

POLITECNICO DI TORINO

Master's Degree in Mechanical Engineering

A.Y. 2025/2026



**Politecnico
di Torino**

Master's Degree Thesis

Computational Fluid Dynamics Modelling of Carbon Capture in a Spray Tower Using Reactive Liquid Absorption

Supervisor:

Prof. Luca Brandt

Co-supervisor:

Prof. Cristophe Duwig

Candidate:

Daniele Crescimbeni

July 2026

Abstract

The increasing need to limit the effects of climate change and comply with stringent CO₂ emission targets has driven industrial interest in post-combustion carbon capture technologies. Among these, spray towers represent a promising solution due to their simple design, low pressure drop, and direct gas–liquid contact.

This thesis develops a Computational Fluid Dynamics (CFD) model of a pilot-scale, multi-nozzle spray tower designed to capture CO₂ from the exhaust gases of a waste-to-energy facility using a 15 wt% aqueous NaOH solution as the absorbent. The objectives are to investigate the influence of the main operating parameters on the absorption process, to integrate an evaporation model and assess its impact on the overall process behaviour, and to identify strategies for improving the overall CO₂ capture efficiency under industrial conditions.

The simulations are carried out in STAR-CCM+ using an Euler–Lagrange framework and the Dispersed Multiphase Model. The continuous gas phase and the dispersed liquid droplets were solved simultaneously, accounting for droplet transport, chemical reactions between absorbed CO₂ and NaOH within the liquid phase, interphase mass transfer, and energy exchange. A quasi-steady droplet evaporation model is implemented to evaluate the relevance of evaporation and its dependence on operating conditions. The model captures complex phenomena within the tower, including droplet dispersion, turbulent mixing, and interphase mass transfer, which are difficult to investigate experimentally. Different liquid-to-gas ratios, droplet-size ranges, and nozzle configurations are examined.

The results indicate that increasing the liquid-to-gas ratio generally improves overall capture efficiency by providing more absorbent, extending droplet residence time, and promoting CO₂ mass transfer. Smaller droplets further enhance absorption efficiency due to their better dispersion, higher surface-area-to-volume ratio, and longer residence time. Multi-nozzle configurations contribute to a more uniform liquid distribution and improved gas–liquid contact within the tower. Evaporation is more significant for smaller droplet-size ranges, whose longer residence times and higher reaction rates lead to a stronger temperature rise within the droplets, promoting evaporation. By contrast, its influence remains limited for larger droplets and lower liquid-to-gas ratios. Nevertheless, the inclusion of evaporation affects droplet temperature, diameter, reaction rate, and local mass-transfer conditions and therefore contributes to a more complete description of the absorption process.

Overall, the developed model provides a computational tool for analysing reactive CO₂ absorption in pilot-scale spray towers. Although droplet deformation, coalescence, collisions, and internal circulation are neglected to ensure computational feasibility, future work could improve droplet modelling by incorporating these phenomena and adopting interface-resolving approaches to improve the prediction of liquid phase dynamics, reaction rates, and heat and mass transfer.

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1 Introduction

Over the past decades, attention to global climate change has increased due to growing concern about global warming and the intensification of extreme weather events. In particular, 2024 was the warmest year recorded since 1850, with a global average surface temperature of 1.46 °C above pre-industrial level [1]. The scientific community recognizes that the increase in greenhouse gases (GHG) concentrations is a primary cause of this trend [2]. GHGs, including carbon dioxide (CO₂), methane (CH₄), nitrous oxide (N₂O), ozone (O₃) and Water Vapour, are naturally present in the Earth's atmosphere and contribute to the greenhouse effect. Because of their radiative properties, these gasses absorb part of the outgoing infrared radiation emitted by the Earth's surface and re-emitting it downward, warming the lower atmosphere and the surface and helping to maintain planetary energy balance; without this effect, Earth's average surface temperature would be below the freezing point of water, making life as we know it impossible [3].

However, since the Industrial Revolution, atmospheric GHG concentrations have been increasing, due to anthropogenic emissions from fossil fuel combustion and industrial processes, leading to a rise in global average surface temperature [4]. Among all GHGs, CO₂ is the largest contributor and is long-lived in the atmosphere [5],[6]making its reduction a global priority. In 2024 fossil carbon dioxide emissions reached 37.4 billion tonnes, 0.8% higher than in 2023, [7] leading to record atmospheric CO₂ concentrations of 422.5 ppm (parts per million), about 50% higher than the pre-industrial level [8].

In response to the continued growth of anthropogenic emissions, the United Nations has promoted international agreements aimed at reducing GHG emissions. Starting from Kyoto Protocol (1997), which required industrialized countries to reduce emissions by 5% below 1990 levels over the period 2008–2012 [9], to the Paris Agreement (2015), which aims to limit warming to 1.5 °C above pre-industrial levels through collective action by all parties in order to reduce climate risks and avoid irreversible impacts [10]. In addition, in 2018 the European Commission submitted a long-term strategy to reduce GHG emissions and reach climate neutrality (net-zero GHG emissions), defining seven strategic priorities, including the deployment of Carbon Capture and Storage (CCS) systems [11].

1.1 Motivation

Despite the policies adopted to reduce carbon dioxide emissions, global CO₂ emissions increased again in 2024. The main drivers were the continued rise in global energy demand by 2.2 %, with the power sector leading the increase. Electricity consumption grew particularly in emerging and developing economies, which accounted for more than 80% of the additional demand. After several years of reduction, energy use in advanced economies also rose by almost 1% in 2024. The growth in electricity demand is driven by higher cooling necessities due to extreme temperatures, increased industrial electricity use, the electrification of transport and the expansion of the data centre sector [8]. Beyond electricity and heat generation, which emit CO₂ through fossil fuel combustion, anthropogenic emissions derive from process products industries such as cement, steel, and chemicals, transportation, manufacturing and construction sector, buildings sector and land-use change and forestry [12]. The distribution of CO₂ emissions across the main sectors is reported in Figure 1.1 [13].

1 Introduction

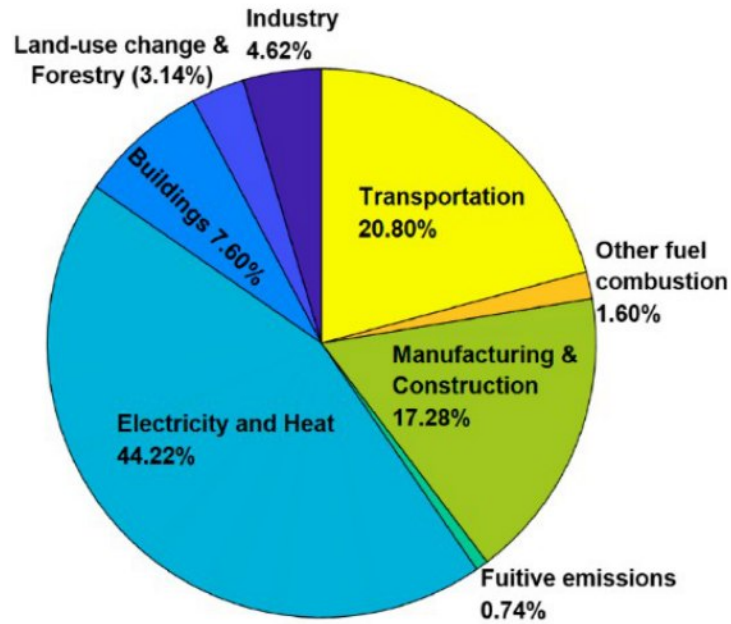


Figure 1.1 - Distribution of global CO₂ emission [13]

CCUS is widely recognized as essential for meeting the targets settled by the normative and to reach net-zero emission regulations. The International Energy Agency (IEA) and the Carbon Sequestration Leadership Forum (CSLF) estimate that achieving net-zero emissions in the energy sector by 2050 will require global CCS capacity to grow from 2020 levels by 10–15 times that level by 2030 and about 100 times by 2050 [14]. Carbon capture technology will play a central role in decarbonizing hard-to-abate industrial sectors where renewables are not economically viable [15].

As of 2025, 77 CCUS facilities are operational worldwide and 734 more in the development, increasing from 50 and 628, respectively, in the previous year. While this indicates growing interest in the technology, this is still not enough to meet climate targets. Additionally, challenges persist, such as unstable policy and financing frameworks, as well as low public acceptance [16]. The capture process accounts for about 70% of total CCUS investment costs, and the absence of mature financial policies, incentives, and government funding slows down cost reductions from technological progress and learning-by-doing effects. CCUS costs include capital cost for the plant, which can be reduced retrofitting the technology in existing plant, and operational cost, from efficiency drop of around 9% due to the capture process [17]. Therefore, studying the capture process and evaluating methods to increase its efficiency is crucial to reduce overall cost, accelerating technology adoption, and helping large emission sources meet regulatory goals.

1.2 Carbon Capture Utilisation and Storage

Carbon Capture, Utilisation and Storage (CCUS) refer to the capture of CO₂, typically from extensive point sources such as power generation plants or industrial facilities, which use fossil fuels or biomass. The captured CO₂ can be used directly on-site after the sequestration; otherwise it is compressed and transported by pipeline, ship, truck, or rail for utilisation in different applications. Alternatively, it can be injected for long-term storage into deep geological formations [18]. Three main CO₂ capture routes are commonly used in

fossil-fuel power plants classified, based on the process stage at which CO_2 is separated: pre-combustion, oxy-combustion, and post-combustion capture, each of them with distinct advantages and disadvantages in terms of efficiency, cost, and applicability. Pre-combustion CO_2 capture involves separating carbon dioxide from fossil or biomass fuels before the final combustion step. Oxyfuel combustion involves burning fuel with pure oxygen instead of air, producing flue gas primarily consisting of CO_2 and H_2O , which can easily be purified by condensation of the water vapor. Post-combustion capture involves separating CO_2 from flue gas primarily consisting of N_2 , O_2 , and H_2O after fossil fuel combustion. This work will focus on post-combustion capture plants as it is the most mature and promising, with proven applicability to both existing and new power plants. Extensive studies including laboratory and pilot-scale experiments, modelling, and simulations have been carried out, and new ones continue to advance the technology [19]. Capturing the CO_2 from the flue gas can be achieved through different methods, including absorption, adsorption, cryogenic separation, membrane separation, and chemical looping each with its own strength and limitations [20]. Currently, absorption methods, remain the most reliable for CO_2 capture offering high processing capacity and extensive industrial operational experience, in particular chemical absorption dominates industrial applications thanks to its high efficiency, proven reliability, and moderate solvent requirements [21]. Carbon capture by chemical absorption is further investigated in the following sections as functioning principle of the carbon capture plant proposed as case study.

Absorber typology is a key aspect of CO_2 capture by chemical absorption, as it determines gas-liquid contact behaviour and thus overall performance of the process. The available options for the absorber are: tray columns, packed bed columns and spray columns [22]. Their main characteristics and differences are discussed in Chapter 2. In this work, attention is focused on the spray column configuration.

1.3 Computational Fluid Dynamics

CO_2 capture using aqueous NaOH solution involves a multiphase gas-liquid flow, specifically a dispersed phase flow where discrete droplets of the solvent solution are carried through a continuous flue gas phase [23]. To study the evolution of the process, a Computational Fluid Dynamics (CFD) analysis is used for this work. CFD predicts fluid flow, mass transfer, chemical reactions, and related phenomena by numerically solving the governing equations. This approach allows to obtain numerical approximate solutions of fluid flows and complements lab testing and experimentation while cutting total effort.

The CFD approach starts with a mathematical model of the physical problem. The conservation equations for mass, species, momentum, and energy are solved across the domain using suitable initial and boundary conditions. Fluid properties are modelled empirically, and physics are simplified to keep the problem tractable. In the fluid region, the governing differential equations are discretized into algebraic equations using numerical methods. The domain is divided into a finite set of control volumes or cells to create the mesh. These algebraic equations are solved simultaneously and iteratively for flow variables in each cell until convergence. CFD simulations offer key advantages over physical experiments: relatively low cost, high computational speed but dependent on the problem, the ability to simulate real or ideal conditions, especially those difficult to test physically, and comprehensive data across the entire domain of interest. Limitations include reliance on physical models for turbulence, compressibility, chemistry and multiphase flow, unavoidable numerical errors and the need for well-defined boundary and

initial conditions [24]. The mathematical models and numerical methods used to simulate the process are presented in Chapters 3 and 4.

1.4 Case study

The system modelled in this work consists of a pilot-scale carbon capture unit developed and constructed by Grimaldi Development AB in collaboration with KTH Royal Institute of Technology at the Tekniska verken waste-to-energy plant in Linköping, Sweden. A schematic representation of the system is shown in Figure 2.7, where the main components are the spray tower, centrifugal separator, humidifier, and solvent circulation tanks. In this work, only the 4-meter-tall spray tower section is modelled and simulated, operating at atmospheric pressure and with flue gas containing 11% mol CO_2 . The solvent used is a 15 wt% NaOH solution, sprayed in a multi-nozzle counter-current configuration.

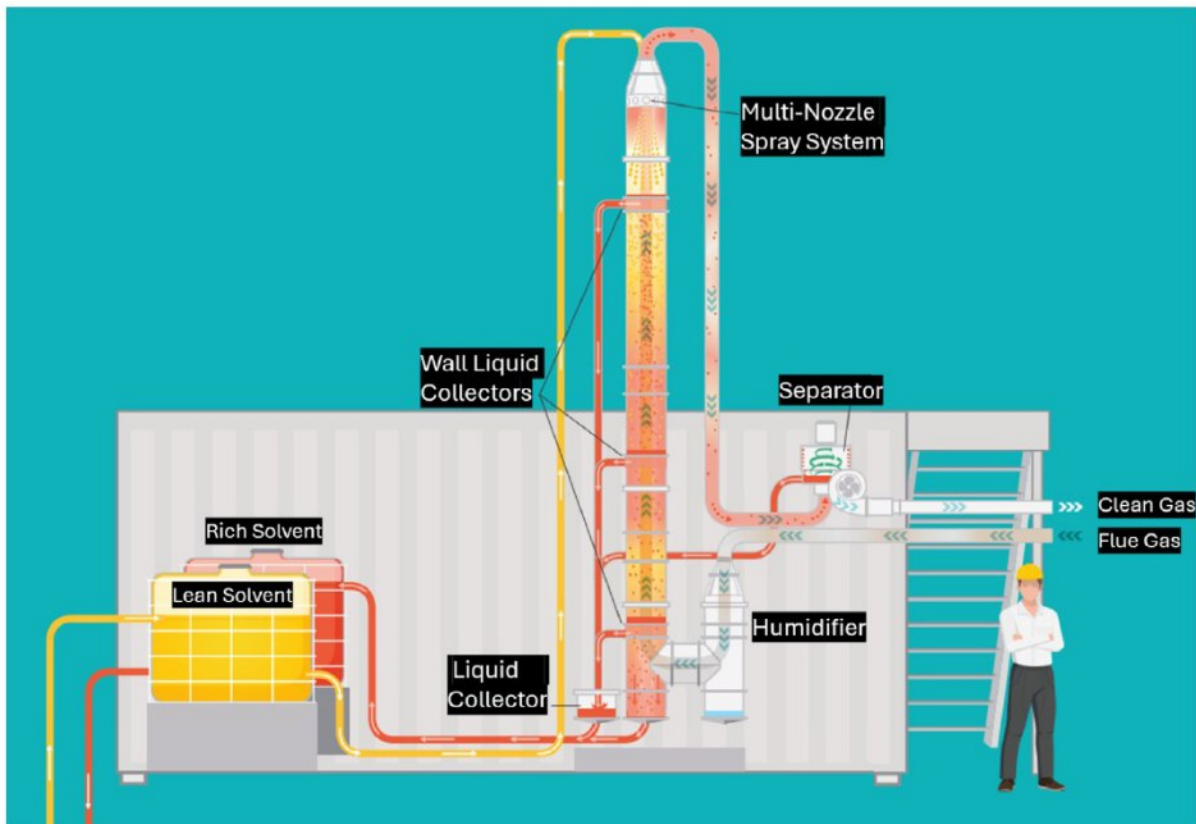


Figure 1.2 - Schematic representation of spray tower system [25]

1.5 Aim of the thesis

The aim of this thesis is to develop a detailed computational fluid dynamics (CFD) analysis adopting a Euler-Lagrange multiphase approach to simulate post-combustion CO_2 capture via chemical absorption with NaOH solvent in a spray-tower. Building on the work of A. Najarneshadmashtadi et. al. (2025) [25], whose model was validated against experimental data from a pilot-scale plant, this study incorporates droplet evaporation, which was initially neglected, representing a step forward toward more realistic operating conditions. The model enables the evaluation of capture process performance under varying design and operating parameters, such as gas and liquid flow rates, droplets diameters and nozzles configuration. Furthermore, it captures the complex phenomena occurring during

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the process, which are difficult to detect experimentally, and it simulates reactive gas-liquid interaction, mass transfer, and droplet motion under industrial conditions.

The main objectives of this study are:

- Development of a 2D Eulerian-Lagrangian multiphase spray tower model including droplet evaporation for single and multi-injector configurations.
- Evaluate the impact of varying the main operating parameters, as liquid-to-gas ratio, nozzles configuration, and droplet diameter distributions, on the absorption process.
- Quantify the impact of droplets evaporation on key capture process outputs by comparing the results to the baseline model without evaporation.
- Analyse the effects of main operating parameters on droplets evaporation.

Research on spray towers has been dominated by laboratory- and pilot-scale experiments that neglect real industrial conditions, such as high gas-liquid flow rates and multiple spray layers, which significantly affect absorption performance. Although numerical models for CO₂ capture have emerged in recent years, they typically focus on single or few droplets rather than the full process. Large-scale CFD simulations under industrial conditions remain scarce in the literature, despite their ability to reveal experimentally inaccessible phenomena while saving time and cost. In addition aqueous NaOH represents a promising non-amine alternative due to the low-cost, still scarce investigate in spray towers under industrial conditions. Developing a model that couples gas-liquid hydrodynamics, heat/mass transfer, chemical reactions, and droplet evaporation is therefore essential for advancing spray-tower technology.

2 Theoretical Background

2.1 Post-combustion carbon capture with chemical absorption

A schematic of a post-combustion capture system is illustrated in Figure 2.1. The capture unit is located downstream of purification systems, necessary to remove SO_x, NO_x, and particulates from the stream gas. Post combustion is the most applied method in current power generation facilities. However, its main weakness is the low mass transfer due to the low CO₂ partial pressure, as the content is between 13-15% [20].

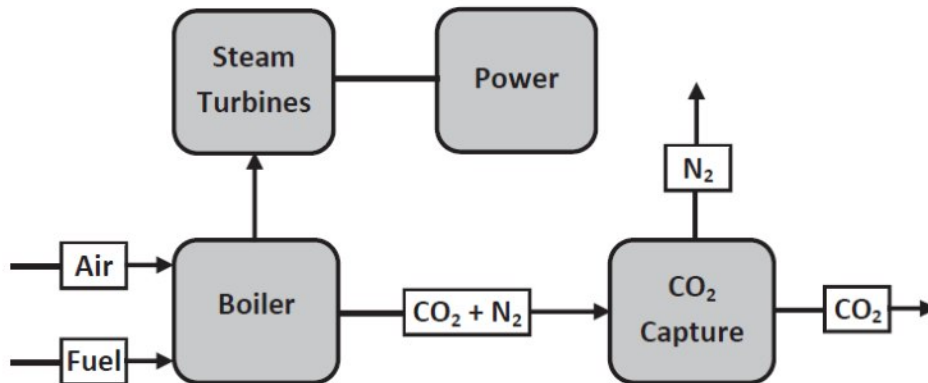


Figure 2.1 - Schematic of Post-combustion CO₂ Capture system [26]

2.1.1 Chemical absorption

Chemical absorption is the most established and widely studied method for post-combustion CO₂ separation. In this process, CO₂-rich flue gas is contacted with a solvent, forming a weakly bound intermediate compound through a reversible chemical reaction; successive regeneration of the solvent releases a concentrated CO₂ stream [27]. The main advantages of the method are:

- high CO₂ capture efficiency (typically 90–98%);
- easy retrofitting to existing plants,
- the highest technology maturity among CO₂ separation methods;
- effective operation at low CO₂ partial pressures.

However, chemical absorption also has drawbacks, including sensitivity to impurities like moisture, NO_x, and SO_x, high environmental impacts from solvent toxicity and degradation, dependence on CO₂ concentration for efficiency, substantial capital costs for equipment, and high energy demands for solvent regeneration [28]. CO₂ capture represents an added cost to electricity generation, so energy penalties, operational, and capital expenses must be justified primarily by environmental benefits. To enhance the chemical absorption process, strict control of oxygen, SO_x, NO_x, fly ash, and soot in the flue gas stream is required, since these pollutants negatively affect absorber performance. In addition, an accurate selection of a low-volatile solvent with low degradation rates and regeneration energy, as well as good thermal and chemical stability, is fundamental for achieving high absorption efficiency [29]. These aspects are discussed in the following subchapters.

2.1.2 Process description

A CO₂ capture system including all major equipment is shown in Figure 2.2 [30]. The process consists of two main steps, the absorption of CO₂ by the solvent in the absorber, and the regeneration of the rich liquid solution in the stripper with CO₂ release [28].

Prior to absorption, flue gas pretreatment removes process contaminants that can affect the process. SO₂ is removed via flue gas desulfurization to below 10 ppm and NO_x is reduced using selective catalytic reduction, selective non-catalytic reduction, or low-NO_x burners, as these contaminants form heat-stable salts with solvents. Fly ash and particulates are captured by filters or electrostatic precipitators. Oxygen levels are also controlled to prevent corrosion and solvent degradation, or corrosion inhibitors are added [31].

After these steps, flue gas is cooled to 45–50°C in a direct contact cooler prior to entering the absorber, to enhance CO₂ absorption and minimize solvent losses due to evaporation [31]. Inside the absorber, at low temperature and high pressures, gas-liquid contact occurs and the liquid solvent absorbs CO₂. The CO₂-rich solution exits the absorber bottom, while flue gas depleted of CO₂ is released to atmosphere after water wash at the top, which cools the column and recovers solvent vapours [21]. The exothermic absorption heats the solvent which, before entering the stripper top, flows through a lean/rich cross-heat exchanger and is preheated by hot CO₂-lean solvent from the stripper.

The endothermic desorption requires high regeneration temperatures of 100–120°C and low pressures, shifting the chemical equilibrium to release CO₂ at ~1–2 bar. Steam from a reboiler, typically extracted from turbine or generated via heat recovery steam generator, provides the required heat; this results in energy penalties of up to 20% reduced power output and 10% lower plant efficiency [29],[30]. Regenerated lean solvent exits the stripper bottom, is cooled through the heat exchanger and a cooler, and pumped back to the absorber top. The purified gas, rich in CO₂, is cooled and sent to a flash drum, where condensed solvent and water are separated and recycled. CO₂ is then compressed, dried, and transported for sequestration [31].

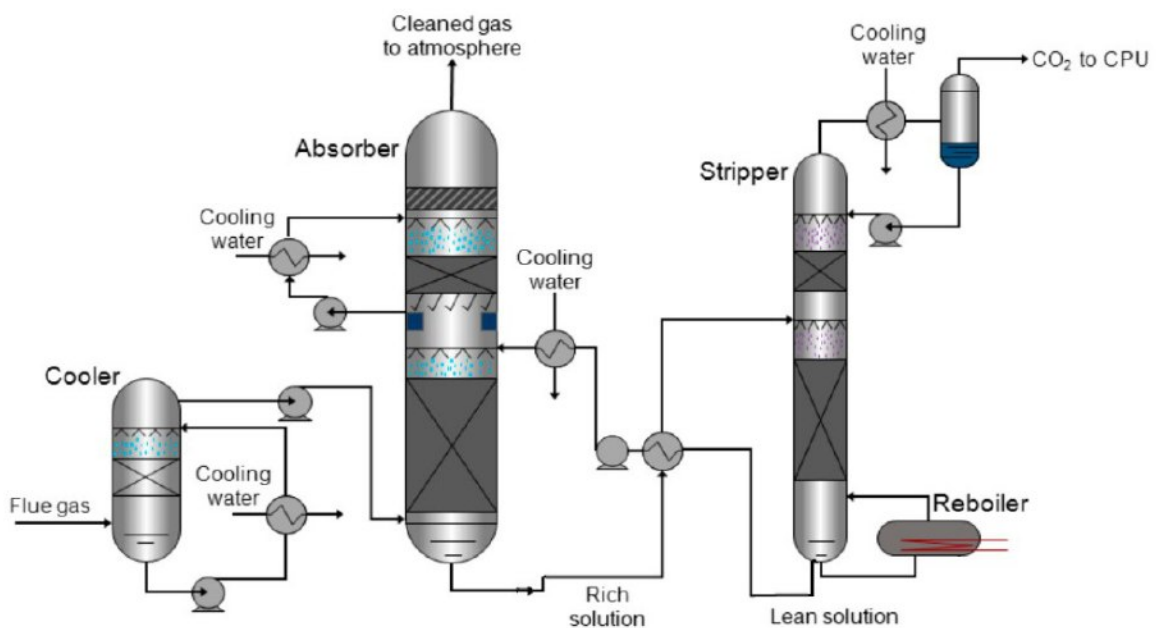


Figure 2.2 - Chemical absorption capture process equipment [30]

2.1.3 Solvents for chemical absorption

Chemical solvents react reversibly with CO_2 in the absorber, binding it selectively and releasing it during stripper regeneration. Chemical reaction enhances CO_2 absorption rate for two reasons [32]:

- Liquid-phase reaction lowers equilibrium partial pressure, increasing mass transfer driving force;
- CO_2 consumption at gas-liquid interface raises the concentration gradient.

