

Interactive effects of spray drying characteristics on SO₂ capture using limestone sorbent: a response surface methodology study

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Abstract

This study involved an investigation to assess the performance of limestone sorbent for spray dry scrubbing of sulphur dioxide from simulated flue gas using a lab-scale spray dryer. The project comprised of detailed experimental testing utilizing Central Composite Design (CCD) to investigate the influence of spray drying parameters on the removal of SO₂ from flue gas. Several experiments were performed to analyse the influence of stoichiometric ratio (0.5 - 2.5), flue gas flowrate (24 - 36 m³/h), and inlet gas temperature (120 - 200 °C) on SO₂ removal efficiency of limestone. A predictive quadratic model was established based on experimental findings to correlate independent variables and the response. The model exhibited a strong fit to the data, indicated by R-squared coefficient of 0.98. The experimental findings revealed that the stoichiometric ratio (SR) had a large impact on the removal efficiency of SO₂ in the spray dryer while the flue gas flowrate through the scrubber exhibited minimal influence. The combined effects of the spray drying variables were found to significantly impact the removal efficiency. A high removal efficiency of 70% was achieved when employing a higher stoichiometric ratio (SR=2) and lower temperature (T=140 °C). The lowest removal efficiency was recorded at a high temperature (T=180 °C) coupled with lower stoichiometric ratio (SR=1). The characterization of the dry collected products revealed evidence of desulphurization, manifesting in the formation of hannebachite (Ca₂SO₃·2H₂O) and unreacted sorbent.

Keywords

Spray dry scrubbing, SO₂, limestone, Response Surface Methodology, interactive effects.

Introduction

The mitigation of harmful gases like sulphur dioxide (SO₂) and nitrous oxide (NO_x) has been a subject of significant attention over the years. These gases are predominantly released during the combustion of coal containing sulphur, leading to the emission of pollutants in power plants. These gases among others generated from combustion of coal contribute to severe environmental pollution and detrimental effects on hu-

man health (Almetwally *et al.* 2020). As a consequence, strict legislations and regulations have been established to address industrial air pollution, specifically targeting SO₂ emissions (World Health Organization, 2021). This has consequently mandated the implementation of flue gas scrubbing methods, specifically flue gas desulphurization (FGD) units, in thermal power plants using coal. In recent years, spray drying absorption (SDA) has emerged as a promising semi-dry flue gas

desulfurization (FGD) technology for coal-fired power plants. It offers several advantages over wet FGD systems, including a smaller footprint, simplified by-product management/disposal, lower capital costs, ease of retrofitting to existing plants, and reduced water usage (Carpenter, 2017; Rezaei *et al.* 2015; Roy and Sardar, 2015). In the SDA process, the sorbent is delivered into the absorption chamber via a spray nozzle. The absorption chamber allows the interaction between the droplets containing the sorbent and SO₂ present in the hot flue gas stream, to form Ca₂SO₃ and Ca₂SO₄ (Hrdlička and Dlouhý, 2019; Li *et al.* 2022a). The resulting dry desulphurization residue is collected below the chamber and in a downstream particle collection device.

The spray drying process in FGD is primarily governed by adsorption, and its effectiveness is influenced by several key factors. These factors encompass the surface properties of the sorbent, particle size, concentration of solid in the slurry, approach to saturation temperature and humidity levels, among other variables (Gassner *et al.* 2014). Extensive experimental investigations have been carried out with a primary focus on analyzing the operational parameters that influence the removal of SO₂ in a spray dryer (Du *et al.* 2020; França *et al.* 2020; Goncaloglu *et al.* 2009; Wey *et al.* 2003; Yi *et al.* 2020). Du *et al.* (2020) developed an integrated process that simultaneously addresses desulphurization and denitrification under optimized conditions. High removal rates for both SO₂ (>90%) and NO (90.5%) were achieved under the determined optimal conditions for the spray dryer. Yang and Kim (2000) conducted an assessment of spray characteristics within a pilot-scale spray drying absorber, utilizing a hydrated lime sorbent. Their study determined the optimal droplet diameter for effective desulphurization based on specific stoichiometric ratios and temperature conditions. Similarly, Ollero *et al.* (1997) employed a pilot plant to investigate the impact of variables such as Ca/S ratio, approach to saturation temperature, unit load changes, and makeup water in SO₂ scrubbing processes. Their research highlighted the importance of maintaining a minimum Ca/S ratio to optimize operational costs. Understanding the effects of operational parameters is crucial for enhancing overall plant performance, as reported by Scala *et al.* (2004). Optimal performance of the spray dryer absorber in the desulphurization process relies on careful optimization of these operating parameters. Although the commercial utilization of SDA has demonstrated the capability to exceed regula-

