

Article

Computational Fluid Dynamics Modelling of a Laboratory Spray Dry Scrubber for SO₂ Removal in Flue Gas Desulphurisation—Effect of Drying Models

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Abstract: Spray dry scrubbing is widely used for SO₂ abatement, but high removal efficiencies are required for economical operation. Whereas SO₂ removal dependence on the drying rate has been investigated, available modelling work has not addressed the impact of selected drying models on the removal efficiency; instead, a single drying model is often assumed. In the present work, computational fluid dynamics is used to numerically model the SO₂ removal in a laboratory-scale spray dry scrubber. The Euler–Lagrangian framework is used to simulate the multiphase interaction and two drying models are used: the widely used classical *D*₂-law model and the mechanistic model. In addressing shortcomings from previous works, this study also provides a comprehensive model development and robust model validation with quantifiable metrics for goodness-of-fit, including R². Also presented are key parameters associated with SO₂-removal efficiency, including the exit product moisture content and droplet dynamics. The mechanistic model gave a better representation of the SO₂-removal efficiency. The latter was found to be dependent on the inlet temperature, the calcium-to-sulphur and liquid-to-gas (L/G) ratios, with a high L/G ratio having the most significant impact on the removal efficiency, although resulting in a higher product outlet moisture content.

Keywords: flue gas desulphurisation; FGD; spray dry scrubbing; numeric modelling; computational fluid dynamics; CFD; SO₂ removal; desulphurisation efficiency; droplet drying



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1. Introduction

Pollutants such as sulphur dioxide (SO₂), oxides of nitrogen, and particulate matter are produced during the combustion of fossil fuels for power generation and in industries such as iron and steel making during the sintering process [1,2]. These pollutants have negative environmental effects—inhaleation thereof can have adverse effects on heart and lung health, which may even be fatal. Worldwide, governments are imposing stricter legislation to combat the release of these pollutants into the atmosphere [3]. Much work is, therefore, required to ensure that current abatement technologies meet these increasingly more stringent emission standards. The process of SO₂ removal from flue gas is often referred to as flue gas desulphurisation (FGD). Several desulphurisation technologies are employed in the power industry; the most common are scrubber techniques comprising wet FGD, semi-dry FGD, and dry FGD technologies [4,5]. In these technologies, hot flue gas is contacted with an alkali-based sorbent for the removal of SO₂ from the gas phase [6,7].

For the wet and semi-dry processes, the sorbent is a slurry, while the dry process utilises dry sorbent [5,8]. The wet FGD process produces a wet concentrated slurry product

that is dewatered and sold as gypsum, while the semi-dry and dry processes produce a dry product that is normally landfilled [9,10]. The main advantage of the wet FGD method is the high desulphurisation efficiencies (>90%) achievable with this technology; however, the handling of acidic slurries downstream generates waste water that requires comprehensive treatment [11]. The semi-dry FGD usually has lower efficiencies but uses about 60% less water than an equivalent wet FGD plant and no waste water is produced because the product exits the scrubber as a dry product [10,12]. It is the preferred method in regions where water scarcity is an issue. The semi-dry FGD method is commonly implemented in a spray dry scrubber (SDS), where the sorbent slurry is atomised into a spray of droplets for high heat and mass transfer rates. A circulating fluidised bed (CFB) may also be used; however, spray dry scrubbing (SDSCR) still constitutes a large percentage of the available semi-dry processes [9,12]. The focus of this study is on semi-dry FGD in an SDS—also referred to as SDSCR.

Spray dry scrubbing is a multiphase, multiphysics, and multiscale process that requires convoluted models. In recent years, because of the availability of increased computing capabilities, computational fluid dynamics (CFD) has been used to comprehensively model the convoluted details [3,4]. There is agreement that the desulphurisation efficiency in the semi-dry FGD process is a result of the competing processes of droplet drying and SO₂ absorption [13,14]. Therefore, the three key phenomena that must be described in modelling this process are the hydrodynamics, the evaporation or drying of the atomised droplets, and the resultant SO₂ absorption [4]. Current advances in FGD are focused on high-performance scrubbers able to achieve >98% removal efficiencies [9]. As, for the general SDSCR processes, achievable desulphurisation efficiencies hardly exceed 70% for a calcium-to-sulphur (Ca/S) ratio of 1–2, additional work is required to further enhance the technology's desirability [13,15]. Comprehensive modelling is, therefore, necessary to capture the detailed phenomena of the multiphase heat, mass, and momentum transfer occurring within the SDS, towards then improving process optimisation aimed at increasing SO₂-removal efficiency.

Research has shown that most of the CFD modelling of the FGD process has been conducted for the wet FGD process. Significant work has been conducted by Marocco and Inzoli [16] and, more recently, by Qu et al. [17]. The trend is changing, however, with complementary research on the semi-dry FGD, particularly in CFBs [18,19] and spouted beds [20,21], coming to the fore. The continued research on SDS [22–24], nonetheless, testifies to the importance of this technology in the abatement of SO₂ emissions from coal- and fossil-based power plants, which continue to be the dominant source of energy in both developed and developing economies. Katolicky and Jicha [15] used CFD to model SO₂ removal in a laboratory SDS that had been fitted with a two-fluid nozzle. They later extended their work to investigate SO₂ removal in an industrial-scale SDS that had been fitted with a rotary atomiser [25]. This work assumed the initial particle size distribution (PSD) of the atomised droplets while other droplet parameters were taken from previous studies. Further recent studies on modelling an industrial SDS include those of Liu et al. [24] and Mei et al. [26]. Liu et al. [24] investigated the SO₂-removal characteristics in an industrial FGD reactor. In their work, they investigated mono-sized droplets of the slurry. This approach holds the advantage of simplifying the analysis work, although it does not fully represent the real process where particles of various sizes are always produced by the injection nozzle. They based their model verification work on earlier studies carried out by Hill and Zank [14]. Mei et al. [26] used a rotary atomiser in their process. Similarly, a model simplification of the atomisation process was achieved through the use of single-sized particles. It is thus evident that further work should be carried out in an effort to achieve adequate representation and analysis of the SDSCR process.

There are generally two approaches that are used for modelling the drying kinetics in SDSCR. One approach treats the slurry droplets as pure water and thus focuses on evaporation being the medium for water removal, assuming no effect from either the dissolved or suspended solids [27,28]. The evaporation models in this category include

the perfect shrinkage model, the classical D^2 -law model, and further amendments of the classical D^2 -law model. The second category takes into account the different drying regimes, incorporating the effect of solids hindrance on the water-removal process [29]. These are also referred to as the hindered-drying models. They include the lumped-parameter models, the internal diffusion-based models, and mechanistic models. A detailed review of the different drying models applicable in spray drying and SDSCR was recently reported by the authors [4], giving further details on the derivation and applicability of the two modelling approaches presented above [4].

A major challenge associated with the second modelling approach is the complexity linked with its implementation and the computational cost. This has led to the first-mentioned modelling approach finding prominence in published CFD works, such as work by Liu et al. [24], Mei et al. [26], and Katolicky and Jicha [25]. Amongst the simplified evaporation models, the classical D^2 -law model has found greater usage in published works partly due to increased accuracy emanating from its further variation to incorporate the Stefan flux in the removal rate [30,31]. Of the hindered drying models, the diffusion-based models are the most complex—the lumped-parameter models require additional experimentation—leaving the simpler mechanistic models as the most viable option when considering the computational cost. In 2000, Hill and Zank [14] proposed a simple mechanistic model; however, it has not found wide usage in the SDSCR environment. Selecting the appropriate drying model capable of accurately describing the multiphase flow process in SDSs is crucial for a full understanding of the process. Nevertheless, in previously published studies on SDSs, some authors have selected a single model with the assumption that the chosen model remains valid for the particular case being investigated. Given that water evaporation is important in determining the desulphurisation efficiency, the present work does not assume a single model but rather investigates both the classical D^2 -law model and mechanistic model in an effort to more fully understand the impact of drying on SO_2 -removal efficiency.

Notwithstanding the foregoing discussion on the ongoing application of CFD to SDSCR processes, there remains a paucity of investigations into achieving a more comprehensive understanding of the complex processes in SDSCR—a problem wherein CFD modelling is better suited to provide answers, given the ever-increasing computation power and modelling capability that had presented a challenge to earlier researchers. A further concern associated with the modelling work carried out to date is the limited validation of the modelling results with real or experimentally measured process data. Our study now seeks to close these gaps—it now presents both modelling work and associated validation experiments. Furthermore, it develops a comprehensive model that considers the three key phenomena characterising the SDSCR process: process hydrodynamics, droplet drying and SO_2 absorption. Sensitivities are investigated in terms of the Ca/S ratio as well as the effect of varying the inlet liquid-to-gas (L/G) ratio on SO_2 -removal efficiency. The sorbent utilisation and final product moisture content are also analysed, alongside the realised SO_2 -removal efficiency.

2. Materials and Methods

The Buchi B-290 laboratory spray dryer (with a spray chamber height and diameter of 0.48 m and 0.154 m, respectively) was used to study the desulphurisation of flue gas using limeslurry (with the lime having an assay of 99 wt % calcium hydroxide and supplied by Kayla Africa Suppliers and Distributors, Johannesburg, South Africa). Artificial flue gas was generated by dosing SO_2 (99 vol % assay grade and supplied by Afrox, Vanderbijlpark, South Africa) in air (with known humidity and temperature), as per the set-up shown in Figure 1. The flue gas, supplied by an air blower having a rated capacity of $35 \text{ m}^3/\text{h}$, was introduced into a heated section of the column (fitted with a 2.3 kW electrical coil). Heating control was achieved using a PT100 fuzzy logic controller having a control accuracy of $\pm 2^\circ\text{C}$. The gas was then co-currently contacted with a spray of lime slurry droplets in the spray chamber or column. A two-fluid nozzle (0.7-mm nozzle tip and 1.5-mm cap size) was

used to effect the atomisation. Droplet drying and SO_2 absorption occur simultaneously in the column and the droplets that exit as dry particles are separated from the gas stream using a cyclone located downstream of the column.

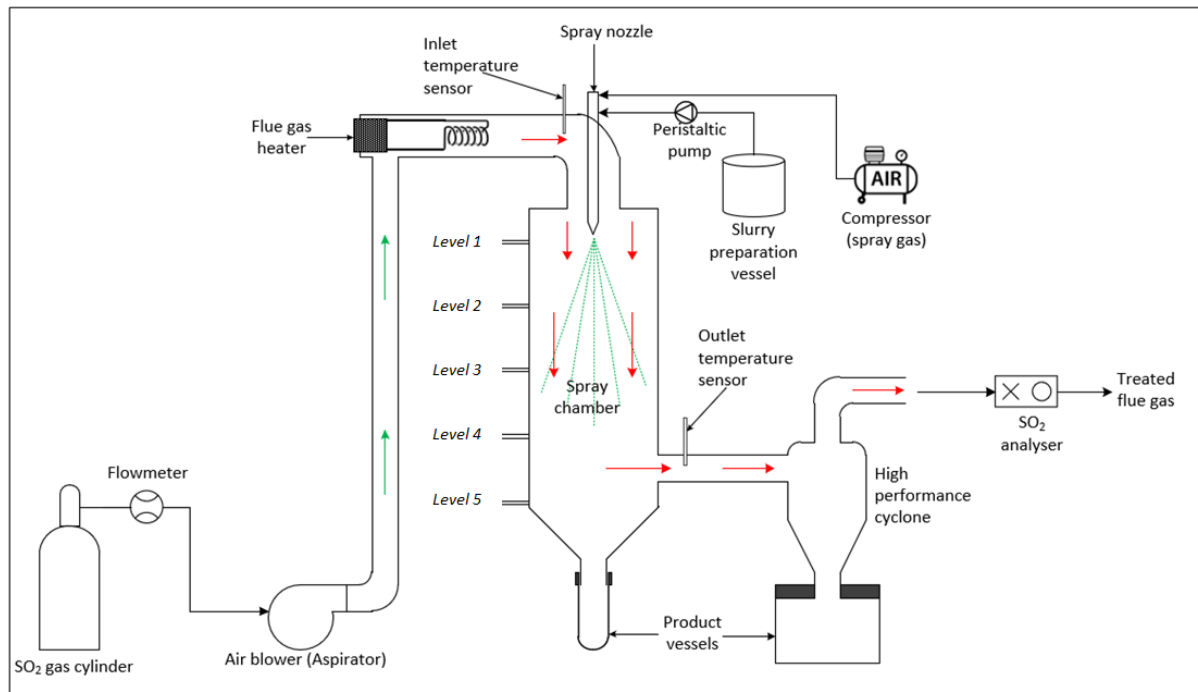


Figure 1. Experimental set-up used for spray dry scrubbing of SO_2 showing in green, cold streams and in red, hot streams (adapted, in part, with permission: B-290 Mini Spray Dryer operation manual [32]; copyright 2018 BÜCHI Labortechnik AG).