Capital and operating costs of conventional solvents remain high due to high energy demand for regeneration, equipment corrosion, solvent losses from evaporation, and degradation by oxygen. Targeted solvent selection reduces costs, and is guided by feed gas properties such as composition, pressure, and temperature, along with treated gas specifications. CO_2 partial pressure in feed and product directly affects solvent circulation rate, which determines equipment size and regeneration energy duty, thereby defining overall capital and energy costs. The Ideal properties of the solvents are the following [33]:

- High reaction kinetics for fast absorption and regeneration;
- Low heat of reaction to minimize regeneration energy;
- High CO_2 equilibrium solubility, for a given partial pressure, for reduced circulation and easier stripping ;
- Minimal corrosivity to lower capital costs;
- Excellent stability to avoid degradation treatment.

However, ideal solvents are challenging to develop, as desirable traits involve trade-offs, for instance, high absorption capacity often matches with poor stripping performance. Major chemical absorbent groups include [34], [35]:

- Amines: Primary (MEA), secondary (MMEA, DEA, etc.), tertiary (DMAE, MDEA)
- Carbonates: Calcium carbonate CaCO_3 , Sodium carbonate Na_2CO_3 and Potassium carbonate K_2CO_3 .
- Hydroxides: Calcium hydroxide $\text{Ca}(\text{OH})_2$ and Sodium Hydroxide NaOH
- Ionic liquids: Imidazolium-, ammonium-, phosphonium-based and task-selective ionic liquids (TSILs).
- Amino acid salts: natural (taurine, proline, and arginine) and synthetic (piperazine derivatives, etc.)

In addition the using novel hybrid absorbents, by blending different solvent types, incorporating promoters, or adding performance enhancers permits mixing the best features from different families, improving CO_2 capture performance. Popular hybrids solvents include alkanolamine-ionic liquid (IL) blends (MEA-IL, MDEA-IL), amino acid-amine mixtures (Proline/Choline + PZ, Taurine + MEA), and non-aqueous/blended amines (nanoparticle-amine, PZ + AMP) [34].

Another solvent option to capture is ammonia (NH_3), which offers a cost-effective alternative with high CO_2 capacity, low regeneration energy from low reaction heat, low corrosivity, and good oxygen resistance. Ammonia processes differ by absorber temperature into two variants: aqueous NH_3 at ambient temperature, which suffers from

NH₃ slip and evaporation losses, or chilled NH₃ at 0–20°C to reduce slip, volatility, and energy penalty [36].

Among different absorbents, MEA, a primary amine, remains the reference for carbon dioxide capture research due to its strong basicity, fast kinetics, good CO₂ capacity, high efficiency, and chemical reactivity, forming stable carbamates that decompose at high temperature to release CO₂. However, the primary drawback is high regeneration energy, accounting for 70 to 80 percent of operating expenses. 30%wt MEA solution constitutes the benchmark for optimization research [37]. This work uses NaOH as solvent, which has lower cost and higher CO₂ absorption capacity than MEA. In addition it is less sensitive to CO₂ concentration of the inlet gas and reaches high efficiency with lower solvent concentration, which is favourable for industrial applications, decreasing operational costs and environmental impact [34], [38]

2.1.4 Absorber typology

In addition to solvent selection, absorber configuration optimizes CO₂ capture via gas-liquid contacting efficiency. The available options for the absorber are [22]:

- Tray column: vertical shell with several perforated trays designed to guide the liquid solvent down from the top injection point to the bottom. Meanwhile, the gas rises from the bottom, contacting the liquid as it passes through the column. High pressure drop, makes them prone to flooding, which reduces efficiency and mass transfer; in addition, they suffer corrosion, with high maintenance costs.
- Packed bed column: column full of material packed between perforated plates. The injected liquid solvent flows through the packing, creating liquid films with large interfacial area, promoting absorption. Despite large gas residence time, which enhances capture efficiency, high pressure drop, high packing replacement costs and flooding risk present serious drawbacks.
- Spray column: empty columns with solvent injected by one or multiple nozzles that atomize it into numerous droplets interacting with rising gas. Absorption benefits from the large contact gas-liquid contact area, due to fine droplet size with high surface area-to-volume ratio. Advantages include also very low pressure drop, low equipment cost, and possibility to work with smaller columns.

Figure 2.6 shows a schematic of a counter-current spray tower used for gas absorption. Post-combustion carbon capture has been dominated by research on tray and packed bed columns; instead, spray towers have been studied primarily in flue gas desulfurization units for sulphur dioxide (SO₂) sequestration [39]. Only in recent years, the interest in spray towers for CO₂ capture has been increasing, leading to several studies and experimental investigations. Comparison between packed bed and spray columns in laboratory-scale experiments using MEA as a solvent highlight the higher mass-transfer capability of the latter under various operating parameters, such as CO₂ partial pressure, gas and liquid flow rates, MEA concentration, CO₂ loading, and nozzle size. This improved performance is primarily due to the larger interfacial area and the availability of three principal diffusion directions generated by nozzle atomization, as opposed to the single diffusion direction across the liquid film in packed beds [39], [40]. Despite of this, spray towers exhibit higher non-uniformity in parameters such as gas velocity, droplet size distribution, spatial dispersion, droplet collision, and wall adhesion. Consequently, the overall CO₂ capture efficiency, both at laboratory and pilot scales, is typically higher in packed columns, which display more

2 Theoretical Background

uniform hydrodynamic behaviour [41]-[43]. Nevertheless, improving the configuration of spray towers and their components, particularly the design of the nozzles, has demonstrated potential for enhancing efficiency and process performance. Moreover, the lower gas-phase pressure drop and reduced construction and maintenance costs have further increased the attractiveness of spray towers, making them a promising technology for carbon capture applications [43].

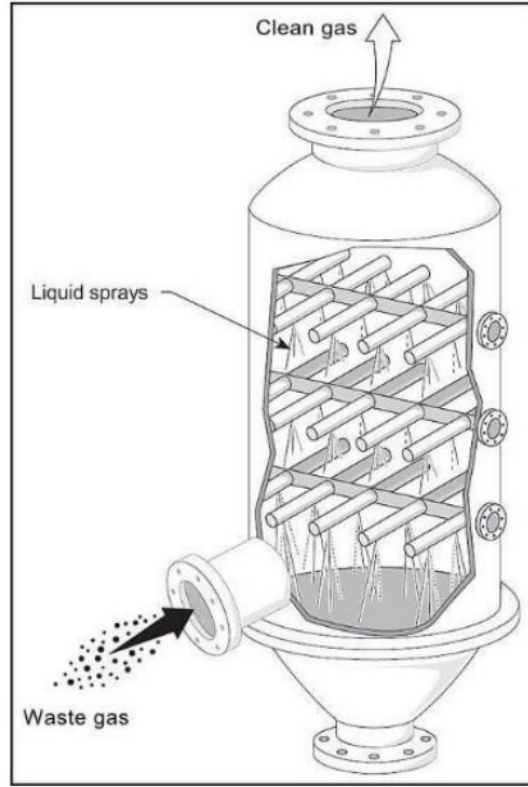


Figure 2.3 - Counter-current spray tower system [44]

2.2 Fluid dynamics governing equations

Fluid dynamics is the branch of continuum mechanics that studies the motion of fluids; it is governed by the conservation equations of mass, momentum, and energy. These equations represent the mathematical statements of three fundamental physical principles: conservation of mass (continuity equation, (2.1)), Newton's second law (momentum equation, (2.2)), and the first law of thermodynamics (energy equation, (2.3)) [45]. The differential formulation is presented in the following equations, where i, j , and k are the spatial coordinate:

- Continuity equation:

$$\frac{\partial \rho}{\partial t} + \frac{\partial(\rho u_j)}{\partial x_j} = 0 \quad (2.1)$$

Where ρ is the fluid density, t is the time, x_j is the j -th spatial coordinate, and u_j is the j -th component of the velocity vector.

- Momentum equation:

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$$\frac{\partial(\rho u_i)}{\partial t} + \frac{\partial(\rho u_j u_i)}{\partial x_j} = -\frac{\partial p}{\partial x_i} + \frac{\partial \tau_{ij}}{\partial x_j} + f_i \quad (2.2)$$

Where p is the fluid pressure, τ_{ij} is the viscous stress tensor, and f_i is the body force per unit volume.

- Energy equation:

$$\frac{\partial(\rho e)}{\partial t} + \frac{\partial(\rho e u_i)}{\partial x_i} = -p \frac{\partial u_i}{\partial x_i} + \frac{\partial}{\partial x_j} \left(\lambda \frac{\partial T}{\partial x_j} \right) + \Phi + S_E \quad (2.3)$$

Where e is the total energy per unit mass, λ is the conductive heat coefficient, T is the temperature, Φ is the viscous dissipation function and S_E is a volumetric energy source term.

These five partial differential equations, mass conservation, x-,y- and z- momentum equations and energy equations, describe the motion of a fluid in three dimensions. The total energy e is related to the internal energy i through the relation (2.4):

$$e = i + \frac{1}{2} \rho u^2 \quad (2.4)$$

Through thermodynamic equilibrium assumption, is possible to relate the thermodynamic variables of the fluid, ρ , T , p and i with the equations of state (2.5) and (2.6) and close the system of equations [45]:

$$p = \rho R T \quad (2.5)$$

$$i = c_v T \quad (2.6)$$

Where c_v is specific heat at constant volume, R is the universal constant for gas.

Since the case study involves a multiphase reacting flow with both the gas and liquid phases are mixtures of different components, the governing equations must be extended to account for chemical species transport. These species also participate in chemical reactions, so the corresponding reaction rates need to be explicitly modelled. Each species k is characterized by its mass fraction Y_k for $k = 1, \dots, N$, denoting the number of species, defined in equation (2.7):

$$Y_k = \frac{m_k}{m} \quad (2.7)$$

where m_k is the mass of species k in a given control volume and m is the total mass of the mixture in that volume. The conservation equation for the mass fraction of species k is presented in equation (2.8):

$$\frac{\partial(\rho Y_k)}{\partial t} + \frac{\partial(\rho u_i Y_k)}{\partial x_i} = \frac{\partial}{\partial x_i} \left(\rho D_k \frac{W_k}{W} \frac{\partial X_k}{\partial x_i} \right) + \dot{\omega}_k \quad (2.8)$$

D_k denotes the effective mass-diffusion coefficient of species k in the mixture, W_k and W are the molecular weights of species k and of the mixture, respectively, and X_k is the molar fraction of species k . $\frac{\partial(\rho u_i Y_k)}{\partial x_i}$ is the convection term, while $\frac{\partial}{\partial x_i} \left(\rho D_k \frac{W_k}{W} \frac{\partial X_k}{\partial x_i} \right)$ is the diffusion term. The source term $\dot{\omega}_k$ represents the net production rate

of species k per unit volume due to chemical reactions. X_k is the molar fraction, defined by equation (2.9) [46]:

$$X_k = \frac{W}{W_k} Y_k \quad (2.9)$$

2.3 Turbulent flows

The flow considered in this work, like most flows encountered in engineering applications, is turbulent. Equation (2.10) define the Reynolds number, which distinguish turbulent and laminar regimes [46]:

$$Re = \frac{\rho UL}{\mu} \quad (2.10)$$

It represents the ratio between inertial and viscous forces, where ρ is the fluid density, U a characteristic velocity, L a characteristic length scale, and μ the dynamic viscosity. At low Reynolds numbers, viscous forces dominate and the flow remains laminar, whereas at sufficiently high Reynolds numbers inertial effects prevail and the flow becomes turbulent [45][46]. Turbulent flows exhibit several characteristic properties:

- Highly unsteady in time.
- Three-dimensional.
- Characterized by intense vorticity, with vortex stretching acting as a key mechanism for turbulence amplification.
- Greatly enhancing of mixing of conserved quantities through turbulent diffusion, which increase the effective transport of mass, momentum, and energy.
- Turbulent diffusion is dissipative so the interactions between fluid elements with different momentum lead to irreversible loss of mean kinetic energy, which is converted into internal energy by viscous effects.
- Presence of coherent structures that play a central role in mixing and momentum transfer.
- High fluctuations over a wide range of length and time scales.

Turbulence can intensify mixing by orders of magnitude, which is advantageous for processes requiring rapid chemical mixing and heat transfer, such as spray-tower absorption. However, this behaviour is inherently coupled with an increase in frictional forces and higher pressure drop

There are different approaches to predict turbulent flows, like direct numerical simulation (DNS), Reynolds averaged Navier–Stokes (RANS), and large eddy simulation (LES). Starting from DNS, which solves the governing equations (2.1) to (2.9) for all the motions in space and time. DNS is the most accurate turbulence-simulation strategy, since no modelling or averaging of the governing equations is introduced beyond numerical discretisation. However, it must resolve the smallest dissipative scales, of the order of the Kolmogorov length η , where viscous effects dominate, so the grid spacing must be comparable to η . As a result, the number of required grid points scales very steeply with Reynolds number increase and DNS is therefore restricted to relatively simple geometries and moderate Reynolds numbers. DNS method is therefore used for highly resolved statistical analysis and validation of turbulence models [47].

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The second major approach is based on decomposing the instantaneous flow variables into mean and fluctuating components, which leads to a set of unclosed equations that require turbulence modelling. This can be done in two main ways:

- By time or ensemble averaging the Navier–Stokes equations, obtaining a system of partial differential equations known as the Reynolds averaged Navier–Stokes (RANS) equations, in which all turbulent scales are modelled through additional closure relations.
- By applying a spatial filter to the Navier–Stokes equations, explicitly resolving the large-scale motions while modelling only the small-scale eddies, which leads to the large eddy simulation (LES) equations.

In this work, the RANS formulation is adopted to model turbulence and it's presented in the next chapter [47].

2.4 NaOH chemical reactions mechanism

The CO₂ absorption process in aqueous NaOH is governed by a sequence of fast reactions described in the literature [48],[49]. First, NaOH is a strong alkaline which fully dissociates in pure water into sodium ions (Na⁺) and hydroxide ions (OH⁻). Before the chemisorption, when gaseous CO₂ enters the droplet, it is physically dissolved in the liquid phase and forms aqueous CO₂, as described in equation (2.11):



Equations (2.12) and (2.13) display the subsequent reaction of dissolved CO₂ with OH⁻ to form bicarbonate (HCO₃⁻) and, consequently, carbonate (CO₃²⁻):



Reactions (2.12) and (2.13) are reversible and fast in the high-pH range. Since the CO₂ concentration is assumed constant in this work, reaction (2.12) is treated as a first-order reaction, even though the true elementary reaction is second order. Reaction (2.13) is performed right after reaction (2.12), therefore during overall reaction aqueous CO₂ does not accumulate because it reacts immediately with OH⁻ after formation. At the beginning of the absorption process, the dominant reaction is (2.13) due to the strong alkalinity of the solution, implying strong conversion of CO₂ and OH⁻ into CO₃²⁻ in respect of HCO₃⁻ formation. This leads to rapid consumption of OH⁻, a decrease in pH during the initial period, and a fast increase in CO₃²⁻ concentration. Therefore, the main reaction in the initial stage of CO₂ absorption is (2.14), describing the formation of carbonate Na₂CO₃, from dissolved CO₂ and hydroxide, which exists as dissociated Na⁺ and CO₃²⁻ in the absorbent.:



The second reaction regime is characterized by continued CO₂ absorption by the solvent, which causes further OH⁻ depletion and accumulation of carbonate species through reaction (2.12) and forward reaction (2.13). This also makes the reverse reaction (2.13) more favourable, resulting in an increase in OH⁻ concentration and accelerating reaction (2.12) more and leading to a further rise in carbonate formation. Equation (2.15) describes the net reaction:



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As the reaction (3.15) approaches equilibrium, a small amount of CO_2 is still absorbed by the solvent because part of the CO_2 remains physically dissolved in water without immediately reacting. Eventually, NaOH becomes the limiting reactant, and the final reaction (2.16) which is the net reaction of equations (2.14) and (2.15), summarizes the overall CO_2 absorption process in aqueous NaOH :



3 Physical Model

The following sections present the main fluid-dynamics equations and the sub-models used to simplify the physics, as well as mass transfers mechanism governing the absorption process.

3.1 Reynolds-averaged Navier-Stokes (RANS) methods

RANS methods to model turbulence flows, use different strategies depending on the flow characteristics [47]:

1. Steady flow: The temporal fluctuations are averaged out and treated as part of the turbulence, leading to time-independent governing equations for the mean flow, commonly referred as steady RANS equations.
2. Unsteady flow: The equations are ensemble-averaged over a set of statistically identical realizations, so random fluctuations are implicitly included in the turbulence, while the mean field remains time-dependent. This formulation is known as unsteady or transient RANS (URANS/TRANS).

For a statistically steady flow, any instantaneous quantity can be decomposed into a time-averaged value $\bar{\phi}$ and a fluctuation from the mean value ϕ' , as shown in equation (3.1):

$$\phi(x_i, t) = \bar{\phi}(x_i) + \phi'(x_i, t) \quad (3.1)$$

The time average is defined in equations (3.2) and (3.3):

$$\bar{\phi}(x_i) = \lim_{T \rightarrow \infty} \frac{1}{T} \int_0^T \phi(x_i, t) dt \quad (3.2)$$

$$\overline{\phi'}(x_i) = \lim_{T \rightarrow \infty} \frac{1}{T} \int_0^T \phi'(x_i, t) dt = 0 \quad (3.3)$$

Where t is the time and T is the averaging interval, which must be large compared with the characteristic time scale of the turbulent fluctuations ϕ' . Under this condition, $\bar{\phi}$ does not depend on the choice of averaging starting time.

For a statistically unsteady flow, the mean of a quantity is defined by ensemble averaging procedure, illustrated in equation (3.4):

$$\bar{\phi}(x_i) = \lim_{N \rightarrow \infty} \frac{1}{N} \sum_{n=1}^N \phi(x_i, t) \quad (3.4)$$

Where N is the number of the ensembles, which must be sufficiently large to eliminate the effect of random turbulent fluctuations. Figure 3.1 schematically illustrates the decomposition into mean and fluctuating components and the distinction between time and ensemble averaging [47].

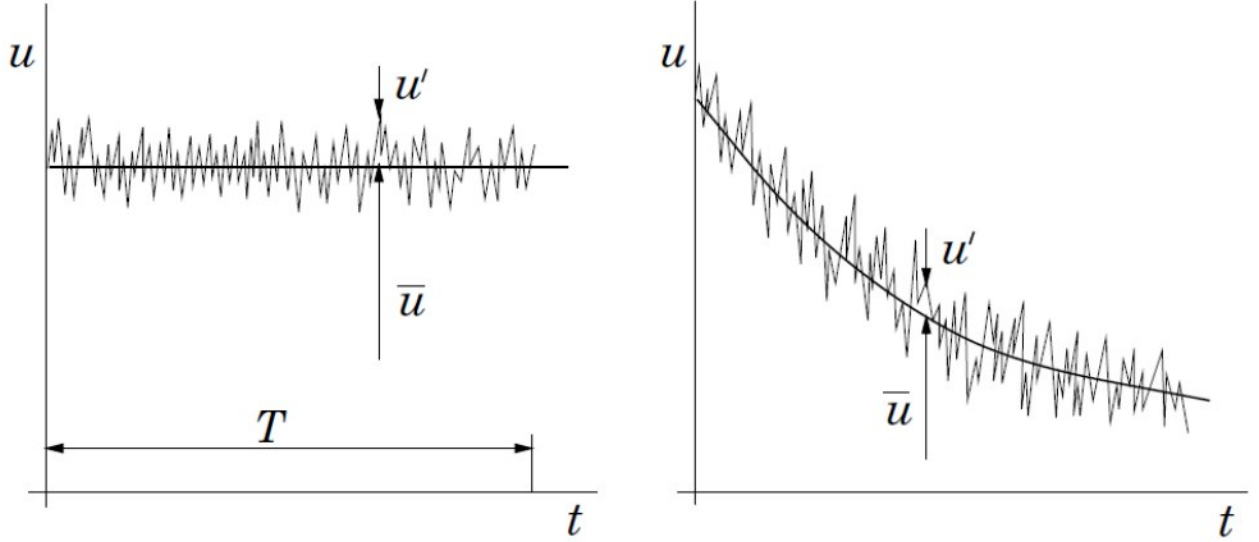


Figure 3.1 - Time averaging for steady flow and ensemble averaging for unsteady flow [45]

Equation (3.5) define the averaging procedure of a quadratic nonlinear term, which is decomposed into the product of the mean values plus a covariance term:

$$\overline{u_i \phi} = \overline{(\bar{u}_i + u'_i)(\bar{\phi} + \phi')} = \bar{u}_i \bar{\phi} + \overline{u'_i \phi'} \quad (3.5)$$

The last term is equal to zero if the fluctuating quantities are uncorrelated, which is rare in turbulent flows. Consequently, averaging the conservation equations introduces additional terms involving correlations of fluctuations that cannot be expressed uniquely in terms of the mean fields alone. As the resulting averaged governing equations are unclosed, with more unknowns than equations, they require turbulence closure models to provide suitable approximations [47].

In flows with significant density fluctuations, such as CO₂ absorption in spray towers where heat is released by chemical reactions, the classical Reynolds averaging introduces additional correlation terms between density and other quantities, which bring additional complexity to the problem solution. A density-weighted (mass-weighted) averaging procedure, known as Favre averaging, is adopted to overcome the problem. In this approach, any flow variable ϕ is decomposed as illustrated in equation (3.6):

$$\phi = \tilde{\phi} + \phi'' \quad (3.6)$$

Where the Favre (mass-weighted) mean is defined by equations (3.7) and (3.8):

$$\tilde{\phi} = \frac{\overline{\rho \phi}}{\bar{\rho}} \quad (3.7)$$

$$\tilde{\phi}'' = \frac{\overline{\rho(\phi - \tilde{\phi})}}{\bar{\rho}} = 0 \quad (3.8)$$

with $(\bar{\cdot})$ denoting the conventional Reynolds average, $(\tilde{\cdot})$ a Favre-averaged quantity and ρ the local density [50].

In this work, the governing equations for the gas phase are therefore written in Favre-averaged form to represent the turbulent behaviour of the variable-density flow.

3.2 Eulerian-Lagrangian framework

There are several approaches to model a multiphase system composed of air and liquid droplets. The main formulations used in spray applications are the Eulerian–Eulerian and Eulerian–Lagrangian models. Both are based on RANS conservation equations and require an appropriate turbulence-closure model. Interface-capturing methods, such as Volume of Fluid, level-set, and phase-field, or interface-tracking methods, such as front-tracking, are typically used when the interface between the phases must be explicitly resolved to capture its motion, deformation, and the evolution of interfacial properties [51].

In the Eulerian–Eulerian model, both phases are treated as interpenetrating continua within the control volume, and the conservation equations are solved in the Eulerian framework for each phase. The droplet properties are averaged over a representative volume to obtain mean liquid-phase values, making this model less computational expensive than the Eulerian-Lagrangian one. If the particle-size distribution must be resolved, a separate set of governing equations must be solved for each size class, which increases the computational cost. The Eulerian–Eulerian approach is therefore preferred for dense dispersed two-phase flows, such as fluidized beds and bubble columns [51].

In the Eulerian–Lagrangian approach the conservation equations for the continuous phase are solved in the Eulerian framework, while the dispersed phase is treated in a Lagrangian framework by tracking the motion of individual particles. The particle trajectories are obtained by solving the equations of motion for the droplets and accounting for the forces, heat transfer, and mass transfer acting on them. Gas phase properties required for the calculation, such as velocity, pressure, and temperature, must therefore be evaluated at the particle locations. Since these quantities are available only at the cell centres of the computational domain, they are interpolated to the particle positions.

In this work, the multiphase system is modelled using an Eulerian–Lagrangian approach because it makes possible to represent the discrete nature of the droplets, including their polydispersity, and to obtain detailed information on individual particles, such as their trajectories, residence time, mass and heat transfers. However, simulating a large number of particles is computationally expensive, and for fine or dense systems it is often impractical. For this reason, in this work the droplets are represented by parcels, computational entities representing a number of real particles with the same properties, such as velocity, size, and temperature, thereby reducing the computational cost.

A key feature of the Eulerian–Lagrangian approach is the inclusion of interphase mass, momentum, and energy exchange. In the present case, a two-way coupling model is used, so the gas influences the droplets through forces, heat transfer, and mass transfer, while the droplets also affect the continuous phase through corresponding source terms in the governing equations. Unlike a four-way coupling model, particle-particle interactions such as collisions, coalescence, and agglomeration are not accounted for [51],[52].