tory requirements for SO₂ removal, its widespread implementation has been constrained by factors such as costly sorbents (hydrated lime – Ca[OH]₂), low utilization of the sorbent, and lower desulphurization efficiency in comparison to the wet limestone FGD technology (França *et al.* 2020; Li *et al.* 2022b). It is crucial to thoroughly investigate and evaluate the potential of locally available alternative sorbents, with limestone being a particularly promising option. Limestone is readily available and significantly more cost-effective (5-10 times cheaper) compared to hydrated lime, making it a probable sorbent for spray drying purposes (Bontzolis *et al.* 2019). This exploration can pave the way for more efficient and cost-effective solutions in the realm of flue gas desulfurization.

The study was aimed to comprehensively understand the intricate relationship between the operating variables and the efficiency of SO₂ removal in spray drying desulfurization process. The experiments involved the manipulation of independent variables including stoichiometric ratio (ranging from 0.5 to 2.5), inlet flue gas temperature (ranging from 120 to 200 °C), and flue gas flowrate (ranging from 24 to 36 m³/h) using central composite design (CCD). The stoichiometric ratio in spray drying FGD is defined as the ratio between the moles of calcium in the incoming slurry and the moles of SO₂ introduced into the scrubber. The established limits in this study were determined through trial experiments that were conducted, taking into account the SO₂ concentrations (up to 3000 ppm) typically found in industrial flue gas. Response Surface Methodology (RSM) was employed to analyse the interrelationships between the input variables and their impact on the efficiency of SO₂ removal.

Materials and Methods

Materials and experimental

The laboratory-scale spray dryer depicted in Figure 1 was utilized to conduct the experiments in this study. The limestone sorbent used was obtained from PPC Lime, Northern Cape, South Africa. The chemical and physical properties of the limestone can be found in Table 1. All particles of the sorbent passed through a 45 µm size sieve. SO₂ gas of 99.99% purity was supplied by Afrox, South Africa. The preparation of the slurry involved the combination of a predetermined mass of limestone sample with ultra-pure water in a separate

reaction vessel. To ensure homogenous feed slurry, a magnetic stirrer was employed, and the mixing process was carried out for approximately 10 minutes before each test run. Throughout the test runs, the slurry was continuously stirred and pumped into the spray chamber via a spray nozzle. A synthetic flue gas (1000 ppm) was produced by combining 99.99% pure SO₂

with air using an aspirator pump. SO₂ concentration in the synthetic flue gas was adjusted by controlling the flowrate of SO₂ into the pump. The flue gas was subjected to heating within the temperature range of 120 to 200 °C at the chamber inlet. The heated gas then enters the spray chamber, where it interacts with the dispersed slurry, facilitating the absorption of SO₂.