The column was modified to incorporate five sampling points at equal distances of 0.095 m axially along the column at the locations shown as Levels 1–5 in Figure 1. These were used for measurements of the desired parameters, including the temperature and SO_2 content of the gas phase inside the column. All values for temperature and inlet gas humidity were measured using a thermocouple and humidity probe (HygroPalm 23: HC2–IC102 probe supplied by Action Instruments South Africa (AISA) (Pty) Ltd., Johannesburg, South Africa), together with the in-built Buchi B-290 spray dryer thermocouples, the locations of which are shown in the figure. Within the column, the measurements were carried out using an amended micro-separator device as designed by Kievet [33]. An SO_2 analyser (Testo 340 analyser supplied by Testo South Africa (Pty) Ltd., Johannesburg, South Africa) was used to intermittently measure the concentration of the SO_2 . All runs were performed in triplicate after obtaining steady temperature and SO_2 concentration values. The mean values of the measured variables are then reported.

Further experiments were conducted. Selected parameters were varied to determine their effects on SO_2 -removal efficiency. These parameters included the inlet flue-gas temperature and the Ca/S ratio at an L/G ratio of 0.024. The ranges of the investigated parameters were selected based on operational parameters in industrial plants as well as previous experimental investigations on SDSCR [13,14]. Here, for the parameter ranges, the rated capacities of the laboratory equipment used were also taken into account. The associated SO_2 -removal efficiency and temperature measurements were conducted at the axial locations shown in Figure 1. Table 1 details the experimental conditions for the seven different case studies undertaken.

Table 1. Process conditions for the validation experiments conducted for an L/G (kg/kg) ratio of 0.024.

Process Variables	Case 1	Case 2	Case 3	Case 4	Case 5	Case 6	Case 7
T _{in} (°C)	108	130	142	108	130	142	142
Ca/S ratio (mol/mol)	1	1	1	1.5	1.5	1.5	2

3. Numerical Model

The SDSCR mechanism may be explained by three key phenomena occurring during the SO₂-removal process; these entail the modelling of hydrodynamics, droplet-drying modelling and SO₂-absorption modelling. This section addresses the numeric modelling and supporting theory for the development of the comprehensive models used in this study considering these three key phenomena.

3.1. Hydrodynamic Modelling

The interaction between gas and droplets was modelled using the Euler–Lagrangian framework where the gas was modelled as the continuous phase (Eulerian approach) with the spray being the dispersed phase (Lagrangian approach). The governing equations in the continuous phase, the equations describing mass, heat, and momentum conservation, are given as Equations (1)–(4), while Equations (10)–(13) give the dispersed-phase formulations.

3.1.1. Continuous Phase

The governing equations of mass, heat, and momentum conservation for the continuous phase are given as follows:

Mass conservation

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \bar{u}) = S_m \quad (1)$$

Species conservation

$$\frac{\partial (\rho w_A)}{\partial t} + \nabla \cdot (\rho w_A \bar{u}) = \nabla \cdot (\rho w_A D_{AB}) + S_{m_A} \quad (2)$$

Energy conservation

$$\frac{\partial (\rho E)}{\partial t} + \nabla \cdot (\rho \bar{u} E) = -P \nabla \cdot \bar{u} + \nabla \cdot (k_c \nabla T) + \rho \bar{u} \bar{g} + S_E \quad (3)$$

Momentum conservation

$$\frac{\partial (\rho \bar{u})}{\partial t} + \nabla \cdot (\rho \bar{u} \times \bar{u}) = -\nabla P + \nabla \cdot (\tau - \tau^R) + \rho \bar{g} + S_{mom} \quad (4)$$

where S is any additional source or sink term in the equations provided.

The shear stress transport (SST) k – ω model has been used successfully in spray drying operations and was used in order to capture the flow turbulence in this work [34,35]. When compared to other turbulence models, the SST k – ω model was selected for this work due to its success in previous studies in spray drying, including in the work of Langrish et al. [35], who studied a similar co-current spray drying operation as opposed to a counter-current contacting system. The SST k – ω model is also advantageous in that it blends the k – ε and k – ω models to best capture the free turbulence with the k – ε model and near-wall turbulence with the k – ω model [34,35]. The turbulence as described by the SST k – ω model is given in Equations (5)–(9), where the kinetic energy (k) is defined as

$$\frac{\partial (\rho k)}{\partial t} + \nabla \cdot (\rho k \bar{u}) = \nabla \cdot \left[\left(\mu + \frac{\mu_t}{\sigma_k} \right) \nabla k \right] + P_k - \rho \beta^* \omega k \quad (5)$$

The specific dissipation rate (ω) is given as

$$\frac{\partial(\rho\omega)}{\partial t} + \nabla \cdot (\rho\omega\bar{u}) = \nabla \cdot \left[\left(\mu + \frac{\mu_t}{\sigma_\omega} \right) \nabla \omega \right] + \alpha P_\omega - \rho\beta\omega^2 + 2\rho(1 - F_B) \frac{1}{\sigma_{\omega,2}\omega} \nabla k \nabla \omega \quad (6)$$

Here, the eddy viscosity (μ_t) is given as

$$\mu_t = \rho \frac{k}{\omega} \min \left(\alpha^*, \frac{a_1 \omega}{\zeta F_E} \right) \quad (7)$$

and the production of the turbulent kinetic energy (P_k) and the dissipation rate (P_ω) described as

$$P_k = \min \left(\mu_t \zeta^2, 10\rho\beta^* k\omega \right) \quad (8)$$

$$P_\omega = \frac{P_k}{\min \left(\frac{k}{\omega}, \frac{a_1 \omega}{\zeta F_E} \right)} \quad (9)$$

F_E and F_B are blending wall functions, β^* and β are turbulence-model constants, and α^* and a_1 are flow-transition constants. σ_k and σ_ω are the turbulent Prandtl numbers for the turbulent kinetic energy and dissipation rate, respectively, and ζ is the invariant strain rate.

3.1.2. Dispersed Phase

The corresponding governing equations for the dispersed phases are given as follows for the k th droplet/particle:

Trajectory

$$\frac{d\bar{x}}{dt} = \bar{v} \quad (10)$$

Mass

$$\frac{dm_d}{dt} = S_{k,m} \quad (11)$$

Momentum

$$m_d \frac{d\bar{v}}{dt} = S_{k,mom} + m_d \bar{g} \quad (12)$$

Energy

$$m_d C_p \frac{dT_d}{dt} = S_{k,E} \quad (13)$$

In this study, the dispersed-phase source terms above were deduced from the equations given below.

The momentum balance source was computed using Equations (14)–(16), where the drag force and gravitational contribution are shown, and any additional forces such as the pressure gradient force, lift force, and added mass force are included as F

$$\frac{d\bar{v}}{dt} = C_D \frac{18\mu}{\rho_p d} \frac{Re_p}{24} (\bar{u} - \bar{v}) + g \left(\frac{\rho_p - \rho}{\rho_p} \right) + F \quad (14)$$

The Schiller–Neumann correlation was used to describe the drag force

$$C_D = \frac{24 \left(1 + 0.15 Re_p^{0.687} \right)}{Re_p} \dots \text{for } Re_p \leq 1000 \quad (15)$$

$$Re_p \equiv \frac{\rho |\bar{u} - \bar{v}| d_d}{\mu} \quad (16)$$

The energy source contribution was computed through the use of Equations (17) and (18), as follows:

$$m_d C_p \frac{dT_d}{dt} = h A_d (T - T_d) + h_{vap} \dot{m}_d \quad (17)$$

where the Ranz and Marshall's Nusselt number (Nu) correlation was used to compute the transfer coefficient, h .

$$Nu = 2 + 0.6 Re_p^{0.5} Pr^{0.33} \quad (18)$$

The species mass transfer involving the water-removal rate as well as that involving SO_2 absorption are given in detail in Sections 3.2 and 3.3. All the source terms were evaluated by volume averaging of the individual dispersed-phase parcel source term contributions within a cell volume.

3.2. Droplet Drying Modelling

The use of the classical D2-law model and the mechanistic model was investigated. The former simplifies the drying phenomenon by disregarding the influence of solids on the water-removal rate. The latter takes into account the hindrance of solids on the water-removal rate [4].

The classical D2-law model is given by Equations (19)–(21) as follows:

$$\frac{dm_d}{dt} = -\rho k_{H_2O, g} A_d \ln(1 + B_M) \quad (19)$$

here B_M is the Spalding transfer number equating to

$$B_M = \frac{w_{H_2O, \infty} - w_{H_2O, s}}{1 - w_{H_2O, \infty}} \quad (20)$$

and $k_{H_2O, g}$ is the gas-phase mass transfer coefficient. It can be determined from the Ranz and Marshall Sherwood number (Sh) correlation as follows:

$$Sh = 2 + 0.6 Re_p^{0.5} Sc^{0.33} \quad (21)$$

The mechanistic model is formulated from Equations (21)–(24) as follows:

$$\frac{dm_{H_2O}}{dt} = \bar{\rho} K'_{H_2O, g} A_d \ln \left[\frac{1 - y_{H_2O, \infty}}{1 - y_{H_2O, s}} \right] \quad (22)$$

where

$$\frac{1}{K'_{H_2O, g}} = \frac{1}{k_{H_2O, g}} + \frac{1}{\left(\frac{D_{H_2O, g} S_{d_i, d_{ag}}}{A_{ag}} \right)} \quad (23)$$

Here, the mass transfer coefficient $K'_{H_2O, g}$ includes an additional resistance to the mass transfer rate—it is implemented only when the falling rate ensues. The A_{ag} is the fixed droplet area post the critical moisture content (it is also referred to as the agglomerate area) and $S_{d_i, d_{ag}}$ is defined as follows:

$$S_{d_i, d_{ag}} = \frac{2\pi}{\frac{1}{d_i} - \frac{1}{d_{ag}}} \quad (24)$$

Here, d_i is the diameter of the liquid interface which recedes inside the wet particle and d_{ag} the diameter of the agglomerate. This process is addressed further in the next section (Section 3.3) when explaining the simultaneous process of water and SO_2 migration.

3.3. SO₂ Absorption Modelling

It is widely accepted that the two-film theory describes the gas–liquid reactions that take place during the absorption of SO₂ into the liquid slurry phase in SDSCR. Figure 2 shows an extension of the two-film theory to include a third phase, in this case, the solid sorbent phase, describing the semi-dry FGD process, where the two-film theory is followed by an unreacted shrinking core model (as presented by Hill and Zank in 2000 [14]).

According to the two-film theory, the rate of SO₂ absorption from the flue gas is proportional to the driving force multiplied by the mass transfer coefficient ($k_{SO_2,g}$) and the interfacial mass transfer area (a):

$$\dot{n}_{SO_2,g} = -k_{SO_2,g}a(p_{SO_2,\infty} - p_{SO_2,i}) \quad (25)$$

On the other hand, the rate of transfer of SO₂ across the liquid film is directly proportional to the difference between the SO₂ concentration in the gas–liquid interface and the bulk liquid:

$$\dot{n}_{SO_2,l} = -k_{SO_2,L}a(c_{SO_2,i} - c_{SO_2,l}) \quad (26)$$

where $k_{SO_2,L}$ is the liquid-side mass transfer coefficient.

One of the assumptions of the two-film theory is that the quantities of the respective species contained in the films are insignificant compared to those passing through. This is due to the films being very thin. As a result, the species that diffuses through the gas film can be assumed to also diffuse through the liquid film. The mass transfer resistances are thus considered to be in series, and the overall transfer rate can be given by

$$\dot{n}_{SO_2} = -K_{SO_2,g}a(p_{SO_2,\infty} - p_{SO_2,i}^*) = K_{SO_2,L}a(c_{SO_2,i}^* - c_{SO_2,l}) \quad (27)$$

where $K_{SO_2,g}$ and $K_{SO_2,L}$ are the overall mass transfer coefficients. $p_{SO_2,i}^*$ and $c_{SO_2,i}^*$ are the equivalent partial pressure and concentration of the bulk gas and bulk liquid, respectively.

