\tau=1.2$.

As the simulation evolves, the number of grid elements in the domain grows to describe thinner laminae. Figure 3.14 shows the number of grid elements of the adapted mesh, N_{elements} , relatively to the initial number, $N_{\text{elements},0} = 2 \times 10^5$, with the flow time. Figure 3.14 shows that, for the simulated flow time, the number of elements increases 20 times and seems to be reaching a maximum value, lower than the maximum corresponding to an uniform grid with a grid spacing of $6.25 \mu\text{m}$. This behavior seems to indicate that all the scales, up to the lengths of the minimum grid spacing, are being described inside the mixing chamber. The number of elements would only increase further if more adaption levels were allowed.

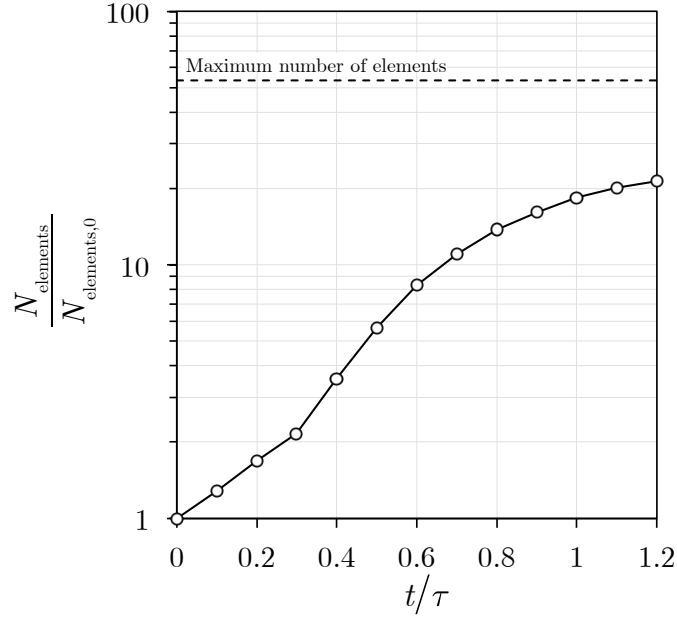


Figure 3.14. Number of grid elements as a function of the non-dimensional flow time.

As expected, with the fast increase of the number of grid elements, the calculation time between iterations also increases significantly. Figure 3.15 shows an exponential increase of the cumulative iteration time, $t_{\text{simulation}}$, with the simulated non-dimensional flow time. As an example, the simulation of a flow time equal to 1.2τ required two months of continuous parallel calculation (using 8 CPU cores). Due to the exponentially increasing computation time and the yet insufficient refinement to simulate all the mixing scales in the flow, the simulation was stopped for a flow time equal to 1.2τ .

The formation of thinner laminae in the flow was studied with 4 levels of grid refinement based on an original grid spacing of $50\ \mu\text{m}$ to obtain $\Delta x_{\text{min}} = 3.1\ \mu\text{m}$. For this analysis only the advection of a small square element of phase A with $0.75 \times 0.75\ \text{mm}$ placed close to the left injector was studied to avoid the nearly exponential increase of grid elements and simulation time to unaffordable values. The remaining chamber was filled with phase B. The flow field was considered equal to the dynamic flow state obtained for a time equal to 10τ in Section 3.4 for $\text{Re} = 300$ and a uniform grid with $\Delta x = 50\ \mu\text{m}$. Boundary conditions remained constant with time. Figure 3.16 shows a dynamic sequence of the advection of the small square

element of phase A (black) in phase B (white) inside the chamber up to 0.21τ , taking about 10 days of continuous calculation with 8 CPU cores. As time advances, the initial square is stretched and folded by the flow, increasing significantly its interfacial area in short flow times. The *horseshoe*-like folding of the black material is a clear indication of the chaotic nature of the flow in RIM.

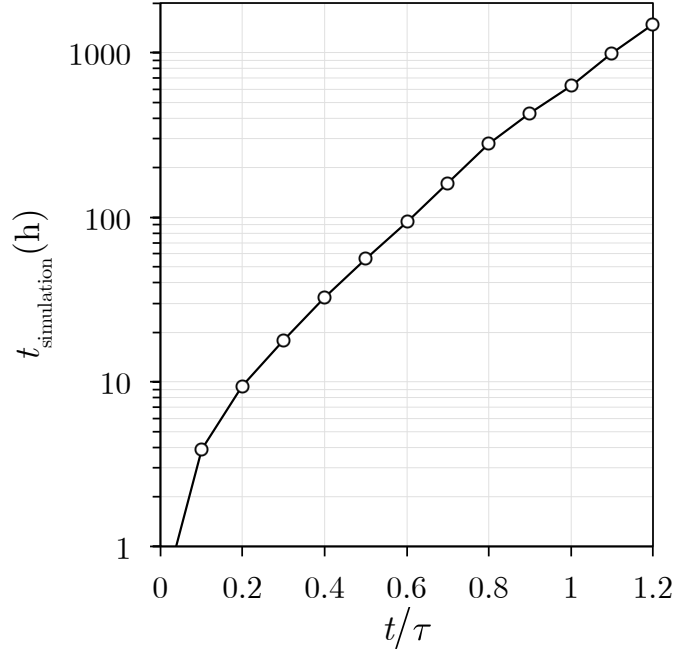


Figure 3.15. Simulation time as a function of the non-dimensional flow time.

Break-up still is observed when laminae reach a thickness of $3.1 \mu\text{m}$, implying that not all scales in the flow are yet described and further reduction would still occur. If deformation is continued, the thickness of some formed laminae uniting the advected blob of phase A are expected to become thin enough to be faded by molecular diffusion still inside the mixing chamber. As laminae reach thicknesses that are not physically realistic in real diffusive systems, diffusion will fade connecting filaments and create isolated zones of material. As also introduced in Chapter 2 from the Eulerian-Lagrangian approach to describe laminae length scales in RIM, these results seem to indicate an additional mixing mechanism of *rupturing* in impingement jets flow due to high material deformation.

An animation of the phase A square material advection in the mixing chamber is available in the DVD (folder `~/Chapter3/Section1.7b`).

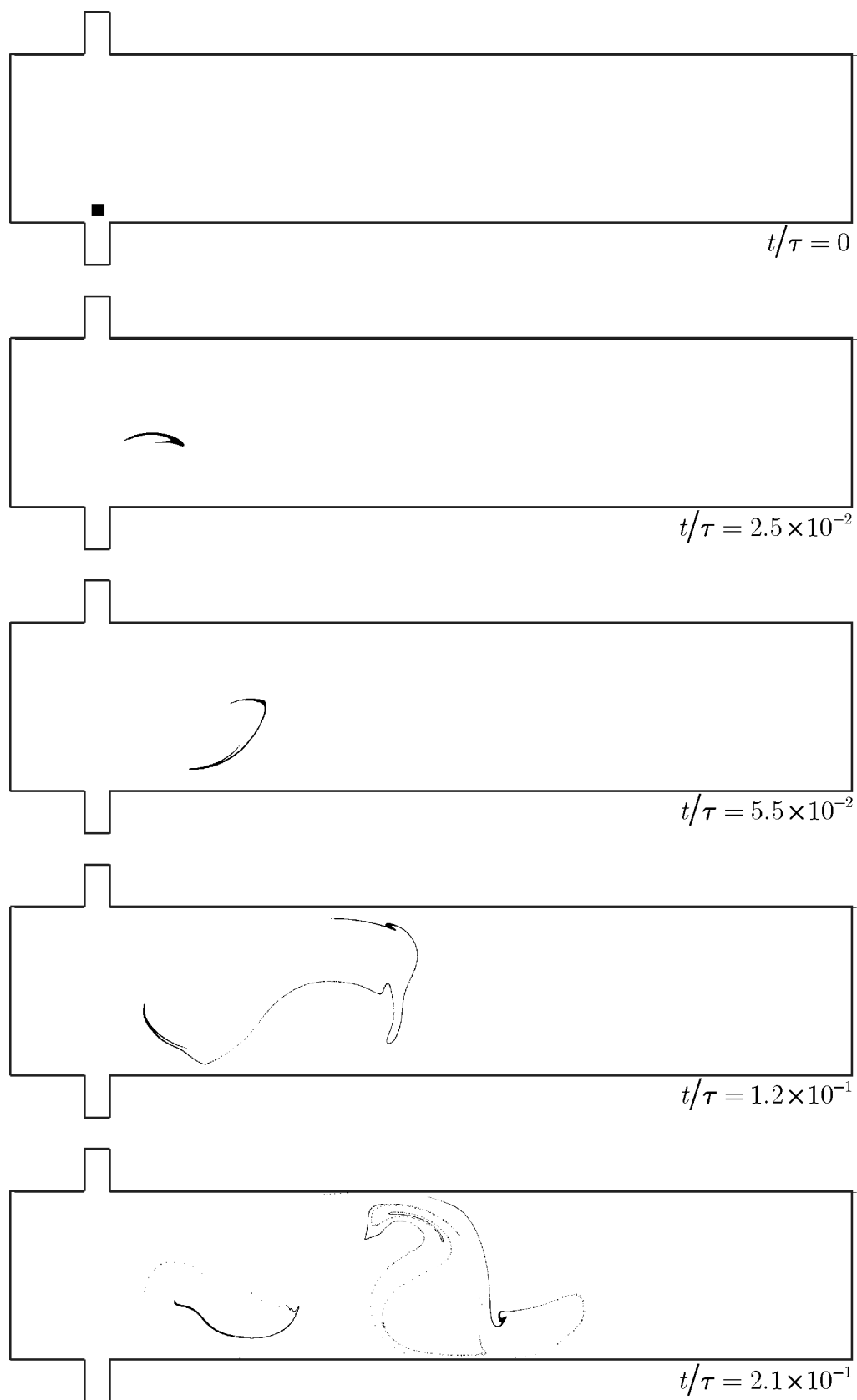


Figure 3.16. Distribution with time of an element on phase A (black) in phase B (white) inside the mixing chamber for an adaptive grid scheme with 4 levels of refinement.

3.8 Conclusions

In this chapter, a 2D model for the simulation of the lamellar structure of the flow in the RIM machine mixing chamber from an Eulerian-Eulerian approach is described. The model describes the advective mixing of two immiscible feeding fluids using the VOF model, without the effect of diffusion.

In agreement with previous 2D CFD simulations of Santos et al. (2005), results of the mixing of two immiscible phases in a RIM mixing chamber have shown that advective mixing occurs for a Reynolds number higher than a critical value placed between 100 and 300:

- For $Re = 100$ a non-symmetrical steady laminar flow, different from the symmetrical initial one, is obtained with complete segregation of the two phases.
- For $Re = 300$ a self-sustained chaotic laminar flow is obtained with formation of counter-rotating vortices that promote the creation of thinner laminae of each phase of smaller dimensions either by stretching and folding of the materials.

From the history of the volume fractions distribution at the outlet, an average length scale of segregation was calculated. It is notorious from the results the decrease of the scale of segregation from the steady laminar flow at $Re = 100$ to the dynamic chaotic laminar flow obtained for $Re = 300$, but the initial selected grids showed to be insufficient to have a complete description of the dependence of the scales of the flow on the Reynolds number.

To evaluate the further reduction of scales and overcome computational resources limitations, a simulation model coupled with an adaptive grid algorithm was developed. Results allowed to identify more thoroughly the mixing mechanisms occurring in the highly deforming flow in RIM and seem to indicate an additional mixing mechanism of *rupturing*.

The issue of a complete description of the mixing length scales in impingement jets mixing still remains unanswered from the results presented in this chapter. Even so, the advective mixing mechanisms visualized in the 2D model of the opposed jet flow in RIM are enough to explain the reduction of the laminae dimensions down to the value of $10\text{ }\mu\text{m}$ reported by Lee et al. (1980) for the complete polymerization to take place in the mold without diffusional limitations. The description of mixing length scales with the VOF model allows the actual visualization of laminar mixing mechanisms, rather than isotropic and homogeneous turbulence assumptions of previous works, which are more physically sounded as far as mixing in RIM is regarded.

From what is known, the use of a VOF model for the analysis of advective mixing in single-phase reactors without diffusion was made for the first time in this study. This approach adds to the two alternative methods to simulate lamellar flow structures already reported in the literature, the use of particle tracking (Roberts, 2010) and the extended map method (Kruijt et al., 2001). Despite the exponential increase in simulation time, the proposed model for the simulation of lamellar structures has the advantage to be easily applied to already existing commercial and open-source CFD codes.

In the future, it is expected that the increasing available computational power will make the simulation of all mixing length scales in a 3D RIM domain affordable from the proposed grid-adaptive Eulerian-Eulerian approach.

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4 Mixing Studies in Opposed Impinging Jets from PLIF Experiments

4.1 Introduction and State of the Art

Over the years, several authors have used flow visualization techniques with passive tracers to study mixing in Confined Impinging Jets (CIJ) mixers, either for Reaction Injection Molding or for other industrial applications. The main concern has been the study of the mixing mechanisms in opposed jets mixing and evaluate the effect of operational conditions or geometrical parameters on mixing quality.

First flow visualization experiments were made by Lee et al. (1980) who have taken high speed photographs of the flow in a transparent Plexiglas mixing chamber, relying in optical effects of small density variations of the injected solutions to observe the mixing patterns. Photographs of the flow were taken at Reynolds numbers $Re = 50, 90$ and 150 , with the Reynolds number defined for CIJ mixers as

$$Re = \frac{\rho du_{inj}}{\mu} \quad (4.1)$$

where d is the injector diameter, u_{inj} the injector mean velocity and ρ and μ the fluid density and viscosity, respectively. All images showed vortex motion in the flow, more intense around the impingement point and dying away downstream.

In the same year Tucker III and Suh (1980) used an ink tracer to detect flow structures in the impingement jets flow. Tucker III and Suh (1980) defined mixing quality from the variance of the photographed concentration fields and observed that poor mixing occurs for $\text{Re} < 150$. An improvement on mixing was observed as Re increased until a value around 1000 where no further improvements were detected. The used method did not allow verifying any impact of the jets' momentum ratio, ϕ_M , on the mixing quality, defined as

$$\phi_M = \frac{\rho_1 d_1^2 u_{\text{inj},1}^2}{\rho_2 d_2^2 u_{\text{inj},2}^2}, \quad (4.2)$$

where subscripts 1 and 2 refer to the left and right injector, respectively. Technological limitations at that time and the rapid movement of the formed vortices hindered the clear visualization of the flow from both methods of Lee et al. (1980) and Tucker III and Suh (1980) for $\text{Re} > 150$, a low value when compared with the range of Re used in the industry to obtain sufficient mixing for complete reaction in the mold.

Sandell et al. (1985) used a more advanced technique with a dye that fades with mixing to visualize the flow inside the chamber. The fading tracer prevents the clouding of the chamber and zones with higher accumulation of tracer and allowed the identification of regions of bad mixing in the flow. Sandell et al. (1985) have shown that mixing quality improves with the increase of Re with a decreasing rate of improvement over the range of 250-720.

Wood et al. (1991) used seeding particles to study the flow from the described paths, obtaining a steady flow regime up to $\text{Re} = 90$. The Reynolds number was incrementally increased to study the onset of flow oscillations. The instability of the

two impinging jets' interface was observed to grow as Re is increased. A stable impingement point position could not be obtained above $Re = 150$ in the experiments.

Johnson et al. (1996) and Johnson and Wood (2000) used several flow visualization techniques including the use of a dye, a fluorescent dye and tracer particles. For $Re < 90$ the flow has been observed to be stationary and unaffected even by sudden temporary perturbations. Above this value the impingement interface of the two opposed jets began to oscillate and the interface structure was no longer observable for $Re \sim 150$. For $Re > 150$ an unsteady three-dimensional flow structure develops. For $Re \sim 300$ mixing scales reduced to the point that the two impinging jets became the only visible flow structures.

Unger and Muzzio (1999) used the Planar Laser Induced Fluorescence (PLIF) technique to study the mixing performance of two impinging jet geometries: a standard symmetric geometry; and an asymmetric geometry where the impinging jets point to the close end of the chamber, with 8° deviation from the chamber axis. The mixing chamber's dimensions were typical of CIJ precipitators with a chamber diameter $D = 8.9$ cm and injectors' diameter $d = 1.3$ cm. The mixing quality was quantified by the calculation of the intensity of segregation. Results have shown that the intensity of segregation in the symmetric geometry drops off sharply from $Re = 150$ to $Re = 200$, decreases roughly linearly from $150 < Re < 200$ and levels off for $Re > 500$. For the asymmetric geometry, the intensity of segregation drops off sharply from $Re = 150$ to $Re = 300$. For $Re > 300$, the intensity of segregation in the asymmetric geometry is slightly lower comparing to the standard one.

Bierdel and Piesche (2001) and Schütz et al. (2005) used a colored tracer in one of the jets to study the formed mixing patterns in two flow regimes for $Re = 50$ where the flow is kept segregated and $Re = 150$ where mixing occurs.

Santos (2003) used the Particle Image Velocimetry (PIV) technique to visualize the flow field in a mixing chamber of a RIM machine, showing that oscillations in the

flow field occur at increasing intensity with Re prior to the actual transition to fully developed chaotic flows at $Re = 120$. From experimental results, Santos (2003) has proposed that the critical Reynolds number should not be identified with the onset of the oscillations but with a global and abrupt change of the flow field.

More recently, flow laser induced fluorescence visualization techniques have also been used by Hoffmann et al. (2006) (μ -LIF) and Sultan et al. (2012) (PLIF) to study different flow regimes and mixing quality in impingement mixing in T-shaped micromixers with rectangular cross-sections.

Table 4.1 presents a summary of experiments with flow visualization techniques for the study of mixing in Confined Impinging Jets mixers.

The present work adds to previous studies a qualitative and quantitative analysis from PLIF experiments of the mixing performance of a RIM machine's confined impinging jets mixer with usual dimensions found in the industry. The effect of the Reynolds number and jets' flow unbalancing on lamellar structure of the flow is studied here. The flow is studied in the range $50 \leq Re \leq 600$ and the parameter for the study of the jets' flow unbalancing is the kinetic energy rate ratio between the two jets, ϕ_K , defined as

$$\phi_K = \frac{\rho_1 d_1^2 u_{inj,1}^3}{\rho_2 d_2^2 u_{inj,2}^3}. \quad (4.3)$$

The parameter ϕ_K was chosen instead of the jets' momentum rate ratio, ϕ_M , (see Equation (4.2)) or the jets' mass flow rate ratio,

$$\phi_{FR} = \frac{\rho_1 d_1^2 u_{inj,1}}{\rho_2 d_2^2 u_{inj,2}}, \quad (4.4)$$

for being the key parameter to control mixing in impinging jets mixers as shown later in Chapter 5.

Table 4.1. Summary of experiments with flow visualization techniques for the study of mixing in Confined Impinging Jets mixers.

Source	Geometry	Mixing chamber dimension (mm)	Injectors dimension (mm)	Experimental technique	Reynolds number range	Critical Reynolds Number
Lee et al. (1980)	Cylindrical	3.18	1	Density variations	50-150	-
Tucker III and Suh (1980)	Cylindrical	22	9.5	Ink tracer	50-2000	>150
Sandell et al. (1985)	Cylindrical	52	9.5	Fading dye with mixing	250-720	-
Wood et al. (1991)	Cylindrical	25.4	2.38	Seeding particles	60-300	135
Johnson et al. (1996) and Johnson and Wood (2000)	Cylindrical, prismatic	20-25	2-4	Seeding particles, dye, fluorescent dye, tracer particles	90-300	Between 90 and 150
Unger and Muzzio (1999)	Cylindrical	890	13	PLIF	150-600	-
Bierdel and Piesche (2001) and Schütz et al. (2005)	Cylindrical	10	1	Colored tracer	50-150	Between 50 and 150
Hoffmann et al. (2006)	Prismatic	0.4	0.1-0.2	μ -LIF	100-375	Different flow regimes
Santos et al. (2008)	Cylindrical	10	1.5	PIV	100-500	120
Sultan et al. (2012)	Prismatic	2-6	0.5-2	PLIF	50-500	150-320
This work	Cylindrical	10	1.5	PLIF	50-600	110

The study of the parameter ϕ_K intends to evaluate the impact on mixing when the RIM machine has to be operated with monomers or pre-polymers formulations with different physical properties or stoichiometry.

The experimental set-up and flow visualization methods are described in Section 4.2. Results are presented and discussed in Section 4.4 where captured images of the top of the chamber reveal the flow structure and mixing mechanisms under different flow conditions. A qualitative analysis of mixing is made from the calculation of the intensity of segregation in the bottom region of the chamber, close to the outlet.

4.2 Methods and Experimental Setup

Planar Laser Induced Fluorescence is a flow visualization experimental technique in which a fluorescent dye marker is added to the flow. When excited by a laser sheet, dyed portions of the flow emit fluorescent light allowing the identification of flow structures. From the intensity of the emitted light this technique makes it possible to obtain the concentration scalar field in the flow system for both qualitative or quantitative analysis. This method is non intrusive and the required dye concentrations are low, thus not changing the fluid properties and disturbing the flow. The PLIF technique was used in this work to study the mixing of the fed streams into the RIM machine mixing chamber, one marked with the fluorescent dye and the other containing pure fluid.

The used experimental set-up is described in Section 4.2.1 and the image acquisition system in Section 4.2.2. The calibration of the method is described in Section 4.2.3.

4.2.1 Experimental Set-up

To allow internal visualization of the flow, a transparent mixing chamber was built from a single $150 \times 40 \times 47.5$ mm Plexiglas block (see Figure 4.1), which was cut and drilled to make the circular holes corresponding to the injectors and the mixing chamber. The block was drilled from side to side to make perfectly aligned injectors.

The injectors are placed at 5 mm from the top of the chamber making an angle of 180° between each other. The mixing chamber has a diameter of 10 mm and a height of 50 mm, equal to the dimensions of the mixing chamber used in the works of Teixeira (2000) and Santos (2003). The mixing chamber was connected to a cylindrical container with 15 cm diameter and 8 cm height, which replicates the operation of mold filling. Figure 4.2 shows a schematic drawing of the mixing chambers and the mold.

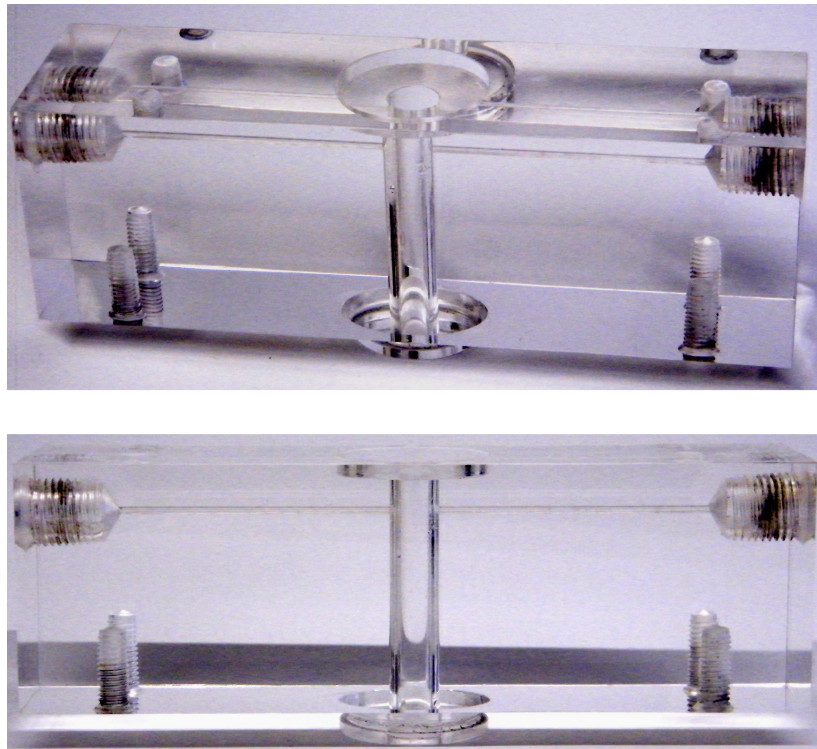


Figure 4.1. Photographic views of the transparent acrylic mixing chamber.

A hydraulic loop (see Figure 4.3) was assembled to transport the working fluid stored in two pressurized tanks until to the mixing chamber, one containing pure working fluid and the other the dyed fluid. The dyed fluid was injected through the left injector and the pure fluid through the right injector in the chamber. The fluorescent dye used in this work is rhodamine 6G. After passing the mixing chamber and the mold, the resulting mixture of dyed and pure working fluids was collected in a third container for proper elimination. Figure 4.3 shows a photograph of the complete hydraulic loop and its schematic representation.

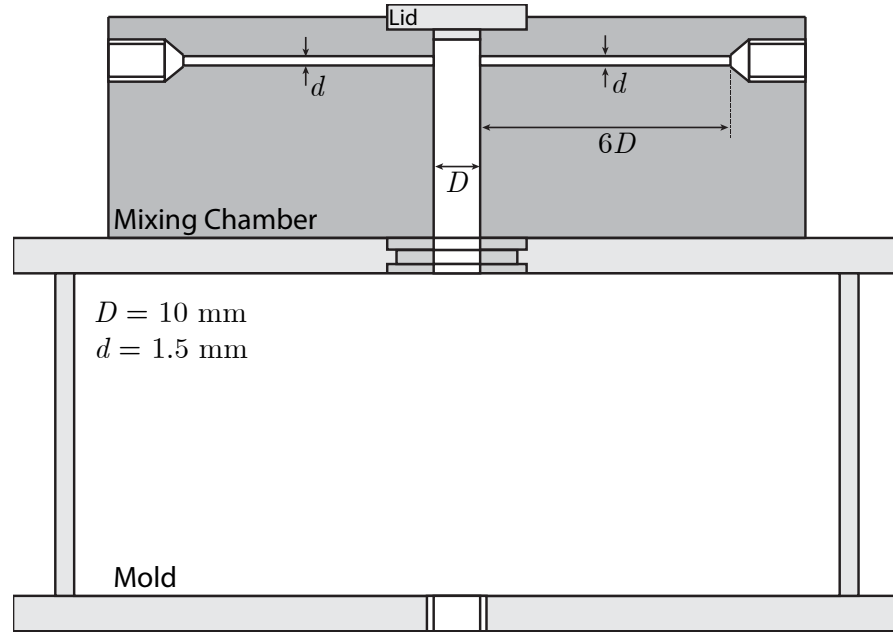
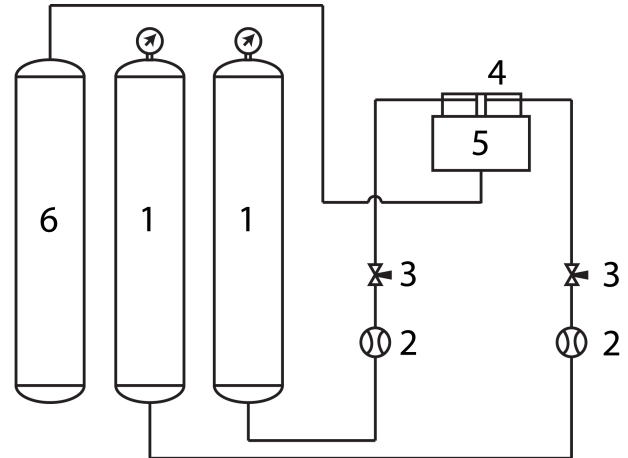
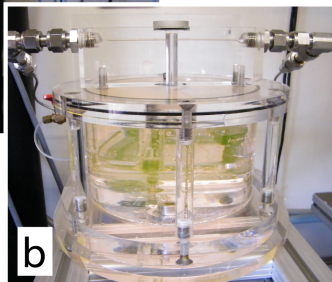


Figure 4.2. Schematic representation of the transparent mixing chambers and the mold.



- 1 - Pressurized container
- 2 - Coriolis mass flow rate meter
- 3 - Needle valve
- 4 - Acrylic mixing chamber
- 5 - Mold
- 6 - Collecting container

Figure 4.3. Image of the hydraulic loop (a-whole setup, b-detail of one transparent mixing chamber and mold) and its schematic representation.

The flow rate at each injector was controlled with needle valves placed in the hydraulic loop between the pressurized tanks and the mixing chamber inlets. The flow rate at each injector was adjusted by the reading in two ROTA-Yokogawa Coriolis mass flow meters (model RCCS33 M01A1SH), allowing to set flow rates with a precision of ± 0.05 g/s in the range from 1.2 g/s to 14.1 g/s of this work, corresponding to a maximum relative error of 4%. Pressurized tanks were used for fluid injection in order to eliminate flow oscillations often originated by pumps. In each experiment, fluid temperature and density were measured with the Coriolis mass flow meters for determination of Re and ϕ_K .

The working fluid used in the experiment was a solution of 70% (w/w) of glycerin in water. The dependence of the solution viscosity on temperature in the range from 15 °C to 35 °C was determined with a cone and plate rheometer (Physica UDS 200) (Figure 4.4).

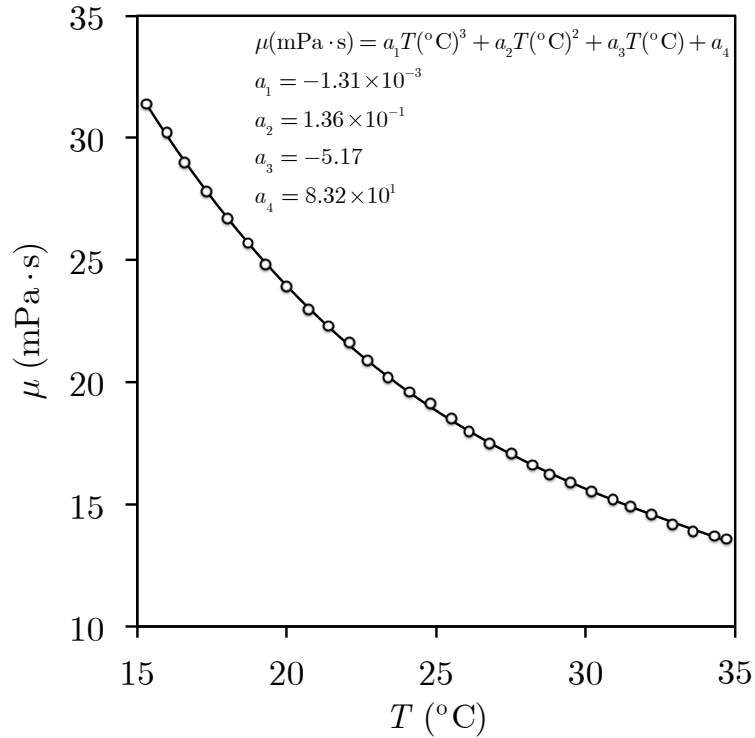


Figure 4.4. Viscosity of glycerin-water solutions (70% w/w) as a function of temperature and fitting function.

4.2.2 Laser System and Image Acquisition

Two-dimensional concentration fields inside the mixing chamber geometry were obtained in a plane resulting from an axial cut of the chamber through the injectors from PLIF. Figure 4.5 shows a photograph of the PLIF experimental set-up and Figure 4.6 shows a schematic representation of the complete PLIF experimental system. The transparent mixing chamber was illuminated with a laser sheet, cutting the chamber axially through the injectors, making the flow only visible in a thin slice, with 1 mm of thickness, of the mixing chamber volume by fluorescence emission. The laser source used is a Nd:YAG double-head pulsed laser from Litron Lasers[®] (Nano L 50-100, which emits 4 ns pulsed beams with maximum energy of 400 mJ/pulse at a maximum rate of 100 Hz and a wavelength of 532 nm. The laser beams were transported to the top of the mixing chamber with a TSI light arm (Model 610015), a telescopic and articulated 1.8 m hollow tube system with internal reflecting mirrors. At the input port of the light arm a collimated lens with a 10 m focal length was placed to control the divergence and prevent clipping of the beams. After traveling a distance corresponding to the light arm length, the laser beams were focused with a 500 mm spherical lens and spread in a sheet with a -15 mm cylindrical lens. The extremity of the laser arm was placed at the top of the mixing chamber at a distance of 500 mm obtaining a laser sheet with approximately 1 mm of thickness.

Three hundred pairs of laser-synchronized frames of the illuminated flow were captured with a CCD camera for each operational condition. The used camera is a TSI Model 630157 with the ability of capturing 1192×1600 pixels images with a 16 bits scale of shades of grey at maximum frame rate of 30 Hz. The frames captured with the CCD camera are transferred to the computer through a frame grabber (TSI model 600067).

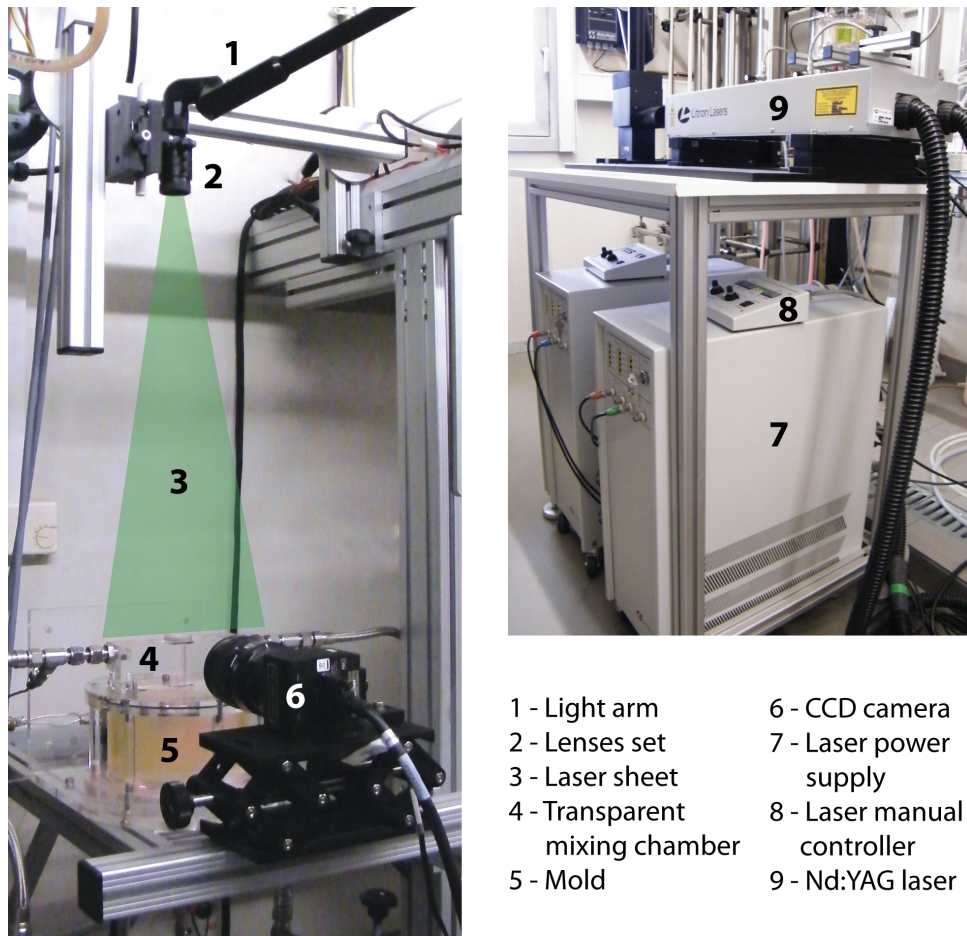


Figure 4.5. Photograph of the PLIF experimental setup.

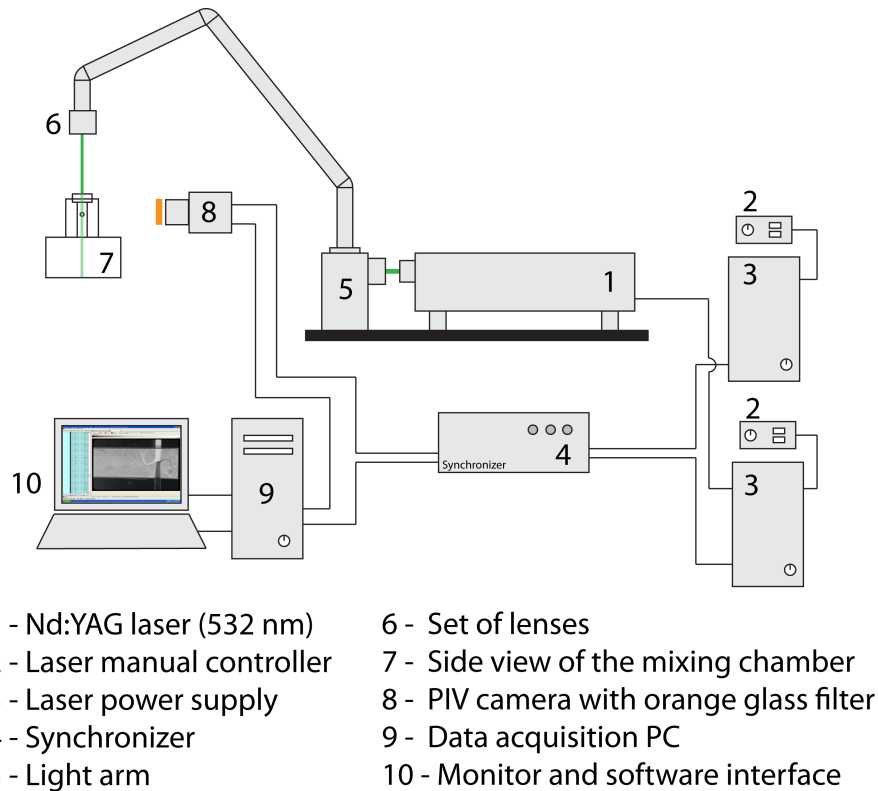


Figure 4.6. Schematic representation of the PLIF experimental setup.

Rhodamine 6G has a maximum of absorption of light close to 526 nm and emits fluorescence in a wavelength range around a maximum value of 564 nm (Magde et al., 2002). Since the emitted fluorescence light is red shifted, an orange glass filter (545 nm) was adapted to the camera lens to filter possible reflections of the green laser light. Apart from the laser light, no other light source was present in the set-up room during the experiments.

In order to capture approximately the same flow time relatively to the chamber residence time in all the experiments, the rate of acquisition of the 300 pairs of images was set accordingly to the Reynolds number. Considering the maximum value of $Re = 600$ for the experiments, the laser frequency, f_{laser} , was set to the closest allowed frequency by the PLIF system according to

$$f_{\text{laser}}(Re) = \frac{Re}{600} f_{\text{laser,max}} \quad (4.5)$$

where $f_{\text{laser,max}} = 30$ Hz. The 300 images of the flow correspond in all experiments to a flow time $t \approx 60\tau$, where the mean residence time, τ , in the chamber is given by

$$\tau = \frac{HD^2}{2d\nu} \frac{1}{Re}. \quad (4.6)$$

A software default value of exposure time of $405 \mu s$ for the image capturing was used since it has shown to be sufficiently small to *freeze* the flow in the images. A TSI synchronizer (model 610035) was the responsible device for the system time management. Image acquisition was done in a Dell® Precision 690 PC with a dual core Intel® Xeon® CPU at 2.33 GHz and 2 GB of RAM with TSI Insight 3G™ software (version 9.0).

4.2.3 Calibration

A concentration of rhodamine 6G equal to $C_{\text{Rhodamine6G}} = 0.4 \text{ g/l}$ was used in the experiments. This concentration was previously tested to verify if a linear response between pixel light intensity and concentration was always obtained. Seventeen solutions of 70% (w/w) glycerin in water with different concentrations of rhodamine 6G in were illuminated with the laser sheet. Images of the illuminated solutions were taken and the pixel light intensity, I , was plotted against the rhodamine 6G concentration (Figure 4.7). When the rhodamine 6G concentration in the solution is zero, no fluorescent light is emitted and, since the orange glass filter was adapted to the camera lens, the captured images showed a pixel intensity of $I \approx 0$. As the concentration is increased, the pixel intensity increases always linearly until the maximum concentration used in this experiment was reached. A linear response of fluorescence with concentration allows a direct transformation of the gray scale field into a field of volume fraction of dyed fluid. The rhodamine dyed fluid volume fraction spatial distribution, $\alpha(\mathbf{x})$, is obtained from the pixels' gray scale intensity in the PLIF images as

$$\alpha(\mathbf{x}) = \frac{I(\mathbf{x}) - \min_{\Omega}(I)}{\max_{\Omega}(I) - \min_{\Omega}(I)} \quad (4.7)$$

over the whole 2D captured domain $\mathbf{x} \in \Omega$.