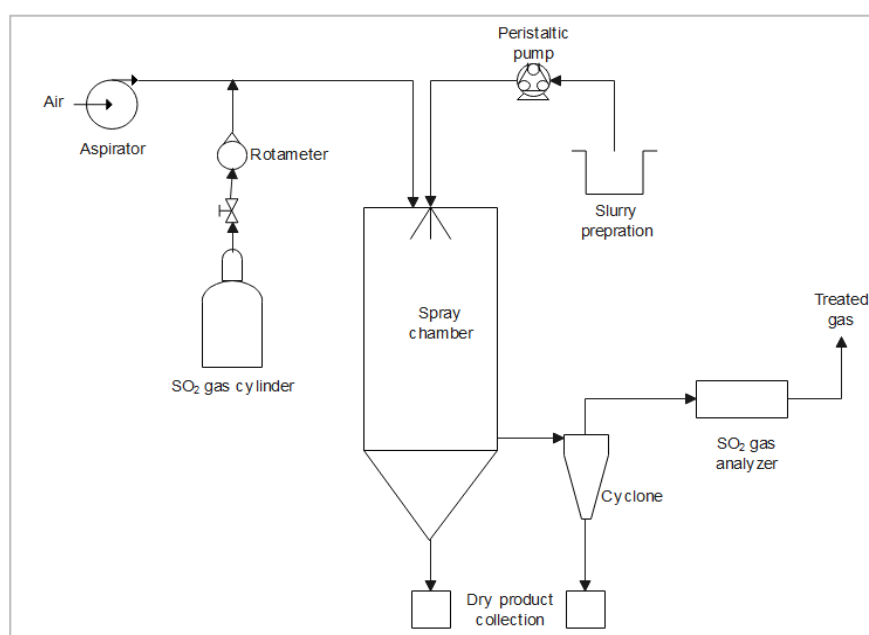


Figure 1

Schematic of the experimental SDS setup

	Components (wt.%)					
	MgO	Al ₂ O ₃	SiO ₂	MgO	CaO	LOI
Limestone (CaCO ₃)	0.58	0.23	0.79	1.17	54.43	42.54

Table 1

Limestone sorbent chemical analysis (XRF analysis)

The experimental setup included a control panel to regulate the flue gas flowrate, slurry flowrate, and inlet gas temperature. The concentration of the solids in the feed slurry was adjusted to meet the desired stoichiometric molar ratio (SR) while maintaining the inlet SO₂ concentration constant at 1000 ppm. Throughout each test run, SO₂ concentration in the exiting flue gas stream was continuously monitored and measured using a Testo 340 flue gas analyzer. The analysis of SO₂ continued until a steady reading was achieved and the results were recorded. To ensure the reliability of the data, each test run was conducted in triplicates. Following the completion of each test run, the collected dry desulphurization residue in the product vessel was stored for subsequent analysis. The desulphurization efficiency was calculated using the

formula shown in equation [1] below:

$$\eta = \frac{N_{SO_2,i} - N_{SO_2,o}}{N_{SO_2,i}} \times 100\% \quad [1]$$

where $N_{SO_2,i}$ is the inlet SO₂ concentration, while $N_{SO_2,o}$ is the outlet SO₂ concentration, both in ppm. Different characterization methods were employed to assess the attributes of both the raw limestone sorbent and the collected desulphurization residue. The specific surface area of the limestone sorbent was determined through N₂ BET analysis using Micromeritics ASAP™ 2020 Porosity Analyzer. XRF analysis was employed to perform elemental analysis of the sorbent. XRD analysis of raw sample was conducted using a Malvern Panalytical Aeris Diffractometer equipped with a

PIXcel detector and fixed slits, employing Co-K α radiation filtered by Fe. Additionally, micrographs of the samples were obtained using a Philips XL-30S scanning electron microscopy (SEM).

Design of experiments and statistical analysis

Response surface methodology (RSM) in the Design Expert software (Ver 13.0.01.0) was used to assess the interactive effects of the spray drying parameters on the desulphurization efficiency. RSM is a combined mathematical and statistical approach that is utilized to model and analyze situations where multiple variables impact a desired response (Myers *et al.* 2016). A central composite design (CCD) with $\alpha=2$ in the Design Expert software was utilized for the design of experiments incorporating three input variables (inlet flue gas temperature, stoichiometric ratio and flue gas flowrate) that were selected for the study. To establish the range of the independent variables, preliminary experiments were conducted to identify their maximum and minimum values. A total of 20 experiments, as presented in Table 2, were then performed, and the corresponding response of SO₂ removal efficiency was recorded.

The experimental data obtained from CCD were used to fit a second-order polynomial model, from which regression coefficients were derived. The response surface analysis employed the second-order polynomial model as shown in equation [2]:

$$\eta = b_0 + \sum_{i=1}^n b_i x_i + \sum_{i=1}^n b_{ii} x_i^2 + \sum_{i=1}^{n-1} \sum_{j=i+1}^n b_{ij} x_i x_j \quad [2]$$

The calculated desulphurization efficiency (%) is represented by η . The significance of the polynomial model (equation 2) was assessed through the application of analysis of variance (ANOVA) and a lack of fit test.

The adequacy of the model was assessed by taking into account the statistic R-squared (R²).