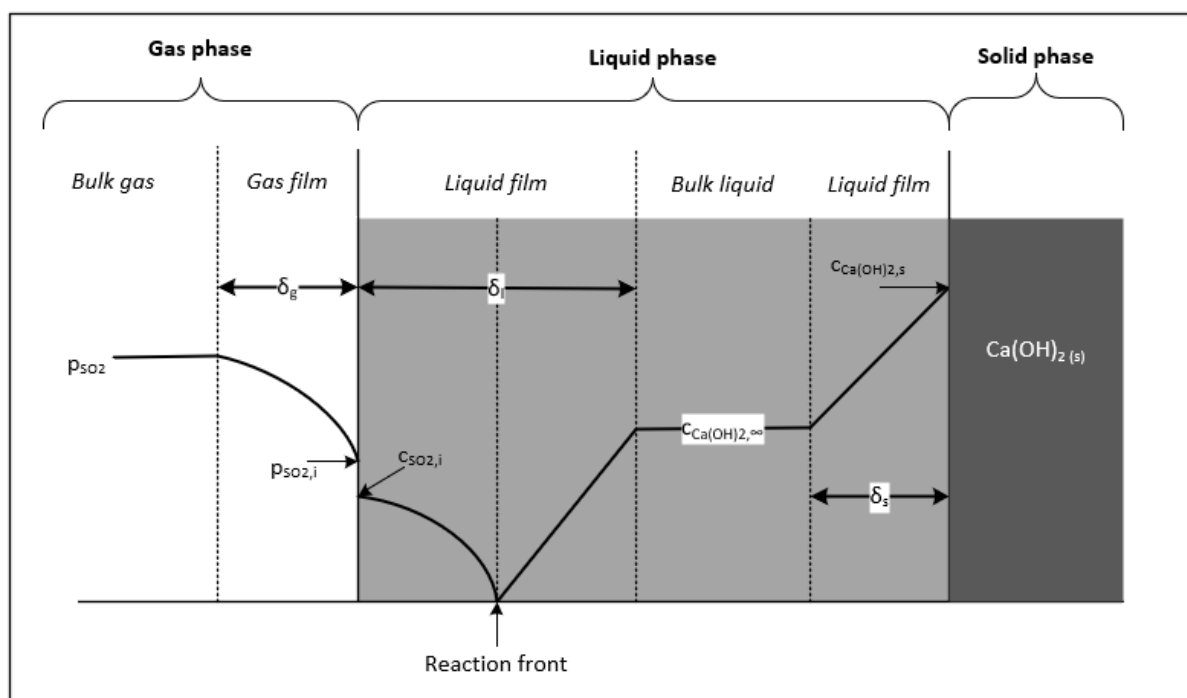


Figure 2. Extended two-film theory representing the absorption of SO₂ in an SDS where i is at the gas–liquid interface and s is at the solid–liquid interface (adapted from Partridge et al. [36]).

The two-film theory as presented above assumes that the dissolution of the hydrated lime sorbent within the droplet is infinite, with the implication that there is always a sufficient supply of calcium ions for an instantaneous reaction with the dissolved sulphurous species. Consequently, a spherical reaction front around the droplet ensues, where there is also precipitation of the hemihydrate with an unreacted core, the size of which decreases as the hemihydrate layer increases during the reaction process. This takes place concurrently with droplet shrinkage as a result of evaporation. The shrinkage is taken to be homogeneous in nature and it continues until the critical moisture content is realised, at which point the droplet size remains fixed (fixed particle agglomerate diameter) and the interface then recedes into the wet particle (Figure 3).

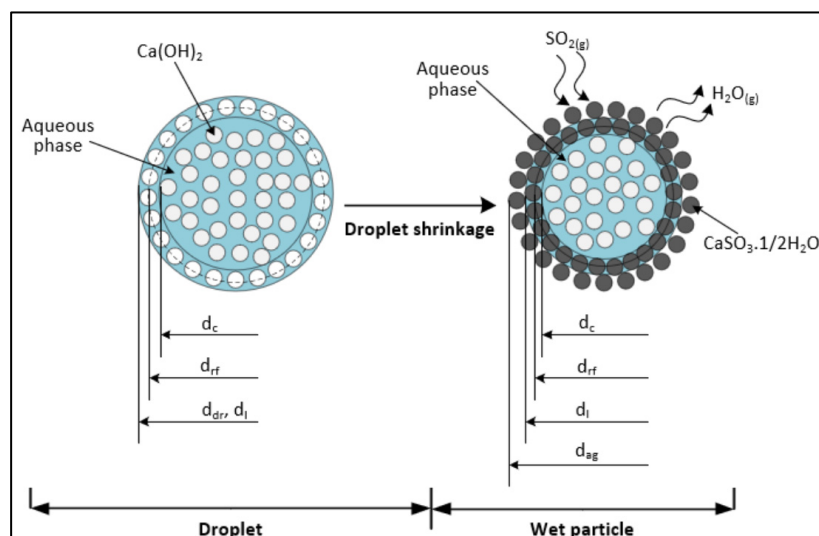
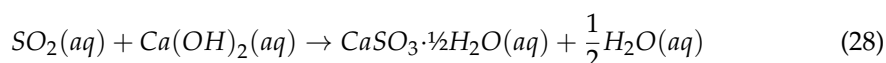


Figure 3. Depiction of the simultaneous droplet drying and SO₂ absorption with chemical reaction, where d_c is the diameter of the core, d_{rf} is the diameter of the reaction front, d_{dr} is the droplet diameter and d_{ag} is the agglomerate diameter (adapted from Hill and Zank [14]).

In terms of the desulphurisation process, the liquid-phase reaction of the dissolved SO_2 with dissolved $\text{Ca}(\text{OH})_2$ is generally presented as follows (Equation (28)):

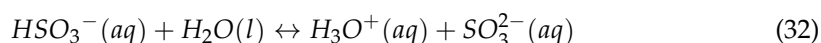
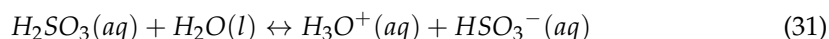
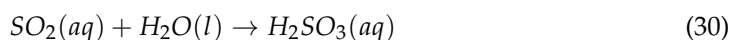


Calcium sulphite hemihydrate is the main product, although, in practice, there are other side reactions that result in the formation of CaSO_4 and $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$. The foregoing reaction as well as the two-film theory presented above show that the reaction progresses in the liquid phase, thus highlighting the importance of the drying rate or the water removal process in determining the efficiency of the SO_2 removal process.

In practice, the above general reaction involves a series of steps as outlined below (Equations (29)–(36)). The reaction sequence commences with the gas-phase diffusion of SO_2 from the bulk gas phase to the droplet surface. This is followed by the absorption of gaseous SO_2 into the aqueous phase



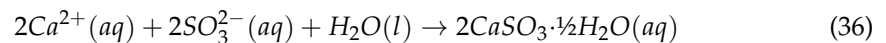
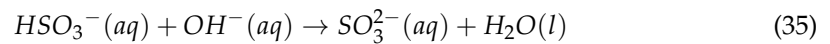
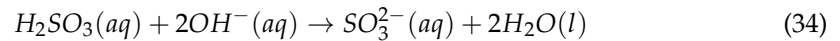
The next step is the formation of the sulphurous acid and its ionisation via the following:



The dissociation of calcium hydroxide then follows



The next step is the liquid-phase diffusion of the sulphurous ions to the reaction front, followed by the liquid-phase diffusion of the calcium ions to the reaction front and then the resultant reaction at the reaction front.



3.4. Effect of pH on the Reaction Chemistry

Sulphur dioxide absorption involves the movement of SO_2 , by diffusion, from the bulk gas phase into the slurry in which chemical reactions (discussed in the previous section) take place. Several authors have shown that different absorption regimes can be defined based on the rate of reaction of SO_2 in the liquid phase [37,38]. Table 2 shows the regimes as adapted from Mchabe et al. [37]. Three regimes are presented, giving different characteristics of SO_2 absorption. These are dependent on the pH of the solution in which the SO_2 reactions take place. Regime I, which is characterised by neutral pH to highly alkaline conditions, is associated with instantaneous dissociation of SO_2 in the liquid phase accompanied by immediate reaction with the alkaline sorbent, thus the mass transfer of SO_2 is controlled by the gas-phase resistance. In Regime II, which refers to the medium acidic pH range, both the gas-phase and liquid-phase mass transfer resistances are applicable, with the absorbed SO_2 undergoing a fast reaction but it is still able to accumulate in the liquid phase. Regime III is characterised by high acidic conditions where there is limited availability of the alkaline sorbent in solution, resulting in limited or no aqueous SO_2 dissociation due to the high concentration of H^{+} ions. In this case, the gas-phase resistance to mass transfer is insignificant. The liquid-phase mass transfer resistance is the controlling mechanism.

Table 2. SO_2 absorption regimes (adapted from Mchabe et al. [37]).

Regime	I	II	III
pH	$\text{pH} \geq 6.83$ at 323.15 K	$6.83 > \text{pH} > 3$ at 323.15 K	$\text{pH} \leq 6.83$ at 323.15 K
Gas-side resistance	Highly significant	Significant	Insignificant
Liquid-side resistance	Insignificant	Significant	Highly significant
Dissociation of aqueous SO_2	Highly favoured	Favoured	Not favoured
HSO_3^{-} concentration	Insignificant	Significant	Highly significant
SO_3^{2-} concentration	Highly significant	Significant	Insignificant

In this work, hydrated lime was used as the sorbent of choice; it gave highly alkaline solutions of calcium hydroxide when mixed with water to form the sorbent slurry. The minimum pH of the alkaline slurry was 12 (as measured in the laboratory), thus corresponding to Regime I (described earlier). The SO_2 absorption was, therefore, characterised by the instantaneous reaction in the liquid phase with the resistance to mass transfer being in the gas phase. Consequentially, in the subsequent analysis, only the gas-phase resistance was used in determining the SO_2 mass transfer rate during the absorption process. In this case, the molar transfer rate of SO_2 may be given by Equations (37) and (38)

$$\dot{n}_{\text{SO}_2} = -\bar{p}k_{\text{SO}_2, g}A_d(y_{\text{SO}_2, \infty} - y_{\text{SO}_2, i}) \quad (37)$$

where k_{g, SO_2} can be determined from the Sherwood number for the classical $D2$ -law model

$$Sh = \frac{k_{SO_2, g} d_d}{D_{SO_2, g}} = 2 + 0.6 Re_p^{0.5} Sc^{0.33} \quad (38)$$

For the mechanistic model, the transfer coefficient is amended to consider the hindrance of the solids on the transfer process, giving Equations (39) and (40)

$$\dot{n}_{SO_2} = -\bar{\rho} K'_{SO_2, g} A_d (y_{SO_2, \infty} - y_{SO_2, i}) \quad (39)$$

where

$$\frac{1}{K'_{SO_2, g}} = \frac{1}{k_{SO_2, g}} + \frac{1}{\left(\frac{D_{SO_2, g} S_{d_i, d_{ag}}}{A_{ag}} \right)} \quad (40)$$

4. Numerical Solution Approach

The numerical solution was implemented in STARCCM+[®] (v.16.02.008) where 2,468,780 volumetrically controlled 3D polyhedral mesh cells with eight prism layers were generated. A multicomponent ideal and compressible gas was used to model the artificial flue gas, wherein SO_2 was introduced as a passive scalar in the continuous phase and another passive scalar introduced in the dispersed phase. The model made use of the interaction of these passive scalars to allow for the passive scalar transfer from the Eulerian to the Lagrangian phase. Thereafter, the desulphurisation or SO_2 -removal efficiency (η) of the process was computed as per Equation (41).

$$\eta = \left(\frac{\dot{n}_{SO_2, inlet} - \dot{n}_{SO_2, i}}{\dot{n}_{SO_2, inlet}} \right) \quad (41)$$

For the investigation of select cases (Table 1: Cases 3, 6 and 7), where the impact of the Ca/S ratio on the desulphurisation efficiency was investigated, the calcium hydroxide utilisation (ζ) was computed from Equation (42) as follows:

$$\zeta = \left(\frac{\dot{n}_{Ca(OH)_2, inlet} - \dot{n}_{Ca(OH)_2, i}}{\dot{n}_{Ca(OH)_2, inlet}} \right) = \frac{\eta}{\lambda} \quad (42)$$

A thermal conductivity of 0.74 W/mK for the scrubber's borosilicate glass (as reported by Hanus and Langrish [39]) and a heat transfer coefficient of 5 W/m²K for natural convection [40] were used to account for the heat loss to the environment. The droplet PSD was modelled by measuring the exit PSD and using the back-calculation method as described by Woo et al. [41], through the Rosin–Rammler description, to determine the mean droplet size and spread parameter. The cone shape and angle were determined from the work of Poozesh et al. [42]. A species mass source on a select volume of cells around the nozzle area was implemented to model the atomisation gas. Together with the described drag force acting on the droplets, the virtual mass and shear lift forces were also included. The Rohsenow correlation was also implemented to account for the stuck droplet heat and mass transfer. Submodels of the classical $D2$ -law and the mechanistic model were implemented using user code. The appended cyclone was also included as a submodel in order to account for the associated pressure drop across the equipment. The model simulated the case investigations given in Table 1; it was also used to predict additional cases to investigate further sensitivities, such as the effect of the L/G ratio on the SO_2 removal rate or desulphurisation efficiency. Further details of the model boundary conditions used in the study are given in Table 3.