3.2.1 Gas phase equations

The conservation equations for mass, momentum, energy, and chemical species mass fractions of the flue-gas mixture are formulated in an Eulerian framework using the RANS approach. Transient simulations are used in this work, even though industrial spray towers operate under steady-state conditions, to analyse key unsteady process features such as

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droplet trajectories, residence times, gas–liquid interactions, turbulent dispersion, and dynamic mass transfer, which cannot be resolved in steady-state simulations. The gas phase is treated as a compressible, turbulent ideal mixture of CO_2 , H_2O , N_2 , and O_2 . Equations from (3.9) to (3.12) represents the governing equations [25], [46]:

- Mass Conservation:

$$\frac{\partial \bar{\rho}}{\partial t} + \frac{\partial}{\partial x_i} (\bar{\rho} \tilde{u}_i) = \overline{S_{\text{mass}}} \quad (3.9)$$

This equation enforces the continuity of mass in the gas phase, where S_m denotes the mass source term used for continuous–dispersed phase coupling and for representing any additional external mass sources or sinks.

- Species Conservation:

$$\frac{\partial}{\partial t} (\bar{\rho} \tilde{Y}_k) + \frac{\partial}{\partial x_i} (\bar{\rho} \tilde{Y}_k \tilde{u}_i) = \frac{\partial}{\partial x_i} \left(\bar{\rho} \bar{D}_k \frac{\partial \tilde{Y}_k}{\partial x_i} - \bar{\rho} \tilde{u}_i'' \tilde{Y}_k'' \right) + \overline{S_{k,\text{mass}}}, k = 1, N \quad (3.10)$$

This transport equation governs the Favre-averaged mass fraction of the k -th chemical species, accounting for both molecular diffusion and turbulence-induced mixing in the reactive flow. The terms $\bar{\rho} \bar{D}_k \frac{\partial \tilde{Y}_k}{\partial x_i}$ represents molecular diffusion, where $\bar{D}_k$ is the molecular diffusivity, the term $\bar{\rho} \tilde{u}_i'' \tilde{Y}_k''$ is the turbulent species fluxes, which need an appropriate closure, and $\overline{S_{k,\text{mass}}}$ denotes the net source term due to chemical reactions or interfacial mass transfer.

- Momentum Conservation:

$$\frac{\partial}{\partial t} (\bar{\rho} \tilde{u}_i) + \frac{\partial}{\partial x_i} (\bar{\rho} \tilde{u}_i \tilde{u}_j) = - \frac{\partial \bar{p}}{\partial x_j} + \frac{\partial \bar{\tau}_{ij}}{\partial x_i} - \frac{\partial}{\partial x_i} (\bar{\rho} \tilde{u}_i'' \tilde{u}_j'') + \bar{\rho} g + \overline{S_{\text{mom}}} \quad (3.11)$$

This momentum equation describes the fluid motion, with the left-hand side representing the unsteady and convective transport of mean momentum, and the right-hand side the pressure gradients $\left(\frac{\partial \bar{p}}{\partial x_j} \right)$ and molecular viscous stresses $\bar{\tau}_{ij}$. The additional term $\bar{\rho} \tilde{u}_i'' \tilde{u}_j''$ represents the turbulent Reynolds stresses, which are closed using the chosen turbulence model, while $\bar{\rho} g$ denotes the gravitational body force and $\overline{S_{\text{mom}}}$ is an external momentum source term accounting for interphase momentum exchange between the gas phase and the dispersed particles.

- Energy Conservation:

$$\begin{aligned} \frac{\partial}{\partial t} (\bar{\rho} \tilde{e}) + \frac{\partial}{\partial x_i} (\bar{\rho} \tilde{e} \tilde{u}_i) = & - \bar{p} \frac{\partial \tilde{u}_i}{\partial x_i} + \frac{\partial}{\partial x_i} \left(\lambda \frac{\partial \tilde{T}}{\partial x_i} - \bar{\rho} \tilde{u}_i'' \tilde{e}'' \right) \\ & + \bar{\tau}_{ij} \frac{\partial \tilde{u}_i}{\partial x_j} - \frac{\partial}{\partial x_i} \left(\bar{\rho} \sum_{k=1}^N D_k \frac{\partial \tilde{Y}_k}{\partial x_i} h_{s,k} \right) + \overline{S_{\text{en}}} \end{aligned} \quad (3.12)$$

This formulation characterizes the evolution of energy in the system, including the work done by pressure $\left(\bar{p} \frac{\partial \tilde{u}_i}{\partial x_i} \right)$ and heat transport by conduction $\left(\lambda \frac{\partial \tilde{T}}{\partial x_i} \right)$, where λ is the

gas thermal conductivity. It also accounts for turbulent energy fluxes $(\overline{\rho u_i'' e''})$, viscous dissipation $(\overline{\tau_{ij} \frac{\partial \tilde{u}_i}{\partial x_j}})$ and energy fluxes associated with species diffusion $(\overline{\rho \sum_{k=1}^N D_k \frac{\partial Y_k}{\partial x_i} h_{s,k}})$, through species diffusion coefficient D_k and sensible enthalpy $h_{s,k}$. $\overline{S_{en}}$ denotes the volumetric energy source term used to model interphase energy exchange between the two phases. The viscous stress tensor τ_{ij} for a Newtonian fluid is given by equation (3.13):

$$\tau_{ij} = \mu \left(\frac{\partial \tilde{u}_i}{\partial x_j} + \frac{\partial \tilde{u}_j}{\partial x_i} - \frac{2}{3} \delta_{ij} \frac{\partial \tilde{u}_k}{\partial x_k} \right) \quad (3.13)$$

With dynamic viscosity μ and δ_{ij} is the Kronecker delta.

3.2.2 Realizable k - ε turbulence model

As is discussed in the previous sections, the averaging procedure introduces additional turbulent correlation terms, so a closure model is required to relate these terms to the mean-flow variables. In this work, the turbulent species fluxes are modelled using the classical gradient-diffusion hypothesis, expressed in equation (3.14) [53]:

$$-\bar{\rho} \overline{u_i'' Y_k''} = -\frac{\mu_t}{Sc_t} \frac{\partial \tilde{Y}_k}{\partial x_i} \quad (3.14)$$

Here μ_t is the turbulent (eddy) viscosity provided by the selected turbulence model, and Sc_t is the turbulent Schmidt number for each species, defined with relation (3.15):

$$Sc_t = \frac{\nu_t}{D_t} \quad (3.15)$$

Where ν_t is the eddy viscosity and D_t is the mass diffusivity and quantifies the ratio between the rates of turbulent transport of momentum and the turbulent transport of mass.

Equation (3.16) illustrate the Reynolds stresses formulation in the momentum equation:

$$\bar{\rho} \overline{u_i'' u_j''} = -\mu_t \left(\frac{\partial \tilde{u}_i}{\partial x_j} + \frac{\partial \tilde{u}_j}{\partial x_i} - \frac{2}{3} \delta_{ij} \frac{\partial \tilde{u}_k}{\partial x_k} \right) + \frac{2}{3} \bar{\rho} k \quad (3.16)$$

The formulation is analogous to the viscous stress tensor for Newtonian fluids, with the presence of turbulent viscosity μ_t and the turbulent kinetic energy k , which is defined with equation (3.17):

$$k = \frac{1}{2} \overline{u_i' u_i'} = \frac{1}{2} (\overline{u_x' u_x'} + \overline{u_y' u_y'} + \overline{u_z' u_z'}) \quad (3.17)$$

Equation (3.18) gives the turbulent energy flux:

$$\overline{\rho u_i'' e''} = -\frac{\mu_t}{Pr_t} \frac{\partial \tilde{e}}{\partial x_i} \quad (3.18)$$

With turbulent Prandtl number Pr_t defined as the ratio between turbulent eddy viscosity and turbulent thermal diffusivity.

To obtain closure, a model for the eddy viscosity μ_t is required and in this work the realizable k - ε turbulence model is adopted, as is widely used in industrial applications and

recommended by previous studies [25], [54], [55]. The model is based on the Boussinesq hypothesis and the expression for turbulent viscosity is shown in equation (3.19):

$$\mu_t = \bar{\rho} C_\mu \frac{k^2}{\varepsilon} \quad (3.19)$$

Where k is the turbulent kinetic energy, ε is its dissipation rate, that is the rate at which turbulent kinetic energy is converted into thermal internal energy due to viscosity and C_μ is a model function, given by equation (3.20):

$$C_\mu = \frac{1}{A_0 + A_s U^{(*)} \frac{k}{\varepsilon}} \quad (3.20)$$

With A_s and $U^{(*)}$ coefficients function of the mean flow and turbulence properties and A_0 is a calibration coefficient. Differently from standard model, which uses a constant C_μ , realizable k - ε model uses a variable coefficient, so the turbulent viscosity responds to the local flow and turbulence conditions. This formulation improves predictive reliability, especially for spray-nozzle flows with strong strain, swirl, and jet-spreading effects.

The realizable k - ε model solves two additional transport equations, (3.21) for k and (3.22) for ε :

- Turbulent Kinetic Energy k :

$$\frac{\partial(\bar{\rho}k)}{\partial t} + \frac{\partial}{\partial x_j}(\bar{\rho}k\tilde{u}_j) = \frac{\partial}{\partial x_j} \left[\left(\mu + \frac{\mu_t}{\sigma_k} \right) \frac{\partial k}{\partial x_j} \right] + G_k + G_b - \bar{\rho}\varepsilon - Y_M \quad (3.21)$$

- Dissipation rate ε :

$$\frac{\partial(\bar{\rho}\varepsilon)}{\partial t} + \frac{\partial}{\partial x_j}(\bar{\rho}\varepsilon\tilde{u}_j) = \frac{\partial}{\partial x_j} \left[\left(\mu + \frac{\mu_t}{\sigma_\varepsilon} \right) \frac{\partial \varepsilon}{\partial x_j} \right] + \bar{\rho}C_1S_\varepsilon - \bar{\rho}C_2\frac{\varepsilon^2}{k + \sqrt{\nu\varepsilon}} + C_{1\varepsilon}\frac{\varepsilon}{k}C_{3\varepsilon}G_b \quad (3.22)$$

Where the model coefficients σ_k , σ_ε , C_1 , C_2 , $C_{1\varepsilon}$, $C_{3\varepsilon}$ and C_μ are assigned their standard values as defined for the realizable k - ε formulation. The left-hand-side terms represent the local rate of change and convective transport of the quantity, while the first term on the right-hand side accounts for diffusive transport. G_k defines generation of k by mean velocity gradients, G_b represents turbulence production due to buoyancy, Y_M is compressibility modification and S_ε is source term for ε [56]. In addition, a two-layer wall treatment is employed to handle turbulence in the near-wall region, where the realizable model is not applicable. This model is discussed in a later subchapter.

3.2.3 Liquid phase equations

The liquid dispersed phase, consisting of an ideal NaOH-H₂O mixture, is modelled in the Lagrangian framework using the Discrete Phase Model (DPM), by which individual droplets are tracked while resolving their physical and chemical properties. Due to high complexity in droplets behaviour during CO₂ absorption process, some assumption for the liquid phase are used, to simplify the analysis:

- 1) Droplets are treated as rigid spheres.
- 2) Agglomeration effects are neglected.
- 3) Droplet interactions, such as collision and subsequent coalescence are neglected.

The trajectory equation (3.23) relates the single droplet position $\vec{x}$ to its velocity $\vec{u}_d$ [57]:

$$\frac{d\vec{x}}{dt} = \vec{u}_d \quad (3.23)$$

By integrating the equation of motion for each particle, it is possible to reconstruct the particle position history and track the motion of the dispersed phase.

The governing equations for force, energy, and mass of an individual droplet i are expressed in equations from (3.24) to (3.26) [57],[58]:

- Equation of motion

$$m_d \frac{d\vec{u}_d}{dt} = \overline{S_{\text{mom}}} + m_d \vec{g} \left(1 - \frac{\bar{\rho}}{\rho_d} \right) \quad (3.24)$$

Where m_d is the mass of the liquid droplet and ρ_d is its density. The terms on the right-hand side represent the net force acting on the particle, including gravity reduced by the buoyancy contribution, which depends on the density ratio between the two phases, $m_d \vec{g} \left(1 - \frac{\bar{\rho}}{\rho_d} \right)$. $\overline{S_{\text{mom}}}$ is the momentum source terms, which accounts for all additional forces acting on the particle.

- Equation of energy

$$m_d c_d \frac{dT_d}{dt} = \overline{S_{\text{en}}} \quad (3.25)$$

Where c_d is heat capacity of the particle and T_d is its temperature. $\overline{S_{\text{en}}}$ is the energy source term, which contains the energy exchange between the two phases.

- Equation of mass

$$\frac{dm_d}{dt} = \overline{S_{k,\text{mass}}} \quad (3.26)$$

Where $\overline{S_{k,\text{mass}}}$ represents the mass source terms, accounting for interphase mass exchange.

The source terms $\overline{S_{\text{mom}}}$, $\overline{S_{\text{en}}}$ and $\overline{S_{k,\text{mass}}}$ describe the coupling between the two phases and are equal as those in the continuous gas-phase governing equations but with opposite signs, to ensure global conservation of mass, momentum, and energy between phases.

3.2.4 Momentum source term

The equation (3.27) formulates the momentum source term present in equations (3.11) and (3.24). It accounts for gas–liquid momentum exchange and represents the forces acting on the droplet in the spray tower. In this work, the dominant contribution is the drag force [57],[58]:

$$F_D = \frac{1}{2} \rho_g C_D |\vec{u} - \vec{u}_d| (\vec{u} - \vec{u}_d) A_D = \overline{S_{\text{mom}}} \quad (3.27)$$

Where C_D is the drag coefficient, ρ_g is the gas density, $\vec{u}$ and $\vec{u}_d$ the velocity of the gas and the particle, respectively, and A_D the droplet area. C_D depends primarily on the particle Reynolds number, defined with relation (3.28) [56]:

$$Re_p = \frac{\bar{\rho} d_d |\vec{u} - \vec{u}_d|}{\mu} \quad (3.28)$$

Where d_d is the droplet diameter and $|\vec{u} - \vec{u}_d|$ the slip velocity. In this work, the Morsi and Alexander correlation (3.29) is used to evaluate C_D [59]:

$$C_D = a_1 + \frac{a_2}{Re_d} + \frac{a_3}{Re_d^2} \quad (3.29)$$

Where a_1 , a_2 and a_3 are constants that depend on the Reynolds-number range.

Other forces acting on the droplet, such as the pressure-gradient force, virtual mass, and Basset history term, are neglected because they are typically of order of gas/liquid density ratio, on the order of 10^{-3} . Since the droplets are not in a high-shear region of the gas flow, lift and Magnus forces are also neglected. Therefore, the momentum source term is taken to be equal to the drag force [57].

3.2.5 Energy source term

The energy source term accounts for convective heat transfer between gas and droplets and latent heat from water evaporation, while radiative heat transfer is negligible in the process and not accounted. The formulation is illustrated in equation (3.30) [57],[58]:

$$\overline{S_{en}} = h(\bar{T} - T_d)\pi d_p^2 + \dot{m}_{H_2O} h_{evap} + h_{rxn} \quad (3.30)$$

Where h represents the convective heat transfer coefficient, $\bar{T}$ and T_d are the gas and droplet temperature respectively, $\dot{m}_{H_2O}$ is the interphase water mass transfer, h_{evap} is the water latent heat and h_{rxn} is the heat reaction rate. h is evaluated by the Ranz-Marshall correlation (3.31) [60]:

$$Nu = \frac{hD}{\lambda} = 2 + 0.6 Re_p^{0.5} Pr^{0.33} \quad (3.31)$$

Where Nu is the Nusselt number, λ is the conductive heat coefficient and Prandtl number Pr is ratio between momentum diffusivity to thermal diffusivity, as expressed in equation (3.32):

$$Pr = \frac{\nu}{\alpha} = \frac{c_p \mu}{\lambda} \quad (3.32)$$

3.2.6 Mass source term

The equation (3.33) gives the formulation for the mass source term, which describes the interphase mass transfer between the phases. In this case it includes CO_2 absorption by the droplet, represented by $\dot{m}_{CO_2}$ and water evaporation from the liquid droplets, represented by $\dot{m}_{H_2O}$ [58]:

$$\overline{S_{k, mass}} = \dot{m}_{H_2O} + \dot{m}_{CO_2} \quad (3.33)$$

3.3 Droplet evaporation

Evaporation involves the phase change of liquid components in the droplets into vapor at the interface, with the resulting mass transferred to the gas phase. In addition, it implicates heat transfer between the phases because of the phase change, and it leads to a reduction in droplet diameter due to mass loss [58]. In this work, a quasi-steady droplet evaporation model is adopted. Originally developed for single-component droplets and later extended

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to multicomponent systems, it assumes that the droplets remain internally homogeneous and consist of an ideal liquid mixture made up of different chemical species, some of which evaporate while others remain inert. The driven force for phase change is the departure from equilibrium point in the liquid-vapor system, which is represented by the difference in concentration of the vapor between the surrounding gas and the droplet surface [61], [62]. It is possible to visualize the equilibrium on an idealized phase diagram, as shown in the Figure 3.2, which is determined by two material properties, the saturation pressure, and the critical temperature.

The equation (3.34) express the mass rate of change of the evaporating component [61]:

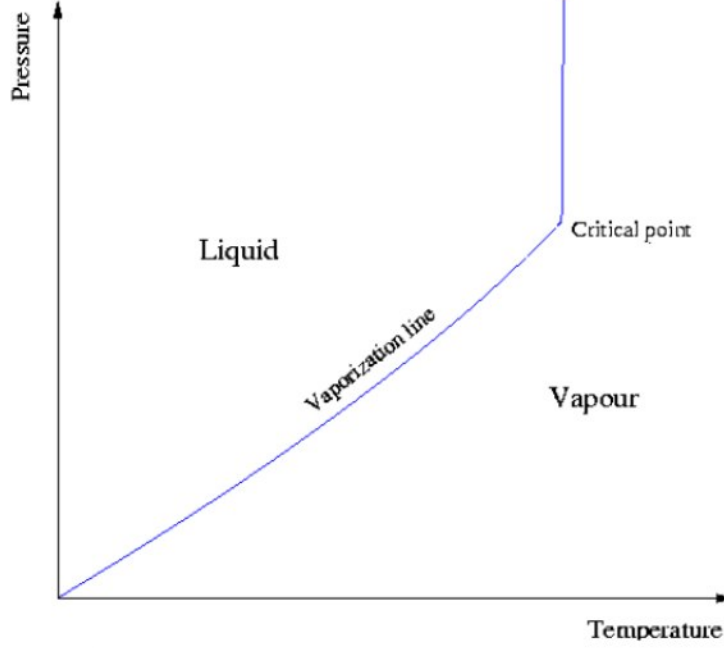


Figure 3.2 - Idealized liquid-vapour phase diagram [63]

$$\dot{m}_{d,i} = -\varepsilon_i g^* A_s \ln(1 + B) \quad (3.34)$$

Where the index i refers to each component of the mixture and ε_i represents the fractional mass transfer rate. In this case, only H_2O evaporates, while $NaOH$ is inert, so $\varepsilon_{NaOH} = 0$ and $\varepsilon_{H_2O} = 1$. The remaining parameters are the mass transfer conductance g^* , which depends on geometrical and mechanical effect of the liquid phase, the Spalding transfer number B , which represents the driving force for evaporation and depends on the thermodynamic properties of both phases, and the droplet surface A_s . With this model, vapour condensation is also possible and in that case the sign of the Spalding transfer number and therefore also the mass-transfer rate, is reversed. But condensation is not considered in this work because is not significant to the process.

The model distinguishes three different evaporation modes, and the values of g^* and B depend on the selected mode [63]:

- Super-Critical Evaporation
- Heat Transfer Limited Evaporation
- Vapor Diffusion Limited Evaporation

3.3.1 Super-Critical evaporation model

Super-Critical Evaporation occurs when the droplet temperature exceeds the critical temperature, as expressed in expression (3.35):

$$T_d \geq T_{c,H_2O} \quad (3.35)$$

Where T_d is the droplet temperature and T_{c,H_2O} is the water critical temperature. When the liquid phase falls within this regime, it results in instantaneous evaporation [61],[63].

3.3.2 Heat transfer limited evaporation model

The expression (3.36) shows the condition for the Heat transfer limited evaporation, that is when vapour at the droplet being subcritical and already saturated:

$$X_{H_2O,s} \geq 1 \quad (3.36)$$

Where $X_{H_2O,s}$ is the equilibrium mole fraction of water at droplet surface, evaluated through Raoult's law (3.37):

$$X_{H_2O,s} = \frac{p_{H_2O,sat}(T_d)}{p} X_{H_2O,p} \quad (3.37)$$

In which $X_{H_2O,p}$ is H_2O molar fraction inside the droplet and p is pressure and $p_{H_2O,sat}(T_p)$ is water saturation pressure at droplet surface temperature, calculated with the empirical Tetens equation (3.38) [64]:

$$p_{H_2O,sat}(T_d) = 0.61078 \exp \left(\frac{17.27(T - 273)}{(T - 273) + 237.3} \right) \quad (3.38)$$

Under this condition, the parameters B and g assume the forms expressed in equations (3.39) and (3.40):

$$B = \frac{c_p(T - T_d)}{h_{evap}} \quad (3.39)$$

$$g^* = \frac{k Nu_p}{c_p d_d} \quad (3.40)$$

Where Nu_p is the Nusselt particle number and k is thermal conductivity of the continuous phase.

In this regime, evaporation is limited by the amount of heat supplied to the droplet, because the water vapour concentration in the gas near the droplet surface is already equal to that at the interface. As a result, the driving force for evaporation depends only on the heat transferred to the droplet, which maintains the droplet surface at the saturation temperature. Consequently, the Spalding number depends on the ratio between the convective heat transfer and the latent heat of vaporization, while the conductance depends on Nu_p and k [61],[65].

3.3.3 Vapor diffusion limited evaporation model

The last evaporation mode occurs when neither of the first two regimes is active, that is, when the droplet surface is subcritical and the surrounding water vapour is not saturated. In this case, evaporation is controlled by diffusion and depends on the transport of vapour

from the droplet surface into the surrounding gas. The Spalding transfer number B is given by equation (3.41) [61],[65]:

$$B = \frac{Y_{H_2O,s} - Y_{H_2O}}{1 - Y_{H_2O,s}} \quad (3.41)$$

In which Y_{H_2O} is the water mass fraction in the continuous phase and $Y_{H_2O,s}$ is the equilibrium water mass fraction at the droplet surface, defined with equation (3.42):

$$Y_{H_2O,s} = X_{H_2O,s} \frac{W_{H_2O}}{W_s} \quad (3.42)$$

With W_{H_2O} and W_s are the molecular weights of the vapour and gas mixture at the droplet surface.

The formulation (3.43) express the conductance g for this regime:

$$g^* = \frac{\rho D_v S h_p}{d_d} \quad (3.43)$$

Where D_v is the molecular diffusivity of the water vapor in air and $S h_p$ is the particle Sherwood number, defined by relation (3.44), as the ratio between convective mass transfer and mass diffusion rate [65]:

$$S h_p = \frac{k_m d_d}{D} \quad (3.44)$$

With convective mass transfer coefficient k_m . The Sherwood number is obtained using the Ranz-Marshall correlation (3.45) [61]:

$$S h_p = 2 \left(1 + 0.3 Re_p^{1/2} Sc^{1/3} \right) \quad (3.45)$$

Where Sc is the Schmidt number of the continuous phase, defined by relation (3.46), as the ratio of momentum diffusivity to mass diffusivity [65]:

$$Sc = \frac{\nu}{D} \quad (3.46)$$

In this work, only the vapor–diffusion-limited evaporation mode is used in the simulations, due to the prevailing gas-phase temperature and H_2O concentration, for which the water vapour is not saturated.

3.4 Mass transfer

To model mass transfer in a system composed of liquid droplets suspended in a gas stream, literature reports that liquid-phase transport and reaction are the rate-limiting steps rather than gas-side transport [66–68]. The reaction-diffusion model assumes negligible internal circulation inside the droplet and treats the system as a pseudo-first-order, irreversible reaction at steady state. Under these assumptions, the CO_2 absorption rate per unit surface area is given by the expression (3.47) formulated by Danckwerts [69]:

$$J_{CO_2} = C K_H \sqrt{D_l k \{OH^-\}} \quad (3.47)$$

Where C is the CO_2 concentration in the gas phase, D_l is the diffusion coefficient of CO_2 in the liquid phase and $\{OH^-\}$ is the hydroxide-ion activity, which depends on the NaOH concentration in solution. K_H is Henry's constant, defined as the ratio between CO_2 concentration in the gas phase and its equilibrium concentration in water [70]. k is the

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reaction rate constant associated with the reaction (3.48), between dissolved CO_2 and hydroxide ions:



Because the hydroxide-ion concentration is high, typically greater than 1 M, the second-order reaction is commonly treated as pseudo-first order with respect to CO_2 [69]. The parameters K_H , $\{\text{OH}^-\}$ and D_l in equation (3.57) depends on the composition of the solution, especially ionic strength, viscosity, and temperature. The diffusion coefficient D_l can be estimated using by the Hayduk and Laudie method, expressed in equation (3.49) [71]:

$$D_l = \frac{13.26 \times 10^{-5}}{\mu_l^{1.4} V^{0.589}} = \frac{D_l^0}{\mu_l^{1.14}} \quad (3.49)$$

Where μ_l is the solution viscosity in centipoise, V is the molar volume and D_l^0 represents the infinite-dilution diffusion coefficient in water [72]. D_l decreases as NaOH concentration increases, mainly because the viscosity of the solution rises. To compute the reaction rate constant k , the model proposed by Pohorecky and Moniuk is adopted [73], because it is valid over a wide temperature range and NaOH concentration range. Equation (3.50) represents the kinetic expression:

$$\log k = \log k_{\text{OH},\infty} + 0.221I - 0.016I^2; \log k_{\text{OH},\infty} = 11.895 - \frac{2382}{T} \quad (3.50)$$

Where I is the ionic strength and $k_{\text{OH},\infty}$ is the reaction rate constant at infinite dilution, which follows an Arrhenius-type temperature dependence, formulated in the equations (3.51) and (3.52) [74],[75]:

$$\log k = \log k_{\text{OH},\infty} + bI \quad (3.51)$$

$$k_{\text{OH},\infty} = A_0 \exp\left(\frac{E_a}{RT}\right) \quad (3.52)$$

Where the constant b depends on the solution properties, A_0 is the pre-exponential factor, E_a the activation energy and R the universal gas constant.