Results and Discussion

Sorbent properties

The chemical analysis (XRF analysis) of the sorbent used in this study is presented in Table 1. The predominant component of the sorbent is calcium, specifically in the form of calcium oxide (CaO), accounting for 54.43% of its weight with 42.54% loss on ignition (LOI). Calcium oxide serves as the active ingredient in the chemisorption reaction. Furthermore, the XRD analysis depicted in Figure 2 demonstrates the presence

of significant calcite (CaCO₃) content, as evidenced by multiple distinct peaks observed. The sorbent specific surface area, obtained through N₂ BET surface area analysis, was determined to be 2.51 m²/g which is sufficient and advantageous for effective desulphurization in the spray drying process.

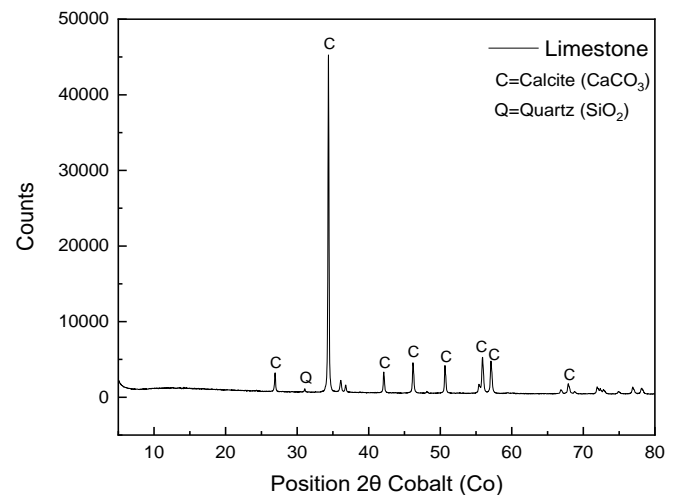


Figure 2. XRD pattern for raw limestone sorbent

Model fitting and significance test

Table 2 presents a complete design matrix of experiments, including all input variables and the response, which is the SO₂ removal efficiency. The range of SO₂ removal observed in these experiments varied from 38% to 74%. Through regression analysis of the data in Table 2 using CCD, a predictive quadratic model (equation [3]) was established correlating the coded factors and the desulphurization efficiency.

$$\eta = 54.95 - 5.5x_1 + 7.5x_2 + 1.75x_3 - 1.75x_1x_2 + 1.85x_1^2 - 0.6477x_2^2 + 0.2273x_3^2 \quad [3]$$

where η = SO₂ removal efficiency (%) – x_1 = inlet flue gas temperature (C°) – x_2 = Stoichiometric ratio (Ca:5) – x_3 = flue gas flowrate (m³/h).

The coded equation provides an insight into the effects of the input variables (spray drying parameters) on SO₂ removal efficiency, by the comparison of the model coefficients.

To assess the significance of the predictive model and its individual terms, ANOVA was performed. Table 3 provides the results of the lack of fit test and ANOVA. The suitability of the selected model was confirmed by a model F-value of 131.54 and a p-value of less than 0.0001, indicating a strong fit.

DOI: [10.6092/issn.2281-4485/17829](https://doi.org/10.6092/issn.2281-4485/17829)**Table 2.** CCD Experimental design matrix of experiments

Run#	x ₁ : Inlet gas temperature (°C)	x ₂ : Stoichiometric ratio (mol/mol)	x ₃ : Flue gas flow (m ³ /h)	SO ₂ Removal Efficiency (%)
1	180	2.0	33	57
2	140	1,0	27	52
3	160	1.5	30	55
4	160	1.5	24	52
5	200	1.5	30	53
6	160	1.5	30	55
7	140	2.0	33	74
8	160	1.5	30	55
9	180	2.0	27	55
10	180	1.0	27	42
11	120	1.5	30	72
12	160	1.5	30	55
13	160	1.5	36	60
14	160	2.5	30	67
15	160	0.5	30	38
16	140	2.0	27	70
17	180	1.0	33	46
18	160	1.5	30	55
19	160	1.5	30	55
20	140	1.0	33	54

Table 3. Analysis of variance (ANOVA) for the model

Source	Sum of squares	F-value	p-value	Significant
Model	1572.53	131.64	< 0.0001	Significant
x ₁ - Inlet temperature	484	364.66	< 0.0001	
x ₂ - Stoichiometric ratio	900	678.08	< 0.0001	
x ₃ – Flue gas flowrate	49	36.92	0.0001	
x ₁ x ₂	24.5	18.46	0.0016	
x ₁ x ₃	0	0	1	
x ₂ x ₃	0	0	1	
x ₁ ²	86.26	64.99	< 0.0001	
x ₂ ²	10.55	7.95	0.0182	
x ₃ ²	1.3	0.9785	0.3459	
Residual	13.27			
Lack of Fit	13.27			
Pure Error	0			
Cor Total	1585.8			