Table 3. Initial and boundary conditions used for the developed CFD models.

Parameter	Units	Value
Continuous phase—Flue gas		
Inlet boundary—velocity inlet		
Inlet temperature	°C	108; 130; 142
Inlet L/G ratio	-	0.014; 0.024; 0.034; 0.049
Inlet Ca/S ratio	-	1; 1.5; 2
Outlet boundary—pressure outlet		Exit gas
Pressure (cyclone exit)	kPa	84.6
Wall		
Convective heat transfer coefficient	W/m ² ·K	5
Thermal conductivity	W/m·K	0.74
Free stream temperature	°C	20
Dispersed phase—Lime slurry		
Inlet boundary—velocity inlet		Atomisation air
Mass flow rate	kg/h	0.473
Dispersed phase—Injection properties		
Mass flow rate	kg/h	0.9
Temperature	°C	20
Parcel number	-	80
Mean diameter	µm	10.5

An unsteady state solution of the single-phase flow of the gas was solved first until convergence and then the slurry droplets were introduced into the domain. Convergence was assessed by monitoring the residual values, the element count within the domain, the exit temperature, and the exit SO₂ mass fraction. Mesh independence studies as well as parcel independence studies were conducted to ascertain that the model solution was independent of these input variables. Three mesh sizes were investigated as part of the conventional grid independence test for this work. These are a course-sized grid of 537,429 cells corresponding to a 0.02 mm cell base size, a medium-sized grid of 2,468,780 cells corresponding to a 0.01 mm cell base size, and a fine grid of 3,178,420 cells corresponding to a 0.005 mm cell base size. The CFD results for selected key process parameters (the SO₂-removal efficiency and the temperature sampled axially along the column length) were used to determine the optimum grid. The results are shown in Figure 4.

From the results in Figure 4, there is no significant change in both the predicted SO₂-removal efficiency and temperature moving from the medium grid to the finer grid and, therefore, the medium-grid resolution was selected as the base for this study, noting the computational complexity that comes with running at the finer mesh size. Furthermore, the model accuracy and optimal grid resolution were judged through comparing the root mean square error (RMSE—defined in Equation (43)) obtained by comparing the measured values to the model-predicted results for the two key parameters. The values obtained for the RMSE for the SO₂-removal efficiency and temperature were 5.7 and 4.1, respectively, for the medium grid, and 4.9 and 3.2, respectively, for the fine grid, thus showing good fit with measured data.

The pressure-velocity coupling was solved using the segregated flow solver. The AMG linear solver with Gauss–Seidel relaxation scheme was used for the segregated flow, species and energy solver with a first-order upwind convection scheme.

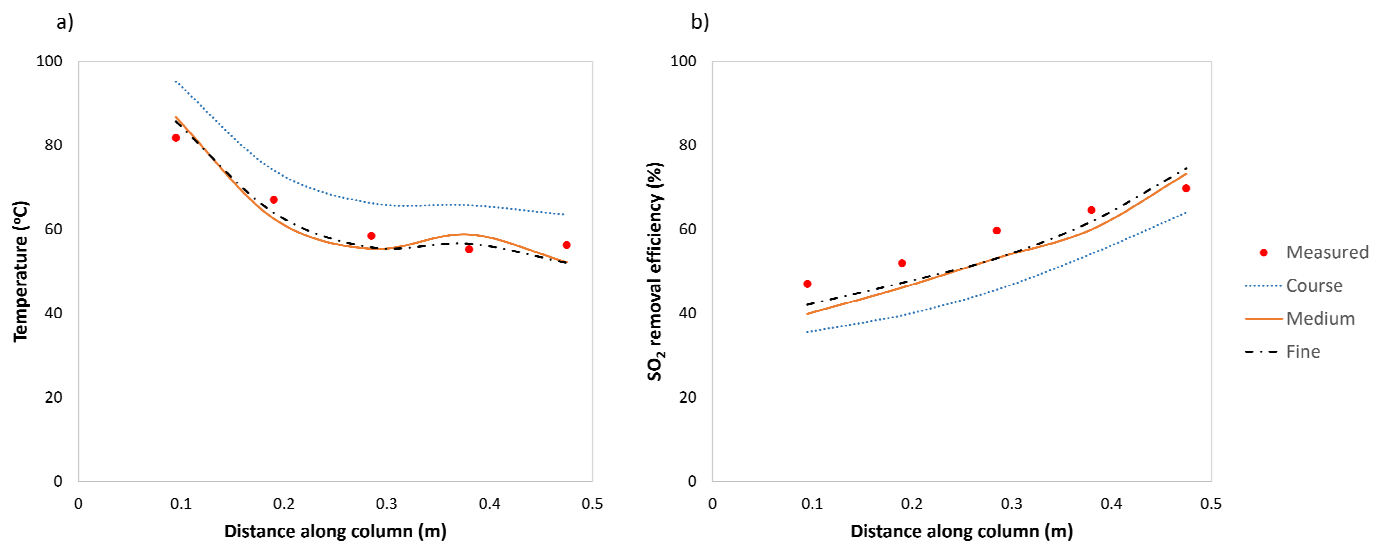


Figure 4. Grid independence analysis showing the variation of the key process parameters along the column for different grid sizes: (a) gas temperature and (b) SO₂ removal efficiency.

5. Results and Discussion

The results of our investigations into the desulphurisation are now presented. First, model-generated profiles of variables are presented, giving insight into the nature and characteristics of the convoluted process dynamics inside the SDS. Second, a comparison between the two drying models (the classical *D*-2 law and the mechanistic) for their predictive capability of the SO₂-removal efficiency, using the root mean square error (RMSE) to judge the goodness-of-fit (Equation (43)), is presented. The percentage error (Equation (44)) is also used to compare some model results to the experimental data as well as the coefficient of determination, R^2 (Equation (45)). The section then concludes with a presentation of the effects of some of the key variables in the SDSCR process, including the Ca/S ratio and L/G ratio, on desulphurisation efficiency, product moisture content, and sorbent utilisation.

$$RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^n (y_i - \hat{y}_i)^2} \quad (43)$$

$$Absolute\ error = |y_i - \hat{y}_i| \quad (44)$$

$$R^2 = 1 - \frac{\sum_{i=1}^n (y_i - \hat{y}_i)^2}{\sum_{i=1}^n (\hat{y}_i - \bar{y}_i)^2} \quad (45)$$

where n is the number of observations, $\hat{y}_i$ is the observed values, and y_i is the model-predicted values, and $\bar{y}_i$ is the average of the observed values.

5.1. SO₂ Removal and Associated Variables

Figure 5 gives the results from the CFD model developed for the SO₂ removal process. Also shown are measurement locations for the evaluation of the removal efficiency (η) and temperature measured (T) at the sampling locations shown in Figure 1. The profiles shown are only for the Case 2 setting, where the following applied: inlet temperature of the flue gas was 130 °C, a Ca/S ratio of 1, and an L/G ratio of 0.024. Furthermore, the mechanistic model was used to describe the drying kinetics and also capture the general trends seen for the other cases. Figure 5 shows that SO₂ efficiency increases down the column, as is evident from the radial and axial profiles shown in Figure 5a–c. Such behaviour is expected as the injected droplets interact with the SO₂-containing gas, where the SO₂ is progressively absorbed while the droplets traverse the column. A closer look at the radial profile of SO₂ efficiency in Figure 5a, for each sectional plane, reveals a central region having low

efficiency, with the efficiency increasing towards the walls of the column. This region of low efficiency is associated with a high-velocity central jet stream (see Figure 5f), which decreases down the length of the column. This decrease is a result of better mixing and recirculation that is present in the lower section of the column. The decrease in the SO_2 content of the gas is accompanied by an increase in the gas humidity and a decrease in the gas temperature, as a result of the simultaneous droplet drying process. The gas profiles also indicate that there is a bias in the overall velocity profile, with recirculation towards one side of the column in the direction opposite to the location of the sampling points. This region of high recirculation is characterised by relatively high humidity and low temperatures (see Figure 5d,e), conditions which are favourable for high SO_2 -removal efficiency.

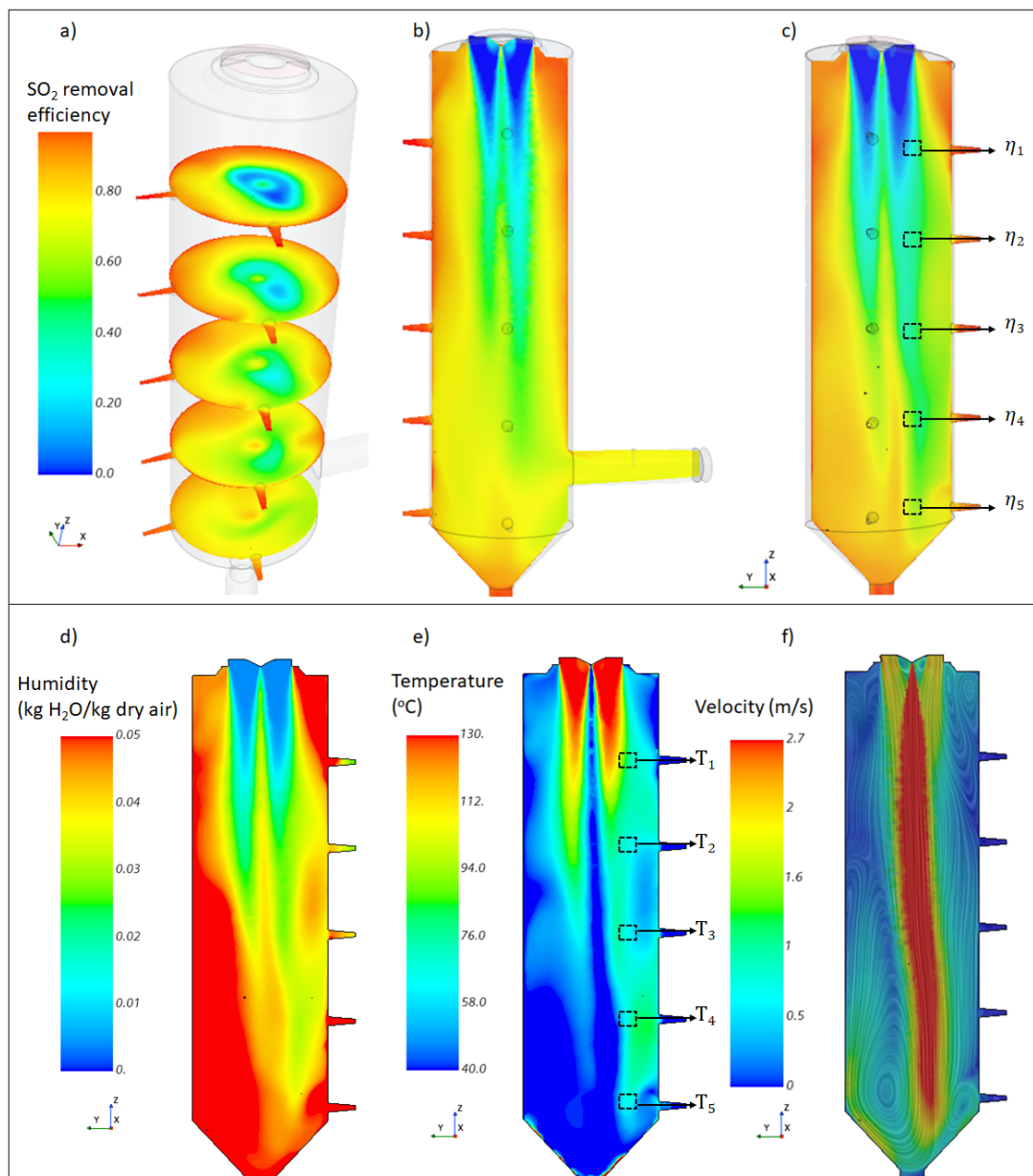


Figure 5. Profiles of the model predicted: (a) radial and axial SO_2 -removal efficiency in the (b) x–z plane and in the (c) y–z plane within the column, and associated y–z profiles of the (d) humidity, (e) temperature and (f) velocity within the column, after 30 s solution time at an inlet temperature of 130 °C, a Ca/S ratio of 1, and an L/G ratio of 0.024.