Figure 3.3 shows the phenomena described by the previous models, which occur within the liquid droplets.

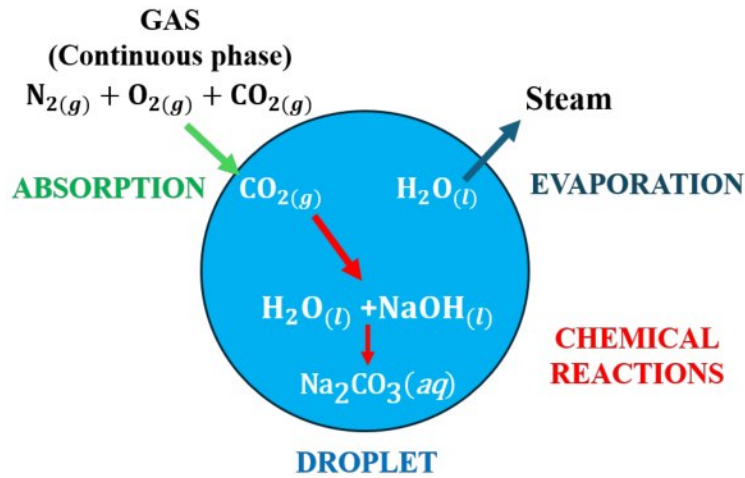


Figure 3.3 - Schematic of the phenomena occurring within the liquid droplets

3.5 Physic sub-models

Several additional sub-models are enabled in the present model to represent physical phenomena occurring during the spray absorption process that would otherwise be too computationally expensive to resolve directly. These sub-models are based on theoretical developments and have been validated for CFD applications, particularly in spray systems.

3.5.1 Two-Layer All y^+ Wall treatment

Flow and turbulence parameters in the near-wall region need to be modelled carefully, since the wall is a major source of vorticity. The near-wall region, usually referred to as the wall boundary layer, is a thin layer adjacent to the wall that is strongly affected by viscosity. Within this layer, the flow velocity increases from zero at the wall to the free-stream value. Its nominal thickness is defined as the normal distance from the wall to the point where the velocity reaches 99% of the free-stream value, beyond which viscous stresses are no longer significant. The turbulent boundary layer is divided into two main regions: an outer layer dominated by turbulent effects and an inner layer, which is further divided into three sublayers [76], as shown in Figure 3.3.

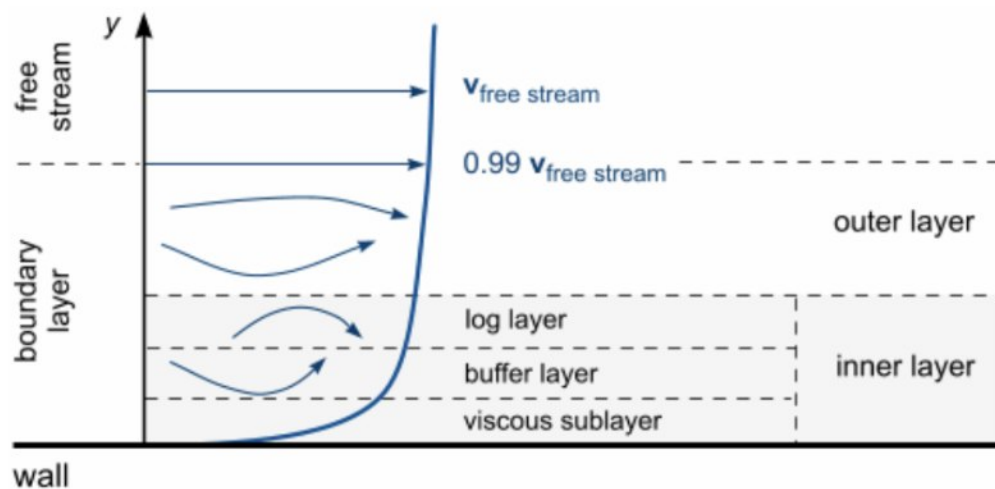


Figure 3.4 - Turbulent boundary layer diagram [63]

The viscous sublayer is in contact with the wall and is dominated by viscous effects. In this region, the mean flow velocity depends only on the fluid density, viscosity, distance from the wall, and wall shear stress. The turbulent log layer, by contrast, is influenced by both viscous and turbulent effects, while the buffer layer acts as a transitional region between the two. Each of them can be modelled with distinct empirical approaches [76].

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The non-dimensional wall distance y^+ is used to define the thickness of these sublayers and to describe the evolution of flow parameters. Figure 3.4, illustrates the evolution of the non-dimensional velocity u^+ , as function of y^+ over the sublayers.

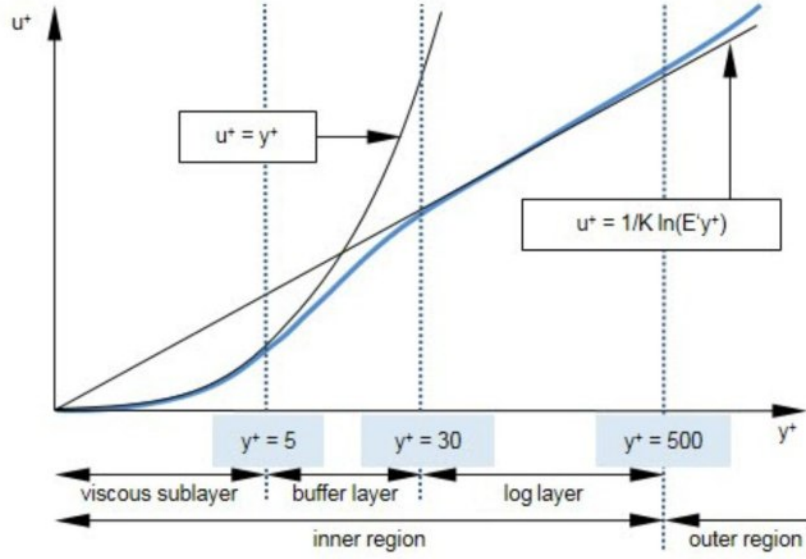


Figure 3.5 - non- dimensional velocity u^+ as function of non-dimensional wall distance y^+ , von Kármán constant K , and log-law constant E [63]

y^+ and u^+ are non-dimensional quantities, independent of the Reynolds number and are used to algebraically approximate the main variables in the inner layer. They may be implemented as standard or blended wall functions, depending on the chosen near-wall treatment, and the expressions (3.53) and (3.54) give their formulation:

$$y^+ = \frac{\gamma \rho u_*}{\mu} \quad (3.53)$$

$$u^+ = \frac{u}{u_*} \quad (3.54)$$

Where u_* is the representative flow velocity and depends on the wall-function formulation. For the Standard wall function equation (3.55) and (3.56) define u_* in the Viscous sublayer and in the Log layer respectively:

$$u_* = \sqrt{\frac{\mu |\mathbf{v}_{\text{tangential}}|}{\rho y}} \quad (3.55)$$

$$u_* = C_\mu^{1/4} k^{1/2} \quad (3.56)$$

Where $\mathbf{v}_{\text{tangential}}$ is the wall-tangential velocity vector, C_μ is the turbulence model coefficient and k is the turbulent kinetic energy.

For Blended wall function, the equation (3.57) gives the expression for u_* in the boundary layer:

$$u_* = \gamma \sqrt{\frac{\mu |\mathbf{v}_{\text{tangential}}|}{\rho y}} + (1 - \gamma) C_\mu^{1/4} k^{1/2} \quad (3.57)$$

Where γ is the blending function defined with relation (3.58):

$$\gamma = \exp\left(-\frac{Re_d}{11}\right) \quad (3.58)$$

With the wall-distance Reynolds number Re_d defined by the equation (3.59):

$$Re_d = \frac{\sqrt{k} d}{\nu} \quad (3.59)$$

Where d is the distance from the wall [76].

Turbulent boundary layers require specific models to capture the physics in the near-wall region. Near-wall turbulence modelling accounts for the local and non-local effects of walls on turbulence within the viscosity-affected region, while wall treatment provides boundary conditions to the flow and energy solvers. In this work, the Shear-Driven Two-Layer approach of Wolfstein is selected, which divides the computational domain into two layers. In the layer next to the wall, the turbulent dissipation rate ε and turbulent viscosity are defined as functions of the wall distance, while in the near-wall region these values are smoothly blended with the values farther from the wall. The turbulent kinetic energy equation is solved across the entire domain. Combined with all y^+ wall treatment, which relates the velocity to the wall shear stress in the first cell adjacent to, this approach enables accurate simulation of both low y^+ for fine mesh and high y^+ for coarse mesh using blended wall functions [77]. It is suitable for a wide range of mesh resolutions, including intermediate meshes in which the wall-cell centroids fall within the buffer-layer region, as shown in Figure 3.5.

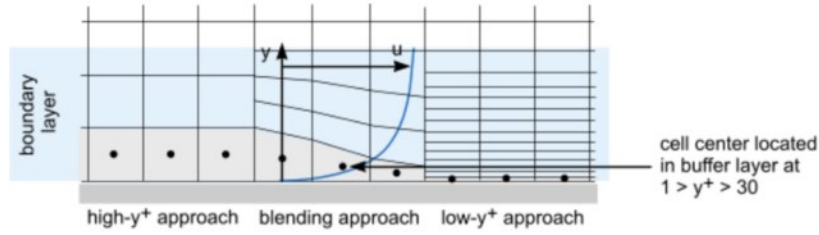


Figure 3.6 - All y^+ wall treatment diagram [63]

3.5.2 Rosin-Rammler distribution model

Liquid injection is modelled by specifying several injector parameters, such as injector type, liquid mass flow rate, velocity direction, and droplet properties including temperature, diameter distribution, and mixture composition. In this study, hollow-cone injectors are used, and the corresponding properties are taken from works in the literature. For the diameter distribution, a polydisperse Rosin–Rammler distribution is selected as is widely used to describe droplet size distributions in sprays processes. It was originally developed to model the volume distribution of particles as a function of diameter and was later extended to represent general cumulative mass, volume, or size distributions. The equation (3.60) define the distribution [78]:

$$F(D) = 1 - \exp\left[-\left(\frac{D}{D_{ref}}\right)^q\right] \quad (3.60)$$

Where D_{ref} is the reference diameter of the distribution and q is the distribution exponent. In addition to these parameters, minimum and maximum diameter limits are also used as model control parameters.

3.5.3 Huh Atomization model

The Huh atomization model is adopted in this work because it reliably describes atomization phenomena in high-pressure full-cone injectors. According to this model, the two main mechanisms governing spray atomization are gas inertia and the internal turbulent stresses generated inside the nozzle [79]. The atomization process is divided into two stages. First, turbulence generated in the nozzle hole perturbs the liquid jet surface as it exits the orifice. Then, after this initial growth stage, the perturbations grow exponentially under the action of pressure forces induced by the surrounding gas, until the jet breaks up into droplets. The atomization process is modelled based on wave-growth theory. The dimensionless number used to determine whether a droplet undergoes primary atomization is the Weber number, defined with relation (3.61):

$$We = \frac{\rho |V_s|^2 d_p}{\sigma} \quad (3.61)$$

Where ρ_g is gas density, V_s between the droplet and the surrounding gas and σ is the surface tension. If the droplet Weber number is lower than the specified critical value, primary atomization is not triggered, and the droplet bypasses this breakup stage [80].

The first stage is described by the average turbulent kinetic energy k_a and its dissipation rate ε_a at the nozzle-hole exit, which are derived from force and energy balances with the equations (3.62) and (3.63):

$$k_a = \frac{U^2}{8L/D} \left(\frac{1}{c_d^2} - K_c - 1 \right) \quad (3.62)$$

$$\varepsilon_a = \frac{K_\varepsilon U^3}{2L} \left(\frac{1}{c_d^2} - K_c - 1 \right) \quad (3.63)$$

Where U is the average injection velocity, L is the nozzle length, D is the nozzle diameter and K_c and K_ε are empirical coefficients, with default values of 0.45 and 0.5, respectively. c_d is the nozzle discharge coefficient, defined with the equation (3.64):

$$c_d = \frac{\dot{Q}}{A_0 \sqrt{\frac{2\Delta p}{\rho_l}}} \quad (3.64)$$

With the volumetric flow rate $\dot{Q}$, the nozzle cross-sectional area A_0 and the pressure drop across the nozzle Δp .

The second stage is described by the dispersion equation for the perturbation amplitude (3.65), which governs the detachment of droplets from the jet surface [81]:

$$(\omega + 2\nu\kappa^2)^2 + \frac{\sigma\kappa^3}{\rho_l} - 4\nu^2\kappa^3 \left(\kappa^2 + \frac{\omega}{\nu} \right)^{1/2} + (\omega + iU\kappa)^2 \frac{\rho}{\rho_l} = 0 \quad (3.65)$$

Where ω is the wave celerity, ν is the kinematic viscosity of the liquid, κ is the wave number, σ is the surface tension and i is the imaginary unit.

The droplet breakup-rate relation (3.66) describes the evolution of a droplet emerging from the nozzle with initial diameter D_d into secondary droplets:

$$\frac{dD_d}{dt} = -\frac{2L_A}{\tau_A K_A} \quad (3.66)$$

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Where L_A is atomization length scale, τ_A is atomization time scale and K_A is a model constant

4 Methods

The fluid-dynamics problem underlying the carbon capture process in the spray tower, as presented in the previous chapter, is governed by a coupled system of nonlinear partial differential equations that cannot be solved analytically. As discussed earlier, the present work uses CFD analysis, and this chapter presents the methods employed to solve the governing equations numerically and to simulate the process.

4.1 CFD Modelling of the Carbon Capture spray tower

4.1.1 Computational tools

Simcenter STAR-CCM+ version 2410 [63], developed by Siemens Digital Industries Software, was used for geometry creation, surface and volume extraction with the built-in CAD modeler, and for pre-processing, simulation, and post-processing. The numerical simulations were carried out on a workstation at the Process Technology Division of the Department of Chemical Engineering at KTH Royal Institute of Technology in Stockholm, Sweden. The results were analysed and visualized using MATLAB R2024b, by MathWorks [82].

4.1.2 Geometry design and operational conditions

The simulation system consists of a pilot-scale carbon capture spray tower with a height of 4 m and a diameter of 0.3 m. It represents the actual unit operating at the 64-MW(e) waste-to-energy facility in Linköping, Sweden, which is capable of processing 200 m³/h of dry flue gas. The spray tower is shown in Figure 4.1.



Figure 4.1 - Experimental unit deployed at Tekniska verken's waste-to-energy plant in Linköping, Sweden [25]

In the simulations, the gas flow rate is varied, and the gaseous phase is composed of 89 mol% dry air and 11 mol% CO₂, which is consistent with values reported for similar processes. The liquid flow rate is also varied, with the aqueous solvent consisting of 15 wt% NaOH. The pilot-scale system is equipped with six solid-cone nozzles positioned at a height of 3.6 m, with the option of operating a variable number of them. In the simulations, different configurations are tested using a variable number of cone injectors at different

positions. As STAR CCM doesn't enable cone nozzles for 2D domain, in this work they are built using a different numbers of point injectors, with varying injection velocity directions; this permits to replicate the cone nozzle behaviour. The entire process is carried out at atmospheric pressure. The operating conditions and geometric characteristics are listed in Table 4.1.

Parameter	Range / Value
Tower height [<i>m</i>]	4.0
Tower diameter [<i>m</i>]	0.3
Gas flow rate [<i>m</i> ³ / <i>h</i>]	75-200
CO₂ concentration at inlet	11 mol%
Liquid flow rate [<i>L/min</i>]	2-12
NaOH concentration in solvent	15 wt%
Number of nozzles	1-3
Operating pressure	Atmospheric

Table 4.1 - Operational parameters values/ranges

4.2 Computational implementation

The pre-processing step consists of creating the simulation domain, generating the mesh, assigning physical properties, models, and boundary conditions, and selecting the solvers used to compute the solution. These are critical steps, as they strongly affect the success and reliability of the simulation results. Two-dimensional (2D) simulations were performed in the present work due to the limited computational power of the workstation; consequently, all subsequent steps and results refer to the 2D case.

4.2.1 Space discretization

The initial 3D geometry of the spray tower was created as a CAD model and sectioned by a plane located along the central symmetry axis, resulting in half of the original volume. The planar surface produced by this cut was then extracted and used as the 2D domain for mesh generation, which subsequently served as the computational domain for the simulations. The domain is discretized into control faces over which the governing conservation equations are integrated. This leads to a Cartesian structured grid with several prism layers in the boundary regions, in order to capture the steep variations of the flow properties in these zones. A mesh-independence study was performed using three different meshes with different cell sizes, and the results are discussed in Chapter 5. to analyse key unsteady droplet trajectories, residence times, gas-liquid interactions, turbulent dispersion, and dynamic mass transfer, which cannot be resolved in steady-state simulations.

4.2.2 Time discretization

Since a transient analysis is performed to capture the unsteady characteristics of the process, such as droplet trajectories and dynamic mass transfer, time must be discretized

in addition to the spatial coordinates. The choice of the time step is a key factor in ensuring the reliability of the simulation. A guiding parameter for this choice is the Courant–Friedrichs–Lewy (CFL) number, defined in the expression (4.1) [47]:

$$CFL = \frac{u\Delta t}{\Delta x} \quad (4.1)$$

where Δt is the time step, u is a characteristic velocity, and Δx is a characteristic length scale of the mesh. The CFL number depends on both the mesh and the flow conditions; therefore, the selection of Δt is intrinsically linked to the mesh size and constrained by the available computational resources. In this work, the CFL number is kept below 0.15 to achieve high temporal accuracy and ensure stable convergence of the solution.

4.2.3 Finite Volume Method

The Finite Volume Method is a numerical method which transforms the governing partial differential equations into a system of algebraic equations through discretization in space and time. Although the problem domain in this work is two-dimensional, STAR-CCM+ applies the Finite Volume Method by treating the discretized domain as a one-cell-thick volumetric mesh. The equations are applied to each mesh cell, ensuring conservation of the relevant quantities within each control volume and over the entire domain. A multigrid solver is then employed to obtain the solution by solving the resulting linear system. The formulation of the method applied to the Eulerian phase is given by the equations from (4.2) to (4.4) [45]:

The conservation equation for a generic scalar quantity ϕ can be written in integral form as:

$$\begin{aligned} \int_V \int_t^{t+\Delta t} \frac{\partial(\rho\phi)}{\partial t} dt dV + \int_V \int_t^{t+\Delta t} \nabla \cdot (\rho\phi\mathbf{u}) dt dV \\ = \int_V \int_t^{t+\Delta t} \nabla \cdot (\Gamma_\phi \nabla \phi) dt dV + \int_V \int_t^{t+\Delta t} S_\phi dt dV \end{aligned} \quad (4.2)$$

By considering the boundary of the differential control volume dV and applying the Gauss divergence theorem to the convective and diffusive flux terms, the following expression is obtained:

$$\begin{aligned} \int_t^{t+\Delta t} \int_V \frac{\partial(\rho\phi)}{\partial t} dV dt + \int_t^{t+\Delta t} \int_{\partial V} (\rho\phi\mathbf{u}) \cdot \mathbf{n} dA dt \\ = \int_t^{t+\Delta t} \int_{\partial V} (\Gamma_\phi \nabla \phi) \cdot \mathbf{n} dA dt + \int_t^{t+\Delta t} \int_V S_\phi dV dt \end{aligned} \quad (4.3)$$

The values of the flow properties, such as ϕ and ρ are evaluated at the cell centres, whereas the gradients at the cell faces are computed using a least-squares approximation. Convective fluxes are discretized with a second-order upwind scheme, which reduces numerical diffusion and improves accuracy. The source term depends on the transported variable and the transient term is discretized using a first-order implicit Euler scheme. The overall discretization procedure leads to a system of algebraic equations that can be written in the general form:

$$a_p \phi_p = \sum a_{nb} \phi_{nb} + S_u \quad (4.4)$$

where ϕ_p and ϕ_N denote the values of the variable at the cell center and at the neighbouring cells, respectively, a_p and a_N are the corresponding coefficients, and S_u

represents the source terms contribution. For cells adjacent to the domain boundaries the general discretised equation is modified to include boundary conditions [45].

4.2.4 Boundary conditions

with the y-axis representing the vertical coordinate. The base of the spray tower is located at $y = 0$ m, where the gas enters the column through a mass flow inlet boundary. At the top of the tower, the gas leaves the domain through a pressure outlet boundary. The liquid injectors are positioned at a height of $y = 3.6$ m and are modelled as liquid inlets. All solid walls are treated as no-slip walls with escape boundary conditions for the dispersed phase. Figure 4.2 shows the discretized domain and the applied boundary conditions for the case with three cone injectors. The zoomed view at the top highlights the mesh refinement and the prism boundary layers near the wall in the high-viscosity regions.

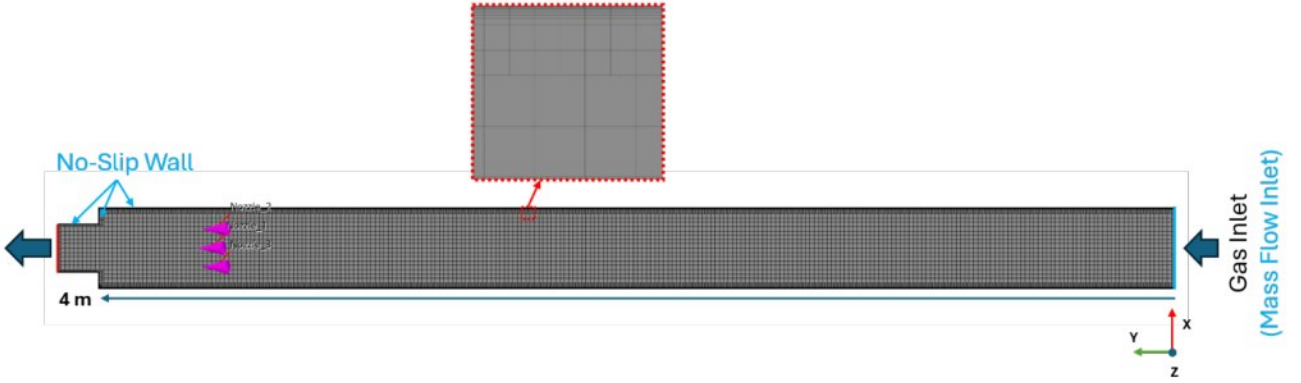


Figure 4.2 – Discretized domain with boundary conditions

4.2.5 Solution procedure

The problem under consideration is non-linear and therefore must be solved using an iterative method. The algebraic system for a generic variable ϕ is expressed in delta form in the equation (4.5) [45]:

$$\frac{a_p}{\omega} \Delta\phi_p + \sum_{nb} a_{nb} \Delta\phi_{nb} + S_u = r \quad (4.5)$$

Where $\Delta\phi_p = \phi_p^{k+1} - \phi_p^k$ with the variable at current iteration $k + 1$ and at previous iteration k . The summation is over all neighbouring cells, and ω is an under-relaxation factor, which controls the weighting of the update between successive iterations. The term r is defined by expression (4.6) as:

$$r = \left| a_p \phi_p - \left(\sum_{nb} a_{nb} \phi_{nb} + S_u \right) \right| \quad (4.6)$$

This term represents the residual, defined as the difference between the left- and right-hand sides of the discretized equation (4.4). The iterative procedure would ideally continue until the residual for all transported quantities decreases to zero, indicating full convergence; in practice, a sufficiently small value, typically below 10^{-4} , is accepted as the convergence criterion [45]. Two levels of iteration are employed during the simulation: an outer iteration loop that updates the nonlinear solution, and an inner loop that governs the iterative solution of the linear system. The outer iterations are repeated several times, while the inner linear systems are solved approximately using an Algebraic Multigrid (AMG) method, which accelerates the convergence of the linear solver [63].

4.2.6 Pressure-velocity coupling

To correctly resolve the momentum equations, an accurate solution of the pressure field is required. Since there is no explicit governing equation for pressure, the pressure and velocity fields are coupled and must be obtained iteratively, allowing the pressure field to be computed in a way that yields a velocity field satisfying the momentum equations [45]. The pressure-velocity coupling works in conjunction with the segregated flow solver, which solves the nonlinear governing equations sequentially. The algorithm first predicts the pressure and velocity components from the momentum equations, then constructs a pressure-correction equation from the continuity equation; this correction is applied to enforce the mass conservation constraint [63].