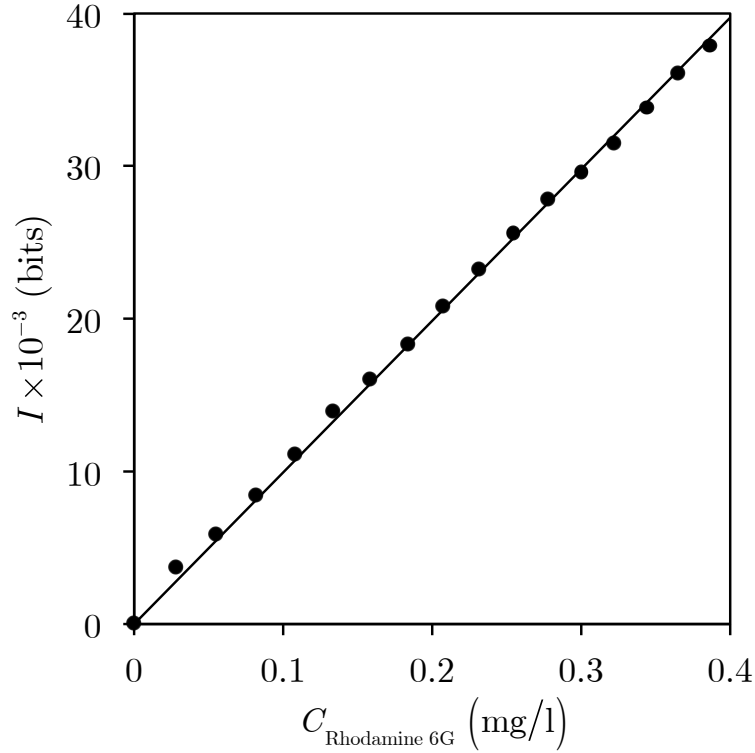


Figure 4.7. Pixel light intensity as a function of rhodamine 6G concentration in a glycerin-water solution (70% w/w).

4.3 Experiments

Different sets of experiments were performed to evaluate both the impact of Reynolds number and the jets' unbalancing on mixing. Images were captured from the top and bottom parts of the chamber to study the lamellar structure of the flow in the two different flow zones observed in Chapter 2: impingement mixing in the top of the chamber and parallel flow next to the exit. Images of the flow for the different conditions were captured at the top of the chamber to study qualitatively the mixing mechanisms and mixing degree. In the bottom of the chamber, images were captured to quantify the extent of mixing since it will be representative of the quality of the mixing that is discharged into the mold in RIM operation. Figure 4.8 shows a photograph of the transparent mixing chamber with the indication of the two captured zones. The images of the lamellar structure in Figure 4.8 correspond to the observed for $Re = 111$.

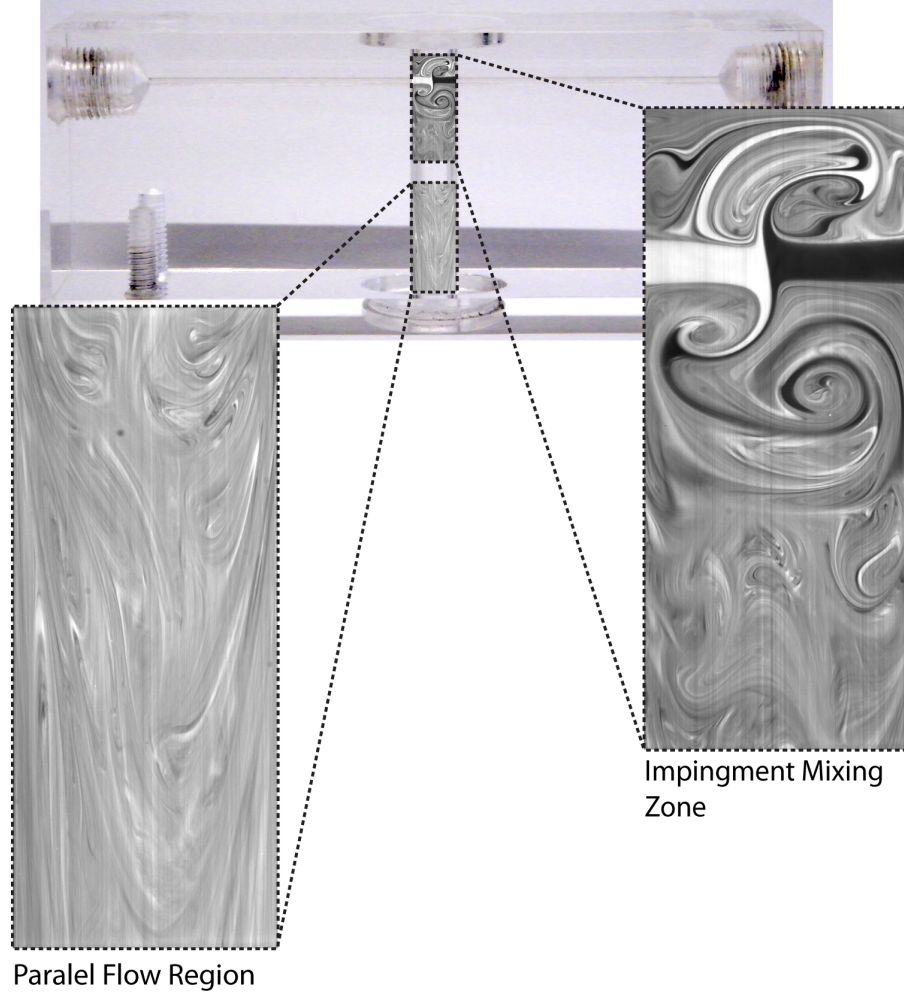


Figure 4.8. Indication of the captured domain for the qualitative analysis of the mixing mechanisms and degree.

Table 4.2 and Table 4.3 resume the list of the experimental conditions to study the effect of the Reynolds number on mixing in the top and bottom parts of the mixing chamber, respectively. Table 4.4 and Table 4.5 resume the list of the experimental conditions to study the effect of ϕ_K on mixing in the top and bottom parts of the mixing chamber, respectively. Since the chamber has jets with equal diameter and the two streams have the same physical properties, Equation (4.3) simplifies here to

$$\phi_K = \frac{u_{\text{inj},1}^3}{u_{\text{inj},2}^3}. \quad (4.8)$$

4. Mixing Studies in Opposed Impinging Jets from PLIF Experiments

Table 4.2. Experimental conditions to study the effect of Re on mixing in the top of the mixing chamber.

Frame	$\dot{m}$ (g/s)	T ($^{\circ}\text{C}$)	$\rho \times 10^{-3}$ (kg/m ³)	μ (mPa · s)	Re	f_{laser} (Hz)
1 - 300	1.2	23.8	1.18	19.8	51	2.5
301 - 600	2.4	23.8	1.18	19.8	103	5
601 - 900	2.4	24.2	1.18	19.5	104	6
901 - 1200	2.6	23.8	1.18	19.8	111	6
1201 - 1500	2.8	23.8	1.18	19.8	120	6
1501 - 1800	3.5	23.8	1.18	19.8	150	7.5
1801 - 2100	4.7	23.8	1.18	19.8	201	10
2101 - 2400	7.1	23.8	1.18	19.8	304	15
2401 - 2700	9.4	23.8	1.18	19.8	402	20
2701 - 3000	11.8	23.8	1.18	19.8	505	25
3001 - 3300	14.1	24.3	1.18	19.4	617	30

Table 4.3. Experimental conditions to study the effect of Re on mixing in the bottom of the mixing chamber.

Frame	$\dot{m}$ (g/s)	T ($^{\circ}\text{C}$)	$\rho \times 10^{-3}$ (kg/m ³)	μ (mPa · s)	Re	f_{laser} (Hz)
1 - 300	1.2	24.3	1.18	19.4	52	2.5
301 - 600	2.4	24.2	1.18	19.5	104	5
601 - 900	2.5	23.9	1.18	19.8	107	6
901 - 1200	2.6	24.3	1.18	19.4	114	6
1201 - 1500	2.8	23.9	1.18	19.8	120	6
1501 - 1800	3.5	23.9	1.18	19.8	150	7.5
1801 - 2100	4.7	24.1	1.18	19.6	204	10
2101 - 2400	7.1	24.1	1.18	19.6	308	15
2401 - 2700	9.4	24.1	1.18	19.6	407	20
2701 - 3000	11.8	24.1	1.18	19.6	511	25
3001 - 3300	14.1	24.0	1.18	19.7	608	30

Table 4.4. Experimental conditions to study the effect of the jets' kinetic energy rate ratio on mixing in the top of the mixing chamber.

Frame	$\dot{m}_1$ (g/s)	$\dot{m}_2$ (g/s)	ϕ_K	ϕ_M	ϕ_{FR}	T ($^{\circ}\text{C}$)	$\rho \times 10^{-3}$ (kg/m ³)	μ (mPa · s)	f_{laser} (Hz)
1 - 300	7.1	6.2	1.50	1.31	1.15	25.3	1.18	18.6	15
301 - 600	7.1	6.8	1.14	1.09	1.04	25.3	1.18	18.6	15
601 - 900	7.1	7.3	0.92	0.95	0.97	25.3	1.18	18.6	15
901 - 1200	7.1	8.1	0.67	0.77	0.88	25.3	1.18	18.6	15

Table 4.5. Experimental conditions to study the effect of the jets' kinetic energy rate ratio on mixing in the bottom of the mixing chamber.

Frame	$\dot{m}_1$ (g/s)	$\dot{m}_2$ (g/s)	ϕ_K	ϕ_M	ϕ_{FR}	T (°C)	$\rho \times 10^{-3}$ (kg/m ³)	μ (mPa · s)	f_{laser} (Hz)
1 - 300	7.1	6.2	1.50	1.31	1.15	25.0	1.18	18.8	15
301 - 600	7.1	6.8	1.14	1.09	1.04	25.0	1.18	18.8	15
601 - 900	7.1	7.3	0.92	0.95	0.97	25.0	1.18	18.8	15
901 - 1200	7.1	8.1	0.67	0.77	0.88	25.0	1.18	18.8	15

4.4 Results

Section 4.4 summarizes the results of the qualitative and quantitative analysis to the effect on mixing in RIM of the Reynolds number (Section 4.4.1) and the jet's flow unbalancing (Section 4.4.2).

4.4.1 Reynolds Number Effect

4.4.1.1 Flow Visualization

With the experimental method and set-up described in Section 4.2, 2D concentration fields were captured for the study the effect of Re in the mixing degree with the conditions of Table 4.2. Figure 4.9 to Figure 4.19 show randomly chosen images from the 300 captured in each experiment of the instantaneous concentration field at the top of the mixing chamber, as indicated in Figure 4.8, to illustrate the dynamic nature of the mixing process. For better visualization, animations of all the captured images are included in the DVD accompanying this work (folder ~/Chapter4/Section4.4.1/Top). In the PLIF images, rhodamine 6G dyed feeding fluid is seen as white, while pure glycerin solution is seen as black.

Some vertical striations are visible in the captured images resulting from shadowing effects of the flow and from physical striations present in the acrylic material of the chamber. These striations could have been removed by Fast Fourier Transform (FFT) imaging techniques by ignoring all striation frequencies in the vertical

direction (inverse of the distance separating them) smaller than the observed vertical striations. However, this technique induces additional blurring to the images, fading striations oriented with the vertical direction. For the described reasons, images were analyzed as they were captured and the vertical striations were considered as second order experimental noise that affects equally all the experiments.

For $Re = 51$ (Figure 4.9) and $Re = 103$ (Figure 4.10) the flow remains mainly segregated. The right side of the chamber is continuously filled with dyed fluid (seen as white), while the left side of the chamber with pure working fluid (seen as black). The only interfacial area created between the two streams is equal to the resulting vertical plane normal to the injectors axis cutting the chamber in two.

As Re is slightly increased for $Re = 104$ (see Figure 4.11), the sharp onset of a dynamic flow regime is observed. For $Re = 104$ the two impinging jets' interface is disturbed and the flow develops to a self-sustained oscillatory regime characterized by the formation of detaching vortices downstream of the inlet. The term *self-sustained* is used here to reinforce that this dynamic state is achieved and maintained with constant inlet conditions in time. Material exchange between the two sides of the chamber and a generation of smaller laminae scales is observed due to the formed detaching vortices but mixing is highly localized in the zone downstream the injectors. This flow regime was also identified in 3D Computational Fluid Dynamics (CFD) simulations of the flow in Chapter 2.

If Re is continuously increased to values higher than 104, the vortices formed in the chamber downstream of the impingement point, start to appear earlier, that is closer to the impingement point, until the point when they start to interact with the impinging jets.

At this point ($Re \geq 104$) the flow develops to a self-sustained chaotic regime with strong mixing dynamics, forming a lamellar mixing structure by the continuous folding and stretching of the two injected streams (see Figure 4.12 to Figure 4.19). Mixing at this flow regime is global, occurring in the whole chamber top volume.

Although the inlet flow conditions are maintained constant in time, the jets' impingement point is observed to change its position in the chamber, both in the normal and tangential directions to the captured 2D plane, when the self-sustained chaotic laminar flow regime is achieved.

As Re is increased, thinner lamellar structures are observed in the images of the flow due to the increase of the degree of mixing. As also observed by Santos (2003) from CFD simulations with transport of species, Figure 4.9 to Figure 4.20 show that the greatest change on laminae size with the increase of Re is observed until $Re = 300$. From $Re = 300$ until $Re = 600$, a reduction of laminae thicknesses with Re is still observed but in a smaller extent. When $Re \sim 600$, very few mixing structures of pure feeding fluids are detected in the captured images, with the only exception of the two impinging jets still visible in the almost homogeneous remaining concentration field.

The observed laminae thicknesses and the continued reduction of striation thicknesses with the increase of Re contrast with models for the prediction of mixing length scales in impingement flow based on the statistical theory of turbulent diffusion. In the model of Tucker III and Suh (1980) an average striation thickness in impingement jets mixing is determined by the smallest hydrodynamic length scale in the flow, the Kolmogorov scale. The models of Baldyga and Bourne (1983) and May (1996), also based on the statistical theory of turbulent diffusion do not predict dependence of mixing length scales with Re .

Figure 4.20 shows zoomed views of a 1×1 mm square portion of the captured domain for $Re = 111$, 304 and 617. The zones were chosen in the region downstream the injectors and in zones where the widest range of laminae scales could be observed. The zoomed images were processed to increase sharpness and contrast.

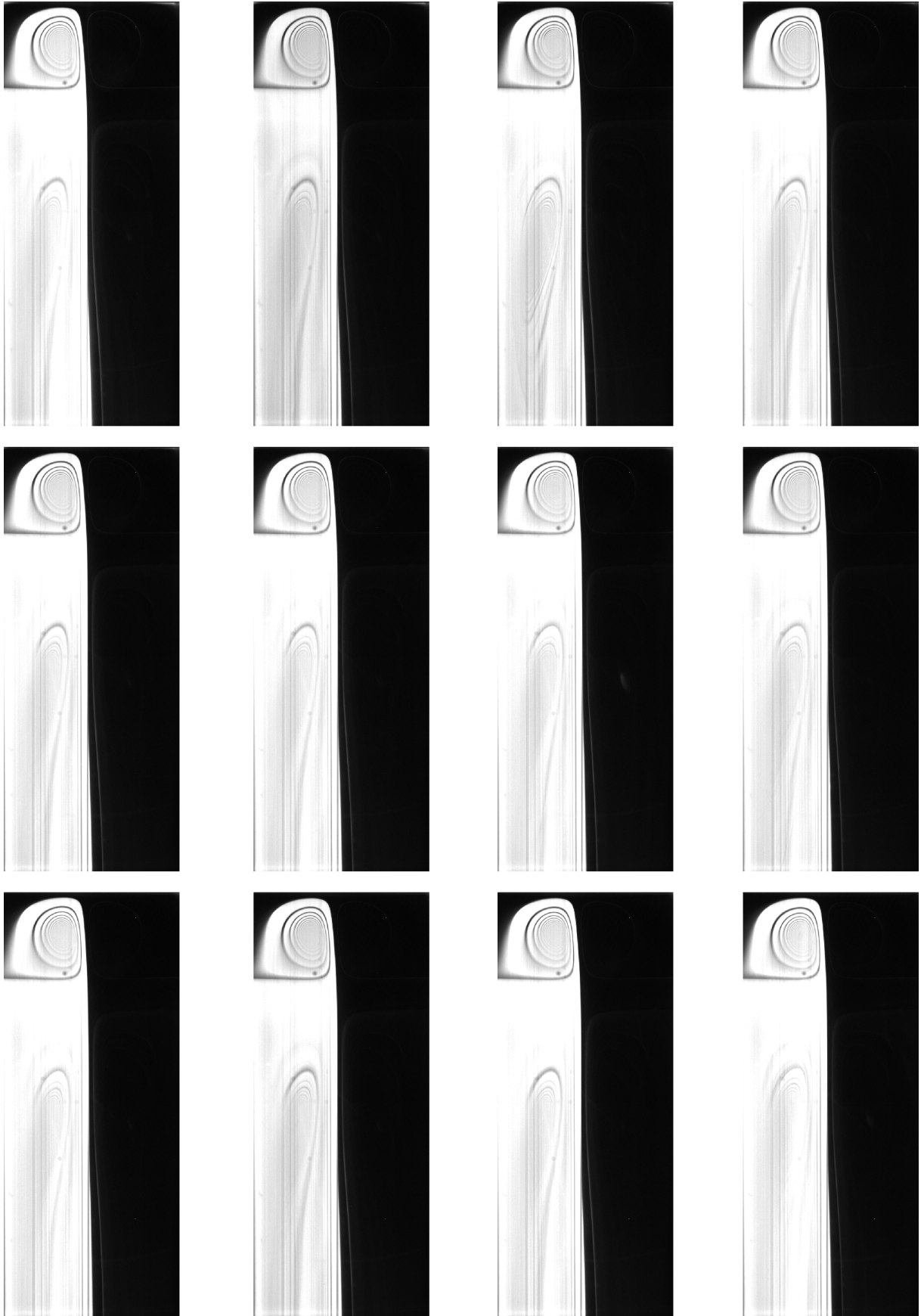


Figure 4.9. Instantaneous concentration fields at the top of the mixing chamber for $Re=51$.

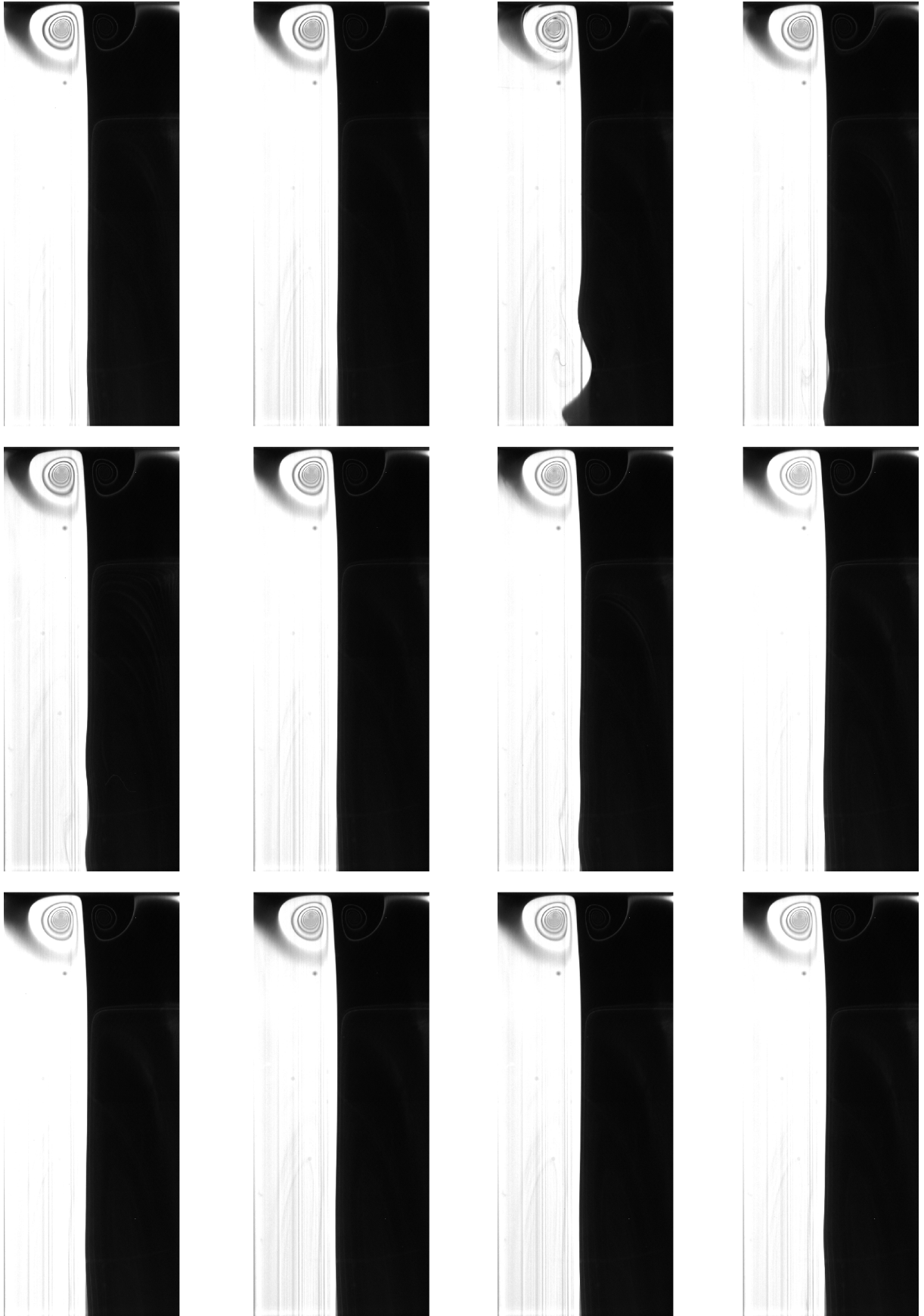


Figure 4.10. Instantaneous concentration fields at the top of the mixing chamber for $Re=103$.

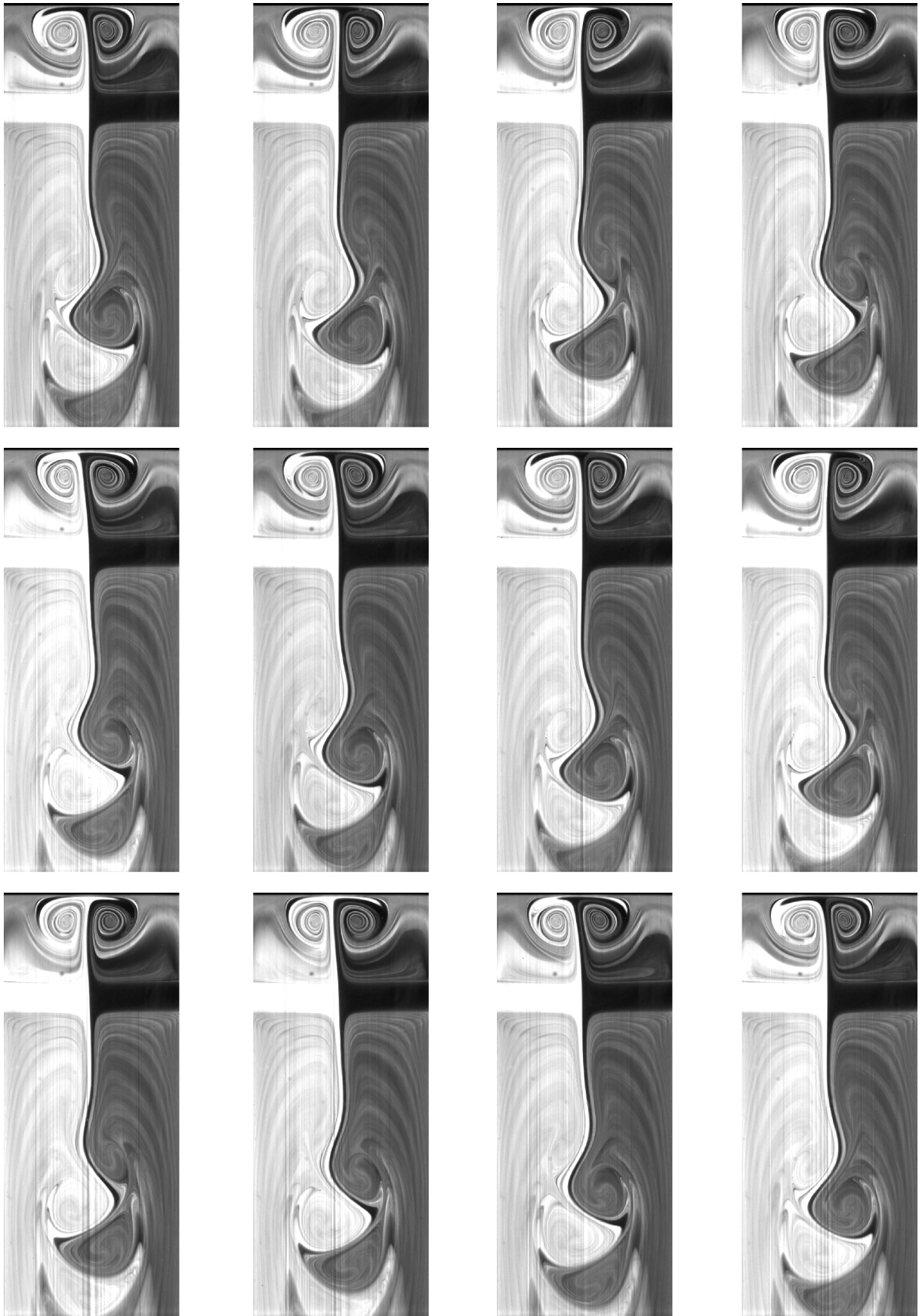


Figure 4.11. Instantaneous concentration fields at the top of the mixing chamber for $Re=104$.

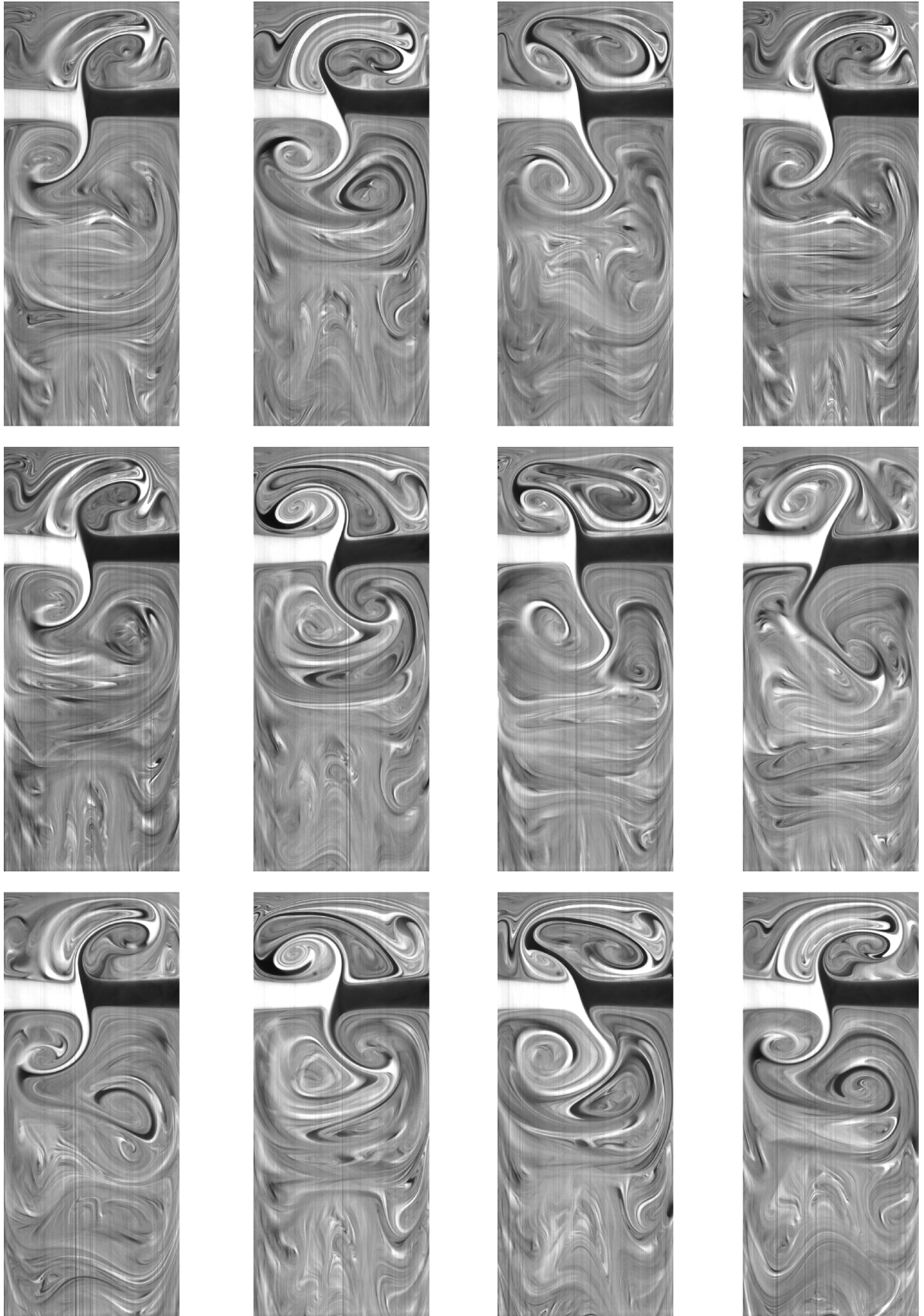


Figure 4.12. Instantaneous concentration fields at the top of the mixing chamber for $Re=111$.

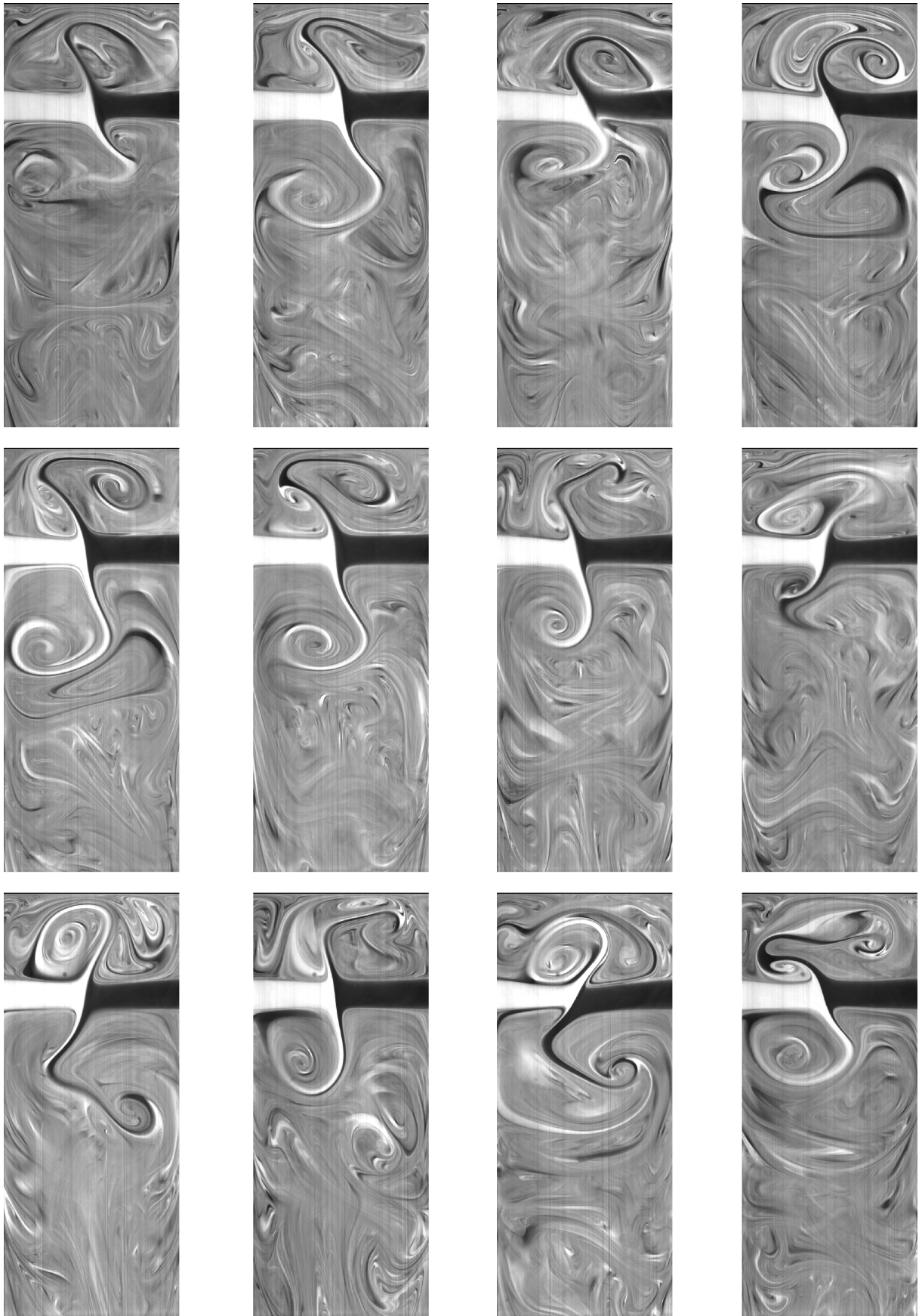


Figure 4.13. Instantaneous concentration fields at the top of the mixing chamber for $Re=120$.

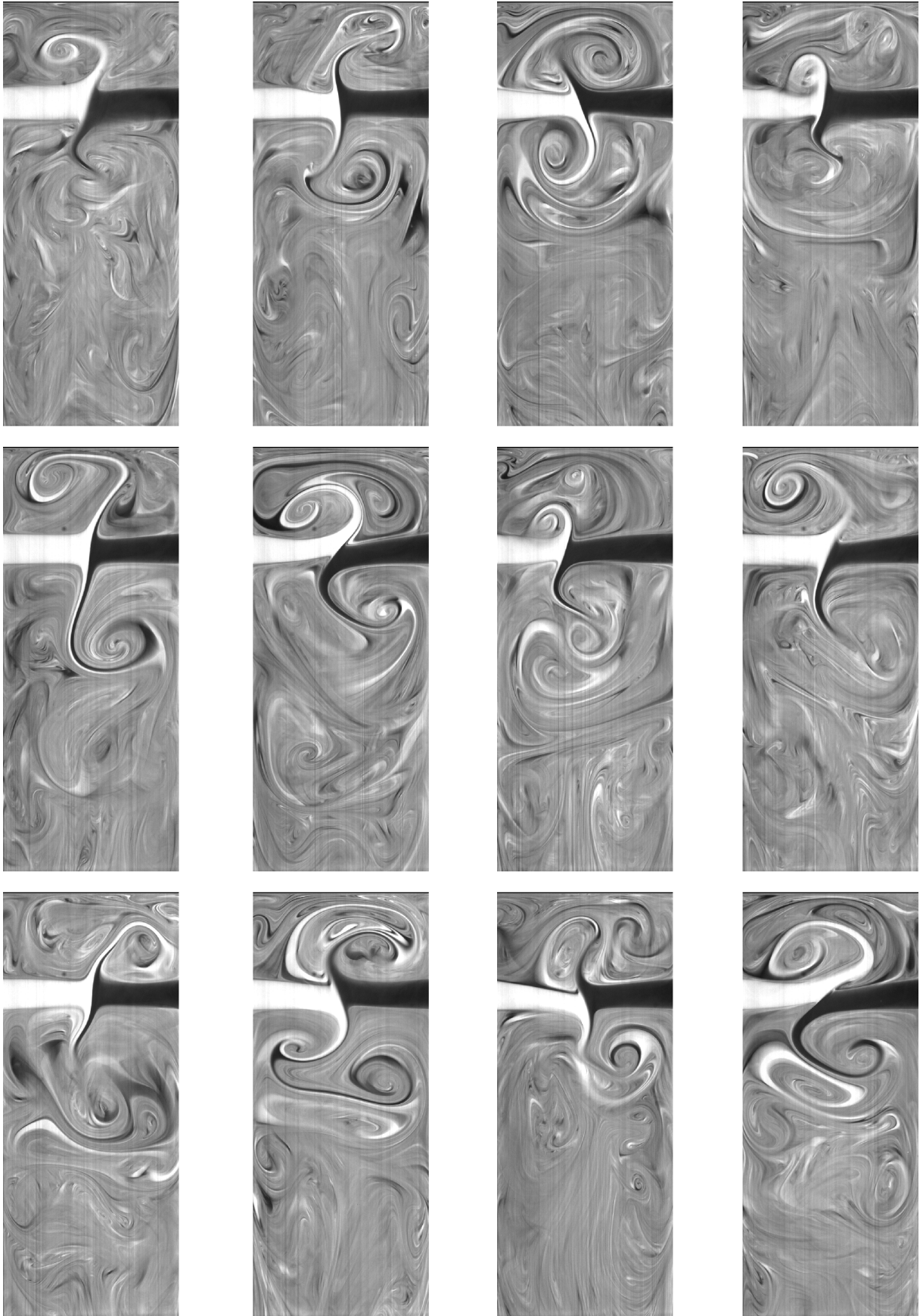


Figure 4.14. Instantaneous concentration fields at the top of the mixing chamber for $Re=150$.

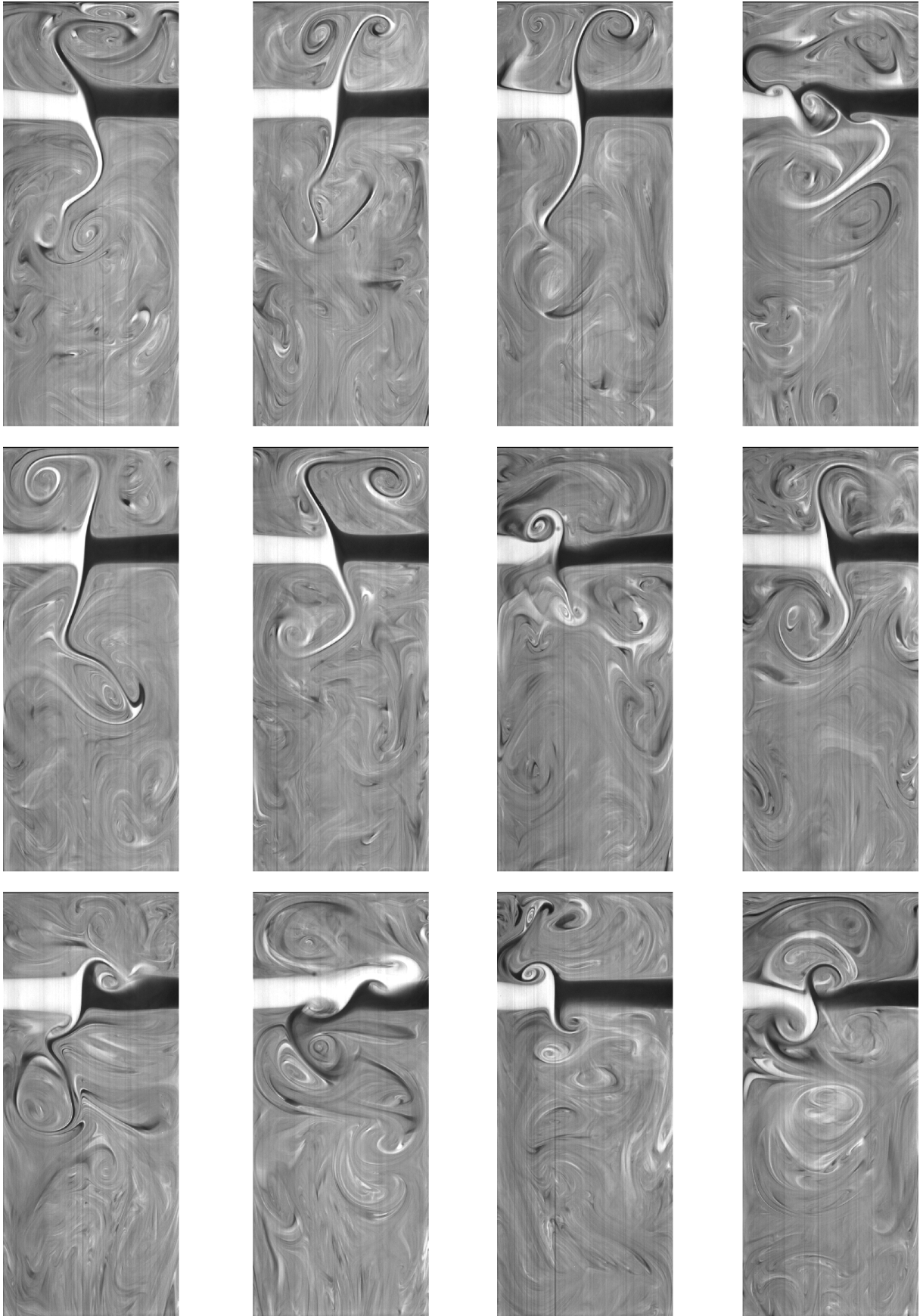


Figure 4.15. Instantaneous concentration fields at the top of the mixing chamber for $Re=201$.