Table 4 compares the actual measured values of the SO₂-removal efficiency and flue-gas temperature to the model-predicted values down the column and at the sampling points as shown in Figure 5. The information in the table is in line with the profiles shown in the figure—there is an increase in SO₂-removal efficiency down the column, accompanied by a general decrease in the gas temperature. The RMSE is also used to indicate the goodness-of-fit values for the model SO₂-removal efficiency and gas temperature: values were 5.7 and 4.1, respectively. A point to note from the data in the table is that there are higher absolute errors towards the top of the column compared to the bottom of the column. This is due to the initial mixing of fresh, cooler, unreacted slurry (together with atomisation air) with the hotter inlet flue gas, as well as recirculating streams. This commences at the top section of the column, continuing in the axial direction, and thus leading to a large variation in the measured properties, which decreases with further mixing.

Table 4. Comparison of the model-predicted and experimentally determined axial SO₂-removal efficiency and flue-gas temperature for the sample points shown in Figures 1 and 5, determined at an inlet temperature of 130 °C, a Ca/S ratio of 1, and an L/G ratio of 0.024.

Measured Variable	Axial Distance from the Top of the Column					RMSE
	Level 1	Level 2	Level 3	Level 4	Level 5	
Model SO ₂ -removal efficiency	40.0	46.2	53.2	60.0	73.2	5.7
Measured SO ₂ -removal efficiency	47.1	52.0	59.8	64.6	69.8	
Absolute error (Efficiency)	7.14	5.82	6.63	4.83	3.35	
Model T (°C)	86.7	62.6	55.4	58.8	52.2	4.1
Measured T (°C)	81.8	67.1	58.5	55.3	56.3	
Absolute error (Temperature)	4.9	4.5	3.0	3.5	4.1	

Figure 6 shows the corresponding droplet profiles inside the column. The concentration of absorbed SO₂ is shown in the figure. It should be noted here that, in reality, some of the absorbed SO₂ would dissociate further and react as shown in Equations (29)–(36). Therefore, the reported SO₂ concentration is the overall absorbed SO₂ for the associated droplet or particle residence time. The profiles indicate an increase in the droplet absorbed SO₂ from the point of atomisation towards the bottom of the column. The profiles also show that high SO₂ capture is associated with high particle moisture content and low particle residence time. This is because reactions between the SO₂ and sorbent take place in the presence of water. The droplet profiles also show high particle residence times at the top of the column, indicating the presence of recirculation in the column. This region is characterised by very low mass fractions of particle water and, in turn, low SO₂ content of particles. The expectation is that particles that have resided for longer periods inside the column would have higher SO₂ content. However, this is not the case, as is evident from the particle profiles. This is likely a result of the recirculation of particles of smaller diameter, which dry quickly and are entrained in the recirculating flow. Subsequently, prior to drying, these particles do not absorb SO₂ significantly.

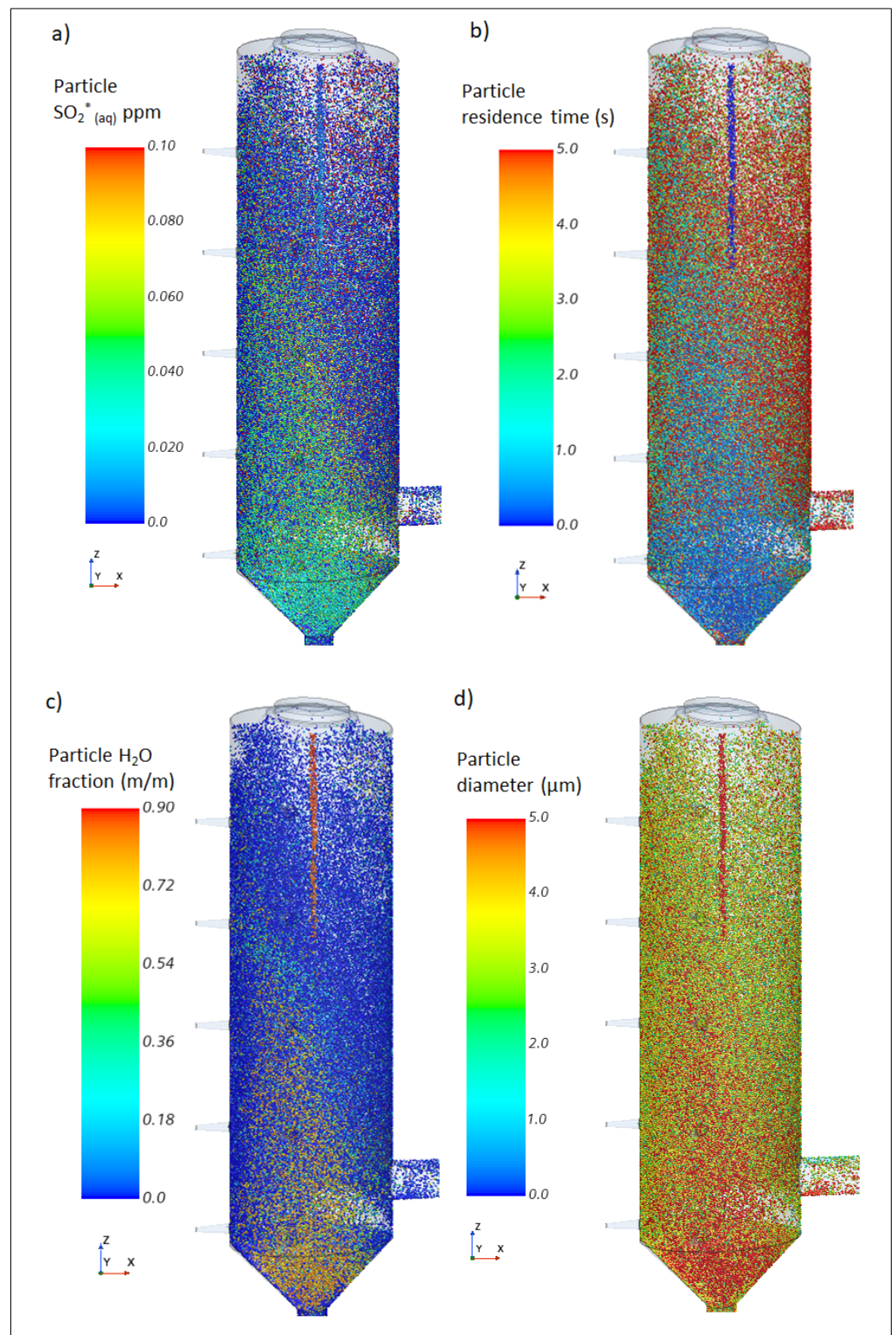


Figure 6. Axial cross-sectional snapshot of droplets along the x–z plane after 30 s solution time, for an inlet temperature of 130 °C, a Ca/S ratio of 1, and an L/G ratio of 0.024: (a) the absorbed SO₂ mass fraction [ppm] of the droplets with the asterisk (*) representing the overall absorbed SO₂ in the droplet, (b) the droplet residence time [s], (c) the mass fraction of water of the droplets and (d) the droplet or particle diameter [μm].

5.2. Validation of the Drying Model

Validation of the two investigated drying models, the mechanistic model and the classical D^2 -law model, is now addressed. Figure 7 shows the variation profiles of SO_2 -removal efficiency within the column for the two model cases at an inlet temperature of 130°C , a Ca/S ratio of 1, and an L/G ratio of 0.024.

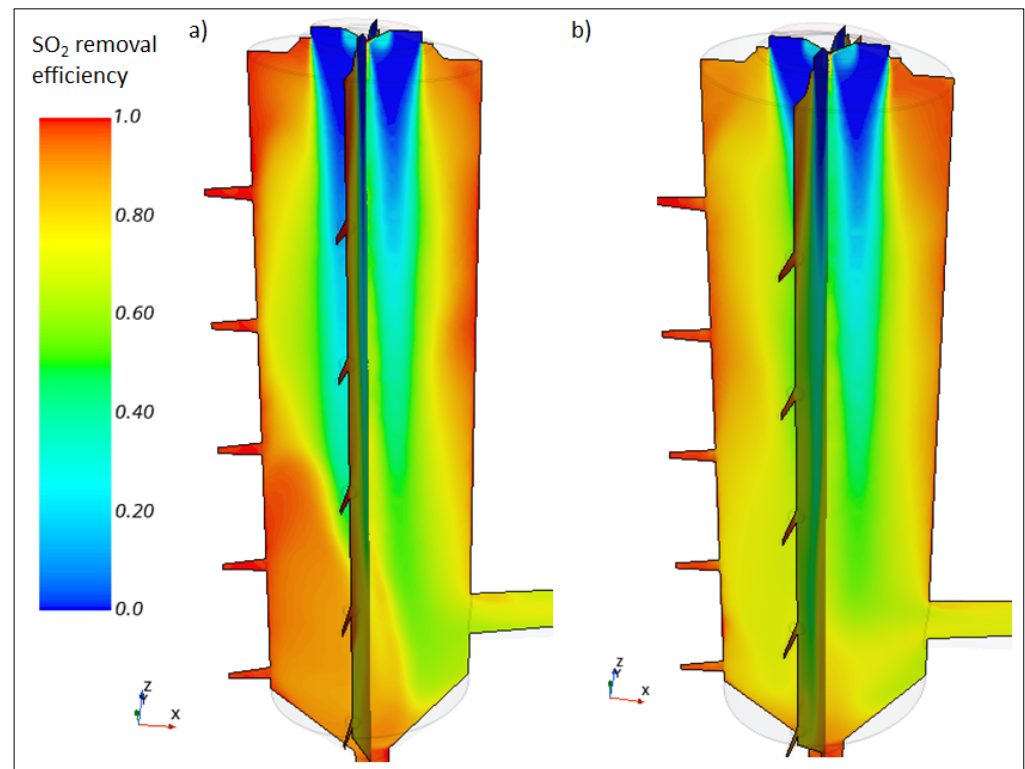


Figure 7. Axial profiles of the SO_2 -removal efficiency within the column showing both the x–z plane and y–z plane for (a) the classical D^2 -law model and (b) the mechanistic model at an inlet temperature of 130°C , a Ca/S ratio of 1, and an L/G ratio of 0.024.

It is evident from Figure 7 that higher SO_2 -removal efficiency is predicted for the classical D^2 -law model than for the mechanistic model, as shown from the plane profiles of the column. This rather unexpected behaviour, given the rigour in the development of the mechanistic model—hence it is expected to give higher efficiencies—is explained in more detail below, where a comparison of both the model predictions with experimental data is presented. Both the classical D^2 -law and mechanistic models show a marked increase in the SO_2 -removal efficiency at the top half of the column where the droplets first interact with SO_2 -laden flue gas, in a region characterised by a high concentration gradient for mass transfer between the continuous phase and the dispersed phase. This behaviour is limited in the lower sections of the column where the droplets already contain appreciable amounts of absorbed SO_2 . Overall, both models show a general increase in SO_2 -removal efficiency down the column. This is in line with the experimental data, which shows a similar behaviour, as well as previous work reported by Hill and Zank [14] and Katolicky and Jicha [15]. Figure 8 shows a comparison of the model-predicted SO_2 -removal efficiency for the classical D^2 -law and mechanistic models with the values measured as part of this work.

