



UNIVERSITÀ DEGLI STUDI DI PALERMO

PhD Course in Chemical, Environmental, Biomedical, Hydraulic and Materials Engineering
Dipartimento di Ingegneria
Settore Scientifico Disciplinare (ING-IND/25)

Modelling and Simulation of multiphase and supercritical fluid systems

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XXXVII CYCLE
GRADUATION YEAR 2025

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ACKNOWLEDGEMENTS

I would like to express my deepest gratitude to the individuals and institutions that have significantly contributed to the completion of this thesis.

First and foremost, I would like to thank my supervisor, Prof. Francesca Scargiali, for her invaluable guidance, constant encouragement, and unwavering belief in my abilities. Her intellectual support, exemplary dedication, and passion for academia have been fundamental to my journey. My heartfelt thanks also go to my co-supervisor, Prof. Michele Ciofalo, who, despite being retired, followed me throughout this path with invaluable advice, teachings, and entire afternoons spent solving numerous challenges.

I am sincerely grateful to Professors Giuseppe Caputo and Franco Grisafi for their support and advice throughout these three years.

A special acknowledgment is also due to Prof. Giorgio Domenico Maria Micale, coordinator of my program, for his role in organizing and supervising the doctoral course.

I extend my gratitude to Prof. Joelle Aubin, my supervisor during my time abroad at the "*École Nationale Supérieure des Ingénieurs en Arts Chimiques et Technologiques*" in Toulouse, France, for welcoming me into her university and research group and for her step-by-step support in my work. I am also grateful to Prof. David Fletcher, who was always available to help when needed.

I am thankful to my colleagues and friends at the Department of Engineering whom I met during these three years of doctoral studies: Pasquale, Serena, and Alessandro, with whom I shared my daily experiences; Noemi, Arima, Francesco, Sara,

Rita, and Georgina for their constant moral support, stimulating discussions, and collaboration during challenging times. Their friendship has made this journey more enjoyable and enriching.

Last but not least, I want to thank my family for their unconditional love and patience during these intense years of study, as well as those who supported me consistently. Gregory, Emanuela, Valentina and Cristina: without your encouragement and sacrifices, this achievement would not have been possible.

My heartfelt thanks to all.

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SUMMARY

This doctoral thesis, entitled "Modelling and Simulation of Multiphase and Supercritical Fluid Systems," presents an extensive investigation into the numerical and experimental study of fluid behavior within complex multiphase systems. The primary objective is to advance the understanding of both multiphase and supercritical fluid dynamics and to offer valuable insights for enhancing industrial process efficiency. The research is structured around three main areas of investigation, each pivotal to the advancement of theoretical and practical aspects of fluid mechanics.

The first major focus of this work is the extraction of omega-3 fatty acids from fish oil using supercritical carbon dioxide (sc-CO₂). This study integrates experimental methodologies with Computational Fluid Dynamics (CFD) approaches to analyze the hydrodynamic conditions within the extraction column, particularly examining flooding conditions. CFD simulations employ the Volume of Fluid (VOF) method to accurately model the complex phase interactions and monitor the behavior of sc-CO₂ within the system. The simulation results exhibit a strong agreement with experimental data, validating the VOF method's effectiveness in capturing the dynamic and intricate nature of multiphase flows in supercritical extractions. This combined approach not only enhances the understanding of sc-CO₂ behavior but also provides guidelines for the design and optimization of industrial-scale extraction processes.

The second area of study investigates the transition between non-aerated and aerated regimes in Uncovered Unbaffled Stirred Tanks (UUSTs). Through CFD simulations, this research accurately predicts the hydrodynamic transitions occurring

within the vessel, including vortex formation and bubble dispersion as the impeller speed increases. Special emphasis is placed on the variations in power consumption associated with these regimes. The simulations deliver detailed insights into how aeration affects mixing efficiency, energy usage, and the overall behavior of the system. The predictive capabilities of the CFD models developed in this study serve as a robust tool for engineers aiming to optimize mixing processes in chemical and biochemical applications.

The third focal point is the investigation of fluid dynamics within vibromixers, an innovative technology used for process intensification in industrial applications. Comprehensive CFD analyses are conducted to examine how the vibrational motion of mixing plates influences fluid flow and mass transfer. A significant finding is the high shear rate observed during biodiesel production processes, impacting the formation of the required emulsion and attributed to the unique flow patterns generated by the vibromixer. Additionally, the study explores the behavior of solid-liquid suspensions under varying operational conditions, providing valuable data on suspension stability and distribution. These findings underscore the potential of vibromixer technology to enhance reaction rates and overall process efficiency.

The collective outcomes of these three studies underscore the practical applications of advanced CFD techniques in the design and optimization of complex fluid systems. By modeling and simulating detailed interactions between phases in supercritical and multiphase environments, with a particular focus on the VOF method when appropriate, this work lays the groundwork for more efficient and effective

engineering solutions in the chemical, biochemical, and energy sectors, providing a deeper analysis of heat and mass transfer.

Thesis Structure

➤ **Chapter 1** introduces the fundamental principles of multiphase flow and the computational approaches utilized in this research. It sets the stage by discussing the key challenges associated with modeling dynamic interfaces in complex multiphase systems and explores the mathematical tools and numerical techniques employed, with a focus on the VOF method.

➤ **Chapter 2** details the experimental and computational investigation of Supercritical Fluid Extraction (SFE) of omega-3 fatty acids. This chapter outlines the experimental setup, properties of the sc-CO₂ system, and the corresponding CFD model used to simulate the extraction process under various operational conditions. The analysis highlights the alignment between computational results and experimental fluid dynamics data, reinforcing the reliability of the modeling approach.

➤ **Chapter 3** explores the transition from non-aerated to aerated regimes in UUSTs. This chapter provides an in-depth examination of the CFD simulations performed to analyze vortex formation, power consumption, and bubble dispersion under different operational states. The findings contribute to a deeper understanding of mixing dynamics and guide the optimization of unbaffled stirred tanks for enhanced process performance.

➤ **Chapter 4** focuses on the CFD analysis of fluid dynamics within vibromixers. It discusses the operational principles of vibromixers, the computational setup, and the results of simulations aimed at optimizing mass transfer during biodiesel production as well as solid-liquid suspension behavior. The chapter concludes with an assessment of how vibromixer technology can be leveraged to improve process intensification.

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LIST OF PUBLICATIONS

The research work conducted has thus far resulted in the following publications:

L. Schembri, G. Caputo, M. Ciofalo, F. Grisafi, S. Lima, F. Scargiali. “*CFD simulations of the transition between non-aerated and aerated conditions in uncovered unbaffled stirred bioreactors*”. Chem. Eng. Sci. vol. 302, part A, 120824, 2025, <https://doi.org/10.1016/j.ces.2024.120824>.

L. Schembri, G. Caputo, M. Ciofalo, F. Grisafi, S. Lima, and F. Scargiali, “*Experimental and Computational Study of Supercritical Fluid Extraction (SFE) of Omega-3 Components from Fish Oil in Structured Packing*,” Chem. Eng. Trans., vol. 100, pp. 337–342, 2023, <https://doi.org/10.3303/CET23100057>

In Preparation:

S. Almasi, L. Schembri, M. Gimba, F. Scargiali, M. Poux, J. Aubin “*Vibromixer for FAME Synthesis through Transesterification Reaction*”. (2025).

Other Publications:

S. Lima, V. Amari, L. Schembri, F. Grisafi, G. Caputo, and F. Scargiali, “*Kinetic of the Sewage Treatment: The Consumption of Organic Carbon of The Microalga Chlorella sp.*,” Chem. Eng. Trans., vol. 99, pp. 325–330, 2023, <https://doi.org/10.3303/CET2399055>

S. Lima, A. Brucato, L. Schembri, F. Grisafi, G. Caputo, and F. Scargiali, “*Modelling Nannochloropsis gaditana Growth in Reactors with Different Geometries, Determination of Kinetic Parameters and Biochemical Analysis in Response to Light Intensity*,” Appl. Sci. 2022, 12, 5776. <https://doi.org/10.3390/app12125776>

CONFERENCE, SEMINAR, AND DOCTORAL SCHOOL

3rd Baldyga Technical Seminar: *“Mixing challenges in biochemical and chemical industries”*

- September 2024, London, University College London, United Kingdom.

GRICU Winter School 2024: *“Chemical engineering of surfaces and interfacial phenomena and advanced numerical tools for chemical engineering”*

- January 2024, Bardonecchia (TO), Olympic Village Bardonecchia, Italy.

17th European Conference on Mixing

- July 2023, Porto, Fundação Dr. António Cupertino de Miranda, Portugal.

ICHEAP16, 16th International Conference on Chemical and Process Engineering

- May 2023, Naples, Hotel Ramada, Italy.

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- February 2023, Zurich, ETH Zürich, Switzerland.

19th UIT Summer School: *“Heat and mass transfer in turbulent flows: modeling and measurement techniques”*

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GRICU Conference & School 2022: *“Fundamentals of Electrochemistry Processes and their Application and Heterogeneous Catalysis”* - *“The Central Role of Chemical Engineering in a Changing World”*

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- **Poster**

GRICU 2022: Modeling of the growth of *Nannochloropsis gaditana* in different reactor geometries, experimental kinetic parameters from batch cultures, and biochemical analysis of the obtained biomass.

- **Virtual Poster**

GRICU 2024: CFD simulations of the transition between non-aerated and aerated conditions in uncovered unbaffled stirred bioreactors.

Presentations at International Conferences:

- **Oral Presentation**

ICHEAP16: Experimental and computational study of Supercritical Fluid Extraction (SFE) of omega-3 components from fish oil in structured packing.

- **Oral Presentation**

3rd Baldyga Technical Seminar: Vibromixer for FAME Synthesis through Transesterification Reaction.

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Mar. - Apr. 2022, Speaker: Prof. Giuseppe Aiello, University of Palermo.

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Preparation and Characterization of (Bio)Polymer-Based Micro and Nanostructured Systems

Apr. 2022, Speakers: Prof. R. Scaffaro, Ing. A. Maio, University of Palermo.

Nanoscience and Nanomaterials: Pushing the Boundaries of Technology

Apr. 2022, Speakers: Prof. Clelia Dispenza, Ph.D. Emanuela Muscolino, University of Palermo.

Green Hydrogen

May 2022, Speaker: Ph.D. Fabrizio Ganci, University of Palermo.

Bio-coatings for Implanted Devices

Jun. 2022, Speakers: Prof. Rosalinda Inguanta, Ph.D. Claudio Zanca, University of Palermo.

European Charter for Researchers: Innovative Start-Ups

Jun. 2022, Speaker: Dr. Stefano Gargaliano, University of Palermo.

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Nov. 2022, Speaker: Dr. Dan Wiesenfeld, University of Palermo.

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Jun. 2024, Speaker: Prof. Li Puma, University of Palermo.

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Plant Design for Process Sustainability

Mar. May 2024, Speaker: Prof. Giuseppe Caputo, University of Palermo.

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NOTATION

English

a_i	interfacial area
A	area
A	amplitude
A_{pcs}	projected cross-sectional area of particle
C_{am}	added mass coefficient
C_D	drag coefficient
d	diameter
$\mathcal{D}_1, \mathcal{D}_2$	deformation tensor
$\mathcal{D}_i$	momentum drift flux tensor
$\hat{e}$	unit vector
Eo	Eötvös number
Eu	Euler number
f	liquid volumetric fraction
f	frequency
f	force per unit volume
$\mathbf{F}$	force
g	acceleration of gravity
$\mathbf{g}$	gravity vector
H	mean curvature
$\mathbb{I}$	unit tensor

$\mathbf{j}$	superficial velocity
k	turbulent kinetic energy
m_p	particle mass
M_1, M_2	momentum transfer terms
Mo	Morton number
N_p	number of particles
n_p	particle number density
$\hat{\mathbf{n}}_1, \hat{\mathbf{n}}_2$	unit vector to the interface pointing in phase 1 and 2, respectively
p	pressure
R	radius
Re	Reynolds number
S_i	phase interface
S_t	Stokes number
t	time
$\mathbb{T}$	stress tensor
U	velocity of continuous phase
V	velocity of disperse phase, averaging volume
V_c	volume of mesh cell
$\mathbf{V}_p$	translational velocity of center-of-mass of particle
$\mathcal{V}$	volume
$\mathbf{v}$	velocity field
We	Weber number

$\mathbf{x}$	position vector
X_k	phase indicator function
$\mathbf{X}_p$	position vector to center-of-mass of particle

Greek

α_1, α_2	volume fraction
Γ_1, Γ_2	mass transfer across interface due to phase change
Γ_μ	viscosity ratio
Γ_ρ	density ratio
δ	Dirac delta function
ε	dissipation rate of turbulent kinetic energy
κ	interface curvature
μ	dynamic viscosity
ρ	density
$\Delta\rho$	absolute value of density difference
σ	coefficient of surface tension
τ_p	particle response time
φ	level-set function
ψ	sphericity
Ω	domain

Subscripts

1	phase 1 (continuous phase)
2	phase 2 (disperse phase)
B	bubble
body	body force
buoy	buoyancy
c	continuous
d	disperse
eq	equivalent
f	cell face
grav	gravity
h, hydr	hydrodynamic
i	interface
k	phase index ($k = 1, 2$)
m	mixture
p	particle
q	phase
r	relative
surf	surface
turb	turbulent
T	terminal velocity

Superscripts

sgs	subgrid stress
T	transposed vector or tensor

Fonts

$\mathbf{f}, \mathbf{F}$	vector
f, F	scalar ($f = \mathbf{f} , F = \mathbf{F} $)
$\hat{\mathbf{f}}$	unit vector
$\mathbb{F}$	tensor

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CHAPTER 1: Multiphase Flow and Computational Approach

1.1 Introduction

This chapter provides an overview of the fundamental physics governing multiphase flow, along with its mathematical representation and physical modeling for numerical computation through Computational Fluid Dynamics (CFD) codes. The modeling and numerical simulation of multiphase flows introduce specific challenges, predominantly due to the presence of dynamic interfaces that separate different phases or immiscible fluids. Typically, the geometry of these interfaces and the spatial distribution of phases are not predefined but emerge as part of the solution. Key modeling challenges relate to physical transfer processes occurring across these interfaces, including momentum, heat, and mass transfer, as well as phase changes. Additionally, numerical challenges arise from the moving nature of the interface, where properties such as density, viscosity, and pressure often exhibit discontinuities.

The focus of this chapter is primarily on gas-liquid and liquid-liquid flows, with a specific emphasis on the hydrodynamic aspects of multiphase flows. Topics such as heat transfer, mass transfer, and phase change—often associated with two-phase engineering applications—are beyond the scope of this discussion. Instead, this chapter centers on fluid dynamic modeling methods, particularly the Volume of Fluid (VOF) Method, which is explored in detail in the first two case studies of this

thesis: Supercritical Fluid Extraction (SFE) and fluid dynamics within Uncovered Unbaffled Stirred Tanks (UUSTs).

1.1 Fundamentals of multiphase flow

1.2.1 Notions

In thermodynamic terms, a “*phase*” refers to a distinct state of matter, which may be solid, liquid, or gas. Multiphase flow involves the simultaneous flow of multiple phases. These phases may comprise a single chemical component, such as water and its vapor (steam), or multiple chemical components, such as oil and water.

Within multiphase terminology, a phase is classified as *continuous* if it occupies continuously connected regions of space, whereas it is termed *disperse* when it occupies disconnected regions of space. The continuous phase may be either gaseous or liquid, while the disperse phase consists of *particles*. The term particle may denote either a rigid (solid) particle or a fluid particle. Fluid particles arising from a gas phase are referred to as *bubbles*, while those originating from a liquid phase are known as *drops*. In this chapter, the continuous phase is indicated by the subscript “*c*” and the disperse phase by the subscript “*d*”. Alternatively, the phases may be denoted by subscript k , where $k=1, 2$. Phase one ($k = 1$) is considered continuous, while phase two ($k = 2$) may be either continuous or disperse (Bird et al., 2007).

In two-phase or multi-fluid flows, the dynamics of all phases are of interest. This focus contrasts with free surface flows, in which the influence of gas density

and viscosity is typically neglected, and only the motion within the liquid phase is considered. This foundation in multiphase flow terminology and phase classification is essential to understanding the modeling and simulation approaches discussed in subsequent sections (H.D. Baehr, 1998).

1.2.2 Morphology and gas-liquid flow regimes in pipes

Gas-liquid two-phase flows can manifest in various topological or morphological configurations, commonly referred to as flow regimes or flow patterns. The importance of identifying the flow regime lies in the fact that the physical transfer processes occurring across the phase interface are significantly influenced by the specific flow regime. Consequently, selecting appropriate models for interfacial transport necessitates the prior identification of the prevailing flow regime. **Figure 1.1** and **Figure 1.2** illustrate typical flow regimes that occur in horizontal and vertical pipes, respectively.

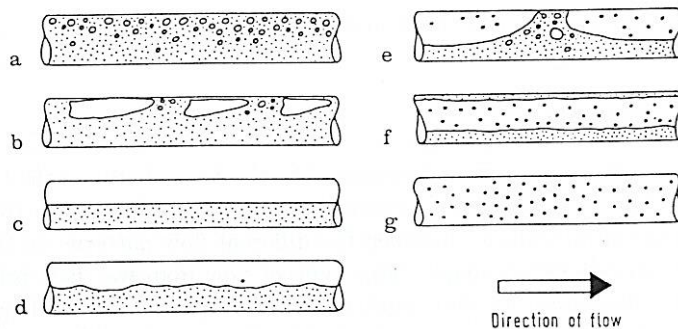


Figure 1.1: Flow patterns in a horizontal, unheated tube: a) bubble flow; b) plug flow; c) stratified flow; d) wavy flow; e) slug flow; f) annular flow; g) spray or drop flow. (H.D. Baehr, 1998)

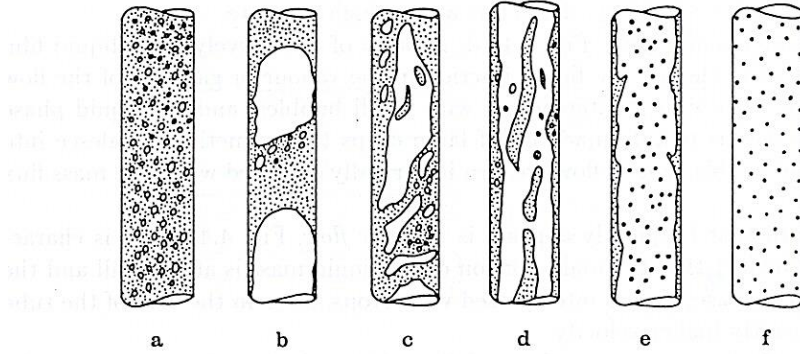


Figure 1.2: *Flow types in a vertical, unheated tube with upward flow: a) bubble flow; b) plug flow; c) churn flow; d) wispy - annular flow, e) annular flow; f) spray or drop flow (H.D. Baehr, 1998).*

The conventional parameters used to identify flow regimes are the superficial velocities of the phases, which represent the volumetric flow rate of each respective phase divided by the channel cross-sectional area. However, the superficial velocities alone are usually not sufficient to characterize the geometrical configuration of two-phase flows. Generally, the flow regime depends on the physical properties of the phases or fluids as well as the channel geometry, such as the pipe diameter.

1.2.3 Fundamental forces in multiphase flow

Any fluid motion originates from forces acting on fluid elements. In general, forces can be classified in three different categories. *Volume forces* (also called body forces) act on a volume element of size $\mathcal{V} \propto L^3$, surface forces act on a surface or area element of size $\mathcal{V} \mathcal{A} \propto L^2$, and line forces act on a curve element of size $\mathcal{C}$

$\propto L$, where L is a linear dimension. In Table 1 we summarize the forces that are important in two-phase flow.

The **pressure force** acts on area or surface elements, accelerating the fluid in the direction of the pressure gradient. The inertia force, a volume force, tends to maintain the current direction and magnitude of motion. The **viscous force** acts on a surface or area element, promoting uniformity in the flow field and reducing velocity differences. The **gravity force** accelerates the fluid in the direction of the gravity vector, with the buoyancy force being related to gravity; it represents the difference between the gravity force and the Archimedes force. Buoyancy reflects the net effect of gravity when density varies across the fluid. In single-phase flows, density variation often results from temperature differences, leading to natural convection. In two-phase flows, non-uniform density arises from the presence of different phases. Finally, the **surface tension force** acts on a line or curve element, working to minimize the surface area of the interface, and is specific to gas-liquid or liquid-liquid two-phase flows (Wörner, 2003).

1.2.4 Non-dimensional groups

In two-phase flows, the relative significance of various forces can vary considerably. Prior to approaching a flow problem numerically, it is beneficial to estimate the magnitude of the forces involved. A practical method for identifying the dominant forces, as well as those that may be considered negligible, is through the use of dimensionless groups, which express the ratio between two forces.

From the six fundamental forces presented in **Table 1.1**, five independent dimensionless groups can be derived.

Table 3.1: Force in multiphase flow and their magnitude.

Force	Type	Magintude of	
		Force	Force per unit volume
Pressure force	Surface force	$F_p \propto \mathcal{A} \Delta p$	$f_p \propto \Delta p L^{-1}$
Inertia force	Volume force	$F_I \propto \mathcal{V} \rho \bar{v}^2 L^{-1}$	$f_I \propto \rho \bar{v}^2 L^{-1}$
Viscous force	Surface force	$F_V \propto \mathcal{A} \mu \bar{v} L^{-1}$	$f_V \propto \mu \bar{v} L^{-2}$
Gravity force	Volume force	$F_G \propto \mathcal{V} g \rho$	$f_G \propto g \rho$
Buoyancy force	Volume force	$F_B \propto \mathcal{V} g \Delta \rho$	$f_B \propto g \Delta \rho$
Surface tension force	Line force	$F_S \propto \mathcal{C} \sigma$	$f_S \propto \sigma L^{-1}$

These groups are often named after prominent scientists, with the Reynolds number being perhaps the most well-known. The **Reynolds number** represents the ratio between inertial and viscous forces:

$$Re = \frac{F_I}{F_V} = \frac{f_I}{f_V} = \frac{\rho L \bar{v}}{\mu} \quad (1.1)$$

The **Euler number** represents the ratio between pressure gradient and inertia forces:

$$Eu = \frac{F_p}{F_I} = \frac{f_p}{f_I} = \frac{\Delta p}{\rho \bar{v}^2} \quad (1.2)$$

The **Froude number** characterizes the ratio of inertia and gravity forces:

$$Fr = \frac{F_I}{F_G} = \frac{f_I}{f_G} = \frac{\bar{v}^2}{g L} \quad (1.3)$$

Next, the dimensionless groups specific to gas-liquid and liquid-liquid two-phase flows are introduced, as these flows involve the surface tension force. The **Weber number**, for instance, characterizes the ratio of inertial forces to surface tension forces, offering insight into the behavior of the interface between phases under varying flow conditions.

$$We = \frac{F_I}{F_S} = \frac{f_I}{f_S} = \frac{\rho L \bar{v}^2}{\sigma} \quad (1.4)$$

and the **Eötvös number** the ratio between buoyancy forces and surfaces tension forces:

$$Eo = \frac{F_B}{F_S} = \frac{f_B}{f_S} = \frac{\Delta \rho g L^2}{\sigma} \quad (1.5)$$

From the above non-dimensional groups further groups can be defined. An example is the **Capillary number**

$$Ca = \frac{F_V}{F_S} = \frac{f_V}{f_S} = \frac{\mu v}{\sigma} = \frac{We}{Re} \quad (1.6)$$

which represents the ratio between viscous and surface tension forces. A particular useful number for gas-liquid and liquid-liquid flows is the **Morton number** which involves the continuous phase density and viscosity and is defined as

$$Mo = \frac{F_B F_V^4}{F_I^2 F_S^3} = \frac{g \Delta \rho \mu_c^4}{\rho_c^2 \sigma^3} = \frac{Eo We^2}{Re^4} \quad (1.7)$$

The Morton number involves only fluid properties. Thus, for a given isothermal incompressible two-phase system it is a constant. For real fluids the Morton number ranges between 10^{-14} for gas bubbles in liquid metals and 10^8 for gas bubbles in viscous oils. For air bubbles in water the value of the Morton number is $Mo = 2.48 \cdot 10^{-11}$.

$$Mo = \frac{F_B F_V^4}{F_I^2 F_S^3} = \frac{g \Delta \rho \mu_c^4}{\rho_c^2 \sigma^3} = \frac{Eo We^2}{Re^4} \quad (1.8)$$

Two further non-dimensional groups are the density and viscosity ratio of the phases

$$\Gamma_\rho = \frac{\rho_d}{\rho_c}, \quad \Gamma_\mu = \frac{\mu_d}{\mu_c} \quad (1.9 \text{ a, b})$$

Thus far, the characteristic length and velocity scales, L and U , used in the aforementioned dimensionless groups, have not been explicitly defined. Selecting appropriate values for these parameters depends on the specific flow type under analysis. For the motion of rigid or fluid particles influenced by gravity, a meaningful characteristic velocity scale is the terminal vertical velocity, V_T , representing the particle's rise or fall rate. An appropriate length scale in this context is the volume-equivalent diameter, d_{eq} , defined as the diameter of a sphere with the same volume, V_p , as the particle itself.

$$d_{eq} \equiv \left(\frac{6 V_p}{\pi} \right)^{\frac{1}{3}} \quad (1.10)$$

Similarly, a surface equivalent diameter can be defined

$$d_s \equiv \left(\frac{\mathcal{A}_p}{\pi} \right)^{\frac{1}{2}} \quad (1.11)$$

which is the diameter of a sphere that has the same surface area, $\mathcal{A}_p$, as the particle.

Another characteristic diameter that is commonly used is the **Sauter diameter**

$$d_{32} \equiv \frac{6 V_p}{\mathcal{A}_p} \quad (1.12)$$

For a sphere the above diameters are all equivalent and are equal to the diameter of the sphere. The square of the ratio of equivalent diameter to surface diameter constitutes the **sphericity**:

$$\psi = \frac{\text{surface area of volume-equivalent sphere}}{\text{surface area of particle}} = \frac{\pi d_{eq}^2}{\pi d_s^2} = \left(\frac{d_{eq}}{d_s} \right)^2 \quad (1.13)$$

which represents a shape factor. As a spherical particle has the smallest possible area per unit volume the maximum value of the sphericity is 1. Thus, for any particle shape the values of the sphericity are in the range $0 < \psi \leq 1$.

1.2.5 Boundary conditions at a fluid interface

The boundary delineating the bulk regions of two fluids is commonly referred to as the *interface*. This interface represents a material region across which physical quantities vary continuously. Typically, the interface is only a few

molecules thick; however, given that this thickness is much smaller than other relevant length scales, it is often convenient to conceptualize a “*functional interface*.” The functional interface is idealized as having zero thickness but is associated with an interfacial tension, denoted by σ . Nevertheless, the finite thickness of the interface may become significant in certain cases, such as in moving contact lines and in systems involving supercritical fluids (Anderson et al., 1998).

In this thesis, the discussion is limited to functional interfaces, allowing the interface to be treated as a discontinuity within the density and viscosity fields. At a functional interface, the phases are interconnected by both a kinematic condition and a dynamic condition. For simplicity, it is assumed throughout that there is no interfacial molecular diffusion (mass transfer) or phase change occurring at the interface. The kinematic condition states that the velocity normal to the interface is continuous

$$\mathbf{v}_{1i} \cdot \hat{\mathbf{n}}_1 = \mathbf{v}_{2i} \cdot \hat{\mathbf{n}}_1 \quad (1.14)$$

If both fluids are viscous then also the tangential velocity at the interface is continuous so that

$$\mathbf{v}_{1i} = \mathbf{v}_{2i} \quad (1.15)$$

The **dynamic condition** states that at the interface the pressure and viscous stresses are in equilibrium with forces due to surface tension

$$-(p_{1i} - p_{2i}) \hat{\mathbf{n}}_1 + (\mathbb{T}_{1i} - \mathbb{T}_{2i}) \cdot \hat{\mathbf{n}}_1 = 2 H \sigma \hat{\mathbf{n}}_1 + \nabla_s \sigma \quad (1.16)$$

Here, $\nabla_s = \mathbb{I}_s \cdot \nabla = (\mathbb{I} - \hat{\mathbf{n}}_1 \hat{\mathbf{n}}_1) \cdot \nabla$ is the surface gradient operator. In the thesis, only incompressible Newtonian fluids are considered, such that the constitutive equation for viscous stress applies:

$$\mathbb{T}_k = 2 \mu_k \mathbb{D}_k, \quad \text{where } \mathbb{D}_k = \frac{1}{2} (\nabla \mathbf{v}_k + (\nabla \mathbf{v}_k)^T) \quad (1.17)$$

Insertion of Eq. (1.17) in Eq. (1.16) and projection in direction of the unit normal vector $\hat{\mathbf{n}}_1$ gives:

$$-(p_{1i} - p_{2i}) + [\mu_1 (\nabla \mathbf{v}_1 + (\nabla \mathbf{v}_1)^T) - \mu_2 (\nabla \mathbf{v}_2 + (\nabla \mathbf{v}_2)^T)]_i \cdot \hat{\mathbf{n}}_1 \hat{\mathbf{n}}_1 = 2 H \sigma \quad (1.18)$$

while projection in the direction of a unit tangential vector $\hat{\mathbf{t}}$ gives:

$$[\mu_1 (\nabla \mathbf{v}_1 + (\nabla \mathbf{v}_1)^T) - \mu_2 (\nabla \mathbf{v}_2 + (\nabla \mathbf{v}_2)^T)]_i \cdot \hat{\mathbf{n}}_1 \hat{\mathbf{t}} = (\nabla_s \sigma) \cdot \hat{\mathbf{t}} \quad (1.19)$$

Here, H is the mean curvature:

$$H \equiv \frac{1}{2} \left(\frac{1}{R_{min}} + \frac{1}{R_{max}} \right) = -\frac{1}{2} \nabla_s \cdot \hat{\mathbf{n}}_1 \quad (1.20)$$

and R_{min} and R_{max} are the principal radii of curvature. For constant surface tension and in the absence of gravity and any motion the fluid particle has a spherical shape and Eq. (1.18) reduces to the well-known **Young-Laplace equation**

$$p_2 - p_1 = \frac{2\sigma}{R} \quad (1.21)$$

where R is the radius of the spherical bubble or drop.

In multiphase flow systems, the coefficient of surface tension across a fluid interface generally exhibits variability. This variation, denoted as σ , can primarily arise due to non-uniformities in either temperature or the concentration of an absorbed material at the interface. From Eq. (1.19), it becomes apparent that the resulting tangential force is balanced solely by a non-zero tangential viscous stress. Consequently, a non-uniform surface tension coefficient invariably induces motion, as it is infeasible to satisfy Eq. (1.19) with stationary fluids.

In free surface flows, the viscosity of phase 2 is typically disregarded. When surface tension remains uniform, this yields a condition of zero tangential stress. If surface tension is entirely neglected, alongside the density of the gas phase, the dynamic boundary condition on the gas side of the interface simplifies to a state of constant pressure, denoted as p_2 . This neglect of gas viscosity and density can be a practical simplification in cases of bubbles rising within a liquid. Under these conditions, the zero-tangential stress boundary condition of a deformable bubble significantly differs from that of a rigid, non-deformable particle, where a no-slip condition applies. These distinctions between bubbles and rigid particles profoundly influence vorticity generation at the surface, thereby affecting the structure of wake flows and the forces exerted by the surrounding fluid.

1.2 Continuous field formulation of multiphase flow

Below, various methods for the numerical modelling of two-phase flows are presented. This section specifically addresses methods that utilize a continuum

description of all phases involved. The continuum approach is applicable to liquid, gaseous, and solid phases and is grounded in an averaging procedure. Multiple averaging techniques are discussed within the literature, including time averaging (Ishii, 1975), volume averaging (Whitaker, 1999), and ensemble averaging (Drew and Passman, 1999), with ensemble averaging representing the most comprehensive method. Each averaging approach produces averaged equations that share a formally similar structure. This analysis focuses on the volume averaging procedure.

In this thesis, the Volume of Fluid (VOF) method plays a fundamental role in identifying the interface between phases within multiphase flow systems. The VOF method utilizes volume fractions to represent the presence and distribution of different phases within each computational cell, focusing specifically on tracking the volume fraction of fluid phases to locate and describe the behavior of interfaces between immiscible fluids, such as liquid and gas. In the VOF approach, volume averaging techniques are central, as they provide a means to describe multiphase regions where precise phase boundaries are challenging to resolve on a sub-cellular level.

Through volume averaging in interface tracking, the VOF method treats the interface as a “*diffuse*” region, where phase properties are averaged across multiple cells. This averaging produces a smooth transition in volume fractions across the interface region rather than a sharply defined boundary. While less precise than certain interface-tracking methods, this approach effectively captures multiphase

boundaries within a continuous field framework, allowing for efficient modeling of complex interactions between phases.

This section begins by outlining essential mathematical prerequisites in Section 1.3.1, followed by the derivation of volume-averaged conservation equations in Section 1.3.2. These equations are not inherently self-contained; therefore, additional model assumptions are introduced to achieve closure. In two-phase flows, relative motion between phases is common. Within the continuous field approach, three primary modeling concepts exist to capture this relative motion, each with distinct complexity and applicability. These models are presented in ascending order of complexity: the homogeneous model in Section 1.3.4 and the two-fluid model in Section 3.5.

1.3.1 Mathematical prerequisites

1.3.1.1 Phase indicator function X_k

Two distinct fluids or phases are considered, each occupying a disjoint, time-dependent domain $\Omega_k(t), k = 1, 2$ such that the union of these domains $\Omega = \Omega_1(t) \cup \Omega_2(t)$ is constant in time. The boundaries of these respective domains are denoted by $\partial\Omega_1, \partial\Omega_2$ and $\partial\Omega$. The interface separating the fluids is consequently defined by:

$$S_i(t) = (\partial\Omega_1 \cup \partial\Omega_2)/\partial\Omega \quad (1.22)$$

For each phase, a phase indicator function or characteristic function $X_k(x, t)$ is defined. This function takes a value of unity when the position vector x is located within fluid k at time t and zero otherwise

$$X_k(x, t) = \begin{cases} 1, & \text{if } x \in \Omega_k(t) \\ 0, & \text{otherwise} \end{cases} \quad (1.23)$$

The two-phase indicator functions are related through the condition

$$X_1 + X_2 = 1 \quad (1.24)$$

The gradient of the phase indicator function is connected to the interface unit normal vector $\hat{\mathbf{n}}_k$ and the Dirac delta function of the interface $\delta(x - x_i, t)$ by relation:

$$\nabla X_k = \hat{\mathbf{n}}_k \delta(x - x_i, t) \quad (1.25)$$

where x_i is a point on the interface. Another important relation is the *topological equation*

$$\frac{\partial X_k}{\partial t} + \mathbf{v}_i \cdot \nabla X_k = 0 \quad (1.26)$$

which expresses the fact that the material derivative of X_k is zero (Drew and Passman, 1999).

1.3.1.2 Volume averaging operators

The method of volume averaging (Drew and Passman, 1999; Whitaker, 1999) is introduced by considering a general representative volume element V , which is bounded by the surface ∂V and remains invariant with respect to both time and space. The centroid of

the volume element V is positioned at $\mathbf{x}$, and a relative position vector $\boldsymbol{\eta}$ is used to specify any point within V relative to this centroid, such that $\mathbf{x}_\eta = \mathbf{x} + \boldsymbol{\eta}$. Consequently, the volume fraction of phase k within the volume element V is determined as

$$\alpha_k \equiv \frac{1}{V} \int_V X_k(\mathbf{x} + \boldsymbol{\eta}, t) d\mathbf{x}_\eta = \frac{V_k}{V} \quad (1.27)$$

Two volume averaging operators are next defined for a general scalar or vector quantity, ψ_k , one over the entire volume element V and the other over the sub-volume, V_k , which is exclusively occupied by phase k , respectively:

$$\begin{aligned} \overline{\psi_k}^V &\equiv \frac{1}{V} \int_V \psi_k(\mathbf{x} + \boldsymbol{\eta}, t) X_k(\mathbf{x} + \boldsymbol{\eta}, t) d\mathbf{x}_\eta \\ \overline{\psi_k}^k &\equiv \frac{1}{V_k} \int_{V_k} \psi_k(\mathbf{x} + \boldsymbol{\eta}, t) X_k(\mathbf{x} + \boldsymbol{\eta}, t) d\mathbf{x}_\eta \end{aligned} \quad (1.28 \text{ a, b})$$

For each phase these averages are linearly related to one another through the respective volume fraction, namely

$$\overline{\psi_k}^V = \alpha_k \overline{\psi_k}^k \quad (1.29)$$

1.3.1.3 Gauss and Leibniz rule

In preparation for the derivations of volume-averaged equations to be presented in Section 1.3.2, two fundamental rules for volume averaging are introduced. The first of these is the Gauss rule for volume averaging, expressed as follows (Ishii, 1975):

$$\overline{X_k \nabla \psi_k}^V = \nabla \overline{X_k \psi_k}^V - \overline{\psi_k \nabla X_k}^V = \nabla \overline{X_k \psi_k}^V - \frac{1}{V} \int_{S_i \cap V} \hat{\mathbf{n}}_k \psi_{ki}(\mathbf{x} + \boldsymbol{\eta}, t) dS \quad (1.30)$$

Here, ψ_k is a general scalar or vector quantity and ψ_{ki} is the value of this quantity on the k -side of the interface. Furthermore, $S_i \cap V$ denotes the part of the interface that is within the averaging volume V .