In this work, the PISO (Pressure-Implicit with Splitting of Operators) algorithm is employed for pressure-velocity coupling, owing to its superior performance in unsteady simulations compared with other methods and its proven reliability for multiphase flows [46],[76]. The algorithm consists of an initial predictor step followed by a user-defined number of corrector steps. For two corrector steps, the expression (4.7) defines the quantities $\mathbf{u}$ and p :

$$\phi^{***} = \phi^{**} + \phi'' = \phi^* + \phi' + \phi'' \quad (4.7)$$

where ϕ denotes a generic variable, ϕ' and ϕ'' the first and second corrected values, and ϕ^* and ϕ^{**} represent the first and second predicted values obtained from the predictor step and the first corrector step, respectively. The overall sequence of operations in the PISO algorithm can be summarised as follows [46,76]:

- 1) A trial value for the pressure field p^* is assumed;
- 2) The momentum equations are solved to obtain an intermediate velocity field u^* ;
- 3) The corresponding intermediate mass fluxes at the cell faces $\dot{m}_f^*$ are computed;
- 4) Solve the pressure correction equation and compute the values for pressure correction p' ;
- 5) The pressure field is updated accordingly to expression (4.8):

$$p^{n+1} = p^n + \omega p' \quad (4.8)$$

Where ω is the pressure under-relaxation factor, which is set to one in the last corrector step;

- 6) The pressure correction is applied at the boundaries to update the boundary pressures p'_b ;
- 7) The face mass fluxes are updated using the corrected pressure field, presented by the equation (4.9):

$$m_f^{n+1} = \dot{m}_f^* + \dot{m}_f' \quad (4.9)$$

- 8) The cell-centre velocities are corrected by the equation (4.10):

$$v_p^{n+1} = v_p^* - \frac{V\omega\nabla p'}{a_p^v} \quad (4.10)$$

Where ω is the under-relaxation factor for velocity, $\nabla p'$ is the gradient of the pressure correction, V is the cell volume, and a_p^v is the vector of coefficients of the discretised momentum equation;

- 9) The density is corrected to account for the updated pressure field;
- 10) The PISO-coupled transport equations for species and energy are solved using the updated flow field;
- 11) The density is updated from the equation of state (except in the last corrector step);
- 12) The predictor–corrector sequence is repeated until the prescribed number of PISO corrector steps is completed.

4.3 Numerical approach

The entire CO₂ absorption process is simulated using an interphase-coupling approach that simultaneously resolves momentum, heat, and mass transfer between the phases and accounts for evaporation and chemical reactions inside the liquid droplets. The simulation procedure consists of the following steps [25]:

- 1) The continuous phase is modelled in a Eulerian framework by solving the governing equations (3. 9)–(3.22). The gas flow is computed first, without liquid injection, until a steady state is reached to achieve convergence of the turbulent fluctuations.
- 2) Once convergence of the gas-phase flow is obtained, the Lagrangian phase is injected into the tower through the nozzles, and the liquid droplets are tracked as discrete parcels. Equations (3.23)–(3.26) are solved to compute the properties of the dispersed phase.
- 3) In parallel, the interphase source terms are evaluated using Equations (3.27)–(3.33), while the chemical reaction model, governed by Equations (2.11)–(2.16) is solved to update the concentration of the dissolved chemical species within the liquid phase.
- 4) At the same time, the vapor–diffusion-limited evaporation model described by Equations (3.41)–(3.46) is implemented as a pseudo reaction that transfers H₂O from the liquid to the gas phase, with the mass transfer rate given by Equation (3.41), since STAR-CCM+ does not allow the chemical reaction and evaporation models to be used simultaneously within the Lagrangian phase.
- 5) Two-way coupling terms are then applied to account for the influence of the droplets on the gas phase in an iterative manner. The properties of the liquid and gas phases are solved alternately, with continuous updating of the interphase source terms and governing parameters.

The described procedure is repeated iteratively until specified convergence criteria are satisfied, based on two main performance parameters: the absorption efficiency η , formulated in equation (4.11) and the tower pressure drop ΔP .

$$\eta = \left(\frac{y_{CO_2,in} - y_{CO_2,out}}{y_{CO_2,in}(1 - y_{CO_2,out})} \right) \times 100\% \quad (4.11)$$

where y_{in} and y_{out} are the inlet and outlet mole fractions of CO₂ in the gas stream, respectively. The simulation is considered fully converged when the variation of η between successive iterations remains below 0.1%, and the fluctuations in ΔP do not exceed 20 Pa.

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5.1 Mesh independency analysis

A mesh-independence analysis was performed to evaluate the sensitivity of the model to the computational cell size. The simulations were carried out using three different base cell sizes of 40 mm, 20 mm, and 10 mm, denoted as L1, L2, and L3, respectively. Table 5.1 summarizes the corresponding mesh configurations.

Mesh Configuration	Base Cell size [mm]
L1	40
L2	20
L3	10

Table 5.1 - Base cell size for each adopted mesh configuration

To assess the influence of mesh refinement on the process behaviour, two parameters are evaluated along the vertical centreline on the y-axis of the spray tower for the three mesh configurations. Figures 5.1 and 5.2 show the corresponding profiles of mole fraction and gas temperature in the spray tower.

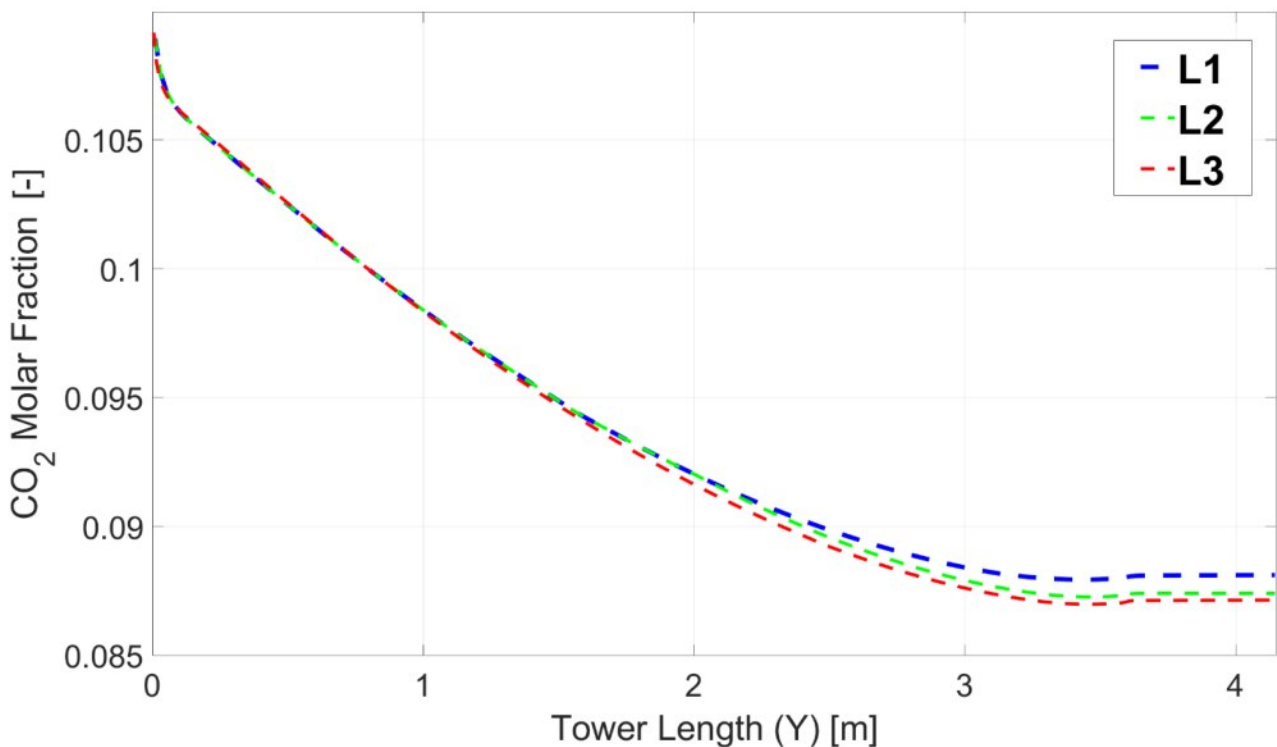


Figure 5.1 - CO₂ mole fraction profiles along the tower centreline for three different cell sizes

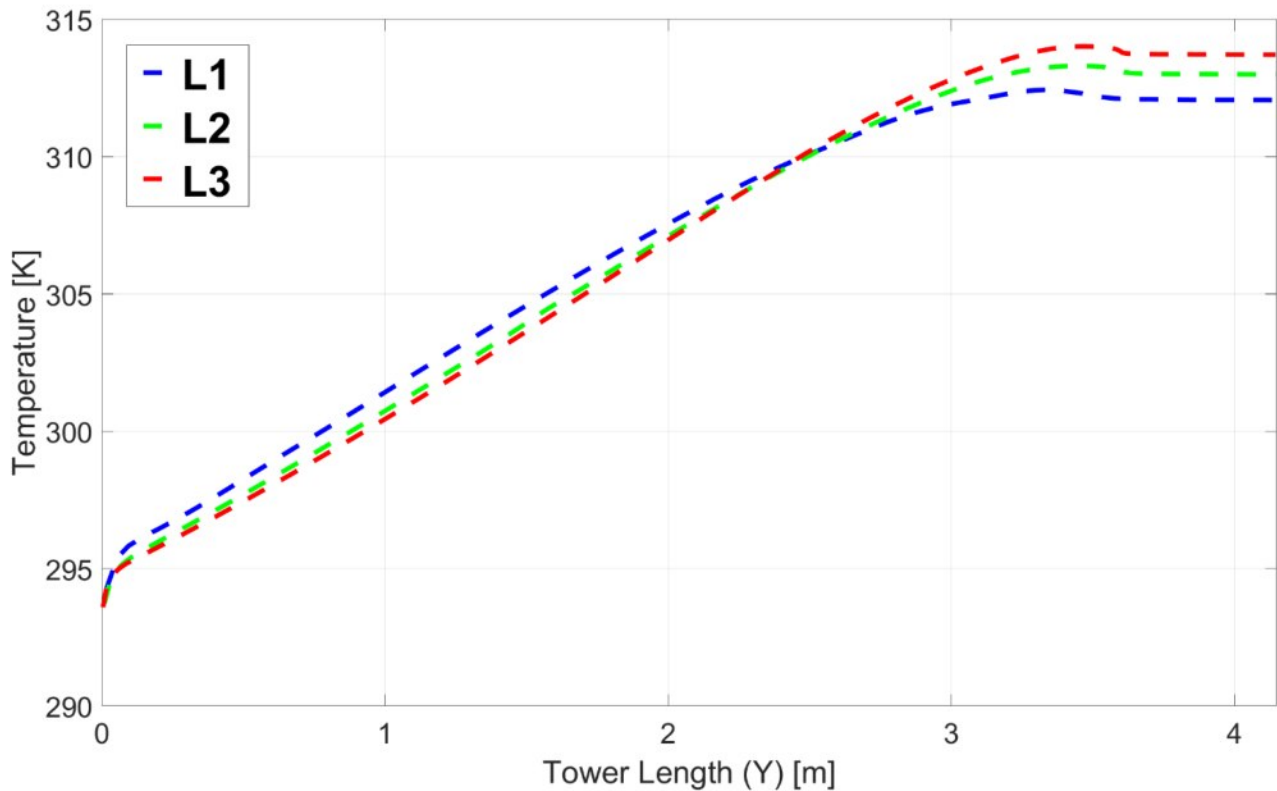


Figure 5.2 - Temperature profiles along the tower centreline for three different cell sizes

Starting from the CO_2 mole fraction profiles, all three mesh configurations exhibit the same trend, with the mole fraction decreasing towards the top of the tower and stabilising in the final section. This behaviour is consistent with the decrease of gas-phase CO_2 concentration from its inlet value due to absorption by the liquid solvent, which is particularly enhanced in the droplet injection zone. The relative small differences between the curves indicate a high degree of mesh convergence, especially between the L2 and L3 cell sizes.

The temperature profiles exhibit the opposite behaviour, increasing from the initial flue gas temperature at the inlet to a nearly constant value in the final section of the column. CO_2 This evolution is explained by absorption of CO_2 into the liquid droplets, which initiates exothermic reactions with NaOH in the liquid phase and leads to an increase in the temperature of both the liquid and the surrounding gas. In this case as well, the small variation of $\pm 2K$ is considered acceptable and indicates good mesh independence.

These results indicate that the model exhibits relatively low sensitivity to the mesh size, which is important because it demonstrates that the simulation results are determined by the physical models and boundary conditions rather than by the chosen mesh size. While the coarser mesh L1 may not capture the phenomena with sufficient accuracy, the refined meshes L2 and L3 show only minor differences. Moving from L2 to L3 halves the base cell size, which results in approximately four times more cells in the two-dimensional simulation domain. This involves a significant increase in the computational time required for the simulations, due to the limited processing power of the workstation and the impossibility of running them on high-performance computing systems. Consequently, the L2 mesh is used for the results presented in the following sections, as it offers a good compromise between computational cost and accuracy, with performance comparable to the more refined mesh.

5.2 Comparison with experimental data

Figure 5.3 presents a comparison between simulation results and experimental data for CO₂ absorption efficiency at the outlet, for gas flow rates Q_g of 200, 150, and 100 m³/h and two liquid flow rates Q_l of 8 and 12 L/min.

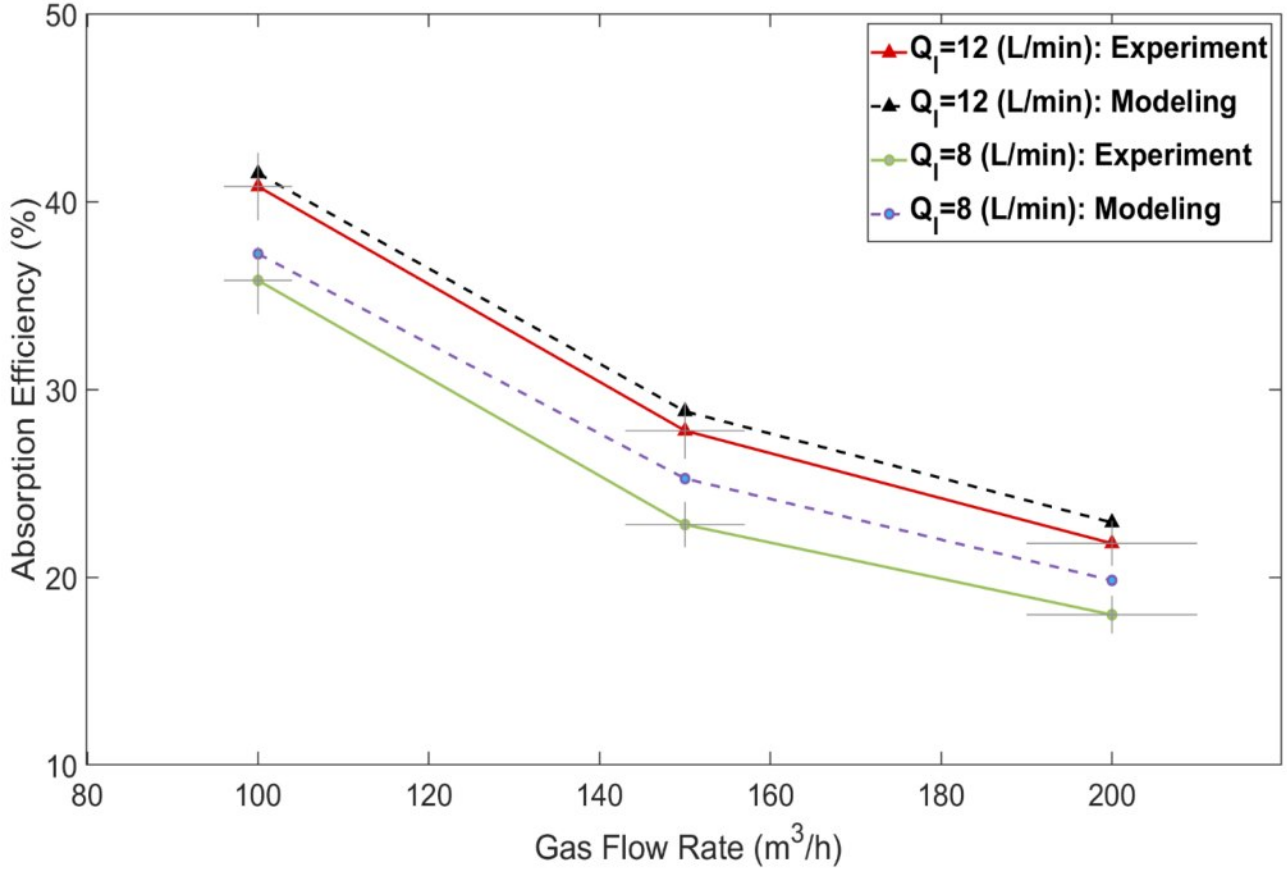


Figure 5.3 – Comparison between experimental data and simulation results for CO₂ absorption efficiency for gas flow rates of 100–200 m³/h and liquid flow rates of 8 and 12 L/min

The results show a good agreement between the experimental data and the simulation, especially for the higher liquid flow rate of 12 L/min. For both experiments and simulations, the absorption efficiency decreases as the gas flow rate increases. Moreover, for all tested gas flow rates, the absorption efficiency is higher at the larger liquid flow rate. The physical explanation of these trends is discussed in detail in Subchapter 5.3.3.

The experimental values are taken from the work of Ali et al., who performed the same analysis for a 3D model without evaporation, shown in Figure 5.4, and correspond to the outlet values measured on the pilot-scale tower developed by Grimaldi Development AB in collaboration with KTH Royal Institute of Technology at the Tekniska verken waste-to-energy plant in Linköping, Sweden. Comparing Figures 5.3 and 5.4 shows that the 3D model reproduces the experimental data very closely, particularly for $Q_l = 12$ L/min. A major difference, however, lies in the computational time required to reach convergence, which is higher for the 2D model used in this work. This is mainly because the 3D model can better resolve the three-dimensional evolution of gas and liquid flows, droplet dispersion, and radial mixing, and because it includes six nozzles and a lateral gas inlet, providing a more accurate representation of the real CO₂ capture system. Nevertheless, the 2D model still reproduces

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the system with good agreement, as the converged flow in the tower is sufficiently axisymmetric. Since the main objective of this thesis is to develop and assess the evaporation model rather than to validate the model as an exact representation of the pilot unit, the agreement obtained is sufficient for the purposes of this study.

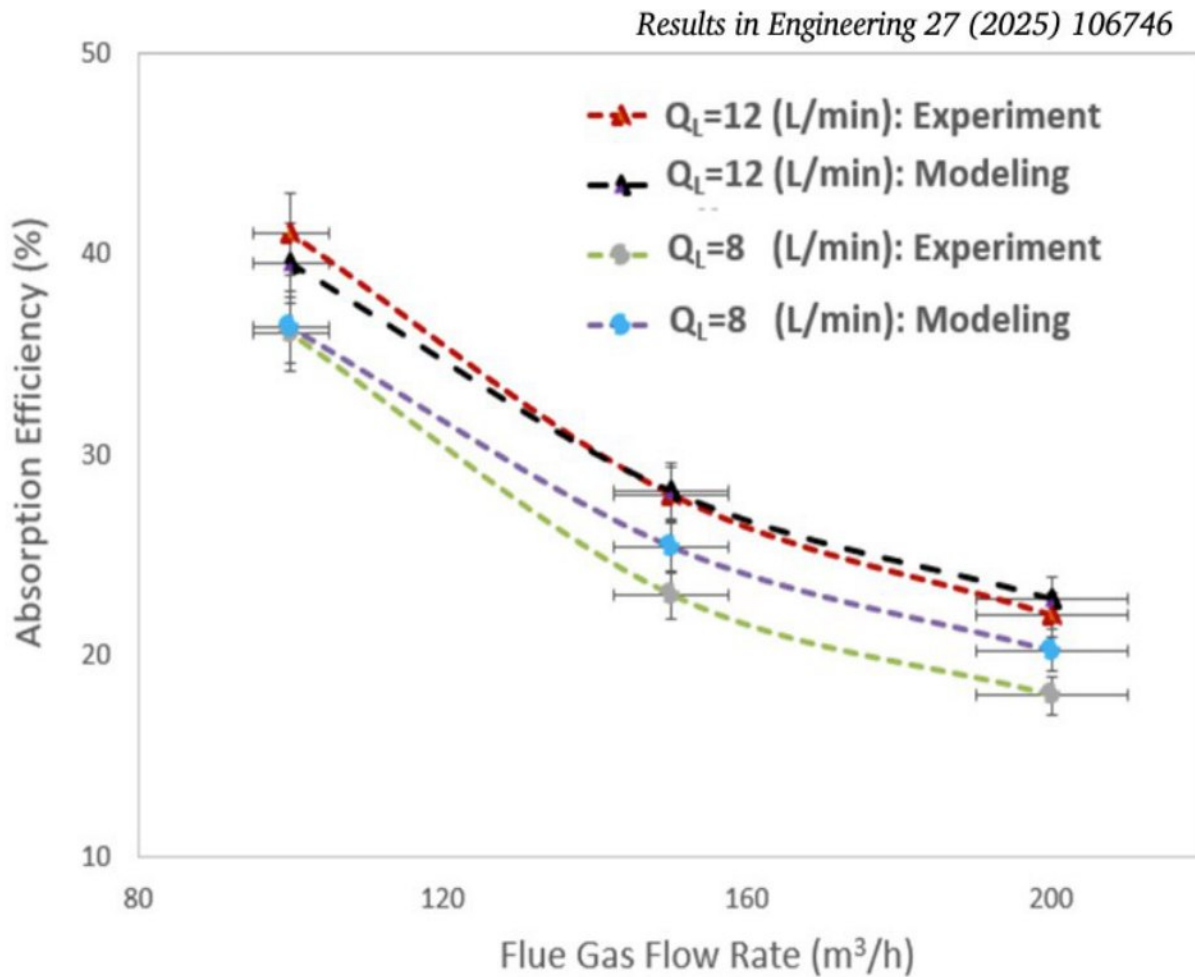


Figure 5.4 - Comparison between experimental data and simulation results of the 3D model of the Ali et al. work for CO₂ absorption efficiency with varying gas and liquid flow rates [25]

5.3 Carbon Capture performance analysis without evaporation model

The carbon capture process is first analysed without the evaporation model, in order to settle a baseline behaviour and enable subsequent comparison with the case where evaporation is included. The performance is then evaluated by varying the number and configuration of nozzles (from one to three), the liquid-to-gas ratio (L/G), and the droplet diameter distribution.

5.3.1 Effect of nozzle configuration on CO₂ capture performance

Two different nozzle configurations were tested in this work: a base case with a single cone nozzle and an improved configuration with three cone nozzles. In both cases, the nozzles have the same characteristics, with an injection angle of 30°, but in the second configuration the lateral injectors are oriented towards the centre of the tower at the same angle, in order to limit the number of droplets directed towards the walls. The simulation were performed with a gas flow rate Q_g of 75 m³/h, a liquid flow rate Q_l of 4 L/min, and a droplet diameter range of 1500-2000 μ m. Figure 5.5 shows the gas temperature field inside the tower and the droplet temperature for the two configurations.