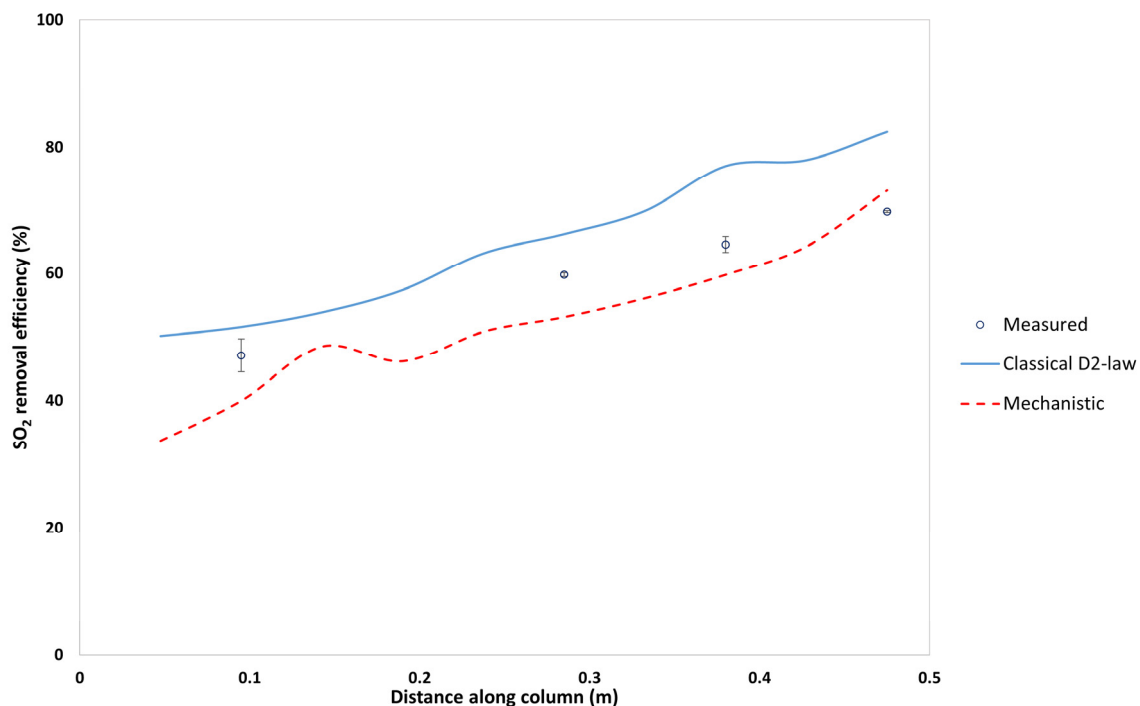


Figure 8. Axial SO₂-removal efficiency: comparison of the classical D2-law and mechanistic models prediction, with experimental data (inlet temperature 130 °C, Ca/S ratio 1, L/G ratio 0.024).

Figure 8 shows that the classical D2-law model over-predicts SO₂ efficiency compared to the measured data. This is attributed to the inability of the model to account for the solid's hindrance on the SO₂ mass transfer during the drying process—as the droplet dries beyond the critical moisture content, the liquid interface recedes inside the droplet, providing increased resistance to the mass transfer of not only the water from the liquid surface but also to the SO₂ transfer into the liquid surface. The classical D2-law model does not take this phenomenon into consideration because the assumption has always been that there is insignificant absorption during this latter part of the drying progression [13,43] and thus its impact on the absorption rate has not been much explored. On the other hand, the mechanistic model slightly under-predicts the removal efficiency. This can be explained by the fact that the model, in its formulation, does not consider the local droplet moisture gradients during the evaporation process, as explained earlier (Introduction section) and in an earlier review of the drying models by the authors [4]. The model nonetheless still captures the receding water front inside the droplet, but the drying rate over-prediction, when compared to the actual rate, leads to an under-prediction in observed SO₂-removal efficiency. The resultant reduction in SO₂-removal efficiency is due to SO₂ absorption reaction dependence on the water availability in the droplet as explained previously. The RMSE was calculated to evaluate the goodness-of-fit. The values obtained for the classical D2-law model and the mechanistic model were 9.7 and 5.7, respectively. Thus, the mechanistic model best predicted the SO₂ removal rate. Consequentially, in later work on looking into sensitivity analyses, only the mechanistic model results will be discussed.

5.3. Sensitivity Analysis

The SO₂-removal efficiency was investigated further in terms of the effect of inlet flue-gas temperature at various Ca/S and L/G ratios. The sorbent utilisation was also calculated for the different Ca/S ratios. A comparison of model-predicted values with experimental values was also undertaken. Results revealed that the RMSE of the desulphurisation efficiency ranged between 2.6 and 6.0, and the R² ranged between 0.77 and 0.95 as shown in Table 5 (the details of the various cases are given in Table 1). This means that for our

investigations, the modelling of the SDSCR process as conducted was accurate to within an average of 4.8 percentage points when compared to the actual process occurring within the SDS. Overall, the high values of the coefficient of determination indicate a good fit between the model and measured data, with variability in the inlet temperature and the Ca/S ratio being adequately reflected in the prediction of desulphurisation efficiency. The lowest value of 0.77 agrees with the corresponding highest RMSE as noted for Case 3.

Table 5. Goodness-of-fit of the validated case studies (Details of the various cases are given in Table 1).

Process Variable	Case 1	Case 2	Case 3	Case 4	Case 5	Case 6	Case 7
RMSE of the efficiency	4.7	5.7	6.0	5.5	5.6	3.8	2.6
Coefficient of determination (R^2)	0.92	0.90	0.77	0.85	0.89	0.94	0.95

5.3.1. Effect of Inlet Temperature on SO₂-Removal Efficiency

In terms of the effect of temperature on SO₂-removal efficiency, Figure 9 shows that an increase in temperature from 108 °C to 142 °C at a constant Ca/S of 1.5 is accompanied by a decrease in removal efficiency. This is evident in both the experimental and model-predicted values. It can be explained to be a result of the increased water evaporation from the droplet, which increases at higher temperatures. The presence of water in the droplets as explained previously helps to improve SO₂ absorption. Similar behaviour was observed for the other Ca/S ratios investigated in this work. The observation explains why relatively low temperatures are used in practice for the SDSCR process which necessitates the cooling of the generally hotter flue gas prior to the desulphurisation process.

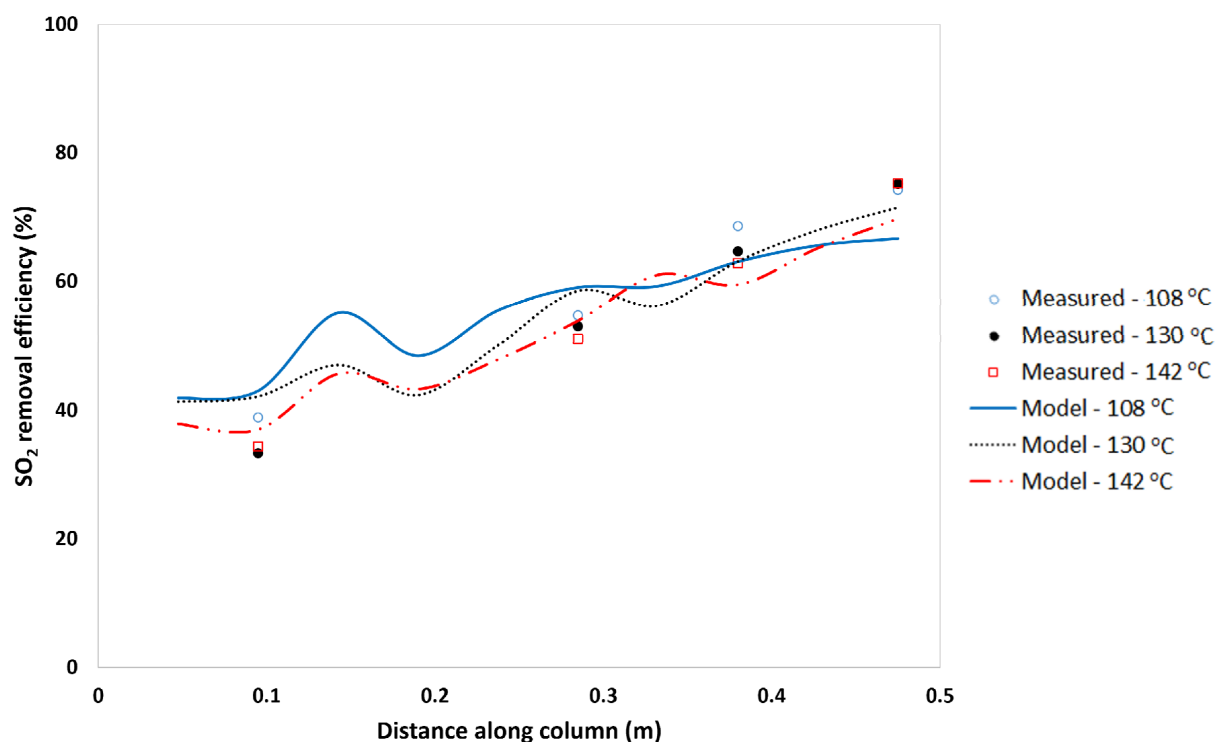


Figure 9. Change in the axial SO₂-removal efficiency along the column at various inlet flue-gas temperatures at a Ca/S ratio of 1.5 and an L/G ratio of 0.024.

5.3.2. Effect of the Ca/S on SO₂-Removal Efficiency and Sorbent Utilisation

Figure 10 shows the effect of increasing the Ca/S ratio from 1 to 2 at a fixed inlet temperature of 142 °C. Both the measured and model-predicted values show that an

increase in the Ca/S ratio results in an increase in SO₂-removal efficiency. A marked increase from a ratio of 1.5 to 2 was seen, compared to that from 1 to 1.5. In practise, the Ca/S ratio may be increased by either increasing the calcium content in the slurry for a fixed SO₂ content in the gas phase or reducing the SO₂ amount in the gas phase in contact with the slurry having a fixed calcium content. In the former case, the improvement in efficiency with increased sorbent injection emanates from reduced water evaporation due to increased solids hindrance from the added calcium in the slurry—taking into account that SO₂ absorption occurs in the presence of water. On the other hand, for the latter case, SO₂-removal efficiency increases because of the corresponding increase in the available mass transfer area per unit amount of SO₂ present in the system. These explanations hold for the case where physical absorption is the rate-limiting step as applicable for this study. Similar behaviour was observed for the other temperatures studied in this work.

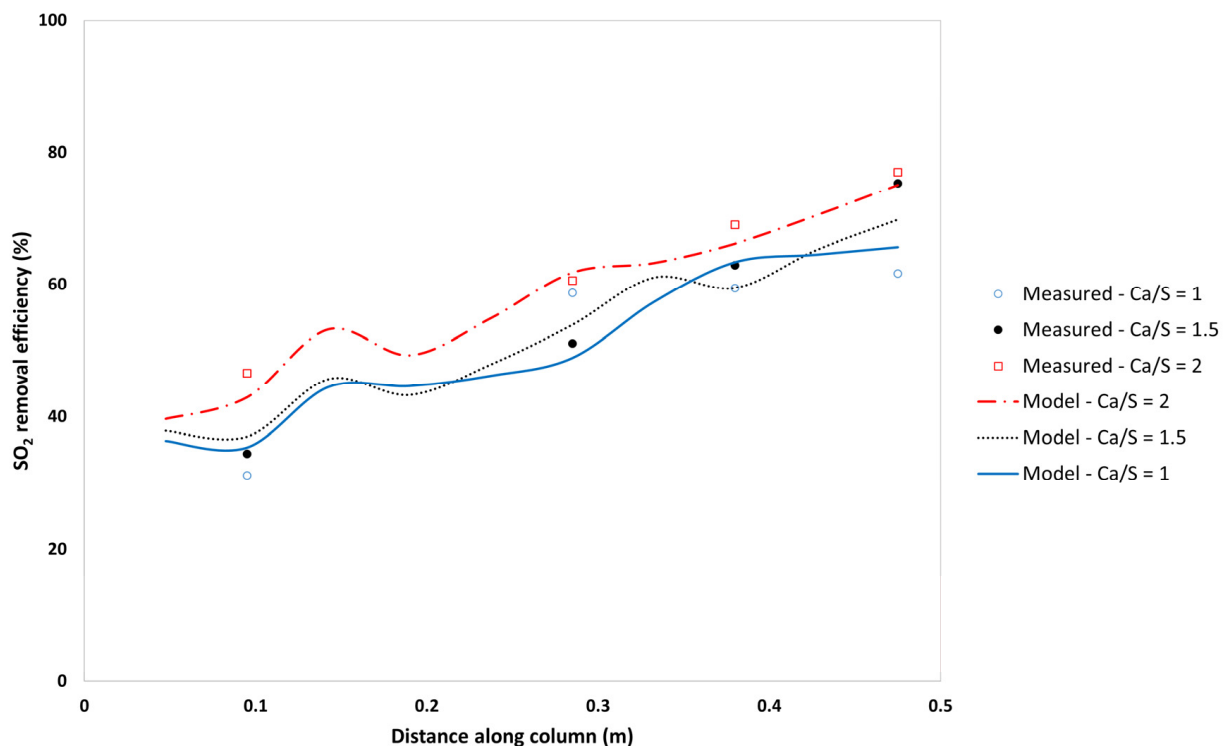


Figure 10. Change in the axial SO₂-removal efficiency along the column at 142 °C flue-gas inlet temperature and an L/G ratio of 0.024, with various Ca/S ratios.