Analogous to Eq. (1.30), the following result is obtained for averages involving time derivatives:

$$\overline{X_k \frac{\partial \psi_k}{\partial t}}^V = \frac{\partial}{\partial t} \overline{X_k \psi_k}^V - \overline{\psi_k \frac{\partial X_k}{\partial t}}^V \quad (1.31)$$

Using the topological equation (1.26) to replace $\partial X_k / \partial t$ we obtain the

Leibniz rule for volume averaging (Ishii, 1975)

$$\begin{aligned} \overline{X_k \frac{\partial \psi_k}{\partial t}}^V &= \frac{\partial}{\partial t} \overline{X_k \psi_k}^V - \overline{\psi_k \mathbf{v}_i \cdot \nabla X_k}^V \\ &= \frac{\partial}{\partial t} \overline{X_k \psi_k}^V + \frac{1}{V} \int_{S_i \cap V} \hat{\mathbf{n}}_k \cdot \mathbf{v}_i \psi_{ki}(\mathbf{x} + \boldsymbol{\eta}, t) dS \end{aligned} \quad (1.32)$$

1.3.2 Volume averaged equations

This section provides a summary of the derivation of volume-averaged equations for a two-fluid system. To simplify the analysis, the focus is restricted to isothermal, incompressible fluids, with constant densities ρ_1 and ρ_2 . The fluids are

assumed to flow with velocities $\mathbf{v}_1(\mathbf{x}, t)$ and $\mathbf{v}_2(\mathbf{x}, t)$. The local conservation equations for mass and momentum, applicable within $\Omega_k(t)$ are given by

$$\frac{\partial \rho_k}{\partial t} + \nabla \cdot \rho_k \mathbf{v}_k = 0 \quad (1.33)$$

and

$$\frac{\partial(\rho_k \mathbf{v}_k)}{\partial t} + \nabla \cdot \mathbf{v}_k \mathbf{v}_k = -\nabla p_k + \rho_k \mathbf{g} + \nabla \cdot \mathbb{T}_k \quad (1.34)$$

respectively, where $k \in 1, 2$ and the viscous stress tensor $\mathbb{T}_k$ is given by

$$\mathbb{T}_k = 2 \mu_k \mathbb{D}_k, \quad \text{where } \mathbb{D}_k \equiv \frac{1}{2} (\nabla \mathbf{v}_k + (\nabla \mathbf{v}_k)^T) \quad (1.35)$$

1.3.2.1 Volume averaged equations for individual phase

Multiplying each of the Eqs. (1.34) and (1.35) by the respective phase indicator function X_k and performing the average over V we obtain

$$\frac{\partial(\alpha_k \rho_k)}{\partial t} + \nabla \cdot \alpha_k \rho_k \overline{\mathbf{v}_k}^k = \Gamma_k \quad (1.36)$$

$$\frac{\partial(\alpha_k \rho_k \overline{\mathbf{v}_k}^k)}{\partial t} + \nabla \cdot \alpha_k \rho_k \overline{\mathbf{v}_k \mathbf{v}_k}^k = -\nabla(\alpha_k \overline{p}^k) + \alpha_k \rho_k \mathbf{g} + \nabla \cdot \alpha_k \overline{\mathbb{T}_k}^k + \mathbf{M}_k \quad (1.37)$$

The terms Γ_k and $\mathbf{M}_k$ are due to the Gauss and Leibnitz rules. The term Γ_k represents the transfer of mass across the interface due to phase change

$$\Gamma_k \equiv \overline{\rho_k (\mathbf{v}_{kl} - \mathbf{v}_l) \cdot \nabla X_k}^V \quad (1.38)$$

where, due to the conservation of mass, the terms Γ_1 and Γ_2 satisfy the jump condition

$$\Gamma_1 + \Gamma_2 = 0 \quad (1.39)$$

Since phase change is not considered in this context, the kinematic boundary condition (1.14) imposes the requirement that the velocity remains continuous at the interface. Thus, $\mathbf{v}_{1i} = \mathbf{v}_{2i} = \mathbf{v}_i$ and accordingly $\Gamma_1 = \Gamma_2 = 0$.

The term $\mathbf{M}_k$ represents the momentum transfer across the interface:

$$\mathbf{M}_k = -\overline{(-p_k \mathbb{I} + \mathbb{T}_k) \cdot \nabla X_k}^V \quad (1.40)$$

The sum of $\mathbf{M}_1$ and $\mathbf{M}_2$ satisfies the jump condition:

$$\mathbf{M}_1 + \mathbf{M}_2 = \frac{1}{V} \int_{S_i \cap V} (2 H \sigma \hat{\mathbf{n}}_1 + \nabla_s \sigma) dS \quad (1.41)$$

which follows from volume averaging of the dynamic boundary condition (1.16). In the following analysis, it is assumed that the surface tension coefficient, σ , remains constant, rendering the second term under the integral equal to zero. While Eqs (1.33) and (1.34) are applicable only within Ω_k , the field Eqs. (1.36) and (1.37) are valid across the entire domain Ω .

The motion of the interface or turbulence may induce velocity fluctuations within each fluid. To represent local spatial deviations from the volume-averaged mean value, a fluctuating velocity field is introduced for each phase.

$$\mathbf{v}'_k \equiv \mathbf{v}_k - \overline{\mathbf{v}}_k^k \quad (1.42)$$

Due to the non-linear convective term, this decomposition gives rise to a sub-grid stress (sgs) term in the volume averaged momentum equation (1.37), namely

$$\alpha_k \rho_k \overline{\mathbf{v}_k \mathbf{v}_k^k} = \alpha_k \rho_k \overline{\mathbf{v}_k^k} \overline{\mathbf{v}_k^k} - \alpha_k \mathbb{T}_k^{sgs} \quad (1.43)$$

where:

$$\begin{aligned} -\mathbb{T}_k^{sgs} &\equiv \rho_k \left(\overline{\mathbf{v}_k^k \mathbf{v}_k^k}^k - \overline{\mathbf{v}_k^k} \overline{\mathbf{v}_k^k} \right) + \rho_k \left(\overline{\mathbf{v}_k^k} \overline{\mathbf{v}'_k^k} + \overline{\mathbf{v}'_k^k} \overline{\mathbf{v}_k^k} \right) + \rho_k \overline{\mathbf{v}'_k \mathbf{v}'_k^k} \\ &\left\{ \begin{array}{l} \mathbb{L}_k \equiv \rho_k \left(\overline{\mathbf{v}_k^k \mathbf{v}_k^k}^k - \overline{\mathbf{v}_k^k} \overline{\mathbf{v}_k^k} \right) \\ \mathbb{C}_k = \rho_k \left(\overline{\mathbf{v}_k^k} \overline{\mathbf{v}'_k^k} + \overline{\mathbf{v}'_k^k} \overline{\mathbf{v}_k^k} \right) \\ \mathbb{R}_k = \rho_k \overline{\mathbf{v}'_k \mathbf{v}'_k^k} \end{array} \right. \quad (1.44) \end{aligned}$$

The tensors $\mathbb{L}_k$, $\mathbb{C}_k$, $\mathbb{R}_k$ represent the Leonard term, the cross term, and the subgrid-scale Reynolds stress term, respectively. It is noted that, in the case of time-averaging, the subgrid-scale stresses are replaced by the corresponding turbulent stress tensor.

In this section, a set of two scalar and two vector equations has been derived, representing the conservation of mass and momentum for the two phases. By applying the volume averaging operator to Eq. (1.24), a condition is obtained for the sum of the volume fractions.

$$\alpha_1 + \alpha_2 = 1 \quad (1.45)$$

The set of equations derived here is unclosed and thus requires the introduction of model assumptions.

1.3.2.2 Volume averaged equations for two-phase mixture

The equations presented in Section 1.3.2.1 describe the behavior of the individual phases. Equations that characterize the behavior of the two-phase mixture can be derived by summing the equations of the individual phases. By summing up Eq. (1.33) for $k = 1$ and $k = 2$ the result for the mass conservation of the two-phase mixture is obtained as follows:

$$\sum_{k=1}^2 \left[\frac{\partial(\alpha_k \rho_k)}{\partial t} + \nabla \cdot \alpha_k \rho_k \overline{\mathbf{v}_k}^k \right] = 0 \quad (1.46)$$

Defining a mixture density

$$\rho_m \equiv \alpha_1 \rho_1 + \alpha_2 \rho_2 \quad (1.47)$$

and a center-of-mass velocity

$$\mathbf{v}_m \equiv \frac{1}{\rho_m} \sum_{k=1}^2 \alpha_k \rho_k \overline{\mathbf{v}_k}^k = \frac{\alpha_1 \rho_1 \overline{\mathbf{v}_1}^1 + \alpha_2 \rho_2 \overline{\mathbf{v}_2}^2}{\alpha_1 \rho_1 + \alpha_2 \rho_2} \quad (1.48)$$

and introducing these definitions into Eq. (1.46) reduces it to the compact form

$$\frac{\partial \rho_m}{\partial t} + \nabla \cdot \rho_m \mathbf{v}_m = 0 \quad (1.49)$$

This equation takes the same form as that of single-phase flow; however, the single-phase density and velocity are replaced by the mixture density and the center-of-mass velocity, respectively. By summing the volume-averaged momentum Eq. (1.37) for both phases and applying the jump condition (1.41), the following form of the volume-averaged single-field momentum equation is obtained after further manipulations:

$$\begin{aligned} & \frac{\partial(\rho_m \mathbf{v}_m)}{\partial t} + \nabla \cdot (\rho_m \mathbf{v}_m \mathbf{v}_m + \mathbb{D}_i) \\ &= -\nabla \sum_{k=1}^2 \alpha_k \overline{p_k^k} + \rho_m \mathbf{g} + \frac{1}{V} \int_{S_i \cap V} 2 H \sigma \hat{\mathbf{n}}_1 dS \\ &+ \nabla \cdot \mu_m (\nabla \mathbf{v}_m + (\nabla \mathbf{v}_m)^T) + \nabla \cdot \mathbb{T}_i + \nabla \cdot (\alpha_1 \mathbb{T}_1^{sgs} + \alpha_2 \mathbb{T}_2^{sgs}) \end{aligned} \quad (1.50)$$

To clarify the various terms in Eq. (50), the concept of relative velocity is introduced first:

$$\mathbf{v}_r = \mathbf{v}_{21} \equiv \overline{\mathbf{v}_2}^2 - \overline{\mathbf{v}_1}^1 \quad (1.51)$$

which provides a measure of the mean velocity difference between the phases within the averaging volume V .

The term $\mathbb{D}_i$ is the momentum drift-flux tensor

$$\mathbb{D}_i \equiv \frac{\alpha_1 \rho_1 \alpha_2 \rho_2}{\rho_m} \mathbf{v}_r \mathbf{v}_r \quad (1.52)$$

It expresses the difference between the total resolved momentum flux and the flux of the averaged momentum due to the center-of-mass velocity. The definition of the interfacial friction tensor $\mathbb{T}_i$ which arises from viscous forces is

$$\mathbb{T}_i \equiv \rho_1 \alpha_2 \mu_2 \left(\nabla \frac{\alpha_1 \mathbf{v}_r}{\rho_m} + \left(\nabla \frac{\alpha_1 \mathbf{v}_r}{\rho_m} \right)^T \right) - \rho_2 \alpha_1 \mu_1 \left(\nabla \frac{\alpha_2 \mathbf{v}_r}{\rho_m} + \left(\nabla \frac{\alpha_2 \mathbf{v}_r}{\rho_m} \right)^T \right) \quad (1.53)$$

It is noted that the elements of the tensors $\mathbb{D}_i$ and $\mathbb{T}_i$ are nonzero only at or near the interface.

1.3.3 Closure problem

In the previous section, eight scalar partial differential equations were derived from the conservation of mass and momentum for the phases. Additionally, the algebraic constraint given in Eq. (1.45) applies to the volume fractions, along with the three momentum jump conditions (one for each coordinate direction). Assuming that the density and viscosity of each phase, as well as the coefficient of surface tension, are known, there are a total of 12 scalar equations for 28 scalar unknowns. These unknowns include, for each phase, the volume fraction, the three velocity components $\overline{v}_k^k$, the pressure $\overline{p}_k^k$, the three components of the momentum transfer term $\mathbf{M}_k$ and the six components of the subgrid-scale stress tensor $\mathbb{T}_k^{sgs}$.

or the remainder of this section, the subgrid-scale stresses will be neglected, i.e., it is assumed that:

$$\mathbb{T}_1^{sgs} = \mathbb{T}_2^{sgs} = \mathbb{T}_m^{sgs} = 0 \quad (1.54)$$

so that there are only 16 unknowns left. One assumption which is very common in the modelling of two-phase flow by the continuous field or interpenetrating field approach is that both phases share the same pressure field, i.e.

$$\overline{p}_1^{-1} = \overline{p}_1^{-2} = p_m \quad (1.55)$$

This assumption reduces the number of unknowns to 15, indicating that three additional equations are required to close the system of equations. The following section introduces the most commonly used approach for establishing these three closure relations.

1.3.4 Homogeneous model

1.3.4.1 Mathematical formulation

The homogeneous model is the most simple model within the continuous fields approach. It assumes that the phases move with the same velocity so that

$$\overline{\mathbf{v}}_2^{-2} = \overline{\mathbf{v}}_1^{-1} \quad (1.56)$$

The phases or fluids are in what is referred to as mechanical equilibrium. By definition (1.51), this implies that the relative velocity is zero in such cases.

$$\mathbf{v}_r = 0 \quad (1.57)$$

Eq. (1.56) respectively Eq. (1.57) provides the three additional scalar equations needed to close the system of equations. In the homogeneous model, therefore, only one velocity field needs to be computed. Because of Eq. (1.56) it is $\bar{\mathbf{v}}_1^1 = \bar{\mathbf{v}}_2^2 = \mathbf{v}_m$.

Thus, the velocity field is obtained by solving the single-field Navier-Stokes equation (1.50) for the center-of-mass velocity. Because the relative velocity $\mathbf{v}_r$ vanishes, so do the components of the tensors $\mathbb{D}_i$ and $\mathbb{T}_i$. With assumptions (1.54) and (1.55) the equations of the homogeneous model are then given by

$$\frac{\partial(\alpha_1 \rho_1)}{\partial t} + \nabla \cdot \alpha_1 \rho_1 \mathbf{v}_m = 0 \quad (1.58)$$

$$\frac{\partial(\alpha_2 \rho_2)}{\partial t} + \nabla \cdot \alpha_2 \rho_2 \mathbf{v}_m = 0 \quad (1.59)$$

$$\begin{aligned} \frac{\partial(\rho_m \mathbf{v}_m)}{\partial t} + \nabla \cdot \rho_m \mathbf{v}_m \mathbf{v}_m \\ = -\nabla p_m + \rho_m \mathbf{g} + \nabla \cdot \mu_m (\nabla \mathbf{v}_m + (\nabla \mathbf{v}_m)^T) \\ + \frac{1}{V} \int_{S_i \cap V} 2 H \sigma \hat{\mathbf{n}}_1 dS \end{aligned} \quad (1.60)$$

It is noted that, instead of using Eq. (1.58) or Eq. (1.59), the conservation equation for the two-phase mixture, Eq. (1.49), may be applied.

When the disperse fluid particle is fully inside the averaging volume V so that the interface does not cut the boundary ∂V of the averaging volume V , then the

integral over the surface tension term drops out from Eq. (1.60) and the interface-momentum-transfer terms all cancel out. In this case, the momentum equation (1.60) is essentially a single-phase transport equation, with variable density and viscosity.

From a computational perspective, the homogeneous model's assumption that the two phases share the same velocity field is reasonable under two notably distinct conditions. The first applies to finely dispersed flow, while the second pertains to flows in which the phases are well separated. It is noted that the distinction between dispersed and separated phases is primarily dependent on the length scale at which one intends to resolve the flow, i.e., the size of the averaging volume. This averaging length scale is typically related to the grid size selected for computation.

Originally, the homogeneous model was developed for finely dispersed flow, where particle size is significantly smaller than both the averaging volume and the mesh cell size. However, with the substantial increase in computational power, it is now possible to employ a high number of grid points and fine meshes, thereby enabling the application of the homogeneous model to separate flow as well.

1.3.5 Two-fluid model or Euler-Euler model

1.3.5.1 The standard two-fluid model

The mathematical basis of the two-fluid model is given by the volume-averaged mass conservation equation (1.36) and momentum conservation equation (1.37) for each phase, or by the time-averaged counterparts of these equations, respectively.

The momentum equations are coupled through the jump condition (1.41) for the transfer terms $\mathbf{M}_1$ and $\mathbf{M}_2$. In the two-fluid model the three scalar equations needed to close the set of equations are provided by a constitutive equation either for $\mathbf{M}_1$ or $\mathbf{M}_2$.

The definition of $\mathbf{M}_k$, given by Eq. (1.40), is revised below:

$$\mathbf{M}_k = -\frac{1}{V} \int_V (-p_k \mathbb{I} + 2\mu_k \mathbb{D}_k) \cdot \mathbf{X}_k dV \quad (1.61)$$

Recalling that the gradient of the phase indicator function is associated with the Dirac delta function of the interface, as referenced in Eq. (1.25), it can be expressed as follows:

$$\mathbf{M}_k = -\frac{1}{V} \int_{S_i \cap V} (-p_k \mathbb{I} + 2\mu_k \mathbb{D}_k) \cdot \hat{\mathbf{n}}_k dS \quad (1.62)$$

To close the two-fluid model, a constitutive equation is required for the integral of the instantaneous pressure and viscous stress distribution over the interface within the averaging volume, V . It is evident that equation (1.61) is closely related to the force, $\mathbf{F}_{surf}$, exerted by the surrounding fluid on a particle, as described by equation:

$$\mathbf{F}_{surf} = \oint_{\mathcal{A}_p} (-p_1 \mathbb{I} + 2\mu_1 \mathbb{D}_1) \cdot \hat{\mathbf{n}}_1 dS \quad (1.62bis)$$

An essential distinction is that the integral in Eq. (1.63) is closed, i.e., it encompasses the entire surface area, A_p , of the particle, whereas the integral in

equation (1.62) pertains only to the portion of the interfacial surface within the averaging volume, V .

The pressure of phase k can be divided into a mean interfacial pressure and a fluctuating pressure.

$$p_k \equiv \overline{p_k}^{S_i} + p'_k \quad (1.63)$$

The mean interfacial pressure is given by

$$\overline{p_k}^{S_i} = \frac{\int_{S_i \cap V} p_k dS}{\int_{S_i \cap V} dS} = \frac{1}{A_i} \int_{S_i \cap V} p_k dS \quad (1.64)$$

where A_i is the area of $S_i \cap V$. Introducing the interfacial area concentration

$$\begin{aligned} a_i &= \frac{A_i}{V} = \frac{1}{V} \int_{S_i \cap V} dS \\ &= \frac{1}{V} \int_V \delta(x - x_i) dV \\ &= \frac{1}{V} \int_V \hat{\mathbf{n}}_k \cdot \hat{\mathbf{n}}_k \delta(x - x_i) dV = \overline{\hat{\mathbf{n}}_k \cdot \nabla X_k}^V \end{aligned} \quad (1.65)$$

equation (1.64) can be written as

$$\overline{p_k}^{S_i} = \frac{1}{A_i} \int_{S_i \cap V} p_k dS = \frac{\overline{p_k \hat{\mathbf{n}}_k \cdot \nabla X_k}^V}{\overline{\hat{\mathbf{n}}_k \cdot \nabla X_k}^V} \quad (1.66)$$

Introducing the splitting of Eq. (1.64) into Eq. (1.40), it is obtained that:

$$\begin{aligned}
 M_k &= \overline{\overline{p}_k^{S_i} \mathbb{I} \cdot \nabla X_k}^V = \overline{(-p'_k \mathbb{I} + \mathbb{T}_k) \cdot \nabla X_k}^V = \overline{p_k^{S_i} \nabla X_k}^V + \mathbf{M}_{k,h} \\
 &= \overline{p_k^{S_i} \nabla \alpha_k} + \mathbf{M}_{k,h}
 \end{aligned} \tag{1.67}$$

The term $\overline{p_k^{S_i} \nabla \alpha_k}$ then contains the force contributions due to a hydrostatic pressure gradient and any external pressure gradient. It gives a net contribution only in case when there are any gradients of the volume fraction. On the other hand, the term $\mathbf{M}_{k,h}$ includes only hydrodynamic contributions and thus is non-zero only if there exists a relative motion between the phases.

Inserting Eqs. (1.67) and Eq. (1.43) in Eq. (1.37), taking advantage of the product rule

$$\nabla(\alpha_k \overline{p_k^k}) = \alpha_k \nabla \overline{p_k^k} + \overline{p_k^k} \nabla \alpha_k \tag{1.68}$$

and assuming that the interfacial-area averaged pressure is equal to the volume averaged pressure

$$\overline{p_k^{S_i}} \approx \overline{p_k^k} \tag{1.69}$$

we obtain the momentum equation for the two phases in the form

$$\begin{aligned}
 \frac{\partial(\alpha_k \rho_k \overline{\mathbf{v}_k^k})}{\partial t} + \nabla \cdot \alpha_k \rho_k \overline{\mathbf{v}_k^k \mathbf{v}_k^k} \\
 = -\alpha_k \nabla \overline{p_k^k} + \alpha_k \rho_k \mathbf{g} + \nabla \cdot \alpha_k \overline{\mathbb{T}_k^k} + \nabla \cdot \alpha_k \mathbb{T}_k^{sgs} + \mathbf{M}_{k,h}
 \end{aligned} \tag{1.70}$$

Introducing Eq. (1.67) in the momentum jump condition (1.41) and using assumption (1.69) the jump condition for the hydrodynamic momentum transfer terms becomes

$$\mathbf{M}_{1,h} + \mathbf{M}_{2,h} = \frac{1}{V} \int_{S_i \cap V} (2H\sigma \hat{\mathbf{n}}_1 - \nabla_s \sigma) dS \quad (1.71)$$

To close the two-fluid model, a constitutive equation for one of the two hydrodynamic momentum transfer terms is still required, which must be investigated on a case-by-case basis.

1.3.5.2 Closure relations for non-disperse two-phase flow

While disperse flows are of particular interest in chemical and nuclear engineering, as illustrated in Figures 1.1 and 1.2, there are numerous other flow regimes in which the phases flow separately. In the oil industry, for instance, flow regimes characteristic of pipelines, such as stratified flow, wavy flow, and slug flow, are of significant importance. It is evident that the closure assumptions for disperse flows, cannot be applied to these flow regimes. Simulations of slug flow in horizontal and nearly horizontal pipes were conducted by Issa and Kempf (Issa and Kempf, 2003) using a one-dimensional transient two-fluid model. For this purpose, a model was developed by the authors to account for the frictional forces between the phases at the interface.

1.3.6 Modern extensions of the two-fluid model

Using the conventional two-fluid model for disperse flows, only globally mono-disperse flows or flows with a globally constant number density can be computed. However, in actual disperse two-phase flows, a spectrum of bubble sizes is frequently present. When breakup or coalescence occurs, neither the bubble diameter nor the number density remains spatially uniform or constant over time. To address these limitations, the two-fluid model must be conceptually extended. The transport equation for interfacial area concentration, population balance equations, and the four-field two-fluid model are three different concepts currently under development that enable this type of study. In the following section, only the first concept, developed and applied in the case studies within this thesis, will be presented.

1.3.6.1 Transport equation for interfacial area concentration

The initial approach, as introduced in (Ishii and Mishima, 1981), is predicated on the intuition that, in a two-phase flow, not only the volumetric concentration of the phases is of importance but also the concentration of the interfacial area. The available interfacial area influences both the viscous and form drag and holds particular significance when heat or mass transfer processes occur at the interface. The concentration of the interfacial area can be defined through statistical averaging, time averaging, and volume averaging, as referenced in (Morel et al., 1999). In this study, the volumetric interfacial area concentration, denoted as a_i , is

considered, which represents the interfacial area per unit volume and, for a disperse flow, is given by:

$$a_i = \frac{1}{V} \sum_{j=1}^{N_p} \mathcal{A}_p^j \quad (1.72)$$

For a mono-disperse system, the above equation is simplified to $a_i = n_p A_p$. In this instance, the number density is associated with the interfacial area concentration, the void fraction, and the sphericity ψ (refer to Eq. (1.13)) through the following expression

$$n_p = \frac{\psi^3}{36 \pi} \frac{a_i^3}{a_2^2} \quad (1.73)$$

$$n_p = \frac{N_p}{V} = \frac{N_p V_p}{V} \frac{1}{V_p} = \frac{\alpha_2}{V_p} = \frac{6\alpha_2}{\pi d_{eq}^3} \quad (1.73bis)$$

Therefore, when the sphericity ψ is known or assumed and the interfacial area concentration is also known, Eq. (1.73) can be used in place of Eq. (1.73bis). It should be emphasized that the advantage of using Eq. (1.73) over Eq. (1.73bis) is realized only when the interfacial area concentration is a local quantity. This corresponds to a local equivalent diameter through the following relation.

$$d_{eq} = \frac{6}{\psi} \frac{\alpha_2}{a_i} \quad (1.74)$$

Thus, the assumption of a *global* mono-disperse flow in the standard two-fluid model is replaced by the assumption of a *local* mono-disperse flow.

Ishii (Ishii, 1975) proposed to solve a scalar transport equation to determine local values for a_i . With α_2 and a_i given, for a locally mono-disperse flow with spherical bubbles the local number density $n_p(\mathbf{x}, t)$ respectively the local equivalent diameter can be computed. Thus, solving the transport equation for the interfacial area concentration presents the advantage that the bubble diameter required for specifying the interfacial transfer terms is no longer a global parameter but rather a local quantity.

The interfacial area transport equation can be derived by considering the fluid particle number density equation, which is analogous to Boltzmann's transport equation (Kocamustafaogullari and Ishii, 1995). Its form is as follows:

$$\frac{\partial a_i}{\partial t} + \nabla \cdot (a_i \mathbf{v}_{a_i}) = \Phi_{a_i} \quad (1.75)$$

Here, $\mathbf{v}_{a_i}$ is the transport velocity of the volumetric interfacial area and Φ_{a_i} a source term taking into account the different phenomena creating or destroying interfacial area, such as coalescence or breakup of bubbles or droplets, phase change or interfacial stretching. Morel et al. (Morel et al., 1999) showed that Eq. (1.75) can be derived without assuming any particular geometrical configuration of the interfaces, so that Eq. (1.75) can be applied independently of the flow regime.

The major problem in development of the transport equation for the interfacial area concentration is that of formulating adequate models for the source/sink term Φ_{a_i} (Ishii et al., 2003). It has been demonstrated that the one-dimensional interfacial area transport equation can give a good prediction for bubbly

flows in vertical tubes with tube diameters in the range 25–50 mm (Hibiki and Ishii, 2002).

Even if the standard two-fluid model is extended by the inclusion of a transport equation for the interfacial area, a significant disadvantage remains: the sphericity of the bubble must still be specified, as indicated in Eq. (1.73). Consequently, while a transport equation for the interfacial area concentration allows for the modelling of changes in particle size, it does not account for changes in particle shape.

To address this limitation, the concept of two-group interfacial area transport has recently been proposed by Ishii et al. (Ishii et al., 2003) as a more comprehensive approach. In this framework, bubbles are classified into two distinct groups, and for each group, a separate transport equation for the interfacial area concentration is solved. The first group comprises spherical and distorted bubbles (e.g., ellipsoidal), whereas the second group includes cap-type and elongated bubbles, which are characteristic of slug flow.

It is anticipated that this approach will replace the conventional flow regime maps and regime transition criteria that do not dynamically represent changes in the interfacial structure. Instead, the two-phase flow structure changes are predicted mechanistically through the introduction of the interfacial area transport equation for the two groups.

1.4 Interface resolving methods

In this paragraph, the flow of two fluid phases, namely gas-liquid and liquid-liquid flow, is considered. For these flows, the diffusion model, the two-fluid model, and the Euler-Lagrange model all require a priori information regarding the flow regime. For instance, in the dispersed flow regime, the standard models necessitate prior knowledge of the shape of the bubbles or drops. The standard two-fluid model further requires information on the size of the bubble or drop.

Conversely, interface-resolving simulation or direct numerical simulation seeks to describe in detail the deformation and topological evolution of the phase interface separating the two fluids. Physically, the deformation of the phase interface is closely linked with the surface tension force. Therefore, any method used for direct numerical simulation of gas-liquid or liquid-liquid flow must rely on an accurate representation of the surface tension force.

The main difficulty of interface resolving simulations is the moving interface whose shape is part of the solution. In addition, physical quantities such as density, viscosity and pressure are discontinuous across the interface. Mathematically, the interface is treated as a surface of discontinuity. Loosely spoken it is “sharp”. To keep the interface numerically sharp, i.e. to avoid any artificial smearing of the interface during the computation, special numerical methods have been developed. Nowadays mainly three methods are used. The volume-of-fluid method (VOF) (Hirt and Nichols, 1981) is the oldest one while the level-set method (Sethian, 1999) and the front-tracking method (Tryggvason et al.,

2001) have been developed in the last decades. All these methods are formulated for incompressible fluids only and solve only one single momentum equation. In detail, the Volume of Fluid method will be presented in the following paragraph.

1.4.1 Volume-of-fluid method

The Volume of Fluid (VOF) model is a well-established computational approach for simulating the behavior of immiscible fluid phases within a defined computational domain. By solving a unified set of momentum equations, this model proficiently tracks the volume fraction of each phase throughout the domain, enabling an accurate depiction of fluid interfaces. The VOF method is particularly suited to complex multiphase flow scenarios such as jet breakup, the movement of large bubbles in liquid media, liquid flow following dam breaks, and both transient and steady-state behaviors of liquid-gas interfaces. These attributes render the VOF model a crucial tool for predicting interactions between immiscible fluids in a range of engineering and scientific contexts.

In the context of this thesis, the application of the VOF model is analyzed as implemented within the commercial software ANSYS® *Fluent*, which is employed for all presented simulations. The VOF model in ANSYS® *Fluent* presents several inherent restrictions that must be considered during computational analyses. The model is exclusively compatible with the pressure-based solver and cannot be used with the density-based solver. All computational cells must be occupied by either a

single fluid phase or a combination of phases, as void regions where no fluid is present are not permitted within the VOF framework. Although the model supports compressible ideal gases as one of the phases, the inclusion of compressible liquids is feasible only through the application of user-defined functions (ANSYS Inc., 2021).

1.4.1.1 Steady-State and Transient VOF Calculations

The Volume of Fluid (VOF) formulation is frequently utilized for time-dependent simulations of multiphase flows. Nevertheless, in scenarios where achieving a steady-state solution is the primary objective, a steady-state VOF calculation is also feasible. A steady-state approach is suitable only when the solution exhibits independence from initial conditions and distinct inflow boundaries are defined for each phase involved. For instance, in cases where the configuration of the free surface, such as within a rotating vessel, relies on the initial fluid level, the implementation of a time-dependent formulation becomes essential. Conversely, scenarios such as water flow in a channel with an overlying air layer and a separate air inlet can justify the use of a steady-state VOF calculation.

The VOF method is predicated on the assumption that the individual phases -each representing a different fluid- are immiscible. To represent each additional phase incorporated into the model, a variable known as the volume fraction is introduced, which indicates the proportion of that phase present within each computational cell. The summation of the volume fractions of all phases within any

given control volume must equal unity. The physical properties and solution variables within the model are shared across the phases and are volume-averaged, contingent upon the volume fractions being determined at every location within the computational domain. As such, the properties within a computational cell can correspond to a single phase or a mixture of phases, depending on the values of the volume fractions.

The volume fraction, denoted as α_q , defines the phase state within a computational cell in relation to the q^{th} phase, yielding three possible conditions:

$\alpha_q = 0$: The cell contains no fluid of phase q^{th} .

$\alpha_q = 1$: The cell is completely filled with fluid of phase q^{th} .

$0 < \alpha_q < 1$: The cell contains an interface between phase q^{th} and one or more other phases.

Based on the local volume fraction values, the appropriate fluid properties and variables are assigned to each control volume throughout the computational domain. This allows for accurate modeling of the interactions between different fluids and the prediction of complex multiphase flow phenomena.

1.4.1.2 Volume Fraction Equation

The tracking of phase interfaces is achieved by solving the continuity equation for the volume fraction of one or more phases present in the system. For

a given phase, the continuity equation governing the volume fraction takes the following form:

$$\frac{1}{\rho_q} \left[\frac{\partial}{\partial t} (\alpha_q \rho_q) + \nabla \cdot (\alpha_q \rho_q \vec{v}_q) \right] = S_{\alpha_q} + \sum_{p=1}^n (\dot{m}_{pq} - \dot{m}_{qp}) \quad (1.76)$$

where $\dot{m}_{qp}$ is the mass transfer from q phase to phase p and $\dot{m}_{pq}$ is the mass transfer from phase p to phase q . By default, the source term on the right-hand side of Eq. (1.76), S_{α_q} , is zero, but it can be specified as a constant or user-defined mass source for each phase. The volume fraction equation will not be solved for the primary phase; the primary-phase volume fraction will be computed based on the following constraint:

$$\sum_{q=1}^n (\alpha_q) = 1 \quad (1.77)$$

The volume fraction equation may be solved either through implicit or explicit time formulation.

1.4.1.3 The Implicit Formulation

When employing the implicit formulation, the discretization of the volume fraction equation is performed as follows:

$$\begin{aligned} \frac{\alpha_q^{n+1} \rho_q^{n+1} - \alpha_q^n \rho_q^n}{\Delta t} V + \sum_f (\rho_q^{n+1} Q_f^{n+1} \alpha_{q,f}^{n+1}) \\ = \left[S_{\alpha_q} + \sum_{p=1}^n (\dot{m}_{pq} - \dot{m}_{qp}) \right] V \end{aligned} \quad (1.78)$$

Where:

$n + 1$ represents the index for the current time step,

n represents the index for the previous time step,

α_q^{n+1} denotes the volume fraction within the cell at time step $n + 1$

α_q^n denotes the volume fraction within the cell at time step n ,

$\alpha_{q,f}^{n+1}$ denotes the volume fraction at the cell face at time step $n + 1$,

Q_f^{n+1} represents the volume flux through the cell face at time $n + 1$,

V is the volume of the cell.

Since the volume fraction at the current time step depends on other quantities evaluated at the same time step, a scalar transport equation is iteratively solved for each of the secondary-phase volume fractions at every time step. Face fluxes are usually interpolated based on the selected spatial discretization scheme. The implicit formulation is typically applicable for both time-dependent and steady-state calculations (ANSYS Inc., 2023).

1.4.1.4 The Explicit Formulation

The explicit formulation is time-dependent and the volume fraction is discretized in the following equation:

$$\frac{\alpha_q^{n+1} \rho_q^{n+1} - \alpha_q^n \rho_q^n}{\Delta t} V + \sum_f (\rho_q^n Q_f^n \alpha_{q,f}^n) = \left[S_{\alpha_q} + \sum_{p=1}^n (\dot{m}_{pq} - \dot{m}_{qp}) \right] V \quad (1.79)$$

where:

$n + 1$ = index for new (current) Time step,

n = index for previous time step,

$\alpha_{q,f}$ = face value of the q^{th} volume fraction,

V = volume of cell,

Q_f = volume flux through the face, based on normal velocity.

The volume fraction at the current time step is directly determined based on known values from the preceding time step. This explicit formulation eliminates the necessity for iterative solutions of the transport equation at each time step, thereby improving computational efficiency. Face fluxes can be interpolated using various interface tracking or capturing schemes, such as Geo-Reconstruct, CICSAM, Compressive, and Modified HRIC. These schemes are specifically developed to manage the motion of the interface between phases with precision.

The integration of the volume fraction equation involves an automatic refinement of the time step, with the possibility of adjustments through the modification of the Courant number. The volume fraction update can be performed either once per time step or iteratively within a time step, depending on the selected configuration. This adaptability facilitates control over the balance between

simulation accuracy and computational expense, providing a nuanced approach to managing the trade-offs inherent in computational multiphase flow simulations.

1.4.1.5 Interpolation near the Interface

The control-volume formulation necessitates the computation of convection and diffusion fluxes through the faces of the control volume, which must be balanced with the source terms contained within the control volume itself. In the geometric reconstruction and donor-acceptor schemes, ANSYS Fluent implements a specialized interpolation technique for cells located near the interface between two phases. Figure 1.6 illustrates the actual interface shape alongside the interfaces that are assumed during computations by these two methods.

Cells near phase interfaces are treated using consistent interpolation schemes, similar to those used for cells entirely occupied by one phase. This approach involves various methodologies such as the First-Order Upwind Scheme, Second-Order Upwind Scheme, QUICK Scheme, Modified HRIC Scheme, the Compressive Scheme, and Interface-Model-based Variants, as well as the CICSAM (Compressive Interface Capturing Scheme for Arbitrary Meshes). These schemes provide the necessary flexibility and precision for modeling complex multiphase interactions, ensuring stability and accuracy in simulations without the need for distinct treatments beyond these established schemes.

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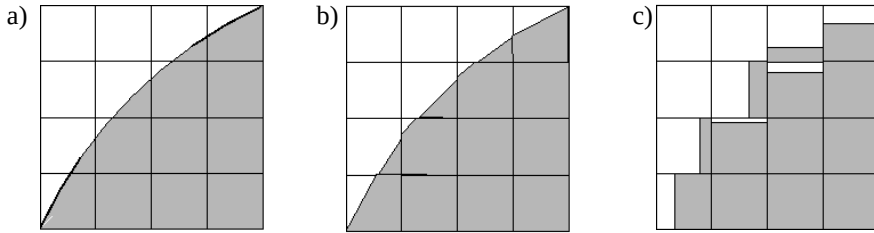


Figure 1.4: Comparison of interface calculation techniques. A) actual curved interface between two fluids. b) the interface reconstruction using the geometric Volume of Fluid method, where the interface is approximated with piecewise-linear segments. c) the donor-acceptor method, where the interface is represented in a stair-stepped manner, leading to less accurate results for curved interfaces.

1.4.1.6 The Geometric Reconstruction Scheme

In the geometric reconstruction approach, the standard interpolation schemes employed in ANSYS Fluent are utilized to determine face fluxes when a cell is completely filled with one phase or another. When a cell is located near the interface between two phases, the geometric reconstruction scheme is applied.

The interface between fluids is represented using a piecewise-linear approach in the geometric reconstruction scheme. In ANSYS Fluent, this scheme is recognized as the most accurate and is suitable for general unstructured meshes. The generalization of this scheme for unstructured meshes is based on the work of Youngs (Youngs, 1982). It is assumed that the interface between two fluids has a linear slope within each cell, and this linear configuration is used to calculate the advection of fluid through the cell faces.

The initial step in the reconstruction scheme involves calculating the position of the linear interface relative to the center of each partially-filled cell, using information about the volume fraction and its derivatives within the cell. The subsequent step involves determining the amount of fluid advected through each face, utilizing the computed linear interface representation and the distribution of normal and tangential velocities on the face. The final step involves calculating the volume fraction within each cell based on the flux balance calculated in the previous step.

1.4.1.7 The Compressive Scheme and Interface-Model-based Variants

The compressive scheme is characterized as a second-order reconstruction scheme based on the application of a slope limiter. Slope limiters are utilized in spatial discretization schemes to prevent spurious oscillations or wiggles, which would otherwise arise with high-order spatial discretization schemes due to abrupt variations within the solution domain. The theory presented below is applicable to zonal discretization and phase-localized discretization, both of which employ the framework of the compressive scheme:

$$\alpha_f = \alpha_d + \beta \nabla \alpha_d \cdot d\vec{r} \quad (1.80)$$

Where:

α_f = face VOF value

α_d = donor cell VOF value

β = slope limiter value

$\nabla \alpha_d$ = donor cell VOF gradient value

$d\vec{r}$ = cell to face distance

The slope limiter is constrained to values between 0 and 2 (inclusive). For values less than 1, the spatial discretization is represented by a low-resolution scheme. For values between 1 and 2, the spatial discretization is represented by a high-resolution scheme. The slope limiter values and their discretization schemes are shown in the table below.

Table 1.4: *Slope limiter values and their discretization schemes*

Slope Limiter Value β	Scheme
0	first order upwind
1	second order reconstruction bounded by the global minimum/maximum of the volume fraction
2	compressive
$0 < \beta < 1$ &	blended: where a value between 0 and 1 means blending of the first order and second order and a value between 1 and 2 means blending
$1 < \beta < 2$	of the second order and compressive scheme

The discretization of the compressive scheme is dependent on the selection of the interface regime type. When the sharp interface regime modeling is selected, the compressive scheme is suited solely for modeling sharp interfaces. However,

when the sharp/dispersed interface modeling is chosen, the compressive scheme becomes appropriate for modeling both sharp and dispersed interfaces.

1.4.1.8 Material Properties

The properties appearing in the transport equations are determined by the presence of the component phases in each control volume. In a two-phase system, for example, if the phases are represented by the subscripts 1 and 2, and if the volume fraction of the second of these is being tracked, the density in each cell is given by

$$\rho = \alpha_2 \rho_2 + (1 - \alpha_2) \rho_1 \quad (1.81)$$

In general, for an n -phase system, the volume-fraction-averaged density takes on the following form:

$$\rho = \sum \alpha_q \rho_q \quad (1.82)$$

All other properties are computed in this manner.