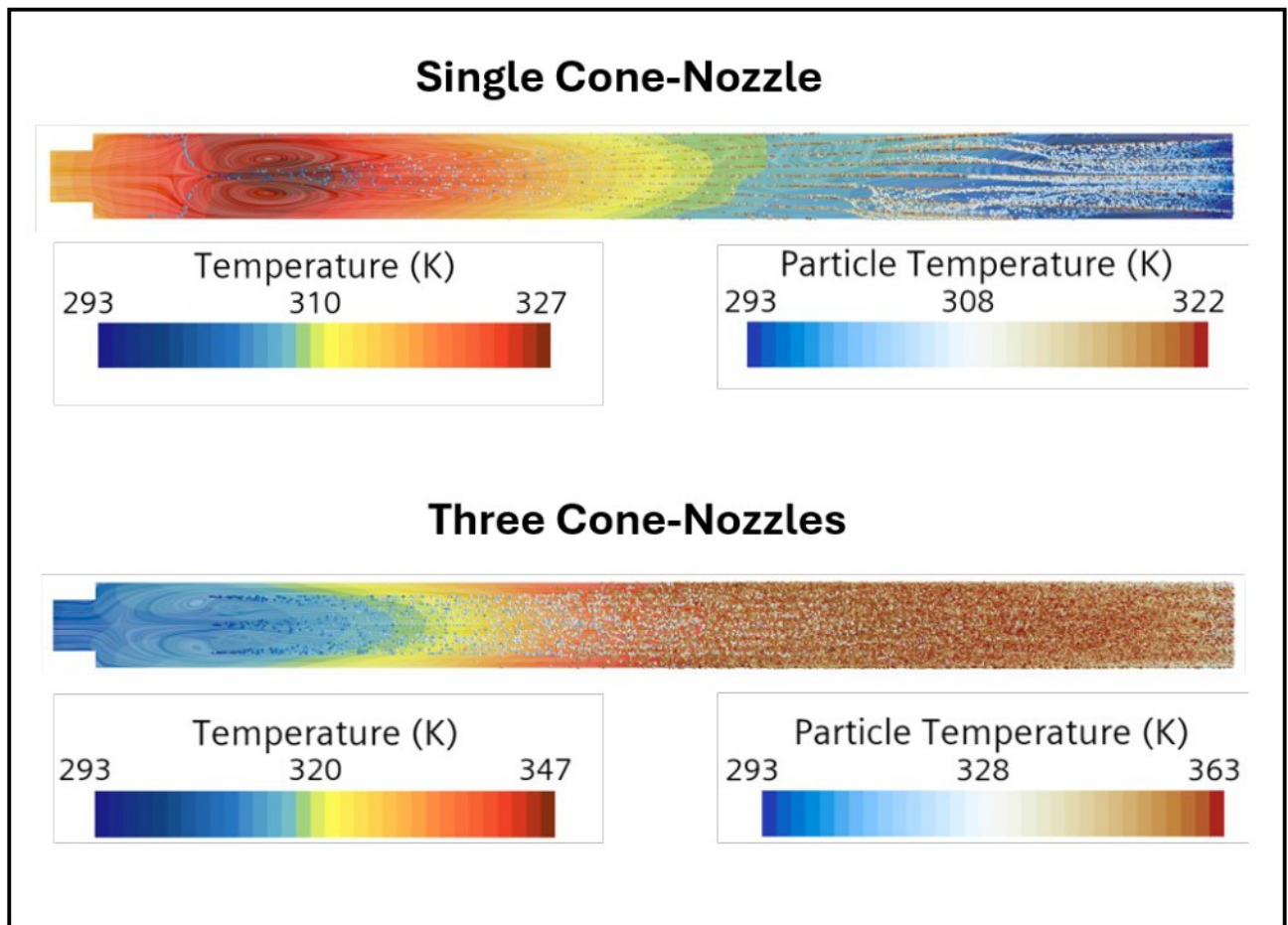


Figure 5.5 - Gas and liquid phase temperatures for single- and three-nozzle configurations

The two configurations show clear differences in the temperature field and in the droplet distribution. In both cases, the droplet temperature increases from the top to the bottom of the tower because absorption and the subsequent reaction increase the droplet temperature. The main difference lies in the spatial distribution of the droplets: in the three-nozzle

configuration the liquid phase better covers the lower part of the tower, which increases the gas–liquid interfacial area and enhances CO_2 absorption. In the single-nozzle configuration, the droplet dispersion is less uniform and many droplets impact the wall, which reduces the effective absorption capacity due to the escape boundary condition. These differences result in an overall lower droplet temperature for the single-nozzle configuration. The gas-phase temperature also differs between the two cases: in the three-nozzle configuration, higher temperatures and a more uniform temperature field are observed, with the maximum located in the lower part of the tower due to the higher droplet concentration and intensified reactions. Moving towards the top, the temperature decreases because the CO_2 concentration in the gas phase becomes lower, which weakens the absorption mechanism. In contrast, in the single-nozzle case the maximum gas temperature occurs in the injection zone, since the reduced droplet concentration in the lower part of the tower leads to less absorption there and the gas reaching the upper section has a higher CO_2 content.

The effects discussed above are summarised in Figure 5.6, which shows the outlet CO_2 molar fraction for the two configurations as a function of time, which denotes the computational time steps used in the transient numerical simulation and does not correspond to real physical time.

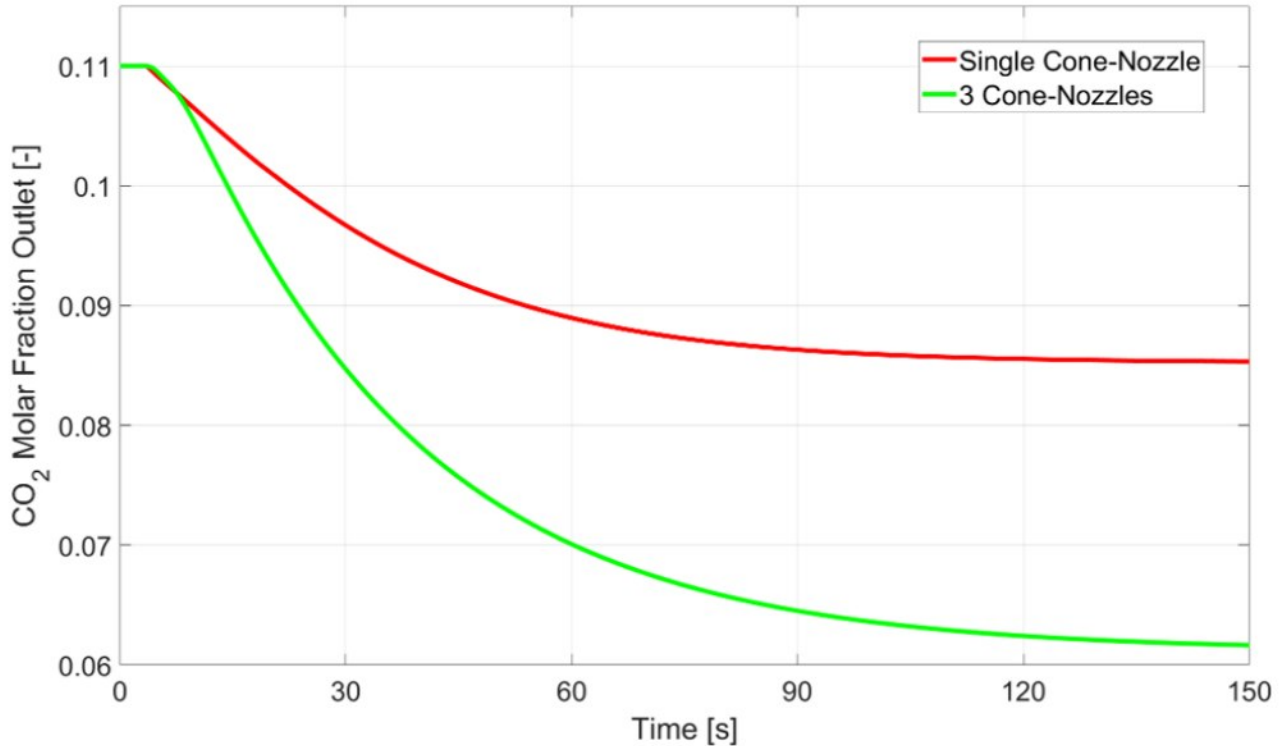


Figure 5.6 - CO_2 molar fraction at the outlet versus time for the two nozzle configurations

The lower outlet CO_2 molar fraction obtained with the three-nozzle configuration is consistent with the previous observations, showing higher absorption in this case due to the better droplet dispersion and larger gas–liquid interfacial area. Other than higher absorption efficiency, the optimised configuration requires a longer time to reach convergence, as a consequence of the increased capture. Although the three-nozzle configuration entails a higher computational cost because of its greater complexity, it is adopted for the subsequent analyses, since it is more representative of real industrial operation.

5.3.2 Gas and liquid flow-rate sensitivity

Figure 5.7 presents the sensitivity analysis of the CO₂ capture performance with respect to the gas flow rate. Three gas flow rates are investigated, 75, 100, and 150 m³/h, using two liquid flow rates of 4 and 2 L/min and a droplet diameter range of 1500-2000 μm. The plots show the temporal evolution of the CO₂ mole fraction at the outlet of the tower.

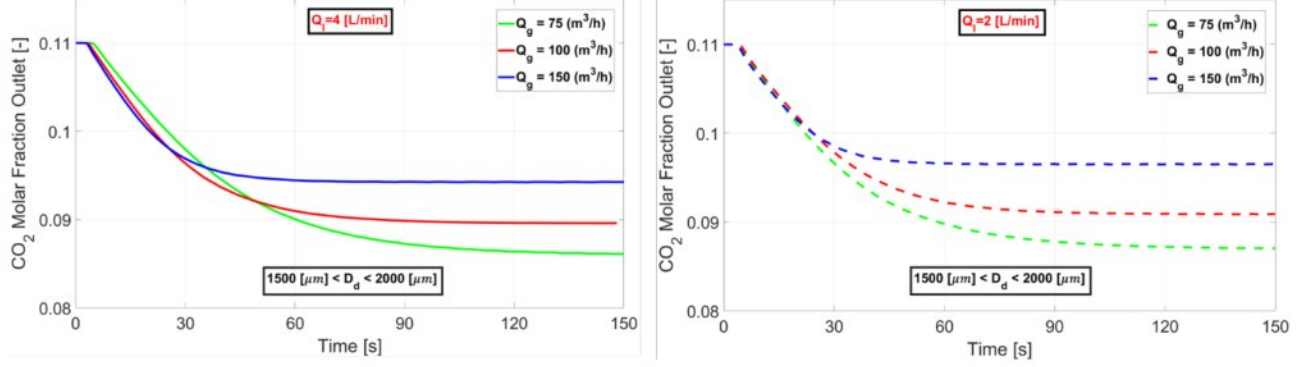


Figure 5.7 - CO₂ molar concentration at the outlet over time for different gas flow rates of 75, 100, and 150 m³/h and two liquid flow rates of 4 and 2 L/min

As shown in the nozzle-configuration comparison, the CO₂ molar concentration at the tower outlet decreases continuously over time in all simulation cases, due to absorption by the liquid solvent. For both Q_L , the highest gas flow rate of 150 m³/h involves the largest outlet molar fraction, corresponding to the lowest capture efficiency. Using higher gas flow rates increases the gas velocity, which enhances turbulence and droplet mixing, leading to better liquid dispersion and a larger interfacial area for mass transfer. On the other hand, the higher velocity reduces the droplet residence time and thus the gas-liquid contact time, which leads to a lower overall absorption efficiency.

Figure 5.8 shows the contour of carbon capture efficiency η , defined by formulation (5.1), within the spray tower domain, together with the droplets Na₂CO₃ molar concentration for two different gas flow rates. The operating conditions are Q_L of 12 L/min, a droplet diameter range of 200-1500 μm droplets diameter ranges, and Q_g of 100 and 200 m³/h.

$$\eta = \left(\frac{y_{CO_2, in} - y_{CO_2}}{y_{CO_2, in}(1 - y_{CO_2})} \right) \times 100\% \quad (5.1)$$

5 Results

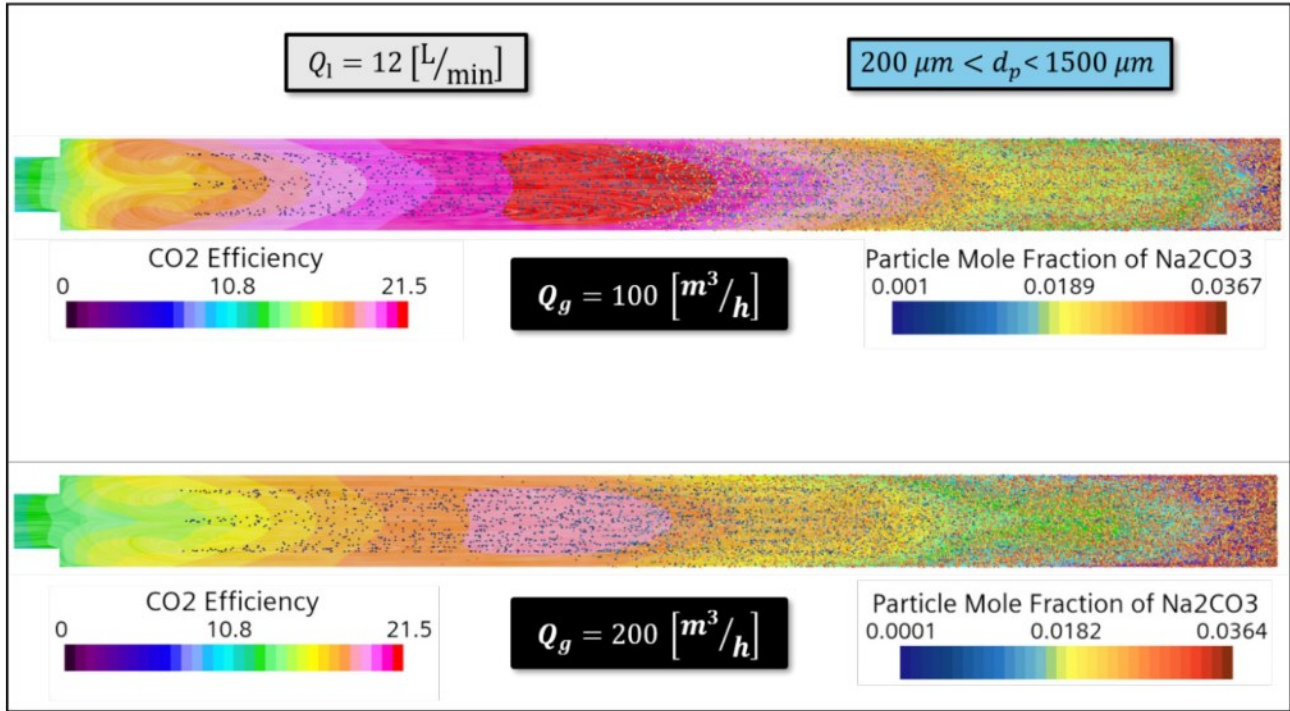


Figure 5.8 - CO₂ capture efficiency in the spray tower and Na₂CO₃ droplet molar concentration for two gas flow rates at constant liquid flow rate

Lower Q_g conditions ($100 \text{ m}^3/\text{h}$, upper image) exhibit higher absorption efficiency, with a maximum in the region just below the injectors, shown in red, in agreement with the previous results. At the higher Q_g ($200 \text{ m}^3/\text{h}$, bottom image), droplet acceleration increases dispersion and radial mixing, but the residence time decreases, which reduces the overall absorption efficiency.

Na₂CO₃ droplet molar concentration is a key indicator of absorption performance, as it represents the final product of reactions (2.11)–(2.16) in the liquid phase between CO₂ and NaOH. The figure shows an increase in droplet CO₂ molar concentration from the injection zone towards the bottom of the tower, highlighting the continuous mass transfer of CO₂ from the gas phase to the liquid and the subsequent reactions.

To analyse the influence of varying liquid flow rates, Figure 5.9 presents six cases, combining three Q_g of 75, 100, and $150 \text{ m}^3/\text{h}$, with two Q_l of 4 and 2 L/min .

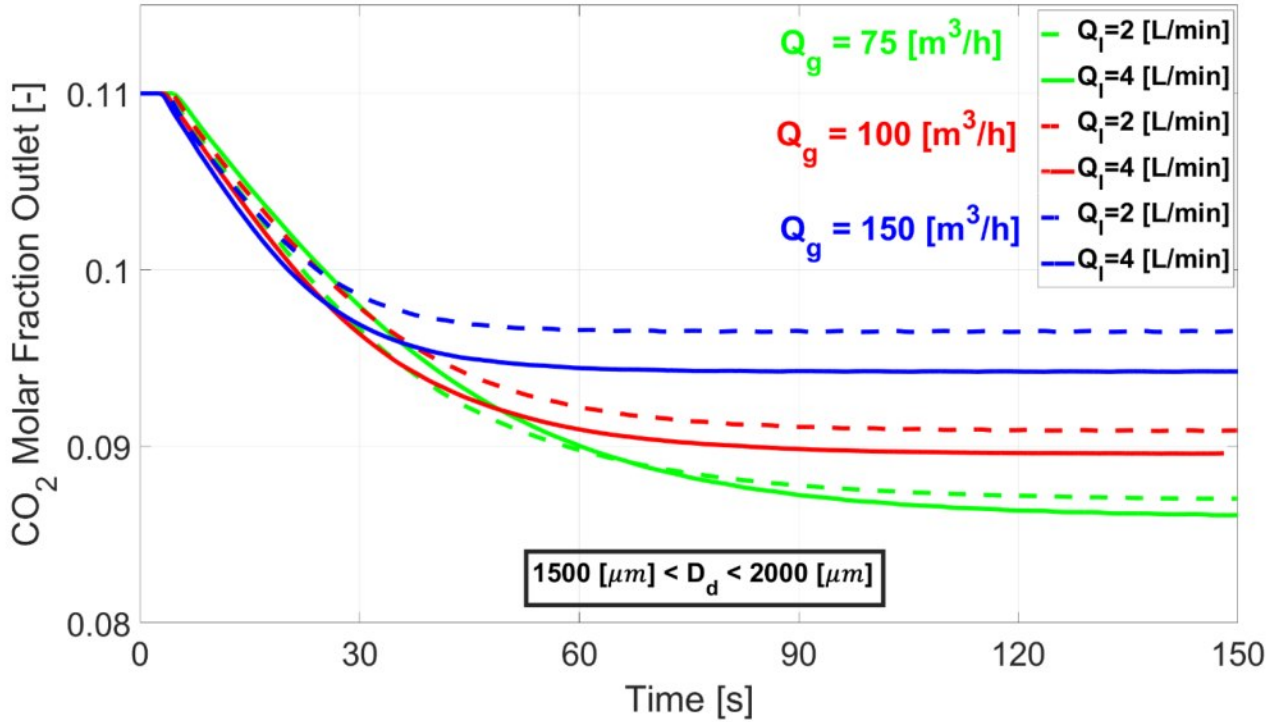


Figure 5.9 – CO₂ molar concentration at the outlet over time for varying gas and liquid flow rates

The outlet CO₂ molar fraction profiles exhibits higher absorption for the cases with the higher L/G ratio, compared to the lower ones. The improved capture performance at higher L/G ratio is due to the larger amount of solvent available to contact the gas stream, which enhances the transfer of CO₂ to the liquid phase. In addition, the higher liquid flow rate increases the number of droplets injected into the tower and, due to the higher injection pressure, produces smaller droplets. These effects increase the gas–liquid interfacial area and therefore lead to improved absorption.

Moreover, variations in liquid flow rate affect turbulence intensity and momentum exchange between the phases, due to the different droplet injection velocities. Figures 5.8 and 5.9 show the spatial distribution and the centreline evolution of the turbulent kinetic energy (TKE) for two cases with the same Q_g of 200 m³/h and two Q_l of 4 and L/min. In Figure 5.8, the TKE distribution exhibits a maximum in the injection zone, between the lateral gas stream and the nozzles, where shear and momentum exchange are high and droplet velocities are larger, as indicated by their blue colour. Moving towards the bottom of the column, the droplets decelerate and the TKE decreases accordingly.

The main difference between the two cases lies in the absolute values of TKE and droplet velocity magnitude, which are higher for the 4 L/min liquid flow rate, leading to stronger mixing, more intense gas–liquid interaction, and enhanced mass transfer. The zoomed views in Figure 5.10 highlight how the different injection velocities influence droplet distribution inside the tower: the lower liquid flow rate produces lower droplet velocities, causing droplets to accumulate more than in the higher-flow case. Droplets in contact with the gas stream are slowed down, whereas droplets with higher velocity (shown in blue) correspond to larger

droplets, which are less affected by the gas stream because they are heavier and tend to maintain their velocity and trajectory.

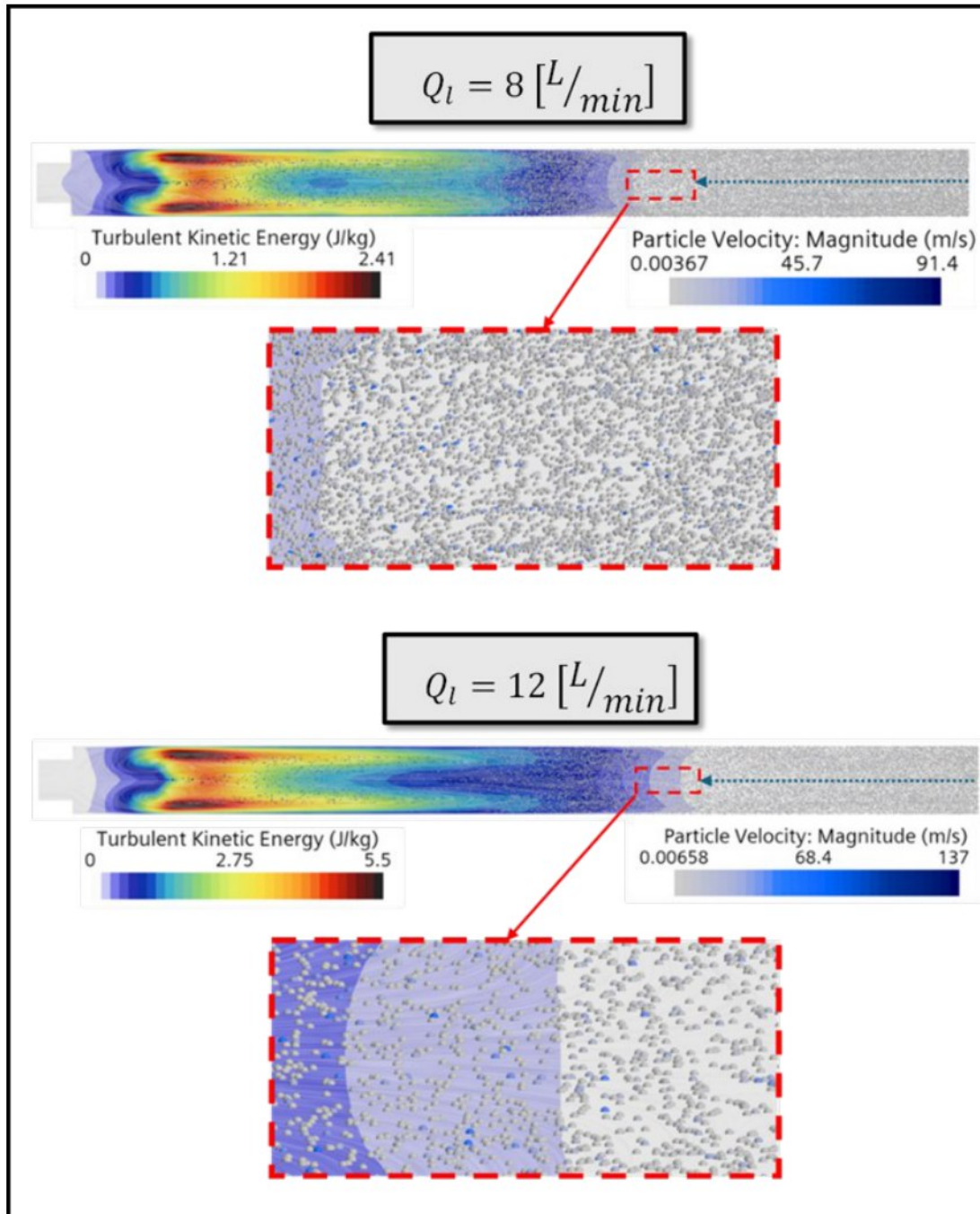


Figure 5.10 – Contours of turbulent kinetic energy TKE (J/kg) for two liquid flow rates, overlaid with droplet velocity magnitude

Figure 5.11 shows the TKE profiles along the vertical centreline, which, in agreement with the previous analysis, are higher for the case with the larger Q_l and display a peak between 3.0 and 3.5 m in the injection zone, where TKE is enhanced.

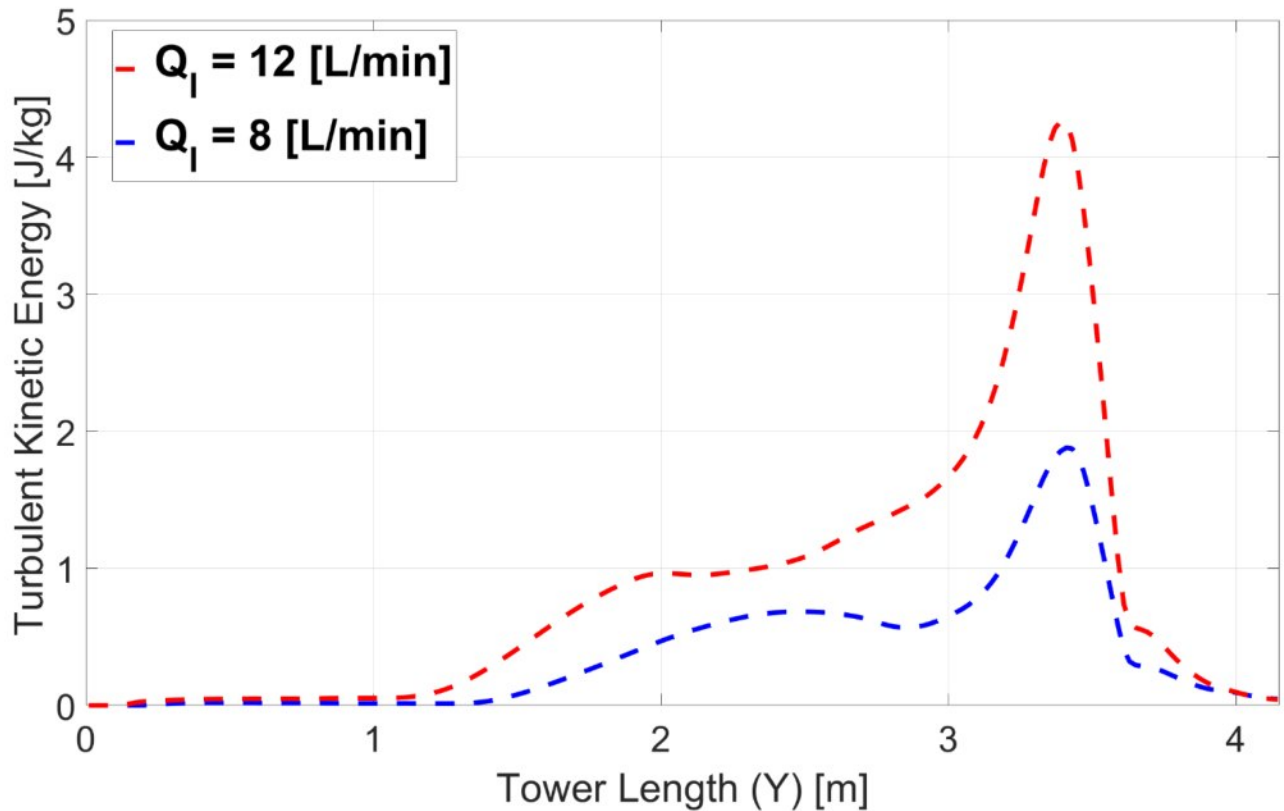


Figure 5.11 – TKE profiles along the tower vertical centreline for two liquid flow rate cases

Figure 5.12 examines the same configurations as in the previous analysis, focusing on how the different liquid flow rates influence droplet trajectories and residence time.