Sorbent utilisation is defined in Equation (38) and gives an indication of how effectively a sorbent is utilised in the SO₂ removal process. A high utilisation value is particularly important for the SDSCR process due to not only the high cost of the sorbent used but also to the high cost of the disposal of the solid product. Figure 11 shows the calculated sorbent utilisation for the different Ca/S ratios investigated in this work at a constant inlet temperature of 142 °C and an L/G ratio of 0.024. From the plot, the sorbent utilisation is seen to increase down the column, resulting in an increase in SO₂-removal efficiency down the column, as shown in Figure 9 for all the Ca/S ratios investigated. What is also evident from the plot is a decrease in the sorbent utilisation with an increase in the Ca/S ratio. The increase in the Ca/S ratio increases SO₂-removal efficiency at the cost of higher sorbent utilisation. This is further explained below.

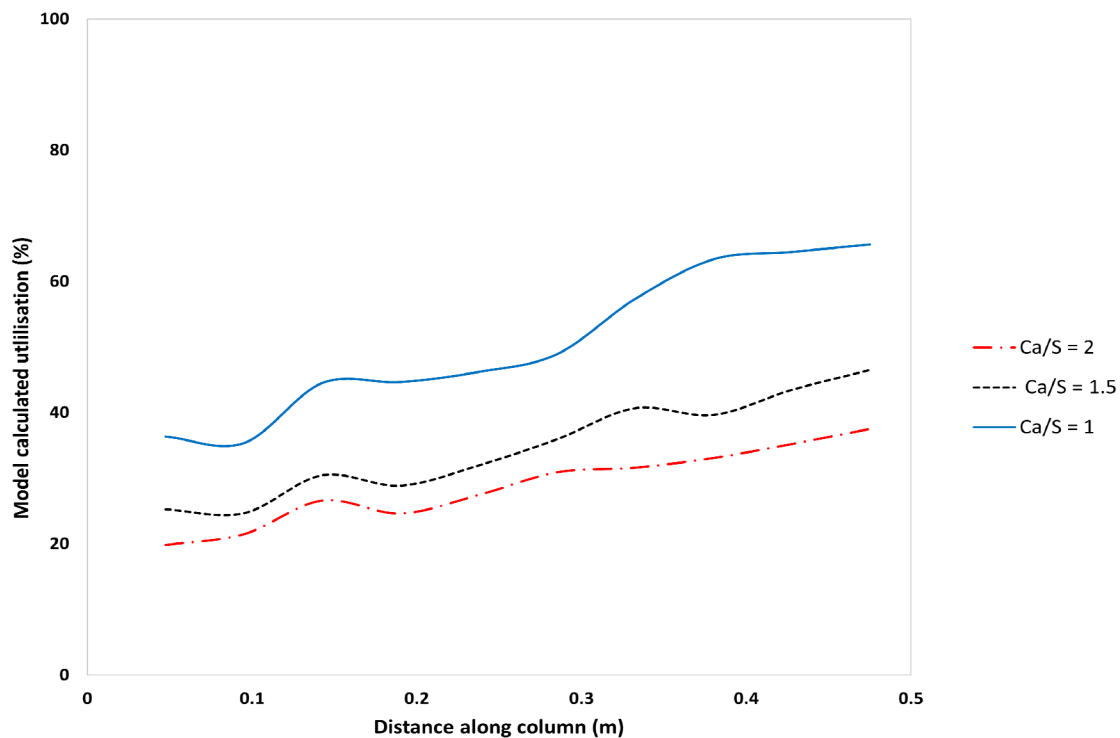


Figure 11. Calculated sorbent utilisation along the column as a function of the Ca/S ratio at a constant inlet flue-gas temperature of 142 °C and an L/G of 0.024.

Table 6 reports the exit SO₂-removal efficiency and calculated sorbent utilisation for the three cases investigated: Cases 3, 6, and 7. Results show that despite the increase in SO₂-removal efficiency with an increase in the Ca/S ratio, the utilisation decreases. This is because increasing the Ca/S ratio beyond the stoichiometric requirement for a fixed inlet SO₂ concentration, as was the case for the results in Table 6, will invariably lead to a reduction in the sorbent utilisation, although accompanied by an increase in removal efficiency. A 10% increase in the removal efficiency is accompanied by a >30% reduction in the sorbent utilisation. This indicates that improved efficiency may thus be forfeited for improved utilisation, where conditions allow for such a trade-off. Increasing the amount of sorbent for improved efficiency may thus be utilised when sorbent recycling is employed to improve the low utilisation.

Table 6. Comparison of model outlet SO₂-removal efficiency and calculated sorbent utilisation for various Ca/S ratios at a constant temperature of 142 °C and L/G of 0.024.

Process Variable	Case 3	Case 6	Case 7
Ca/S ratio	1	1.5	2
Model outlet efficiency (%)	70.7	73.9	78.6
Model calculated outlet utilisation (%)	70.7	49.3	39.3

5.3.3. Effect of the L/G Ratio on the SO₂-Removal Efficiency

The effect of increasing the L/G ratio in the scrubber is presented in Figures 12 and 13. Figure 12 reports SO₂-removal efficiency along the axial sampling locations used in the study for four different L/G ratios. An increase in the L/G ratio was seen to result in a marked increase in SO₂-removal efficiency. The highest efficiencies were observed for an L/G ratio of 0.049. For one of the L/G ratios (Case 2: L/G 0.024), validation of the measured data compared to model data was conducted. A good fit, as shown previously,

was obtained with a resultant RMSE of 5.7 (shown in Table 5), giving confidence in the other model predictions at different L/G values.

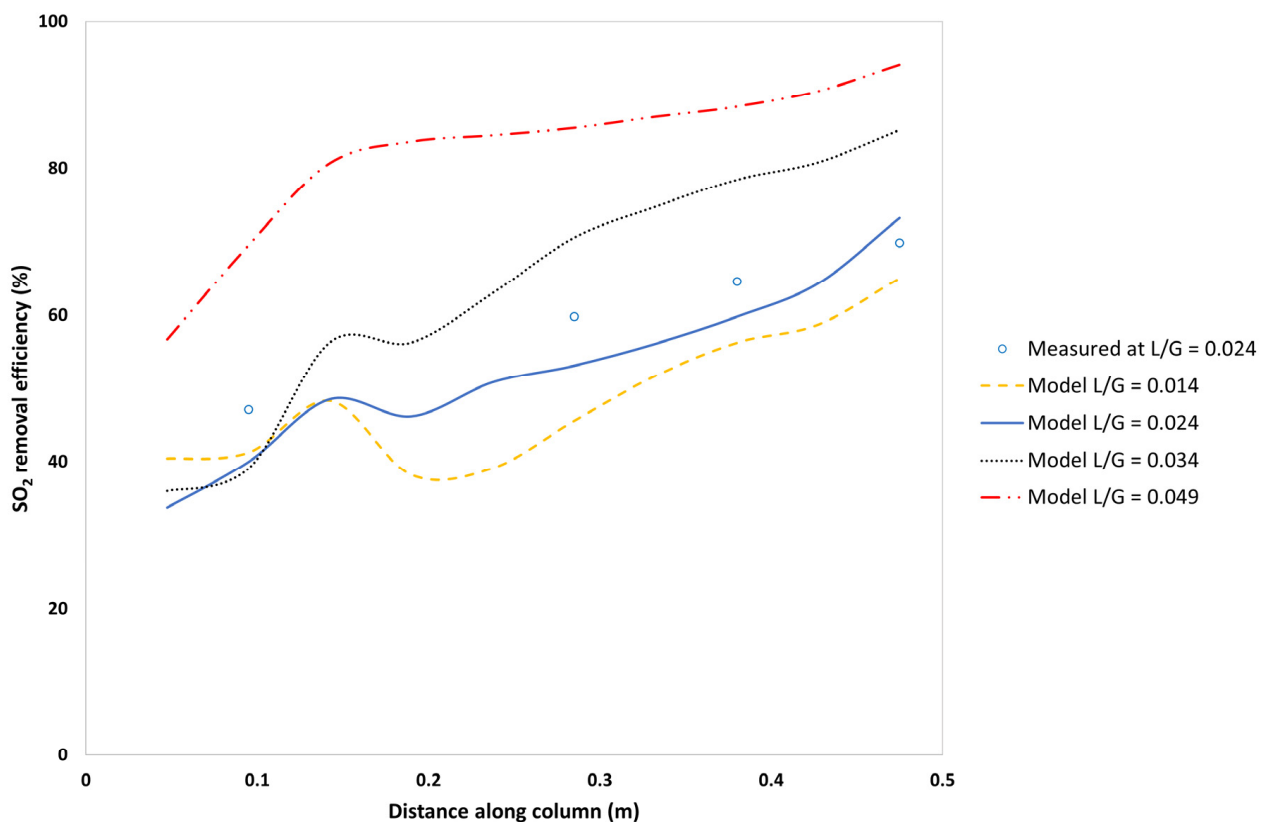


Figure 12. Axial variation in SO₂-removal efficiency along the column with change in the L/G ratio at an inlet flue-gas temperature of 130 °C and Ca/S of 1.

Figure 13 shows the axial plane profiles for L/G ratios of 0.014 and 0.049. Higher SO₂-removal efficiencies are also shown for the higher L/G ratio. It is notable that at the high L/G ratio, high efficiencies of >50% are observed midway in the column, in contrast to the low L/G ratio case where lower efficiencies are evident, even at the bottom of the column. Increasing the L/G ratio increases the amount of slurry droplets contacting the SO₂-laden flue gas, and hence the available overall surface area for mass transfer, leading to an increase in the mass transfer taking place within the scrubber. Taking note that the rate-limiting step for the reaction is SO₂ absorption into the liquid phase, increasing the available liquid slurry increases the amount of SO₂ ending up in the liquid phase from where it instantaneously reacts to form the hemihydrate salt.

Figure 14 shows the profile of the moisture content within the column for the lower L/G ratio (0.014) and the higher L/G ratio (0.049). Both profiles show particles with low moisture content at the top of the column. This is because of the recirculation towards this section of the column; thus, the particles in this area generally have low moisture and longer residence times. In the lower section of the column, particles with a higher moisture content are seen, and for the higher L/G ratio, a larger proportion of higher moisture particles are evident—the same particles that also report to the outlet leg.

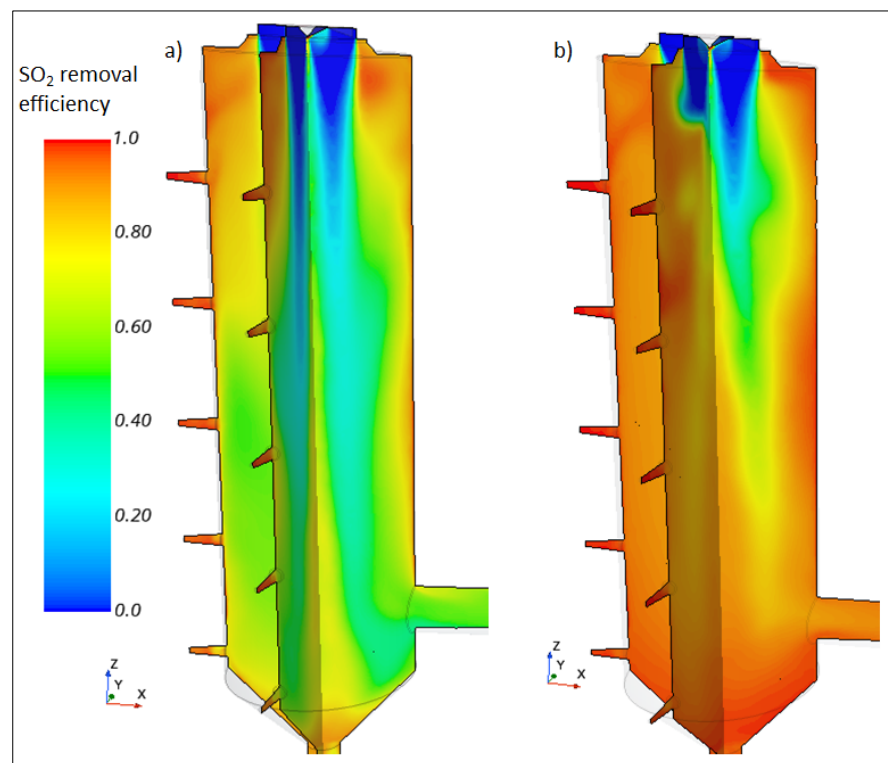


Figure 13. Axial profiles of SO₂-removal efficiency within the column showing both the x–z plane and the y–z plane for (a) an L/G ratio of 0.014 and (b) an L/G ratio of 0.049 at an inlet flue-gas temperature of 130 °C and a Ca/S ratio of 1.