1.4.1.9 Momentum Equation

A single momentum equation is solved throughout the domain, and the resulting velocity field is shared among the phases. The momentum equation, shown below, is dependent on the volume fractions of all phases through the properties p and μ .

$$\frac{\partial}{\partial t}(\rho \vec{v}) + \nabla(\rho \vec{v} \vec{v}) = -\nabla p + \nabla \cdot [\mu(\nabla \vec{v} + \nabla \vec{v}^T)] + \rho \vec{g} + \vec{F} \quad (1.83)$$

One limitation of the shared-fields approximation is that in cases where large velocity differences exist between the phases, the accuracy of the velocities computed near the interface can be adversely affected.

1.4.1.10 Energy Equation

The energy equation, also shared among the phases, is shown below.

$$\begin{aligned} \frac{\partial}{\partial t}(\rho E) + \nabla \cdot (\vec{v}(\rho E + p)) \\ = \nabla \cdot \left(k_{eff} \nabla T - \sum_q \sum_j h_{j,q} \vec{j}_{j,q} + (\vec{\tau}_{eff} \cdot \vec{v}) \right) + S_h \end{aligned} \quad (1.84)$$

where k_{eff} is the effective conductivity ($k + k_t$, where k_t is the turbulent thermal conductivity, defined according to the turbulence model being used), $\vec{j}_j$ is the diffusion flux of species j , $h_{j,q}$ is enthalpy of species j in phase q and $\vec{j}_{j,q}$ is diffusive flux of species in phase. The first three terms on the right-hand side of Eq. (1.84) represent energy transfer due to conduction, species diffusion, and viscous dissipation, respectively. S_h includes volumetric heat sources that you have defined but not the heat sources generated by finite-rate volumetric or surface reactions since species formation enthalpy is already included in the total enthalpy calculation.

The VOF model treats energy, E , as a mass-averaged variable

$$E = \frac{\sum_{q=1}^n \alpha_q \rho_q E_q}{\sum_{q=1}^n \alpha_q \rho_q} \quad (1.85)$$

In Eq. (1.84):

$$E_q = h_q - \frac{p}{\rho_q} + \frac{v^2}{2} \quad (1.86)$$

where h_q for each phase is based on the specific heat of that phase and the shared temperature. The properties ρ, k_{eff} (effective thermal conductivity) and μ_{eff} (effective viscosity) are calculated by volumetric averaging over the phases. The source term, S_h , contains contributions from radiation, as well as any other volumetric heat sources.

As with the velocity field, the accuracy of the temperature near the interface is limited in cases where large temperature differences exist between the phases. Such problems also arise in cases where the properties vary by several orders of magnitude.

1.4.1.11 Surface Tension and Adhesion

In the commercial software ANSYS Fluent, it is possible to include the effects of surface tension along the interface between a pair of phases in your simulation. It is possible to specify a surface tension coefficient as a constant, as a function of temperature through a polynomial, or as a function of any variable through a

User-Defined Function (UDF). The solver will include the additional tangential stress terms (causing what is termed as Marangoni convection) that arise due to the variation in surface tension coefficient. Variable surface tension coefficient effects are usually important only in zero/near-zero gravity conditions.

Wall adhesion effects can be included by the additional specification of the contact angles between the phases and the walls, as well as at porous jumps.

1.4.1.11 Surface Tension

Surface tension arises from the attractive forces between molecules within a fluid. For instance, in an air bubble immersed in water, the net force on a molecule due to its neighbouring molecules within the bubble is zero. However, at the surface, the net force is directed radially inward. The combined effect of these radial force components across the entire spherical surface causes the surface to contract, which increases the pressure on the concave side of the surface. Surface tension, which acts solely at the surface, is the force required to maintain equilibrium under such conditions. It functions to balance the radially inward intermolecular attractive forces with the radially outward pressure gradient force across the surface. When two fluids are separated but not in the form of spherical bubbles, surface tension works to minimize the free energy by reducing the interfacial area.

In ANSYS® Fluent, two surface tension models are available: the *Continuum Surface Force* (CSF) model and the *Continuum Surface Stress* (CSS)

model. Detailed descriptions of these models are provided in the subsequent sections.

1.4.1.12 The Continuum Surface Force Model

The *Continuum Surface Force* (CSF) model proposed by Brackbill et al. interprets surface tension as a continuous, three-dimensional effect across an interface, rather than as a boundary value condition on the interface. Surface tension effects are modeled by adding a source term in the momentum equation. To understand the origin of the source term, consider the special case where the surface tension is constant along the surface, and where only the forces normal to the interface are considered. It can be shown that the pressure drop across the surface depends upon the surface tension coefficient, σ , and the surface curvature as measured by two radii in orthogonal directions, R_1 and R_2 :

$$p_2 - p_1 = \sigma \left(\frac{1}{R_1} + \frac{1}{R_2} \right) \quad (1.86)$$

where p_1 and p_2 are the pressures in the two fluids on either side of the interface. The surface curvature is computed from local gradients in the surface normal at the interface. Let n be the surface normal, defined as the gradient of α_q , the volume fraction of the q^{th} phase.

$$n = \nabla \alpha_q \quad (1.87)$$

The curvature, κ , is defined in terms of the divergence of the unit normal, $\hat{n}$:

$$\kappa = \nabla \cdot \hat{n} \quad \text{with} \quad \hat{n} = \frac{\mathbf{n}}{|\mathbf{n}|} \quad (1.88)$$

The surface tension can be written in terms of the pressure jump across the surface. The force at the surface can be expressed as a volume force using the divergence theorem. It is this volume force that is the source term that is added to the momentum equation. It has the following form:

$$F_{vol} = \sum_{\text{pairs } i, j, i < j} \sigma_{ij} \frac{\alpha_i \rho_i \kappa_i \nabla \alpha_j + \alpha_j \rho_j \kappa_j \nabla \alpha_i}{\frac{1}{2}(\rho_i + \rho_j)} \quad (1.89)$$

This expression allows for a smooth superposition of forces near cells where more than two phases are present. If only two phases are present in a cell, then $\kappa_i = -\kappa_j$ and $\nabla \alpha_i = -\nabla \alpha_j$, and Eq. (1.89) simplifies to

$$F_{vol} = \sigma_{ij} \frac{\rho \kappa_i \nabla \alpha_i}{\frac{1}{2}(\rho_i + \rho_j)} \quad (1.90)$$

where ρ is the volume-averaged density computed using Eq. (1.82). Eq. (1.90) shows that the surface tension source term for a cell is proportional to the average density in the cell.

1.4.1.13 The Continuum Surface Stress Model

The *Continuum Surface Stress* (CSS) method is an alternative way to modeling surface tension in a conservative manner, unlike the non-conservative

formulation of the *Continuum Surface Force* (CSF) method. CSS avoids the explicit calculation of curvature and could be represented as an anisotropic variant of modeling capillary forces based on surface stresses.

In the CSS method, the surface stress tensor due to surface tension is represented as

$$T = \sigma(I - \hat{n} \otimes \hat{n}) |\hat{n}| \quad (1.90)$$

$$\vec{n} = \nabla \alpha \quad (1.91)$$

$$\hat{n} = \frac{\vec{n}}{|\vec{n}|} \quad (1.92)$$

where

I = unit tensor

σ = surface tension coefficient

$\otimes$ = tensor product of the two vectors: the original normal and the transformed normal

α = volume fraction

$\vec{n}$ = volume fraction gradient

$$T = \sigma \left(|\nabla \alpha| I - \frac{\nabla \alpha \otimes \nabla \alpha}{|\nabla \alpha|} \right) \quad (1.93)$$

The surface tension force is represented as

$$F_{CSS} = \nabla \cdot T \quad (1.94)$$

1.4.1.14 The importance of surface tension

The importance of surface tension effects is determined based on the value of two dimensionless quantities: the Reynolds number, Re , and the Capillary number, Ca ; or the Reynolds number, Re , and the Weber number, We , presented in paragraph 1.2.4. For $Re \ll 1$, the quantity of interest is the capillary number and for $Re \gg 1$ the quantity of interest is the Weber number. Surface tension effects can be neglected if $Ca \gg 1$ or $We \gg 1$.

1.4.1.15 Wall Adhesion

The effects of wall adhesion at fluid interfaces in contact with rigid boundaries in equilibrium can be readily estimated within the framework of the CSF model. Instead of imposing this boundary condition directly at the wall, the contact angle that the fluid is assumed to form with the wall is employed to adjust the surface normal in cells adjacent to the wall. This dynamic boundary condition leads to the modification of the surface curvature near the wall. If θ_w is the contact angle at the wall, then the surface normal at the live cell next to the wall is

$$\hat{n} = \hat{n}_w \cos \theta_w + \hat{t}_w \sin \theta_w \quad (1.95)$$

where $\hat{n}_w$ and $\hat{t}_w$ are the unit vectors normal and tangential to the wall, respectively. The combination of this contact angle with the normally calculated surface normal one cell away from the wall determines the local curvature of the surface, and this curvature is used to adjust the body force term in the surface tension calculation.

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CHAPTER 2: Experimental and computational study of SFE **in structured packing**

Part of the content in this section was previously published in this publication:

L. Schembri, G. Caputo, M. Ciofalo, F. Grisafi, S. Lima, and F. Scargiali,
“*Experimental and Computational Study of Supercritical Fluid Extraction (SFE) of Omega-3 Components from Fish Oil in Structured Packing*,”
Chem. Eng. Trans., vol. 100, pp. 337–342, 2023,
<https://doi.org/10.3303/CET23100057>

2.1 Summary

The benefits of polyunsaturated fatty acids and their implications for human health have gained scientific attention to their extraction from biological sources, not being produced by the human body. Most known industrial productions of omega-3 fatty acids often work under operating conditions that may degrade these components and they often use toxic or flammable solvents that can adversely affect human health. In this sense, innovative and interesting prospects are provided by Supercritical Fluid Extraction (SFE).

In this chapter, two parallel studies were carried out: an experimental activity in a laboratory apparatus using supercritical carbon dioxide (sc-CO₂) and preliminary computational fluid dynamics (CFD) simulations, limited to the hydrodynamic aspects of the process. In the experimental apparatus a Sulzer® EX structured packing, made up of corrugated metal gauze sheets, was used as the column filler. The study made it possible to identify the optimal operating conditions

leading to an enrichment of the starting mixture in Eicosapentaenoic acid (EPA) and Docosahexaenoic acid (DHA), target products. CFD simulations were based on the Volume of Fluid (VOF) approach, suitable to the present complex multiphase system with two phases in close contact (transesterified fish oil and sc-CO₂). The meatus created by the corrugations of the metal gauze was chosen as the calculation domain representative of the system. The computations were performed by the commercial software ANSYS® Fluent, which allowed the prediction of the hydrodynamic evolution of the system through transient simulations. CFD predictions were in qualitative agreement with the experimental results.

2.2 General Overview

The growing attention to the health and psycho-physical well-being of humans and the optimistic evaluation of the benefits associated with the intake of omega-3 caused a global increase in the demand for products containing Poly-Unsaturated Fatty Acids (PUFA). For its market, estimated in 2021 equals to 2.19 billion dollars, a forecast of increase, for the period 2022-2030, is expected with a Compound Annual Growth Rate (CAGR) of 8.0% (Polaris, 2022). In previous years awareness policies were implemented by the various governments like USA and Europe to influence the trend of the demand of these compounds. The interest in PUFAs arises from the variety of biochemical processes in which they are involved. Numerous studies have reported physiological effects of PUFA, in fact omega-3 may influence several pathological conditions associated with metabolic, inflammatory

and oxidative processes. PUFAs may regulate the antioxidant signaling pathway and modulate inflammatory processes. They also influence hepatic lipid metabolism and physiological responses of other organs, including the heart (Djuricic and Calder, 2021). The Omega-3 product industry is driven by the growing demand in the Active Pharmaceutical Ingredient (API) market, linked to the growing awareness of the effective action of these products, in particular on chronic and cardiovascular diseases.

Nowadays one of the problems of extraction and purification is linked to the fact that known industrial processes work under operating conditions that inevitably induce the degradation of these components and the use of toxic or flammable solvents that can have adverse effects on human health, as well as for the environment. In this sense, liquid-solvent extraction, that makes use of CO₂ in supercritical conditions as a solvent, offers innovative, attractive and more environmentally friendly chances. Supercritical conditions are used throughout the process to increase the CO₂'s solvating power. The Omega-3 industry is commonly divided into two categories, based on the origin of the raw material (marine sources and plant sources) (Lima et al., 2022b, 2020). One of the possible starting matrices is an oil obtained from waste of the fishing industry such as tuna, sardines, or shark.

The problem with this matrix is that more than 40 different fatty acids are present in these oils, which makes them a much more complex and articulated mixture (Wen et al., 2023). In fact, that mixtures consist of linear chain fatty acids having 14 to 22 carbon atoms, with several unsaturations typically ranging from 0

up to 6. Another problem is the lack in the literature of studies and models of the Supercritical Fluid Extraction (SFE) phenomenon that would allow a more optimal separation without the need to work with an energy-intensive plant at high pressures. This work takes part on the aforementioned aspects by proposing a fluid dynamics computational study to optimize the enrichment of the matrix in compounds with high added value.

2.3 Experimental methods and materials

There are several methods traditionally used for Omega-3 concentration and purification such as urea adduction, chromatographic methods, fractional crystallization at low temperatures, distillation, enzymatic and integrated methods and SFE (Caputo et al., 2016). The last one, employing a continuous operating equipment, is based on the pioneering experiments carried out by Brunner and co-workers (Riha and Brunner, 2000). In the last twenty years, there have been many improvements in this process, which has increasingly taken hold in the world of industry since it guarantees a product with high purity and thermally undegraded (Abou Elmaaty et al., 2022). Figure 2.1 shows the process flow diagram of the pilot plant where the present experimental activities were conducted. The plant is equipped with a 2.40 m active height extraction column (T-1). The column has an inner diameter of 21.2 mm and is filled with a Sulzer® EX packing layer. Liquid CO₂ at 40 bar, stored in vessel S-2, is compressed via pump P-2 to the supercritical state at 145 bar (extraction pressure). The sc-CO₂ is heated using a hairpin heat exchanger

(H-3) to the desired temperature of 60 °C. Afterwards, the CO₂ enters the extraction column from the bottom and leaves it from the top, where it is expanded to 40 bar by a manual backpressure regulator valve entering the separation vessel S-1. Here the extracted compound can be removed by a needle valve from the bottom of the vessel. The gaseous CO₂ flows through a plate condenser (C-1) back into the CO₂ vessel (S-2). The single-phase flow rate of the sc-CO₂ was measured by a flowmeter Bronkhorst®, mini CORI-FLOW® Series, model M14 with a nominal range between 0.1 e 30 kg h⁻¹ and a mass flow accuracy for liquids of ±0,2%. The oil phase is fed into the central region of the column via pump P-1 and, after being processed, is taken from the system as a fraction enriched in the heavier components in the bottom of the column and as a light fraction from the bottom of the S-1 tank. A part of this is fed back into the column as a reflux current using pump P-3.

The entire process is presented in detail:

The transesterified fish oil (stream 1) is compressed through pump P-1 and subsequently heated by exchanger H-1 to reach operating conditions of 60°C and 145 bar before entering the column. Inside the column, the oil encounters an upward flow of supercritical CO₂, which, as previously mentioned, serves to extract the lighter components, carrying them towards the column head. The heavier components, which are less soluble in the solvent, reach the bottom of the column and exit as raffinate in stream 8.

Stream 2, composed of the fraction of extracts mixed with the solvent, exits the top of the column and, upon passing through the back-pressure valve, expands

from 145 bar to 40 bar. This depressurization is necessary to separate the oil from the CO₂, which is no longer in a supercritical state but now gaseous. Since this expansion is adiabatic, the fluid undergoes rapid cooling due to the Joule-Thomson effect, reducing its temperature from 60°C to around -6°C. Therefore, to avoid freezing of the mixture, the stream passes through a heater immediately downstream of valve BPV-1, raising the temperature to 35°C. This value was selected as a suitable compromise between separation efficiency and subsequent cooling of the CO₂ exiting the separator, as this must be temporarily condensed before being sent back into the column.

Stream 3, downstream of the heater, is not a single phase, as the pressure and temperature are below the solvent's critical values. Therefore, it is a biphasic mixture with a gas phase primarily composed of CO₂ and a liquid phase containing the extracted esters. Exploiting the density difference between the two phases, the expanded stream is sent to the gravimetric separator S-1, where a gas stream exits the top and a liquid stream exits the bottom. Part of the liquid phase is withdrawn, while another part is refluxed back into the column (stream 11) after being recompressed and reheated to the desired conditions through pump P-3 and heater H-4.

The gaseous solvent exiting the separator must be reconditioned before being recycled into the column. Specifically, stream 4, consisting of CO₂ in the gas phase, is directed to the condenser C-1, where it is condensed and subcooled to -5°C to prevent cavitation in the pump (the equilibrium temperature at 40 bar is 5.5°C).

The utility fluid that removes enthalpy from the solvent is provided by an external system using a mixture of water and ethylene glycol. The condensed CO₂ is then sent to buffer tank S-2, whose primary function is to monitor the solvent hold-up throughout the plant by tracking the liquid level within. To maintain a constant temperature inside the buffer tank, it is equipped with a cooling jacket fed by the same cold utility. Subsequently, stream 5, consisting of the solvent in the liquid phase, is fed to pump P-2 to restore it to the column pressure.

The last step for the carbon dioxide before being recycled to the column is heating in exchanger H-3, which raises the fluid to the pre-set column temperature. The reconditioned stream 7 is then sent directly to the extraction section, where it serves as a solvent for the extraction process.

As shown in the PFD in Figure 2.1, there are lines in two different colors: the red lines indicate high-pressure streams, while the black lines indicate medium-low pressure streams. The solvent undergoes a closed cycle, initially introduced into the system through stream 12, and once the desired hold-up is reached, additional CO₂ is only added to compensate for losses.

2.3.1 Determination of physical properties

Simple equations of state are not able to accurately estimate the fluids' properties over the large interval of pressure and temperature of the present study and multi-parameter equations of state are required. In regard to CO₂ density, among several models present in the literature (Bisotti et al., 2019), the most suitable for our case was found to be the Bender equation of state (Brunner, 2013):

$$p = RT\rho_f + B\rho_f^2 + C\rho_f^3 + D\rho_f^4 + E\rho_f^5 + F\rho_f^6 + (G + H\rho_f^2)\rho_f^3 \exp(-a_{20}\rho_f^2) \quad (2.1)$$

This equation calculates the pressure of a generic fluid as a function of density using the seven constants B, C, D, E, F, G, H, which in their turn are functions of temperature via 20 parameters, to be fitted on property data and must be inverted to provide the density as a function of pressure and temperature. The Bender equation of state is considered a good compromise between simple and more elaborate equations and it is highly reliable for homogeneous fluids in the supercritical state, particularly for well-known compounds as CO₂ with a low acentric factor (Ghazouani et al., 2005).

For the evaluation of the sc-CO₂ viscosity, the following formula was used (Heidaryan et al., 2011):

$$\mu_v = \frac{(A_1 + A_2\rho_f + A_3\rho_f^2 + A_4(\ln(T))^2 + A_6(\ln(T))^3)}{1 + A_7\rho_f + A_8 \ln(T) + A_9(\ln(T))^2} \quad (2.2)$$

To evaluate the interfacial tension of marine fatty acid ethyl esters (FAEE), which constitute the fish oil used, in contact with sc-CO₂, the model proposed by Seifried (Seifried and Temelli, 2010) was employed, adapted with a proper corrective factor for the present numeric simulation.

$$\gamma(P, T, \rho_{CO_2}) = \gamma_0(T) - k_1(T)(p - 0.1) - k_2(T) * (1 - \exp(k_3(T)(\rho_{CO_2} - \rho_{CO_2}^0))) \quad (2.3)$$

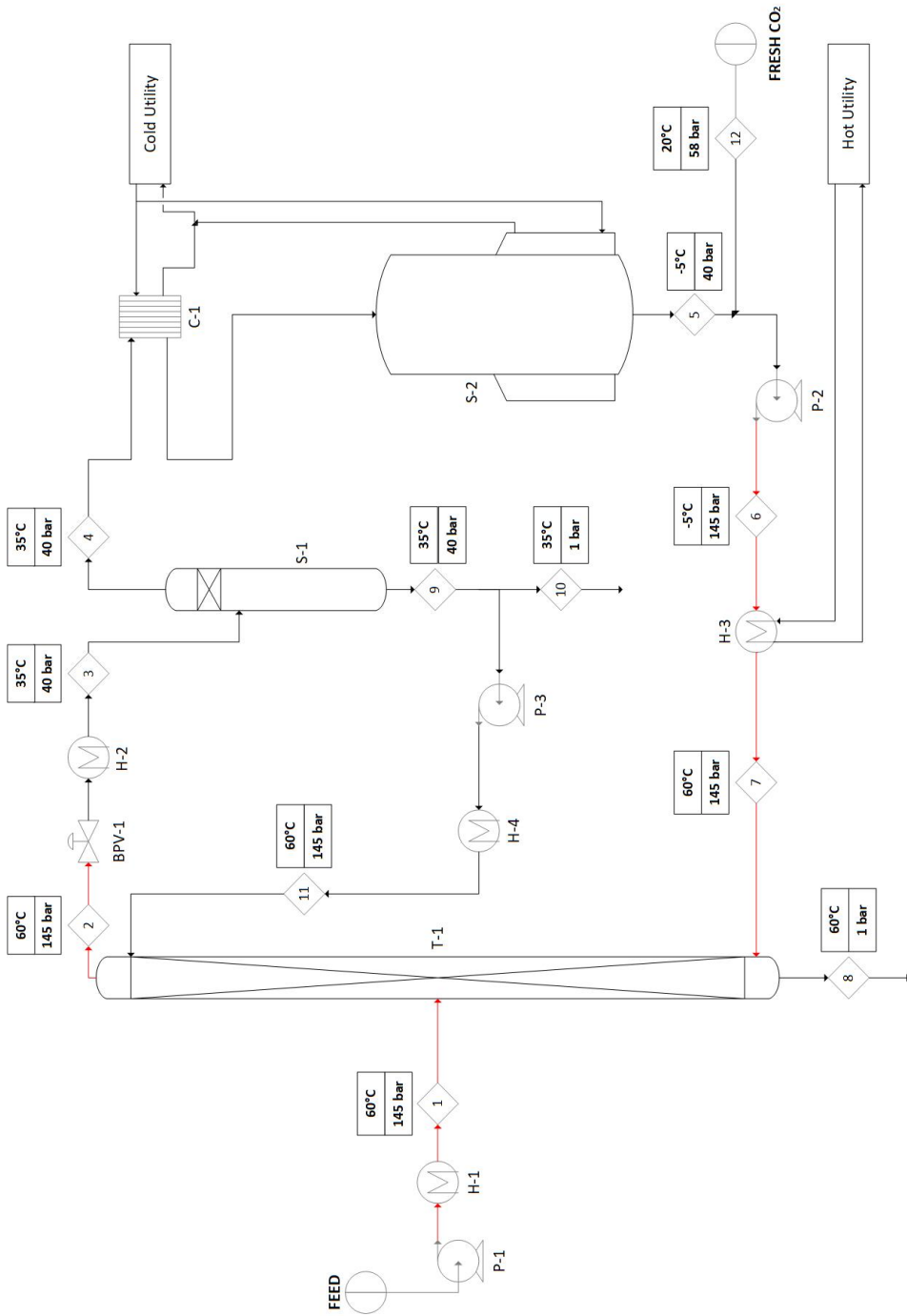


Figure 2.1: Schematic diagram of the experimental setup and operating conditions for every streamline

As the contact angle, the value estimated by Fernandes (Fernandes et al., 2009). The thermodynamic properties of transesterified fish oil (LISY- Ethyl Esters 33/22 HE002) were experimentally measured under the present operating conditions. For the evaluation of density, a previously calibrated graduated cylinder was filled with the substance and weighted by an analytical balance Kern® ABS-ABJ.

The viscosity of fish oil was measured employing an Ubbelohde suspended-level capillary viscometer 532 11 Ia-Schott Mainz-JenaERGlas (diameter of 0.95 mm) provided with a ViscoClock® instrumentation and immersed in a thermo-static bath Lauda®015T. All data are given in Table 2.1.

Table 2.1: *Physical properties of sc-CO₂/fish oil for the selected operating conditions ($P = 145$ bar and $T = 60$ °C).*

	Sc-CO ₂	Transesterified Fish Oil
Density [kg m⁻³]	582.61	884
Viscosity [Pa s]	$4.36 \cdot 10^{-5}$	$2.72 \cdot 10^{-3}$
Surface Tension [N m⁻¹]		$5.68 \cdot 10^{-4}$
Contact Angle [deg]		30

2.4 Numerical simulation

The VOF formulation, as described in paragraph 1.4.1, models two or more immiscible phases by solving the momentum equations of each of the fluids by using the commercial CFD code ANSYS® *Fluent* V.23 R2 (ANSYS Inc., 2023). In this study, the VOF model solved the transport equations for two Eulerian phases, sc-

CO₂ and transesterified fish oil. The volume fraction is solved by the explicit method, which exhibits a better numerical accuracy compared to the implicit formulation. This assessment has been evaluated through personal experiences and tests on the same system. The maximum Courant number allowed near the free surface is set to 0.25. To model the interface between the two phases, the sharp interface model is used, since a distinct interface is present between the supercritical and the liquid phases. The volume fraction cutoff is set to 10⁻⁶. In vector formulation, the momentum (Navier-Stokes) equations solved are:

$$\frac{\partial}{\partial t}(\rho \vec{v}) + \nabla \cdot (\rho \vec{v} \vec{v}) = -\nabla \cdot p + \nabla \cdot [\mu (\nabla \vec{v} + \nabla \vec{v}^T)] + \rho \vec{g} + \vec{F} \quad (2.4)$$

In Eq (2.4), ρ and $\vec{v}$ define the local density and the velocity vector, μ is the viscosity of the fluid, $\vec{g}$ is the gravitational acceleration vector. p defines the pressure and $\vec{F}$ is the source term. $\vec{F}$ also contains model-dependent source terms. The local density is calculated by:

$$\rho = \alpha_{sc-CO_2} \rho_{sc-CO_2} + \alpha_{oil} \rho_{oil} \quad (2.5)$$

where ρ_{sc-CO_2} specifies the density of phase sc-CO₂ and α_{sc-CO_2} represents the local volume fraction of sc-CO₂ phase. The volume fraction must be solved only for one of the phases, since for the other phase it is determined from the constraint:

$$\sum_{i=1}^2 \alpha_i = 1 \quad (2.6)$$

The tracking of the interface between the phases is performed by solving the continuity equation for the volume fraction of one of the phases:

$$\begin{aligned} \frac{\partial}{\partial t} (\alpha_{sc-CO_2} \rho_{sc-CO_2}) + \nabla \cdot (\alpha_{sc-CO_2} \rho_{sc-CO_2} \vec{u}_{sc-CO_2}) \\ = S_{\alpha_{sc-CO_2}} + (\dot{m}_{oil/sc-CO_2} - \dot{m}_{sc-CO_2/oil}) \end{aligned} \quad (2.7)$$

$S_{\alpha_{sc-CO_2}}$ is a mass source or mass sink; $\dot{m}_{psc-CO_2}$ and $\dot{m}_{sc-CO_2p}$ are the mass transfer rates from sc-CO₂ phase to oil phase and vice versa. The continuum surface force (CSF) model proposed by Brackbill (Brackbill et al., 1992), presented in chapter 1, is implemented such that the addition of surface tension to the VOF calculation results in a source term $\vec{F}$ in the momentum equation. For each simulation carried out, the fluids are assumed to be Newtonian, isothermal and incompressible; hence the properties are kept constant. The values for the thermophysical properties implemented in computational simulations, including the values for the surface tension and contact angle are reported in Table 2.2.

2.4.1 Packing geometry and computational domain

Sulzer® EX structured gauze packing and its geometric characteristics are shown in Figure 2.2 and Table 2.2. The characteristic length of the packing is defined by the diameter of its channels, $d_p = 6(1 - \varepsilon)/a_s$, where ε is the void fraction and a_s is the surface area per unit volume of packing. The hydraulic diameter, d_h , is defined as $d_h = 4\varepsilon/a_s$. The calculations were carried out in the computational domain shown in Figure 2.2.b. Its geometry corresponds to the space between two neighboring packing sheets of a packing element.

Table 2.2: *Geometric data of the Sulzer® EX packing.*

Packing - Sulzer® EX	
Column internal diameter [mm]	21.2
Material	SS. gauze
Fractional void volume, ε	0.86
Specific surface area, a_s [$\text{m}^2 \text{m}^{-3}$]	1710
Inclination angle, θ [degrees]	45
Number of corrugated gauzes per single element	11
Height per single element [mm]	54.6
Total height of tower [mm]	2400

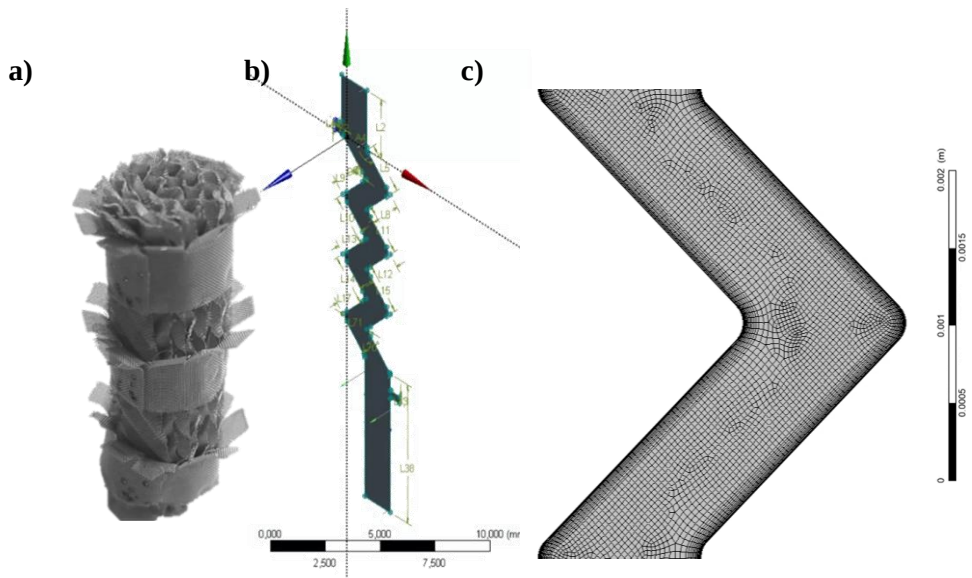


Figure 2.2: a) *Photography of the Sulzer® EX structured gauze packing;* b) *Computational domain for the fluid dynamics calculations;* c) *Zoomed detail of the computational mesh.*

It develops mainly in two dimensions; three volumes were placed along the third direction, so that to make the system three-dimensional in order to be able to use the same model in more complex applications.

For simplicity reasons, along the longitudinal direction the computational domain includes only three and a half repetitive crisscross units for a total length of ~10 mm (not including upper and lower headers), while the real equipment includes more than 800 such units and is ~2.4 m high. The grid was refined toward the walls so that at least 12–15 grid points were placed in the liquid film. Structured hexahedral meshes with 60,000, 120,000, 160,000 and 480,000 total volumes (one third of which in the main simulation plane) were tested. Preliminary results showed that the mesh with 160,000 volumes gave the best compromise between the mesh quality (in terms of aspect ratio, orthogonality and uniformity), and the computational burden required by the simulation. The criterion used to choose the mesh was based on the observation of the shape of the liquid film on the solid wall of the packing. While the first two coarser meshes didn't result in the formation of a uniform film on the solid surface, the last two gave results almost identical to each other and in agreement with the expected continuous film behavior, so the 160,000 volumes mesh was chosen. More details are shown in Appendix A.

The walls were defined as no-slip boundary conditions. Both the liquid and sc-CO₂ inlet zones were defined as uniform inlet velocity (Dirichlet) boundary conditions. The outlets for sc-CO₂ and liquid were defined as pressure-outlet boundaries with a gauge pressure of 0 Pa.

2.4.2 Computation details

For the time-dependent CFD a time step of $1 \cdot 10^{-4}$ s was chosen with 30 iterations per time step. The initialization was made with the entire domain filled with sc-CO₂ and as a viscous model, taking into account the low Reynolds number present in the system for both phases, a laminar model was chosen. A phase-coupled PISO algorithm was used to solve the pressure-velocity coupling. The volume fraction was discretized with a compressive scheme, while pressure was discretized with a PRESTO! scheme. The momentum equations were discretized with second-order upwind schemes and the Least Squares Cell Based gradient method was chosen. The simulations were usually protracted up to a real time of about 120 s.

2.5 Results and discussion

2.5.1 Experimental Results

The experimental activity was carried out in order to investigate the optimal conditions of supercritical extraction. Attention was paid to the flow rate of sc-CO₂ to be sent in the column that allows the best extraction in reference to the two compounds of interest with high added value: Eicosapentaenoic acid (EPA) and Docosahexaenoic acid (DHA). The best results were obtained with an extraction pressure of 145 bar and a temperature of 60 °C. The transesterified fish oil flow rate was set to 0.90 kg h⁻¹ and different solvent flow rate values were investigated. Using a flow rate of 30.00 kg h⁻¹ as reported in the literature (Riha and Brunner, 2000), the experiments showed a total lack of product in the refined section and the consequent

malfunction of the entire process: the feed was not properly separated into light and heavy compounds as required, but was dragged as a whole into the separation unit downstream of the extraction column.

Therefore, the solvent flow rate was gradually reduced; the maximum separation, as verified by chromatographic analysis, was obtained for a value of 9.00 kg h^{-1} . In the corresponding experimental trial, the product was present in the bottom-column and correctly enriched as designed. To investigate what happened inside the extraction column with the initial flow rate and what changed with the reduced flow rate, a computational fluid dynamics study of the multiphase system was carried out as will be discussed below.

2.5.2 CFD simulations

The transient simulation illustrated in Figure 2.3 and Figure 2.4 was carried out for $p=145 \text{ bar}$, $T= 60 \text{ }^{\circ}\text{C}$ imposing an oil mass flow rate of 0.9 kg h^{-1} (inlet velocity $1.428 \cdot 10^{-4} \text{ m s}^{-1}$). The sc-CO_2 mass flow rate was kept equal to 9.00 kg h^{-1} (inlet velocity $3.098 \cdot 10^{-2} \text{ m s}^{-1}$) from $t=0$ to $t=70 \text{ s}$ and then was made to increase stepwise to 30 kg h^{-1} (inlet velocity 0.1033 m s^{-1}). The lower (sampling) outlet was opened at $t=26 \text{ s}$ and closed at $t=76 \text{ s}$ to mimic the experimental history. Figure 2.3 shows the contours of liquid holdup obtained during the whole transient.

As it can be seen, in the first part of the transient (instants t_1 - t_4), the formation of the film starts at the beginning of the run and the film progressively and regularly wets the full extent of the packing sheet. In the second part of the transient (instants t_5 - t_8) the increase of sc-CO_2 flow rate immediately leads to a thickening of the oil

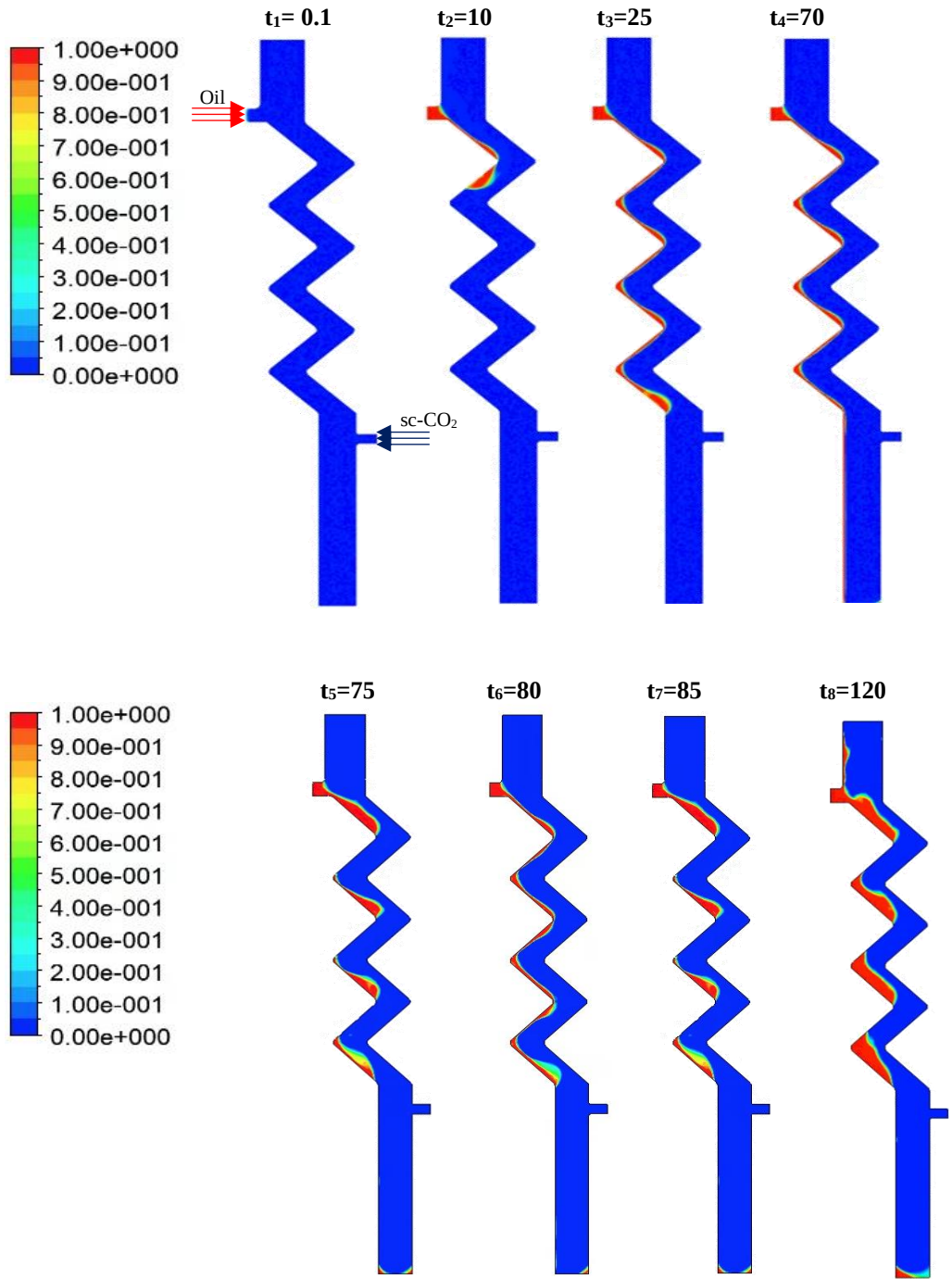


Figure 2.3: Contours of liquid holdup obtained for $p=145$ bar, $T= 60$ °C. Time is expressed in seconds

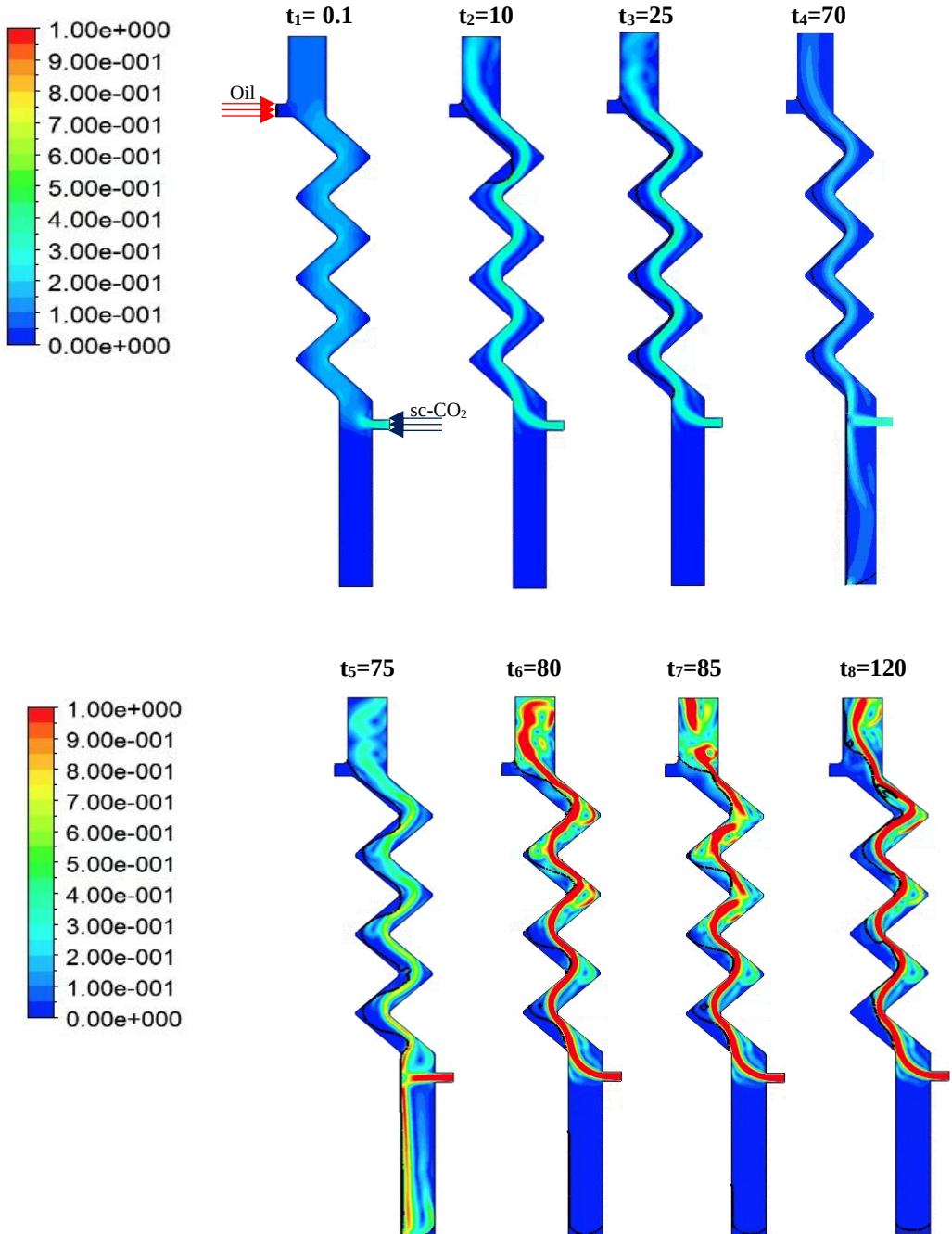


Figure 2.4: Contours of velocity normalized by the maximum inlet velocity of sc-CO₂ (0.103 m s^{-1}). A thin black line represents the interface. Time is expressed in seconds.

film, previously uniformly distributed, in some areas of corrugation. Following the closure of the lower (sampling) outlet at $t=76$ s, the continuity of the film and its normal outflow towards the section bottom are interrupted and the oily phase is entrained towards the upper outlet. This is clearly shown in Figure 2.4, which reports velocity contours normalized by the inlet-velocity of sc-CO₂ at the highest flow rate.