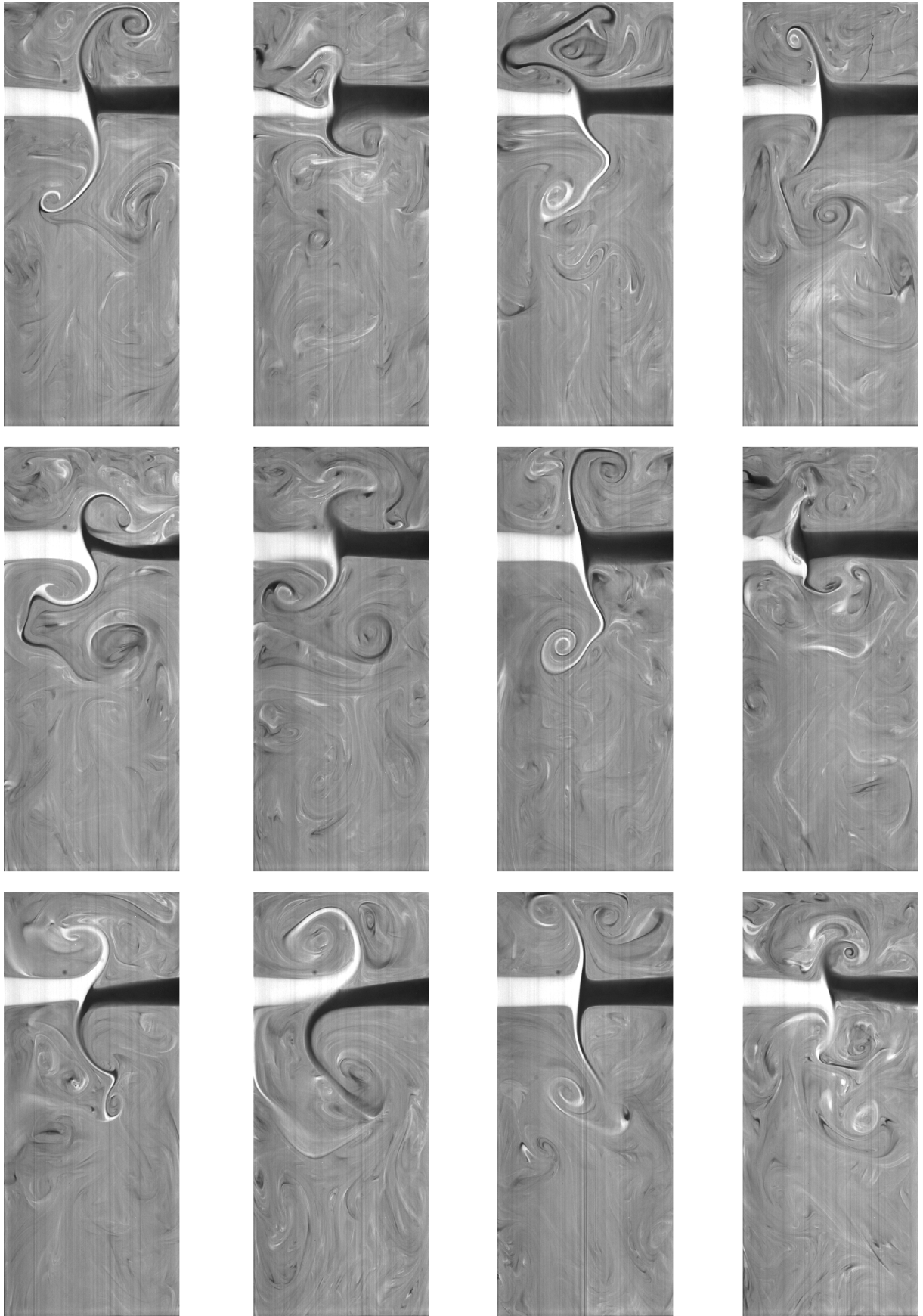


Figure 4.16. Instantaneous concentration fields at the top of the mixing chamber for $Re=304$.

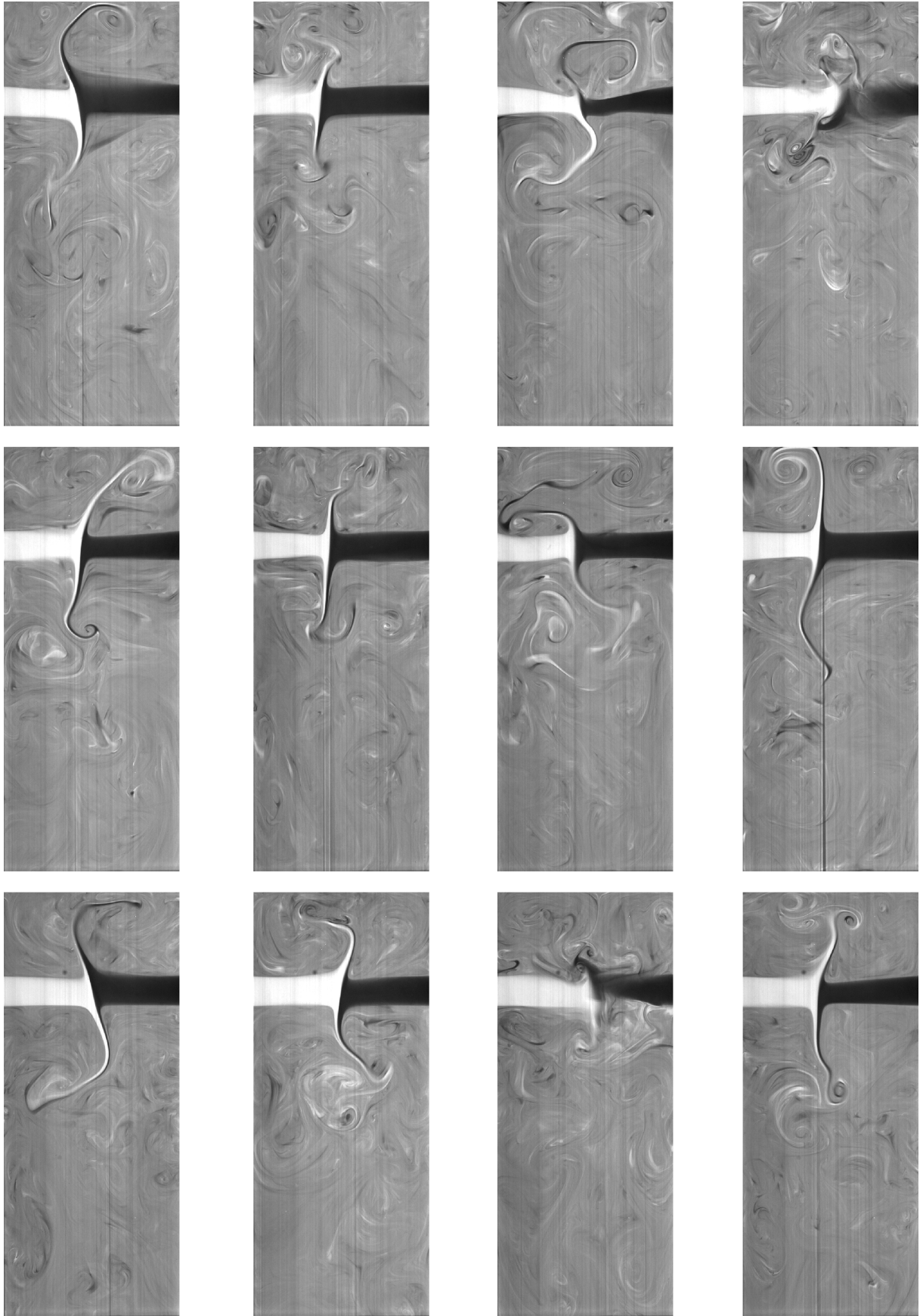


Figure 4.17. Instantaneous concentration fields at the top of the mixing chamber for $Re=402$.

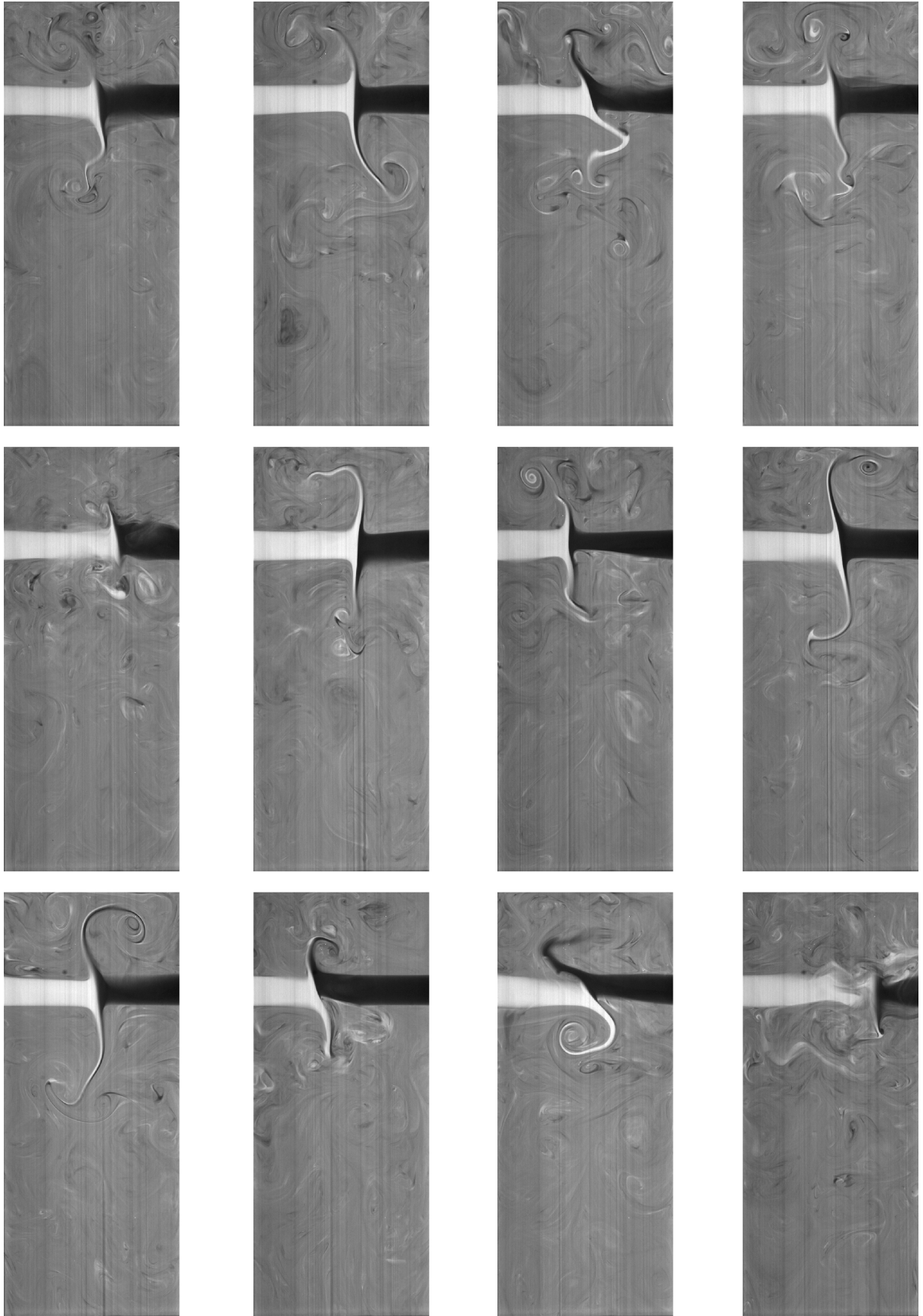


Figure 4.18. Instantaneous concentration fields at the top of the mixing chamber for $Re=505$.

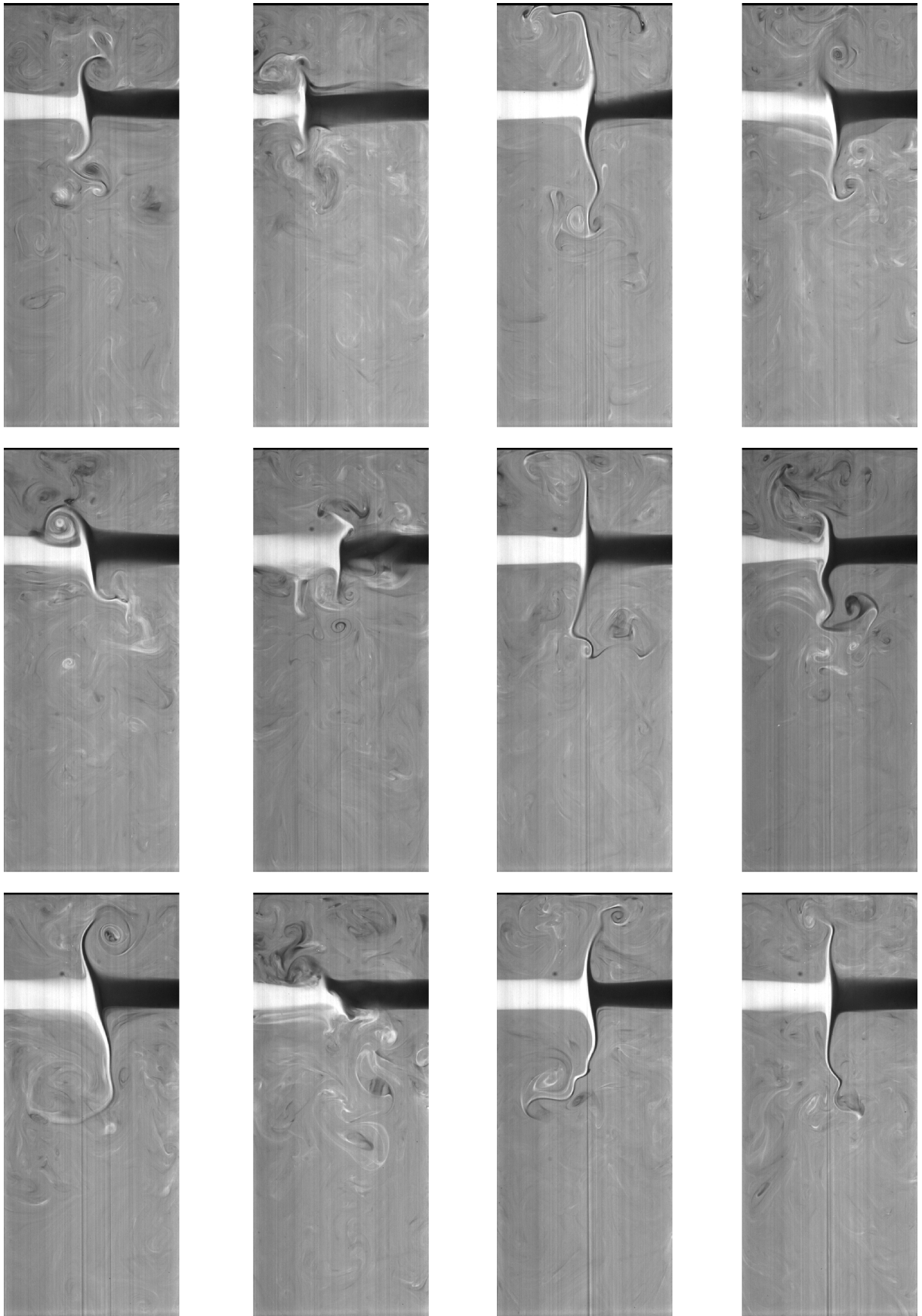


Figure 4.19. Instantaneous concentration fields at the top of the mixing chamber for $Re=617$.

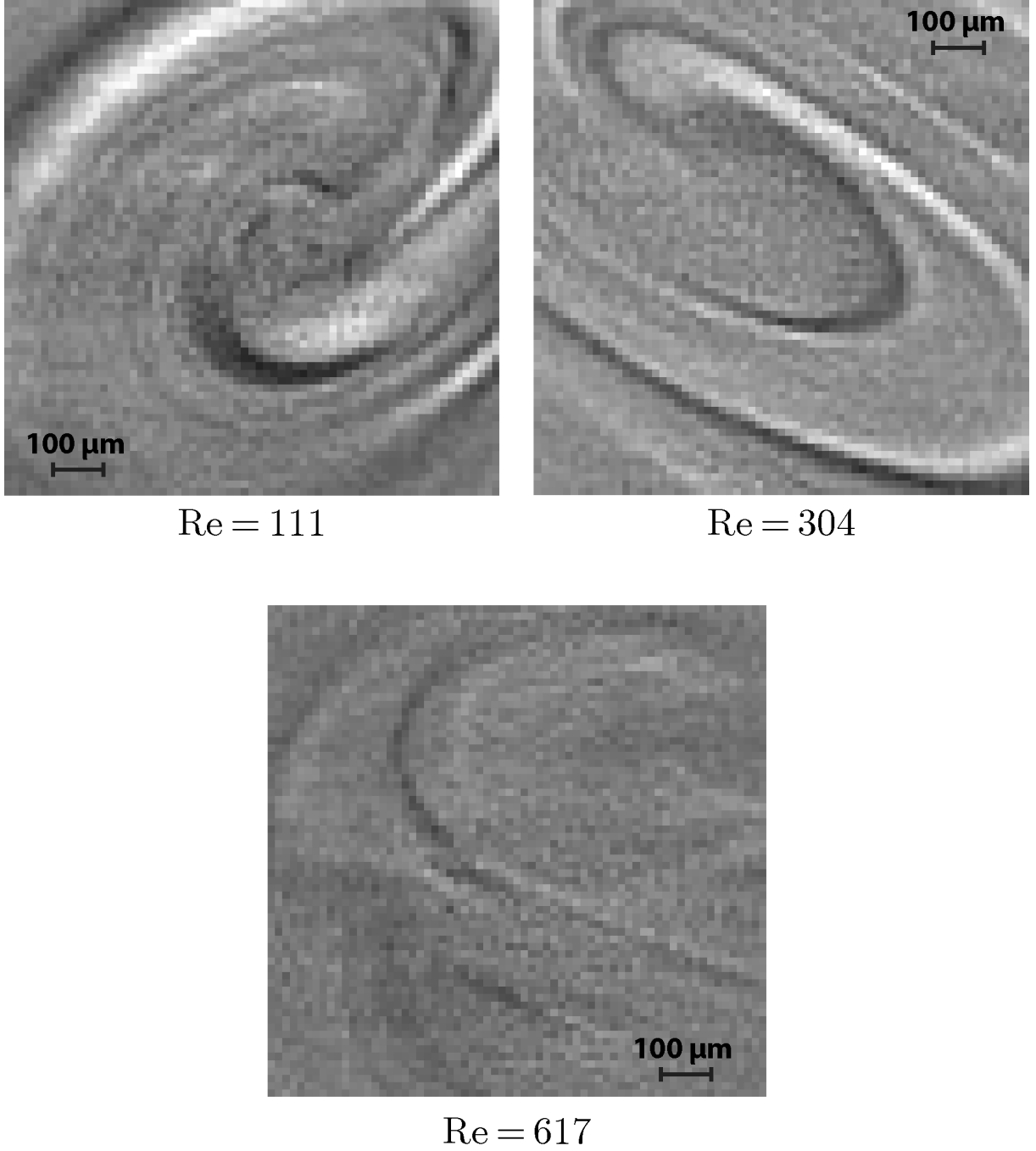


Figure 4.20. Zoomed views of the lamellar structure of the flow for three different Re .

For the selected Reynolds numbers, the Kolmogorov length scale, estimated from a mechanical energy balance to the mixing chamber as described in Chapter 2, are $\lambda_K = 190\ \mu\text{m}$, $90\ \mu\text{m}$ and $53\ \mu\text{m}$, respectively for $Re = 111$, 304 and 617 . The model of Baldyga and Bourne (1983) predicts a striation thickness distribution ranging from a thickness of $50\ \mu\text{m}$ to the injectors' diameter, with an average value of $150\ \mu\text{m}$. The estimated mixing length scale values consider that impingement mixing occurs in

top of the chamber until a length $\sim 2.5D$ from the top. In Figure 4.20 it is visible, up to the value of $15\ \mu\text{m}$ permitted by the image resolution, a distribution of striation length scales smaller than the values predicted by turbulence modeling. As already concluded in Chapter 2, the results of the visualization of the lamellar structure from PLIF strongly indicate that the flow is laminar and that mixing is obtained by laminar advective mechanisms of folding and stretching. The distribution of flow scales in the impingement mixing zone is not homogeneous, and so isotropic and homogeneous turbulence considerations for the modeling of mixing scales do not provide a correct physical description of the flow.

4.4.1.2 Mixing Quantification

Although the analysis of mixing from the visualization of the species volume fraction maps provides very good insight on the mixing mechanisms and the mixing degree, it lacks quantification. Danckwerts (1952) introduced two measurements of mixing: the scale of segregation and the intensity of segregation. While the scale of segregation is intended to quantify the reduction of mixing length scales promoted by advective flow mechanisms of stretching and folding, the intensity of segregation measures homogenization of the mixture at a molecular level by molecular diffusion.

In real polymeric systems with low diffusivity ($\mathcal{D} \sim 10^{-11}\ \text{m}^2\text{s}^{-1}$) (Bourne and Garcia-Rosas, 1985) diffusion has a limited impact on the mixing degree achieved inside the RIM machine's mixing chamber. As already explored in Chapter 2, diffusion can only fade striation thicknesses $\sim 1\ \mu\text{m}$ in the formed lamellar structure for a flow time of the order of the mean residence time.

In the present experimental study the analysis of a distribution of striation scales is conditioned by the imaging resolution ($\sim 15\ \mu\text{m}$) and the action of diffusion of rhodamine in the thinnest laminae. On one hand, for a diffusivity of rhodamine in the glycerine solution of $\mathcal{D} \sim 5 \times 10^{-11}\ \text{m}^2\text{s}^{-1}$ (Gendron et al., 2008), a value one order of magnitude higher than diffusivity in polymers, diffusion is able to fade the lamellar

structure of the flow when the striation thickness reaches values of the order of the pixels' dimension for a time equal to the mean residence time, τ , of the fluid inside the mixing chamber. On the other hand, when mixing length scales are reduced to values below the pixels' dimensions, even if diffusion has as little impact as in actual polymeric systems, it is not any longer possible to identify points of pure dyed and not dyed working fluid, i.e., pixels of pure white or pure black in the images. The resulting pixel grey scale intensity will be given by the fraction of each species contained in that pixel, even if diffusion has not faded laminae interfaces. From previous studies (Lee et al., 1980) and the results presented in Chapter 2, striation thicknesses inferior to the imaging resolution are predicted to be formed in the mixing chamber.

Since a complete description of the mixing length scales in the chamber cannot be made with the present imaging method, mixing was quantified by the calculation of the intensity of segregation, here as a measure of the extent of the formation of laminae thinner than the imaging resolution. The more these smaller scales are present in the captured plane, the more zones with volume fraction of rhodamine dyed fluid $0 < \alpha < 1$ are detected, decreasing the intensity of segregation. The intensity of segregation is defined as

$$I_s = \frac{\sigma_\alpha^2}{\bar{\alpha}(1 - \bar{\alpha})} \quad (4.9)$$

where $\bar{\alpha}$ is the spatial mean value of the volume fraction of rhodamine dyed fluid in Ω ,

$$\bar{\alpha} = \frac{\int_{\Omega} \alpha(\mathbf{x}) \, dA}{A} = \frac{\sum_{i=1}^{N_r} \sum_{j=1}^{N_c} \alpha(i, j)}{N_r N_c} \quad (4.10)$$

and σ_α^2 the variance of α in Ω ,

$$\sigma_{\alpha}^2 = \frac{\int_{\Omega} (\alpha(\mathbf{x}) - \bar{\alpha})^2 dA}{A} = \frac{\sum_{i=1}^{N_r} \sum_{j=1}^{N_c} (\alpha(i, j) - \bar{\alpha})^2}{N_r N_c}. \quad (4.11)$$

N_r and N_c are the number of rows and columns of pixels in the captured PLIF images. For complete segregation, $\alpha(\mathbf{x})$ at any point in the domain is either 0 or 1 and Equation (4.9) returns $I_s = 1$. When perfect mixing is achieved, the concentration at any point in the domain is $\alpha(\mathbf{x}) = \bar{\alpha}$, $\forall \mathbf{x} \in \Omega$, and Equation (4.9) returns a value of $I_s = 0$.

Figure 4.21 shows the time average intensity of segregation, $\bar{I}_s$, as a function of Re at the bottom region of the chamber, as indicated in Figure 4.8, calculated from the 300 captured frames. Animations of all the captured images in the bottom of the mixing chamber and for the conditions of Table 4.3 are included in the DVD (folder ~/Chapter4/Section4.4.1/Bottom). The dashed line indicates a trend of the points and the error bars indicate the standard deviation of I_s , σ_{I_s} , in the 300 measurements, defined as

$$\sigma_{I_s} = \sqrt{\frac{1}{300} \sum_{i=1}^{300} (I_s(i) - \bar{I}_s)^2}. \quad (4.12)$$

For Re lower than the critical value for the onset of the dynamic chaotic flow regime, Figure 4.21 shows complete segregation of the two inlet streams with $I_s \approx 1$. As Re increases, I_s decreases sharply with the onset of the dynamic regime for $Re \approx 105$. A further reduction of mixing scales as Re is increased occurs in a linear trend, as also observed in previous studies of Tucker III and Suh (1980) and Unger and Muzzio (1999). This continued reduction of mixing scales with Re is also predicted from the numerical analysis of the flow of Chapters 2 and 3 and contradicts previous assumptions of Baldyga and Bourne (1983) and May (1996) based on the statistical theory turbulent diffusion.

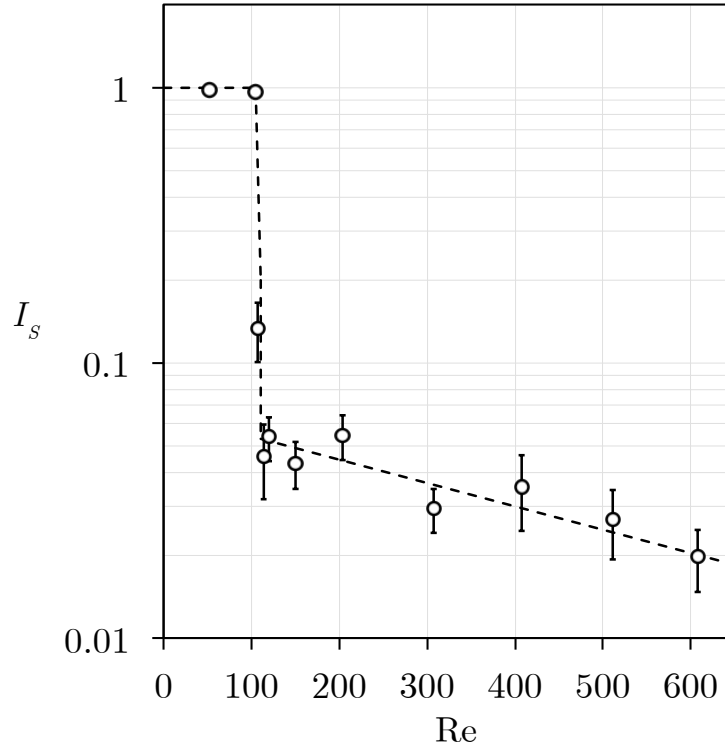


Figure 4.21. Intensity of segregation in the bottom region of the mixing chamber as a function of Re .

4.4.2 Kinetic Energy Rate Ratio Effect

4.4.2.1 Flow Visualization

As in Section 4.4.1, 2D rhodamine 6G volume fraction fields were captured for the conditions of Table 4.4, but here for the study of the effect of the two impinging jets' balancing in the mixing degree. The Reynolds number of the left injector was kept constant ($Re \simeq 300$) and the flow rate of the left injector was changed to study the flow at different jets' kinetic energy rate ratio. From these experiments it is intended to study the impact on mixing when the RIM machine has to be operated with monomers or pre-polymers formulations with different physical properties and stoichiometry. The kinetic energy rate ratio will be shown in Chapter 5 to be the main parameter to control mixing in systems with feeding streams with different physical properties or volumetric stoichiometric ratios. Figure 4.22 to Figure 4.25

show randomly chosen images from the 300 captured in each experiment of the concentration field at the top of the mixing chamber, as indicated in Figure 4.8, to illustrate the dynamic nature of the mixing process. For better visualization, animations of all the captured images are included in the DVD accompanying this work (folder ~/Chapter4/Section4.4.2/Top).

Results (Figure 4.22 to Figure 4.25) show that mixing is highly affected by the jet's balancing. As the impingement point is more deviated from the center, comparing with the concentration field in Figure 4.16, larger and more frequent portions of unmixed inlet streams materials are detected in the flow, even if, in the studied range of ϕ_K and for $Re \sim 300$, a dynamic flow regime has always been observed.

The observation of the decrease on the mixing quality for $\phi_K \neq 1$, even when it results of the increase of the energy rate of one of the jet streams, contrasts with the results obtained from previous models for a description of the lamellar structure of the flow, either from flow kinematics considerations (Lee et al., 1980) or from isotropic and homogeneous turbulence considerations (Tucker III and Suh, 1980; Baldyga and Bourne, 1983; May, 1996). All the models predict practically the same mixing scales or the formation of thinner lamellae if the kinetic energy of one of the jets is increased, independently of jets' impingement point position in the mixing chamber in those flow conditions, which is not in agreement with the experimental observations.

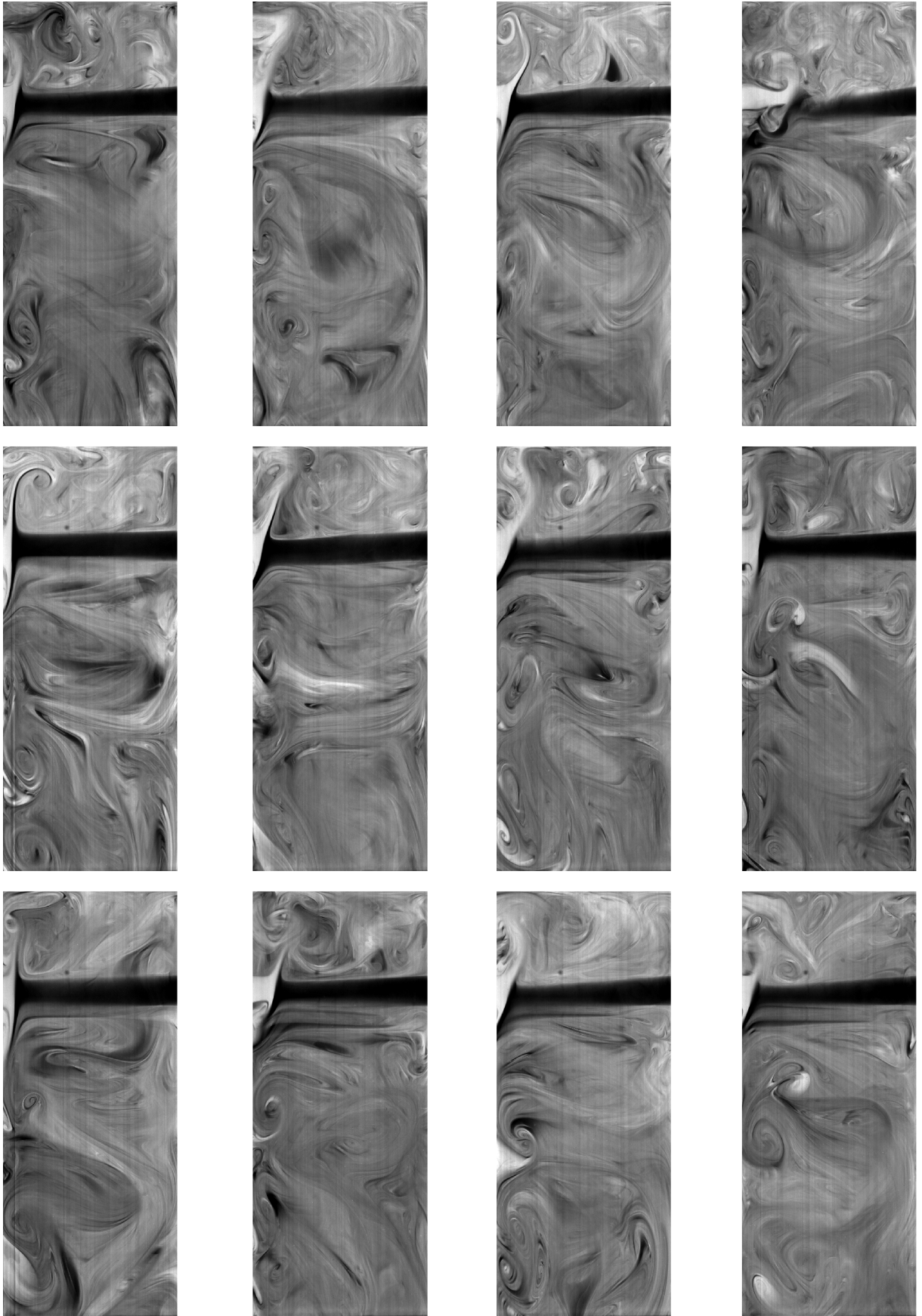


Figure 4.22. Instantaneous concentration fields at the top of the mixing chamber for $Re_1=324$ and $\phi_K=0.67$.

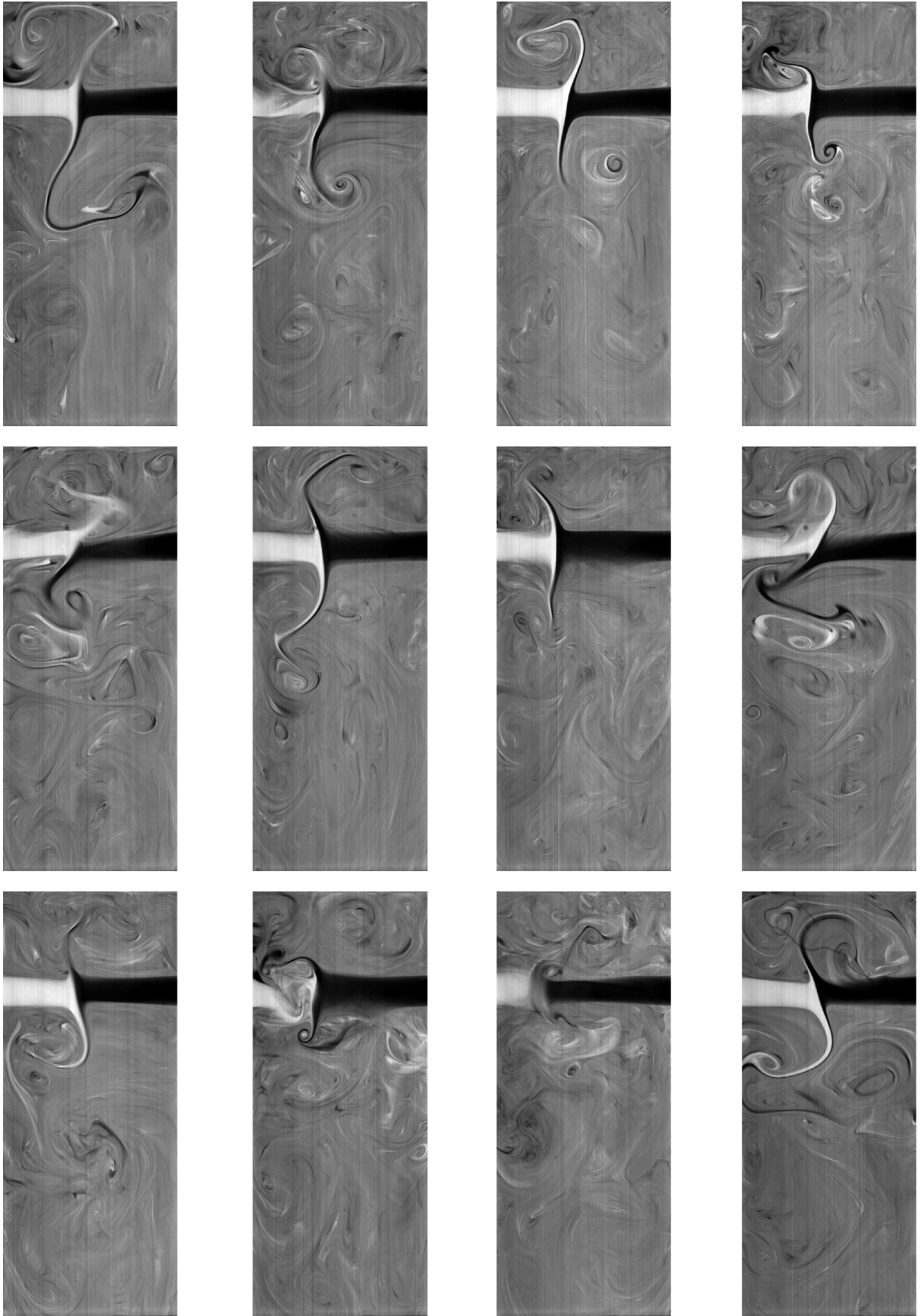


Figure 4.23. Instantaneous concentration fields at the top of the mixing chamber for $Re_1=324$ and $\phi_K=0.92$.

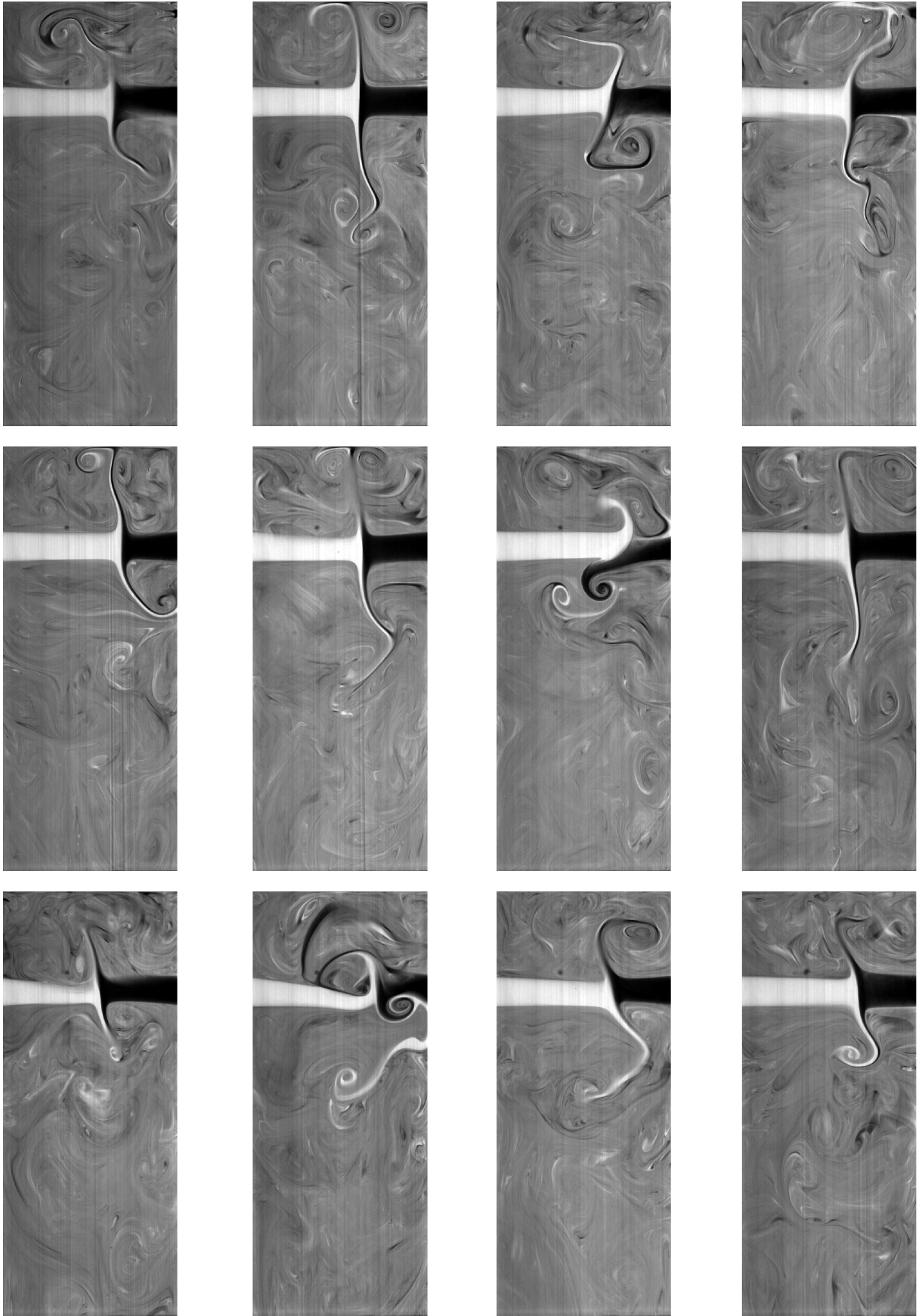


Figure 4.24. Instantaneous concentration fields at the top of the mixing chamber for $Re_1=324$ and $\phi_K=1.14$.