The significance of the model terms was assessed using p-values, with values below 0.0500 indicating their significance. In this case, all three model terms (x_1 , x_2 and x_3) showed considerable effect on SO₂ absorption efficiency. Among the interaction effects, only x_1x_2 was found to be significant, while the rest had p-values greater than 0.1000, indicating non-significance. Furthermore, the analysis revealed that the quadratic effects of all variables, shows that x_1^2 and x_2^2 are significant but x_3^2 is non-significant, with p-value greater than 0.0500. To validate the accuracy of the established model, the linearity between the actual (experimental) values and the predicted values was examined. Figure 4 illustrates a plot of the actual and predicted values, demonstrating a satisfactory alignment of the plot data alongside the line with a unit gradient. This yielded a high R-squared (R^2) value of 0.98 obtained for the model.

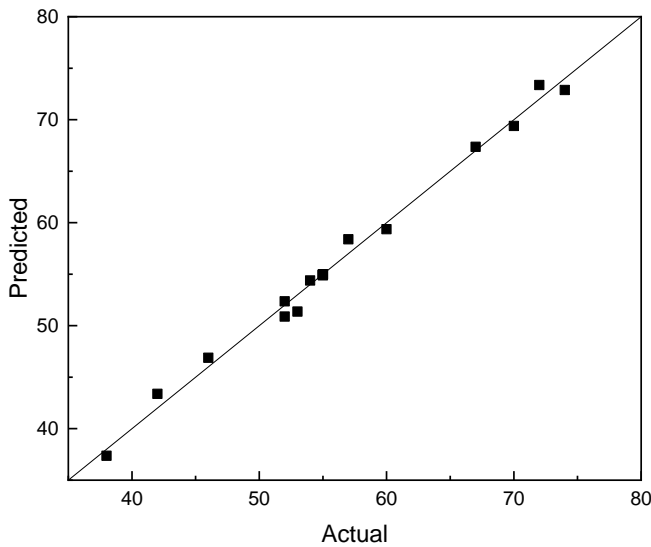


Figure 4. A plot of the predicted vs actual values

Effects of spray drying variables.

The interactive effects of the input parameters on SO₂ removal efficiency were visualized through 3D surface plots obtained from CCD. Each plot depicted the variation of two independent parameters while maintaining one variable constant at its central point. Figures 5-7 illustrates 3D surface response plots, offering a visual representation of how the independent variables interactively influence the efficiency of SO₂ removal.

Figure 5 shows the combined effects of the inlet flue gas temperature (x_1) and the stoichiometric molar ratio (x_2) on the desulphurization rate while maintaining a constant flue gas flowrate of 30 m³/h. The figure shows a substantial increase in the desulphurization rate with

increasing stoichiometric ratio. This is ascribed to the higher concentration of the reactive species (Ca²⁺), which helps to sustain the reaction at the surface of the droplets, resulting in a decrease in resistance to mass transfer within the liquid phase (Gao *et al.* 2011; Tavan and Hosseini, 2017). The impact of temperature on SO₂ removal efficiency in the spray dryer is demonstrated in Figure 5, showing a decrease in efficiency as the inlet flue gas temperature was increased. This consistent trend is further supported by Figure 6. It is known that high temperatures significantly reduce the solubility of both SO₂ and CaCO₃ within the droplet (Córdoba, 2015; Fakhari *et al.* 2017).

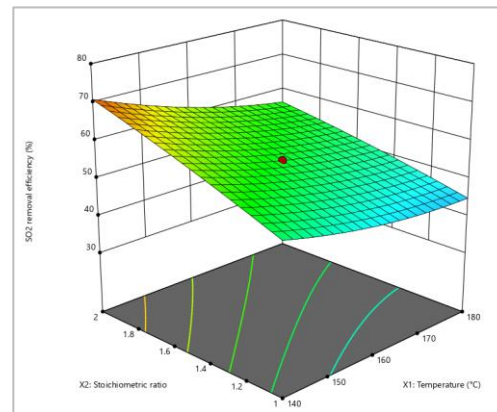


Figure 5 Effects of stoichiometric ratio and inlet flue gas temperature on the desulphurization efficiency.