2.6 Conclusions

Experimental results for SFE in a column filled with a Sulzer® EX packing were obtained at different sc-CO₂ (solvent) mass flow rates. The best results to maximize the omega-3 extraction were acquired at an extraction pressure of 145 bar, a temperature of 60 °C and a sc-CO₂/oil flow rate ratio of 100. CFD simulations of a simplified version of the experimental system were also carried out to investigate possible fluid-dynamics improvements of the system. Simulation results were in qualitative agreement with the observed behavior of the system and could predict the transition to flooding conditions. The model can be used in the future to evaluate the optimal operating conditions under a different matrix, a more complex calculation domain and, in conjunction with a predictive model of mass transport, can be used to predict the SFE yield in terms of components with high added value.

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CHAPTER 3: CFD simulations of non-aerated to aerated transitions in UUSTs

Part of the content in this section was previously published in this publication:

L. Schembri, G. Caputo, M. Ciofalo, F. Grisafi, S. Lima, F. Scargiali.
“*CFD simulations of the transition between non-aerated and aerated conditions in uncovered unbaffled stirred bioreactors*”. Chem. Eng. Sci.
vol. 302, part A, 120824, 2025, <https://doi.org/10.1016/j.ces.2024.120824>.

3.1 Summary

In this chapter, the transition between non-aerated and aerated regimes in uncovered unbaffled stirred tanks (UUSTs) was investigated using computational fluid dynamics (CFD). The Volume of Fluid (VOF) method was employed to model the free surface dynamics under various operational conditions. The simulations were able to predict the peculiar experimentally observed behavior of UUSTs and revealed that at velocities below a critical threshold ($N < N_{crit}$), the system remains non-aerated, while exceeding this threshold ($N \geq N_{crit}$) induces bubble ingestion, leading to significant changes in power consumption and flow patterns. The CFD simulations accurately predicted the behavior of the Power Number (N_p) as well as the vortex shape inside the tank both in subcritical and supercritical regimes and showed good agreement with original experimental data and correlations from the literature. Additionally, the modeling of the aerated regime successfully predicted

the vortex shape, the bubble dispersion within the tank, and the cavities formed behind the blades.

3.2 General Overview

Uncovered Unbaffled Stirred Tanks (UUSTs) are gaining significant attention for their interesting performance across diverse industrial sectors such as biochemical, food, oil, chemicals, pharmaceuticals, paper, and power industries. This includes applications in biotechnology, powder dissolution, nuclear fuel processing, wastewater treatment, and so on (Lima et al., 2020; Ramírez-Cruz et al., 2020). These mixers were rarely utilized in industry due to the inherent shortcomings they exhibit: UUSTs, in fact, are limited in mixing effectiveness due to the swirling motions within the fluid, where tangential velocity dominates the flow, resulting in reduced gradients and inadequate axial flow, compared to standard baffled tanks. This swirling motion generates a hollow in the liquid surface within UUSTs, influenced by factors such as impeller design and fluid properties. Notwithstanding these challenges, UUSTs hold promise for specific applications. UUSTs are employed in food and pharmaceutical industries, where vessel cleanness has a primary importance. Another significant advantage of UUSTs is their low power consumption compared to baffled tanks under the same operating conditions. In addition, the gentle stirring in an unbaffled mixing system offers a low shear environment, which promotes the growth of biomass that is normally shear-sensitive (Aloi and Cherry, 1996; Lamberto et al., 1996; Scargiali et al., 2017). Unbaffled

vessels are preferred over baffled systems for highly viscous fluids, where baffled systems may lead to the formation of undesired dead zones. Finally, their ability to provide easy aeration and a gentler impact on solids in crystallizers makes them a valuable choice in many scenarios.

Busciglio et al. (Busciglio et al., 2014) investigated mixing rates in unbaffled vessels under free surface vortex using the planar laser induced fluorescence (PLIF) technique. Their results revealed two distinct, partially segregated zones leading to dual mixing dynamics. Unbaffled vessels showed poorer mixing efficiency, with mixing times 2-3 times longer than baffled tanks for the same power input. However, mixing times (10-100 seconds) were suitable for slower processes. When the vortex bottom approached the impeller plane, mixing efficiencies matched those of baffled tanks, highlighting the broader potential of unbaffled vessels.

As highlighted by Ciofalo et al. (Ciofalo et al., 1996), the depth and shape of the free surface in UUST primarily depend on the impeller's rotational speed. At the same time, additional parameters, for example impeller type and diameter, off-bottom clearance, and physical properties such as viscosity and surface tension, also play significant roles. Busciglio et al. (Busciglio et al., 2013) have classified the flow into various operating conditions based on the position of the lower end of the vortex in relation to the impeller.

Two distinct operational regimes have been identified: sub-critical and super-critical. The sub-critical regime occurs when the rotational speed of the impeller is below a critical threshold ($N < N_{crit}$), meaning that the free surface vortex

has not yet reached the impeller blades. In such circumstances, the formation and subsequent bursting of bubbles inside the reactor can be effectively prevented, as air in the headspace is not drawn within the impeller stream (Prakash et al., 2019). This characteristic makes unbaffled vessels potentially helpful for foaming gas-liquid systems, given that the process rates and gas consumption requirements align with the relatively constrained gas transfer rates reachable under these conditions (Scargiali et al., 2014a; Tamburini et al., 2021). In contrast, the super-critical regime is characterized by rotational speeds exceeding the critical threshold ($N > N_{crit}$), where the free surface vortex reaches the impeller, allowing the gas phase to be ingested and dispersed inside the reactor. This regime is particularly beneficial in three-phase mixing operations where blockage of gas spargers by solid particles is a frequent problem (Tamburini et al., 2016). Solid particles may cause wear of sparger holes or form a muddy solid residue that blocks the holes, adversely affecting the sparger's performance (Lima et al., 2022a). Operating an unbaffled reactor under aerated conditions at velocities above N_{crit} , using a self-ingesting device, can prevent such blockages.

However, it is important to note that under specific circumstances, surface instability phenomena may occur in the use of these mixers, leading to oscillations on the free surface, and potentially adversely affecting agitation and vessel support mechanisms (Busciglio et al., 2016). An increase in impeller speed can cause the system to transition from a sub-critical into a super-critical regime, resulting in significant gas bubble ingestion into the tank. Such a phenomenon also proves to be

disadvantageous in certain processes like polymerization, where bubble adhesion to droplets can lead to the formation of poor-quality polymer beads (Tanaka et al., 1986). However, it is noteworthy that this bubble injection phenomenon can be leveraged as a vortex-ingesting (self-induced) system, offering advantages in enhancing mass transfer for bio-hydrogen production (Ngo et al., 2012) or serving as a surface aerator in wastewater treatment applications (Lima et al., 2024; Moucha et al., 2003). However, there is a paucity of studies examining the hydrodynamics of UUSTs compared to standard baffled systems. Research in this area can be categorized into three main domains. Firstly, research focuses on studying and/or modeling the free surface dynamics and on comparing mean tangential velocity profiles with the classical Rankine vortex, as reported by Nagata et al. (Shinji Nagata, 1975). The second set of studies explores employing UUST hydrodynamics in various processes, such as aeration and crystallization. Lastly, the research explores into understanding and enhancing the mixing process within UUSTs, using computational fluid dynamics as well. Numerical modeling of flow in UUSTs presents complexities, as emphasized by Ciofalo et al. (Ciofalo et al., 1996), primarily attributed to strong turbulence anisotropy, streamline curvature, and flow rotation throughout the tank. Unlike baffled tanks, where turbulence is primarily anisotropic only within the impeller stream and largely isotropic in the overall fluid flow, UUSTs display a more complex flow regime. Moreover, conventional approaches like the Reynolds-Averaged Navier–Stokes (RANS) method employing

common eddy viscosity turbulence models struggle to accurately capture secondary flows and impeller discharge areas in UUSTs.

Numerous numerical studies existing in literature adopt the RANS approach. Haque et al. (Haque et al., 2006), Glover et al. (Glover and Fitzpatrick, 2007) and Mahmud et al. (Mahmud et al., 2009) have attempted to improve accuracy in the numerical studies. Haque et al. and Mahmud et al. employed a second-moment differential Reynolds-stress turbulence Model (RSM) and the Shear Stress Transport (SST) eddy viscosity model, demonstrating a more accurate prediction of bulk fluid motion. Mahmud et al. (Mahmud et al., 2009) demonstrated that these models often underestimate the size of the discharge zone and fail to accurately capture the dynamics of the free surface. While the predicted overall shape of the liquid free surface matches well with measurements, the vortex depth is not accurately predicted. The RSM model shows a good match between the measured and predicted axial and tangential velocities, but it significantly underestimates the radial velocity. Predictions of turbulent kinetic energy align reasonably well with measurements in the bulk flow region, although there is a noticeable discrepancy near the reactor wall where this quantity is underpredicted.

Additionally, the RSM model provides better predictions of the locations of the velocity peak, i.e., the boundary between the forced- and free-vortex regions. Furthermore, the RSM model predicted axial and radial velocities in better agreement with measurements than the SST model. Recently, the Unsteady Reynolds-Averaged Navier–Stokes (URANS) method has been utilized with these

types of problems. The advantage of URANS lies in its capability to capture part of the unsteady behavior periodically induced by the impeller motion, resulting in more accurate outcomes compared to the conventional RANS approach (Zamiri and Chung, 2017).

Large-Eddy Simulation (LES) has emerged as a powerful tool for resolving flow dynamics within stirred tanks, as demonstrated by Derksen and Van den Akker (Derksen and Van Den Akker, 1999). Alcamo et al. (Alcamo et al., 2005) further showed the utility of LES in understanding complex turbulent flow fields within industrial-scale, unbaffled tanks stirred by a Rushton turbine. Their study utilized the Smagorinsky model to capture unresolved subgrid scales, achieving numerical predictions that closely matched experimental data obtained via Particle Image Velocimetry (PIV) and literature results, particularly in terms of mean tangential velocities. LES excels in capturing macro instabilities, flow anisotropy, and trailing vortex structures near the impeller blades, pivotal to the mixing process. These attributes make LES particularly effective for modeling the flow within Uncovered Unbaffled Stirred Tanks. Despite being promising, there remains a lack of studies utilizing LES for UUST analysis. Lamarque et al. (Lamarque et al., 2010) worked with LES application in UUSTs, investigating a flow precipitator mixed with a rod at the tank bottom. Their research produced promising outcomes, as the average and fluctuating velocities closely matched experimental Laser-Doppler velocimetry measurements and coherent turbulent structures revealing insights into mixing dynamics using the Q-criterion, which identifies regions where the fluid follows a

closed rotational path. Similarly, Bertrand et al. (Murielle Bertrand et al., 2012) investigated into UUST flow analysis, highlighting anisotropic regions within the tank using Lumley's Triangle (Escudié and Liné, 2006) and elucidating their impact on the precipitation process. Notably, these studies, while informative, deviate from industry-standard impeller configurations.

Moreover, the computational mesh often lacks the resolution necessary to precisely capture both mean and fluctuating velocities in the central region of the tank near the impeller. Deshpande et al. (Deshpande et al., 2017) employed Large-Eddy Simulations (LES) in a vessel stirred by a Rushton turbine using the free open-source computational fluid dynamics (CFD) software package OpenFOAM (Open Source Field Operation and Manipulation), and likened power consumption with the findings of Scargiali et al. (Scargiali et al., 2014b). Although LES proves to be effective in resolving flow within UUSTs, its computational demands are challenging. Firstly, the mesh size required for a high-quality solution is significant. Secondly, considerable time is needed to develop the flow and deform the free surface, increasing computational costs. While, in LES or URANS studies of standard baffled tanks, approximately twenty revolutions of the impeller are typically needed to establish flow, UUST simulations may require over 100 impeller revolutions, emphasizing the importance of simulation time (Ramírez-Cruz et al., 2020). Furthermore, accurate modeling of the free surface in UUSTs is essential due to flow pattern alterations induced by surface deformation driven by the impeller-induced flow dynamics. Thus, as highlighted by Ciofalo et al. (Ciofalo et al., 1996),

accurately predicting the surface shape in Uncovered Unbaffled Stirred Tanks is complex and serves as a crucial validation metric for overall flow field simulations.

Their pioneering work attempted to model UUSTs using a body-fit method to simulate liquid surface deformation, necessitating an iterative process to achieve a steady-state solution under a zero-stress condition in the normal direction. Subsequent research has favored the Volume of Fluid (VOF) method for solving flow dynamics within UUSTs (Jiao et al., 2024). Additionally, a hybrid VOF-Eulerian-Eulerian model was employed by Haque et al. (Haque et al., 2006) and Mahmud et al. (Mahmud et al., 2009). The primary challenge with these methods arises from representing the liquid volume fraction across a broad range from 0 to 1. Consequently, the validation test can be somewhat misleading. In contrast, Bertrand et al. (M. Bertrand et al., 2012) employed a Lagrangian approach known as Discontinuous Front-Tracking (DFT), which can achieve high accuracy given a finely detailed mesh. However, this precision may come at the cost of increased computational demands.

The primary objective of this study is to assess the fluid dynamics established in UUSTs under both sub-critical and supercritical conditions and to reproduce the unique experimental behavior, previously observed by the same authors, of power consumption under varying operating conditions, such as changes in the rotational speed of the Rushton impeller and relevant aeration conditions. Additionally, the study aims to correctly predict the vortex shape in these systems under both non-aerated and aerated conditions. To achieve this objective, various

computational fluid dynamics (CFD) simulations were conducted using the Volume of Fluid method. These simulations enable adequate assessments of the free surface shape in a biphasic system in the absence and presence of bubble ingestion and the flow patterns generated by this radial impeller design.

3.3 Materials and Methods

The mixing system investigated in this study is shown in Figure 3.1.a. The system comprises a flat-bottomed cylindrical tank equipped with a standard Rushton turbine for stirring. The working fluid utilized was water, with a density $\rho_l = 998 \text{ kg m}^{-3}$ and a viscosity $\mu_l = 0.00102 \text{ Pa} \cdot \text{s}$. The initial liquid level was set to $H = T$. Detailed geometrical dimensions of the stirred vessel are provided in Table 3.1.

To account for the formation of a vortex at the liquid surface during stirring, an air zone with a density $\rho_g = 1.225 \text{ kg m}^{-3}$ and a viscosity of $\mu_l = 1.79 \times 10^{-5} \text{ Pa} \cdot \text{s}$ was introduced above the liquid level. This air zone had a height of $H_g = 0.11 \text{ m}$ to prevent the liquid from spilling out of the tank.

3.3.1 Geometrical modeling

The initial phase of the simulation involved geometric modeling and the creation of body-fitted grids using the mesh tools within ANSYS® Workbench software (ANSYS Inc®, USA). The fluid domain was partitioned into two sections, namely, *fluid-domain-zone* and *fluid-subdomain-zone*, containing the impeller.

Table 3.1: *Dimensions of the stirred tank utilized.*

Geometry	Symbol	Value
Tank diameter	T	0.19 m
Tank height	H_T	0.30 m
Liquid level	H	T
Impeller diameter	D	$T/3$
Off-bottom clearance	c	$T/3$
Blade height	a	$D/5$
Blade width	b	$D/4$
Disk diameter	d	$\frac{3}{4} D$
Shaft diameter	d_s	0.017 m
Blade Angle	θ	60°

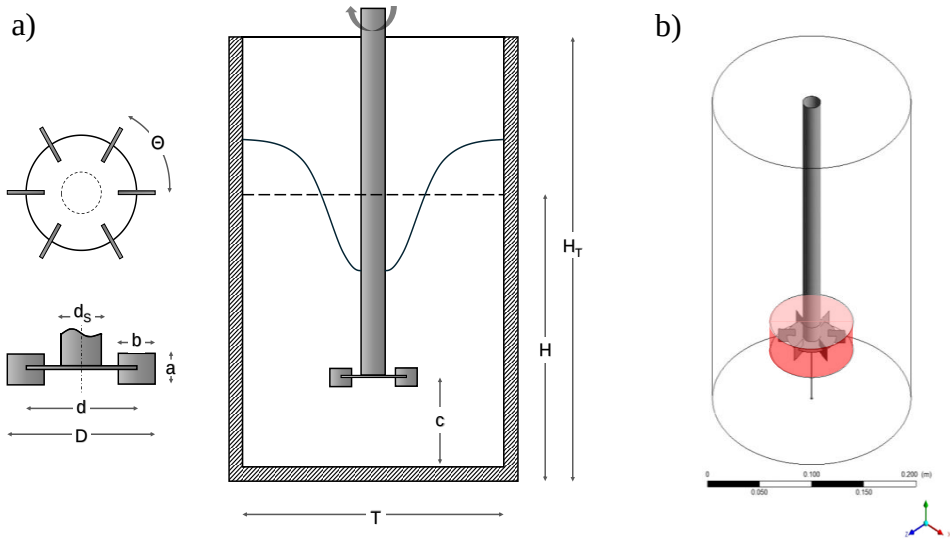


Figure 3.1: *The adopted mixing system: (a) Stirred tank and Rushton turbine; (b) Geometrical domain with the fluid-subdomain-rotating-zone highlighted in red.*

To simulate the impeller rotation, a moving reference frame was applied to the impeller zone and a sliding mesh approach was used to manage the relative motion between the rotating and stationary zones. As shown in Figure 3.1.b., the common interface of the two zones was placed at $r = 0.04$ m and $0.049 \text{ m} < z < 0.077 \text{ m}$. The region of the subdomain circumscribed to the impeller has been highlighted in red.

3.4 Mathematical model

3.4.1 The governing equations

The following momentum and continuity equations were solved across the entire fluid domain:

$$\frac{\partial \rho}{\partial t} = \nabla \cdot (\rho \vec{v}) = 0 \quad (3.1)$$

$$\frac{\partial}{\partial t}(\rho \vec{v}) + \nabla \cdot (\rho \vec{v} \vec{v}) = -\nabla p + \nabla \cdot \left[\mu \left(\nabla \vec{v} + \nabla \vec{v}^T \right) \right] + \rho \mathbf{g} + \mathbf{F} \quad (3.2)$$

where t is the time, $\mathbf{u}$ is the mixture velocity, p is the static pressure, $\mathbf{g}$ is gravitational acceleration and $\mathbf{F}$ is body force. ρ and μ are respectively mixture density and dynamic viscosity, described in the following section.

3.4.2 The Volume of Fluid model

Under stirring conditions, near to the free surface, the formation of a central vortex occurred which curved the surface itself in the direction of the impeller (Li et

al., 2017; Phusa et al., 2021). By solving a set of continuity equations, the VOF method can model two or more immiscible *Eulerian Phases* to find out the volume fraction (α) in each computational cell (Li and Xu, 2017). The properties and variables of any given cell are affected by its volume fraction values. Each cell is characterized by either air ($\alpha_g = 1$), water ($\alpha_g = 0$) or a blend of water and air ($0 < \alpha_g < 1$). Properties and variables were allocated to each cell within the solution domain according to the local volume fraction of each phase. The conservation of volume fraction for the generic phase i (α_i) can be determined for incompressible phases and no inter-phase mass transfer by:

$$\frac{\partial \alpha_i}{\partial t} + \vec{v} \cdot \nabla(\alpha_i) = 0 \quad (3.3)$$

In each control cell, the mixture's volume fraction and physical properties were constrained as follows:

$$\alpha_g + \alpha_l = 1 \quad (3.4)$$

$$\rho = \rho_l \alpha_l + \rho_g \alpha_g \quad (3.5)$$

$$\mu = \mu_l \alpha_l + \mu_g \alpha_g \quad (3.6)$$

3.4.3 The turbulence model

Turbulent flows are characterized by fluctuating velocity fields, which in turn lead to fluctuations in the transported variables. These fluctuations encompass a wide range of scales and frequencies, making direct modeling difficult. As an alternative, to describe the turbulent flow one can use Reynolds-averaged Navier-

Stokes (RANS) models, including eddy viscosity models (such as $k-\varepsilon$ or $k-\omega$), and Reynolds Stress transport models (RSM) (Montante et al., 2006).

Before evaluating grid independence, a systematic study was conducted to select the most appropriate turbulence model. This was achieved by comparing the free surface profile evaluated through the Nagata correlation (Shinji Nagata, 1975), as refined by Ciofalo et al. (Ciofalo et al., 1996), with solutions obtained from a series of preliminary simulations using various turbulence models with an intermediate grid: the standard $k-\varepsilon$ model (Launder and Spalding, 1974), the $k-\omega$ SST (Shear Stress Transport) model (Menter et al., 2003), and two versions of a Reynolds Stress transport model (RSM), using either a linear pressure-strain correlation approximation (RSM-LPSM) or a quadratic pressure-strain correlation approximation (RSM-QPSM). The former version is essentially that proposed in 1975 by Launder et al. (Launder et al., 1975a), the latter is the variant introduced in 1991 by Speziale et al (Speziale et al., 1991). All the above models are implemented as standard options in ANSYS-Fluent® (ANSYS Inc., 2021). A preliminary mesh with approximately 2 million volumes was used, which was essentially equivalent to the final mesh selected through grid independence analysis, except for a refinement applied near the wall regions.

Comparative results concerning the vortex shape are shown in Figure 3.2 for 400 RPM, liquid viscosity equal to 0.001 Pa·s. The eddy-viscosity RANS models ($k-\varepsilon$ and SST) exhibit inefficiencies in predicting the vortex shape accurately. The seven-equation Reynolds Stress Models, especially RSM-LPSM, yield a better

agreement with the empirically-based profile reported by Nagata. The RSM-QPSM predicts an irregular diffused liquid-air interface which is not exhibited by the experiments at this rotational speed and liquid viscosity (Michalcová and Kotrasová, 2020). In addition to vortex shape analysis, the evaluation included consideration of the Np value, which can be assessed either from the torque exerted by the impeller or from the energy dissipation (ε), which represents the rate at which the fluid's turbulent kinetic energy is converted into thermal energy due to viscous forces. This comparison is shown in Figure 3.3. The values of Np computed by the two independent methods practically coincide only when the RSM models are used, while they broadly diverge when eddy-viscosity models (especially the $k-\varepsilon$) are adopted. In addition, only the RSM models yield Np values in agreement with the experimental data and relevant empirical correlation proposed by Scargiali et al. (Scargiali et al., 2013) ($Np = 19.5 Re^{-0.3}$). As a result of the study, the RSM LPSM emerged as the optimal turbulence model. Therefore, it was selected for the grid independence assessment.

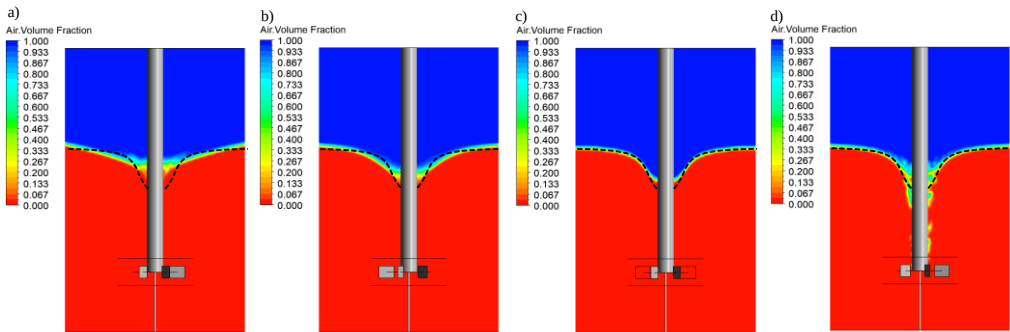


Figure 3.2: Air volume fraction maps at 400 RPM (liquid viscosity of $0.001 \text{ Pa}\cdot\text{s}$) for different turbulence models considered: a) $k-\varepsilon$; b) $k-\omega$ SST; c) RSM LPSM; d) RSM QPSM. Black dashed line: Nagata correlation.

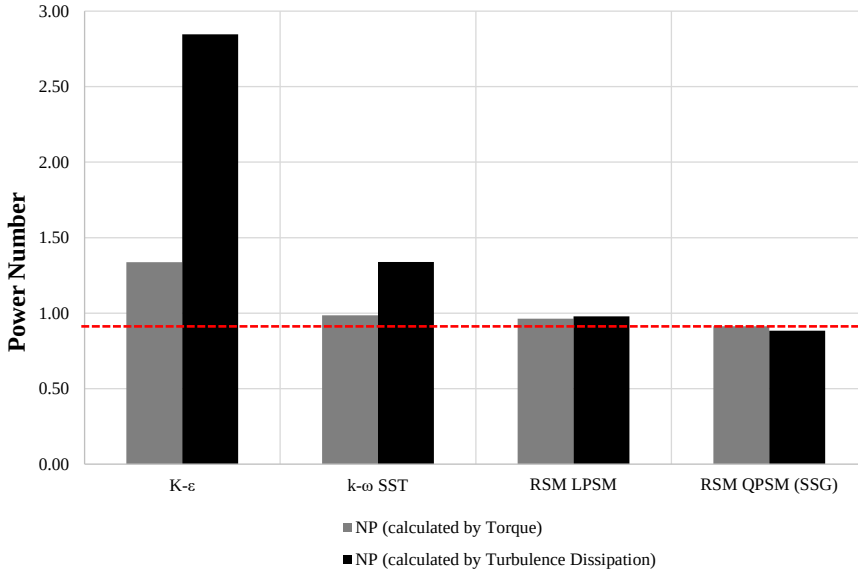


Figure 3.3: Power Number (Np) at 400 RPM for different turbulence models: k - ϵ , k - ω SST, RSM LPSM, and RSM QPSM. Grey bars: Np calculated using torque; black bars: Np calculated using turbulence dissipation. Red dashed line: reference Power Number ($Np = 19.5 Re^{-0.3} = 0.918$).

The Reynolds stress transport equations in the RSM-LPSM model are (Launder et al., 1975b):

$$\frac{\partial \tau_{ij}}{\partial t} + \frac{\partial u_k \tau_{ij}}{\partial x_k} = D_{T,ij} + D_{L,ij} + P_{ij} + \phi_{ij} + \epsilon_{ij} + F_{ij} \quad (3.7)$$

where τ_{ij} is the generic Reynolds Stress, defined as $\tau_{ij} = \rho \overline{v'_i v'_j}$. $D_{T,ij}$ is the turbulent diffusion, $D_{L,ij}$ is molecular diffusion, P_{ij} is the stress production. The term ϕ_{ij} accounts for the pressure-strain correlation, ϵ_{ij} represents dissipation and F_{ij} is the production due to system rotation. The terms $D_{L,ij}$, P_{ij} and F_{ij} do not necessitate any modeling, and have the following expressions:

$$D_{L,ij} = \frac{\partial}{\partial x_k} \left[\mu \frac{\partial}{\partial x_k} (\overline{v'_i v'_j}) \right] \quad (3.8)$$

$$P_{ij} = -\rho \left(\overline{v'_i v'_k} \frac{\partial u_j}{\partial x_k} + \overline{v'_j v'_k} \frac{\partial u_i}{\partial x_k} \right) \quad (3.9)$$

$$F_{ij} = -2\rho\Omega_k (\overline{v'_j v'_m} \varepsilon_{ikm} + \overline{v'_i v'_m} \varepsilon_{jkm}) \quad (3.10)$$

The impeller region was simulated using a rotating reference frame, where Ω_k is equal to the impeller rotational velocity. The terms $D_{T,ij}$, ϕ_{ij} , ε_{ij} required modelling to close the set of Eq. (3.7). For the turbulent diffusion $D_{T,ij}$ the simplified model authored by Daly and Harlow (Daly and Harlow, 1970) was adopted:

$$D_{T,ij} = \frac{\partial}{\partial x_k} \left(\frac{\mu_t}{\sigma_k} \frac{\partial \overline{v'_i v'_j}}{\partial x_k} \right) \quad (3.11)$$

where σ_k (turbulent Prandtl number for k) was set to 0.82 (Lien and Leschziner, 1994).

The pressure-strain term ϕ_{ij} was divided into three components as proposed by Gibson and Launder (Gibson and Launder, 1978), Fu et al. (Fu et al., 1988), and Launder (Launder, 1989):

$$\phi_{ij} = \phi_{ij,1} + \phi_{ij,2} + \phi_{ij,w} \quad (3.12)$$

where $\phi_{ij,1}$ is labelled slow pressure-strain term, $\phi_{ij,2}$ rapid pressure-strain term, and $\phi_{ij,w}$ wall-reflection term. The slow and rapid pressure-strain terms are modeled as:

$$\phi_{ij,1} = C_1 \rho \frac{\varepsilon}{k} \left[\overline{v'_i v'_j} - \frac{2}{3} \delta_{ij} k \right] \quad (3.13)$$

$$\phi_{ij,2} = C_2 \left[(P_{ij} + F_{ij} - C_{ij}) - \frac{1}{3} \delta_{ij} (P_{kk} - C_{kk}) \right] \quad (3.14)$$

where $C_1 = 1.80$, $C_2 = 0.60$. P_{ij} and F_{ij} are defined by Eqs. (3.9) and (3.10). C_{ij} is the convection term:

$$C_{ij} = \frac{\partial}{\partial x_k} (\rho u_k \overline{v'_i v'_j}) \quad (3.15)$$

The term $\phi_{ij,w}$ is responsible for the redistribution of normal stress near the wall and is represented by the following model:

$$\begin{aligned} \phi_{ij,w} = & C'_1 \frac{\varepsilon}{k} \left(\overline{v'_k v'_m} n_k n_m \delta_{ij} - \frac{3}{2} \overline{v'_i v'_k} n_j n_m \right. \\ & \left. - \frac{3}{2} \overline{v'_j v'_k} n_j n_k \right) \frac{C_l \kappa^{\frac{3}{2}}}{\varepsilon d} \\ & + C'_2 \frac{\varepsilon}{k} \left(\phi_{km,2} n_k n_m \delta_{ij} - \frac{3}{2} \phi_{ik,2} n_j n_m \right. \\ & \left. - \frac{3}{2} \phi_{jk,2} n_j n_k \right) \frac{C_l \kappa^{\frac{3}{2}}}{\varepsilon d} \end{aligned} \quad (3.16)$$

where $C'_1 = 0.50$, $C'_2 = 0.30$. n_k is the x_k component of the unit vector normal to the wall, d is the normal distance to the wall, and $C_l = C_\mu^{\frac{3}{4}}/k$, where $C_\mu = 0.09$ and κ is the von Kármán constant, assumed equal to 0.4187. The dissipation tensor ε_{ij} is modeled as isotropic:

$$\varepsilon_{ij} = \frac{2}{3} \delta_{ij} \rho \varepsilon \quad (3.17)$$

The scalar dissipation rate ε can be determined using a transport equation like that in the standard k - ε model.

3.4.4 Mesh and grid independence study

The fluid domain was discretized using structured hexahedral grids, chosen for their ability to yield a satisfactory convergence with a moderate number of finite volumes. This type of grid offers higher accuracy and computational efficiency compared to tetrahedral grids, which is particularly beneficial in simulations involving advection-dominated flows where reduced numerical diffusion is critical (Michalcová and Kotrasová, 2020). Hexahedral grids align better with primary flow directions, thereby improving solution accuracy and stability.

To ensure the simulation's accuracy, the grids were refined in critical areas such as blades and tank walls. Grid independence was rigorously tested by approximately halving and doubling the number of volumes compared to the Fine grid configuration (Fig. 4) along each of the three cylindrical directions (azimuthal, vertical and radial). This investigation spanned a range from 990,000 to 4,000,000 finite volumes. The analysis concerned the comparison of a global quantity, such as the power number. As shown in Table 3.2 the relative error between the fine and the very fine grids was found lower than 4 %, supporting the selection of the fine grid as a balanced choice in terms of accuracy and computational efficiency.

Furthermore, the power number obtained with the chosen grid showed good agreement with the experimental value of 0.93 (Scargiali et al., 2013), confirming the reliability of the simulation results with this configuration.

Table 3.2 *Grid independence study for CFD simulations performed using the RSM LPSM turbulence model, with an impeller speed of 400 RPM and a liquid viscosity of 0.001 Pa s. “Coarse grid” and “very fine grid” indicate approximately the halving and doubling of the number of volumes compared to the Fine grid configuration.*

Mesh	Number of volumes	Power Number	Relative error (%)
1 – Coarse grid (azimuthal)	794,304	1.481	59.9%
2 – Coarse grid (vertical)	992,880	1.343	45.0%
3 – Coarse grid (radial)	992,880	1.414	52.7%
4 – Fine grid	1,985,760	0.962	3.9%
6 – Very fine grid (azimuthal)	3,971,520	0.926	0.0%
7 – Very fine grid (radial)	3,971,520	0.938	1.3%
5 – Very fine grid (vertical)	3,971,520	0.955	3.1%

3.4.5 Computational details

Time-dependent computational fluid dynamics simulations were conducted using the commercial software *ANSYS® Fluent V23 R2* by *ANSYS® Inc* (ANSYS Inc., 2021). The initial time step was set to 0.00083 seconds, corresponding to the time required for a 2-degree rotation, which aligned with a circumferential division of the mesh when the impeller operates at a speed of 400 RPM. Each time step

involved 15 iterations. A phase-coupled SIMPLEC algorithm was employed to manage the velocity-pressure coupling. The volume fraction was discretized using a compressive scheme, while pressure was discretized using a PRESTO! scheme. Second-order upwind schemes were employed to discretize the momentum equations, turbulence kinetic energy, and turbulence dissipation rate. Initially, the fluid was at rest and the free liquid surface was located at a height $H = T$.

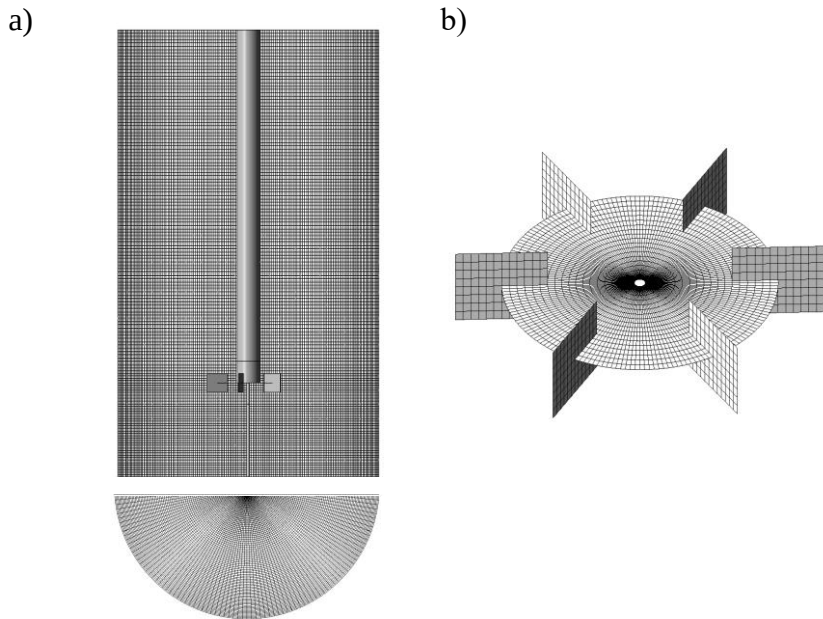


Figure 3.4: *Computational Domain: (a) Adopted Mesh; (b) Detailed view of the impeller mesh, specifically focusing on the simulated Rushton Turbine.*

Regarding the boundary conditions, the velocities at the tank's bottom and sidewalls were fixed at zero, and a standard wall function was utilized to simulate fluid flow near the walls. The impeller zone was resolved using a sliding mesh approach, with a specific rotational velocity imposed on the impeller. In contrast, the

stationary zone was resolved using a stationary reference frame. Within the moving reference frame, the impeller blades were treated as stationary walls. The top of the vessel was designated as a pressure-outlet boundary condition with a gauge pressure of zero.