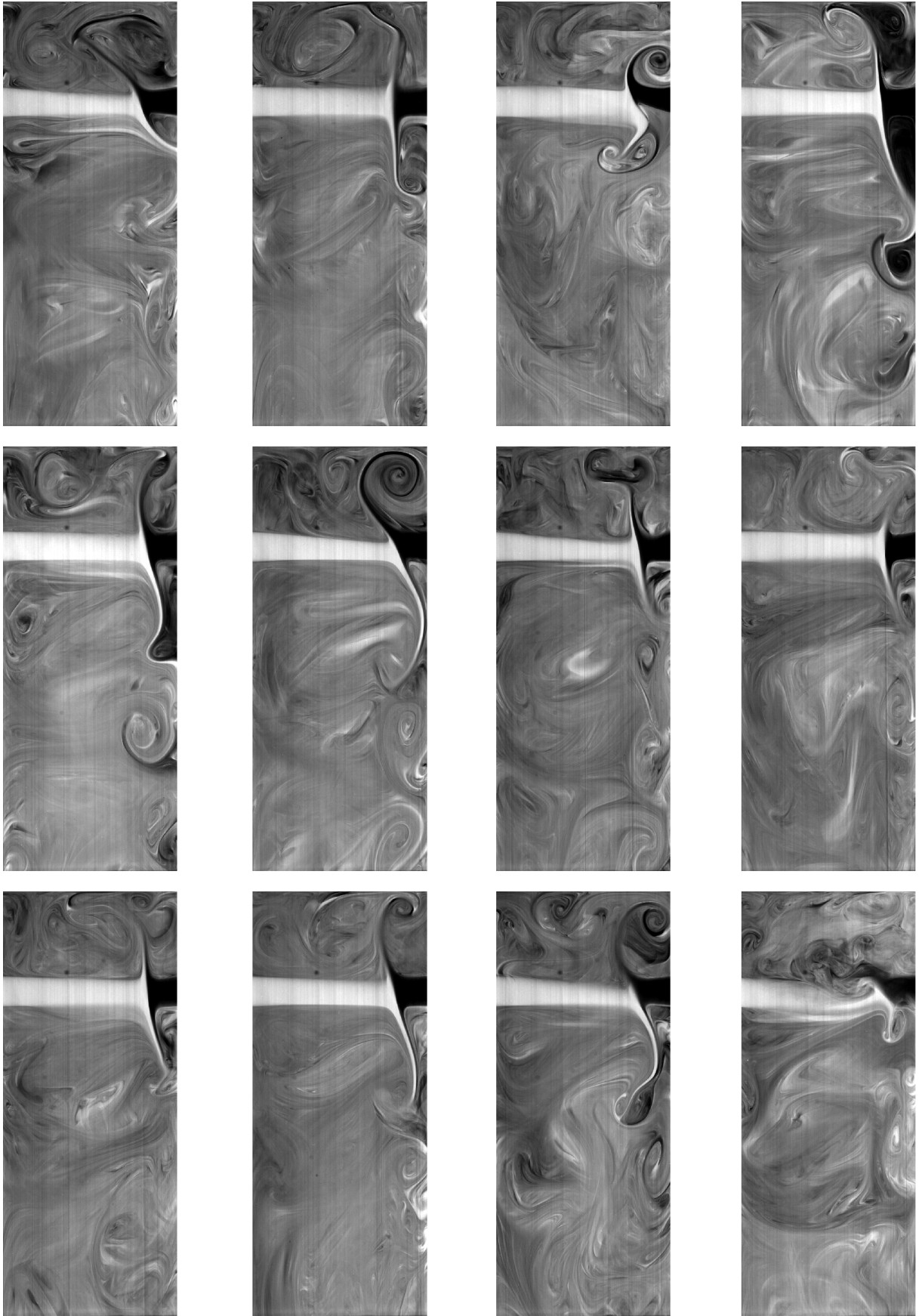


Figure 4.25. Instantaneous concentration fields at the top of the mixing chamber for $Re_1=324$ and $\phi_K=1.5$.

4.4.2.2 Mixing Quantification

Mixing was quantified, as in Section 4.4.1 by the calculation of the intensity of segregation as a measure of the extent of the formation of laminae thinner than the imaging resolution. Figure 4.26 shows the time average normalized intensity of segregation, $I_s/I_{s,0}$, at the bottom region of the chamber, as indicated in Figure 4.8, calculated from the 300 captured frames as a function of ϕ_K . Figure 4.26 also shows the mass flow rate relative deviation, $|\dot{m}_1 - \dot{m}_2|/\dot{m}_1$ for each value of ϕ_K . $I_{s,0}$ is here the value of the intensity of segregation for $\phi_K = 1$ and $Re = 304$ in Figure 4.21. The error bars indicate the standard deviation of $I_s/I_{s,0}$ in the 300 measurements. Animations of all the captured images in the bottom of the mixing chamber are included in the DVD (folder ~/Chapter4/Section4.4.2/Bottom).

From Figure 4.26 the impact of the jets' unbalancing on the mixing degree becomes clear; the more the impingement point is deviated from the center, the more segregated spatially and more heterogeneous in time (standard deviation of I_s increases) the resulting mixture of the two streams becomes. It should be pointed that a decrease in the mixing degree of the two injected streams is observed even for deviations as small as 5% in the flow rate. These results contrast with the observations of Tucker III and Suh (1980) where the jets' momentum ratio does not seem to affect mixing quality.

The observation of the dependence of the mixing degree with the jets' balance condition led to a more detailed study of the dependence of the jets impinging point position and the jets' interaction under different operating conditions, presented in Chapter 5.

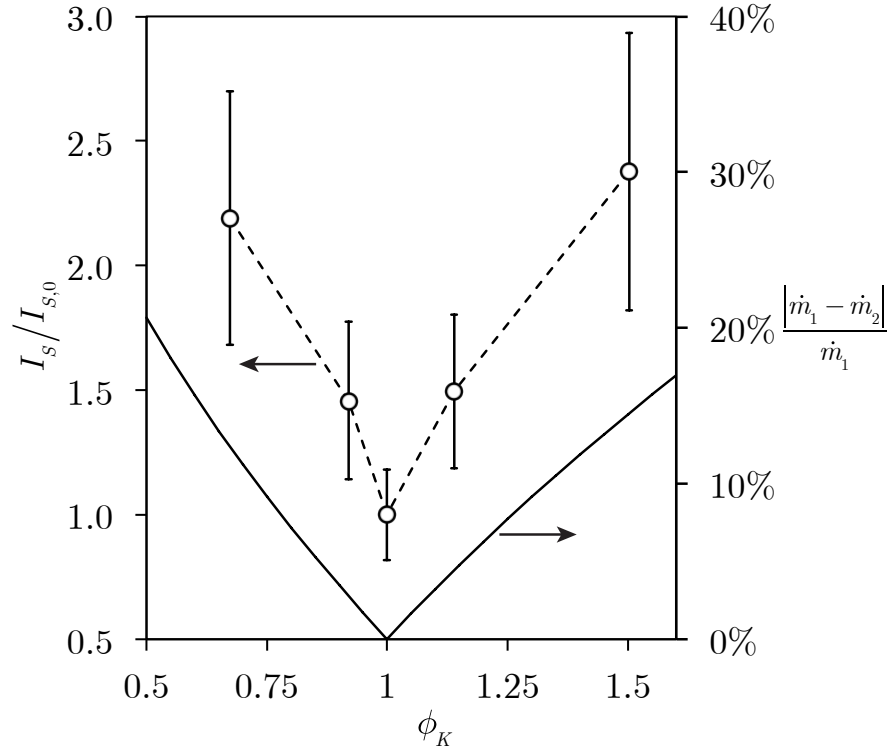


Figure 4.26. Normalized intensity of segregation and mass flow rate relative deviation as a function of the jets kinetic energy rate ratio for $Re=300$.

4.5 Conclusions

The PLIF experimental technique was used in this Chapter to obtain 2D concentration fields at the top and bottom of the RIM machine's mixing chamber. The obtained fields were used to analyze qualitatively and quantitatively the effect of Re and the two impinging jets' balancing on mixing. One of the injected fluids was marked with rhodamine 6G which emits fluorescence when excited by laser light, allowing to detect the mixing patterns in the flow. The imaging resolution allowed visualization of the lamellar structure of the flow with thicknesses down to $15 \mu m$. The intensity of segregation of the dyed fluid volume fraction field in the bottom of the mixing chamber, as defined by Danckwerts (1952), was used as a measure of the extent of the formation of laminae thinner than the imaging resolution.

Based on the results of Section 4.4.1 three different flow regimes were identified:

- For $Re \leq 103$ the flow is segregated, with each side of the mixing chamber containing mainly one of the fluids;
- For $Re = 104$ the flow evolves to an *oscillatory periodic laminar regime* characterized by the disturbance on the two impinging jets' interface and the formation of alternated detaching vortices in the mixing chamber downstream the inlet;
- For $Re \geq 111$ the formation of the interface vortices occurs earlier in the flow, i.e. closer to the impingement point, and start to interact with the inlet jets, developing a *self-sustained chaotic laminar regime* with strong mixing dynamics.

As in 3D computational simulations of the flow in Chapter 2, advective mixing in RIM is observed to occur above a critical Reynolds number $Re_c > 110$.

As Re is increased above Re_c , in the studied range, fewer structures of pure injected fluids are identified in the flow due to the increase of the mixing degree and the formation of thinner thicknesses. The greatest change on laminae size with the increase of Re is observed until $Re = 300$. From $Re = 300$ until $Re = 600$, a reduction of laminae thicknesses with Re is still observed but in a smaller extent.

The observed patterns and distribution of lamellae thicknesses indicate that the flow in the studied range of Re and for $Re \geq Re_c$ is laminar and that mixing is achieved by the advective mechanisms of stretching and folding. The results of this chapter indicate that isotropic and homogeneous turbulence considerations for the modeling of mixing scales do not provide a correct physical description of the flow.

The PLIF results for different jets' kinetic energy rate ratio have shown the importance of maintaining the jets balanced and the impingement point in the center of the chamber. When the jets' impingement point is deviated from the center of the mixing chamber, by increasing or decreasing one of the jets' flow rate, the mixing

degree between the two feeding streams decreases and becomes more inconsistent with time, even when it corresponds to increasing the kinetic energy rate of one of the streams.

4.6 References

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5 Impinging Jets Interaction

5.1 Introduction and State of the Art

Mixing quality in RIM depends strongly in ensuring proper operational conditions to continuously maintain the impinging jets' oscillation and its interface unstable in the mixing chamber. The folding and stretching of the interface between two or more monomers or pre-polymers streams are the responsible mechanisms for the reduction of mixing scales down to the necessary values for complete reaction in the mold.

As show in Chapters 2 and 4 there is a critical Reynolds number value that marks the onset of the dynamic regime. However, even when the jets' Reynolds number is above this critical value, differences in the monomers' physical properties or small deviations on the flow rate of one injector may result in poor mixing, as shown in Chapter 4, or even in complete dampening of the jets' oscillation. The difficulty in maintaining the continuous presence of the self-sustained chaotic flow regime in the mixing chamber during the machine operation is one of the main reasons to obtain structural or aesthetical defects in the produced plastic parts, which lead, eventually, to the parts rejection. The polymerization reaction itself often adds a constrain to mixing control in RIM since the jets' mass flow ratio must be set accordingly to the

stoichiometry. This may be difficult to accomplish simultaneously, the iso-momentum condition in standard RIM machine mixing chambers, for example, when the monomers' densities are different. Daily and even seasonal temperature changes, by altering in a different manner the monomers' physical properties, will eventually require the machine tuning for operation in the best conditions for complete and efficient reaction of the monomers.

The problem of maintaining the mixing regime during RIM operation or other Confined Impinging Jets Reactors (CIJR) applications has been the object of study of several authors. Malguarnera and Suh (1977) were the first to show experimentally with a RIM machine prototype that, in addition to satisfying the desired mass flow rate ratio, set by stoichiometry, the momentum ratio of the fluid components must be equal to 1. The jets' momentum rate ratio, ϕ_M , and the jets' flow rate ratio, ϕ_{FR} , are respectively defined as

$$\phi_M = \frac{\rho_1 d_1^2 u_{inj,1}^2}{\rho_2 d_2^2 u_{inj,2}^2} \quad (5.1)$$

and

$$\phi_{FR} = \frac{\rho_1 d_1^2 u_{inj,1}}{\rho_2 d_2^2 u_{inj,2}} \quad (5.2)$$

where ρ is the fluid density, d the injector diameter and u_{inj} the mean velocity at the injectors. The indices 1 and 2 indicate the left and the right side injectors, respectively. Malguarnera and Suh (1977) proposed that when a volumetric stoichiometric ratio different from 1 is desired, it is necessary to use different section area injectors.

Sebastian and Boukobbal (1986) studied experimentally the effect of the jets momentum ratio on the actual polymerization. Their study has shown that a higher adiabatic temperature raise, which is directly linked to product yield and mixing

efficiency in exothermic reactions, is obtained when the momentum ratio is closer to one.

Hosseinalipour and Mujumdar (1997) studied numerically the effect of unequal momenta jets impinging in two-dimensional confined opposing reactor in steady laminar regime. In this work the inequality in the jets' momenta is achieved using equal or unequal widths of the two dimensional jets by adjusting the average velocity appropriately, showing the effect in the flow and its thermal characteristics.

Johnson (2000) observed a significant alteration of the flow field under unequal flow rate conditions in RIM from numerical flow simulations and experimentally for an equal injectors diameter mixing chamber. Changing the flow rate of one injector moves the impingement point towards the lower momentum rate jet, stabilizing the flow field to some extent. Johnson (2000) has shown the formation of large areas of recirculation in the mixing chamber that lead to poor mixing due to the possibility of unmixed fluid leaving the chamber and the increase in the residence time of the mixing fluids.

Santos et al. (2002) and Santos et al. (2010) have also shown from Laser Doppler Anemometry (LDA) and computational simulations of the flow that the momentum ratio between the jets is a critical parameter for the dynamic behavior of the system where the jets oscillations were only observable on a very narrow interval of the momentum ratio. This work has also shown that, by changing the injector diameters, a sustainable chaotic flow regime can be obtained for flow rate ratios different from the unity as long as the diameter change imposes a unitary value for the momentum ratio.

Siddiqui et al. (2009) also studied numerically and experimentally the effect of unequal flow on the average energy dissipation in a CIJR, showing a sharp decrease of energy dissipation in the a CIJR, as the difference in flow rates increases, to approximately 40% of the value for equal flow rates for lower flow rates ($Re \sim 2100$). For higher flow rates $Re \geq 3500$, Siddiqui et al. (2009) reports a transition to a

turbulent flow regime stabilizing the decrease of energy dissipation with the increase of flow rate difference. Under these conditions, energy dissipation retained 84% of the value in balanced flow all the way to a 30% difference in flow rates, suggesting that reservations about the practicality of operating CIJRs under non-ideal plant conditions and varying inlet flow rates can be set aside. Despite these results, the range of industrial interest for RIM operation goes only to Reynolds numbers up to 600, as high monomers' viscosities would imply an excessive use of pumping energy for higher Re , making these recommendations not applicable.

Steffen Schütz et al. (2009) studied the impact of the injectors' viscosity ratio in the flow, and observed that the jets' impingement point position is shifted from the center towards the injector where the higher viscous fluid is injected due to the increased frictional forces acting on the higher viscous jet, leading to a quicker jet expansion. They have also observed that an asymmetric velocity field results in a worse distribution of shear and elongation forces and in an incomplete usage of the mixing volume.

More recently, Nunes et al. (2012) for RIM and (Krupa et al., 2012) for T-shaped CIJRs have also shown experimentally with consecutive-competitive reactions, specifically used in micromixing characterization, that mixing quality decreases when the flow rate ratio is different from unity.

The present chapter describes a theoretical, computational and experimental study of the interaction between the two impinging jets in the RIM machine mixing chamber under different geometrical and operational conditions. The main goal is to offer deeper insight about the system's behavior under unbalanced conditions. Results offer ruler for setting operational and design parameters to optimize the plastic production for any RIM monomer formulation.

This chapter is organized into five sections. An elastic analogue model of the jets interaction is described in Section 5.2. In Section 5.3 an axisymmetric Computational Fluid Dynamic (CFD) model of the flow in the mixing chamber is described and the

obtained results for the jets impinging point position are compared with the predicted by the elastic analogue model. In Section 5.4, Particle Image Velocimetry (PIV) experimental results of the flow regime and jets interaction in the mixing chamber are shown and discussed.

5.2 Impinging Jets' Elastic Analogue

To predict the impingement point position of the two opposed jets in the RIM machine mixing chamber an elastic analogue model of the jets' is proposed. The jets' system is here conceptualized to behave as a system of two connected springs, mimicking the two oscillating jets, which are forced to compress from an initial length equal to the mixing chamber diameter, D , to an equilibrium length, l . A previous analogy of impinging jets with springs was presented by Xiaojin et al. (2007) to study the flow's oscillation. Figure 5.1 shows a schematic representation of the considered elastic system, where index 1 refers to left side of the mixing chamber and index 2 to the right side.

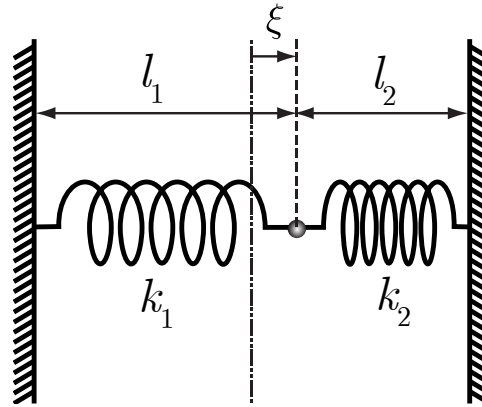


Figure 5.1. Schematic representation of the elastic analogue of the impinging jets system.

Each competing spring applies a force to the other spring so that, in equilibrium, Spring 1 will be deformed to a length l_1 and Spring 2 to a length l_2 , with $l_1 + l_2 = D$. Considering that the springs behave ideally, the force applied to Spring 2 by Spring 1, $F_{1 \rightarrow 2}$, and the force applied to Spring 1 by Spring 2, $F_{2 \rightarrow 1}$, can be related to the springs' displacement by Hooke's law

$$\begin{cases} F_{2 \rightarrow 1} = k_1 (D - l_1) = k_1 l_2 \\ F_{1 \rightarrow 2} = k_2 (D - l_2) = k_2 l_1 \end{cases}, \quad (5.3)$$

where k_1 and k_2 are the springs' constants, and can be considered to be a function of the injector diameter, jet velocity and the fluid physical properties. When the two springs are in equilibrium, $F_{1 \rightarrow 2}$ must be equal to $F_{2 \rightarrow 1}$. The present analysis only focus on an average impingement point position and neglects its oscillation with time around the average point as observed in Chapter 4. Considering the equilibrium condition and combining the two terms of Equation (5.3), the springs' constants ratio is equal to

$$\frac{k_1}{k_2} = \frac{l_1}{l_2}. \quad (5.4)$$

The springs' compression will result in an increase of its stored elastic potential energy. The potential energy stored by Spring 1, $\Delta E_{P,1}$, and Spring 2, $\Delta E_{P,2}$, are given by the work performed by the forces along the compression length

$$\begin{cases} \Delta E_{P,1} = \frac{1}{2} k_1 l_2^2 \\ \Delta E_{P,2} = \frac{1}{2} k_2 l_1^2 \end{cases}. \quad (5.5)$$

The potential energy stored at each instant by one spring in the elastic system can be assumed to be proportional, in the continuous jets flow system, to the kinetic energy rate, $\dot{E}_K$, of the opposed jet forcing the spring to compress. With this assumption, combining Equations (5.4) and (5.5) results in

$$\frac{\Delta E_{P,2}}{\Delta E_{P,1}} = \frac{\dot{E}_{K,1}}{\dot{E}_{K,2}} = \frac{k_2}{k_1} \frac{l_1^2}{l_2^2} = \frac{l_1}{l_2}. \quad (5.6)$$

However, as the jets travel in the mixing chamber from the inlet until the impingement point, they expand and dissipate kinetic energy by entrainment of surrounding fluid from the sides and formation of vortices at the top of the chamber. For this reason Equation (5.6) should be seen as

$$\frac{\dot{E}_{K,1}(l_1)}{\dot{E}_{K,2}(l_2)} = \frac{l_1}{l_2}. \quad (5.7)$$

where $\dot{E}_K$ is a function of the length traveled by the jets from the inlet until the impingement point.

The jets' kinetic energy with the position inside the chamber can be estimated from the simplified model of a narrow axisymmetric jet (NAJ) described by Bird et al. (2002) and White (2006). The NAJ model describes a jet that enters a larger medium from a circular hole with enough momentum to remain narrow and grow slowly. In the RIM machine mixing chamber the jets have high inlet velocities (10 to 150 m·s⁻¹) and travel maximum distances of 1 to 2 cm, so it's valid to assume that, apart from the impingement point where the jets are forced to bend and spread radially, radial variations are much larger than axial variations, $\partial_r \gg \partial_x$, and that the main jet motion is in the axial direction, $u \gg v$. Here x and r represent the axial and radial coordinates, respectively and u and v represent the axial and radial velocities, respectively. Pressure is considered uniform inside the chamber. The use of this simplified model neglects the effect of the chamber walls and the additional kinetic energy dissipation due to unsteady vortex formation and detachment in the unsteady chaotic regime. Some divergence is thus expected between the experimental and the CFD simulations data as the Reynolds number is increased and as the jets' impingement point position approaches the chamber walls.

The equations that describe the mass conservation and the axial momentum transport in the steady narrow jet flow are, respectively

$$\partial_x u + \frac{1}{r} \partial_r (rv) = 0 \text{ and} \quad (5.8)$$

$$v \partial_r u + u \partial_x u = \frac{\nu}{r} \partial_r (r \partial_r u), \quad (5.9)$$

where $\nu = \mu/\rho$ is the kinematic viscosity. These conservation equations are to be solved with the following boundary conditions

$$\begin{cases} v = 0 \text{ at } r = 0, \\ \partial_r u = 0 \text{ at } r = 0, \text{ and} \\ u \rightarrow 0 \text{ at } x \rightarrow \infty. \end{cases} \quad (5.10)$$

The exact solution of Equations (5.8) and (5.9) for a point source of momentum is given by Bird et al. (2002) and White (2006):

$$u(x, r) = \frac{3J}{8\pi\mu x} \left[1 + \frac{1}{4} \left(\frac{Cr}{x} \right)^2 \right]^{-2} \quad (5.11)$$

for the axial velocity and

$$v(x, r) = \left(\frac{C\nu}{x} \right) \left[\frac{Cr}{x} - \frac{1}{4} \left(\frac{Cr}{x} \right)^3 \right] \left[1 + \frac{1}{4} \left(\frac{Cr}{x} \right)^2 \right]^{-2} \quad (5.12)$$

for the radial velocity, where μ is the fluid viscosity and C an integration constant equal to

$$C = \sqrt{\frac{3\rho J}{16\pi\mu^2}}. \quad (5.13)$$

The fed jets' momentum rate, J , considering a fully developed parallel parabolic velocity profile at the inlet is given by

$$J = \int_0^R \left(2u_{\text{inj}} \frac{R^2 - r^2}{R^2} \right)^2 2\pi\rho r \, dr = \frac{\pi}{3} \rho u_{\text{inj}}^2 d^2 \quad (5.14)$$

where $R = d/2$ is the injector radius.

Knowing the jet velocity profile from the NAJ model, the kinetic energy rate ratio of Equation (5.7) can be estimated as

$$\frac{\dot{E}_{K,1}(l_1)}{\dot{E}_{K,2}(l_2)} \equiv \frac{1/2 \int_0^\infty u_1(l_1, r)^3 2\pi\rho_1 r \, dr}{1/2 \int_0^\infty u_2(l_2, r)^3 2\pi\rho_2 r \, dr} = \frac{l_1}{l_2} \quad (5.15)$$

which results in

$$\sqrt{\phi_K \frac{\text{Re}_1}{\text{Re}_2} \frac{d_1}{d_2}} = \frac{l_1}{l_2} \quad (5.16)$$

where ϕ_K is the jets' kinetic energy feeding rate ratio defined as

$$\phi_K = \frac{\rho_1 d_1^2 u_{\text{inj},1}^3}{\rho_2 d_2^2 u_{\text{inj},2}^3}, \quad (5.17)$$

where u_{inj} is the average jet inlet velocity.

As mentioned, the NAJ model assumes a virtual source point of momentum that is located inside the injector tube, non-coincident with the chamber inlets. The distance from the source point to the chamber inlet, l' , is chosen in this analysis to be equal to the distance traveled by the jet for it to have a kinetic energy equal to the feeding kinetic energy, i.e.,

$$\frac{1}{2} \int_0^\infty u(l', r)^3 2\pi\rho r \, dr = \frac{1}{2} \int_0^R \left(2u_{\text{inj}} \frac{R^2 - r^2}{R^2} \right)^3 2\pi\rho r \, dr. \quad (5.18)$$

Solving Equation (5.18) for l' returns

$$l' = \frac{\text{Re}}{20} d. \quad (5.19)$$

The existence of momentum point sources from the NAJ model replaces the initial hypothesis that $l_1 + l_2 = D$ by the relation $l_1 + l_2 = D + l'_1 + l'_2$, i.e., the total sum of the two springs lengths must now be equal to the chamber diameter plus the distances from the inlets to the momentum source points. Despite this new consideration for the springs' total length, it can be seen that Equations (5.3) to (5.5) and all the model derivation up to this point remain unchanged. This consideration was not introduced earlier only to allow a clearer explanation of the model.

Defining each spring/jet length from the momentum point source until the impingement point position, x_{IP} , as

$$\begin{cases} l_1 = \frac{D}{2} + x_{IP} + l'_1 \\ l_2 = \frac{D}{2} - x_{IP} + l'_2 \end{cases} \quad (5.20)$$

Equation (5.16) becomes

$$\frac{D/2 + x_{IP} + l'_1}{D/2 - x_{IP} + l'_2} = \sqrt{\phi_K \frac{\text{Re}_1 d_1}{\text{Re}_2 d_2}}. \quad (5.21)$$

From Equation (5.21) a non-dimensional impingement point position, $\xi = x_{IP}/(D/2)$, can then be estimated as

$$\xi = \frac{\left(\frac{\text{Re}_2 d_2}{10 D} + 1 \right) \sqrt{\phi_K \frac{\text{Re}_1 d_1}{\text{Re}_2 d_2}} - \left(\frac{\text{Re}_1 d_1}{10 D} + 1 \right)}{\sqrt{\phi_K \frac{\text{Re}_1 d_1}{\text{Re}_2 d_2}} + 1}, \quad (5.22)$$

valid for $|\xi| \leq 1$ since the jets are bounded by the chamber walls and cannot expand or shrink until the momentum source points, which are located outside the mixing chamber volume.

This model makes it possible to set operational and geometrical conditions, according to the monomers' or pre-polymers physical properties or volumetric stoichiometry, to ensure that the two jets impinge in the center of the mixing chamber.

The jets' behavior predicted by the elastic analogue model will be compared and validated in Section 5.3 with CFD simulations and in Section 5.4 with PIV experimental results.

5.3 Axisymmetric CFD Simulations

In this section, the interaction of two opposed confined impinging jets is studied from axisymmetric CFD simulations. The impingement point position under different operational conditions and for different injectors diameters is obtained from the simulations and compared with the value predicted by the elastic analogue model. The studied operational variables were the jets Reynolds number and the jets' kinetic energy rate ratio in mixing chambers with jets with equal and unequal diameters.

The model, geometrical domain and numerical methods are described in Sections 5.3.1 to 5.3.4. The results from the simulations of the jets' impingement are presented and compared with the elastic analogue model in Section 5.3.5.

5.3.1 Simulation Domain

To study the interaction of the two opposed confined impinging jets, an axisymmetric CFD model of the flow in the mixing chamber was developed. Physically, the model describes the impingement of the two axisymmetric jets between two parallel plates, which can be seen as a simplification of the flow in the RIM machine mixing chamber, not taking into account the mixing chamber wall curvature. This model also does not allow radially non-symmetric solutions of the flow field, and thus is not

suited for dynamic flow simulations. Focus is given here to the jets and the region surrounding the impingement point, and it is assumed that secondary flows in the chamber do not have relevant impact in the jets' impingement point position. The developed axisymmetric CFD model offers a faster and simpler method to study the impact of different geometrical and operational parameters and to validate the elastic analogue analysis described in Section 5.2.

The physical domain is schematized in Figure 5.2. It consists of a T-shaped 2D domain that, by rotation around the axis, corresponds to the volume between two parallel plates with two cylindrical injectors connected normally to the plates. The distance from the rotation axis to the outlet boundary was set equal to $3D$ to ensure enough length for the flow to exit parallel to the walls. The injectors were simulated with a length equal to D . The geometrical parameters of Table 5.1 were assumed.

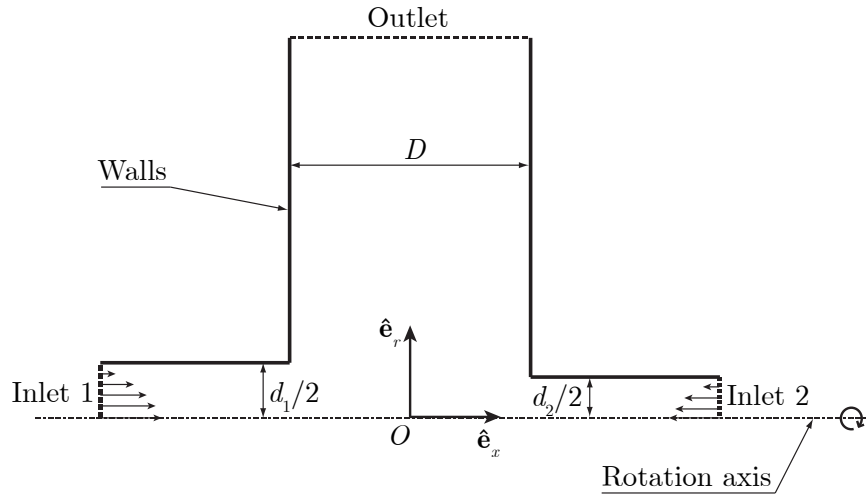


Figure 5.2. Schematic representation of the axisymmetric model used in the CFD simulations.

Table 5.1. Geometrical parameters of the mixing chambers for the axisymmetric CFD simulations.

Chamber No.	D (mm)	d_1 (mm)	d_2 (mm)
1	10	1.5	1.5
2	10	1	2
3	10	2.5	2.5
4	10	2	1.5
5	20	1.5	1.5

5.3.2 Governing Equations and Boundary Conditions

The governing equations that describe the steady laminar flow of the two incompressible impinging axisymmetric jets in the simulation domain are

$$\partial_x u + \frac{1}{r} \partial_r (r v) = 0, \quad (5.23)$$

$$\rho \left(u \partial_x u + v \partial_r u \right) = -\partial_x p + \mu \left[\partial_{xx} u + \frac{1}{r} \partial_r (r \partial_r u) \right] \text{ and} \quad (5.24)$$

$$\rho \left(u \partial_x v + v \partial_r v \right) = -\partial_r p + \mu \left[\partial_{xx} v + \frac{1}{r} \partial_r (r \partial_r v) - \frac{v}{r^2} \right] \quad (5.25)$$

for mass, axial momentum and radial momentum conservation, respectively. Here x and r represent the axial and radial coordinates, respectively, p the pressure and u and v represent the axial and radial velocities, respectively.

The conservation equations are to be solved for the following boundary conditions. A fully developed parabolic velocity profile at the inlets given by

$$\mathbf{u} = -u(r)\mathbf{n} = -2u_{\text{inj}} \frac{\left(\frac{d}{2}\right)^2 - r^2}{\left(\frac{d}{2}\right)^2} \mathbf{n} \quad (5.26)$$

for $\mathbf{u} = u \hat{\mathbf{e}}_x + v \hat{\mathbf{e}}_r$. The vector $\mathbf{n}$ represents here the normal unitary vector to the outside normal unitary vector to the boundaries of the domain and $r \in [0, d/2]$ is the radial position at the injectors' circular section area. A uniform and constant reference pressure was set at the outlet, $p = p_0$, equal to the atmospheric pressure. The no-slip condition was assumed at the chamber walls, $\mathbf{u} = \mathbf{0}$, and axisymmetry at the axis setting $v = 0$ and $\partial_r u = 0$. A density of $\rho = 1.18 \times 10^3 \text{ kg/m}^3$ and a viscosity of $\mu = 2 \times 10^{-2} \text{ Pa} \cdot \text{s}$ were assumed for the working fluid physical properties, similar to the fluid properties of the experiments described in Section 5.4.

5.3.3 Discretization

The simulation domain was discretized with uniformly sized quadrilateral finite volumes with $25 \mu\text{m}$ of length. The chosen grid element size was compared with the smallest structure possible in a turbulent flow, the Kolmogorov scale, to ensure that all the hydrodynamic scales were being correctly simulated without the need for turbulence modeling. The Kolmogorov scale, λ_K , in an isotropic and homogeneous turbulent flow can be estimated from

$$\lambda_K = \left(\frac{\nu^3}{\varepsilon} \right)^{\frac{1}{4}} \quad (5.27)$$

where ε is the energy dissipation rate of the flow into heat. The energy dissipation rate can be estimated from a balance to the kinetic energy rate over the simulation domain boundary surface, $\partial\Omega$, neglecting the contribution of friction at the walls. The energy dissipation rate is approximately given by

$$\varepsilon \approx \frac{\iint_{\partial\Omega} \frac{|\mathbf{u}|^2}{2} \rho (\mathbf{u} \cdot \mathbf{n}) dS}{\bar{\rho} V_{\text{dissipation}}} \quad (5.28)$$

where $\bar{\rho}$ is the average density weighted by the volumetric flow rate, Q , given by

$$\bar{\rho} = \frac{Q_1 \rho_1 + Q_2 \rho_2}{Q_1 + Q_2}, \quad (5.29)$$

and $V_{\text{dissipation}} = \left(\frac{\pi}{4} D^2 \right) (nD)$ is the volume of the cylindrical mixing chamber where dynamic impingement mixing occurs and nD , the distance from the top of the mixing chamber to the point where dynamic mixing still occurs, has been observed to be $nD \approx 2D$ to $3D$ (Kolodziej et al., 1982). Despite the differences in geometry from the axisymmetric and cylindrical chambers, $V_{\text{dissipation}}$ is here assumed to be close in both cases. Since the outlet velocity is at least one order of magnitude smaller than

the inlet velocities due to the difference of flow areas, the kinetic energy rate of the exiting fluid was also neglected. Assuming fully developed parabolic velocity profiles at the inlets, Equation (5.28) results in

$$\varepsilon \approx \frac{\int_0^{d_1/2} \left(2u_{\text{inj},1} \frac{(d_1/2)^2 - r^2}{(d_1/2)^2} \right)^3 \rho_1 \pi r \, dr + \int_0^{d_2/2} \left(2u_{\text{inj},2} \frac{(d_2/2)^2 - r^2}{(d_2/2)^2} \right)^3 \rho_2 \pi r \, dr}{\bar{\rho} V_{\text{dissipation}}}. \quad (5.30)$$

Assuming equal density and viscosity for both fluids, the condition of the present computational study, the smallest hydrodynamic scale of the flow can be estimated from

$$\lambda_K = \left(\frac{D^3 n d_1}{1 + 1/\phi_K} \right)^{\frac{1}{4}} \text{Re}_1^{-\frac{3}{4}} \quad (5.31)$$

where the feeding kinetic energy rate ratio, ϕ_K , is given by Equation (5.17).

For $\text{Re} = 600$ (the highest value of Re in the range of industrial practice), $n = 2.5$, $d_1 = 1.5 \text{ mm}$, $D = 10 \text{ mm}$ (typical mixing chamber dimensions) and $\phi_K = 1$, a value around $50 \text{ } \mu\text{m}$ is estimated from Equation (5.31). This estimated flow scale shows that, even if one were dealing with homogeneous and isotropic turbulence, which is not the case for the simulated flow regimes, all the hydrodynamic flow scales would be correctly simulated with the selected grid spacing, without the need for further refinement or turbulence modeling. A conservative element size of $25 \text{ } \mu\text{m}$ was chosen to have a better spatial discretization for the impingement point position determination.

Geometrical domains and simulation grids for the simulations were generated with the software packages DesignModeler and Meshing included in Ansys® 14 suite.

5.3.4 Numerical Solution

The flow equations, Equations (5.23) to (5.25), were solved with the finite-volume commercial CFD solver ANSYS Fluent[®] 14 in an Intel[®] Xeon[®] 3 GHz 8-core PC with Windows 7 64-bits. The advective terms of Equations (5.23) to (5.25) were discretized with the Third-Order MUSCL scheme for momentum and the PRESTO! scheme for pressure. The pressure-velocity coupling scheme was set to Coupled and gradients were computed with the Least Squares Cell Based scheme. All flow variables were initialized with a value equal to zero and solution was accepted for residues lower than 10^{-6} .

5.3.5 Results

In this section the results from the axisymmetric CFD simulations of the flow are shown and compared with the model proposed in Section 5.2. Steady state simulations of the flow were obtained for different injectors and mixing chamber diameters. The effect of the jets' feeding kinetic energy rate ratio at different Reynolds numbers was also studied. The Reynolds numbers was maintained at low values to be possible to obtain steady state solutions. The simulated operational are shown in Table 5.2 to Table 5.6.

Table 5.2. Operational conditions considered in the axisymmetric CFD simulations for Chamber no. 1.

Chamber No.	Re_1	Re_2	$u_{inj,1}$ (m/s)	$u_{inj,2}$ (m/s)	ϕ_K	ϕ_M	ϕ_{FR}
1	50	108	0.565	1.22	0.10	0.22	0.46
1	50	85	0.565	0.966	0.20	0.34	0.58
1	50	63	0.565	0.712	0.50	0.63	0.79
1	50	50	0.565	0.565	1.00	1.00	1.00
1	50	40	0.565	0.448	2.00	1.59	1.26
1	50	29	0.565	0.330	5.00	2.92	1.71
1	50	23	0.565	0.262	10.0	4.64	2.15
1	100	215	1.13	2.43	0.10	0.22	0.46
1	100	171	1.13	1.93	0.20	0.34	0.58
1	100	126	1.13	1.42	0.50	0.63	0.79
1	100	100	1.13	1.13	1.00	1.00	1.00
1	100	79	1.13	0.897	2.00	1.59	1.26
1	100	58	1.13	0.661	5.00	2.92	1.71
1	100	46	1.13	0.524	10.0	4.64	2.15

5. Impinging Jets Interaction

Table 5.3. Operational conditions considered in the axisymmetric CFD simulations for Chamber no. 2.

Chamber No.	Re_1	Re_2	$u_{inj,1}$ (m/s)	$u_{inj,2}$ (m/s)	ϕ_K	ϕ_M	ϕ_{FR}
2	50	136	0.847	1.15	0.10	0.14	0.18
2	50	108	0.847	0.913	0.20	0.22	0.23
2	50	79	0.847	0.673	0.50	0.40	0.31
2	50	63	0.847	0.534	1.00	0.63	0.40
2	50	50	0.847	0.424	2.00	1.00	0.50
2	50	37	0.847	0.312	5.00	1.84	0.68
2	50	29	0.847	0.248	10.0	2.92	0.85
2	100	271	1.69	2.30	0.10	0.14	0.18
2	100	215	1.69	1.83	0.20	0.22	0.23
2	100	159	1.69	1.35	0.50	0.40	0.31
2	100	126	1.69	1.07	1.00	0.63	0.40
2	100	100	1.69	0.847	2.00	1.00	0.50
2	100	74	1.69	0.624	5.00	1.84	0.68
2	100	58	1.69	0.496	10.0	2.92	0.85

Table 5.4. Operational conditions considered in the axisymmetric CFD simulations for Chamber no. 3.