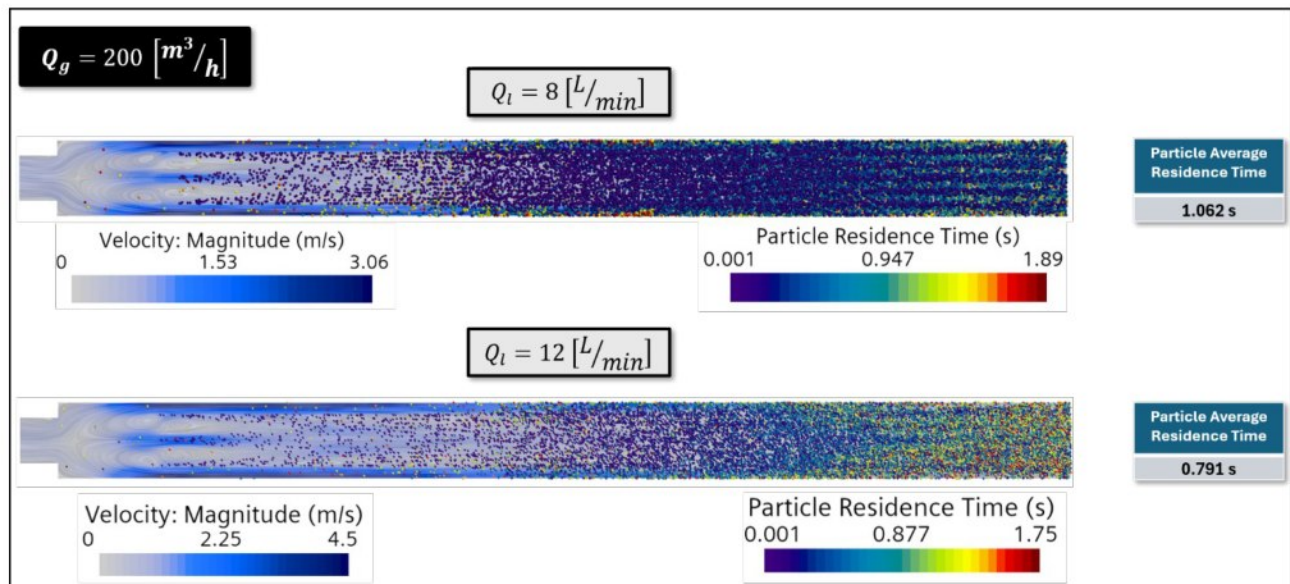


Figure 5.12 – Droplet residence time for two liquid flow rate configurations at constant gas flow rate

The results shown in the figure agree with the previous findings. A lower Q_l leads to lower droplet velocities, which increases the average droplet residence time compared with the

case with a higher liquid flow rate. In Figure 5.10, the gas–liquid interactions inside the tower are also visible, through the lateral dispersion of droplets and their reverse migration toward the outlet. At the lower gas flow rate, finer droplets injected at low velocities are more likely to interact with localized vortices and follow the gas flow. Under higher liquid flow rate conditions, this mechanism is enhanced because of the increased atomization, which produces smaller droplets. Unlike larger droplets that mainly follow gravity-driven paths, smaller droplets are more strongly affected by turbulent mixing. Therefore, droplet size also plays a key role in capture performance by controlling droplet residence time and their interactions with the gas phase.

5.3.3 Effect of droplet size distribution on CO₂ capture performance

To evaluate the effect of droplet size distribution on carbon capture efficiency, three different droplet diameter ranges are tested, all with the same maximum diameter of 2000 μm and decreasing minimum diameters of 1500, 1000, and 500 μm . These ranges are imposed by modifying the parameters of the Rosin–Rammler distribution, given in equation (3.60), in the simulation model. The simulation results are shown in Figure 5.13 for Q_g of 75 m^3/h and Q_l of 4 L/min .

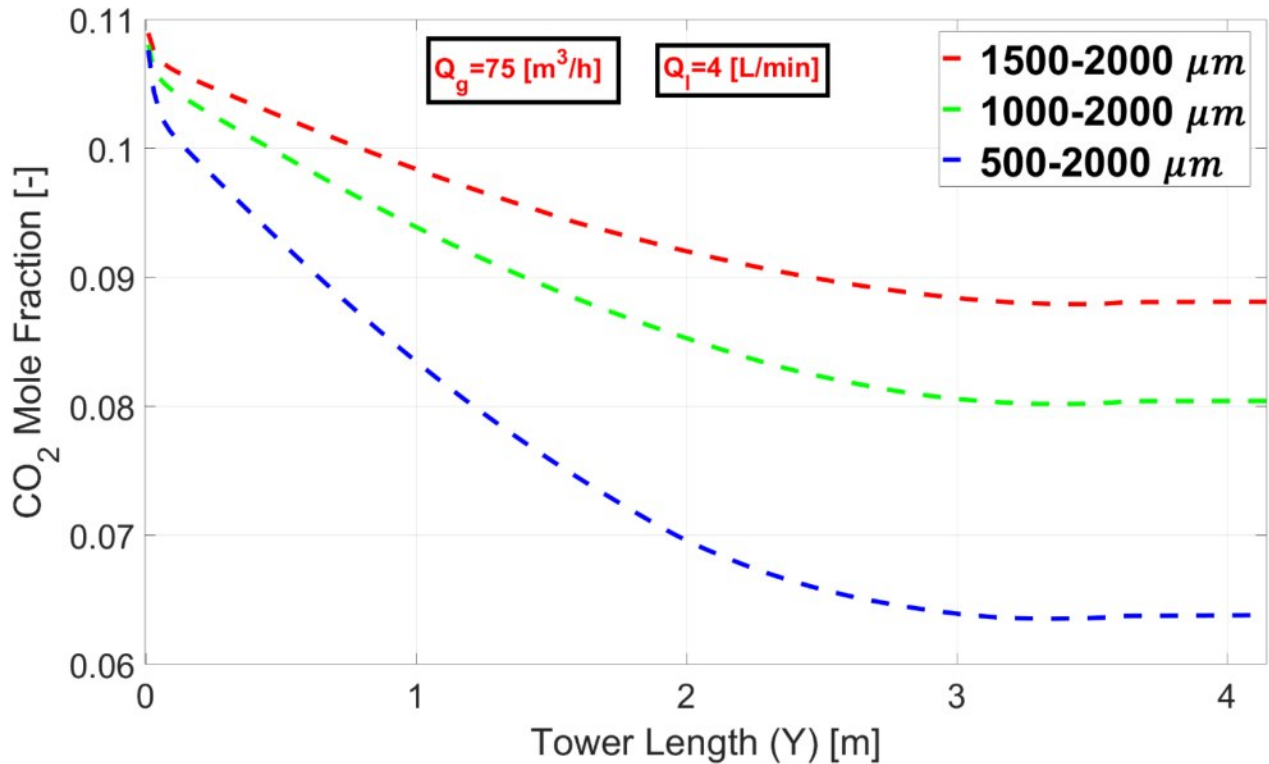


Figure 5.13 - CO₂ molar fraction along the vertical centreline for three droplet diameter distributions

The results show the CO₂ molar concentration along the tower height, with the distribution having the smallest minimum droplet diameter exhibiting the lowest CO₂ concentration within the tower. This highlights the effect of injecting finer droplets, which increases absorption efficiency due to their higher surface-area-to-volume ratio compared with larger droplets, thereby enhancing the gas–liquid interfacial area available for mass transfer. In addition, smaller droplets are lighter and more likely to follow the gas flow, which increases their residence time and allows a greater amount of CO₂ to be absorbed.

Figure 5.14 shows the time evolution of the absorption efficiency, defined in equation (4.11), for the three droplet diameter ranges 1500–2000, 1000–2000, and 500–2000 μm (shown in

blue gradients), together with an additional distribution with a minimum diameter of 200 and increasing maximum diameters of 500, 700, and 1000 μm (shown in red gradients). The simulations are performed with a gas flow rate of 75 m and a liquid flow rate of 4 L/min.

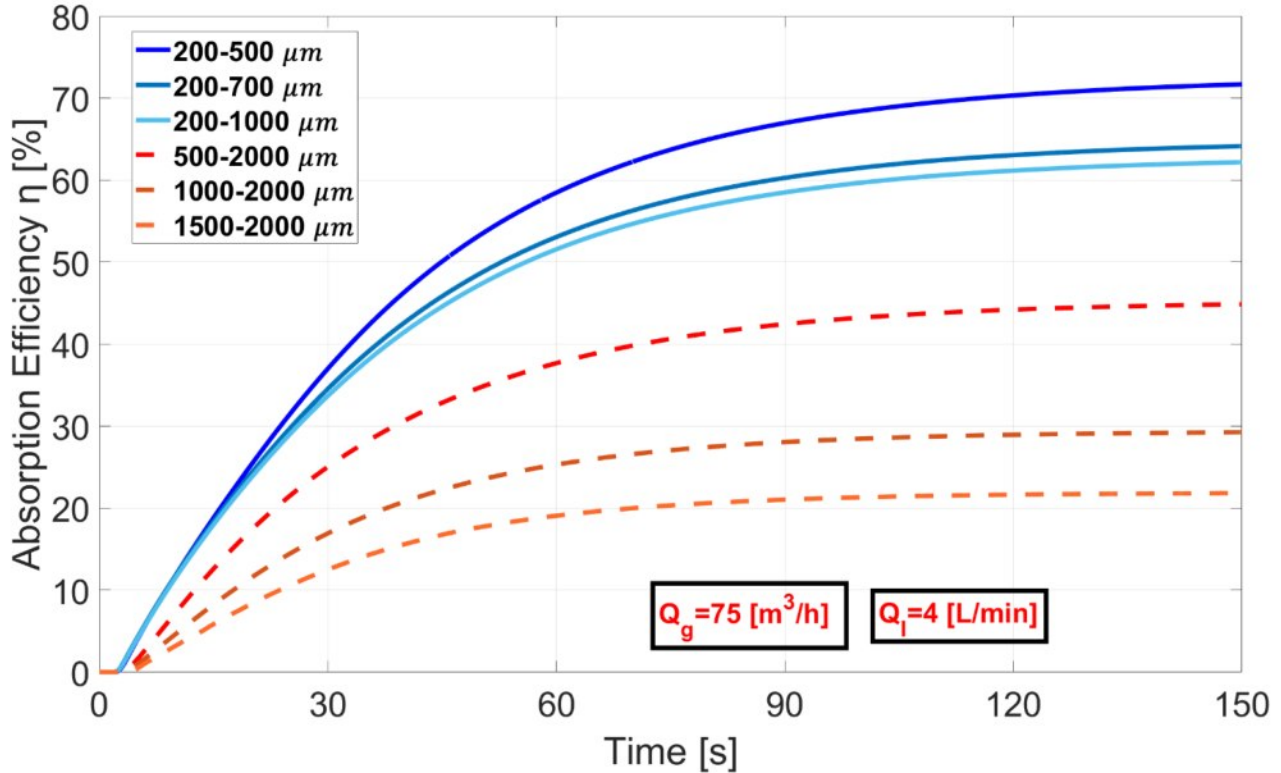


Figure 5.14 – Time evolution of absorption efficiency for six droplet diameter distributions

Lower droplet diameter distributions (the blue curves) show a faster increase in absorption efficiency during the initial phase of the simulations and reach higher final values, indicating that the presence of finer droplets inside the tower enhances the capture process. In particular, the 200–500 μm distribution reaches an efficiency of 72%, in contrast with the much lower efficiency of 22% obtained for the larger droplet distribution of 1500–2000 μm .

Droplet diameter distribution is a key factor in maximizing the performance of the spray tower absorber. It depends mainly on the nozzle design, the injection pressure, and the liquid flow rate, and therefore requires careful optimization to identify the most effective configuration. In contrast, the gas flow rate is dictated by the specific process in which the spray tower operates and may vary over time. As a result, all these parameters, droplet size distribution, liquid flow rate, injection conditions, and gas flow rate, must be optimized together to achieve the best overall capture performance. The conditions that result in the highest absorption efficiency in the simulations are multi-nozzle configuration, with high liquid-to-gas ratio, and fine spray.

5.4 Carbon capture performance analysis with the evaporation model

Figure 5.15 shows the contour of H_2O concentration within the spray tower for a simulation with for Q_g of $75 \text{ m}^3/\text{h}$, Q_l of 4 L/min , and a droplet diameter distribution of $1500\text{--}2000 \mu\text{m}$.

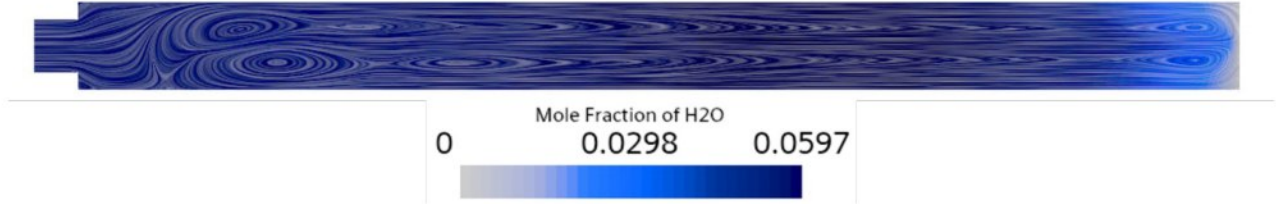


Figure 5.15 – Contour of H_2O mole concentration within the spray tower for a simulation with the evaporation model enabled

The figure shows that at the inlet the water concentration in the gas phase is zero, since dry flue gas is assumed, whereas moving toward the top of the tower the molar fraction of H_2O increases because of the continuous evaporation of droplets and the associated mass transfer of water from the liquid to the gas phase.

The Vapor diffusion limited evaporation model used in this work depends on the Spalding transfer number B and the conductance g , which are sensitive to the temperatures of both gas and liquid phases, the droplet size, and the component concentrations in each phase. To analyse how evaporation influences the capture process, and how different operating configurations affect evaporation, the following subsections present a series of comparative studies.

5.4.1 Effects of evaporation on the carbon capture process

The effect of evaporation in the model is first examined by comparing two simulations with the same configuration, one without the evaporation model and one with it enabled, as shown in Figure 5.16. A gas flow rate of $75 \text{ m}^3/\text{h}$, a liquid flow rate of 4 L/min , and a droplet diameter distribution of $1500\text{--}2000 \mu\text{m}$ are used.

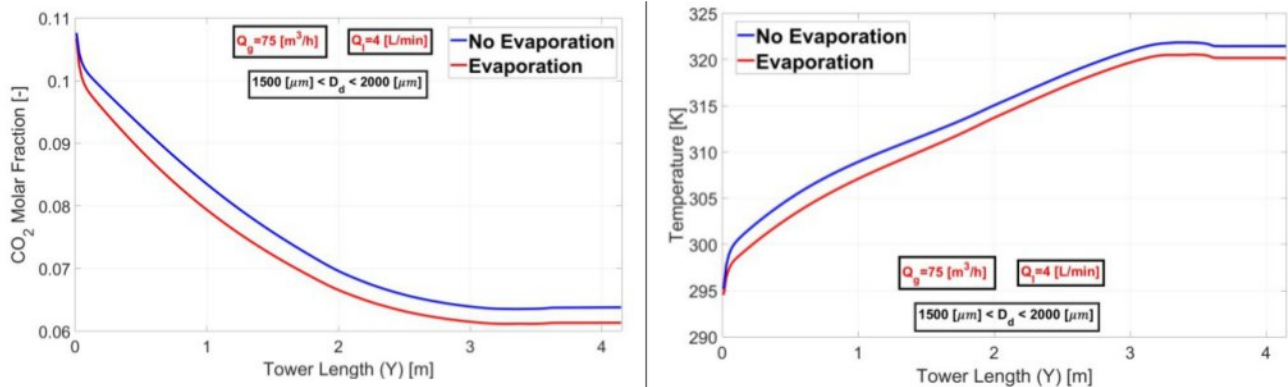


Figure 5.16 – Comparison of CO_2 molar fraction (on the left) and temperature (on the right) profiles along the tower for cases with and without the evaporation model

The left plot shows how introducing the evaporation model leads to a decrease in the overall CO_2 molar concentration within the tower, while the overall trends remain essentially the same. The right plot shows the temperature evolution along the tower height, and also in this case the temperatures are lower throughout the spray tower, with the evaporation enabled.

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This behaviour is consistent with the physics, since droplet evaporation is an endothermic phenomenon that requires energy from the liquid phase and also extracts energy from the surrounding gas, cooling the system.

To better understand the effects of evaporation, the same cases are analysed by comparing other key parameters in the absorption process. Figure 5.17 presents the comparison for average droplet temperature, average droplet surface-area-to-volume ratio, average NaOH concentration within the droplets, and average reaction rate..

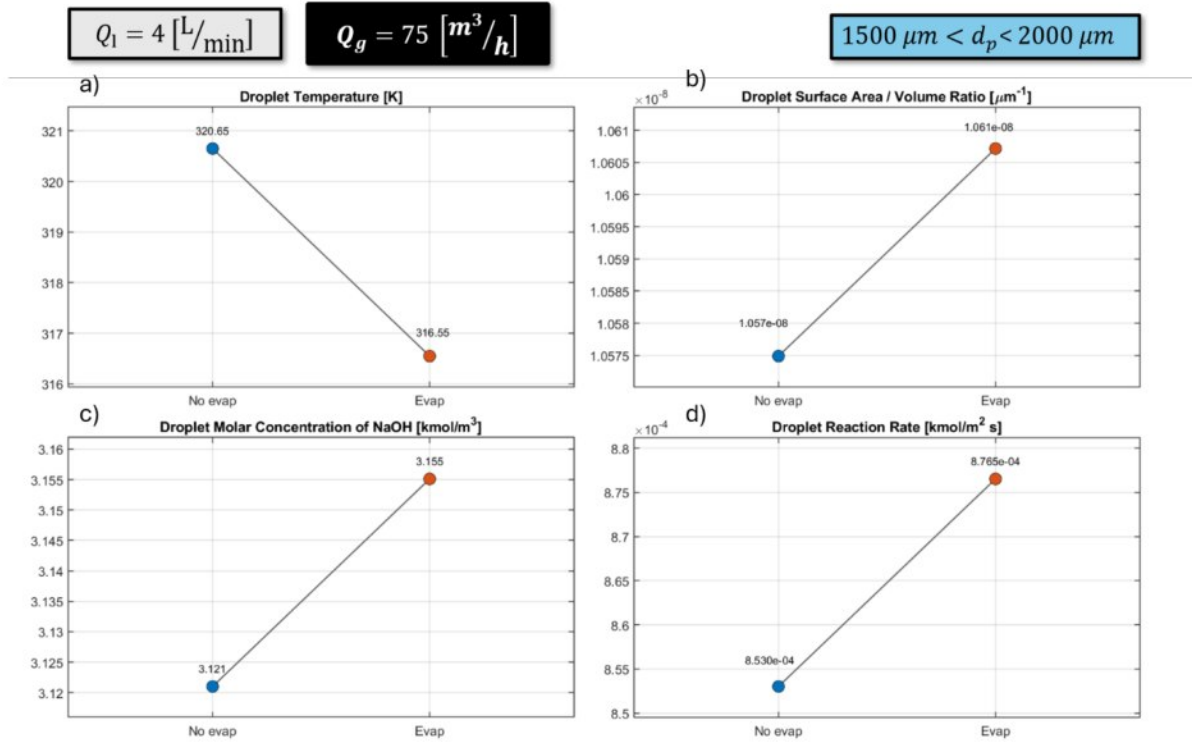


Figure 5.17 - Comparison between of average droplet temperature (a), average surface-area-to-volume ratio (b), average NaOH concentration within the droplets (c) and average reaction rate (d) for two cases with the same configuration, with and without the evaporation model.

Adding the evaporation model to the simulations produces several effects. As discussed above, the gas-phase temperature decreases, and the droplet temperature also decreases, as shown in Figure 5.17(a), due to the endothermic nature of evaporation. In the spray tower considered here, the operating conditions lead to a limited amount of vapour transfer between the phases, so the temperature drops are about 2 K in the gas phase and 4 K in the liquid phase. In addition, evaporation reduces droplet size and thereby increases the average surface-area-to-volume ratio, Figure 5.17(b), which is inversely proportional to droplet diameter. The transfer of water from the droplets to the gas phase also increases the effective solvent content in the liquid phase, as indicated in Figure 5.17(c) by the higher NaOH molar concentration. The reaction rate in equation (3.47) depends on these parameters: it benefits from the higher solvent concentration, but the lower droplet temperature reduces it through the Arrhenius expression in equation (3.52) and the kinetic relation for the reaction-rate constant in equation (3.50). Moreover, the combined decrease in temperature and increase in solvent concentration reduce the diffusion coefficient D_l in equation (3.47) by increasing the solvent viscosity. However, because the temperature drop is relatively small, and because the increase in solvent concentration and in surface-area-to-volume ratio is more significant

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in the evaporative case, the overall reaction rate increases when evaporation is included, as shown in in Figure 5.17(d) leading to a lower CO_2 molar concentration at the outlet.

5.4.2 Effect of gas and liquid flow rates on evaporation

To evaluate the effect of varying the liquid flow rate on evaporation, Figure 5.18 shows the H_2O molar fraction along the tower vertical centreline for two Q_l of 4 L/min (on the left) and 2 L/min (right), each tested at three Q_g of 75, 100, and $150\text{ m}^3/\text{h}$.

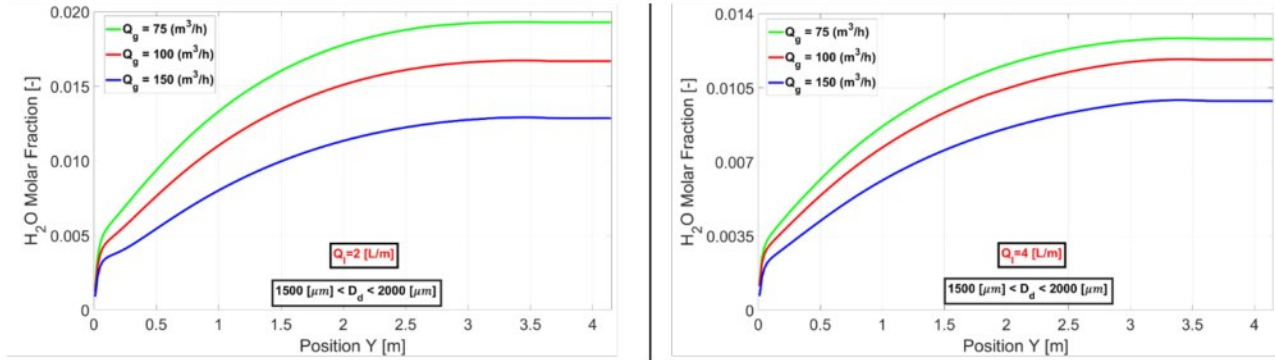


Figure 5.18 – H_2O molar fraction along the vertical centreline for two Q_l (4 L/min on the left, 2 L/min on the right) at Q_g of 75, 100, and $150\text{ m}^3/\text{h}$

The profiles show the same behaviour discussed earlier, with a continuous increase of H_2O molar fraction and a stabilisation in the upper section of the tower. For both liquid flow rates, higher H_2O molar fractions are observed at the lowest gas flow rate of $75\text{ m}^3/\text{h}$ (green curves). This is because lower gas flow rates increase the gas residence time, enhancing CO_2 absorption by the liquid phase and thus promoting reactions between CO_2 and NaOH . The exothermic nature of these reactions releases heat into both phases, raising their temperatures and creating conditions that favour evaporation. In addition, the longer residence time at lower gas velocities allows more time for droplet evaporation. On the other hand, at higher gas flow rates, the lower degree of reaction between CO_2 and NaOH and shorter gas–liquid contact time result in lower amounts of H_2O transferred to the gas phase.

To study the effect of varying the liquid flow rate, Figure 5.19 shows the H_2O molar fraction along the tower vertical centreline for two liquid flow rates, 4 and 2 L/min , at a fixed gas flow rate of $75\text{ m}^3/\text{h}$.