The simulation runtime depends on reaching the steady state for each specific rotational speed adopted (value between 200 to 1200 RPM). The attainment of the steady state was assessed by evaluating the Power Number and its invariance over time. It was observed that the time to reach the steady state condition decreases as the rotational speed increases. Upon achieving final convergence, all the residuals were less than $1 \cdot 10^{-4}$ for all simulations. The simulations took approximately 3÷5 week to run on the following system specifications: Windows 10 Enterprise 64-bit (10.0, build 19045); Processor: Intel(R) Xeon(R) CPU E5-2640 v3 @ 2.60GHz (32 CPUs), ~2.6GHz; Memory: 65536MB RAM.

3.4 Results and discussion

3.4.1 Power consumption

The experimental specific power dissipation values, sourced from Scargiali et al. (Scargiali et al., 2013), were obtained at different agitation velocities using a static frictionless turntable. CFD simulations of the same system were conducted. Four viscosities were selected for this study: 0.001 Pa·s, 0.00285 Pa·s, 0.000707 Pa·s, and 0.00152 Pa·s. The experimental results reported in (Scargiali et al., 2013) are presented in Figure 3.5.a and the four cases are shown from the lowest

viscosity (distilled water, 0.001 Pa·s) to the highest viscosity case (water-polyvinylpyrrolidone solution, 0.0152 Pa·s).

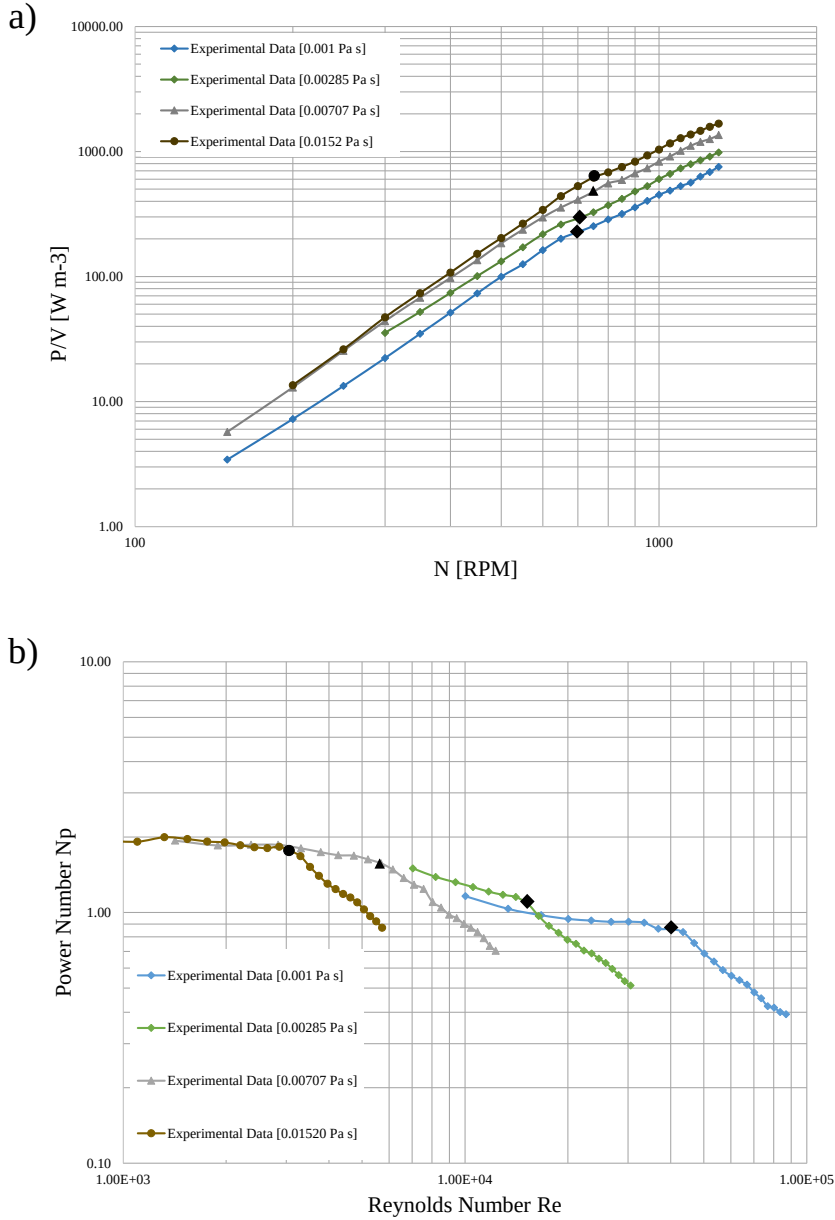


Figure 3.5: Experimental specific power dissipation (a) and the respective power number (b) for liquid viscosities investigated (Scargiali et al., 2013).

As shown, bold black symbols indicate the critical rotational speeds (N_{crit}) when the free surface vortex reached the impeller blade. Experimental observations of the reactor revealed that at rotational speeds below N_{crit} , no bubbles were present in the liquid phase. However, once N_{crit} was exceeded, air was drawn into the system from the free surface vortex, causing a dispersion of bubbles within the liquid. As shown in Figure 3.5.a, a steep decrease in power dissipation with agitation speed occurred across all configurations.

The authors (Scargiali et al., 2013) reported that for rotational velocities below N_{crit} , the specific power dissipation increased with agitation speed, following a power law with an exponent of approximately 3. Above the critical agitation speed, a noticeable decrease in the power law exponent occurred, due to the dispersed gas phase present in the vessel. According to several studies, this behaviour could be attributed to the formation of gas pockets behind the stirrer blades (Maluta et al., 2023; Nienow et al., 1985). Another phenomenon visually observed, which affected the mechanical power demand, was that as velocity increased in uncovered unbaffled tanks, a progressively larger portion of the impeller blades came in contact with the vortex and therefore with the gas phase overlying the liquid. Consequently, these submerged blades were no longer in contact with the liquid phase and thus ceased to contribute to torque generation. The total power dissipation was converted into power number N_p values, defined as:

$$N_p = \frac{P}{\rho_L N^3 D^5} \quad (3.18)$$

where P is agitation power [W], ρ_L is liquid density [kg m^{-3}], N is agitation speed

$[\text{s}^{-1}]$ and D is impeller diameter $[\text{m}]$. Power number results were reported Figure 3.6.b for the four liquid viscosities investigated. The power number remained nearly constant or slightly decreased until the free surface vortex reached the impeller (non-aerated regime, $N < N_{crit}$), consistent with observations by Ruston et al. (Rushton et al., 1950). Subsequently, a distinct decrease in the power number was noted after the vortex reached the impeller and air bubbles started dispersing inside the reactor ($N > N_{crit}$). Upon reaching critical conditions, the dependence of N_p on the Reynolds number follow a power law that closely resembled simple inverse proportionality. In this regime, the phenomena already discussed affecting power demand gave rise to a quite dependence of N_p on Re , leading to a simple proportionality between P and N^2 .

3.4.2 CFD simulation results on Power Number

In order to evaluate the Power Number (N_p) calculated through Eq. (3.18), it was necessary to assess the power consumption value as a function of the impeller's rotational speed and the viscosity of the liquid under consideration.

For each simulation step, the dissipated power was calculated as follows:

$$P = N \cdot \sum_{i=1}^n T_i \quad (3.19)$$

where N is the rotational speed at which the impeller was rotating, measured in radians per second $[\text{rad s}^{-1}]$ and $\sum_{i=1}^n T_i$ is the sum of the moments exerted on the impeller surfaces and the shaft, measured in Newton-meters $[\text{N m}]$ that was obtained

according to the CFD simulation results. The plot of N_p vs Reynolds number (Re) is presented in Figure 3.6, where the mixing Reynolds number in stirred tanks was defined as $Re = (\rho_l D^2 N) / \mu_l$.

The data for the power number (N_p) were meticulously gathered for each specified liquid viscosity (0.001 Pa·s, 0.00285 Pa·s, 0.000707 Pa·s, and 0.00152 Pa·s) while maintaining constant impeller speeds at 200, 400, 600, 800, 1000, and 1200 RPM. This methodology enabled a comprehensive examination of different flow regimes:

- Sub-critical regime: in this regime, the vortex generated by the impeller did not yet reach the impeller itself. This phase was crucial for understanding the initial flow characteristics and the onset of vortex formation. The power number (N_p) follows a well-defined law that was solely a function of the Reynolds number (Re). During this phase, fluid behavior is primarily governed by the Reynolds number, representing the predominance of viscous terms over inertial ones.
- Supercritical regime: In the second regime, the vortex has fully reached the impeller, and the gas phase above the liquid is now being dispersed into the liquid as bubbles. This dispersion was significant as it affected the overall mixing efficiency and the homogeneity of the mixture. The behavior of the power number is influenced by a combination of two parameters: the

Reynolds number (Re) and the Froude number (Fr) evaluated by the following expression $Fr = D N^2 / g$, as reported in literature (Scargiali et al., 2013). The transition between these two regions is easily identifiable as it occurred when the Reynolds value reached a certain critical point, related to a specific critical velocity of the impeller (N_{crit}). This transition was associated with the onset of bubble ingestion and the reaching of the vortex at the impeller. In this phase, fluid behavior was influenced by both viscosity and inertial forces, and the Froude number began to play a significant role.

The distinction between these two regions is of fundamental importance as it indicated when significant changes in fluid behavior can be expected based on operational parameters. This transition was typically highlighted when the Reynolds value crossed a critical threshold, emphasizing the shift from a viscosity-dominated regime to one in which inertial forces and impeller speed significantly influence fluid behavior. As it can be seen, the results of numerical simulations shown in Figure 3.6 exhibited excellent agreement with experimental data previously documented in the literature. This alignment underscored the robustness and accuracy of the computational fluid dynamics (CFD) modelling employed in this study. By capturing the complex flow dynamics and mixing behavior, the simulations validated the reliability of the chosen turbulence models and grid resolutions. This convergence between simulation and experimental results not only enhances confidence in the computational approach but was further corroborated by the

noticeable change in slope observed in the N_{crit} value upon the vortex reaching the Rushton impeller. This underscores the utility of CFD as a predictive tool for optimizing design parameters and operational conditions in industrial applications, ensuring effective and efficient performance of fluid systems.

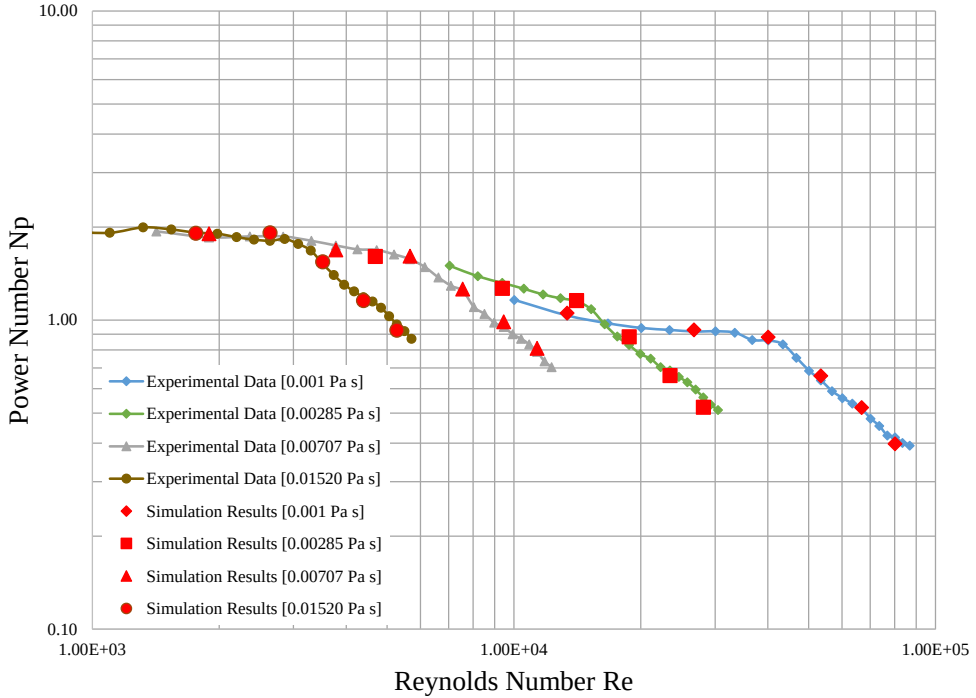


Figure 3.6: Comparison between experimental data (Scargiali et al, 2013) and simulation results on Power number versus Reynolds number for various liquid viscosities.

3.4.3 Vortex shape simulation results

Figure 3.7 shows the impact of impeller speed on the shape of the free surface within a cylindrical agitated vessel for the liquid viscosity equal to 0.001 Pa·s. The simulations confirmed two distinct regimes based on impeller rotational speed. In the subcritical regime, at lower speeds, the vortex generated

inside the vessel did not reach the impeller. The free surface remained relatively stable with minimal deformation. This regime signified more controlled and less turbulent conditions, (impeller velocity of 400 and 600 RPM). At higher speeds, the vortex reached the impeller, causing bubble self-ingestion and deformation of the free surface. This super-critical regime was associated with highly dynamic and turbulent fluid behavior. The analysis effectively visualized the transition between these regimes based on impeller speed. Understanding this relationship was crucial for optimizing fluid behavior within agitated vessels for various industrial processes.

The distinction between subcritical and supercritical regimes significantly impacted process efficiency and performance, influencing decisions about agitation speed, mixing strategies, and equipment design. In Figure 3.8 velocity profiles are also shown within the Uncovered Unbaffled Stirred Tank, confirming findings from the literature of the existence of two toroidal recirculations below and above the impeller, responsible for the vortex formation near the shaft (Xiong et al., 2023).

In this study, the free surface shape was analyzed through computational fluid dynamics simulations and compared with the correlations provided by Nagata, re-elaborated by Ciofalo et al. (Ciofalo et al., 1996) and the Two-Zone Model (2ZM) reported by Busciglio et al. (Busciglio et al., 2013). For the subcritical regime ($N = 600 \text{ RPM}$), the simulations, 2ZM and Nagata models were used to evaluate the free surface profile. Similarly, in the supercritical regime ($N = 800 \text{ RPM}$), the 2ZM and simulation solutions were utilized to determine the vortex shape. The Nagata model employed empirical correlations based on extensive experimental data to predict the

free surface shape. Additionally, the two-zone model divided the tank into two distinct regions: an inner region dominated by rotational flow and an outer region where the flow is more radial.

These models were evaluated in comparison to the CFD simulations, which provide a more detailed and dynamic analysis of the fluid behavior within the tank. Notably, Nagata's predictions were provided solely for sub-critical impeller speeds, as this model was developed specifically for those conditions. The results indicated a high degree of overlap between the solutions under identical conditions, confirming the reliability and accuracy of both models in predicting the free surface profile.

In the subcritical regime (Figure 3.10.a), the agreement among the Nagata model, 2ZM, and simulation results was excellent, demonstrating the robustness of these models in predicting the fluid surface under these conditions. For the supercritical regime (Figure 3.10.b), the 2ZM and simulation results also showed a strong correlation, accurately capturing the complex central vortex formation that occurs at higher agitation speeds. This comprehensive analysis reinforces the validity of using CFD simulations to predict fluid dynamics in UUSTs.

The formation of these air cavities was associated with the ingestion of air into the flow (Figure 3.9), affecting the overall fluid dynamics significantly (Maluta et al., 2021). Figure 3.11 shows the profiles of the free surface evaluated using the models: 2ZM, Nagata and the simulation solution obtained from the implementation of the Volume of Fluid model applying a compressive scheme.

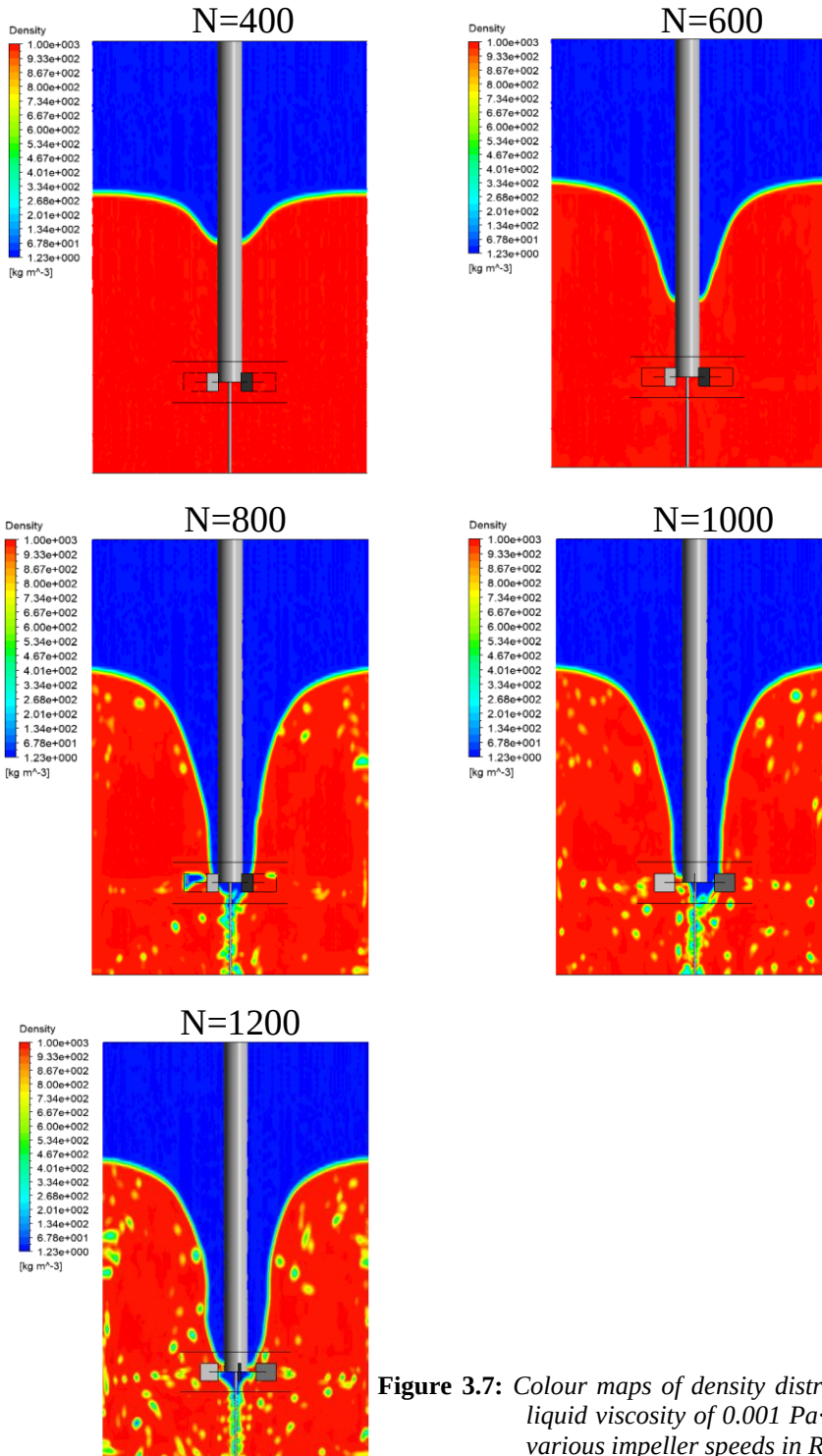


Figure 3.7: Colour maps of density distribution of a liquid viscosity of 0.001 Pa·s and air at various impeller speeds in RPM.

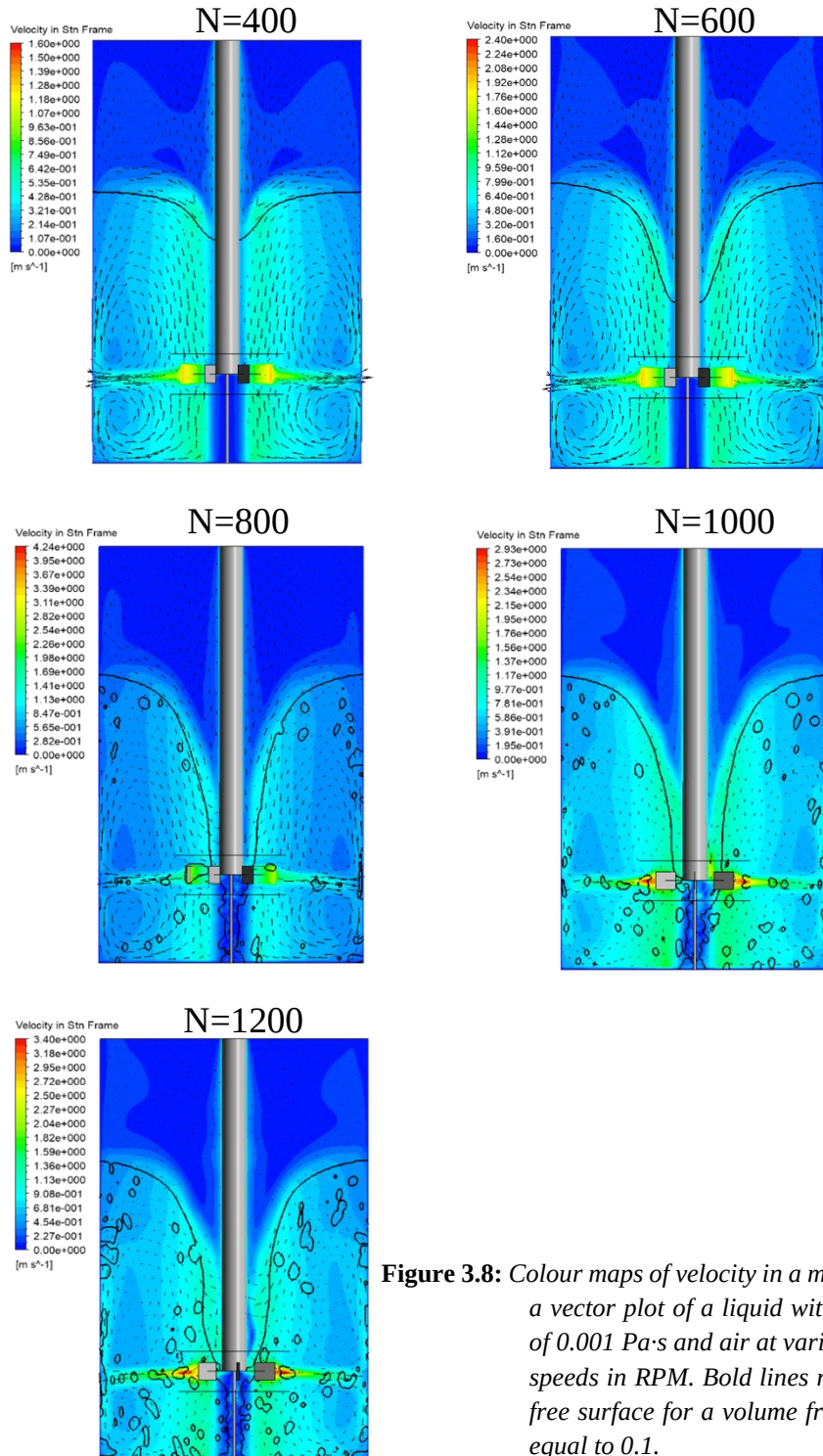


Figure 3.8: Colour maps of velocity in a midplane with a vector plot of a liquid with a viscosity of 0.001 Pa·s and air at various impeller speeds in RPM. Bold lines represent the free surface for a volume fraction of air equal to 0.1.

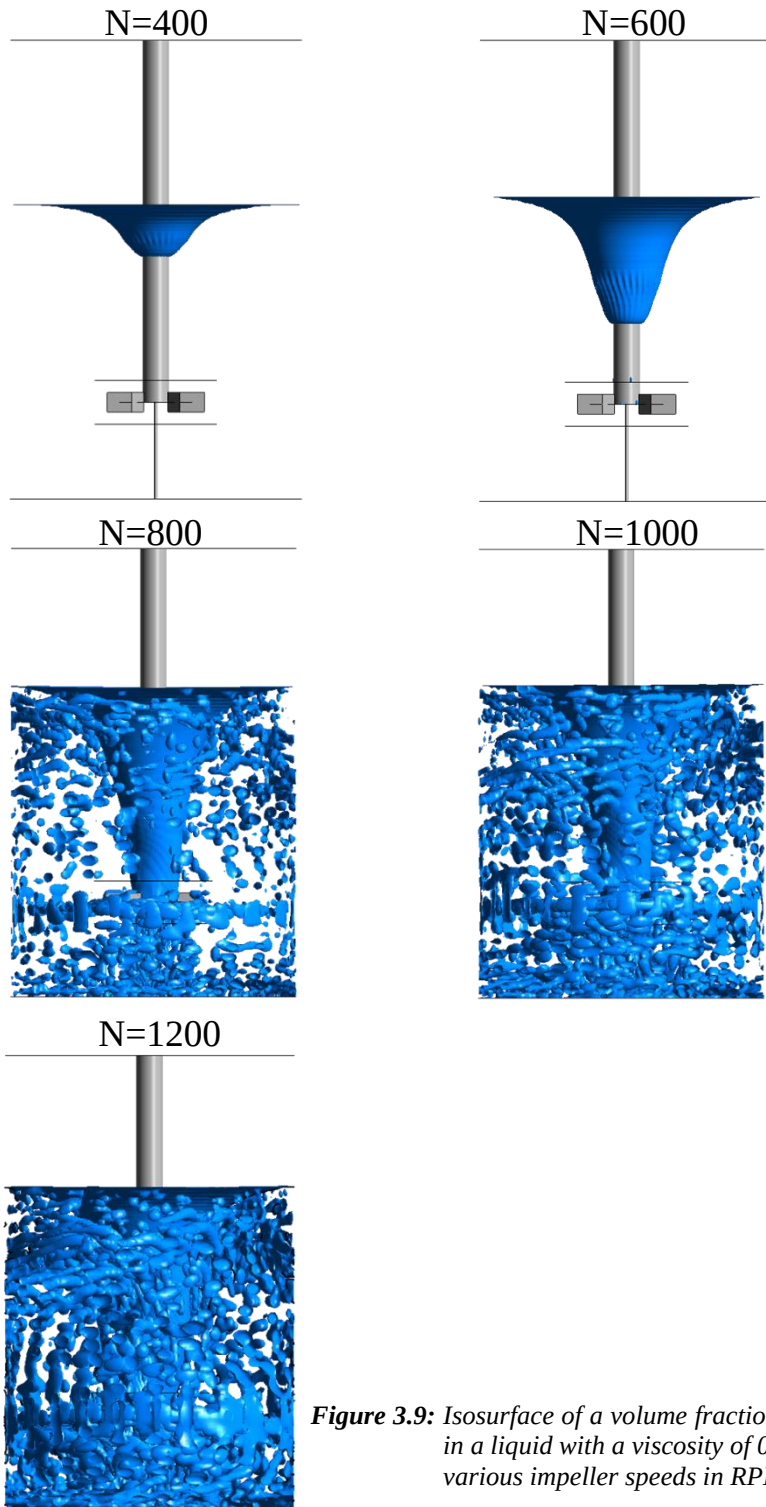


Figure 3.9: Isosurface of a volume fraction of air equal to 0.1 in a liquid with a viscosity of 0.001 Pa·s and air at various impeller speeds in RPM.

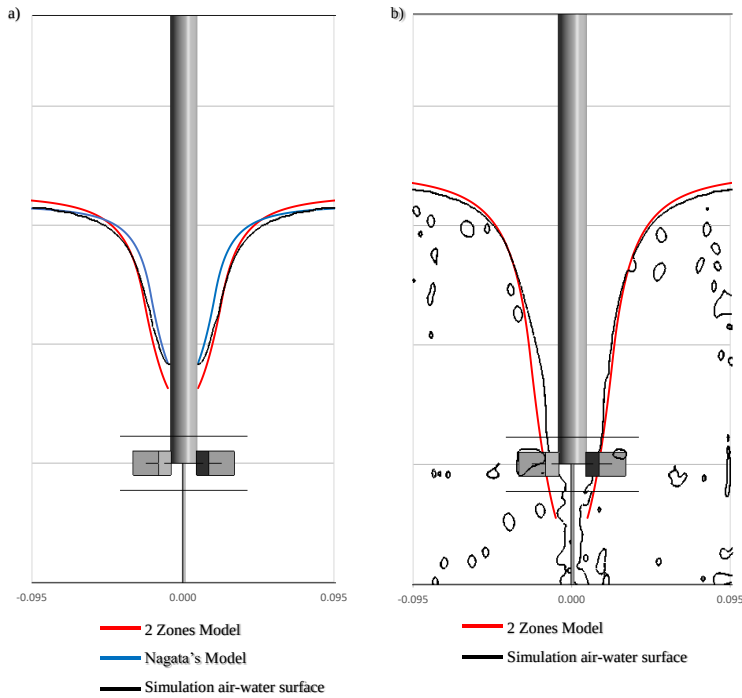


Figure 3.10: Profiles of the free surface evaluated using the models: 2ZM (in red), Nagata (in blue) and the isosurface of a volume fraction of air equal to 0.1 (in black) for a) sub-critical regime (600 RPM) and b) super-critical regime (800 RPM).

They played a pivotal role in the mixing process and mass transfer efficiency within the system. In the supercritical regime, the entrainment of air into the flow behind the impeller blades not only altered the flow patterns but also reduced the effective power consumption. This phenomenon underscored the complex interplay between fluid dynamics and operational parameters in stirred tanks, highlighting the importance of understanding and controlling such phenomena for optimizing mixing efficiency and process performance.

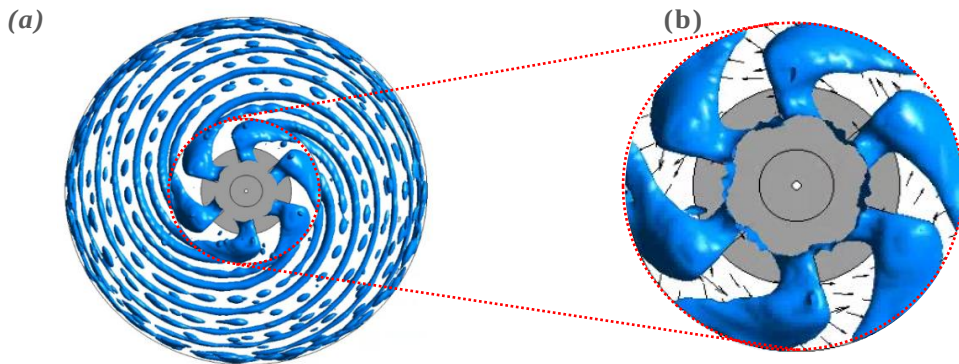


Figure 3.11: *Cavities behind the blades of a Rushton turbine in a supercritical stirred vessel ($N= 800$ RPM): (a) Isosurface of Volume Fraction of air equal to 0.1 on the plane of the impeller; (b) detail of cavities.*

3.5 Conclusions

In this work, Computational Fluid Dynamics (CFD) simulations were employed to investigate the transition between non-aerated and aerated conditions in Uncovered Unbaffled Stirred Tanks (UUSTs). The Volume of Fluid (VOF) method effectively captured the free surface dynamics and vortex shapes under various impeller speeds. Different turbulence models were compared, and the Reynolds Stress Model with Linear Pressure-Strain Model (RSM-LPSM) was selected for its better internal consistency and its ability to predict both the shape and depth of the central vortex, as well as the values of the power number.

The Power Number (N_p), evaluated using ANSYS Fluent[®] fluid dynamics simulations, was plotted against the Reynolds Number (Re) for different liquid viscosities (0.001 Pa·s, 0.00285 Pa·s, 0.000707 Pa·s, 0.00152 Pa·s) at various

impeller speeds (200, 400, 600, 800, 1000, 1200 RPM). The simulation results were able to predict the existence of two distinct regimes: sub-critical and super-critical. In the sub-critical regime ($N < N_{crit}$), the impeller speed was insufficient for the vortex to reach the blades, preventing air bubble entrainment. Conversely, the super-critical regime ($N > N_{crit}$) was characterized by vortex-induced bubble ingestion, leading to significant changes in power consumption and flow patterns. In the super-critical regime, air ingestion significantly altered the flow patterns reducing effective power consumption. This result emphasizes the role of air cavities in influencing power consumption and mixing efficiency within the stirred tank.

The transition from the sub-critical to the super-critical regime was well illustrated. In fact, it was marked by a noticeable change in the slope of the power number vs Reynolds number as the vortex reached the impeller, validating the simulation results against the experimental data.

Additionally, CFD simulations of the free surface shape at impeller speeds of 600 RPM (sub-critical) and 800 RPM (super-critical) were compared with two predictive models (Nagata model and Two-Zone Model, 2ZM). Simulation results showed excellent agreement with both models, confirming the accuracy of the CFD simulations in predicting free surface profiles. These findings underline the importance of CFD simulations for optimizing UUST design and operation, particularly when gas-liquid mass transfer is a key process consideration.

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CHAPTER 4: CFD Analysis of Fluid Motion in a Stirred

Vessel with a Vibrating Plate

Part of the content in this section will be published in the following publication:

S. Almasi, L. Schembri, M. Gimba, F. Scargiali, M. Poux, J. Aubin
“*Vibromixer for FAME Synthesis through Transesterification Reaction*”.
(2025).

4.1 Summary

This chapter investigates the potential of vibromixers to improve liquid-liquid mixing and mass transfer, focusing on biodiesel production. Vibromixers, known for efficient mixing in various industries, offer advantages over traditional stirred tanks by enhancing mass and energy transfer. The study utilizes Computational Fluid Dynamics (CFD) simulations to analyze the impact of vibromixer parameters, such as oscillation amplitude and plate orientation, on fluid motion and dispersion in a pseudo-single-phase system, as well as solid-liquid suspension behavior.

Results from URANS simulations provide insights into flow dynamics, showing unique flow structures and efficient mixing across laboratory and industrial applications. Velocity profiles, pressure distributions, and vector plots confirm complex fluid dynamics within the vibromixer, with single recirculations observed in the flow. Further multiphase simulations explore water-glass bead mixtures, using Eulerian-Eulerian and DEM approaches, laying the foundation for understanding suspension heights and validating experimental data.

Overall, the chapter demonstrates the crucial role of CFD in revealing the behavior within vibromixers, underscoring the importance of parameter optimization for process

improvement. The findings contribute to advancing vibromixer technology and its applications across diverse industries.

4.2 General Overview

As previously mentioned in the third chapter, mixing is a fundamental unit operation in diverse industries, including chemical, pharmaceutical, and food-processing sectors, where the primary objective is to enhance system homogeneity by reducing non-uniformities in composition, phase distribution, or temperature to achieve a specified product. The duration and intensity of mixing processes are known to exert considerable influence on heat and mass transfer rates, operational efficiency, costs, safety, and product quality. The critical link between mixing parameters and product outcomes underlines the need for precise control over mixing parameters to ensure desired process outcomes (Komoda et al., 2000).

In many chemical production processes, conventional stirred-tank reactors face limitations, such as insufficient mixing efficiency and extended processing times, often due to mass-transfer limitations, as observed in reactions like transesterification. As a response to these challenges, intensification technologies, such as vibromixing, have garnered attention due to their potential to enhance mass and energy transfer, thereby increasing reaction conversion rates and reducing processing times compared to traditional methods. Vibromixers have emerged as innovative alternatives to conventional rotating impellers, showing notable advantages across single and multiphase applications in industries ranging from

pharmaceuticals and chemicals to biotechnology, fermentation, and agriculture. Their simple structure, ease of operation, and robust mixing performance make them suitable for processes requiring effective liquid-liquid contacting and homogeneous mixing (Wójtowicz, 2017, 2014; Yakovenko, 1994).

In this chapter, the vibromixer is employed especially for liquid-liquid mixing to promote biodiesel production through enhanced contact and mass transfer in transesterification reactions. The device operates by generating a vertical reciprocating motion via a holed plate attached to a shaft, offering an efficient alternative to traditional impellers. Studies by Lo et al. (Lo et al., 1998) have shown that the amplitude and frequency of oscillations in vibromixers significantly influence drop size distributions, with higher oscillation intensities leading to smaller drop sizes and enhanced mixing efficiency. In their experiments, distilled water and pure xylene served as the continuous and dispersed phases, respectively, while the device operated at oscillation amplitudes of 0.005 to 0.035 meters and frequencies ranging from 0.67 to 33.3 Hz. Findings indicated that as both amplitude and frequency increased, the drop size distribution shifted toward smaller diameters, with the Sauter mean diameter (Eq 1.12). stabilizing after about 30 minutes under specific operating conditions (frequency $f = 4.5$ Hz and amplitude $A = 0.225$ m).

Similarly, Kamiński and Wójtowicz (Kamiński and Wójtowicz, 2003) conducted research into liquid-liquid mixing within a vibromixer setup, where amplitudes and frequencies ranged from 0.02 to 0.05 meters and 0.5 to 3 Hz, respectively. The vibromixer featured a plate diameter of 0.71 to 0.91 times the tank

diameter, an off-bottom clearance of 0.5 times the tank diameter, and a liquid height equal to the tank diameter. Through these experiments, it was observed that increased mixing time decreased drop diameters until a stable size was reached within 5 to 7 minutes. Additionally, the dispersed phase drop size was inversely correlated with oscillation frequency, highlighting that plate diameter and oscillation frequency significantly influence the final droplet sizes, where larger plates at higher frequencies produced finer dispersions.

Although the potential and industrial applicability of vibromixers, their performance, characterization, and design remain relatively underexplored in the current literature. One notable example is the FUNDAMIX[®] vibromixer, produced by DrM[®], Dr. Mueller AG, Switzerland, which has demonstrated significant promise in process intensification, particularly in biodiesel synthesis applications. Orlewski et al. (Orlewski et al., 2018) expanded upon previous studies by examining the impact of geometrical parameters of the FUNDAMIX[®] vibromixer on mass transfer rates in a chemical reaction, demonstrating that increased vibration amplitudes and larger plate sizes enhance mass transfer, effectively reducing mixing times and increasing mixing intensity.

The primary objectives of this chapter are to examine the potential of the vibromixer in enhancing liquid-liquid contact and mass transfer rates to intensify the transesterification reaction in biodiesel production. To provide further quantitative insights into the mixing system, Computational Fluid Dynamics (CFD) simulations. focus on parameters such as oscillation amplitude and plate orientation to capture

detailed flow patterns, local velocity fields, and shear rates—information that remains challenging to obtain through experimental techniques alone. Notably, CFD studies on vibromixing at the high frequencies used in this chapter are limited, making this analysis a significant contribution to understanding vibromixing under conditions applicable to biodiesel production.

The simulations conducted are divided into two primary sections. The first section evaluates biodiesel production considered as *pseudo-single-phase* system, analyzing the impact of vibromixer operation on fluid motion and component dispersion. The second section examines briefly the behavior of solid-liquid suspensions, focusing on the height of the suspension cloud generated by fluid movement within the vibromixer. These analyses aim to elucidate the mechanisms by which vibromixing affects mass transfer and fluid dynamics, offering insights into the practical applications and scalability of vibromixer technology across various industrial mixing and chemical processes. This chapter thus contributes to the broader field of process intensification, providing foundational data for further development of vibromixer-based systems in industrial applications.