Chamber No.	Re_1	Re_2	$u_{inj,1}$ (m/s)	$u_{inj,2}$ (m/s)	ϕ_K	ϕ_M	ϕ_{FR}
3	50	108	0.339	0.730	0.10	0.22	0.46
3	50	85	0.339	0.580	0.20	0.34	0.58
3	50	63	0.339	0.427	0.50	0.63	0.79
3	50	55	0.339	0.373	0.75	0.83	0.91
3	50	50	0.339	0.339	1.00	1.00	1.00
3	50	45	0.339	0.308	1.33	1.21	1.10
3	50	40	0.339	0.269	2.00	1.59	1.26
3	50	29	0.339	0.198	5.00	2.92	1.71
3	50	23	0.339	0.157	10.0	4.64	2.15
3	100	215	0.678	1.46	0.10	0.22	0.46
3	100	171	0.678	1.16	0.20	0.34	0.58
3	100	126	0.678	0.854	0.50	0.63	0.79
3	100	110	0.678	0.746	0.75	0.83	0.91
3	100	100	0.678	0.678	1.00	1.00	1.00
3	100	91	0.678	0.616	1.33	1.21	1.10
3	100	79	0.678	0.538	2.00	1.59	1.26
3	100	58	0.678	0.396	5.00	2.92	1.71
3	100	46	0.678	0.315	10.0	4.64	2.15

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Table 5.5. Operational conditions considered in the axisymmetric CFD simulations for Chamber no. 4.

Chamber No.	Re_1	Re_2	$u_{inj,1}$ (m/s)	$u_{inj,2}$ (m/s)	ϕ_K	ϕ_M	ϕ_{FR}
4	50	98	0.424	1.11	0.10	0.26	0.68
4	50	78	0.424	0.878	0.20	0.41	0.86
4	50	57	0.424	0.647	0.50	0.76	1.16
4	50	50	0.424	0.565	0.75	1.00	1.33
4	50	45	0.424	0.513	1.00	1.21	1.47
4	50	41	0.424	0.466	1.33	1.47	1.62
4	50	36	0.424	0.407	2.00	1.92	1.85
4	50	27	0.424	0.300	5.00	3.54	2.51
4	50	21	0.424	0.238	10.0	5.62	3.16
4	100	196	0.847	2.21	0.10	0.26	0.68
4	100	155	0.847	1.76	0.20	0.41	0.86
4	100	114	0.847	1.29	0.50	0.76	1.16
4	100	100	0.847	1.13	0.75	1.00	1.33
4	100	91	0.847	1.03	1.00	1.21	1.47
4	100	83	0.847	0.934	1.33	1.47	1.61
4	100	72	0.847	0.815	2.00	1.92	1.85
4	100	53	0.847	0.600	5.00	3.54	2.51
4	100	42	0.847	0.477	10.0	5.62	3.16

Table 5.6. Operational conditions considered in the axisymmetric CFD simulations for Chamber no. 5.

Chamber No.	Re_1	Re_2	$u_{inj,1}$ (m/s)	$u_{inj,2}$ (m/s)	ϕ_K	ϕ_M	ϕ_{FR}
5	50	108	0.565	1.22	0.10	0.22	0.46
5	50	85	0.565	0.966	0.20	0.34	0.58
5	50	63	0.565	0.712	0.50	0.63	0.79
5	50	50	0.565	0.565	1.00	1.00	1.00
5	50	40	0.565	0.448	2.00	1.59	1.26
5	50	29	0.565	0.330	5.00	2.92	1.71
5	50	23	0.565	0.262	10.0	4.64	2.15
5	100	215	1.13	2.43	0.10	0.22	0.46
5	100	171	1.13	1.93	0.20	0.34	0.58
5	100	126	1.13	1.42	0.50	0.63	0.79
5	100	100	1.13	1.13	1.00	1.00	1.00
5	100	79	1.13	0.897	2.00	1.59	1.26
5	100	58	1.13	0.661	5.00	2.92	1.71
5	100	46	1.13	0.524	10.0	4.64	2.15

After Equations (5.23) to (5.25) were solved for the conditions of Table 5.2 to Table 5.6, the axial velocity profile along the rotation axis was extracted from the simulation results. The jets' impingement point position was determined as the rotation axis' axial coordinate at which the axial velocity equals zero,

$$u(x_{IP}, r = 0) = 0, x_{IP} \in [-D/2, D/2]. \quad (5.32)$$

Figure 5.3 to Figure 5.8 show the obtained values of the non-dimensional impingement position from the CFD simulations and the values predicted by Equation (5.22) for $Re_1 = 50$ and 100. Values of ξ are shown for the different chamber geometrical parameters and as a function of the jets' kinetic energy rate ratio, the momentum rate ratio and the flow rate ratio.

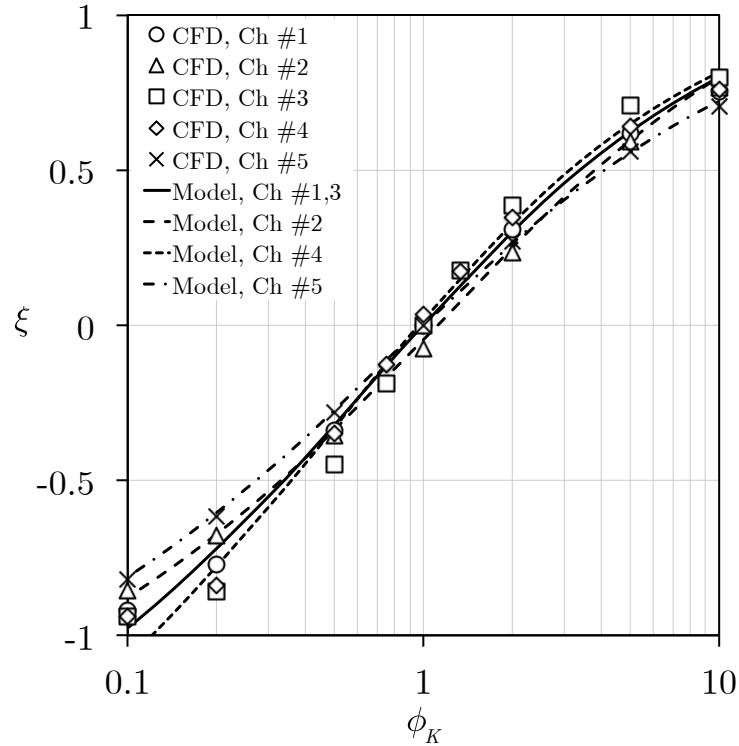


Figure 5.3. Non-dimensional impingement point position from the axisymmetric CFD simulations and elastic analogue model as a function of the jets' kinetic energy rate ratio at $Re_1=50$.

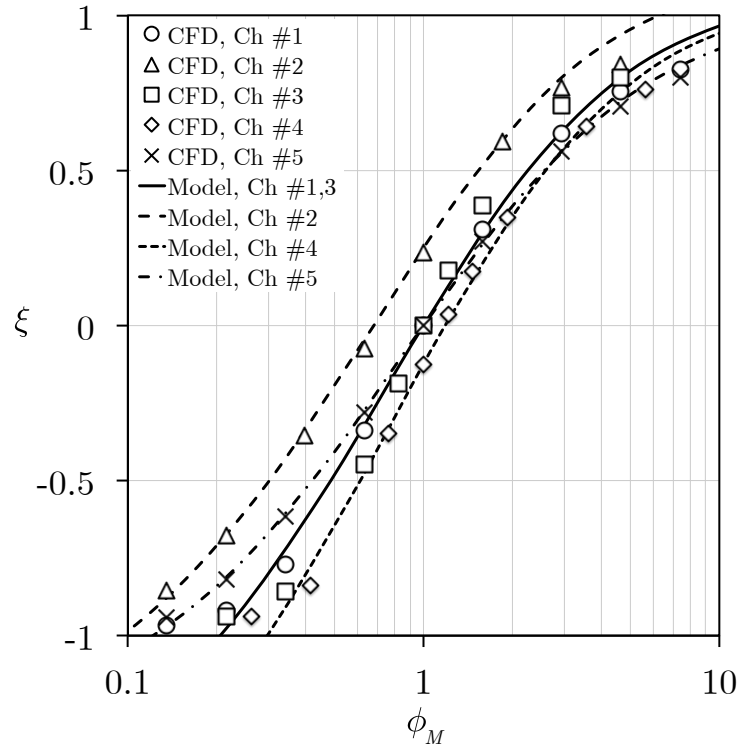


Figure 5.4. Non-dimensional impingement point position from the axisymmetric CFD simulations and elastic analogue model as a function of the jets' momentum rate ratio at $Re_1=50$.

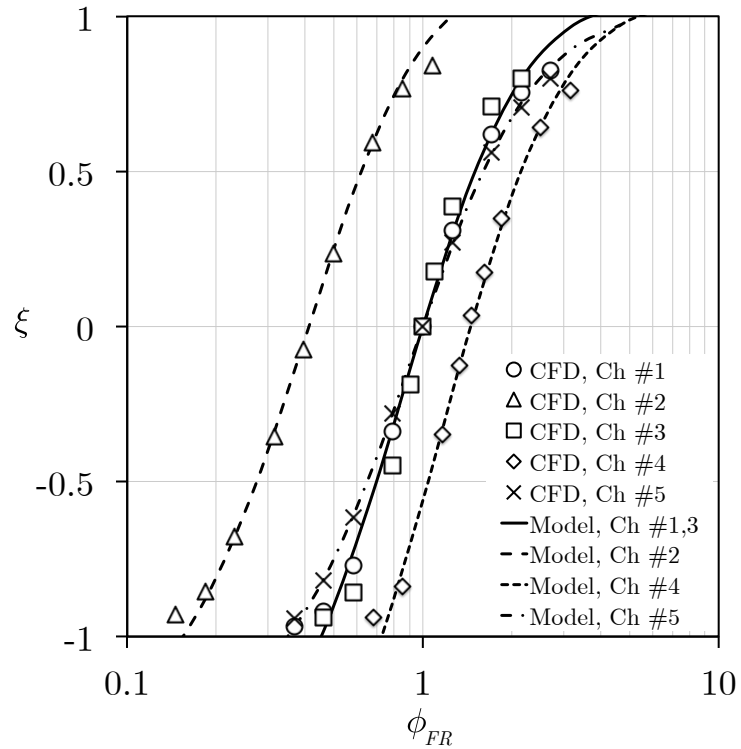


Figure 5.5. Non-dimensional impingement point position from the axisymmetric CFD simulations and elastic analogue model as a function of the jets' flow rate ratio at $Re_1=50$.

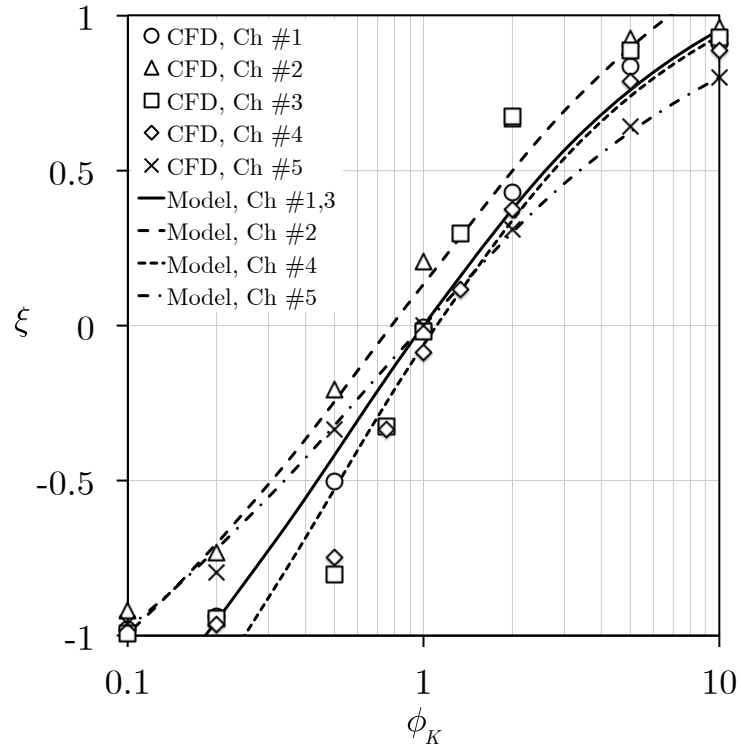


Figure 5.6. Non-dimensional impingement point position from the axisymmetric CFD simulations and elastic analogue model as a function of the jets' kinetic energy rate ratio at $Re_1=100$.

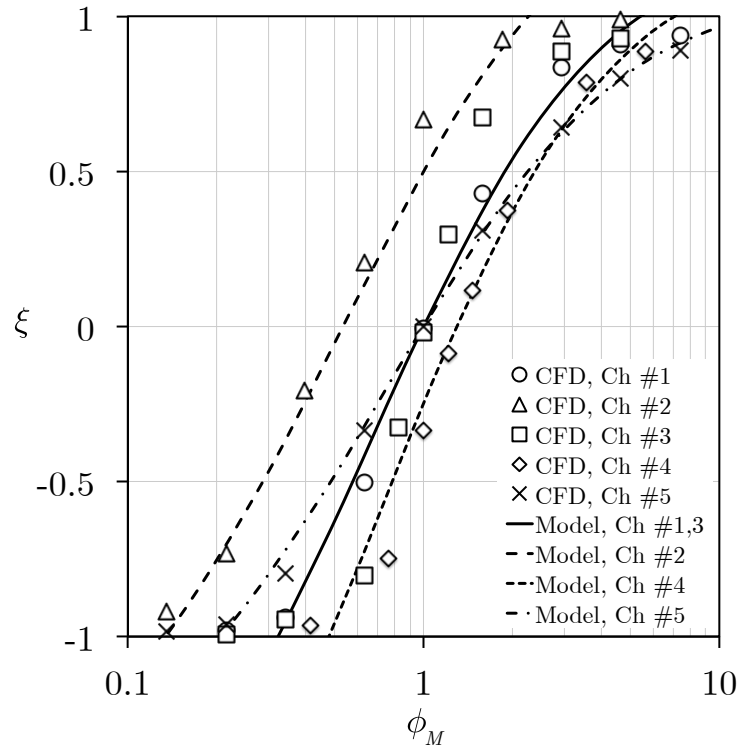


Figure 5.7. Non-dimensional impingement point position from the axisymmetric CFD simulations and elastic analogue model as a function of the jets' momentum rate ratio at $Re_1=100$.

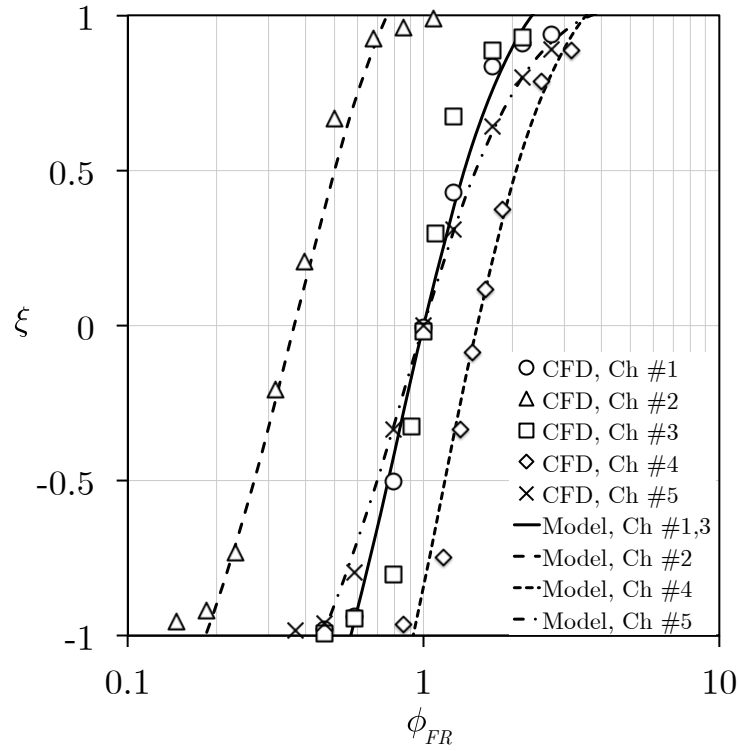


Figure 5.8. Non-dimensional impingement point position from the axisymmetric CFD simulations and elastic analogue model as a function of the jets' flow rate ratio at $Re_1=100$.

As observed in previous works (see Section 5.1), Figure 5.3 to Figure 5.8 show, from both CFD results and the elastic analogue model, that increasing or decreasing one of the jets' flow rate has the effect of displacing the impingement point position from the center of the mixing chamber towards the walls. Results show that the representation of ξ as a function of ϕ_K tends to unify the jets' behavior and overlap the curves for all the chamber designs, with some deviation, however, depending on the chamber's injector diameter ratio. Representing ξ as a function of ϕ_M or ϕ_{FR} tends to separate the curves, showing clearly different behaviors for each chamber with different injector diameter ratio.

Results also show an asymmetric behavior of the impinging jets' interaction. Maintaining one injector fixed and increasing the flow rate of the other does not produce the same impingement point displacement as decreasing the flow rate of the changing injector. For equal injectors it is observed that

$$\xi(\phi_K) \neq -\xi(\phi_K^{-1}), \quad (5.33)$$

for $\phi_K \neq 1$, revealing the role of the jets' expansion/dissipation inside the chamber on its interaction, considered in the elastic model. This asymmetry occurs due to the jets remaining narrower inside the mixing chamber for higher inlet velocities, dissipating less energy per energy fed in the same traveling length. The same is observed for unequal injectors showing asymmetric curves at the left and right of the impingement point at the center of the chamber.

As also reported by Johnson (2000), it is observed that if ϕ_K is maintained constant and different from 1, and Re is increased in both jets accordingly, the impingement point moves closer to the side of the jet with the lowest Re, towards the chamber walls. This phenomenon indicates that in RIM the system becomes more sensitive to small deviations in the monomers flow rates or environmental conditions as Re is increased, demanding a more precise and strict flow rate control.

Overall, Figure 5.3 to Figure 5.8 show good agreement between the CFD simulations results and the elastic analogue model. As expected, the model shows some divergence to the CFD results as the impingement point position gets closer to the walls. Results also seem to indicate that a better description of the jets' interaction from the model is obtained as the distance between injectors increased. The model is also successful in the description of the interaction of jets with different diameters and the prediction of the increasing system sensitivity to flow rate variations at higher Re.

5.4 PIV Experiments

The impinging jets interaction was experimentally studied with Particle Image Velocimetry (PIV), namely the flow regime transition and jet's impingement point position under different operation conditions and mixing chamber designs. The experimental set-up and methods are described in Section 5.4.1 and the experimental

conditions are resumed in Section 5.4.2. Results are shown and discussed in Sections 5.4.3 to 5.4.6.

5.4.1 Experimental Set-Up and Methods

The principles of PIV are based in the measurement of the displacement of small tracer particles carried by the flow during a short time interval (Tropea et al., 2007). Two consecutive images (frames) of a seeded flow with the tracer particles are taken. The images are divided into smaller regions (interrogation regions) and each is processed statistically to obtain a two-dimensional velocity vector field. A 2D velocity vector field inside three different mixing chamber geometries was obtained in a plane resulting from an axial cut of the chamber through the injectors.

The experimental set-up, flow seeding and image acquisition and processing techniques used in the PIV experiments are described next in Sections 5.4.1.1 to 5.4.1.4.

5.4.1.1 Mixing Chambers and Hydraulic Loop

Different transparent mixing chambers were built from single $150 \times 40 \times 47.5$ mm Plexiglas blocks, which were cut and drilled to make the circular holes corresponding to the injectors and the mixing chamber (see Figure 5.9 to Figure 5.11). For the geometries with equal injectors, the blocks were drilled from side to side to make perfectly aligned injectors. For the geometry with unequal injectors, the injectors' length was shortened to guarantee the best alignment possible. In this case, the Plexiglas block was cut and a hole with the smaller injector diameter was drilled from side to side. After this procedure, one of the sides was drilled again to obtain a larger injector diameter. The final diameter and alignment of the injectors was checked with calibrated rods and from macro photographs of the chamber.

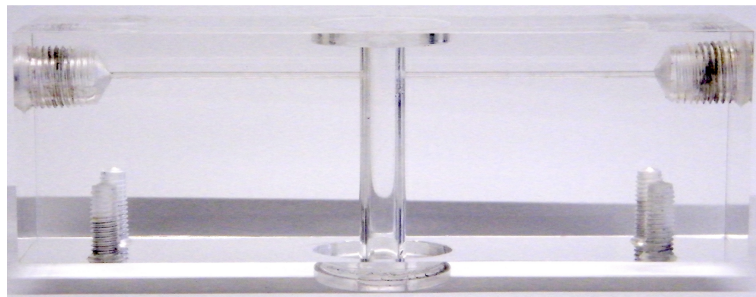


Figure 5.9. Views of the transparent mixing chamber no. 1.

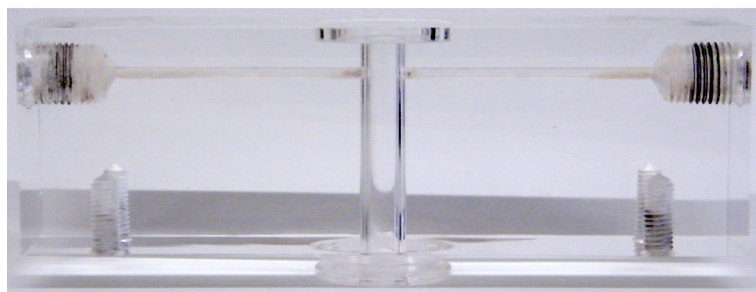
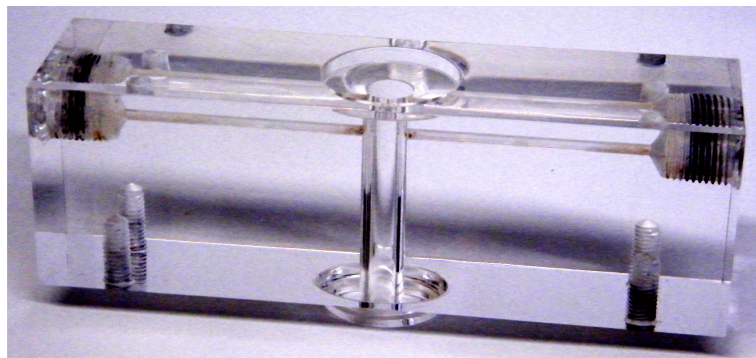


Figure 5.10. Views of the transparent mixing chamber no. 2.

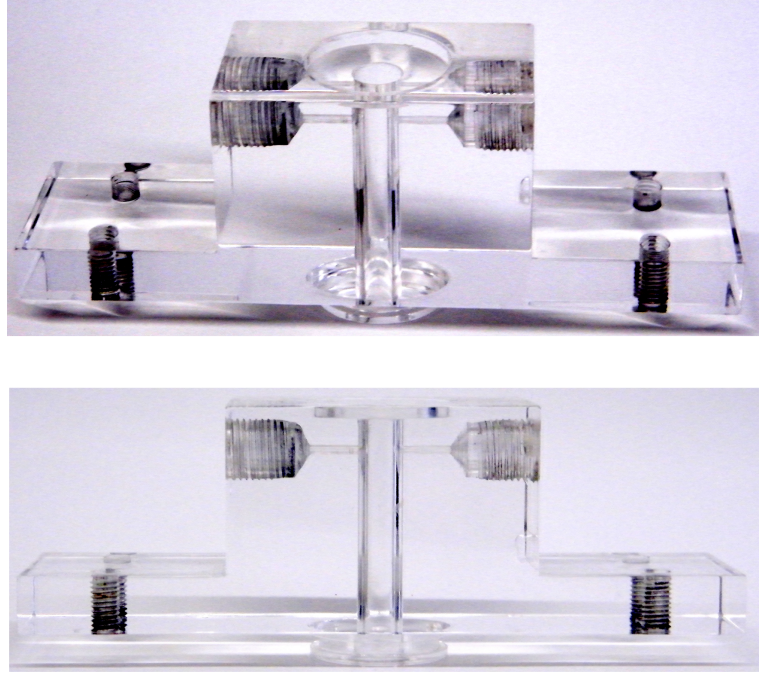


Figure 5.11. Views of the transparent mixing chamber no. 3.

For all the chambers, the injectors are placed at 5 mm from the top making an angle of 180° between each other. The mixing chamber was connected to a cylindrical container with 15 cm diameter and 8 cm height, which replicates the operation of mold filling. Geometrical parameters of the 3 mixing chambers used in the PIV experiments are resumed in Table 5.7. Figure 5.12 show schematic drawings of the mixing chambers and the mold.

Table 5.7. Geometrical parameters of the mixing chambers for the PIV experiments.

Chamber No.	D (mm)	H (mm)	d_1 (mm)	d_2 (mm)
1	10	50	1.5	1.5
2	10	50	2.5	2.5
3	10	50	1.9	1.5

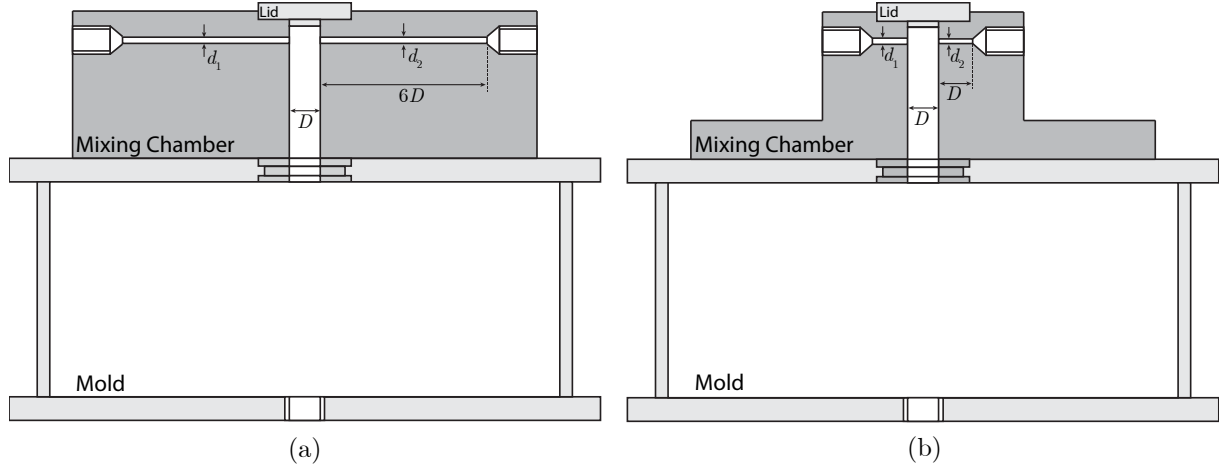


Figure 5.12. Schematic representation of the transparent mixing chambers with injectors of equal (a) and unequal (b) diameters and the mold.

The mixing chambers were fed with the working fluid contained in two pressurized tanks through a hydraulic loop. The flow rate at each injector was controlled with needle valves placed in the hydraulic loop between the pressurized tanks and the mixing chamber inlets. The flow rate at each injector was adjusted using two ROTA-Yokogawa Coriolis mass flow meters (model RCCS33 M01A1SH) with a precision of ± 0.05 g/s in the range from 1.2 g/s to 13.2 g/s of this work, corresponding to a maximum relative error of 10% for the minimum mass flow rate used in the experiments, $\dot{m}_{\min} = 0.5$ g/s.

After passing the mixing chamber and the mold, the working fluid was collected in a third container for reutilization. Figure 5.13 shows a photograph of the experimental set-up and its schematic representation.

The fluid used in the experiment was a solution of 70% (w/w) of glycerin in water. The dependence of the solution viscosity on temperature was determined with a cone and plate rheometer (Physica UDS 200), shown in Figure 5.14 in the range from 15 °C to 35 °C. The Coriolis mass flow meters allowed to measure, for each experiment, the fluid temperature and density.

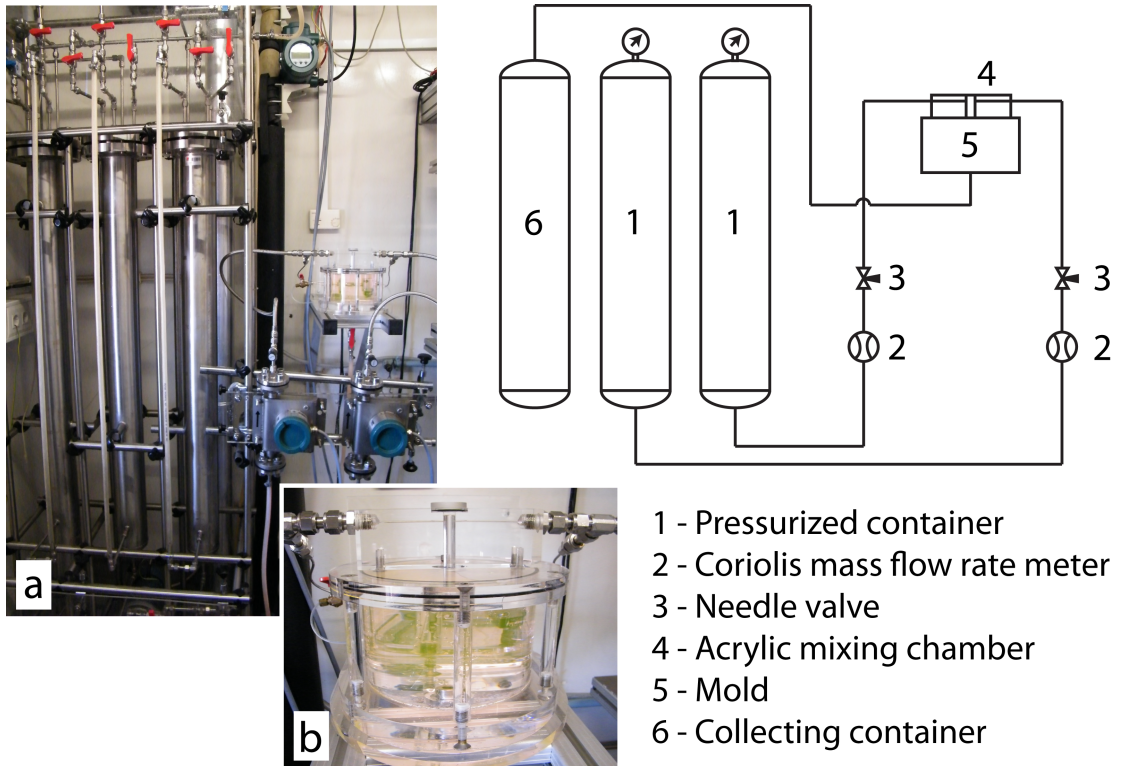


Figure 5.13. Image of the hydraulic loop (a-whole setup, b-detail of one transparent mixing chamber and mold) and its schematic representation.

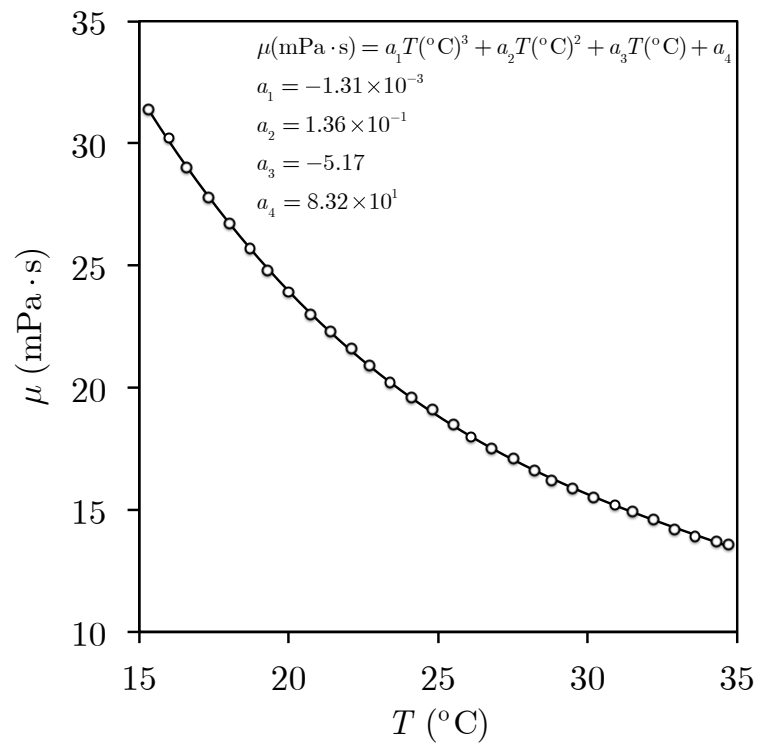


Figure 5.14. Experiments working fluid viscosity as a function of temperature.

5.4.1.2 Seeding

The working fluid was seeded with spherical nylon particles (TSI[®], model 10084) with a concentration that ensured at least 5 to 15 particles per interrogation region, a general rule of thumb for PIV analysis. The spherical nylon particles have a nominal diameter of $d_p = 4 \mu\text{m}$ and density $\rho_p = 1.14 \text{ g} \cdot \text{cm}^{-3}$. Care was taken to confirm that the chosen particles would follow passively the flow, that is guaranteed if the Stokes number, St , is maintained less than 0.1. Under this condition, the particles follow the flow with errors below 1% (Tropea et al., 2007). The Stokes number, the ratio between the particle's characteristic response time and the characteristic flow time scale, can be estimated as (Icardi et al., 2011)

$$St = \frac{\dot{\gamma} \rho_p d_p^2}{12\mu} \quad (5.34)$$

where $\dot{\gamma}$ is a characteristic rate of strain of the flow. For the value of $\dot{\gamma}$ a conservative value equal to the rate of strain at the injectors, given by

$$\dot{\gamma} = \frac{8u_{mj}}{d}, \quad (5.35)$$

was used. For $Re = 600$, a value higher than the maximum considered for the experiments, and an injector diameter of $d = 1.5 \text{ mm}$ at 24°C , the Stokes number is equal to 3×10^{-3} . This value is 3 times smaller than the desired one ensures that the particles followed the flow accurately in the experiments.

5.4.1.3 PIV Image Acquisition System

To obtain the images of the particles for the vector field extraction, the transparent mixing chambers were illuminated with a laser sheet, cutting the chamber axially through the injectors, making the particles only visible in a thin slice of the volume by scattering of the laser light. The laser source used is a Nd:YAG double-head pulsed laser from Litron Lasers[®] Nano L 50-100, which emits 4 ns pulsed beams with

maximum energy of 400 mJ/pulse at a maximum rate of 100 Hz and a wavelength of 532 nm. The laser beams were transported to the top of the mixing chamber with a TSI light arm (Model 610015), a telescopic and articulated 1.8 m hollow tube system with internal reflecting mirrors. At the input port of the light arm a collimated lens with a 10 m focal length was placed to control the divergence and prevent clipping of the beams. After traveling a distance correspondent to the light arm length, the laser beams were focused with a 500 mm spherical lens and spread in a sheet with a – 15 mm cylindrical lens. The extremity of the laser arm was placed at the top of the mixing chamber at a distance of 500 mm obtaining a laser sheet with approximately 1 mm of thickness. Apart from the laser light, no other light source was present in the set-up room during the experiments. Figure 5.15 shows a photograph of the PIV experimental set-up detailing the mixing chamber lighting and image acquisition system. Figure 5.16 shows a schematic representation of the PIV experimental set-up.

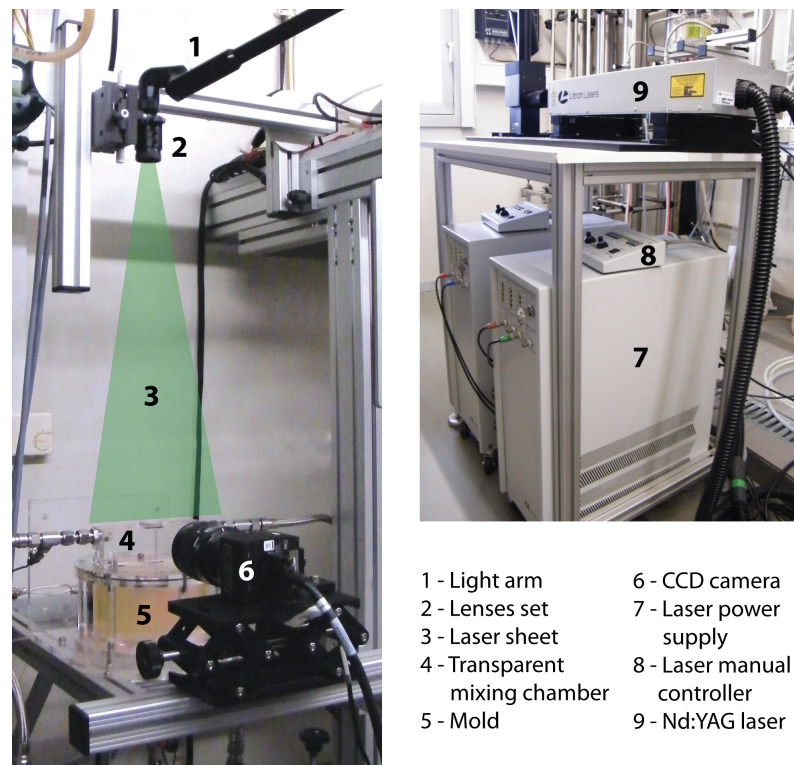


Figure 5.15. Photograph of the PIV experimental setup.

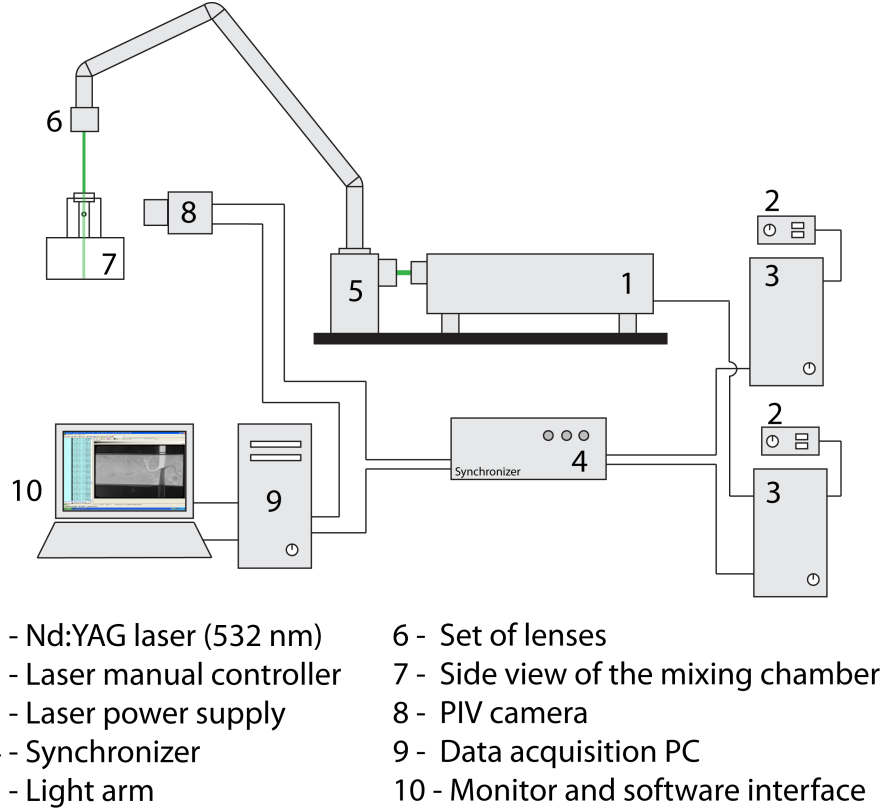


Figure 5.16. Schematic representation of the PIV experimental setup.

For each geometrical and operational condition, 300 pairs of laser-synchronized frames of the illuminated seeded flow were captured with a CCD camera. This camera is a TSI Model 630157 with the ability to capture 1192×1600 pixels images with a 16 bits scale of shades of grey at maximum frame rate of 30 Hz. The frames captured with the CCD camera are transferred to the computer through a frame grabber (TSI model 600067).

In order to capture approximately the same flow time relatively to the chamber residence time in all the experiments, the rate of acquisition of the 300 pairs of images was set accordingly to the Reynolds number. In PIV analysis, the system limits the camera to a frequency of image acquisition of 15 Hz. Considering a maximum value of $Re_1 = 400$ for the experiments, the laser frequency, f_{laser} , was set to the closest allowed frequency by the PIV system calculated from

$$f_{\text{laser}}(\text{Re}_1) = \frac{\text{Re}_1}{400} f_{\text{laser,max}} \quad (5.36)$$

where $f_{\text{laser,max}} = 15\text{Hz}$. Due to RAM limitations, the 300 images of each experiment were taken in two sets of 150, corresponding to a flow time $t \approx 80\tau$. The mean residence time inside the mixing chamber is given by

$$\tau = \frac{D^2 H}{u_{\text{inj},1} d_1^2 + u_{\text{inj},2} d_2^2}. \quad (5.37)$$

5.4.1.4 PIV Image Processing

Each captured pair of frames of the seeded flow was divided into search areas. The displacement of the particles was obtained from cross-correlation of each pair of search areas. The velocity field was found by dividing the particles displacement in each direction by the time interval between frames. The search area size was set to 64×64 pixels for interrogation regions of 16×16 pixels. The time interval between the laser pulse of each frame, Δt_{beams} , was set so that the particles could only move one fourth of a search area in that time interval,

$$\Delta t_{\text{beams}} = \frac{\Delta x_{\text{int. region}}}{2 \max\{u_{\text{inj},1}, u_{\text{inj},2}\}} \quad (5.38)$$

where $\Delta x_{\text{int. region}}$ is the length of each interrogation region, and was calculated from an initial image of the flow converting pixels into physical length. Further information on the PIV technique can be obtained in specialized literature on this topic such as Tropea et al. (2007).