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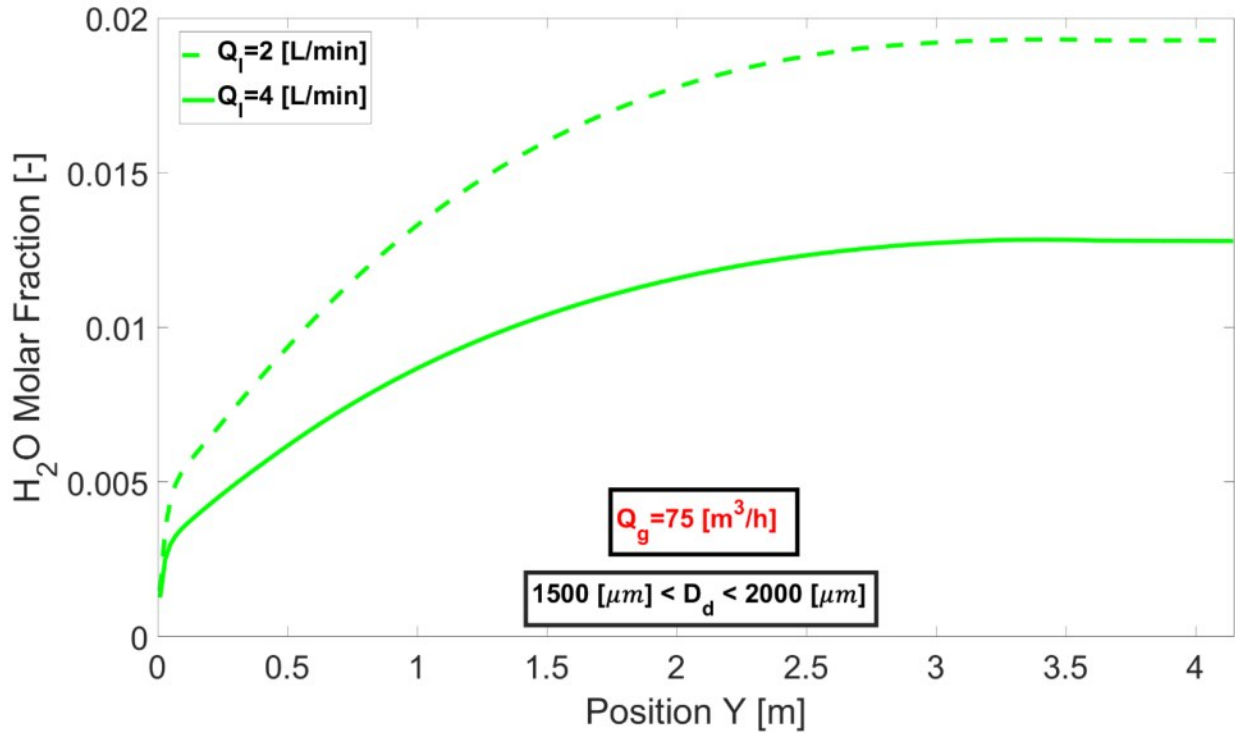


Figure 5.19 – H₂O molar fraction along the vertical centreline for two liquid flow rates of 4 and 2 L/min at a gas flow rate of 75 m³/h.

The results show a higher H₂O molar fraction for the lower liquid flow rate of 2 L/min (dashed line), indicating that a smaller amount of liquid in the spray tower enhances evaporation. This appears to contradict the non-evaporative case, where a lower liquid flow rate was associated with reduced CO₂ absorption and fewer reactions in the liquid phase, leading to a smaller temperature increase and, in principle, less favourable conditions for evaporation. To investigate this behaviour, Figure 5.20 compares the Spalding transfer number B of equation (3.41) and the conductance g of equation (3.43), which together govern the evaporation rate, for the two gas–liquid flow rate conditions.

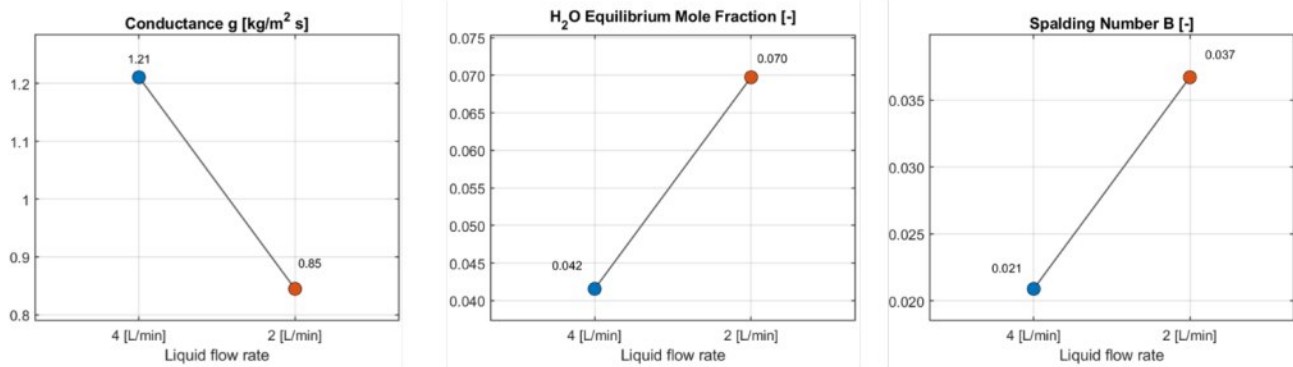


Figure 5.20 – Comparison of conductance, H₂O equilibrium molar fraction, and Spalding number for two different liquid flow rates of 4 and 2 L/min

The results highlight that for 4 L/min the conductance g in equation is higher. This trend can be explained by analysing how the dependent terms vary. The particle Sherwood number Sh_p , of equation (3.45) depends on the particle Reynolds number Re_p , which increases at higher liquid flow rates because the relative velocity is higher, as showed by equation (3.28). In addition, a higher liquid flow rate produces finer droplets, reducing their diameter, which

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appears in the denominator of the conductance expression. The other terms tend to reduce the conductance at higher liquid flow rate, as in this case the overall gas temperature is higher due to more intense reactions, which increases the diffusivity of water in air and decreases the gas density. However, the effects of the enhanced Sherwood number and the reduced droplet diameter are dominant, and they explain the higher conductance for the higher liquid flow rate.

The second plot shows the H_2O equilibrium molar fraction, which is higher for the lower liquid flow rate. With a lower L/G, the smaller number of droplets inside the tower is less cooled by the surrounding liquid than in the higher L/G case, so droplet temperatures remain higher. This increases the saturation temperature in equation (3.37) and thus the equilibrium molar fraction of equation (3.42), leading to a larger Spalding transfer number B . The increase of B at lower liquid flow rate, even though the conductance g is lower, results in a higher overall evaporation rate in this case compared with the higher liquid flow rate.

In addition, as discussed in the previous section, a lower liquid flow rate also leads to lower droplet injection velocities, which increase the residence time. Since the increased gas–liquid contact time provides more time for droplet evaporation, this aspect also play an important role in the behaviour observed above.

To complete the analysis, Figure 5.21 compares the H_2O molar fraction within the tower for the other two gas flow rates of 100 and 150 m^3/h at different liquid flow rates, showing the same trend described previously.

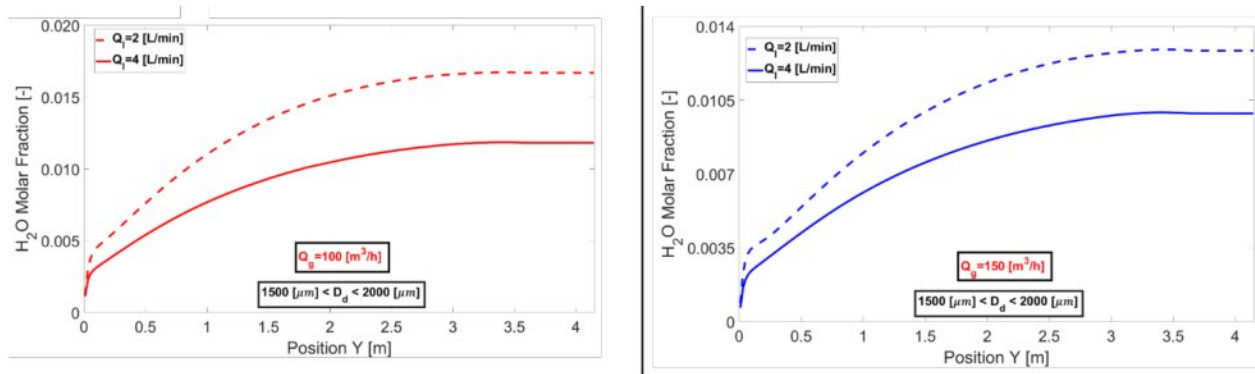


Figure 5.21 – H_2O molar fraction in the tower for gas flow rates of 100 and 150 m^3/h at different liquid flow rates

5.4.3 Effect of droplet diameter distribution on evaporation

Figure 5.22 shows the evolution of the H_2O molar fraction along the tower vertical centreline for three cases with the same gas and liquid flow rates, Q_g of 75 m^3/h and Q_l of 4 L/min , and different droplet diameter distributions. All distributions share the same maximum diameter of 2000 μm , while the minimum diameter decreases from 1500 to 1000 and 500 μm .

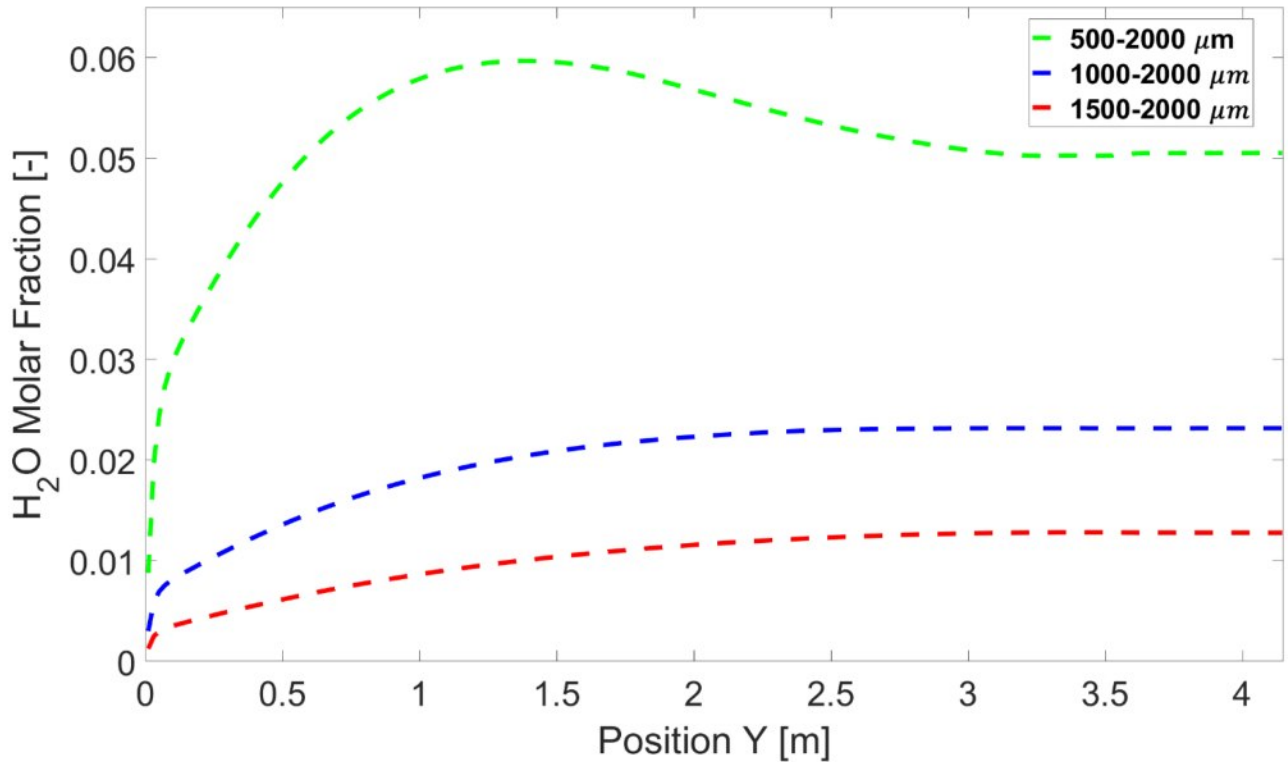


Figure 5.22 – H₂O molar fraction along the tower vertical centreline for three droplet diameter distributions at the same operating conditions

The results highlight that the distribution with the smallest minimum diameter (the green curve) exhibits the highest evaporation, with a clear maximum around a height of 1–1.5 m, whereas the other two distributions show a much flatter profile. In addition, the change from the 1000–2000 μm distribution to the 500–2000 μm one is significantly larger than the change from the 1500–2000 μm to the 1000–2000 μm distribution, indicating a non-linear correlation between droplet size and evaporation. The configuration with finer droplets absorbs more CO₂, for the reasons discussed in the previous subchapters, which intensifies the chemical reactions inside the droplets and raises the temperature of both gas and liquid phases. The higher temperatures enhance evaporation, increasing the Spalding transfer number B , the key parameters governing evaporation, as discussed previously. Finer droplets also have a longer residence time because their lower settling velocity and more effective mixing keep them suspended in the gas phase for longer, as previously analysed, improving evaporative transfer. In contrast, the lower temperatures and shorter gas-liquid contact time in the cases with larger droplets reduce the evaporation rate.

To better understand this phenomenon, Figure 5.23 shows the percentage variation, relative to the case without the evaporation model, for the three evaporation simulations in terms of

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droplet and tower average temperature, average surface-area-to-volume ratio, and average NaOH concentration within the droplets.

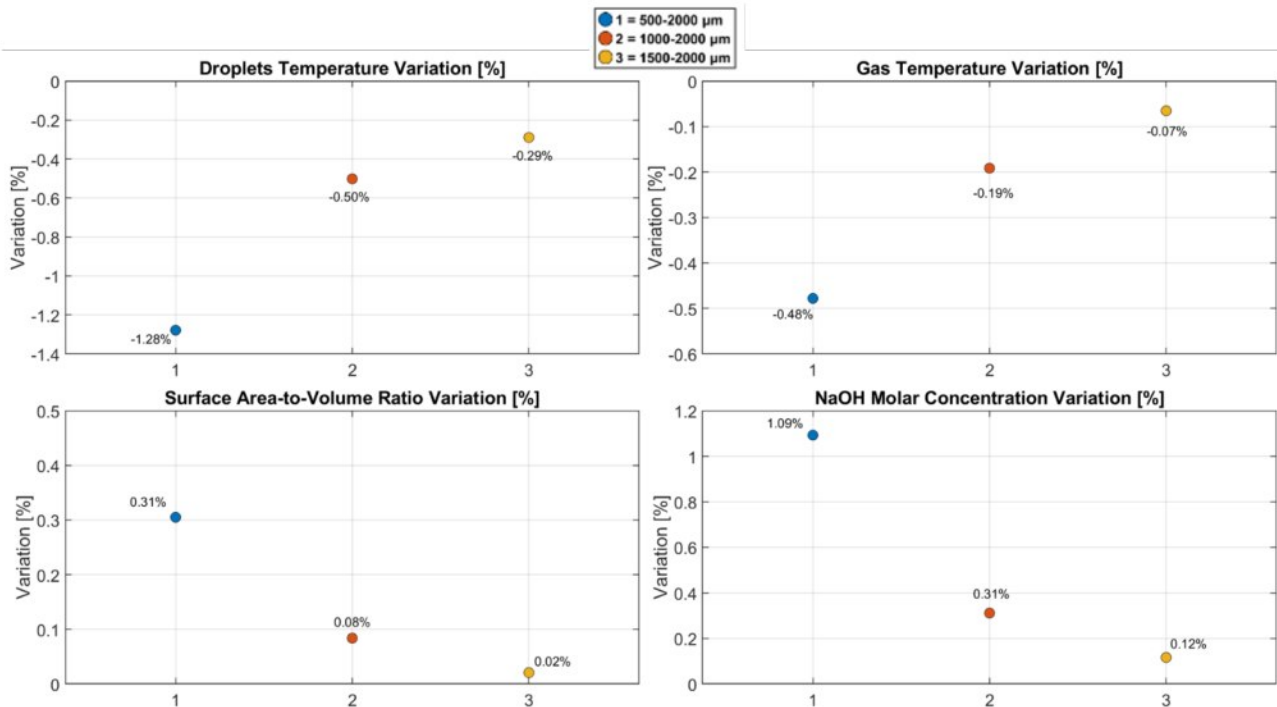


Figure 5.23– Percentage variation, with and without the evaporation model, for three droplet diameter distributions in terms of droplet and tower temperature, surface-area-to-volume ratio, and average NaOH concentration within the droplets

The results show that, although the variations of all parameters are small, they are larger for the cases with smaller droplet sizes and remain very limited for the other two distributions. This behaviour leads to a greater increase in absorption efficiency for the 500–2000 μm distribution, due to increased evaporation, as shown in Figure 5.24.

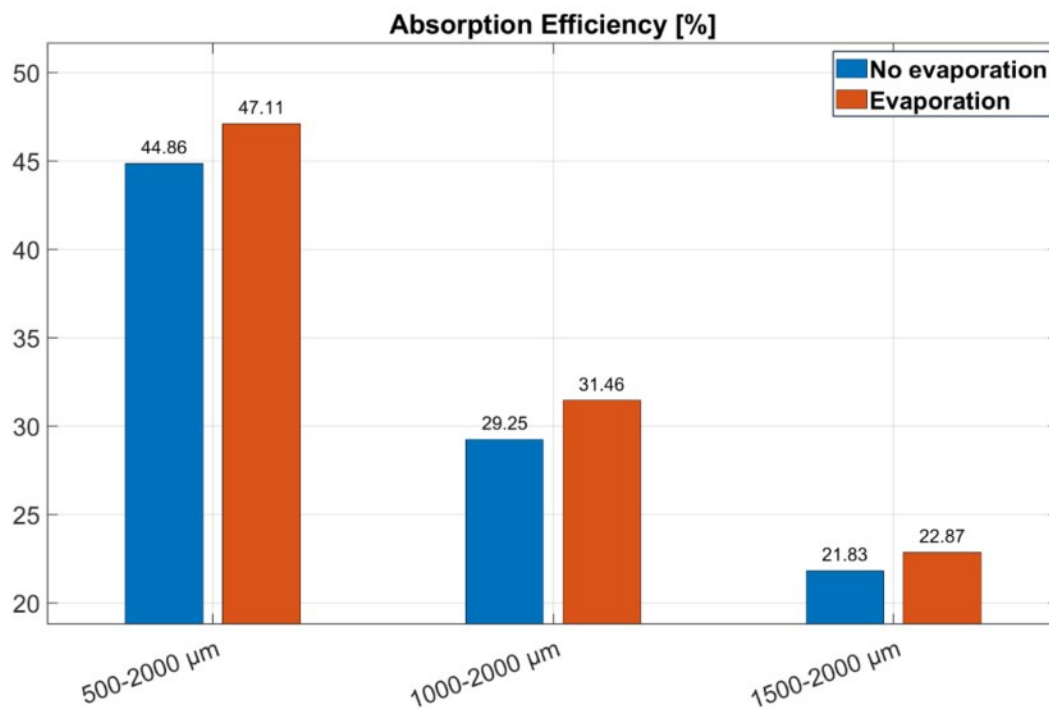


Figure 5.24 - Comparison of absorption efficiency between cases with and without the evaporation model for three droplet diameter distributions

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To conclude, the evaporation model affects the overall CO₂ capture by modifying gas and liquid temperatures, solvent composition, and droplet size, and it produces a slight increase in absorption efficiency. The evaporative mass transfer is controlled by the Spalding transfer number B and the conductance g , as well as by the droplet residence time, all of which are sensitive to the operating conditions. Although evaporation remains limited in these simulations, it is enhanced by lower gas and liquid flow rates and by finer droplet diameter distributions.

6 Conclusions and further developments

Carbon capture is a key option to mitigate anthropogenic CO₂ emissions from industrial activities, particularly from hard-to-abate processes that currently depend on fossil fuels. This work presents a 2D CFD model of a spray tower system that captures CO₂ from flue gas via chemical absorption using a 15 wt% NaOH solvent. The model represents a 4 m-tall pilot-scale carbon capture unit developed and constructed by Grimaldi Development AB in collaboration with KTH Royal Institute of Technology at the Tekniska verken waste-to-energy plant in Linköping, Sweden. The simulations were performed with STAR-CCM+ under real industrial conditions, with a flue gas CO₂ concentration of 11 mol% and gas flow rates ranging from 75 to 200 m³/h, liquid flow rates from 2 to 12 L/min, different numbers of cone nozzles (1 to 3), and various droplet diameter distributions.

The model adopts an Euler–Lagrange multiphase framework with a dispersed multiphase approach and solves the RANS equations with a k – ε turbulence model to capture mass, momentum, and energy transfer between phases, turbulent mixing, and chemical reactions inside the droplets. A quasi-steady evaporation model describes water transfer from liquid to gas, allowing investigation of how evaporation modifies phase properties and the effects on the overall absorption process. Transient simulations were carried out to analyse unsteady phenomena such as droplet trajectories, residence time, gas–liquid interactions, turbulent dispersion, and dynamic mass transfer, which are not accessible in steady-state simulations and are difficult to investigate experimentally. First, simulations without evaporation model were run, to study absorption performance and sensitivity to nozzle configuration, gas and liquid flow rates, and droplet size distributions. The same configurations were then simulated with evaporation enabled to assess both the impact of evaporation on the process and the influence of operating conditions on this phenomenon.

A mesh-independence study was first conducted to ensure that the main results do not depend on grid refinement, and the selected base size provides a good compromise between accuracy and computational cost. The comparison between simulation results and experimental data shows an acceptable level of agreement. This is considered sufficient for the aims of this work, which focus on developing and analysing the evaporation model rather than on providing a fully representative 2D model of the 3D process. The nozzle configuration analysis shows that moving from a single 30° cone nozzle to three cone nozzles with the same angle but oriented towards the tower centre, improves droplet dispersion, reduces liquid losses to the wall, and increases absorption efficiency. The gas–liquid flow rate analysis without evaporation highlights that higher L/G ratios lead to higher capture efficiency. Higher gas flow rates enhance droplet dispersion and turbulent mixing, increasing the interfacial area for mass transfer, but they strongly reduce droplet residence time, which is a key parameter for absorption performance. This explains why the highest CO₂ capture is observed at lower gas flow rates. Increasing the liquid flow rate leads to higher injection velocity and, due to higher nozzle pressure, improved atomisation and a finer spray. This increases the turbulent kinetic energy in the tower and, despite a slightly reduced residence time, improves droplet dispersion because lighter droplets more closely follow the gas flow. The larger amount of solvent in the tower at higher liquid flow rates also favours CO₂ absorption. The analysis of droplet diameter distributions shows that smaller droplet size ranges increase CO₂ capture due to the higher gas–liquid interfacial area available for mass transfer and the longer average residence time.

6 Conclusions and further developments

The comparison between simulations with and without evaporation shows that droplet evaporation tends to lower the temperature of both gas and liquid, due to the endothermic nature of the phase change, while producing a slight increase in absorption efficiency. This occurs because, although lower temperature slows the chemical reactions, the higher NaOH concentration inside droplets and the increased surface-area-to-volume ratio create more favourable conditions for CO₂ absorption. The gas-flow-rate sensitivity analysis indicates that lower gas flow rates promote evaporation due to longer residence time and higher temperatures linked to more intense reactions. Lower liquid flow rates also lead to higher evaporation because the reduced liquid mass weakens the cooling effect on the gas, allowing higher temperatures and thus a stronger driving force for evaporation; in addition, lower droplet velocities increase residence time. Finally, the analysis of different droplet diameter distributions under evaporation shows that evaporation remains limited for larger droplets, while finer distributions exhibit higher H₂O mole fraction in the gas phase, due to higher temperatures, larger interfacial area, leading to greater increase in absorption efficiency.

Despite the model being able to capture the overall absorption process and its sensitivity to operating conditions, several limitations must be recognized. First, the simulations are performed in a two-dimensional domain, which is a strong simplification of the real three-dimensional tower. This choice was made to develop and test the evaporation model in a simpler and less computationally expensive framework, and because previous 3D simulations showed a partially axisymmetric behaviour due to the geometric configuration. The main difference compared with the 3D simulations is the longer computational time required to reach convergence in 2D, while the predicted absorption efficiencies remain similar. Second, assumptions about droplet behaviour introduce further simplifications as droplets are treated as rigid spheres, and agglomeration, collision, and coalescence are neglected, providing only a simplified representation of liquid–liquid and liquid–gas interactions. Moreover, the Euler–Lagrange framework does not resolve sub-droplet-scale transport phenomena, such as internal circulation, concentration gradients inside droplets, or interfacial deformation. These simplifications, although necessary to provide computational feasibility, can influence the reliability of the predictions of interfacial area, mass transfer, reaction rates, and overall capture. Finally, the model represents a specific pilot-scale configuration operated under a limited set of conditions, which may not fully reflect performance in full-scale industrial units, where long-term operational issues and more complex hydrodynamics can play an important role. For these reasons, the present work should be seen as a quantitative benchmark and a basis for future optimisation, rather than as a fully predictive tool for all operating scenarios.

Future developments could include the integration of the evaporation model into a full 3D simulation and validation against experimental data for both absorption efficiency and outlet water content in the flue gas. Relaxing the current assumptions on droplet nature and physical and the integration of interface-resolving models would lead to a more accurate representation of gas–liquid interactions. Extending the study to a broader range of operating conditions and tower configurations, such as an increased number of nozzles or side gas inlets, together with scale-up to full-scale applications, would improve the insights into practical carbon-capture performance. Altogether, these developments would make the simulations more representative of real industrial operation and support more reliable design and optimisation of spray-tower CO₂ capture systems.

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