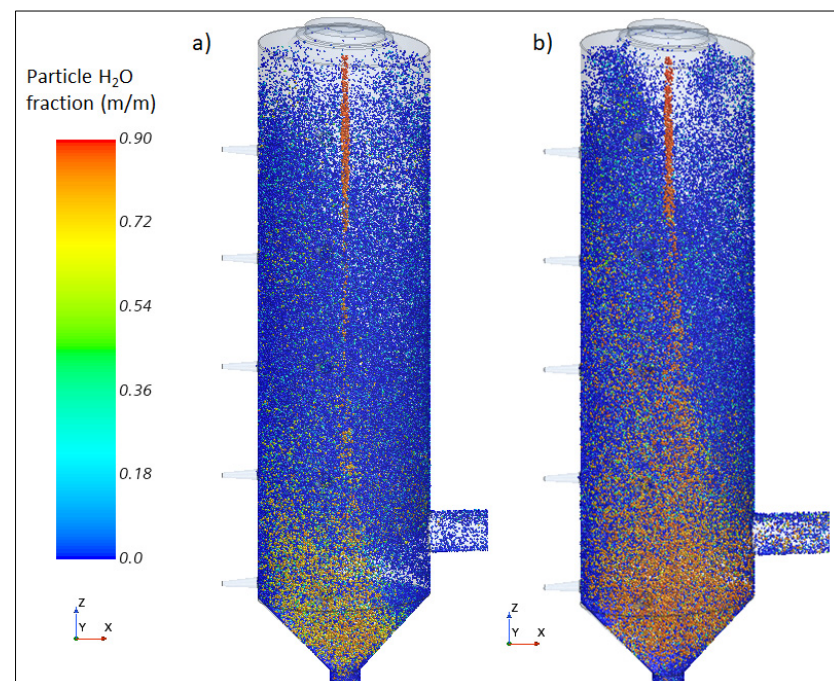


Figure 14. Axial cross-sectional snapshot of droplets after 30 s solution time showing the particle mass fraction, for (a) an L/G ratio of 0.014 and (b) an L/G ratio of 0.049 at an inlet flue-gas temperature of 130 °C and a Ca/S ratio of 1.

The predicted SO₂-removal efficiency at the column outlet as well as the corresponding predicted moisture content of the final product were also determined for various L/G

ratios. See Figure 15. With an increase in the L/G ratio, there is an increase in SO₂-removal efficiency at the column outlet. Concurrently, there is an increase in the outlet product moisture content. This is attributed to the fact that increasing the L/G ratio increases the energy demand for the evaporation process. Given that the inlet temperature remains constant, the overall effect will be a reduction in the amount of water evaporated from the droplets, resulting in an increase in the outlet product moisture content. The slower the drying process, the better the SO₂-removal efficiency—hence the observed simultaneous increase in SO₂-removal efficiency with delayed drying. However, a wet SDS outlet product results in challenges in terms of pneumatic conveying and downstream handling, and an increasing risk of fouling inside the scrubber. Thus, in reality, a compromise has to be reached between ensuring higher efficiencies with the associated disadvantages of handling a wet outlet product and relatively lower efficiencies associated with a dry outlet product and easier handling.

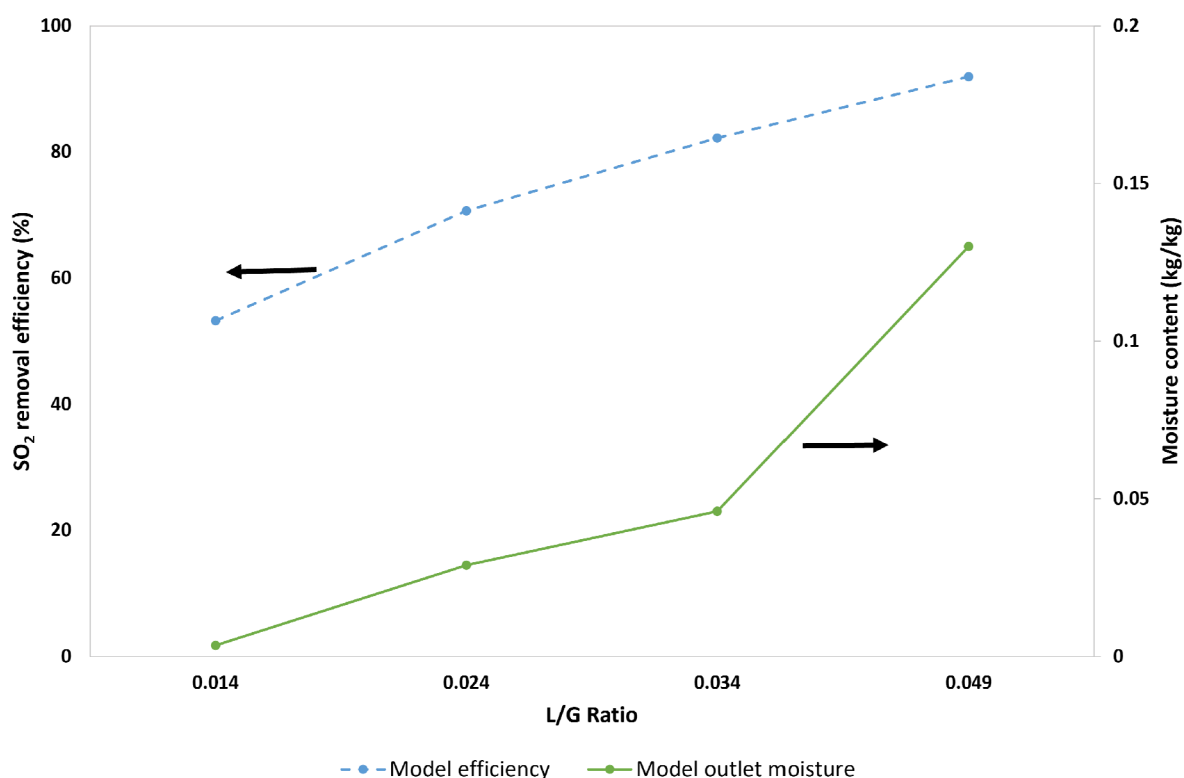


Figure 15. Change in the exit SO₂-removal efficiency and outlet product moisture content with change in the L/G ratio at a constant inlet flue-gas temperature of 130 °C and a Ca/S of 1.

Further increasing the L/G ratio ultimately changes the process to wet FGD where even higher SO₂ removal efficiencies of >90% have been reported. Wet FGD handles a slurry product and thus presents the extreme end in terms of solids handling and associated treatment and disposal costs. A techno-economic evaluation will, therefore, be needed in practice to determine the appropriate SO₂ removal technology. However, in dry environments where water availability is a challenge, wet FGD may not be an option and the degree of water addition (max L/G ratio that can be utilised) will also be determined from techno-economic considerations.

6. Conclusions

In this study, a comprehensive model for the desulphurisation process in a spray dry scrubber was developed. SO₂ absorption was investigated, taking into account the sorbent slurry pH, which determined the relevant mass transfer mechanism for the absorption process. Under conditions of high pH (>12), the rate-controlling mechanism is governed by

the transfer of SO₂ from the bulk gas to the droplet interface, where all the absorbed SO₂ reacts instantaneously with the calcium to form the calcium sulphite product. The resultant absorption equations were incorporated in the overall CFD model as submodels using passive scalars in STARCCM+. The developed comprehensive model was validated by comparing the model-predicted data with experimentally measured values and the RMSE was used as a measure of the goodness-of-fit.

The classical D2-law model and mechanistic drying model were compared. Results indicated over-prediction of SO₂-removal efficiency of the classical D2-law model compared to the mechanistic model. Such over-prediction was deemed to be a result of the classical D2-law model failing to take into account the mass transfer hindrance of the slurry solids on the SO₂ mass transfer. The mechanistic model, which had previously been proven to be the best drying model, gave a better representation of the complete process incorporating SO₂ absorption.

This work has highlighted the need for evaluating the applicability of the different drying models prior to their use in a comprehensive modelling framework for the spray dry scrubbing process. This is because the resultant SO₂-removal efficiency is largely influenced by particle moisture, which in turn is a product of the drying process, and can only be accurately determined by considering the solids hindrance to mass transfer inside the droplet.

Furthermore, sensitivity analysis on key process variables was carried out and their resultant effects on SO₂-removal efficiency were determined. Parameters that resulted in an increase in SO₂-removal efficiency included the following: reducing the inlet flue-gas temperature, increasing the Ca/S ratio, and increasing the L/G ratio. Increasing the latter ratio had the effect of increasing the particle moisture content, which in turn increased the SO₂ absorption rate, resulting in high efficiencies. A high particle moisture content was noted to be undesirable, however, due to problems with particulate handling systems, including downstream pneumatic conveying. A compromise must, therefore, be made between increasing SO₂-removal efficiency at the risk of downstream operating equipment challenges as well as scrubber fouling. Sorbent utilisation, which is a measure of how effective the added sorbent is in removing SO₂ in flue gas, was also determined. There was an inverse relationship between it and an increased Ca/S ratio. An increase in the efficiency resulted in a decrease in the sorbent utilisation, which, in practice, can be mitigated by recycling the used sorbent.

In conclusion, our studies have led to the development of a comprehensive model for the spray dry scrubbing process. Insight has been obtained into the complex chemical and physical processes inside the spray dry scrubber—information that the authors believe will subsequently be invaluable in optimizing both new and existing spray dry scrubbing systems. Potential further work from the study includes conducting a techno-economic assessment between semi-dry FGD as conducted in this work and wet FGD for a case where both technologies are applicable and water availability is not a concern. This stems from the fact that wet FGD is the limiting case for increasing the L/G ratio in semi-dry FGD. The authors also recommend proceeding with the study and evaluating other drying models beyond the two models investigated in this work, in predicting the SO₂-removal efficiency in SDSs. These may include lumped-parameter models and even internal diffusion-based models, taking into consideration the increased computational cost associated with the more complex models.

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Conflicts of Interest: Ignus Le Roux is employed by the company Aerotherm. The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

Nomenclatures

ρ	Density (kg m^{-3})	μ	Viscosity (Pa s)
u	Velocity (m s^{-1})	μ_t	Eddy viscosity (Pa s)
w	Species mass fraction (-)	g	Gravitational term ($\text{m}^1 \text{s}^{-2}$)
D	Mass diffusivity ($\text{m}^2 \text{s}^{-1}$)	Re	Reynolds number (-)
t	Time (s^{-1})	Pr	Prandtl number (-)
E	Energy per unit mass (J kg^{-1})	Sc	Schmidt number (-)
P	Pressure (Pa)	C_D	Coefficient of drag (-)
p	Partial pressure (Pa)	C_p	Specific heat capacity (J K^{-1})
k_c	Thermal conductivity ($\text{W m}^{-1} \text{K}^{-1}$)	h	Heat transfer coefficient ($\text{W m}^{-2} \text{K}^{-1}$)
τ	Stress tensor (Pa)	A	Area (m^2)
τ^R	Reynolds stress tensor (Pa)	h_{vap}	Latent heat of vaporisation (J kg^{-1})
T	Temperature (K)	$k_{i,g}$	Gas-phase mass transfer coefficient for component i (m s^{-1})
ω	Specific dissipation rate (s^{-1})	$K_{i,c}$	Overall mass transfer coefficient for component i (m s^{-1})
k	Turbulent kinetic energy ($\text{m}^2 \text{s}^{-2}$)	$S_{d_i,d_{ag}}$	Geometric parameter (m)
x	Position (m)	A_{ag}	Agglomerate area (m^2)
d	Diameter (m)	ϵ	Tortuosity parameter (m)
v	Particle velocity (m s^{-1})	n	Molar flow rate (mol)
Subscripts		mom	
A	Species A	k	k^{th} particle
B	Species B	g	gas
d	Droplet	l	liquid
p	Particle	∞	Bulk phase
m	mass	E	energy
s	Droplet/particle surface		
Superscripts			
$-$	vector term	$\cdot$	Rate

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