For the time interval between laser pulses of Equation (5.38) the minimum velocity detected in the PIV analysis is equal to

$$u_{\min} = \frac{1}{8} \max \{u_{\text{inj},1}, u_{\text{inj},2}\}, \quad (5.39)$$

corresponding to a particle moving 1 pixel in the image. The average velocity inside the mixing chamber, u_{chamber} , has a smaller value given by

$$u_{\text{chamber}} = \frac{d^2}{D^2} (u_{\text{inj},1} + u_{\text{inj},2}). \quad (5.40)$$

Since focus is here given to the jets and not to the remaining flow, the minimum detectable velocity value was not considered an important parameter. To study with more detail the flow structures in zones with smaller velocities, far from the jets, Δt_{beams} or the number of pixels of the search area could have been increased. The first solution has the inconvenience of compromising the region of the jets and its surroundings since particles can have displacements between the consecutive frames bigger than the search area, making them undetectable. With the second option, a coarser vector map will be obtained and smaller flow structures can be missed. Figure 5.17 shows a captured image of a seeded flow in one of the transparent mixing chamber with the indication of the whole region of analysis (ROI) and the size of the search areas.



Figure 5.17. Photograph of a seeded flow in the transparent mixing chamber with the indication of the ROI and the search area size.

A TSI synchronizer (model 610035) was used for the system time management. Image acquisition and processing were done in a Dell® Precision 690 PC with a dual core Intel® Xeon® CPU at 2.33 GHz and 2 GB of RAM with TSI Insight 3G™ software (version 9.0).

Insufficient seeding, strong velocity gradients, and motion of the particles in and out of the laser sheet can result in low correlation peaks between the two successive frames. This can result in the appearance of “bad” or spurious velocity vectors that generally correspond to a discontinuity in the vector maps (Huang et al., 1997; Nogueira et al., 1997). Spurious vector detection was made by inspecting the smoothness or coherence expected in the velocity field, in which spurious vectors stand out with respect to the overall or local field (Tropea et al., 2007). Its elimination was made by allowing a maximum vector deviation from the field equal to three times the standard deviation computed from all the vectors in the ROI. The post-processing and validation of the vector maps was made with the software TSI Insight 3G™ software (version 9.0). It was observed visually during the analysis that the used settings for vector elimination were enough to comprehend all spurious vectors in the maps. Missing data points in the maps resulting from the validation step were filled with the value resulting from the interpolation of the surrounding velocity vectors.

Although the whole region of interest shown in Figure 5.17 was processed, the maps were only considered for $-0.4 \leq x/D \leq 0.4$ due to the poor quality of the results obtained closer to the chamber walls. Poor quality vectors next to the walls result from optical distortions due to the curvature of the chamber. The distortion problem was subject of analysis of Santos (2003) for the same mixing chamber geometry, having shown that the most extreme optical distortions occur near the chamber walls. In the final selected region of interest, negligible optical distortions occur.

5.4.2 Experiments

Table 5.8 to Table 5.20 resume the list of the experimental conditions with the three transparent mixing chambers to study the effect of jet unbalancing in the flow field.

Table 5.8. List of conditions for the experiments with the transparent mixing chamber no. 1 at $Re_1=50$.

Frames	$\dot{m}_1$ (g/s)	$\dot{m}_2$ (g/s)	ϕ_K	T (°C)	ρ (kg/m ³)	μ (mPa · s)	Δt_{beams} (μ s)	f_{laser} (Hz)
1-300	1.2	2.5	0.11	23.7	1.18	19.9	50	2
301-600	1.2	1.9	0.25	23.7	1.18	19.9	67	2
601-900	1.2	1.5	0.51	23.7	1.18	19.9	85	2
901-1200	1.2	1.3	0.79	23.7	1.18	19.9	97	2
1201-1500	1.2	1.2	1.0	23.7	1.18	19.9	107	2
1501-1800	1.2	1.1	1.3	23.7	1.18	19.9	107	2
1801-2100	1.2	0.9	2.4	23.7	1.18	19.9	107	2
2101-2400	1.2	0.7	5.0	23.7	1.18	19.9	107	2

Table 5.9. List of conditions for the experiments with the transparent mixing chamber no. 1 at $Re_1=100$.

Frames	$\dot{m}_1$ (g/s)	$\dot{m}_2$ (g/s)	ϕ_K	T (°C)	ρ (kg/m ³)	μ (mPa · s)	Δt_{beams} (μ s)	f_{laser} (Hz)
1-300	2.4	3.7	0.27	23.8	1.18	19.8	34	4
301-600	2.4	3.0	0.51	23.8	1.18	19.8	42	4
601-900	2.4	2.6	0.79	23.8	1.18	19.8	49	4
901-1200	2.4	2.4	1.00	23.8	1.18	19.8	53	4
1201-1500	2.4	2.1	1.49	23.8	1.18	19.8	53	4
1501-1800	2.4	1.9	2.02	23.8	1.18	19.8	53	4
1801-2100	2.4	1.5	4.10	23.8	1.18	19.8	53	4

Table 5.10. List of conditions for the experiments with the transparent mixing chamber no. 1 at $Re_1=200$.

Frames	$\dot{m}_1$ (g/s)	$\dot{m}_2$ (g/s)	ϕ_K	T (°C)	ρ (kg/m ³)	μ (mPa · s)	Δt_{beams} (μ s)	f_{laser} (Hz)
1-300	4.7	7.5	0.25	23.8	1.18	19.8	17	8.33
301-600	4.7	5.9	0.51	23.8	1.18	19.8	21	8.33
601-900	4.7	5.2	0.74	23.8	1.18	19.8	24	8.33
901-1200	4.7	4.7	1.00	23.4	1.18	20.2	27	8.33
1201-1500	4.7	4.3	1.31	23.4	1.18	20.2	27	8.33
1501-1800	4.7	3.7	2.05	23.8	1.18	19.8	27	8.33
1801-2100	4.7	3.0	3.85	23.8	1.18	19.8	27	8.33

5. Impinging Jets Interaction

Table 5.11. List of conditions for the experiments with the transparent mixing chamber no. 1 at $Re_1=300$.

Frames	$\dot{m}_1$ (g/s)	$\dot{m}_2$ (g/s)	ϕ_K	T (°C)	ρ (kg/m ³)	μ (mPa · s)	Δt_{beams} (μ s)	f_{laser} (Hz)
1-300	7.1	8.9	0.51	24.5	1.18	19.2	14	11.11
301-600	7.1	7.8	0.75	24.5	1.18	19.2	16	11.11
601-900	7.1	7.1	1.00	24.2	1.18	19.5	18	11.11
901-1200	7.1	6.4	1.37	24.5	1.18	19.2	18	11.11
1201-1500	7.1	5.6	2.04	24.5	1.18	19.2	18	11.11

Table 5.12. List of conditions for the experiments with the transparent mixing chamber no. 1 at $Re_1=400$.

Frames	$\dot{m}_1$ (g/s)	$\dot{m}_2$ (g/s)	ϕ_K	T (°C)	ρ (kg/m ³)	μ (mPa · s)	Δt_{beams} (μ s)	f_{laser} (Hz)
1-300	9.4	10.2	0.78	24.3	1.18	19.4	12	14.29
301-600	9.4	9.7	0.91	24.3	1.18	19.4	12	14.29
601-900	9.4	9.4	1.00	24.3	1.18	19.4	13	14.29
901-1200	9.4	9.0	1.14	24.3	1.18	19.4	13	14.29
1201-1500	9.4	8.7	1.26	24.3	1.18	19.4	13	14.29

Table 5.13. List of conditions for the experiments with the transparent mixing chamber no. 2 at $Re_1=50$.

Frames	$\dot{m}_1$ (g/s)	$\dot{m}_2$ (g/s)	ϕ_K	T (°C)	ρ (kg/m ³)	μ (mPa · s)	Δt_{beams} (μ s)	f_{laser} (Hz)
1-300	2.0	3.1	0.27	23.4	1.18	20.2	112	2
301-600	2.0	2.5	0.51	23.4	1.18	20.2	141	2
601-900	2.0	2.2	0.75	23.4	1.18	20.2	162	2
901-1200	2.0	2.0	1.00	24.2	1.18	19.5	178	2
1201-1500	2.0	1.8	1.37	23.4	1.18	20.2	178	2
1501-1800	2.0	1.6	1.95	23.4	1.18	20.2	178	2
1801-2100	2.0	1.2	4.63	23.4	1.18	20.2	178	2

Table 5.14. List of conditions for the experiments with the transparent mixing chamber no. 2 at $Re_1=100$.

Frames	$\dot{m}_1$ (g/s)	$\dot{m}_2$ (g/s)	ϕ_K	T (°C)	ρ (kg/m ³)	μ (mPa · s)	Δt_{beams} (μ s)	f_{laser} (Hz)
1-300	3.9	5.2	0.42	24.5	1.18	19.2	58	4
301-600	3.9	4.9	0.50	24.5	1.18	19.2	71	4
601-900	3.9	4.4	0.70	24.5	1.18	19.2	80	4
901-1200	3.9	3.9	1.00	23.8	1.18	19.8	89	4
1201-1500	3.9	3.5	1.38	23.8	1.18	19.8	89	4
1501-1800	3.9	3.1	1.99	24.5	1.18	19.2	89	4

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Table 5.15. List of conditions for the experiments with the transparent mixing chamber no. 2 at $Re_1=200$.

Frames	$\dot{m}_1$ (g/s)	$\dot{m}_2$ (g/s)	ϕ_K	T (°C)	ρ (kg/m ³)	μ (mPa · s)	Δt_{beams} (μ s)	f_{laser} (Hz)
1-300	7.9	9.9	0.51	23.6	1.18	20.0	35	8.33
301-600	7.9	9.5	0.58	23.6	1.18	20.0	34	8.33
601-900	7.9	8.8	0.72	23.6	1.18	20.0	34	8.33
901-1200	7.9	7.9	1.00	23.6	1.18	20.0	45	8.33
1201-1500	7.9	7.5	1.17	23.6	1.18	20.0	45	8.33
1501-1800	7.9	7.0	1.44	23.6	1.18	20.0	45	8.33

Table 5.16. List of conditions for the experiments with the transparent mixing chamber no. 2 at $Re_1=300$.

Frames	$\dot{m}_1$ (g/s)	$\dot{m}_2$ (g/s)	ϕ_K	T (°C)	ρ (kg/m ³)	μ (mPa · s)	Δt_{beams} (μ s)	f_{laser} (Hz)
1-300	11.8	13.2	0.71	24.4	1.18	19.3	27	11.11
301-600	11.8	12.5	0.84	24.4	1.18	19.3	28	11.11
601-900	11.8	11.8	1.00	24.4	1.18	19.3	30	11.11
901-1200	11.8	11.1	1.20	24.4	1.18	19.3	30	11.11
1201-1500	11.8	10.5	1.42	24.4	1.18	19.3	30	11.11
1501-1800	11.8	9.9	1.69	24.4	1.18	19.3	30	11.11

Table 5.17. List of conditions for the experiments with the transparent mixing chamber no. 3 at $Re_1=50$.

Frames	$\dot{m}_1$ (g/s)	$\dot{m}_2$ (g/s)	ϕ_K	T (°C)	ρ (kg/m ³)	μ (mPa · s)	Δt_{beams} (μ s)	f_{laser} (Hz)
1-300	1.6	2.3	0.13	24.1	1.18	19.6	55	2
301-600	1.6	1.7	0.32	24.1	1.18	19.6	74	2
601-900	1.6	1.3	0.71	24.1	1.18	19.6	93	2
901-1200	1.6	1.2	0.90	24.1	1.18	19.6	107	2
1201-1500	1.6	1.1	1.2	24.1	1.18	19.6	118	2
1501-1800	1.6	1.0	1.6	24.1	1.18	19.6	130	2
1801-2100	1.6	0.8	3.0	24.1	1.18	19.6	143	2
2101-2400	1.6	0.7	4.5	24.1	1.18	19.6	143	2
2401-2700	1.6	0.5	12	24.1	1.18	19.6	143	2

Table 5.18. List of conditions for the experiments with the transparent mixing chamber no. 3 at $Re_1=100$.

Frames	$\dot{m}_1$ (g/s)	$\dot{m}_2$ (g/s)	ϕ_K	T (°C)	ρ (kg/m ³)	μ (mPa · s)	Δt_{beams} (μ s)	f_{laser} (Hz)
1-300	3.1	3.4	0.3	23.1	1.18	20.5	37	4
301-600	3.1	3.0	0.4	23.1	1.18	20.5	42	4
601-900	3.1	2.7	0.6	23.1	1.18	20.5	47	4
901-1200	3.1	2.4	0.8	23.1	1.18	20.5	53	4
1201-1500	3.1	2.1	1.2	23.1	1.18	20.5	59	4
1501-1800	3.1	1.9	1.7	23.1	1.18	20.5	63	4
1801-2100	3.1	1.8	1.9	23.1	1.18	20.5	66	4
2101-2400	3.1	1.7	2.3	23.1	1.18	20.5	71	4
2401-2700	3.1	1.5	3.4	23.1	1.18	20.5	71	4

5. Impinging Jets Interaction

Table 5.19. List of conditions for the experiments with the transparent mixing chamber no. 3 at $Re_1=200$.

Frames	$\dot{m}_1$ (g/s)	$\dot{m}_2$ (g/s)	ϕ_K	T (°C)	ρ (kg/m ³)	μ (mPa · s)	Δt_{beams} (μs)	f_{laser} (Hz)
1-300	6.3	5.4	0.6	23.8	1.18	19.8	23.0	8.33
301-600	6.3	4.8	0.9	23.8	1.18	19.8	26.0	8.33
601-900	6.3	4.3	1.2	23.8	1.18	19.8	29.0	8.33
901-1200	6.3	3.8	1.7	23.8	1.18	19.8	33.0	8.33
1201-1500	6.3	3.4	2.4	23.8	1.18	19.8	36.0	8.33

Table 5.20. List of conditions for the experiments with the transparent mixing chamber no. 3 at $Re_1=300$.

Frames	$\dot{m}_1$ (g/s)	$\dot{m}_2$ (g/s)	ϕ_K	T (°C)	ρ (kg/m ³)	μ (mPa · s)	Δt_{beams} (μs)	f_{laser} (Hz)
1-300	9.4	7.7	0.7	23.6	1.18	20.0	16	11.11
301-600	9.4	7.2	0.8	23.6	1.18	20.0	18	11.11
601-900	9.4	6.8	1.0	23.6	1.18	20.0	19	11.11
901-1200	9.4	6.4	1.2	23.6	1.18	20.0	20	11.11
1201-1500	9.4	6.1	1.4	23.6	1.18	20.0	21	11.11
1501-1800	9.4	5.7	1.7	23.6	1.18	20.0	22	11.11

The determination of the critical Reynolds number, Re_c , was performed with more detail for the mixing chambers with equal injectors. This study intended to verify if the same Re_c is observable in chambers with different injectors diameters. To study the regime transition, the flow rate for both injectors was maintained equal and increased from the equivalent to $Re=100$ with increments of 0.1 g/s until the dynamic state was observed. The procedure was repeated decreasing the flow rate from the dynamic state until a steady flow was observed, with 300 pairs of images obtained for each flow rate increment. Δt_{beams} was again set so that the seeding particles could move only one fourth of a search area in the time interval as indicated in Equation (5.38). Temperature and fluid density were measured at every image acquisition. The frequency of acquisition of the images was kept constant and equal to 14.29 Hz.

5.4.3 Flow Field Characterization

Velocity vector fields were obtained from the captured images of the seeded flow for the set of experiments of Table 5.8 to Table 5.20. For each experimental condition, the 300 instantaneous velocity vector fields were divided in two sets of 150 and a time averaged velocity field was calculated from each set. To obtain information about the dynamics, the average velocity fields were coupled with a non-dimensional kinetic energy of the velocity fluctuations, $k/u_{\text{inj},1}^2$. The kinetic energy of the velocity fluctuations is defined as

$$k(\mathbf{x}) = \frac{1}{2} \overline{\left(u_x(\mathbf{x}, t) - \bar{u}_x(\mathbf{x})\right)^2} + \frac{1}{2} \overline{\left(u_y(\mathbf{x}, t) - \bar{u}_y(\mathbf{x})\right)^2} \quad (5.41)$$

where x is the horizontal coordinate, y is the vertical coordinate in the 2D velocity map and u_x and u_y are the velocity components in the x -direction and y -direction, respectively. Figure 5.18 to Figure 5.30 show one of two obtained average velocity vector fields for each experimental condition overlapped with kinetic energy of the velocity fluctuations.

All vector maps show that the two jets enter the mixing chamber and impinge distributing radially, normally to the 2D plane. The jets' impingement point is shown to depend on both the operation conditions and the mixing chamber's geometrical parameters. For all chambers, it is observed that the jets get narrower as the flow rate is increased.

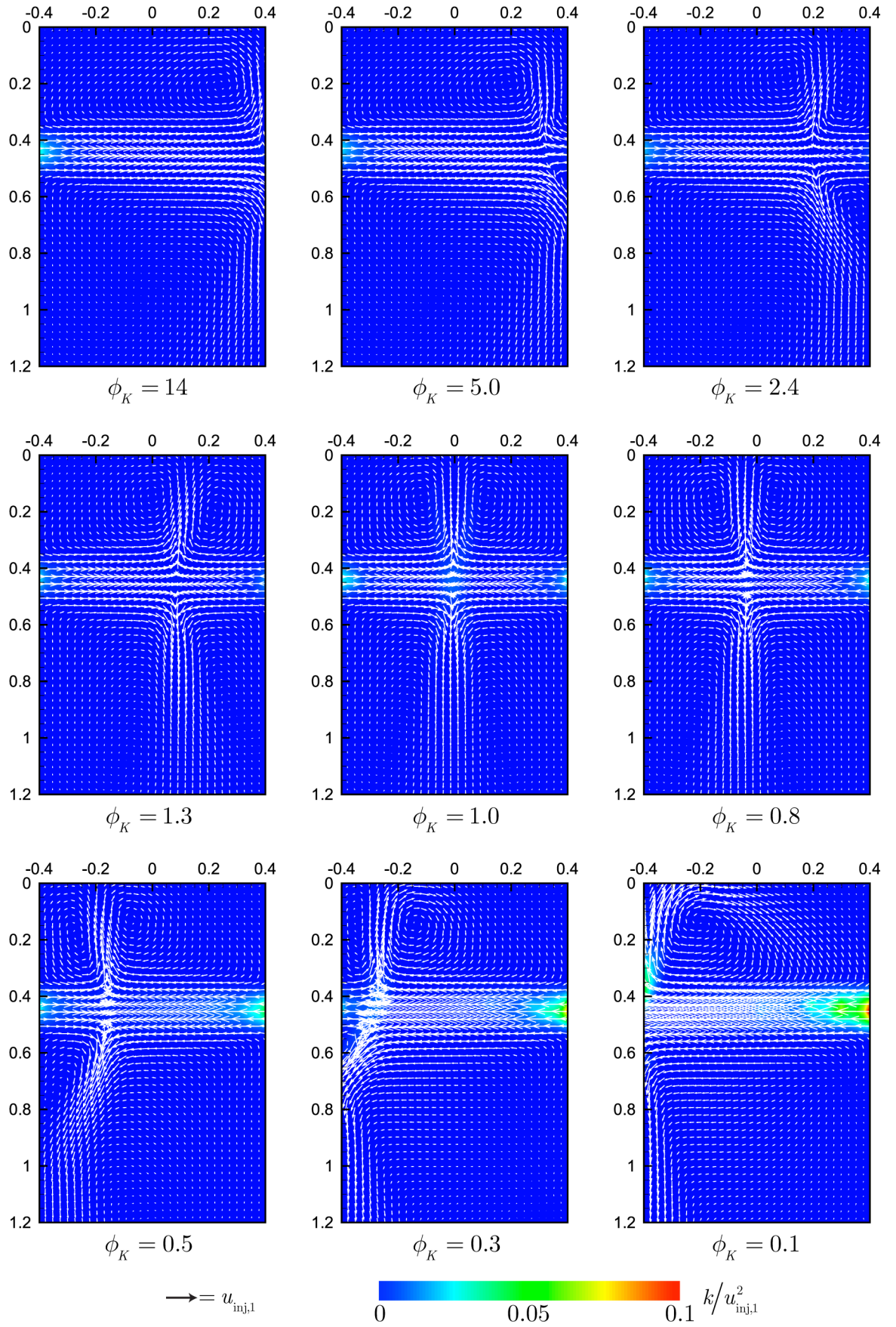


Figure 5.18. Velocity field vector maps and non-dimensional velocity fluctuations contours for Chamber 1 as a function of the kinetic energy rate ratio at $Re=50$. Spatial dimensions relative to the chamber diameter.

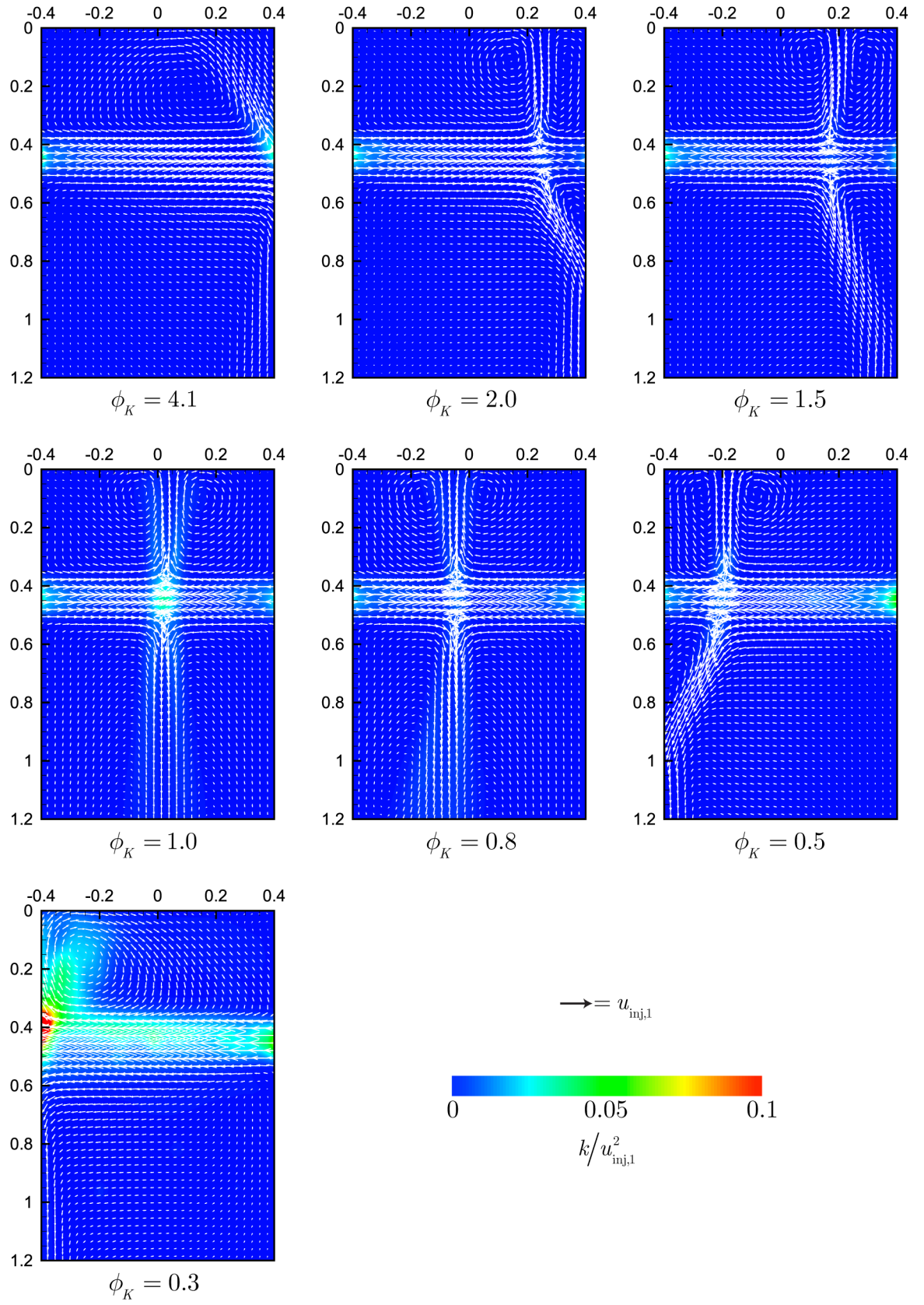


Figure 5.19. Velocity field vector maps and non-dimensional velocity fluctuations contours for Chamber1 as a function of the kinetic energy rate ratio at $Re=100$. Spatial dimensions relative to the chamber diameter.

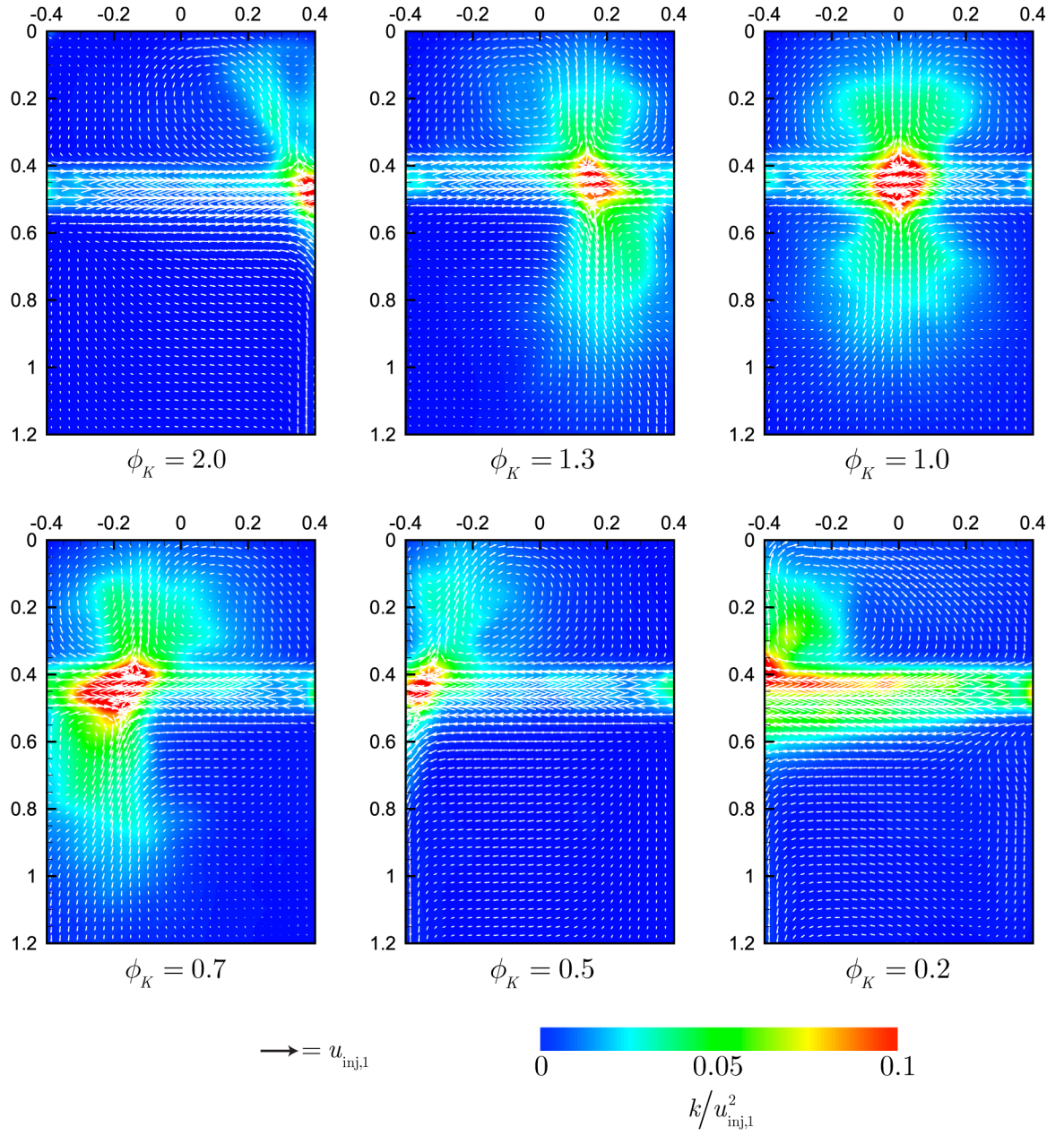


Figure 5.20. Velocity field vector maps and non-dimensional velocity fluctuations contours for Chamber 1 as a function of the kinetic energy rate ratio at $Re=200$. Spatial dimensions relative to the chamber diameter.

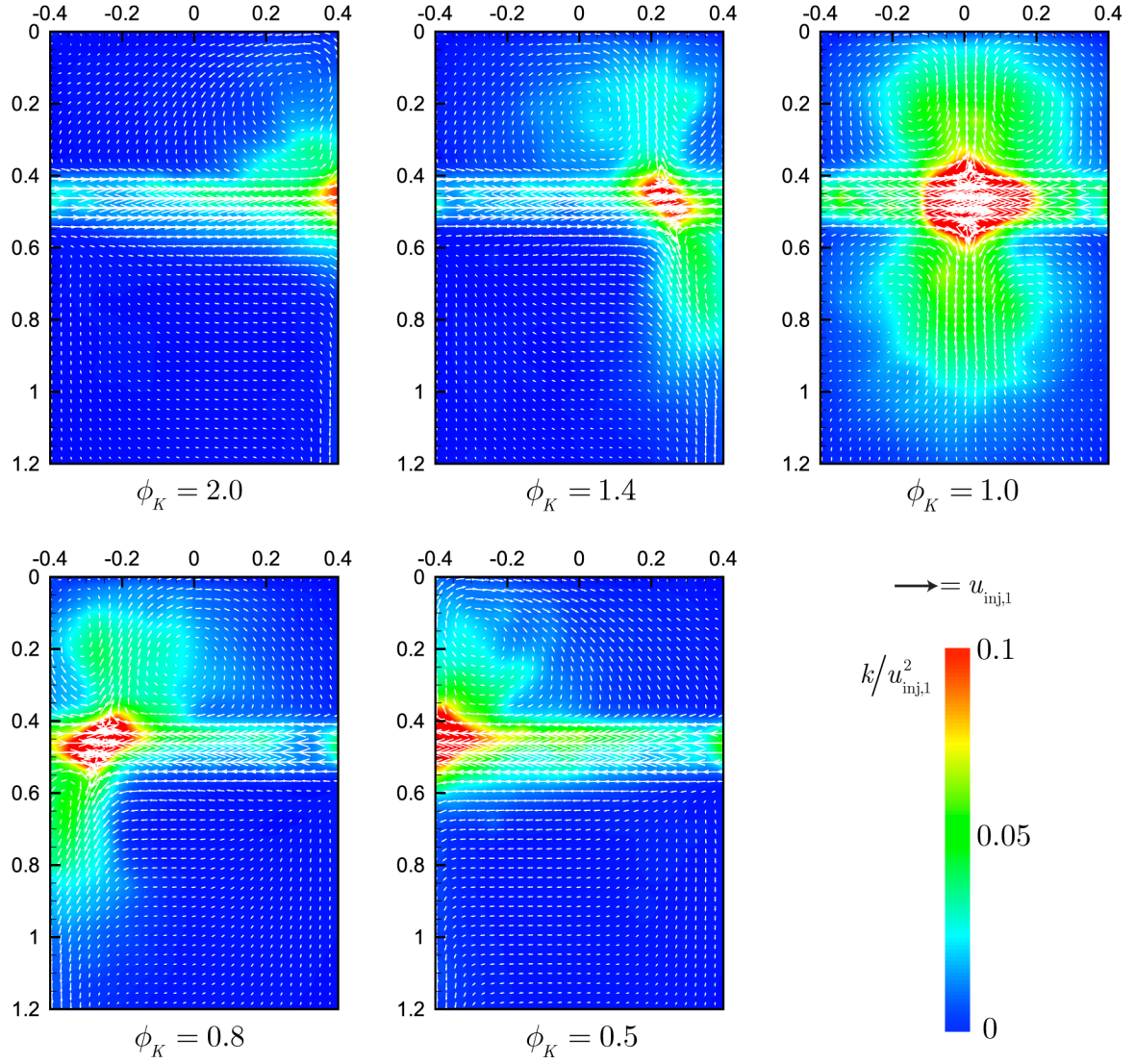


Figure 5.21. Velocity field vector maps and non-dimensional velocity fluctuations contours for Chamber1 as a function of the kinetic energy rate ratio at $Re=300$. Spatial dimensions relative to the chamber diameter.

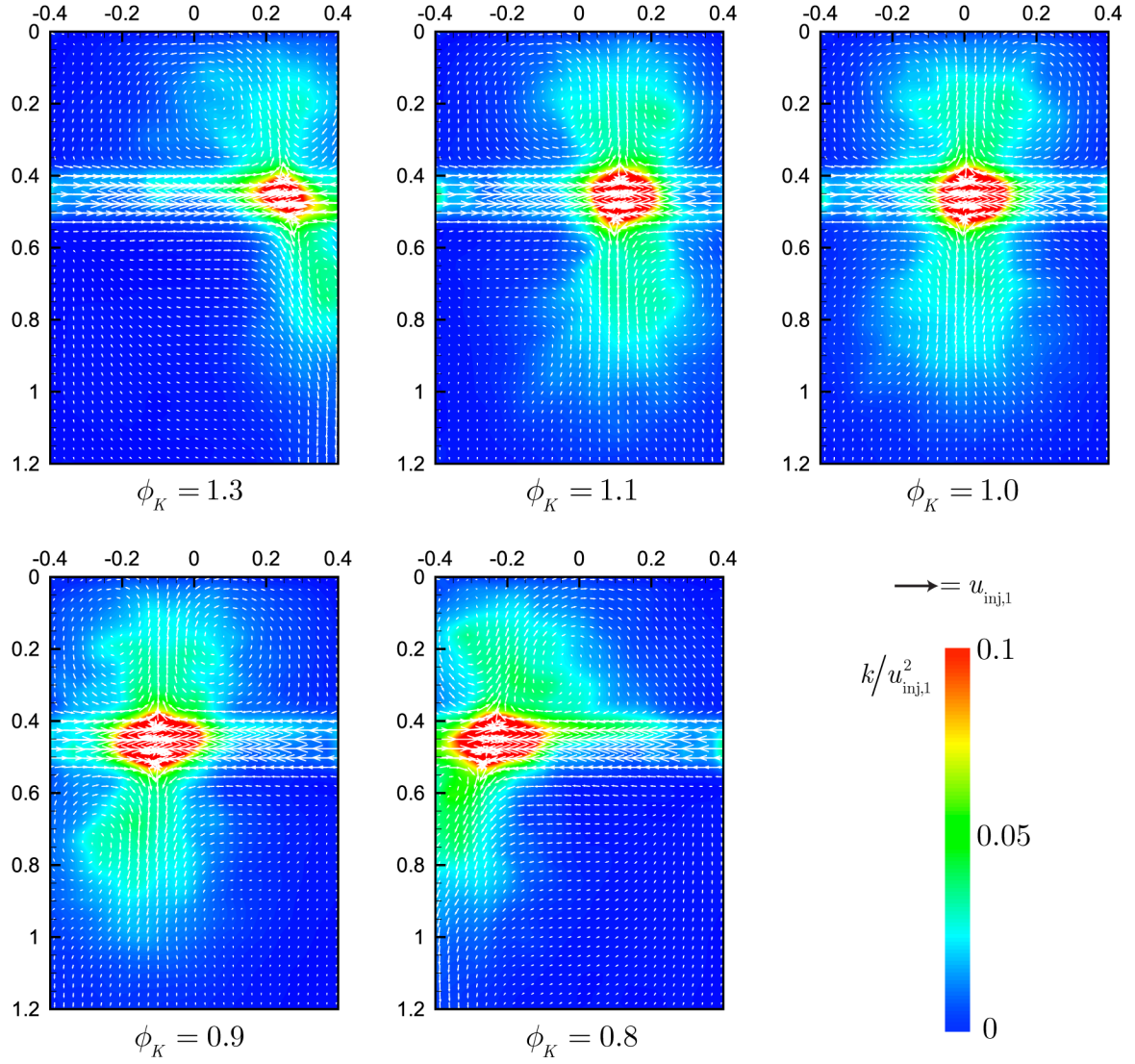


Figure 5.22. Velocity field vector maps and non-dimensional velocity fluctuations contours for Chamber 1 as a function of the kinetic energy rate ratio at $Re=400$. Spatial dimensions relative to the chamber diameter.

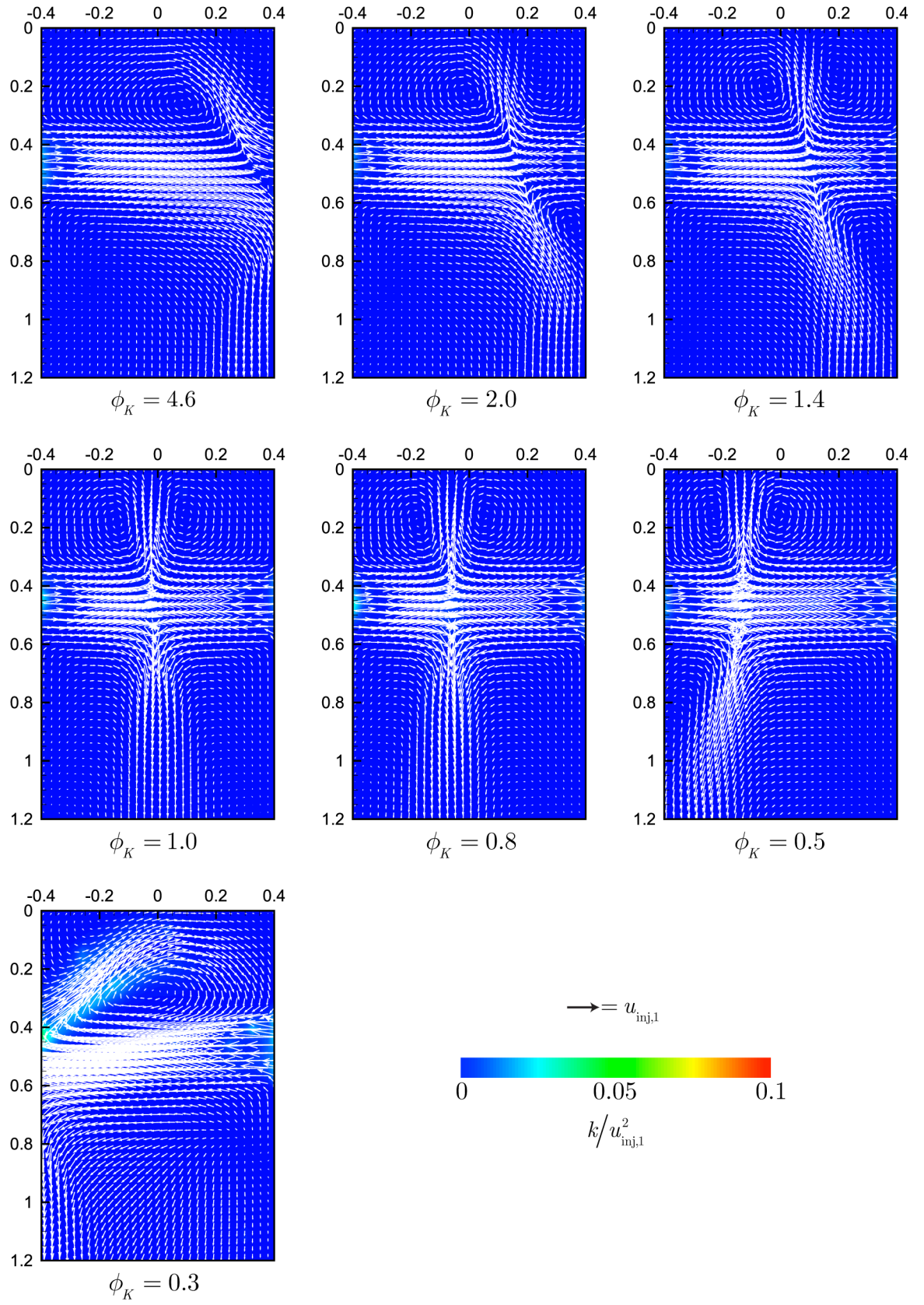


Figure 5.23. Velocity field vector maps and non-dimensional velocity fluctuations contours for Chamber 2 as a function of the kinetic energy rate ratio at $Re=50$. Spatial dimensions relative to the chamber diameter.