4.3 Materials and Methods

Vibromixers have been shown to be effective in achieving efficient and thorough mixing across a range of applications, including liquid reactions, shear-sensitive systems, liquid-liquid dispersion, and solid-liquid suspensions. Several potential advantages are offered by these mixers, such as the mitigation of vortex formation and consequent foam production, the promotion of uniform mixture

distribution due to stable flow patterns within the reactor, the elimination of the need for baffles, and their inherent simplicity, durability, and minimal maintenance requirements.

The integration of Computational Fluid Dynamics (CFD) into vibromixing has facilitated a clearer understanding of the complex fluid dynamics within these systems. Through the employment of CFD simulations, deeper insights into the flow patterns and velocity distributions produced by vibromixers have been obtained. This approach has enhanced the comprehension of fluid behavior, enabling the potential for optimized mixing processes.

The simulations were conducted using ANSYS® *Fluent* V23 R2 software, focusing on modeling the fluid motion within the agitated vessel.

4.3.1 Operating principle of vibromixers

Vibromixers generate vibrational motion through two distinct motor types, as shown in Figure 4.1:

- The first type, driven by an electric motor connected to a flywheel and crankshaft with specified parameters, such as the length of the connecting rod and the dimensionless radial location. This configuration typically exhibits a wider range of center-to-peak amplitude (A) and operates at lower frequencies (f).
- The second type employs an electromagnetic field, generally achieving lower amplitudes and higher frequencies.

These vibromixers can incorporate various types of plates, including non-perforated and perforated configurations, with perforated plates further distinguished by the shape of their holes. The vibrations generated by the motors are transmitted through a shaft to a reciprocating plate, positioned centrally within the vessel with a defined clearance from the walls. The plate's off-bottom clearance is adjustable, impacting the mixing process significantly (Wójtowicz, 2017).

This study specifically examines vibromixers with plates with conical holes operating at high frequencies and low amplitudes, focusing on their mixing mechanisms and advantages, particularly in achieving uniform mixing and process intensification. The orientation of these plates—either *up* or *down pumping*—affects fluid dynamics and recirculation patterns, with potential applications in homogenization, emulsification, sedimentation prevention, and gas dispersion.

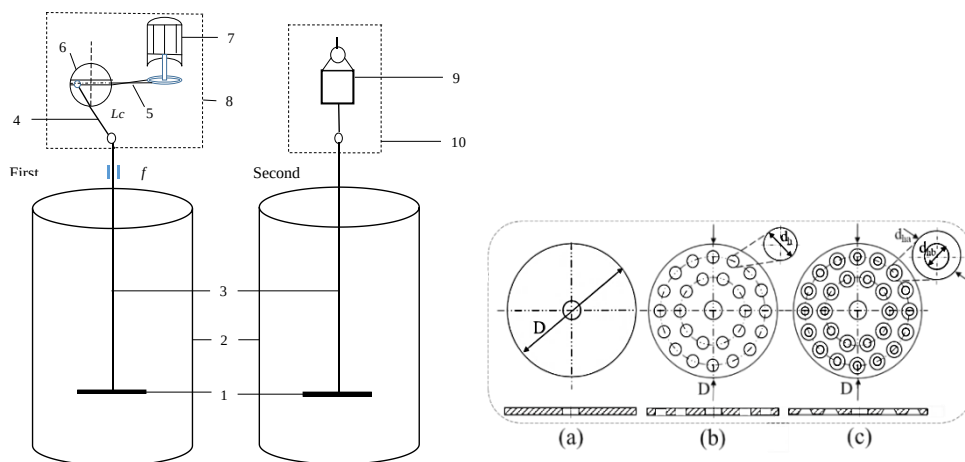


Figure 4.1: Vibromixer set up for type 1 and 2 configurations: (1) disk (plate); (2) vessel; (3) shaft or vibrating-rod; (4) connecting-rod or variable gear (connecting-rod length, L_c); (5) V-belt transmission, (6) flywheel; (7) electric motor; (8) mechanical driving unit; (9) electromagnetic vibrator; (10) electromagnetic driving unit. Right different type of plates; (a): solid plate, (b): perforated plate with cylindrical holes, and (c) perforated plate with conical holes.

In multiphase applications, processes such as breakup, collision, and coalescence influence the interfacial area and mass transfer efficiency within the perforated plates. A schematic of the vibromixer operation, along with various disk types utilized, is illustrated in Figure 4.2. Additionally, the device offers two distinct mixing types depending on the orientation of its conical holes.

Mixer Plate Type A, commonly called *up-pumping configuration*, with conical holes facing upward, is considered the standard choice due to its enhanced efficiency, which promotes an upward flow that permeates the entire vessel. This configuration enables the plate to be positioned very low within the vessel, thereby minimizing the mixable heel volume and optimizing operational effectiveness.

Alternatively, *Mixer Plate Type B*, commonly called *down-pumping configuration*, designed with downward-facing conical holes, is employed for products susceptible to sedimentation or foaming, or when avoiding air introduction during mixing is required. This plate is positioned at a clearance from the vessel's bottom approximately equal to its diameter.

In applications requiring gas dispersion, *Mixer Plate Type B enables gas injection* through the shaft, allowing downward liquid flow to disperse fine gas bubbles within the vessel, resulting in effective gas dispersion. An alternative method injects gas between two plates, which disrupts bubble formation, thus enhancing mixing efficacy.

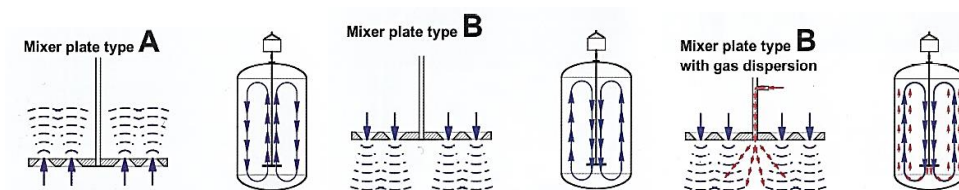


Figure 4.2: Pictures of plate hole orientations (a) upward (b) downward and the corresponding fluid recirculation pattern in a Fundamix vibromixer <https://drmgroup.com/fundamix>.

4.3.2 CFD Analysis of vibromixers technology

CFD enables the simulation of fluid flow and heat or mass transfer phenomena by solving the fundamental mathematical equations that govern these processes. Through CFD analysis, flow patterns, velocity distributions, and temperature profiles within mixing equipment can be accurately predicted and visualized. By establishing suitable boundary conditions and defining the system geometry, CFD serves as a critical tool for engineers and scientists in the optimization of mixing processes, enhancement of efficiency, and assurance of product quality. The following paragraph presents the CFD methodology used for modeling mixing processes within the context of vibromixer technology, specifying the approach, assumptions, and simulation parameters required to achieve reliable analysis and results.

4.3.3 Problem Statement

This study aims to investigate liquid-liquid and solid-liquid mixing through a comprehensive computational campaign. This was accomplished by conducting

two types of simulations: the first focusing on biodiesel production (within a single-phase environment), and the second involving an assessment of solid-liquid mixing, with particular attention to the evaluation of cloud height, a parameter that distinguishes effective suspension in a solid-liquid system.

- The vibromixer's potential to enhance the transesterification reaction in biodiesel production is substantial, as this process technology effectively facilitates liquid-liquid dispersions but remains relatively unexplored in the scientific literature. In this study, experimental investigations into the transesterification reaction are supported by flow simulations, which assist in understanding the hydrodynamics and flow behavior within the vibromixer system.
- Solid-liquid mixing holds critical importance in the chemical industry, finding applications in various unit operations such as the dispersion of solid particles, leaching and dissolution processes, crystallization, precipitation, adsorption and desorption, solid-catalyzed reactions, ion exchange, suspension polymerization, and activated sludge processing. This study focuses on assessing the vibromixer's performance in solid particle suspension, specifically by examining how suspension is formed.

Preliminary simulations were carried out to establish the optimal computational representation for the simulation domain. Figure 4.3 illustrates the geometries utilized for biodiesel production, along with a table of their geometric characteristics (Table 4.1).

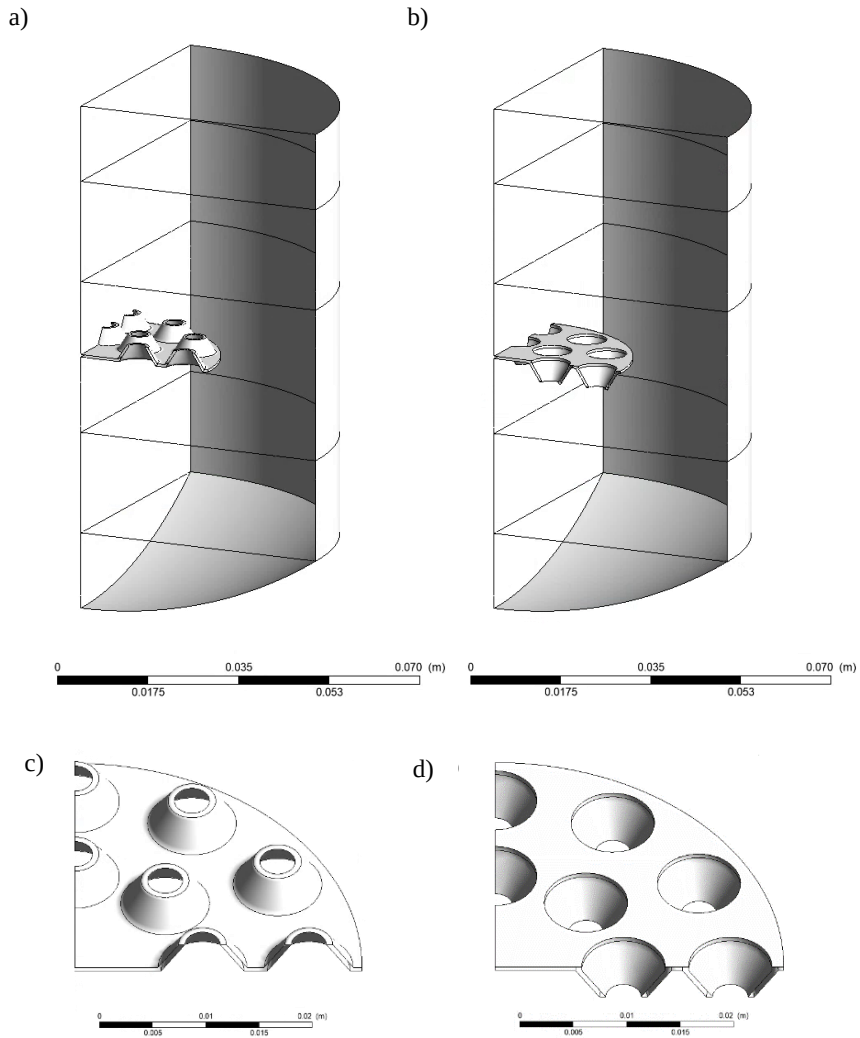


Figure 4.3: Computational domain for simulations concerning biodiesel production: a) Up-pumping configuration (off-bottom clearance of $0.50 H$); b) Down-pumping configuration (off-bottom clearance of $0.50 H$); c) detailed view of the vibrating disk of up-pumping configuration; d) detailed view of the vibrating disk of down-pumping configuration.

Table 4.1: *Geometric data related to fluid dynamics simulations.*

Variable	Symbol	Unit	Value
Height of the liquid	H	mm	100
Diameter of vessel	D	mm	100
Off-bottom clearance	h	mm	0.50 H
Disk size	d	mm	54
Amplitude	λ	mm	1.5*, 3**, 3.5 * for down-pumping configuration ** for solid-liquid suspension analysis
Frequency	f	Hz	100
Number of conical holes	n	-	20 [#] *for the entire domain
Diameter of holes	dh	mm	9 ^{##} and 4.5 ^{###} ##largest and ###smallest diameter of the conical hole
Disk thickness	t	mm	0.5

4.3.4 Computational domain and Mesh

After selecting the resolving setup, as detailed in the following paragraph, four different meshes were created for the two simulated cases. Unstructured tetrahedral meshes were generated across the entire domain to capture the unique geometry of the conical sections of the vibrating disk that define the system. These meshes incorporated inflation layers near the walls, with a transition ratio of 0.272, a maximum of 5 layers, and a growth rate of 1.2. To support the dynamic mesh functionality, a voxel layer was implemented near the moving zone to allow proper deformation of cells during motion, ensuring the volume of modified cells remains above zero.

A grid independence study was performed to confirm the accuracy and reliability of the simulation results. This involved preliminary simulations using four meshes with 2504083, 1191187, 819490 and 629371 cells, respectively, aimed at identifying the optimal mesh size to balance computational cost with accuracy. The meshes were evaluated based on critical numerical parameters, such as velocity profiles, pressure distributions, and dimensionless metrics like the Newton and Circulation numbers.

Upon careful review of the simulation outcomes, it was determined that increasing the mesh size beyond approximately 850000 cells did not significantly affect the numerical results, indicating that grid independence had been achieved. Therefore, a mesh size of approximately 800000 cells was chosen for both up-pumping and down-pumping configurations, resulting in a computational domain with 819490 cells for the up-pumping case and 830332 cells for the down-pumping liquid motion. Further information is provided in Appendix B.

Polyhedral grids with a comparable cell count were also evaluated for performance comparison. However, it was observed that polyhedral meshes offered no advantage over tetrahedral meshes in terms of accuracy or computational efficiency. As a result, unstructured tetrahedral meshes were preferred for their superior handling of the complex geometry and dynamic movement involved.

Figure 4.4 illustrates the meshes used in simulations for biodiesel production and solid-liquid case evaluation. Additionally, the polyhedral mesh was examined for simulations of the solid-liquid system using the Discrete Element Method

(DEM), as shown in the Figure 4.5.

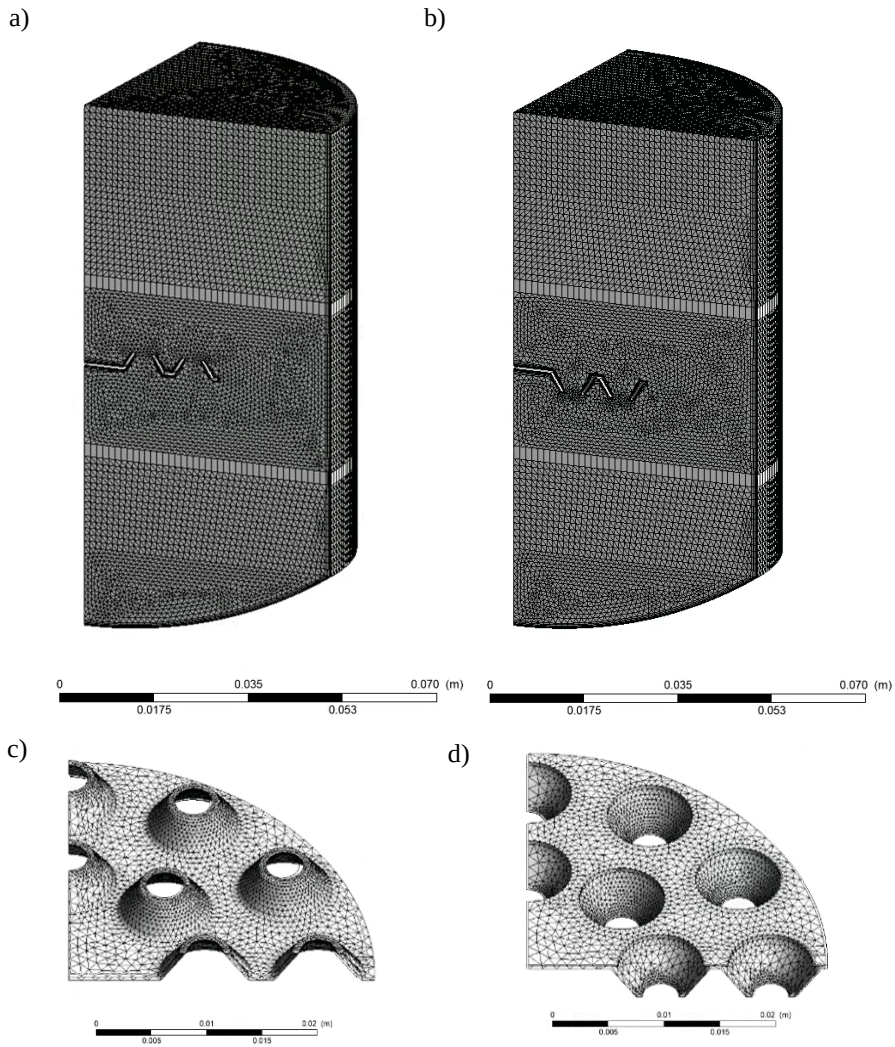


Figure 4.4: Meshes for simulations concerning biodiesel production: a) Mesh for up-pumping configuration vibromixer (819490 voxels); b) Mesh for down-pumping configuration vibromixer (830 332 voxels); c) detailed mesh of the vibrating disk for up-pumping configuration vibromixer; d) Detail of the mesh of the vibrating disk for down-pumping configuration vibromixer.

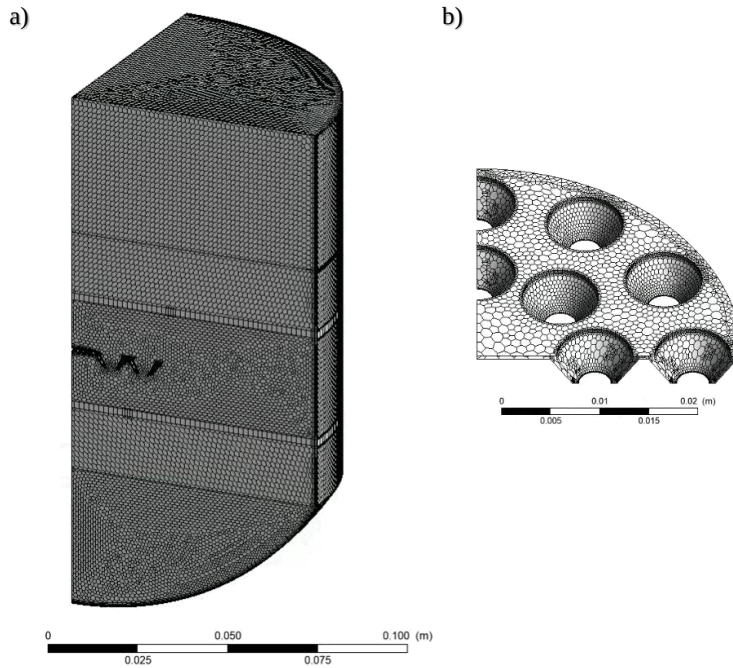


Figure 4.5: Polyhedral mesh for simulations concerning solid-liquid suspension: a) Mesh off-bottom clearance of $0.50 H$ (394758 voxels); b) Detail of the mesh of the vibrating disk.

4.3.5 Numerical Methods

In computational fluid dynamics (CFD), the accurate capture of intricate fluid flow behavior depends critically on the selection and implementation of numerical methods. Precision is achieved by employing double precision for solver options and enhancing computational efficiency through parallel processing on local machines. This approach is further strengthened by the utilization of 12 to 16 concurrent processes.

4.3.5.1 Law of vibration of the disk

The primary challenge in these simulations has been to achieve periodic motion of a perforated disk at a high frequency of 100 Hz. To achieve this, various methods were evaluated, including an overset mesh with motion applied to a portion of the mesh overlaid on a structured grid, or a dynamic mesh approach. Consistent results that align with real-world phenomena were obtained using the latter method.

In this setup, the computational domain is divided into five distinct zones: two static zones positioned at the ends of the vessel, two mesh deformation zones adjacent to the disk zone, and a central zone enclosing the disk, which undergoes rigid body motion governed by the equations described below (Eq. 4.4). Notably, it is possible to input a translational velocity law directly into the software. The parameters for this motion include amplitudes A of 1.5, 3, and 3.5 mm with a frequency f of 100 Hz, dependent on the specific simulation considered.

$$x(t) = A * \sin(2 * \pi * f * t) \quad (4.1)$$

$$v(t) = \frac{d}{dt}x(t) \quad (4.2)$$

$$\frac{d}{dt}(A * \cos(2 * \pi * f * time)) = 2 * \pi * A * f * \cos(2 * \pi * f * t) \quad (4.3)$$

$$v(t) = (-)2 * \pi * A * f * \cos(2 * \pi * f * t) \quad (4.4)$$

Regarding the choice of dynamic mesh: for the two deformation zones, the smoothing (diffusion) mesh method was evaluated according to the standard description.

4.3.5.2 Turbulence Model

For the vibromixing simulation, the selected k - ω SST (Shear-Stress Transport) *turbulence model* is regarded as a robust and versatile choice, well-suited for accurately predicting a broad range of turbulent flow phenomena commonly encountered in engineering applications.

The k - ω SST model integrates features from both the k - ε and k - ω models, providing a balanced approach that addresses various flow regimes effectively. Differential equations for both turbulent kinetic energy k and specific turbulence dissipation rate ω are utilized, enabling this model to capture turbulent behaviors in boundary layers, shear layers, and separated flows. One of the principal advantages of the k - ω SST model is its capability to transition smoothly between near-wall and far-field regions, making it particularly suitable for simulating the intricate fluid dynamics inherent in vibromixing processes.

This adaptability facilitates accurate predictions of turbulent quantities, such as eddy viscosity, turbulent kinetic energy, and dissipation rate, which are essential for representing the complex flow phenomena associated with the vibromixer's operation. The transport equations for the SST k - ω model are:

$$\frac{\partial}{\partial t}(\rho k) + \frac{\partial}{\partial x_i}(\rho k v_i) = \frac{\partial}{\partial x_i} \left(\Gamma_k \frac{\partial k}{\partial x_j} \right) + \widetilde{G}_k - Y_k \quad (4.5)$$

$$\frac{\partial}{\partial t}(\rho \omega) + \frac{\partial}{\partial x_i}(\rho \omega v_i) = \frac{\partial}{\partial x_i} \left(\Gamma_\omega \frac{\partial \omega}{\partial x_j} \right) + G_\omega - Y_\omega + D_\omega \quad (4.6)$$

In these equations, $\widetilde{G}_k$ represents the generation of turbulence kinetic energy due to mean velocity gradients. G_ω represents the generation of ω . Γ_k and Γ_ω represent the

effective diffusivity of k and ω , respectively. Y_k and Y_ω represent the dissipation of k and ω , due to turbulence and D_ω represents the cross-diffusion term.

For simulations where the flow regime falls into the transitional zone, such as when the oscillation amplitude of the disk reduces to 1.5 mm, leading to a decrease in the Reynolds number, a second turbulence hybrid model called SBES (Stress-Blended Eddy Simulation) has been evaluated. SBES is a hybrid turbulence modeling approach that combines elements of Reynolds-Averaged Navier-Stokes (RANS) models and Large Eddy Simulation (LES).

The information regarding the current regime will be validated in due course, once the PIV data enables a more comprehensive study to validate the results presented in this thesis.

In this approach, the simulation dynamically switches between RANS and LES based on the local flow conditions. RANS is used in regions where turbulence is relatively well-behaved and resolved by the grid, while LES is used in regions where turbulence is highly unsteady and requires more detailed modeling.

The comparison of the two simulations using the two different turbulence models will be shown in the following paragraphs.

4.3.5.3 Materials

The two types of simulations involve distinct fluid properties: for the biodiesel production simulation, a fluid characterized by a mixture density of 842 kg m^{-3} and a relative viscosity of $0.0041 \text{ Pa}\cdot\text{s}$ is employed.

In the case of the multiphase simulation, water, with a density of 998.2 kg m^{-3} and a viscosity of $0.001003 \text{ Pa}\cdot\text{s}$, is selected as one phase, while a second phase represents granular material, specifically glass beads, with a density of 2220 kg m^{-3} . This granular phase is initially deposited with a bed void fraction of 0.39 at the bottom, allowing for the simulation of two immiscible Eulerian phases.

For the granular phase properties, the Syamlal-O'Brien model is applied to determine granular viscosity, and the Moraga formulation is adopted for the lift coefficient. Regarding the simulation conducted using the DEM method, 50 elements were introduced into the vessel below the impeller following a pyramidal arrangement, and their motion was analyzed during the transient phase.

4.3.5.4 Boundary conditions

In the field of computational simulations, the establishment of precise boundary conditions is essential to achieve an accurate representation and insightful analysis of the phenomena occurring within a vibromixer. The simulations leveraged the system's inherent symmetry to reduce computational demand.

The walls were modeled as moving walls, set to a zero-velocity relative to the absolute reference frame, in alignment with the manual's recommendations and employing the standard setup for roughness modeling. For the uppermost wall within the computational domain, which represents the free surface, a specified shear stress of 0 Pa was applied across all three components.

4.3.5.5 Methods and times resolution

Only transient simulations were performed. Within the "Pressure-Velocity Coupling" scheme, the SIMPLEC method was applied. For the flux type, the Rhie-Chow approach, momentum-based, was implemented. Spatial discretization followed a Gradient Least Square Cell-Based scheme. Momentum, pressure, turbulent kinetic energy, and specific dissipation rate were each discretized to the second order. Additionally, momentum was discretized using the Second Order Upwind method.

The transient formulation employed a First Order Implicit approach. For the calculation run, time advancement was configured as a fixed type using a frequency-based method. The specified parameters included a frequency of 100 Hz, 125 time steps per period, a time step size of 8×10^{-5} seconds, and a maximum of 15 iterations per time step.

Each simulation was conducted over an approximate duration of two to three seconds, which was sufficient to attain a steady-state condition.

4.3.5.6 Dimensionless numbers

To characterize the flow induced by the vibromixer, it is essential to define the Reynolds number specific to this flow. This requires selecting appropriate length and velocity scales that effectively capture the vibromixer dynamics. Given the time-dependent oscillation of the disk's velocity and the presence of multiple length scales depending on disk size and perforation, several options for these scales are available.

Determining the Reynolds number presents a challenge, as various formulations have been proposed in the literature. Based on experimental analysis, the most reliable formulation is attributed to Orlewsky et al., 2018 (Orlewski et al., 2018).

$$R_e = \frac{v D_H}{\nu} = \frac{f A (d^2 - n d_2^2)}{D + d + n d_1} \quad (4.7)$$

Where D is the diameter of the reactor vessel, measured in meters; d the diameter of the disk-shaped component within the system; d_1 the major diameter of the conical holes; d_2 the minor diameter of the conical holes; n the total number of holes and ν the cinematic viscosity [$\text{m}^2 \text{s}^{-1}$], expressed in square meters per second.

Another essential parameter to calculate is the energy consumed during mixing, as well as the related dimensionless Newton Number. To determine the power consumption in a vibromixer, it is necessary to measure the force exerted on the disk and to have data on either the maximum or mean velocity of the disk or fluid flow. The force acting on the reciprocating disk during its vibrations is typically measured with piezoelectric sensors or transducers, inductive transducers, or strain gauge sensors. The mean power input was calculated as the time integral of the product of the total force acting on the disk, $F(t)$, and the instantaneous disk velocity, $v(t)$.

$$\bar{P} = -f \int_0^{1/f} F(t) v(t) dt \quad (4.8)$$

To calculate the value of this integral, numerical integration of the force acting on the disk over time was performed, multiplied by the disk velocity, using the 3/8 Simpson's rule as follows.

$$\int_0^{1/f} F(t) v(t) dt \approx \frac{3}{8} h \left[F(t_0) v(t_0) + 3 \sum_{i=1,3 \neq i}^{n-1} F(t_i) v(t_i) + 2 \sum_{i=1}^{\frac{n}{3}-1} F(t_i) v(t_i) + F(t_n) v(t_n) \right] \quad (4.9)$$

Given the dissipated power is known, the Newton Number can be calculated.

$$Ne_v = \frac{\bar{P}}{\bar{w}^3 D^2 \rho} \quad (4.10)$$

with the mean disk velocity $\bar{w} = 4Af$.

In the evaluation of vibromixing, assessing the flow number and circulation number is crucial for understanding the fluid dynamics and mixing efficiency within the system. The flow number, generally defined as the ratio of the characteristic velocity of the mixing element to the mean velocity of the fluid, provides insight into the overall flow pattern generated by the mixer. At the same time, the circulation number, obtained from the ratio of the circulation velocity to the characteristic velocity, serves as a metric for fluid recirculation and mixing effectiveness.

By analyzing these parameters, researchers can evaluate the performance of vibromixing setups and refine operating conditions to improve mixing efficiency and achieve the desired product homogeneity. For vibromixers, the Flow Number N_Q and the Circulation Number N_{Q_T} are defined as:

$$N_Q = \frac{\int_S \vec{v} \cdot \hat{n} dS}{f D^3} = \frac{Q}{f D^3} \quad (4.11)$$

$$N_{Q_T} = \frac{\int_{S_T} \vec{v} \cdot \hat{n} dS_T}{f D^3} = \frac{Q_T}{f D^3} \quad (4.12)$$

The considered surface for the evaluation of flow number is depicted in Figure 4.6.

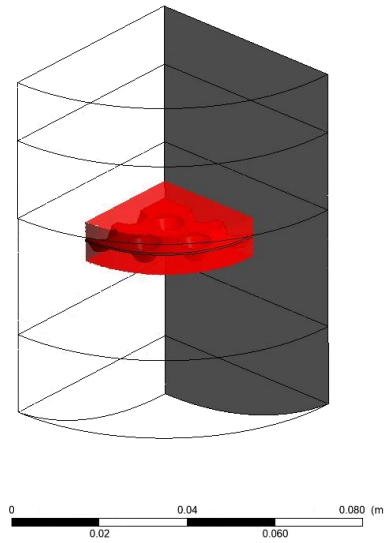


Figure 4.6: Surfaces considered in the evaluation of a) Flow Number and b) Circulation Number.

4.3.5.7 Shear rate analysis in vibromixers

In evaluating the performance of vibromixers, an important parameter to assess is the shear rate within the mixing vessel. The shear rate $|\dot{\gamma}|$ offers insight into the deformation rate of fluid elements, which is crucial for understanding mixing efficiency and evaluating the likelihood of shear-induced reactions or potential

damage to sensitive materials. In a three-dimensional flow field, the shear rate can be calculated using the following formula:

$$|\dot{\gamma}| = \sqrt{\left(\frac{\partial v_1}{\partial x}\right)^2 + \left(\frac{\partial v_2}{\partial y}\right)^2 + \left(\frac{\partial v_3}{\partial z}\right)^2 + 2 * \left[\left(\frac{1}{2}\left(\frac{\partial v_1}{\partial y} + \frac{\partial v_2}{\partial x}\right)\right)^2 + \left(\frac{1}{2}\left(\frac{\partial v_1}{\partial z} + \frac{\partial v_3}{\partial x}\right)\right)^2 + \left(\frac{1}{2}\left(\frac{\partial v_2}{\partial z} + \frac{\partial v_3}{\partial y}\right)\right)^2\right]} \quad (4.13)$$

In this equation, $\frac{\partial v_1}{\partial x}$, $\frac{\partial v_2}{\partial y}$, and $\frac{\partial v_3}{\partial z}$ represent the normal components of the velocity gradient tensor, corresponding to the rate of change of the velocity components v_1 , v_2 , and v_3 in the x , y and z directions, respectively. The terms within the brackets account for the shear components of the velocity gradient tensor, representing the rate of angular deformation. These shear components are expressed by the symmetric combinations of the velocity gradients, such as $\frac{\partial v_1}{\partial y} + \frac{\partial v_2}{\partial x}$.

By calculating the shear rate with this comprehensive formula, the local shear conditions within the vibromixer can be accurately assessed. This information is crucial for optimizing the design and operational parameters of the mixer to ensure efficient and effective mixing while also preventing excessive shear that could damage sensitive materials or compromise the quality of the final product.

4.4 Results and discussion

The results of vibromixing simulations offer a comprehensive view of process dynamics, emphasizing the effectiveness and unique features of this technique in fluid homogenization. By conducting a thorough analysis of the collected data, the system's performance can be evaluated in terms of mixing quality,

particle distribution, and other critical parameters. In the following paragraph, the results of velocity profiles over time are presented, highlighting key structures and practical implications for applying vibromixing at both laboratory and industrial scales.

4.4.1 Biodiesel production

In the context of biodiesel production, several computational fluid dynamics simulations were conducted to thoroughly investigate the various flow patterns within a vibromixer. All simulations performed were URANS (Unsteady-Reynolds-Averaged Navier–Stokes) simulations. Successive time steps of several physical quantities characterizing the system—such as velocity normalized by v_{tip} , pressure, and vector plots—are presented. Additionally, different operating configurations are compared.

The first comparison focuses on the up-pumping and down-pumping configurations of the vibromixers, both operating with a 54 mm disk, a frequency of 100 Hz, and an amplitude of 3.5 mm. In Figure 4.7 and Figure 4.8 the flow pattern and vector plots at various time steps are shown for up-pumping configuration. Distinct toroidal vortex zones can be observed: the first vortex, of greater magnitude, appears near the free surface, while the second, smaller vortex forms near the bottom of the reactor. As anticipated, the highest velocity zone is located near the perforated disk, particularly around the conical holes. In the pressure color map, shown in Figure 4.11, two distinct zones of high and low pressure are observed, varying temporally with the direction of disk movement.

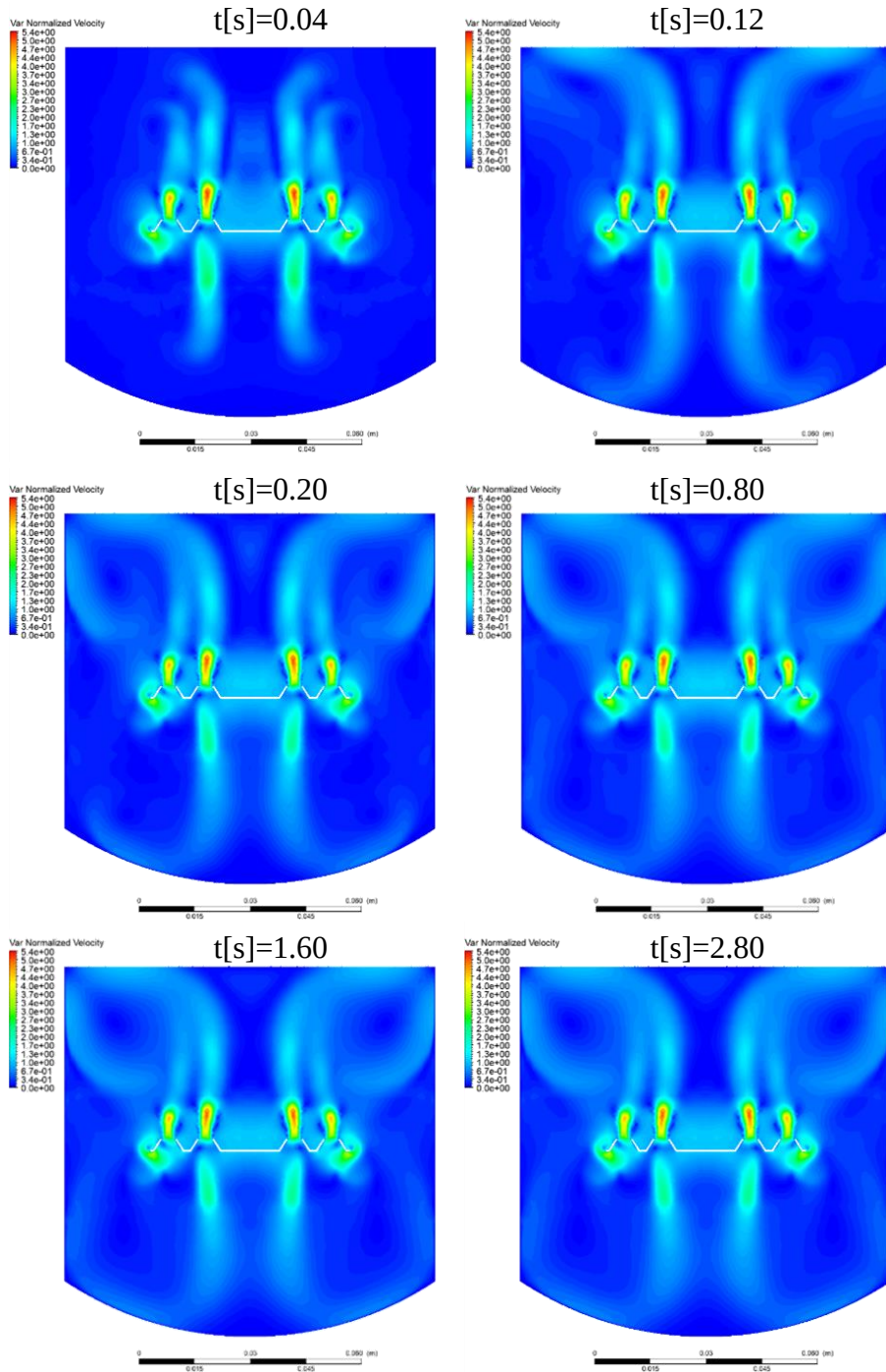


Figure 4.7: Colour maps of velocity magnitude, normalized by maximum disk velocities $w_{\max}=2\pi Af$, for an up-pumping configuration vibromixer at different time steps. Off-bottom clearance is $0.50 H$, frequency is 100 Hz , and amplitude is 3.5 mm .

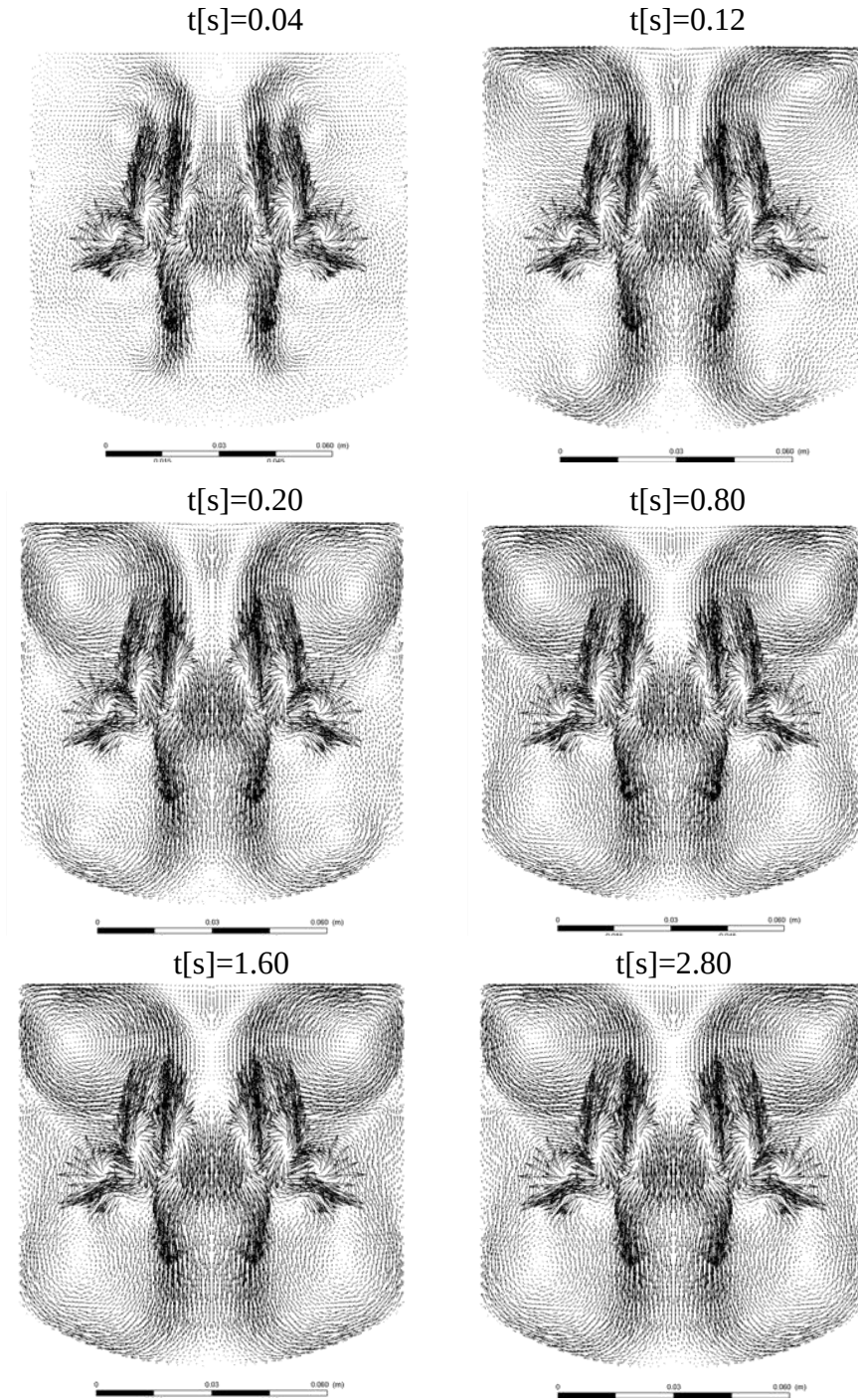


Figure 4.8: Velocity vectors in an up-pumping configuration vibromixer at different time steps. Off-bottom clearance is equal to $0.50 H$, frequency is 100 Hz and amplitude is 3.5 mm.