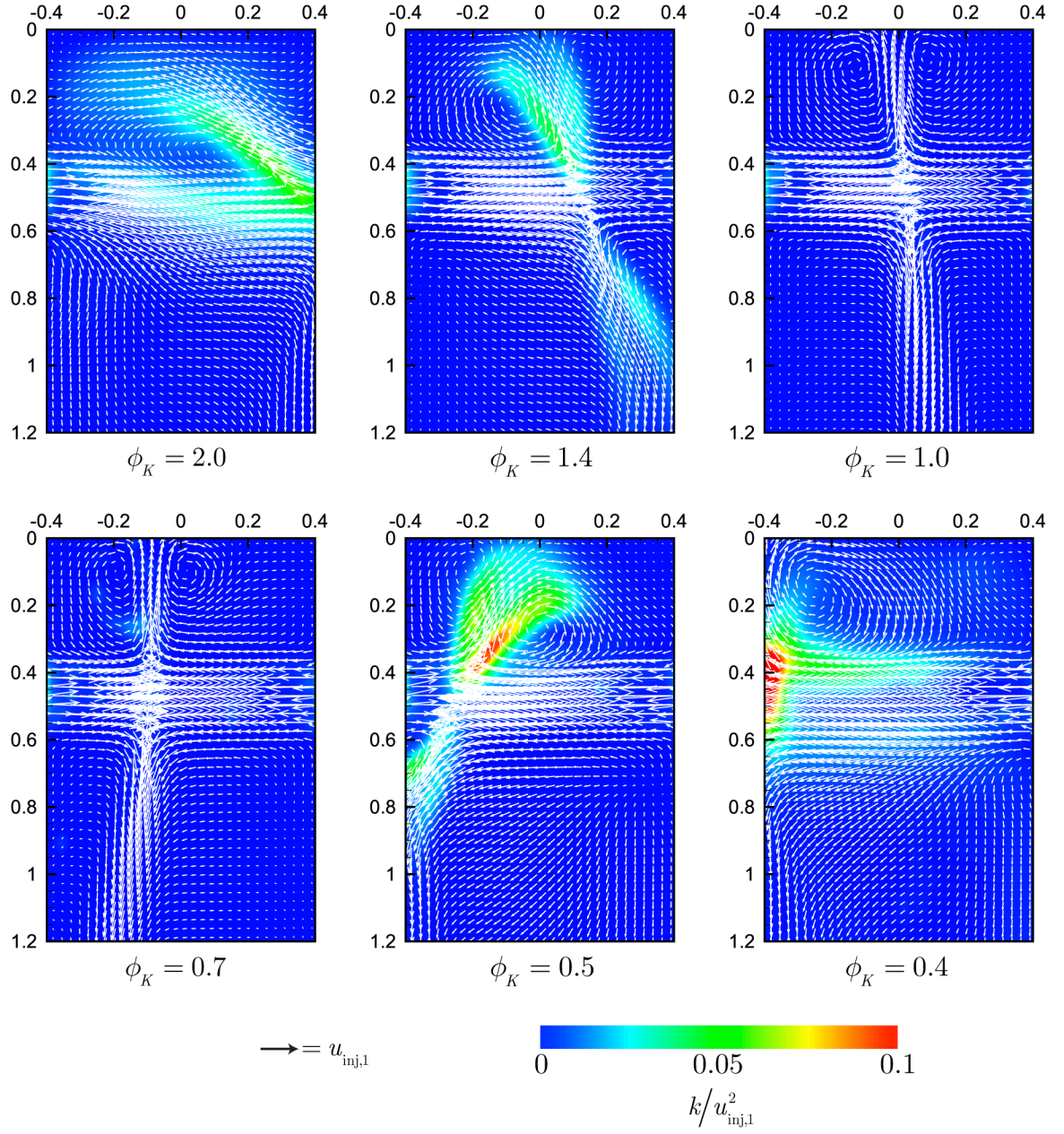


Figure 5.24. Velocity field vector maps and non-dimensional velocity fluctuations contours for Chamber 2 as a function of the kinetic energy rate ratio at $Re=100$. Spatial dimensions relative to the chamber diameter.

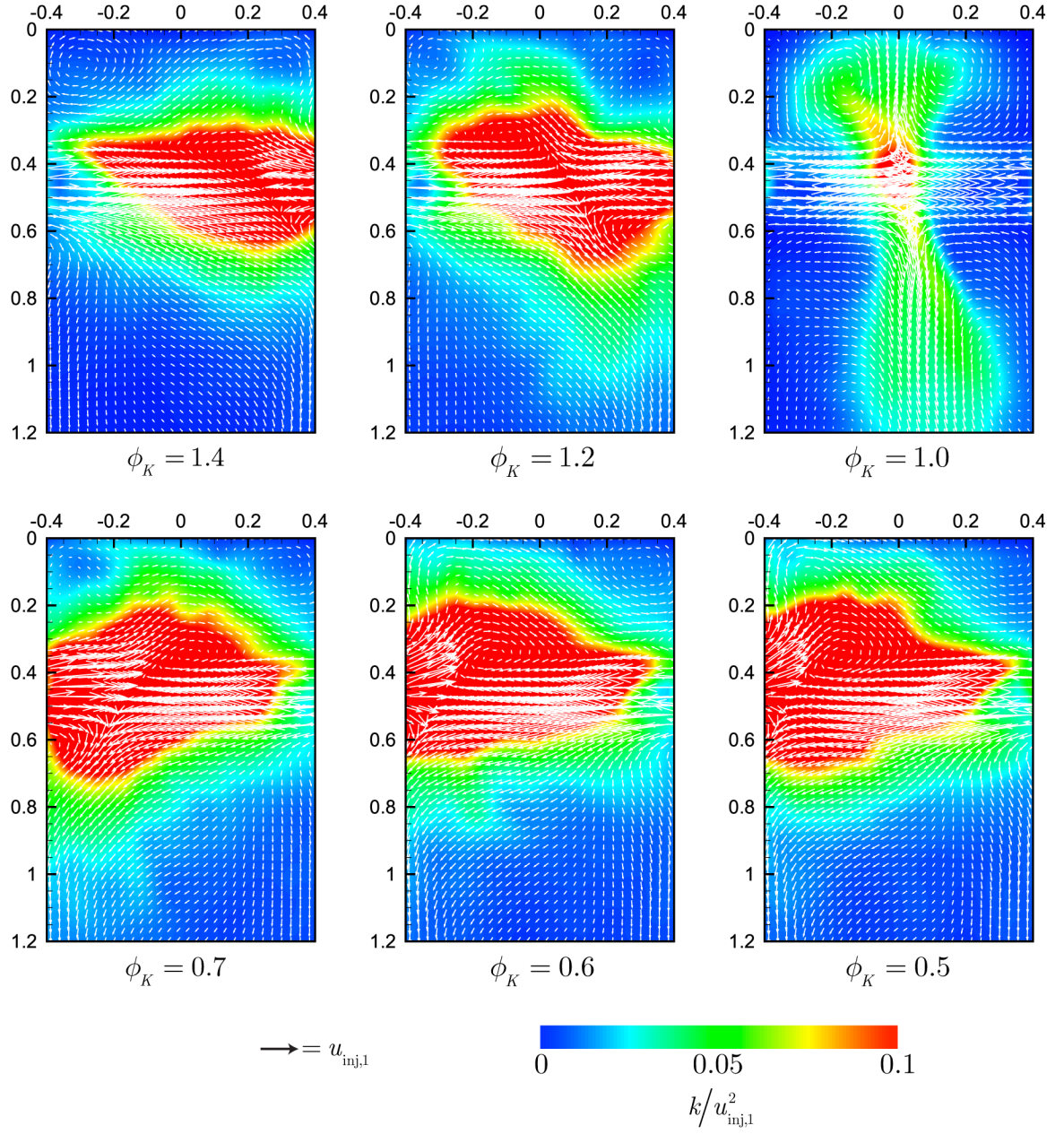


Figure 5.25. Velocity field vector maps and non-dimensional velocity fluctuations contours for Chamber 2 as a function of the kinetic energy rate ratio at $Re=200$. Spatial dimensions relative to the chamber diameter.

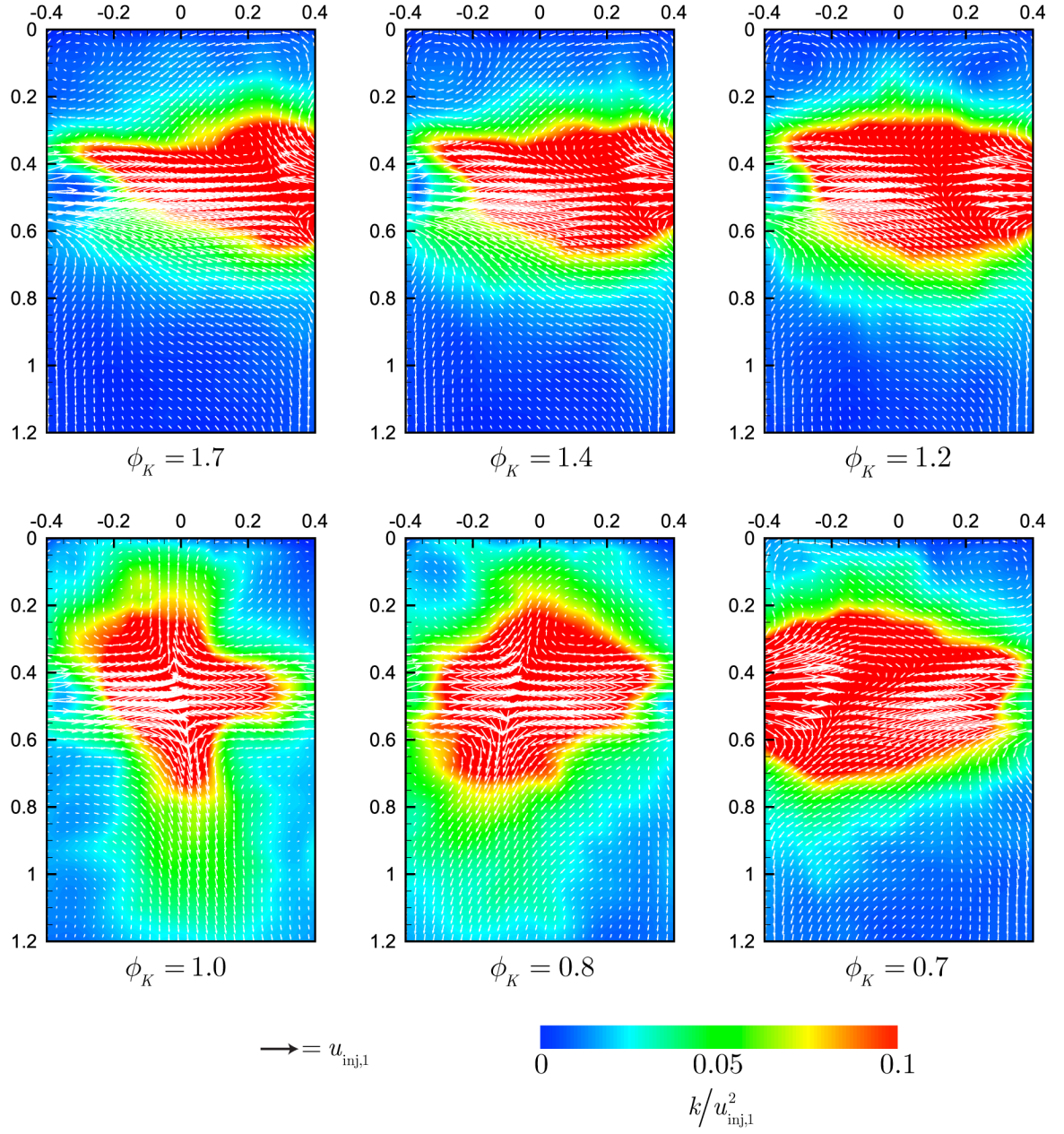


Figure 5.26. Velocity field vector maps and non-dimensional velocity fluctuations contours for Chamber 2 as a function of the kinetic energy rate ratio at $Re=300$. Spatial dimensions relative to the chamber diameter.

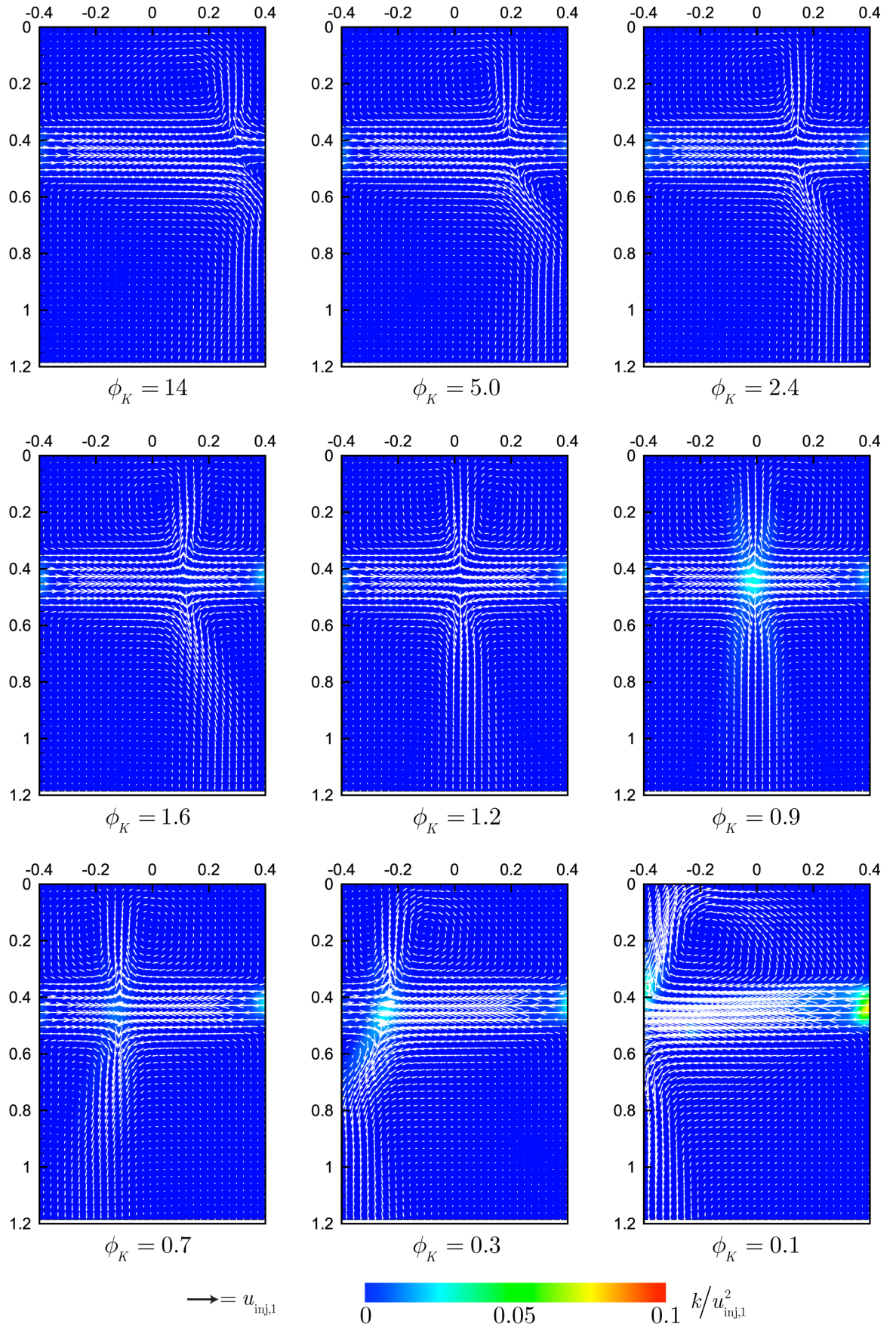


Figure 5.27. Velocity field vector maps and non-dimensional velocity fluctuations contours for Chamber 3 as a function of the kinetic energy rate ratio at $Re=50$. Spatial dimensions relative to the chamber diameter.

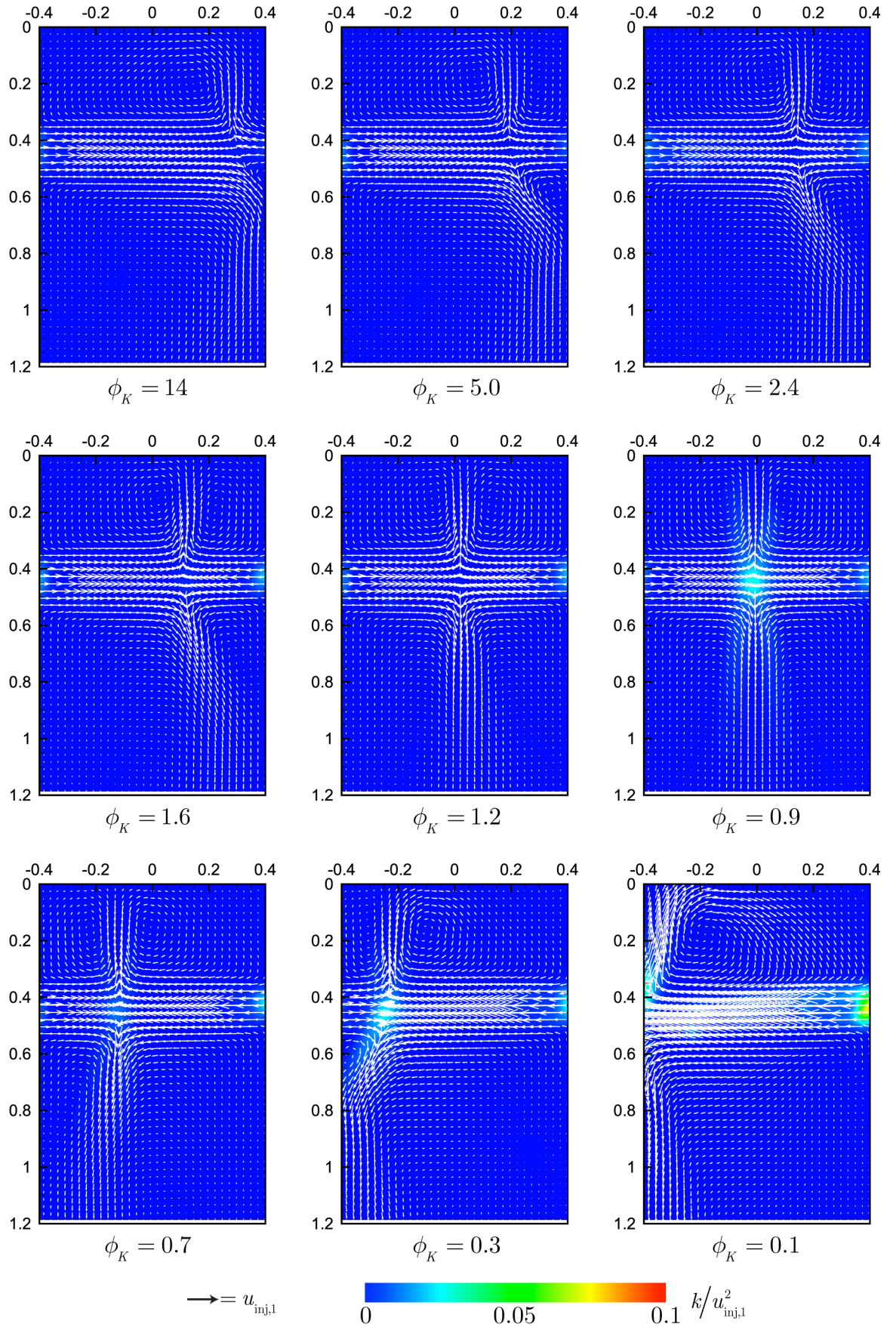


Figure 5.28. Velocity field vector maps and non-dimensional velocity fluctuations contours for Chamber 3 as a function of the kinetic energy rate ratio at $Re=100$. Spatial dimensions relative to the chamber diameter.

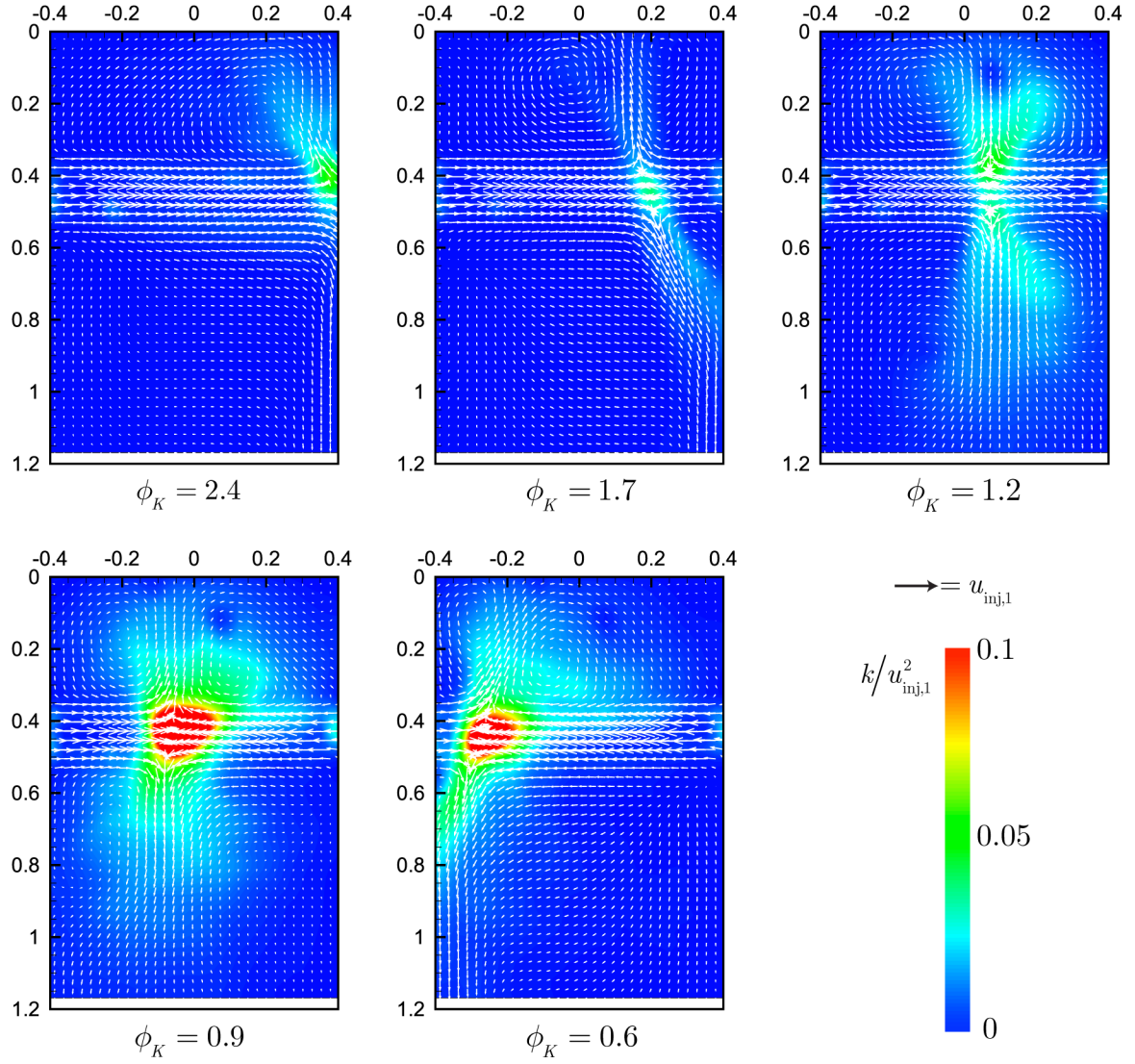


Figure 5.29. Velocity field vector maps and non-dimensional velocity fluctuations contours for Chamber 3 as a function of the kinetic energy rate ratio at $Re=200$. Spatial dimensions relative to the chamber diameter.

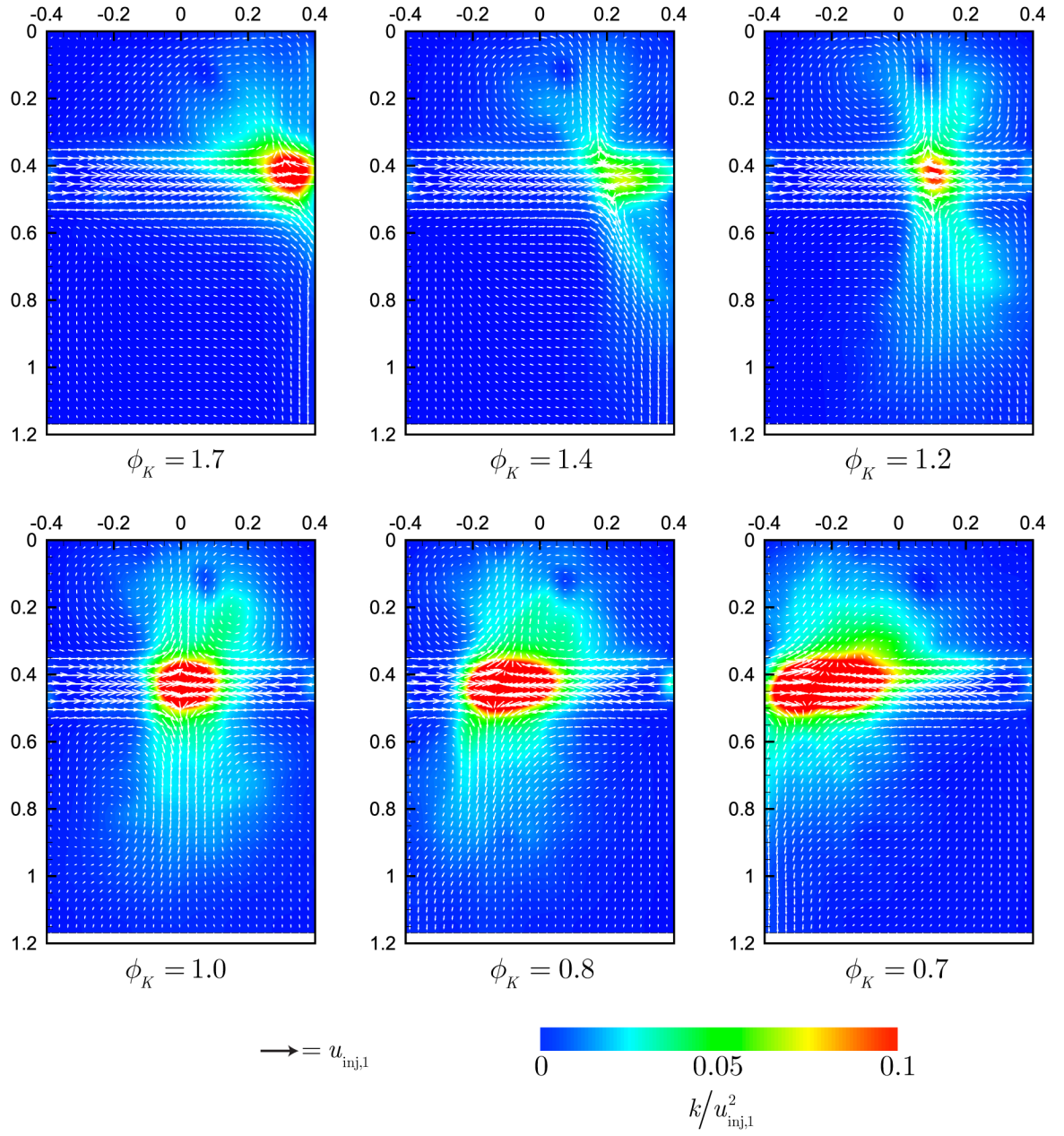


Figure 5.30. Velocity field vector maps and non-dimensional velocity fluctuations contours for Chamber 3 as a function of the kinetic energy rate ratio at $Re=300$. Spatial dimensions relative to the chamber diameter.

For Chamber no. 1 (see Table 5.7), as expected, the jets impinge in the center when the flow rates are balanced and $\phi_K = 1$. Unsteady flow regimes were observed for $Re > 100$ although dynamic behaviors for lower Re , were observed when one of the jets have enough flow rate to impinge directly in the chamber wall. The flow fluctuations are damped as the impingement point of the two jets moves away from the center, with this effect being less observed as Re increases. An asymmetric behavior for the same Re_1 is observed in the impingement point displacement and in the oscillations damping for $\phi_K > 1$ and $\phi_K < 1$.

For Chamber no. 2 (see Table 5.7), a dynamic behavior of the flow was also detected for $Re > 100$, showing again a dynamic behavior for lower Re when one of the jets impinges directly in the chamber wall. In the dynamic regime, comparing with Chamber no. 1 and 3, less discernable flow structures were observed, with the flow entering and leaving the laser plane. This can be caused by small oscillations of the jets in the normal direction to the 2D plane of the figures. Due to this phenomenon, velocity fluctuation maps in Chamber no. 2 will not be discussed. The average velocity fields were used, although, for the determination of the average jets' impingement point. As expected, the jets impinge, on average, in the center of the chamber when $\phi_K = 1$.

For Chamber no. 3 (see Table 5.7), as already predicted from the elastic analogue model and CFD simulations, the jets do not impinge in the center when the jets have equal flow rates or momenta, but when its kinetic energy rate ratio is equal to 1. A dynamic behavior of the flow was also detected for $Re > 100$. Similar to the observed in the other two chambers, the flow showed a dynamic behavior for lower Re when one of the jets impinges directly in the chamber wall. As in Chamber no. 1, the flow fluctuations seem to be damped as the impingement point of the two jets moves away from the center, with this effect being less observed as Re is increased. An asymmetric behavior for the same Re_1 is observed in the impingement point displacement and in the oscillations damping for $\phi_K > 1$ and $\phi_K < 1$.

5.4.4 Impingement Point Position

From the time averaged velocity vector fields an average position of the jets' impingement point was determined. The x -velocity profile was extracted from the PIV results in a vertical line coincident with the jets' axis. As in the CFD analysis of Section 5.3, the impingement point position was determined as the x -position in the inspected profile where the x -velocity is equal to zero. This procedure was done by importing and processing the vector field data with the software MATLAB® R2011b for Mac OS X. Figure 5.31 to Figure 5.48 show the obtained results for the three transparent mixing chambers as a function of ϕ_K , ϕ_M and ϕ_{FR} .

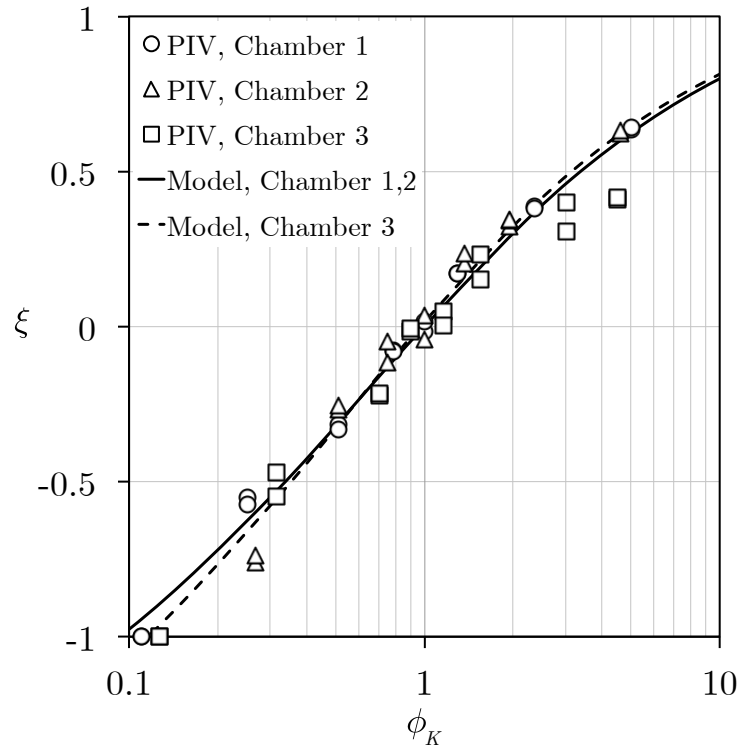


Figure 5.31. Non-dimensional impingement point position from the PIV experiments and elastic analogue model as a function of the jets' kinetic energy rate ratio at $Re_1=50$.

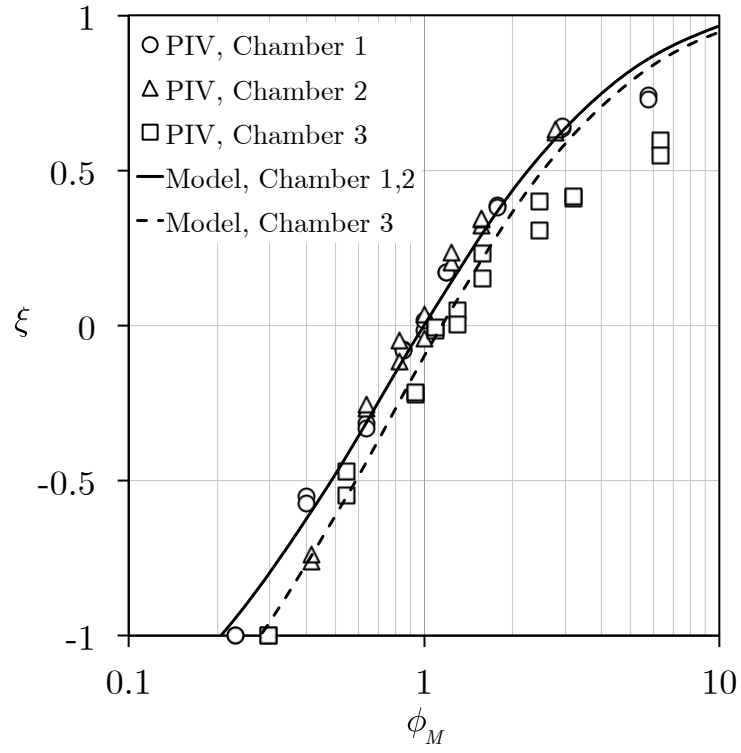


Figure 5.32. Non-dimensional impingement point position from the PIV experiments and elastic analogue model as a function of the jets' momentum rate ratio at $Re_1=50$.

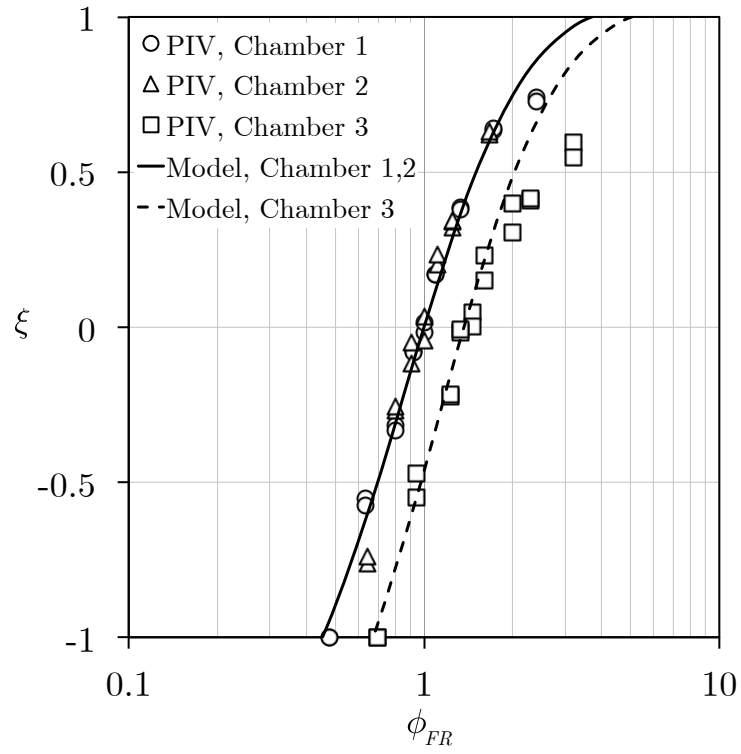


Figure 5.33. Non-dimensional impingement point position from the PIV experiments and elastic analogue model as a function of the jets' flow rate ratio at $Re_1=50$.

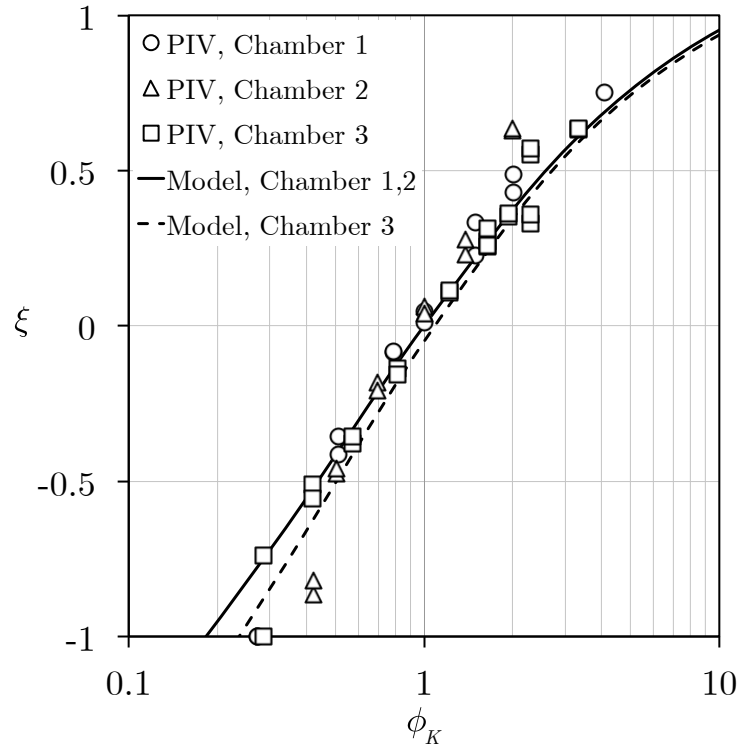


Figure 5.34. Non-dimensional impingement point position from the PIV experiments and elastic analogue model as a function of the jets' kinetic energy rate ratio at $Re_1=100$.

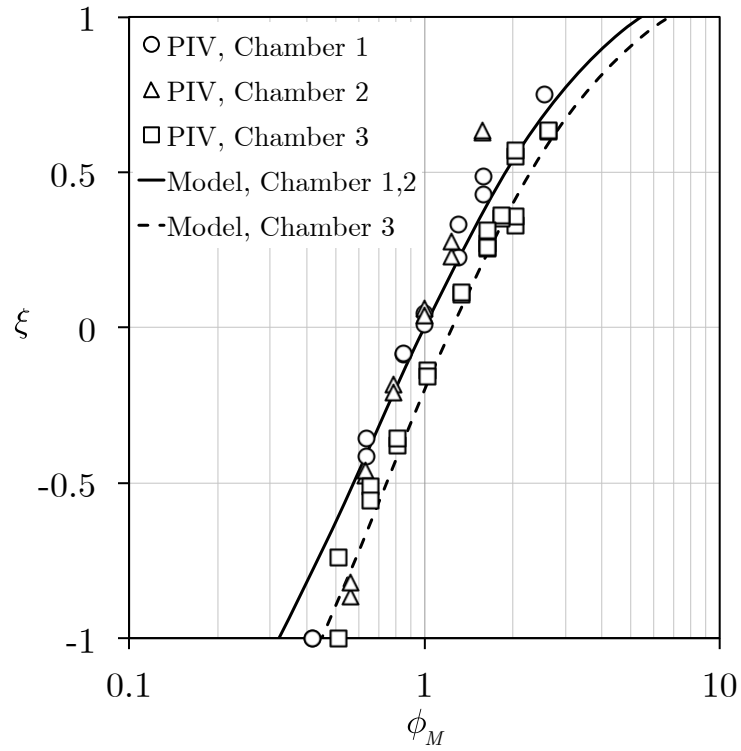


Figure 5.35. Non-dimensional impingement point position from the PIV experiments and elastic analogue model as a function of the jets' momentum rate ratio at $Re_1=100$.

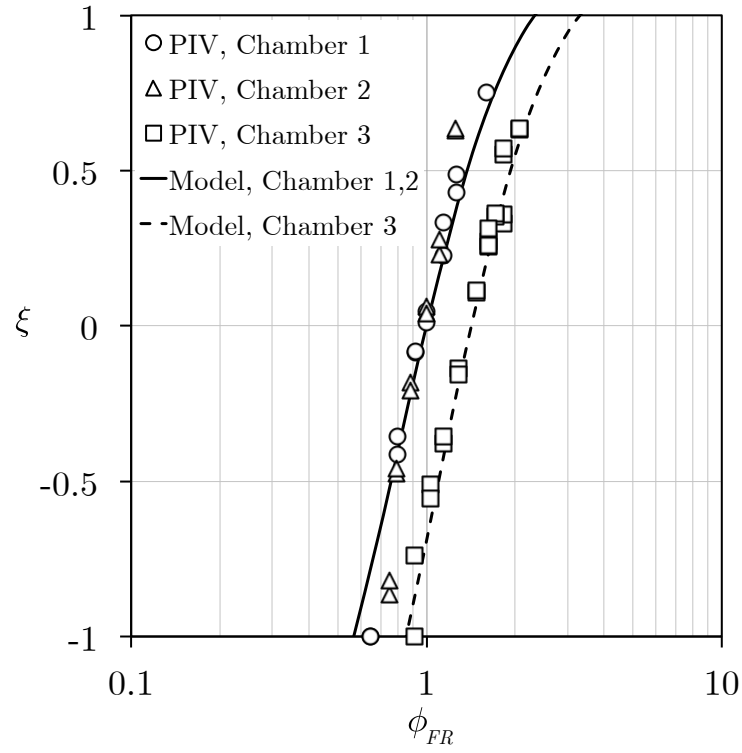


Figure 5.36. Non-dimensional impingement point position from the PIV experiments and elastic analogue model as a function of the jets' flow rate ratio at $Re_1=100$.

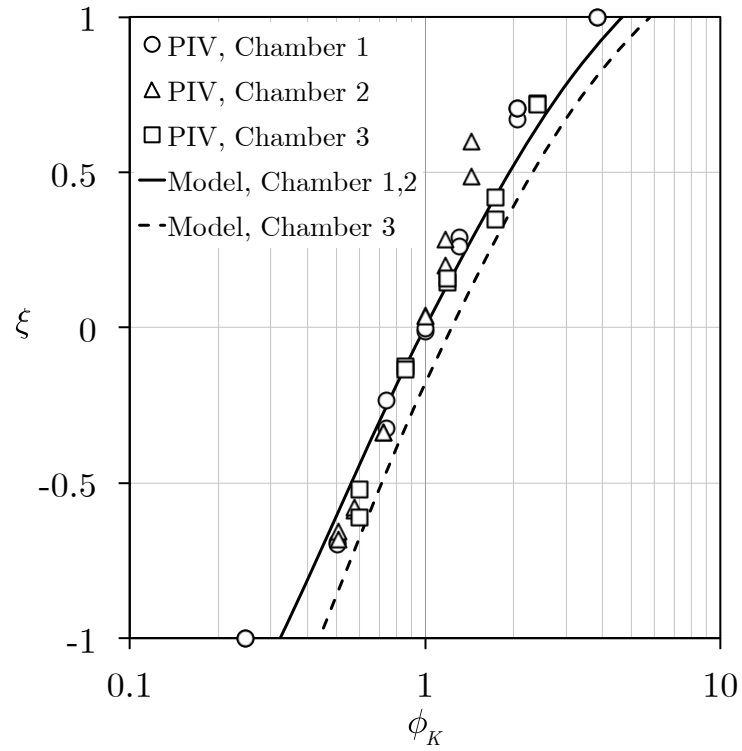


Figure 5.37. Non-dimensional impingement point position from the PIV experiments and elastic analogue model as a function of the jets' kinetic energy rate ratio at $Re_1=200$.

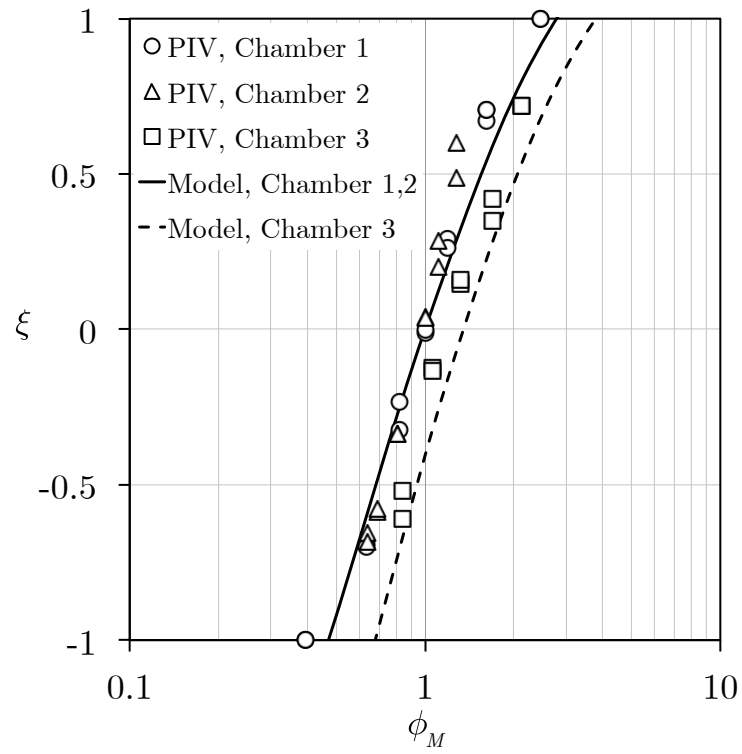


Figure 5.38. Non-dimensional impingement point position from the PIV experiments and elastic analogue model as a function of the jets' momentum rate ratio at $Re_1=200$.

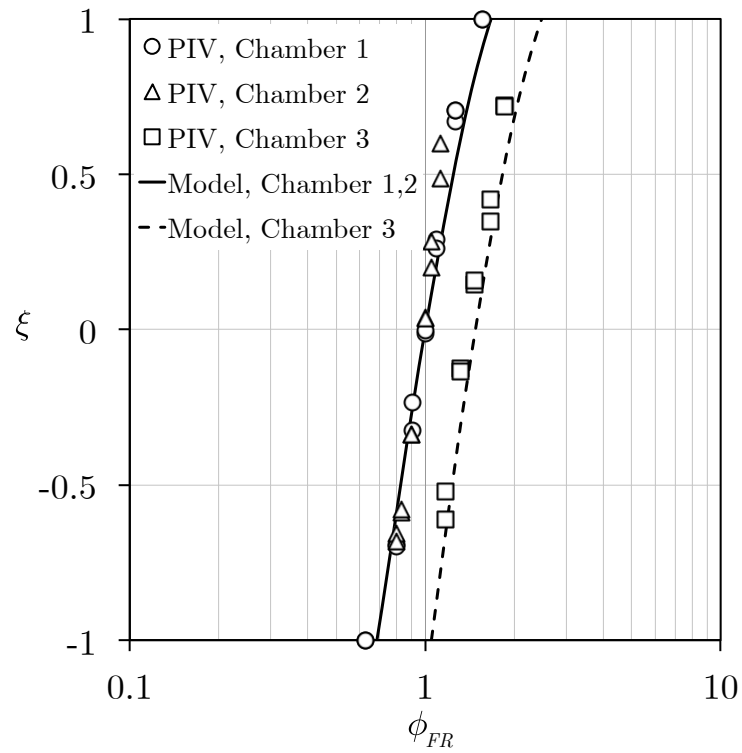


Figure 5.39. Non-dimensional impingement point position from the PIV experiments and elastic analogue model as a function of the jets' flow rate ratio at $Re_1=200$.

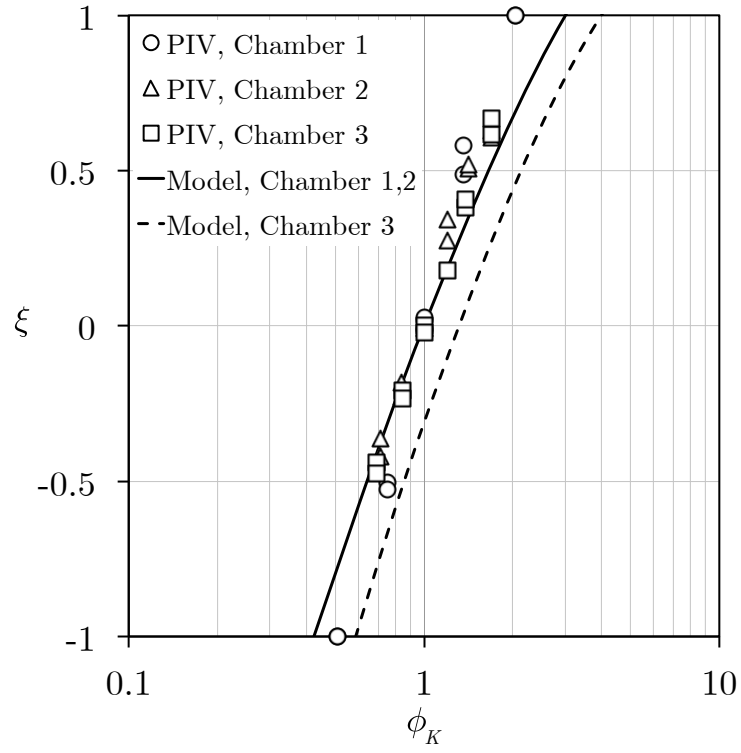


Figure 5.40. Non-dimensional impingement point position from the PIV experiments and elastic analogue model as a function of the jets' kinetic energy rate ratio at $Re_1=300$.

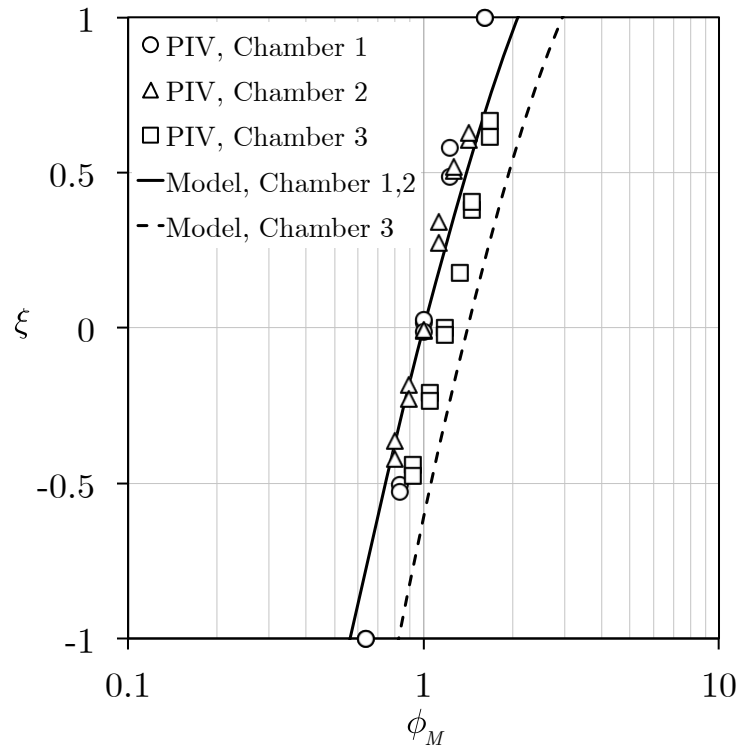


Figure 5.41. Non-dimensional impingement point position from the PIV experiments and elastic analogue model as a function of the jets' momentum rate ratio at $Re_1=300$.

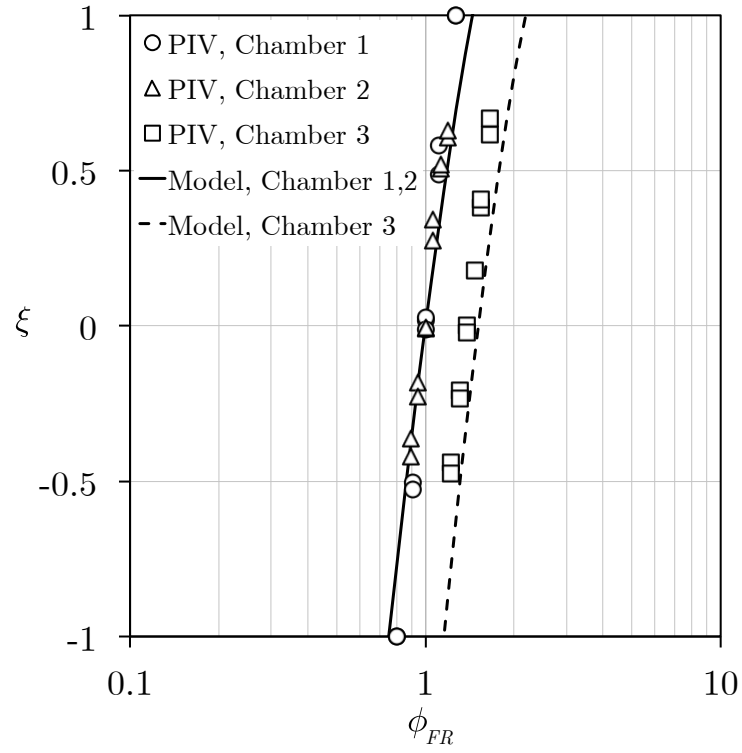


Figure 5.42. Non-dimensional impingement point position from the PIV experiments and elastic analogue model as a function of the jets' flow rate ratio at $Re_1=300$.

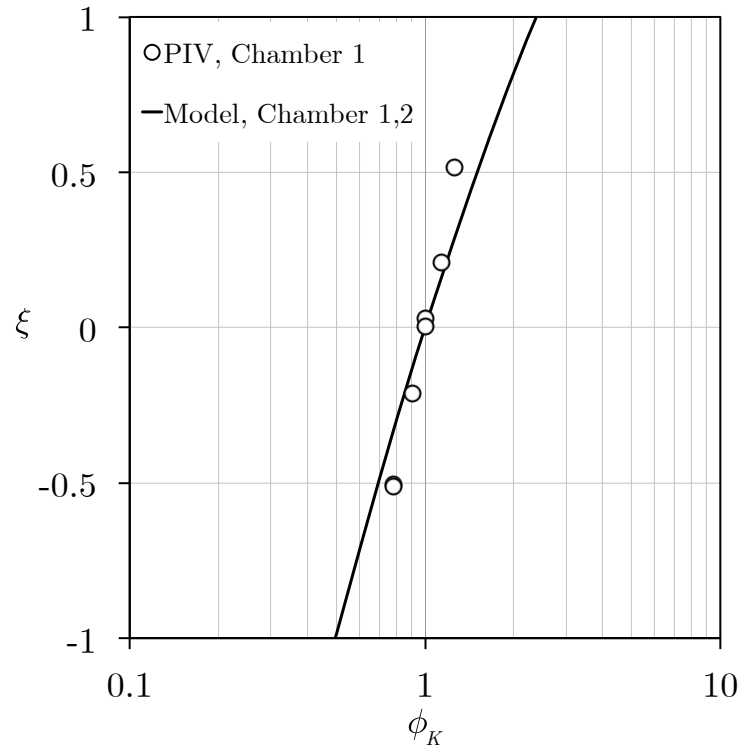


Figure 5.43. Non-dimensional impingement point position from the PIV experiments and elastic analogue model as a function of the jets' kinetic energy rate ratio at $Re_1=400$.

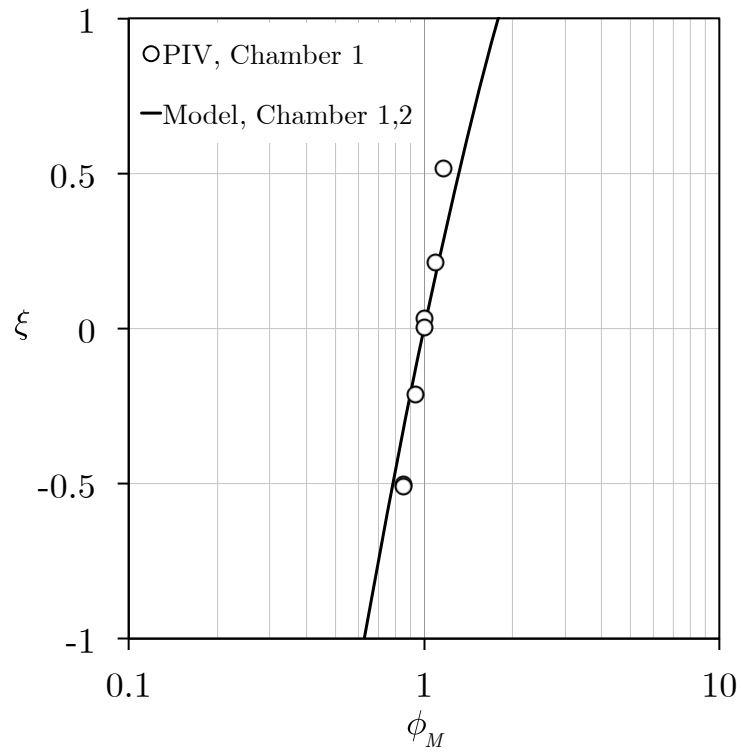


Figure 5.44. Non-dimensional impingement point position from the PIV experiments and elastic analogue model as a function of the jets' momentum rate ratio at $Re_1=400$.

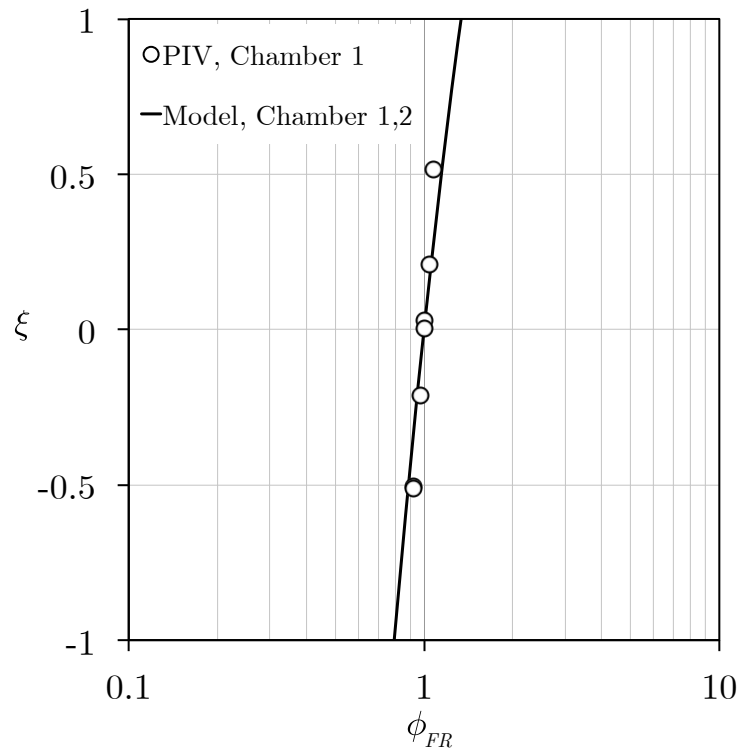


Figure 5.45. Non-dimensional impingement point position from the PIV experiments and elastic analogue model as a function of the jets' flow rate ratio at $Re_1=400$.

As observed from the CFD results in Section 5.3.5 and predicted by the elastic analogue model, increasing or decreasing one of the jets' flow rate has the effect of displacing the impingement point position from the center of the mixing chamber towards the walls. As in the CFD results of Section 5.3.5, the representation of ξ as a function of ϕ_K tends to unify the jets' behavior and overlap the curves for all the chamber designs. Representing ξ as a function of ϕ_M or ϕ_{FR} tends to separate the curves, showing clearly different behaviors for each chamber with different injector diameter ratio.

Figure 5.31 to Figure 5.45, show, as in the computational results, an asymmetric behavior of the impinging jets' interaction when the flow rates are increased or decreased.

Again from the experiments, it is observed that the impingement jets flow becomes more sensitive to small deviations in the flow rates as Re is increased. In the case of RIM, the increase of Re demands a more precise and strict monomers or pre-polymers flow rate control and becomes more sensitive to temperature of the polymers production plant.

Differently from the predicted by the elastic analogue model, the jets' impingement in the center of the mixing chamber is always obtained when both have the same kinetic energy ratio, independently of the jets' diameter ratio. This difference is most observed for higher Re and can be caused by some flow dynamics or tridimensional effects not included in the model. Nevertheless, results show good agreement between the PIV experimental and the elastic analogue model. As expected, the model shows some divergence from the CFD results as the impingement point position gets closer to the walls. The model is also successful in predicting the increasing system sensitivity to flow rate variations at higher Re .

5.4.5 Energy Dissipation

The question arises on whether it is in fact the jets' impingement in the center of the mixing chamber the correct condition to achieve better mixing efficiencies. To answer this question, a time average of the energy dissipation in the 2D PIV velocity field was calculated. The energy dissipated into heat due to changes of fluid shape in the 2D plane, neglecting the gradients in the normal direction, is given by

$$\bar{\varepsilon}_{2D} = \overline{\int_A 2\mu(\mathbf{D} : \mathbf{D}) dA}, \quad (5.42)$$

where $\mathbf{D} = \frac{1}{2} \left(\nabla \mathbf{u} + (\nabla \mathbf{u})^T \right)$ is the stretching tensor. The velocity gradients for the evaluation of $\mathbf{D}$ were determined numerically from the PIV velocity field in MATLAB® R2011b for Mac OS X using central differencing. The energy dissipation field is directly related to the reduction of mixing length scales in the mixing chamber (Siddiqui et al., 2009).

A relative energy dissipation, $\hat{\varepsilon}$, to the amount of kinetic energy fed to the system, $\dot{E}_{K,\text{in}}$, can be defined as a measure of the efficiency of mixing, and calculated from

$$\hat{\varepsilon} = \frac{\bar{\varepsilon}_{2D}}{\dot{E}_{K,\text{in}}}. \quad (5.43)$$

Assuming a fully developed parabolic velocity profile at the inlets,

$$\dot{E}_{K,\text{in}} = \sum_{i=1}^2 \frac{1}{2} \rho_i \int_0^{d_i/2} \left(2u_{\text{inj},i} \frac{(d_i/2)^2 - r^2}{(d_i/2)^2} \right)^3 2\pi r dr, \quad i = \{1, 2\} \quad (5.44)$$

or, solving the integral and assuming equal fluid densities,

$$\dot{E}_{K,\text{in}} = \frac{\pi}{4} \rho \left(d_1^2 u_{\text{inj},1}^3 + d_2^2 u_{\text{inj},2}^3 \right). \quad (5.45)$$

The relative energy dissipation was calculated for the chamber with unequal injector diameters to study separately the behavior of the system at $\phi_M = 1$ and $\phi_K = 1$. Figure 5.46 shows the value of $\hat{\varepsilon}$ as a function of the jets' impingement point position. The impingement point position for equal jet momenta and equal kinetic energy rate is also displayed in Figure 5.46. The lines in this plot were obtained from fitting a quadratic curve to the experimental points with the intent to show its trend.

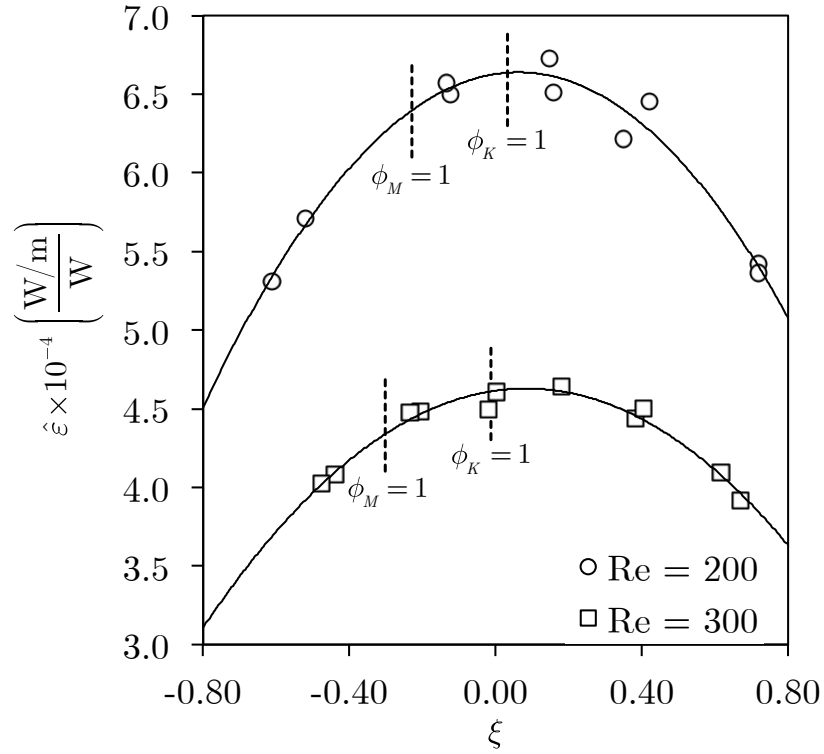


Figure 5.46. Relative energy dissipation for the Mixing Chamber no. 3.

For the unequal injectors mixing chamber, results indicate that a maximum efficiency, i.e., maximum dissipation per energy fed, is observed when the jets' impingement point is located at the center of the chamber. Asymmetric flow fields results in a less efficient use of the fed energy in flow shearing. This result reinforces the assumption that it is in fact the equal jet kinetic energy rate condition, at which the jets impinge in center, rather than the iso-momentum condition, that must be used in the operation of the RIM machine with formulations with volumetric stoichiometric or density ratios different from 1.

5.4.6 Critical Reynolds Number

The determination of the critical Reynolds number, when transition from the steady state flow regime to the self-sustained chaotic laminar state occurs, was performed with smaller flow rate increments for the mixing chambers with equal injector. Flow oscillations were measured from the spatial average of the kinetic energy of the velocity fluctuations,

$$\langle k \rangle = \left\langle \frac{1}{2} \sum_{i=1}^2 \overline{\left(u_i(\mathbf{x}, t) - \bar{u}_i(\mathbf{x}) \right)^2} \right\rangle. \quad (5.46)$$

Figure 5.47 and Figure 5.48 show the space average of non-dimensional kinetic energy of the velocity fluctuations as a function of Re for Chamber no. 1 and Chamber no. 2, respectively. The dashed lines represent the points' trend.

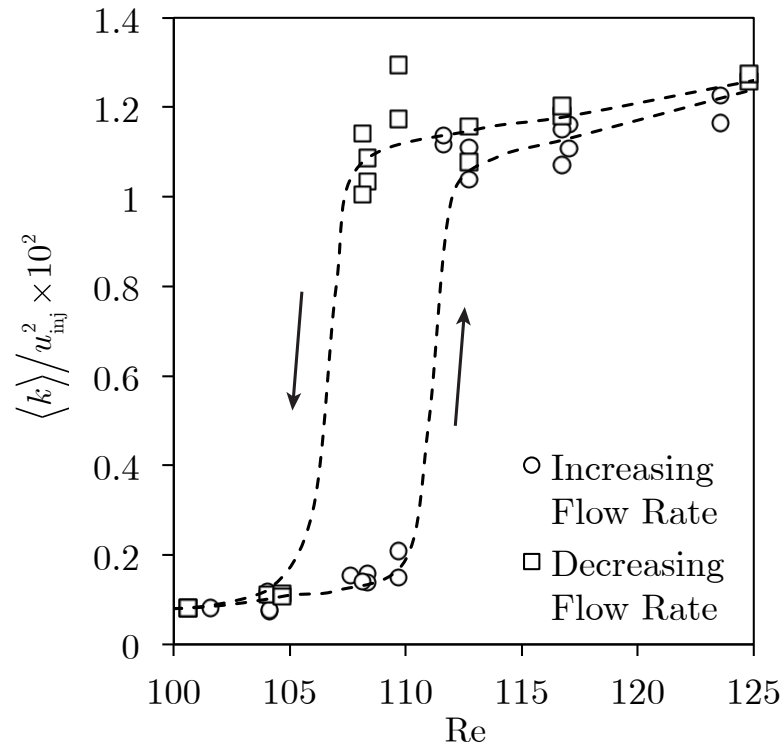


Figure 5.47. Space average of non-dimensional kinetic energy of the velocity fluctuations as a function of Re for Chamber no. 1.

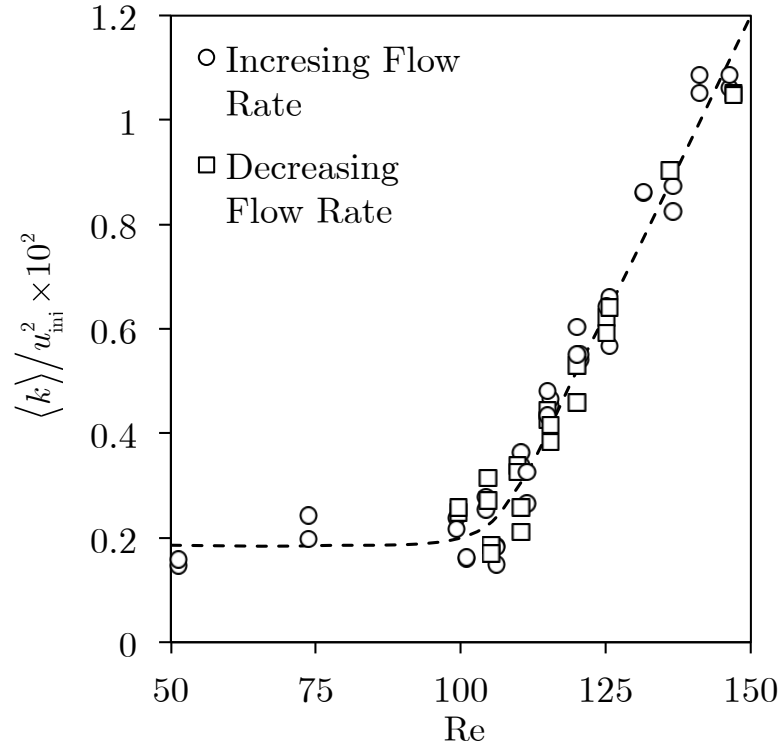


Figure 5.48. Space average of non-dimensional kinetic energy of the velocity fluctuations as a function of Re for Chamber no. 2.

For $Re > 110$ an increase of the flow oscillations from an initial residual value was observed. Independently of the chamber diameter, the flow regime transition from a steady state to a dynamic state was observed to take place at $Re \sim 110$. In Chamber no. 1 a hysteretic behavior was observed when increasing and decreasing flow rates.

5.5 Conclusions

In this chapter the impinging jets' interaction in a RIM machine mixing chamber was studied theoretically, experimentally and numerically. The present study has the goal to offer deeper insight on the system behavior under unbalanced conditions in order to optimize polymer production from monomer formulations with different physical properties or stoichiometric ratios different from 1.

Unbalanced jet conditions were found to affect the flow significantly by moving the impingement point towards the chamber walls. Under some unbalanced conditions, the jets oscillations are partially or completely damped, even when a dynamic flow regime was expected. PIV experiments show the flow to become unsteady for $Re \geq 110$, under balanced jet conditions and independently of the injector diameter. From this value on, the flow undergoes a transition from a steady laminar flow to a self-sustained chaotic flow regime with strong mixing dynamics. For Chamber no. 1 a hysteretic behavior was observed for increasing and decreasing flow rate increments.

If ϕ_K ($\phi_K \neq 1$), is maintained constant and Re is increased in both jets accordingly, the impingement point moves closer to the side of the jet with the lowest Re , towards the chamber walls. This phenomenon indicates that the impinging jets flow becomes more sensitive to small deviations in flow rates as Re is increased. In the case of RIM, the increase of Re demands a more precise and strict monomers or pre-polymers flow rate control and the system becomes more sensitive to temperature of the polymer production plant.

An elastic analogue model of the jets' behavior was developed to predict the impingement point position under different flow conditions, showing good agreement with computational and experimental results of the flow. The present study suggests that higher mixing efficiency is obtained when the operating conditions and mixing chamber design allow for the jets' to impinge at the center of the mixing chamber. Contrarily to the reported by other authors (Malguarnera and Suh, 1977; Sebastian and Boukobbal, 1986; Hosseinalipour and Mujumdar, 1997), optimum conditions are obtained when the kinetic energy rate of both jets are balanced, and not its momenta.

The study of this chapter shows that impingement mixing should occur in the center of the mixing chamber, setting ϕ_K as estimated by the proposed elastic analogue model, and for $Re \geq 110$. Injectors' diameter can be adapted to comply simultaneously with reaction stoichiometry.

5.6 References

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6 Final Remarks

The present chapter is divided in two sections. The first, Section 6.1, summarizes the main conclusions of the present work. The second, Section 6.2, provides a list of suggestions for future work.

6.1 Conclusions

The main goals of this work were the identification of mixing mechanisms and flow regimes and the characterization of the lamellar microstructure of the flow in impinging jets mixers, with applications to the flow in a mixing chamber of a Reaction Injection Molding machine. The flow was studied numerically from Computational Fluid Dynamics (CFD) tools and experimentally from Particle Image Velocimetry (PIV) and Planar Laser Induced Fluorescence (PLIF).

An introduction to impinging jets mixers and Reaction Injection Molding technology and its actual economical impact are presented. It is followed by the motivation and relevance of this dissertation, as also as its objectives and layout.

The mixing length scales and stretching properties of impinging jets flow are studied in a mixing chamber from 3D Computational Fluid Dynamics (CFD) simulations

coupled with a Lagrangian tracking of the position and deformation of passive material fluid elements.

The lamellar structure of the flow is also studied from an Eulerian-Eulerian approach using a 2-dimensional CFD model. The model describes the purely advective mixing of two immiscible feeding fluids using the Volume-of-Fluid (VOF) model, without the effect of diffusion and surface tension. A dynamic adaptive grid algorithm in the region of higher gradients of volume fraction is included in the simulation model to describe smaller mixing length scales without the need to refine equally the entire domain.

Planar Laser Induced Fluorescence (PLIF) flow visualization technique is used to obtain 2D concentrations fields at the top and bottom of the mixing chamber. One of the injected streams was marked with rhodamine 6G which emits fluorescence when excited by laser light, allowing to detect mixing patterns in the flow. The obtained fields are used to analyze qualitatively and quantitatively the effect of the Reynolds number and the impinging jets' unbalancing on mixing.

Finally, the impinging jets' interaction is studied theoretically, experimentally and numerically. An elastic analogy of the two impinging jets behavior is used to predict the impingement point position under different flow conditions, showing good agreement with CFD simulations of the flow and results from PIV experiments. The study offers deeper insight on the system behavior under unbalanced jets flow conditions in order to optimize polymer production from monomers or pre-polymers formulations with different physical properties or stoichiometric ratios different from 1 in RIM.

In the next sections, the main achievements of this dissertation are presented, namely the observed flow regimes (Section 6.1.1), the stretching and striation thickness distribution in the flow (Section 6.1.2) and the conclusions about the impact of the jets' unbalancing on the flow (Section 6.1.3).

6.1.1 Flow Regimes

Three different flow regimes were identified in the range of Reynolds number of industrial interest in the production of polymers from RIM:

- $Re < 105$: The flow is *laminar and steady* with poor mixing properties. The flow remains segregated with each side of the mixing chamber containing mainly one of the injected fluids.
- $105 \leq Re \leq 110$: A dynamic flow regime is onset. The flow evolves to a *self-sustained oscillatory periodic laminar regime* characterized by the disturbance of the two impinging jets' interface and the formation of alternated detaching vortices in the mixing chamber downstream the inlet.
- $110 < Re < 600$: The formation of the vortices occurs earlier in the flow, i.e. closer to the impingement point, and start to interact with the inlet jets. At this point, the flow develops to a *self-sustained laminar chaotic regime* with strong dynamics and good mixing properties.

Results have shown that advective mixing in opposed impinging jets is observed to occur above a critical Reynolds number $Re_c > 110$. This critical Reynolds number value is shown, in Chapter 2 from Particle Image Velocimetry experiments, to be same for two different mixing chambers with different injectors' diameters.

6.1.2 Stretching and Striation Thickness Distributions

From the calculation of the stretching experienced by the advected passive elements in the flow, this work shows that, in the mixing regime, the reduction of scales in opposed impinging jets mixers results from an increase of the stretching rate combined with a strong flow reorientation, both at the top of the mixing chamber. In reactive polymer injection, the combination of the two mechanisms (stretching and reorientation) allows the flow to exponentially generate interfacial area between the two monomers or pre-polymers in RIM and reduce scales to a value that avoids diffusional limitations during the polymerization reaction in the mold. In this regime

the flow is shown to produce nearly exponential average stretching of fluid elements with time, a feature of chaotic flows. Impingement mixing is observed to occur in the top of the chamber until a distance approximately equal to twice the chamber diameter from the top wall. In the remaining chamber the flow develops a nearly parallel profile. The flow in the bottom of the chamber still contributes to further reduction of mixing length scales but linearly in time due to the orientation of trajectories with the streamlines. Further reduction of striation thicknesses in the monomers or pre-polymers mixture is observed with the increase in Reynolds number.

Computational and experimental results of this work are compared to theoretical models existent in the literature for the estimation of laminae scales in impingement jets mixing. Results show to be consistent with the kinematic model of Lee et al. (1980), rather than with models based on the statistical theory of turbulent diffusion (Tucker III and Suh, 1980; Baldyga and Bourne, 1983; Bourne and Garcia-Rosas, 1985; May, 1996). The model of Lee et al. (1980) considers that the impingement process is, on a local scale, a rapid, two dimensional stagnation flow, assuming that the stretching remains in the plane of each lamina.

The observed patterns and distribution of lamellae thicknesses indicate that the flow up to $Re = 600$ and for $Re \geq Re_c$ is laminar and that mixing is achieved by the advective mechanisms of stretching and folding. The results of this dissertation strongly indicate that isotropic and homogeneous turbulence considerations for the modeling of mixing scales do not provide a correct physical description of the flow.

CFD Simulations and PLIF experimental results show the formation of striation scales thin enough to be faded by diffusion during time periods close to the mixing chamber mean residence time. This result suggests an additional mechanism of *rupturing*, associated with stretching and folding mechanisms, capable of generating isolated blobs of material in the medium.

The results of this dissertation offer deeper insight on how impingement mixing in RIM produces striation length scales sufficiently thin for the rate of polymerization reaction in the mold to become chemically controlled.

6.1.3 Jets Flow Unbalancing

Unbalanced jet conditions were found to affect the flow significantly by moving the impingement point towards the chamber walls. Under some unbalanced conditions, the jets' oscillations are partially or completely damped, even when a dynamic flow regime is expected.

The present dissertation suggests that higher mixing efficiency is obtained when the operating conditions and mixing chamber design allow for the jets' to impinge at the center of the mixing chamber. Contrarily to the reported by other authors (Malguarnera and Suh, 1977; Sebastian and Boukobbal, 1986; Hosseinalipour and Mujumdar, 1997), optimum conditions are obtained when the kinetic energy rate of both jets are balanced, and not the respective momenta.

When the Reynolds number is increased, maintaining the jets' kinetic energy rate ratio and at values different from one, the impingement point moves towards the chamber walls, closer to the lowest Reynolds number jet side. This phenomenon indicates that the impinging jets flow becomes more sensitive to small deviations in flow rates as Re is increased. In the case of RIM, the increase of Re demands a more precise and strict monomers or pre-polymers flow rate control and the system becomes more sensitive to temperature of the polymer production plant.

The present dissertations indicates that impingement mixing must occur at the center of the mixing chamber for better mixing efficiency, setting the kinetic energy rate ratio as estimated by the proposed elastic analogue model, and for $Re > 110$. The injectors' diameter of the mixer can be adapted to comply simultaneously with reaction stoichiometry.

6.2 Future Work

The present work offers insight about the mixing mechanisms and the reduction of mixing scales in the Confined Impinging Jet mixer of a RIM machine. Further work can still be suggested, some due to the available computational power during the period of this dissertation. Some suggestions are presented below.

- Use of the Eulerian-Lagrangian approach presented in Chapter 2 to study mixing in impingement jets flow under different jets' kinetic energy rate ratios and evaluate the impact in the distribution of striation thicknesses.
- Use of the CFD mixing model for the description of the lamellar structure of the flow of Chapter 3 to investigate mixing of fluids with different physical properties, specially with high ratios of viscosity usually found in the RIM process.
- Implement the non-diffusive dynamic grid adaptive mixing model described in Chapter 3 in a 3D CFD model of the flow in the mixing chamber. With the fast advances in computer hardware, this simulation is expected to be possible in a close future.
- Evaluation of the actual impact in the final physical properties of a polymer produced from impingement mixing and the reaction kinetics with design parameter and operational conditions set according to the rules suggested in Chapter 5. Such properties could be assessed from Dynamic Mechanical Analysis (DMA) experiments to test the viscoelastic behavior of the polymeric material and the evaluation of the adiabatic rise of temperature in the mold.

6.3 References

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