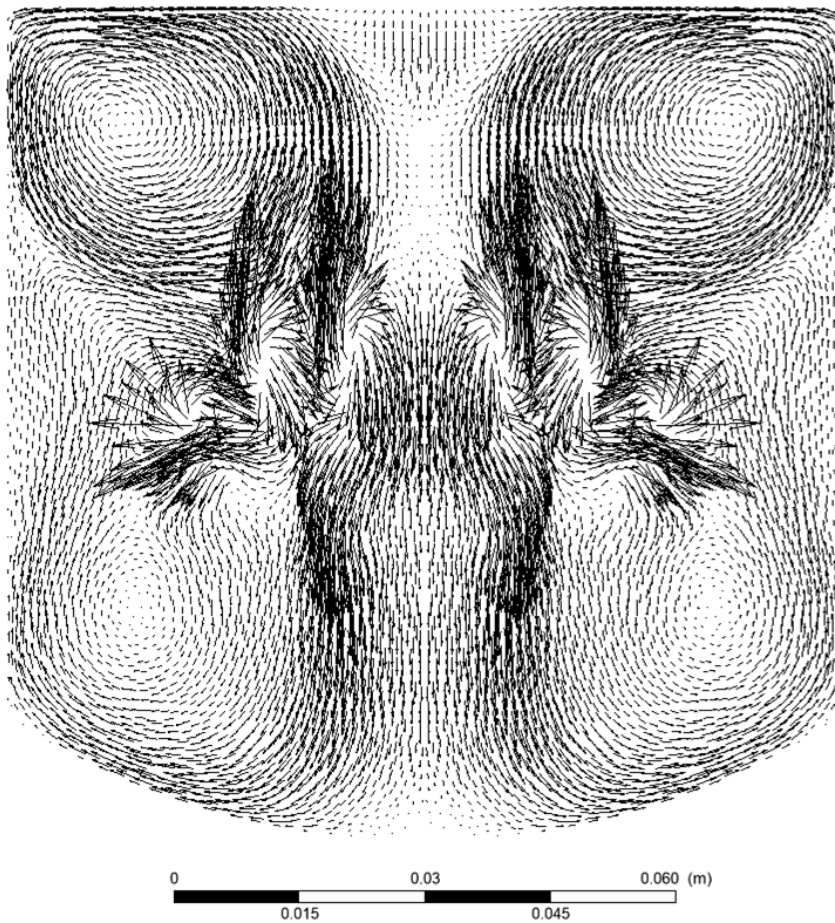


Figure 4.8.bis: Velocity vectors representing the mean velocity magnitude for an up-pumping configuration vibromixer. Off-bottom clearance is $0.50 H$, frequency is 100 Hz, and amplitude is 3.5 mm.

Similarly, Figure 4.9 and Figure 4.11 present the flow pattern and pressure color map at various time instances for the down-pumping vibromixer configuration. In this setup, two distinct toroidal vortex zones are also evident: one near the free surface and another near the bottom of the reactor, with the latter displaying greater magnitude, as confirmed by the vector plot in Figure 4.10.

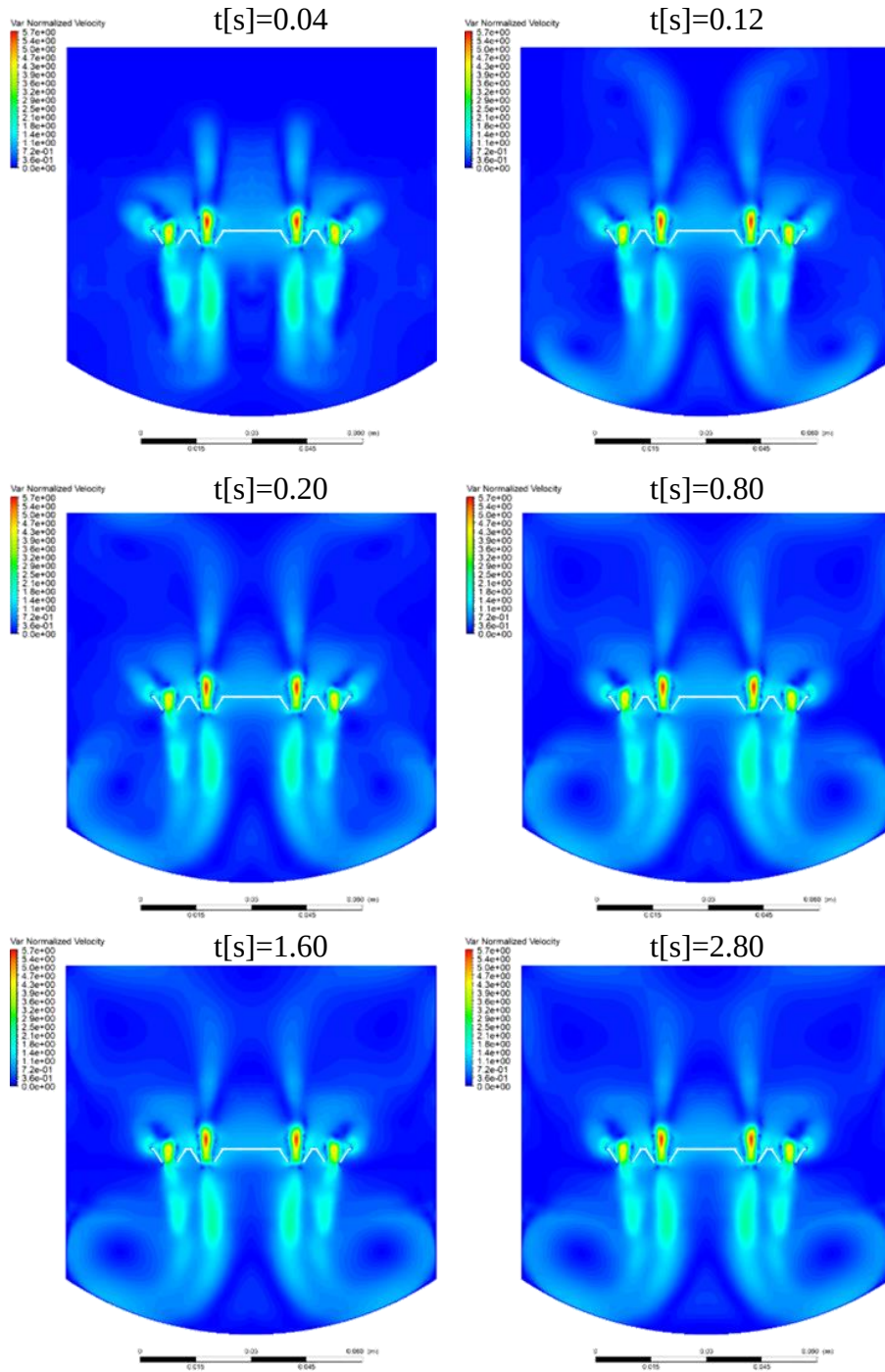


Figure 4.9: Colour maps of velocity magnitude, normalized by maximum disk velocities $w_{max}=2\pi Af$, for a down-pumping configuration vibromixer at different time steps. Off-bottom clearance is $0.50 H$, frequency is 100 Hz , and amplitude is 3.5 mm .

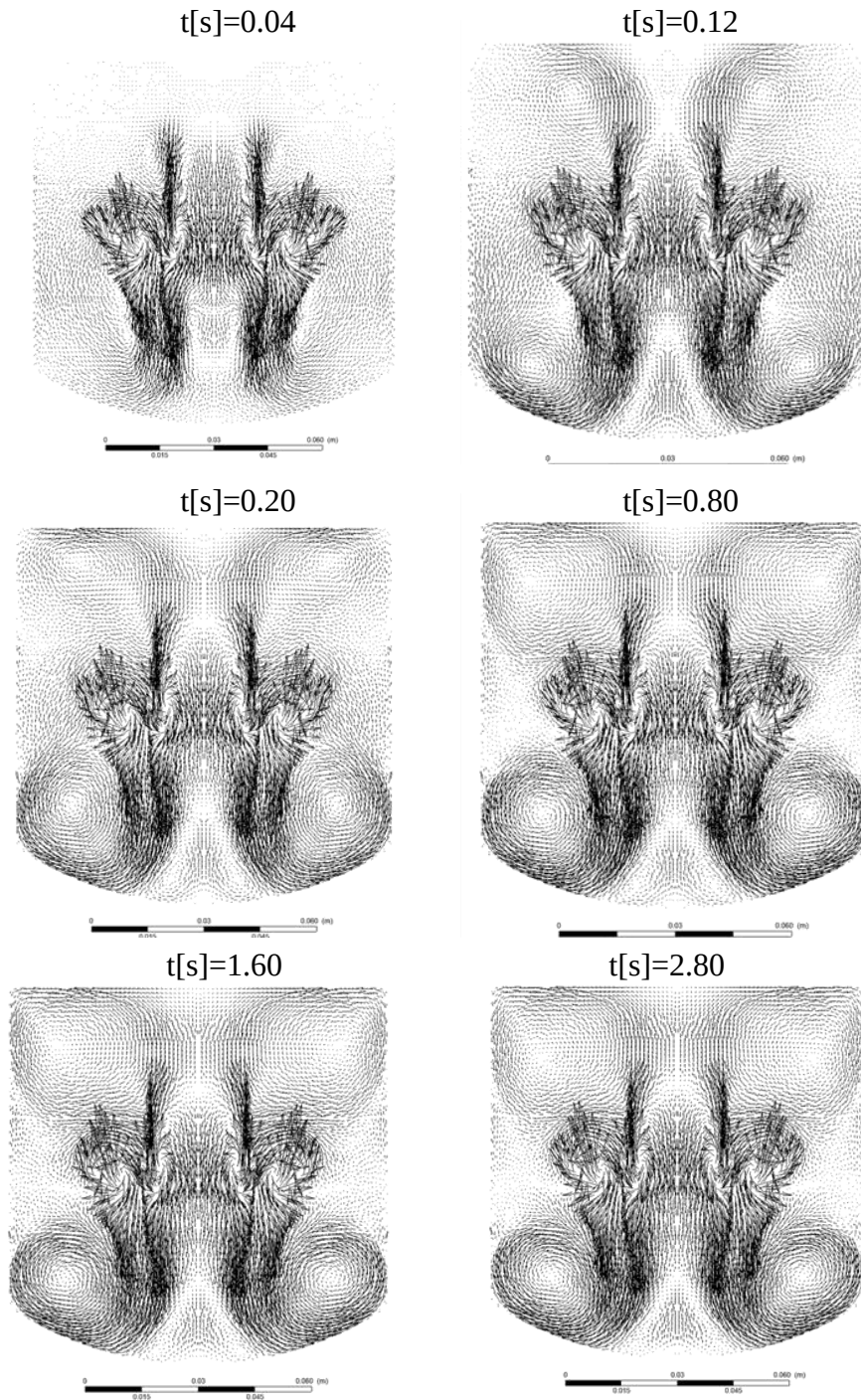


Figure 4.10: Velocity vectors in a down-pumping configuration vibromixer at different time steps. Off-bottom clearance is equal to $0.50 H$, frequency is 100 Hz and amplitude is 3.5 mm .

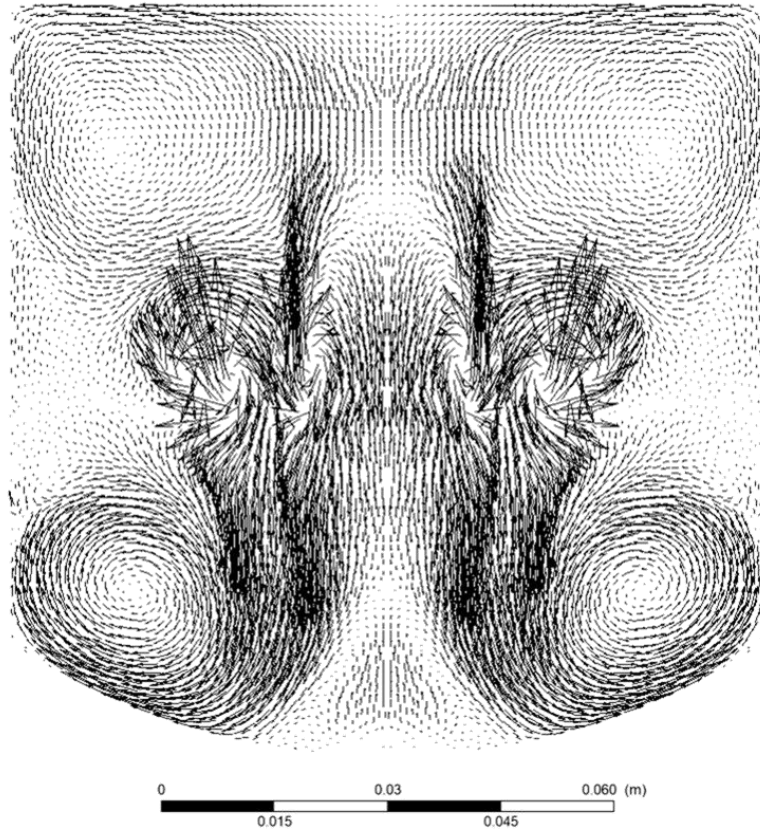


Figure 4.10.bis: *Velocity vectors representing the mean velocity magnitude for a down-pumping configuration vibromixer. Off-bottom clearance is $0.50 H$, frequency is 100 Hz, and amplitude is 3.5 mm.*

To enable an effective comparison of the two simulations, Figure 4.12 provides color maps of the time-averaged velocity profile, where the previously mentioned recirculation patterns can be observed. Contrary to the manufacturer's claims, a single global circulation with up-pumping across the entire liquid volume is not established for the up-pumping vibromixer configuration, neither is a global down-pumping circulation achieved for the down-pumping vibromixer configuration.

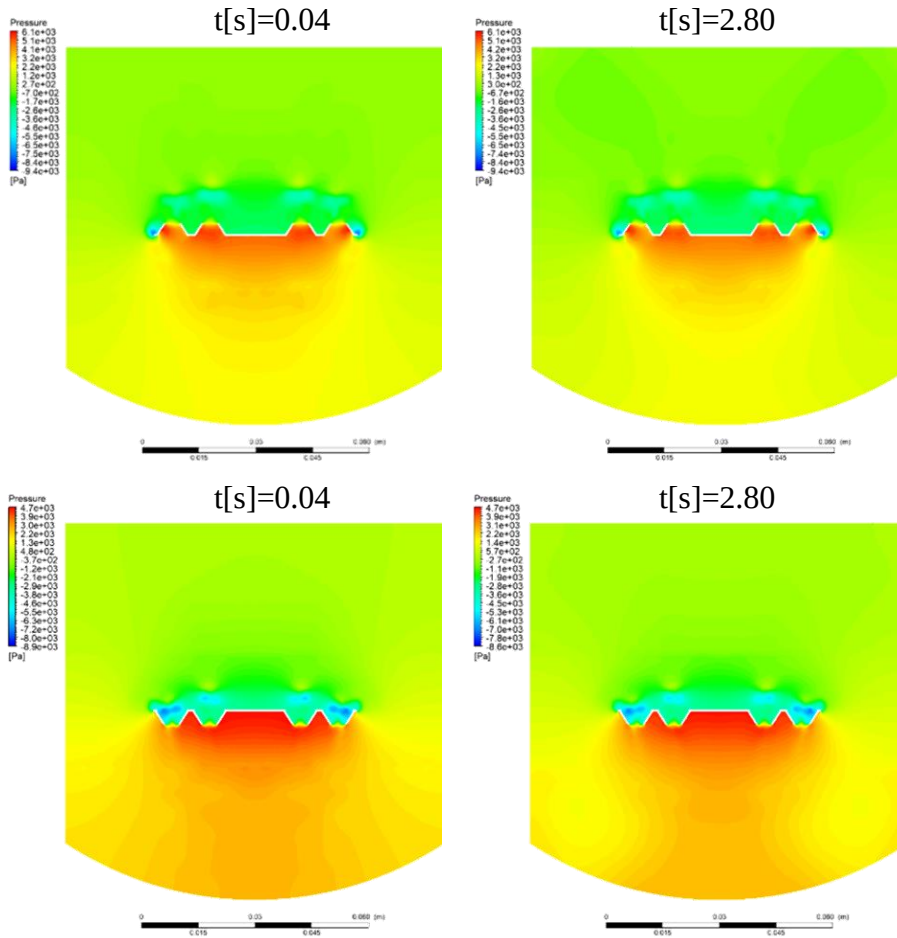


Figure 4.11: Color maps of pressure for an up-pumping configuration vibromixer (first row) and a down-pumping configuration vibromixer (second row) at different time steps. Off-bottom clearance is $0.50 H$, frequency is 100 Hz , and amplitude is 3.5 mm .

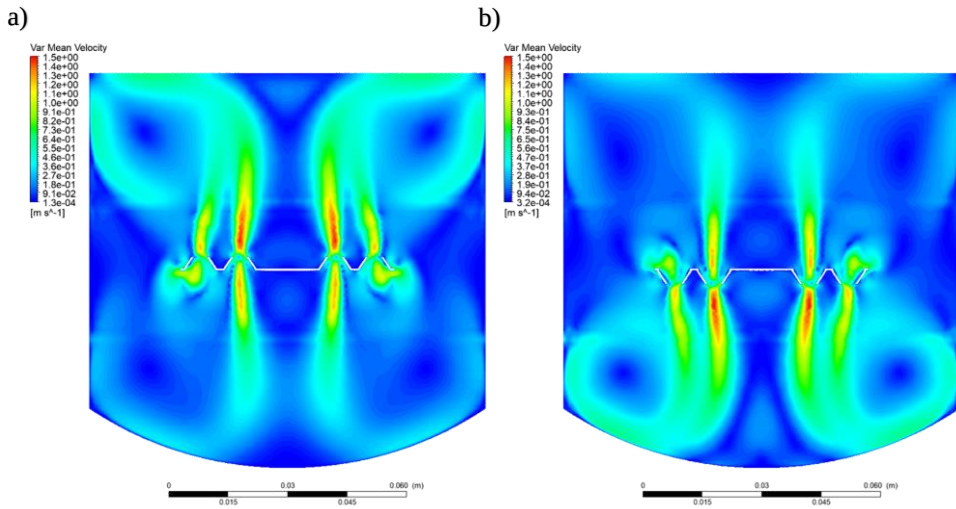


Figure 4.12: Colour maps of mean velocity magnitude for an up-pumping configuration vibromixer (left) and a down-pumping configuration vibromixer (right). Off-bottom clearance is $0.50 H$, frequency is 100 Hz, and amplitude is 3.5 mm.

This can be confirmed through experimental Particle Image Velocimetry (PIV) analysis of the same simulated vibromixer, not present in this thesis.

The following simulations aim to compare two different amplitudes while keeping the geometry and physical parameters constant. Color maps of velocity profiles within the same vibromixer are shown in Figure 4.13 after reaching steady-state, for a vibrating disk with (a) amplitude of 3.5 mm and (b) amplitude of 1.5 mm.

It is immediately visible that the maximum instantaneous velocities achieved in the first configuration are significantly higher than those in the second configuration, as expected. The recirculation regions appear similar in structure but are of lower magnitude in the second case. As before, in Figure 4.14, the time-averaged velocity profiles are presented.

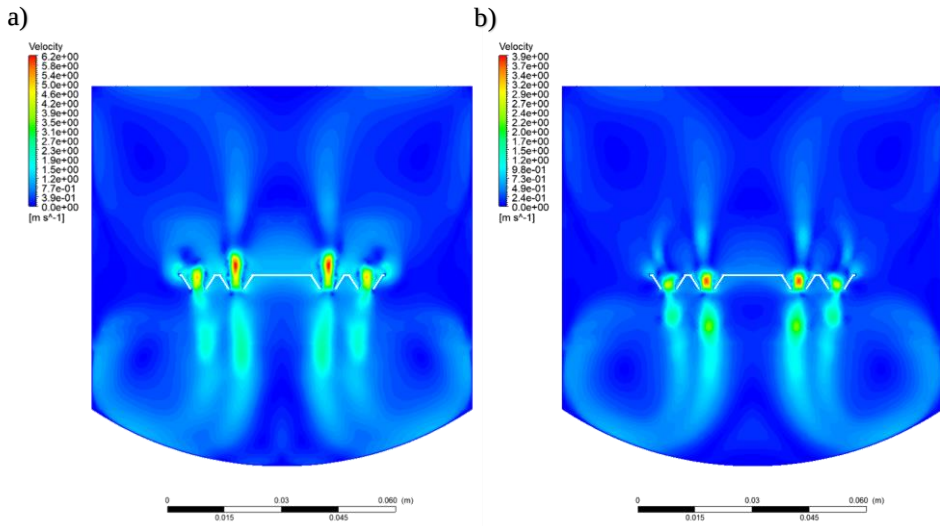


Figure 4.13: Color maps of velocity magnitude for the down-pumping vibromixer configuration with (a) amplitude of 3.5 mm and (b) amplitude of 1.5 mm at the same time step. Off-bottom clearance is 0.50 H, and frequency is 100 Hz.

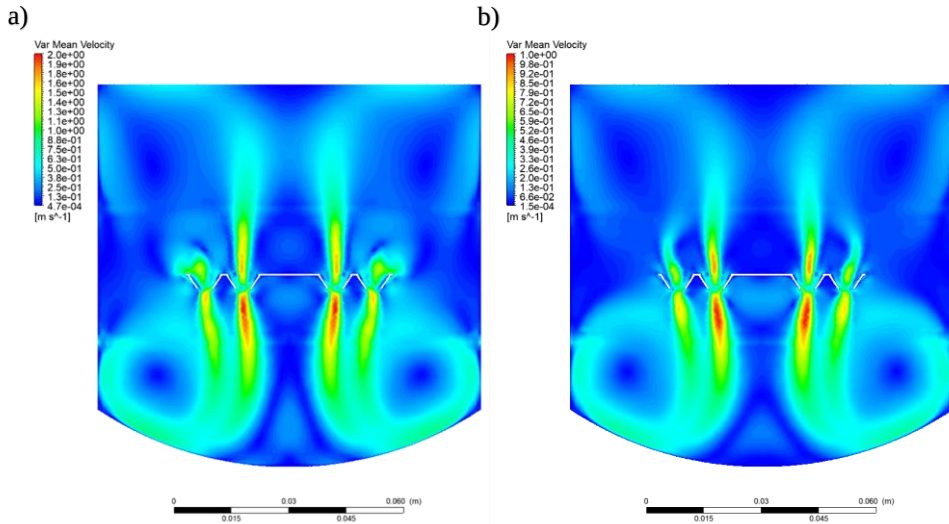


Figure 4.14: Color maps of velocity magnitude for the down-pumping vibromixer configuration with (a) amplitude of 3.5 mm and (b) amplitude of 1.5 mm at the same time step. Off-bottom clearance is 0.50 H, and frequency is 100 Hz.

Figure 4.15 shows color maps of the mean shear rate in the domain of the moving plate at $\bar{A} = 3.5$ mm for a plate with size $D = 0.36T$, located in the middle of the reactor ($C = 0.5T$), with the plate positioned in the downward pumping direction.

This figure provides a clear depiction of the high shear rate gradients observed near the perforated plate, specifically at the locations of the conical holes. The color maps illustrate mean shear rates within the reactor domain, with parameters set at $\bar{A} = 3.5$ mm, frequency $f = 100$ Hz, and the plate positioned at $C = 0.5T$, with a diameter of $D = 0.36T$, oriented in the downward pumping direction.

The data indicates that the conical holes and the adjacent regions of the plate are characterized by intense mixing and fluid motion. This localized high shear rate enhances the breakdown of droplets into finer sizes, significantly improving mass transfer within the reactor.

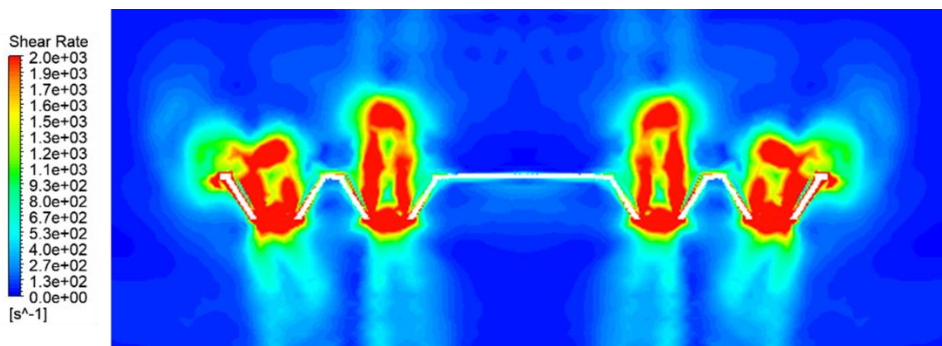


Figure 4.15: Colour maps of mean shear rate in the domain of the moving plate within the reactor at $\bar{A} = 3.5$ mm, frequency $f = 100$ Hz, with the plate located at $C = 0.5T$, size of $D = 0.36T$ in the downward pumping direction.

Consequently, this results in an increased content of fatty acid methyl esters (FAME) due to more efficient emulsification and reaction processes.

Additionally, increasing the amplitude enhances the dispersion process of immiscible mixtures by generating greater inertial stresses and shear forces. Specifically, the mean shear rate in the domain of the moving plate increases from 74 s^{-1} to 156 s^{-1} as the amplitude rises from $A = 1.5 \text{ mm}$ to $A = 3.5 \text{ mm}$. For a plate with a diameter of $D = 0.36T$, positioned at $C = 0.5T$ and oriented in the downward pumping direction, the circulation number is lower at $A = 1.5 \text{ mm}$ (0.112) compared to $A = 3.5 \text{ mm}$ (0.197). This indicates that the circulation time for fluid elements is longer when the amplitude is 1.5 mm than when it is 3.5 mm.

The shorter circulation time at $A = 3.5 \text{ mm}$ results in more rapid fluid movement, which contributes to the higher shear rates and velocities observed with increased amplitude. This enhanced fluid motion and shear rate improve the efficiency of mass transfer processes, leading to a higher concentration of fatty acid methyl esters (FAME) in the reactor. The increased shear rate promotes more effective emulsification and reaction kinetics, thereby optimizing the overall performance of the reactor. Therefore, the performance of the transesterification reaction to produce FAME is influenced by a combination of plate size and amplitude. It is thought that increasing the amplitude and plate size enhances turbulence and the dispersion process of immiscible mixture by generating greater inertial stresses and shear forces, as well as increasing energy dissipation rates.

The final comparison investigates whether the turbulence model used is suitable for a lower Reynolds number than in the previous case, where the amplitude was 3.5 mm, yielding a Reynolds number of approximately 1350. According to the literature, reducing the vibration amplitude lowers the Reynolds number (in this case, approximately 580), moving the system out of the fully turbulent range and into a transitional zone.

Thus, a comparison between the $k-\omega$ SST turbulence model and the $k-\omega$ SST SBES model was conducted, as studies suggest the latter may be more suitable for transitional flow conditions. Figure 4.16 shows the respective color maps of velocity profiles once steady-state conditions are reached in both cases, and Figure 4.17 shows the corresponding color maps of time-averaged velocity profiles.

We can analyze the fluid dynamics simulations by comparing the results in terms of Power Consumption, Newton Number, Reynolds Number, Flow Number, and Circulation Number. These results are displayed in Table 4.2.

Table 4.2: *Numerical comparison between simulations concerning Biodiesel production with a vibromixer.*

	Simulation 1 Up-pumping A: 3.5 mm k-ω SST	Simulation 2 Down- pumping A: 3.5 mm k-ω SST	Simulation 3 Down- pumping A: 1.5 mm k-ω SST	Simulation 4 Down-pumping A: 1.5 mm k-ω SST SBES
Power				
Consumption [W]	8.08	7.74	2.49	2.34
Newton Number	1.19	1.14	4.70	4.41
Reynold Number	1352.4	1352.4	579.6	579.6
Flow Number	0.116	0.165	0.104	0.117
Circulation				
Number	0.18	0.197	0.112	0.139

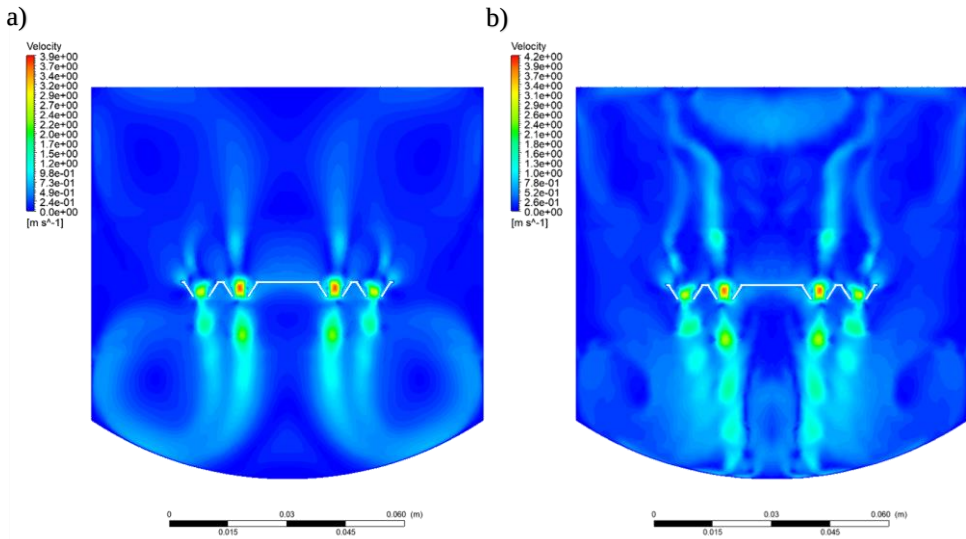


Figure 4.16: Color maps of velocity magnitude for the down-pumping vibromixer configuration using the turbulence models: (a) $k-\omega$ SST and (b) $k-\omega$ SST (SBES) at the same time step. Off-bottom clearance is $0.50 H$, frequency is 100 Hz, and amplitude is 1.5 mm.

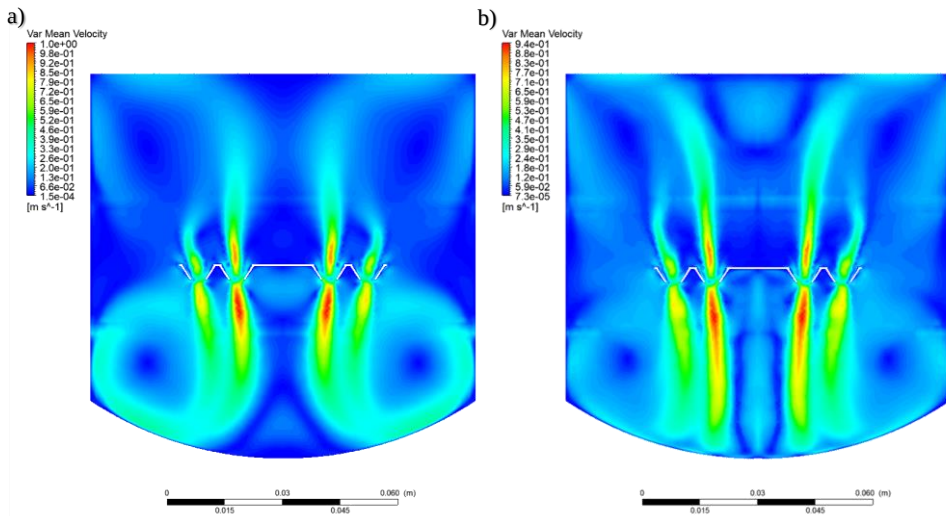


Figure 4.17: Color maps of mean velocity magnitude for the down-pumping vibromixer configuration using the turbulence models: (a) $k-\omega$ SST and (b) $k-\omega$ SST (SBES) at the same time step. Off-bottom clearance is $0.50 H$, frequency is 100 Hz, and amplitude is 1.5 mm.

4.4.2 Solid-Liquid Suspension

With the computational domain, shown in Figure 4.3, two preliminary simulations were conducted for a biphasic system, aimed at streamlining the setup process for subsequent solid-liquid simulations, not investigated due to time constraints.

These solid-liquid simulations involve water as the primary phase and glass beads with a diameter of two millimeters as the secondary phase. Two settings were examined: the first, shown in Figure 4.18, employs the multiphase option, describing the two components as Eulerian phases—the first being a continuous water phase and the second a granular phase with specified particle diameter. The second simulation utilizes the DEM (Discrete Element Method) option, as depicted in Figure 4.19.

For this latter case, 54 spheres were instantaneously injected using an external file specifying their spatial positions and physical and geometric properties. Both simulations demanded a considerable amount of computational time.

In the first simulation, color maps illustrate the volume fraction distribution of glass beads in water for the down-pumping vibromixer configuration at various time steps. As time progresses, the maps show a gradual dispersion of the glass beads, highlighting the mixing effectiveness of the vibromixer and demonstrating how the cloud of suspended solids reaches approximately $3/4$ of the vessel height.

The simulation also presents particle tracking of the glass beads within the same down-pumping vibromixer configuration across successive time steps. The

sequential images reveal the movement and distribution of the beads throughout the vessel, with particles initially concentrated at the bottom and gradually dispersing into the liquid.

Due to the different initial conditions set in the two systems, in the DEM case, the presence of only 54 spheres, while in the case with two Eulerian phases, a bed that almost entirely covered the bottom of the vessel, and the simulated seconds, the two results should be evaluated qualitatively and not quantitatively.

Together, these figures underscore the capability of the down-pumping vibromixer configuration to promote suspension and distribution of solid particles (glass beads) within a liquid medium (water) over time. This demonstrates its efficiency in achieving homogeneous mixing in solid-liquid systems.

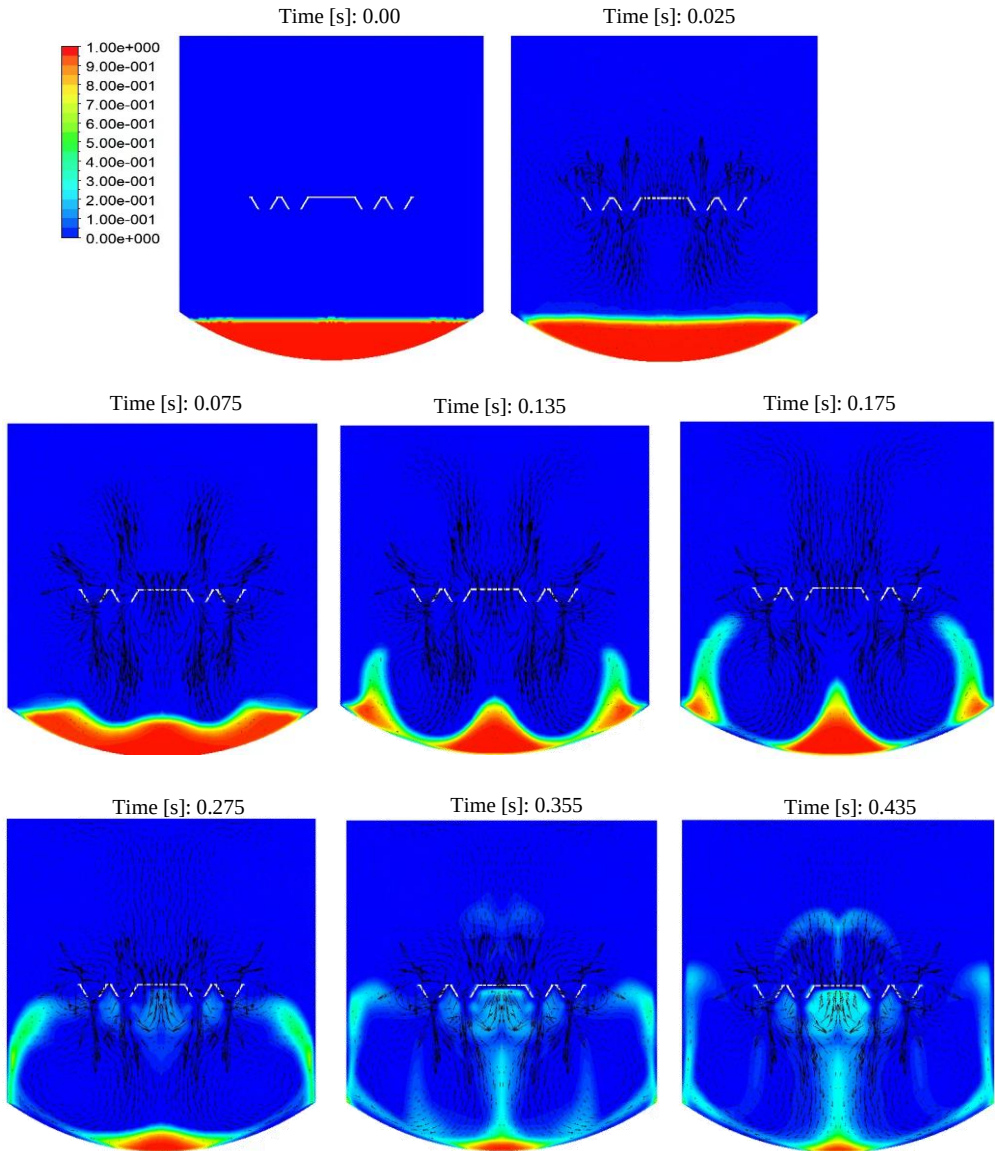


Figure 4.18: Colour maps of volume fraction of glass beads into water for the down-pumping vibromixer configuration at different time steps.

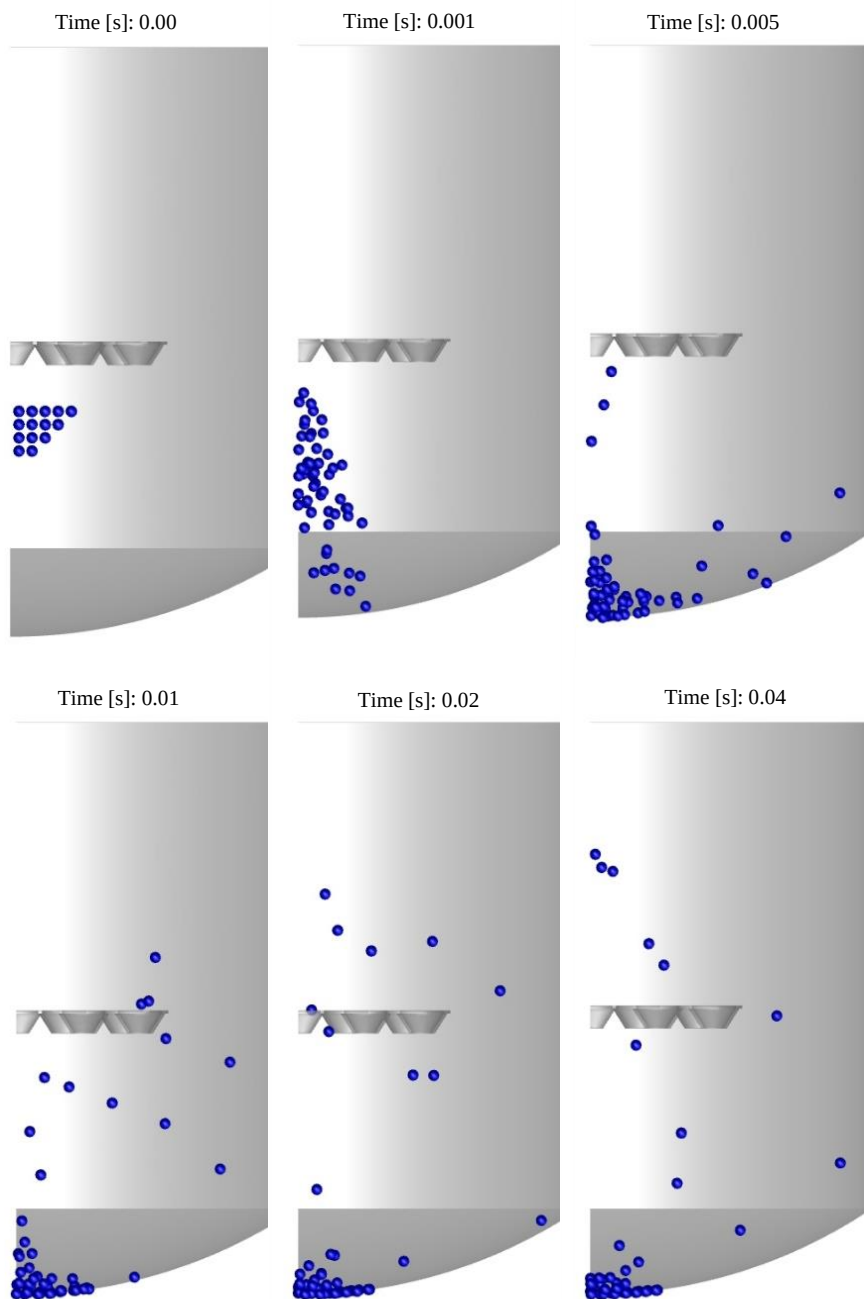


Figure 4.19: Particle tracking representing glass beads for the down-pumping vibromixer configuration at increasing time steps.

4.5 Conclusions

The results obtained from vibromixing simulations offer a comprehensive understanding of process dynamics, underscoring the effectiveness and unique features of this technique in achieving fluid homogenization. Through meticulous data analysis, the system's performance in terms of mixing efficiency, particle distribution, and other relevant parameters has been evaluated.

The velocity profiles over time reveal significant flow structures, providing visual support for understanding the physical phenomena in both laboratory and industrial applications of vibromixing.

In the context of biodiesel production, computational fluid dynamics (CFD) simulations have proven essential for analyzing the flow patterns within a vibromixer. Utilizing URANS simulations, various operational setups were systematically examined, producing crucial velocity profiles, pressure distributions, and vector plots. Comparative analyses of different configurations, such as up-pumping and down-pumping, highlighted flow profile phenomena that diverged from existing literature, confirming single recirculations within the entire fluid and illustrating the complexity of fluid dynamics from initial flow development to steady-state.

Furthermore, investigations into solid-liquid multiphase systems provided valuable insights. Preliminary simulations explored the behavior of water-glass bead mixtures, employing both Eulerian-Eulerian multiphase simulations and the Discrete Element Method (DEM). While these simulations were computationally intensive,

they established a foundation for further exploration of suspension heights and comparison with experimental data.

The examination of vibromixer dynamics uncovered notable phenomena, including the formation of recirculation zones and central jet effects. In particular, the simulations highlighted differences in mixing efficiency between configurations, emphasizing the significance of parameter optimization to enhance process performance.

Overall, the results presented in this chapter underscore the pivotal role of CFD in elucidating the behavior of complex systems such as a perforated plate vibromixer, providing critical insights for process optimization. With ongoing experimental validations and further investigations, these simulation outcomes will undoubtedly advance the understanding of this mixer configuration and its potential applications.

GENERAL CONCLUSIONS & FUTURE PERSPECTIVES

The work presented in this thesis underscores the importance of understanding multiphase flow phenomena in a variety of industrial applications, particularly through the use of computational fluid dynamics (CFD) simulations. By investigating complex systems such as supercritical fluid extraction (SFE), stirred tank reactors, and vibromixing processes, the study emphasizes how CFD can optimize process conditions and improve operational efficiency. Each chapter of the thesis contributes to a broader understanding of fluid dynamics, offering valuable insights into system design, performance, and potential areas for future evaluation.

Chapter 2's experimental results on SFE in a column filled with Sulzer® EX packing revealed that the best conditions for omega-3 extraction were achieved at 145 bar pressure, 60°C temperature, and a sc-CO₂/oil flow rate ratio of 100. CFD simulations of a simplified version of the experimental system provided valuable predictions, with results qualitatively matching experimental observations and identifying the transition to flooding conditions. The model can be further developed to optimize operating conditions for different matrices and under more complex calculation domains. Furthermore, combining this model with a predictive mass transport model could lead to more accurate predictions of SFE yields in terms of enrichment of high-value components, which will be essential for scaling up the process and improving its efficiency in industrial applications.

In *Chapter 3*, the study of Uncovered Unbaffled Stirred Tanks (UUSTs) demonstrated how CFD simulations could effectively capture the dynamics of free

surface behavior and vortex formation under varying impeller speeds. The use of the Reynolds Stress Model with Linear Pressure-Strain Model (RSM-LPSM) was crucial in providing accurate predictions of the power number, the shape and depth of the central vortex, and the transition between sub-critical and super-critical regimes. These findings are critical for optimizing stirred tank reactor design, especially when gas-liquid mass transfer is a key factor. Moving forward, coupling CFD simulations with mass balance equations will allow for a more comprehensive prediction of the gas-liquid Volumetric Mass Transfer Coefficient $k_L a$ in both sub-critical and super-critical regimes. This approach will enable a more precise evaluation of reactor performance, ultimately contributing to more efficient and sustainable chemical processes.

Chapter 4 explored the vibromixing process, revealing the effectiveness of this technique in achieving fluid homogenization, particularly in solid-liquid systems. Through detailed CFD simulations of different operational configurations, the study highlighted the complex flow dynamics within the vibromixer, including recirculation zones and jet formation. These findings provide valuable insights into the mixing efficiency of various setups and their impact on process performance. Future work will focus on further investigating the flow field variations as the Reynolds number changes, as well as conducting more in-depth comparisons with experimental data, such as those obtained through Particle Image Velocimetry (PIV). This will validate the simulation results and provide a more robust understanding of the mixing process, helping to optimize mixer configurations for industrial

applications such as biodiesel production. The continued use of CFD simulations in combination with experimental validation will allow for the development of more efficient mixing systems and improve the overall performance of multiphase processing techniques.

The overall contribution of this thesis lies in its advancement of multiphase flow modeling and its application to real-world industrial processes. By bridging the gap between theoretical modeling and experimental validation, this work provides a foundation for the optimization of various multiphase systems. Moving forward, it will be essential to continue refining the models developed in this thesis, particularly in the context of more complex fluid systems and multiphase interactions. The integration of CFD with advanced predictive models for mass transport and reaction kinetics holds significant potential for improving process design, operational efficiency, and product yield. Additionally, the scalability of the models to industrial applications will require further validation and fine-tuning, particularly in terms of computational cost and simulation accuracy.

Ultimately, this work lays the groundwork for future advancements in the modeling and simulation of multiphase flows, which will enable the optimization of a wide range of industrial processes, from supercritical fluid extraction to mixing and reaction engineering.

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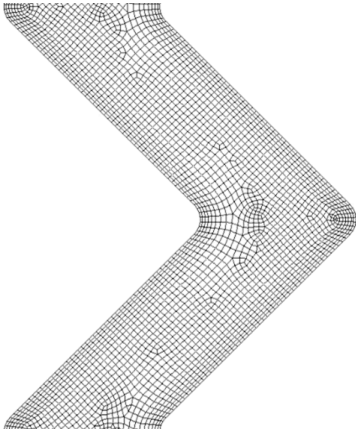
APPENDIX A

Given the challenging nature of observing the actual phenomenon under conditions involving supercritical carbon dioxide at high pressure, it was assumed perfect wettability of the metallic gauze that constitutes the structured packing. It was hypothesized that, during the initial passage of the oil phase, the gauze would not be entirely dried upon subsequent exposure to the sc-CO₂ once the oil phase was no longer continuous.

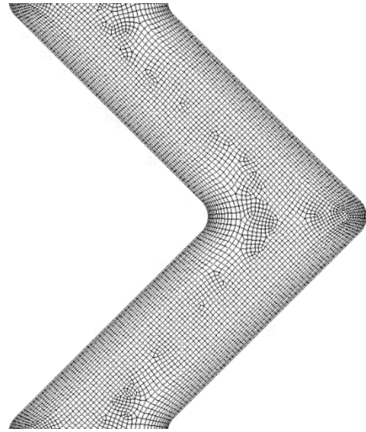
The focus was therefore placed on refining the inflation layer near the wall, as shown in Figure A.1, by testing meshes with 60,000, 120,000, 160,000, and 480,000 volumes. In the first two coarser meshes, the phenomenon was not observed, whereas in the latter two, upon the disruption of the continuous oil phase for a given flow rate, a residual oil layer formed.

This layer was estimated plausible as it facilitated subsequent crossings. For this reason, the 160,000-volume mesh was selected as the optimal configuration for the computational domain.

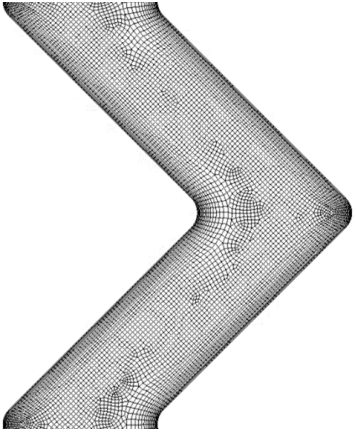
a) ~ 60 000 volumes



b) ~ 120 000 volumes



c) ~ 160 000 Volumes



d) ~ 480 000 Volumes

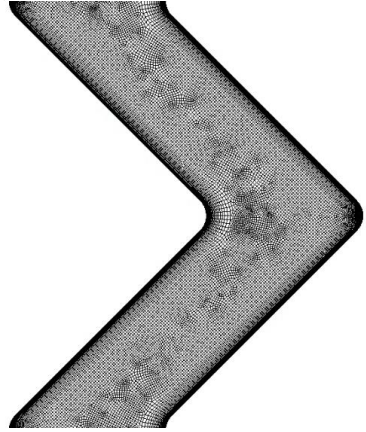


Figure A.1: Structured hexahedral meshes with a) 60,000, b) 120,000, c) 160,000 and d) 480,000 total volumes.

APPENDIX B

A grid independence study was conducted to ensure the accuracy and reliability of the simulation results for the vibromixing analysis. After defining a valid initial resolving setup, as detailed in the subsequent section, four different unstructured tetrahedral meshes were created for the simulated cases. These meshes were designed to capture the intricate geometry of the vibrating disk's conical sections and included inflation layers near the walls.

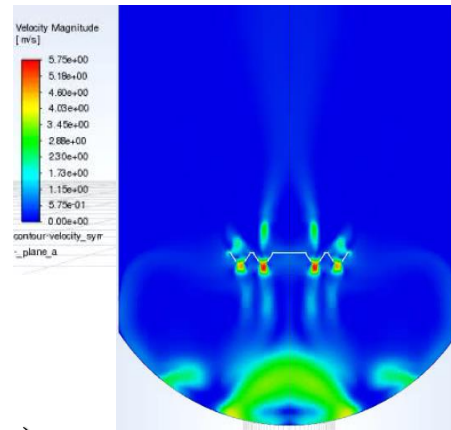
The grid independence study involved testing meshes with 2 504 083, 1 191 187, 819 490, and 629 371 cells, as described in Table B.1. These meshes were evaluated based on critical numerical parameters, including velocity profiles, pressure distributions, and dimensionless metrics such as Newton and Circulation numbers, to determine the optimal mesh size that balances computational cost and accuracy.

During the simulations, once a valid resolving setup was established, also anomalies were observed in the flow field for cases Mesh n°1, Mesh n°2, and Mesh n°4 shown in Figure B.1. At various time instances, the flow field exhibited irregular patterns, resembling external interferences that introduced distortions into the flow field. These anomalies were not observed in Mesh 3, which was ultimately selected as the optimal configuration. This choice was consistent with the findings of the grid independence study, as increasing the mesh size beyond approximately 850 000 cells did not yield significant improvements in numerical accuracy. For the selected

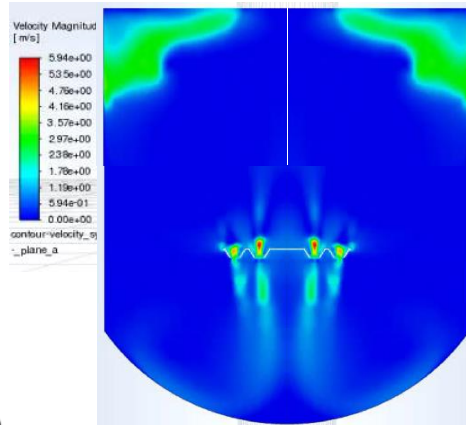
configuration, the computational domain contained 819 490 cells for the up-pumping case and 830 332 cells for the down-pumping configuration.

Table B.1: *Unstructured tetrahedral meshes investigated.*

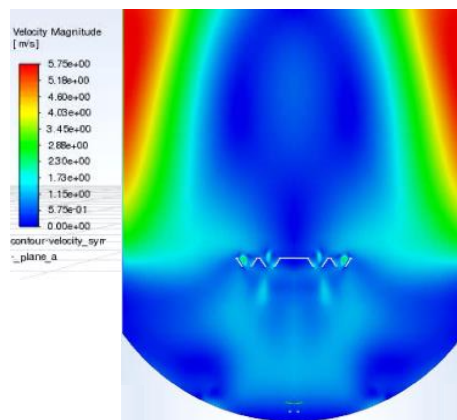
	Mesh n°1	Mesh n°2	Mesh n°3	Mesh n°4
n° cells	2 504 083	1 091 187	830 332	629 371
n° faces	5 219 634	2 302 847	1 721 834	1 305 110
n° nodes	549 205	257 257	175 880	133 313



a)



b)



c)

Figure B.1: Flow patterns for meshes with a) 2 504 083 b) 1 091 187 and c) 629 371 total volumes.

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BIBLIOGRAPHY

- Abou Elmaaty, T., Sayed-Ahmed, K., Elsisy, H., Magdi, M., 2022. Optimization of Extraction of Natural Antimicrobial Pigments Using Supercritical Fluids: A Review. *Processes* 10, 2111. <https://doi.org/10.3390/pr10102111>
- Alcamo, R., Micale, G., Grisafi, F., Brucato, A., Ciofalo, M., 2005. Large-eddy simulation of turbulent flow in an unbaffled stirred tank driven by a Rushton turbine. *Chem. Eng. Sci.* 60, 2303–2316. <https://doi.org/10.1016/j.ces.2004.11.017>
- Aloi, L.E., Cherry, R.S., 1996. Cellular response to agitation characterized by energy dissipation at the impeller tip. *Chem. Eng. Sci.* 51, 1523–1529. [https://doi.org/10.1016/0009-2509\(95\)00307-X](https://doi.org/10.1016/0009-2509(95)00307-X)
- Anderson, D.M., McFadden, G.B., Wheeler, A.A., 1998. Diffuse-interface methods in fluid mechanics, *Annu. Rev. ed.*
- ANSYS Inc., 2023. ANSYS Fluent Th. Guide. ANSYS Inc., USA Release 12.0®.
- ANSYS Inc., 2021. Ansys Fluent Theory Guide. Release 2021 R2 ANSYS, Inc. 1069.
- Bertrand, Murielle, Parmentier, D., Lebaigue, O., Plasari, E., Ducros, F., 2012. Mixing study in an unbaffled stirred precipitator using les modelling. *Int. J. Chem. Eng.* 2012. <https://doi.org/10.1155/2012/450491>
- Bertrand, M., Plasari, E., Lebaigue, O., Baron, P., Lamarque, N., Ducros, F., 2012. Hybrid LES-multizonal modelling of the uranium oxalate precipitation. *Chem. Eng. Sci.* 77, 95–104. <https://doi.org/10.1016/j.ces.2012.03.019>
- Bird, R.B., Stewart, W.E., Lightfoot, E.N., 2007. *Transport Phenomena*, 2nd Editio. ed.

- Bisotti, F., di Pretoro, A., Dell'Angelo, A., Previtali, D., Amaral, A.F., Andoglu, E.M., Manenti, F., 2019. Smart implementation of bender equation of state. *Chem. Eng. Trans.* 74, 715–720. <https://doi.org/10.3303/CET1974120>
- Brackbill, J.U., Kothe, D.B., Zemach, C., 1992. A continuum method for modeling surface tension. *J. Comput. Phys.* 100, 335–354. [https://doi.org/10.1016/0021-9991\(92\)90240-Y](https://doi.org/10.1016/0021-9991(92)90240-Y)
- Brunner, G., 2013. Gas Extraction, *Journal of Chemical Information and Modeling*.
- Busciglio, A., Caputo, G., Scargiali, F., 2013. Free-surface shape in unbaffled stirred vessels: Experimental study via digital image analysis. *Chem. Eng. Sci.* 104, 868–880. <https://doi.org/10.1016/j.ces.2013.10.019>
- Busciglio, A., Grisafi, F., Scargiali, F., Brucato, A., 2014. Mixing dynamics in uncovered unbaffled stirred tanks. *Chem. Eng. J.* 254, 210–219. <https://doi.org/10.1016/j.cej.2014.05.084>
- Busciglio, A., Scargiali, F., Grisafi, F., Brucato, A., 2016. Oscillation dynamics of free vortex surface in uncovered unbaffled stirred vessels. *Chem. Eng. J.* 285, 477–486. <https://doi.org/10.1016/j.cej.2015.10.015>
- Caputo, G., Rubio, P., Scargiali, F., Marotta, G., Brucato, A., 2016. Experimental and fluid dynamic study of continuous supercritical water gasification of glucose. *J. Supercrit. Fluids* 107, 450–461. <https://doi.org/10.1016/j.supflu.2015.09.022>
- Ciofalo, M., Brucato, A., Grisafi, F., Torracca, N., 1996. Turbulent flow in closed and free-surface unbaffled tanks stirred by radial impellers. *Chem. Eng. Sci.* 51, 3557–3573. [https://doi.org/10.1016/0009-2509\(96\)00004-8](https://doi.org/10.1016/0009-2509(96)00004-8)
- Daly, B.J., Harlow, F.H., 1970. Transport Equations in Turbulence. *Phys. Fluids* 13, 2634–2649. <https://doi.org/10.1063/1.1692845>

- Derksen, J., Van Den Akker, H.E.A., 1999. Large eddy simulations on the flow driven by a Rushton turbine. *AIChE J.* 45, 209–221.
<https://doi.org/10.1002/aic.690450202>
- Deshpande, S.S., Kar, K.K., Walker, J., Pressler, J., Su, W., 2017. An experimental and computational investigation of vortex formation in an unbaffled stirred tank. *Chem. Eng. Sci.* 168, 495–506.
<https://doi.org/10.1016/j.ces.2017.04.002>
- Djuricic, I., Calder, P.C., 2021. Beneficial outcomes of omega-6 and omega-3 polyunsaturated fatty acids on human health: An update for 2021. *Nutrients* 13. <https://doi.org/10.3390/nu13072421>
- Drew, D.A., Passman, S.L., 1999. *Theory of Multicomponent Fluids*, pp. 1–10. Wiley, New York.
- Escudié, R., Liné, A., 2006. Analysis of turbulence anisotropy in a mixing tank. *Chem. Eng. Sci.* 61, 2771–2779. <https://doi.org/10.1016/j.ces.2005.09.022>
- Fernandes, J., Lisboa, P.F., Simões, P.C., Mota, J.P.B., Saadjan, E., 2009. Application of CFD in the study of supercritical fluid extraction with structured packing: Wet pressure drop calculations. *J. Supercrit. Fluids* 50, 61–68. <https://doi.org/10.1016/j.supflu.2009.04.009>
- Fu, S., Huang, P.G., Launder, B.E., Leschziner, M.A., 1988. A Comparison of Algebraic and Differential Second-Moment Closures for Axisymmetric Turbulent Shear Flows With and Without Swirl. *J. Fluids Eng.* 110, 216–221.
<https://doi.org/10.1115/1.3243537>
- Ghazouani, J., Chouaieb, O., Bellagi, A., 2005. Evaluation of the parameters of the Bender equation of state for low acentric factor fluids and carbon dioxide. *Thermochim. Acta* 432, 10–19. <https://doi.org/10.1016/j.tca.2004.11.008>

- Gibson, M.M., Launder, B.E., 1978. Ground effects on pressure fluctuations in the atmospheric boundary layer. *J. Fluid Mech.* 86, 491–511.
<https://doi.org/10.1017/S0022112078001251>
- Glover, G.M.C., Fitzpatrick, J.J., 2007. Modelling vortex formation in an unbaffled stirred tank reactors. *Chem. Eng. J.* 127, 11–22.
<https://doi.org/10.1016/j.cej.2006.09.019>
- H.D. Baehr, K.S., 1998. Heat and mass transfer, 1998th ed. Springer Verlag, Berlin.
- Haque, J.N., Mahmud, T., Roberts, K.J., Rhodes, D., 2006. Modeling turbulent flows with free-surface in unbaffled agitated vessels. *Ind. Eng. Chem. Res.* 45, 2881–2891. <https://doi.org/10.1021/ie051021a>
- Heidaryan, E., Hatami, T., Rahimi, M., Moghadasi, J., 2011. Viscosity of pure carbon dioxide at supercritical region : Measurement and correlation approach. *J. Supercrit. Fluids* 56, 144–151.
<https://doi.org/10.1016/j.supflu.2010.12.006>
- Hibiki, T., Ishii, M., 2002. Development of one-group interfacial area transport equation in bubbly flow systems. *Int. J. Heat Mass Trans.*
- Hirt, C.W., Nichols, B.D., 1981. Volume of Fluid (VoF) method for the dynamics of free boundaries. *J. Comput. Phys.*
- Ishii, M., 1975. Thermo-Fluid Dynamic Theory of Two-Phase Flow. Paris.
- Ishii, M., Mishima, K., 1981. Study of two-fluid model and interfacial area, Argonne Na. ed.
- Ishii, M., Sun, X., Kim, S., 2003. Modelling strategy of the source and sink terms in the twogroup interfacial area transport equation. *Ann. Nucl. Energy*.
- Issa, R.I., Kempf, M.H.W., 2003. Simulation of slug flow in horizontal and nearly horizontal pipes with the two-fluid model, *Int. J. Mu.* ed.

- Jiao, Y., Zhang, Z., Gao, F., Han, J., Wang, J., 2024. Effects of Particle Suspension on Surface Vortex in Unbaffled Stirred Tanks through DEM-VOF. *Ind. Eng. Chem. Res.* 63, 4662–4677. <https://doi.org/10.1021/acs.iecr.3c04462>
- Kamiński, J., Wójtowicz, R., 2003. Dispersion of liquid-liquid systems in a mixer with a reciprocating agitator. *Chem. Eng. Process. Process Intensif.* 42, 1007–1017. [https://doi.org/10.1016/S0255-2701\(02\)00214-3](https://doi.org/10.1016/S0255-2701(02)00214-3)
- Kocamustafaogullari, G., Ishii, M., 1995. Foundation of the interfacial area transport equation and its closure relations. *Int. J. Heat Mass Trans.*
- Komoda, Y., Inoue, Y., Hirata, Y., 2000. Mixing performance by reciprocating disk in cylindrical vessel. *J. Chem. Eng. Japan* 33, 879–885. <https://doi.org/10.1252/jcej.33.879>
- Lamarque, N., Zoppé, B., Lebaigue, O., Dolias, Y., Bertrand, M., Ducros, F., 2010. Large-eddy simulation of the turbulent free-surface flow in an unbaffled stirred tank reactor. *Chem. Eng. Sci.* 65, 4307–4322. <https://doi.org/10.1016/j.ces.2010.03.014>
- Lamberto, D.J., Muzzio, F.J., Swanson, P.D., Tonkovich, A.L., 1996. Using time-dependent RPM to enhance mixing in stirred vessels. *Chem. Eng. Sci.* 51, 733–741. [https://doi.org/10.1016/0009-2509\(95\)00203-0](https://doi.org/10.1016/0009-2509(95)00203-0)
- Launder, B.E., 1989. Second-moment closure and its use in modelling turbulent industrial flows. *Int. J. Numer. Methods Fluids* 9, 963–985. <https://doi.org/10.1002/fld.1650090806>
- Launder, B.E., Reece, G.J., Rodi, W., 1975a. Progress in the development of a Reynolds-stress turbulence closure. *J. Fluid Mech.* 68, 537–566. <https://doi.org/10.1017/S0022112075001814>
- Launder, B.E., Reece, G.J., Rodi, W., 1975b. Progress in the development of a Reynolds-stress turbulence closure. *J. Fluid Mech.* 68, 537–566. <https://doi.org/10.1017/S0022112075001814>

- Launder, B.E., Spalding, D.B., 1974. The numerical computation of turbulent flows. *Comput. Methods Appl. Mech. Eng.* 3, 269–289.
[https://doi.org/10.1016/0045-7825\(74\)90029-2](https://doi.org/10.1016/0045-7825(74)90029-2)
- Li, L., Wang, J., Feng, L., Gu, X., 2017. Computational fluid dynamics simulation of hydrodynamics in an uncovered unbaffled tank agitated by pitched blade turbines. *Korean J. Chem. Eng.* 34, 2811–2822.
<https://doi.org/10.1007/s11814-017-0208-9>
- Li, L., Xu, B., 2017. Numerical simulation of hydrodynamics in an uncovered unbaffled stirred tank. *Chem. Pap.* 71, 1863–1875.
<https://doi.org/10.1007/s11696-017-0180-1>
- Lien, F.S., Leschziner, M.A., 1994. Assessment of turbulence-transport models including non-linear rng eddy-viscosity formulation and second-moment closure for flow over a backward-facing step. *Comput. Fluids* 23, 983–1004.
[https://doi.org/10.1016/0045-7930\(94\)90001-9](https://doi.org/10.1016/0045-7930(94)90001-9)
- Lima, S., Brucato, A., Caputo, G., Schembri, L., Scargiali, F., 2022a. Modelling *Nannochloropsis gaditana* Growth in Reactors with Different Geometries, Determination of Kinetic Parameters and Biochemical Analysis in Response to Light Intensity. *Appl. Sci.* 12. <https://doi.org/10.3390/app12125776>
- Lima, S., Cosenza, A., Caputo, G., Grisafi, F., Scargiali, F., 2024. Utilization of native *Chlorella* strain in laboratory-scale raceway reactor for synthetic wastewater treatment: A study in batch and continuous modes with multi-substrate modeling. *J. Water Process Eng.* 60, 105145.
<https://doi.org/10.1016/j.jwpe.2024.105145>
- Lima, S., Lokesh, J., Schulze, P.S.C., Wijffels, R.H., Kiron, V., Scargiali, F., Petters, S., Bernstein, H.C., Morales-Sánchez, D., 2022b. Flashing lights affect the photophysiology and expression of carotenoid and lipid synthesis genes in *Nannochloropsis gaditana*. *J. Biotechnol.* 360, 171–181.
<https://doi.org/10.1016/j.jbiotec.2022.11.012>

- Lima, S., Villanova, V., Grisafi, F., Brucato, A., Scargiali, F., 2020. Combined effect of nutrient and flashing light frequency for a biochemical composition shift in *Nannochloropsis gaditana* grown in a quasi-isoactinic reactor. *Can. J. Chem. Eng.* 98, 1944–1954. <https://doi.org/10.1002/cjce.23776>
- Lo, M.Y.A., Gierczycki, A.T., Titchener-Hooker, N.J., Shamlou, P.A., 1998. Newtonian power curve and drop size distributions for vibromixers. *Can. J. Chem. Eng.* 76, 471–478. <https://doi.org/10.1002/cjce.5450760317>
- Mahmud, T., Haque, J.N., Roberts, K.J., Rhodes, D., Wilkinson, D., 2009. Measurements and modelling of free-surface turbulent flows induced by a magnetic stirrer in an unbaffled stirred tank reactor. *Chem. Eng. Sci.* 64, 4197–4209. <https://doi.org/10.1016/j.ces.2009.06.059>
- Maluta, F., Alberini, F., Paglianti, A., Montante, G., 2023. Hydrodynamics, power consumption and bubble size distribution in gas-liquid stirred tanks. *Chem. Eng. Res. Des.* 194, 582–596. <https://doi.org/10.1016/j.cherd.2023.05.006>
- Maluta, F., Paglianti, A., Montante, G., 2021. Prediction of gas cavities size and structure and their effect on the power consumption in a gas-liquid stirred tank by means of a two-fluid RANS model. *Chem. Eng. Sci.* 241, 116677. <https://doi.org/10.1016/j.ces.2021.116677>
- Menter, F.R., Kuntz, M., Langtry, R., 2003. Ten Years of Industrial Experience with the SST Turbulence Model Turbulence heat and mass transfer. *Cfd.Spbstu.Ru* 4, 625–632.
- Michalcová, V., Kotrasová, K., 2020. The numerical diffusion effect on the cfd simulation accuracy of velocity and temperature field for the application of sustainable architecture methodology. *Sustain.* 12, 1–18. <https://doi.org/10.3390/su122310173>

- Montante, G., Bakker, A., Paglianti, A., Magelli, F., 2006. Effect of the shaft eccentricity on the hydrodynamics of unbaffled stirred tanks. *Chem. Eng. Sci.* 61, 2807–2814. <https://doi.org/10.1016/j.ces.2005.09.021>
- Morel, C., Goreaud, N., Delhaye, J.-M., 1999. The local volumetric interfacial area transport equation: derivation and physical significance, *Int. J. Mu. ed. Int. J. Multiphase Flow*.
- Moucha, T., Linek, V., Prokopová, E., 2003. Gas hold-up, mixing time and gas-liquid volumetric mass transfer coefficient of various multiple-impeller configurations: Rushton turbine, pitched blade and techmix impeller and their combinations. *Chem. Eng. Sci.* 58, 1839–1846. [https://doi.org/10.1016/S0009-2509\(02\)00682-6](https://doi.org/10.1016/S0009-2509(02)00682-6)
- Ngo, T.A., Nguyen, T.H., Bui, H.T.V., 2012. Thermophilic fermentative hydrogen production from xylose by *Thermotoga neapolitana* DSM 4359. *Renew. Energy* 37, 174–179. <https://doi.org/10.1016/j.renene.2011.06.015>
- Nienow, A.W., Warmoeskerken, M.M.C.G., Smith, J.M., Konno, M., 1985. On the flooding/ loading transition and the complete dispersal condition in aerated vessels agitated by a Rushton turbine. *5th Eur. Conf. Mix. Wurzburg, West Ger.* 143–154. [https://doi.org/10.1016/0009-2509\(87\)85067-4](https://doi.org/10.1016/0009-2509(87)85067-4)
- Orlewski, P.M., Wang, Y., Hosseinalipour, M.S., Kryscio, D., Igglund, M., Mazzotti, M., 2018. Characterization of a vibromixer: Experimental and modelling study of mixing in a batch reactor. *Chem. Eng. Res. Des.* 137, 534–543. <https://doi.org/10.1016/j.cherd.2018.08.003>
- Plusa, T., Talaga, J., Duda, A., Duda, P., 2021. Modeling mixing dynamics in uncovered baffled and unbaffled stirred tanks. *AIChE J.* 67, 1–17. <https://doi.org/10.1002/aic.17322>
- Polaris, 2022. Omega 3 Market Share, Size, Trends, Industry Analysis Report.

- Prakash, B., Bhatelia, T., Wadnerkar, D., Shah, M.T., Pareek, V.K., Utikar, R.P., 2019. Vortex shape and gas-liquid hydrodynamics in unbaffled stirred tank. *Can. J. Chem. Eng.* 97, 1913–1920. <https://doi.org/10.1002/cjce.23433>
- Ramírez-Cruz, J., Salinas-Vázquez, M., Ascanio, G., Vicente-Rodríguez, W., Lagarza-Córtés, C., 2020. Mixing dynamics in an uncovered unbaffled stirred tank using Large-Eddy Simulations and a passive scalar transport equation. *Chem. Eng. Sci.* 222, 115658. <https://doi.org/10.1016/j.ces.2020.115658>
- Riha, V., Brunner, G., 2000. Separation of fish oil ethyl esters with supercritical carbon dioxide. *J. Supercrit. Fluids* 17, 55–64. [https://doi.org/10.1016/S0896-8446\(99\)00038-8](https://doi.org/10.1016/S0896-8446(99)00038-8)
- Rushton, J.H., Costich, E.W., Everett, H.J., 1950. Power characteristics of mixing impellers – part II. *Chem. Eng. Prog.* 46, 467–476. <https://doi.org/10.1016/b978-012220855-3/50007-9>
- Scargiali, F., Busciglio, A., Grisafi, F., Brucato, A., 2014a. Mass transfer and hydrodynamic characteristics of unbaffled stirred bio-reactors: Influence of impeller design. *Biochem. Eng. J.* 82, 41–47. <https://doi.org/10.1016/j.bej.2013.11.009>
- Scargiali, F., Busciglio, A., Grisafi, F., Brucato, A., 2014b. Mass transfer and hydrodynamic characteristics of unbaffled stirred bio-reactors: Influence of impeller design. *Biochem. Eng. J.* 82, 41–47. <https://doi.org/10.1016/j.bej.2013.11.009>
- Scargiali, F., Busciglio, A., Grisafi, F., Tamburini, A., Micale, G., Brucato, A., 2013. Power consumption in uncovered unbaffled stirred tanks: Influence of the viscosity and flow regime. *Ind. Eng. Chem. Res.* 52, 14998–15005. <https://doi.org/10.1021/ie402466w>

- Scargiali, F., Tamburini, A., Caputo, G., Micale, G., 2017. On the assessment of power consumption and critical impeller speed in vortexing unbaffled stirred tanks. *Chem. Eng. Res. Des.* 123, 99–110.
<https://doi.org/10.1016/j.cherd.2017.04.035>
- Seifried, B., Temelli, F., 2010. Interfacial tension of marine lipids in contact with high pressure carbon dioxide. *J. Supercrit. Fluids* 52, 203–214.
<https://doi.org/10.1016/j.supflu.2009.12.009>
- Sethian, J.A., 1999. Level set methods and fast marching methods : evolving interfaces in computational geometry, fluid mechanics, computer vision, and materials science. Cambridge University Press.
- Shinji Nagata, 1975. Mixing - principles and applications. Halsted Press, Tokyo, Japan.
- Speziale, C.G., Sarkar, S., Gatski, T.B., 1991. Modelling the pressure–strain correlation of turbulence: an invariant dynamical systems approach. *J. Fluid Mech.* 227, 245–272. <https://doi.org/10.1017/S0022112091000101>
- Tamburini, A., Brucato, A., Ciofalo, M., Gagliano, G., Micale, G., Scargiali, F., 2021. CFD simulations of early- to fully-turbulent conditions in unbaffled and baffled vessels stirred by a Rushton turbine. *Chem. Eng. Res. Des.* 171, 36–47. <https://doi.org/10.1016/j.cherd.2021.04.021>
- Tamburini, A., Cipollina, A., Micale, G., Scargiali, F., Brucato, A., 2016. Particle Suspension in Vortexing Unbaffled Stirred Tanks. *Ind. Eng. Chem. Res.* 55, 7535–7547. <https://doi.org/10.1021/acs.iecr.6b00824>
- Tanaka, M., Noda, S., O'shima, E., 1986. Effect of the Location of a Submerged Impeller on the Enfoldment of Air Bubbles From the Free Surface in a Stirred Vessel. *Int. Chem. Eng.* 26, 314–318.

- Tryggvason, G., Bunner, B., Esmaeli, A., Juric, D., Al-Rawahi, N., Tauber, W., Han, J., Nas, S., Jan, Y.J., 2001. A front-tracking method for the computations of multiphase flow. *J. Comput. Phys.*
- Wen, Y.-Q., Xue, C.-H., Zhang, H.-W., Xu, L.-L., Wang, X.-H., Bi, S.-J., Xue, Q.-Q., Xue, Y., Li, Z.-J., Velasco, J., Jiang, X.-M., 2023. Concomitant oxidation of fatty acids other than DHA and EPA plays a role in the characteristic off-odor of fish oil. *Food Chem.* 404, 134724.
<https://doi.org/10.1016/j.foodchem.2022.134724>
- Whitaker, S., 1999. *The Method of Volume averaging*. Kluwer Academic Publishers, Dordrecht.
- Wójtowicz, R., 2017. Flow pattern and power consumption in a vibromixer. *Chem. Eng. Sci.* 172, 622–635. <https://doi.org/10.1016/j.ces.2017.07.010>
- Wójtowicz, R., 2014. Choice of an optimal agitated vessel for the drawdown of floating solids. *Ind. Eng. Chem. Res.* 53, 13989–14001.
<https://doi.org/10.1021/ie500604q>
- Wörner, M., 2003. *A Compact Introduction to the Numerical Modeling of Multiphase Flows*. Rep. Forschungszentrum Karlsruhe FZKA 38.
- Xiong, X., Wang, S., Liu, P., Tao, C., Wang, Y., Liu, Z., 2023. Numerical investigation on intensified mixing performance with modified dual impeller. *Chem. Eng. Sci.* 274. <https://doi.org/10.1016/j.ces.2023.118698>
- Yakovenko, V.P., 1994. Optimizing agitator shapes for reciprocating mixers. *Chem. Pet. Eng.* 30, 56–61. <https://doi.org/10.1007/BF01147794>
- Youngs, D.L., 1982. *Time-Dependent Multi-Material Flow with Large Fluid Distortion*. Academic Press.
- Zamiri, A., Chung, J.T., 2017. Ability of URANS approach in prediction of unsteady turbulent flows in an unbaffled stirred tank. *Int. J. Mech. Sci.* 133, 178–187. <https://doi.org/10.1016/j.ijmecsci.2017.08.008>