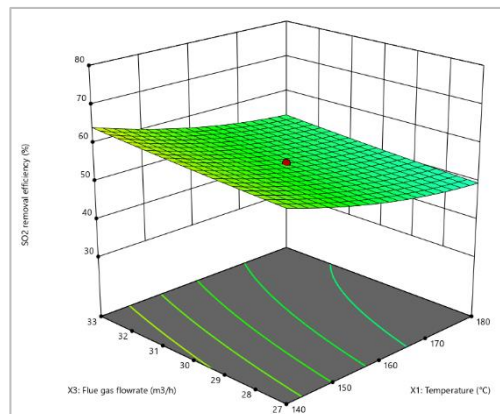


Figure 6 Effects flue gas flowrate and inlet gas temperature on the desulphurization efficiency.

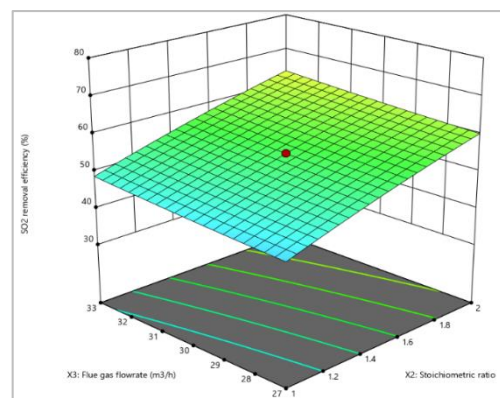


Figure 7 Effects flue gas flowrate and stoichiometric ratio on the desulphurization efficiency.

This reduces the degree of dissolution of CaCO_3 within the droplet and subsequently lowering the rate of SO_2 absorption due to low concentration of the dissolved reacting species. Furthermore, high temperatures accelerate the rate of evaporation in the spray dryer, shortening the necessary time required for absorption. The absorption mechanism in a spray dryer mainly occurs in the liquid phase (within the droplet), which is greatly influenced by rapid evaporation induced by elevated temperatures (Li *et al.* 2018). Figures 6 and 7 provide insights into the impact of flue gas flowrate on the efficiency of SO_2 removal, revealing a minimal effect. Initially, it was anticipated that variation of the flowrate would significantly influence the interaction between the flue gas and the droplets within the reaction chamber. However, there was only a slight enhancement in SO_2 removal efficiency as the flue gas flowrate increased. This suggests that the higher flowrate facilitated improved interaction within the chamber, promoting effective absorption of SO_2 . The findings from the model plots demonstrate that the chosen flowrates of the flue gas in this study caused almost uniform interaction in the spray chamber, resulting in minimal disruption to the absorption of SO_2 in the chamber. Figure 5 demonstrates the significant impact of both the stoichiometric ratio and inlet flue gas temperature on the efficiency of SO_2 removal. The highest removal efficiency of 74% was achieved at a lower temperature of 140 °C coupled with a high stoichiometric ratio of 2.0, which resulted in a 29% improvement compared to the high temperature of 180 °C and low stoichiometric ratio of 1.0. This highlights the pronounced influence of the stoichiometric ratio, as evidenced by its steep increase in removal efficiency, surpassing the effect of temperature. Figure 6 illustrates the interactive effects of the flue gas flowrate and inlet flue gas temperature on SO_2 . The model plot revealed that a high flue gas flowrate of 33 m^3/h combined with a lower temperature of 140 °C resulted in a remarkable SO_2 removal efficiency of 63%. This indicates a notable 14% increase compared to the value achieved with a lower flue gas flowrate of 27 m^3/h and a higher inlet flue gas temperature of 180 °C. Furthermore, Figure 6 highlights that the variation of inlet flue gas temperature has a more significant impact on the efficiency of SO_2 removal than the variation of the flue gas flowrate. These findings further support the conclusion that the spray dryer's removal efficiency of SO_2 was minimally affected by the flue gas flowrate. The combined influence of the flue gas flowrate and the stoichiometric ra-

tion on the removal of SO_2 is depicted in Figure 7. The combined effect of these factors is clearly illustrated, revealing a significant improvement in the removal efficiency of SO_2 . The model plot demonstrates an increase in the removal efficiency from 48% to 63% when transitioning from a lower flue gas flowrate of 27 m^3/h with a stoichiometric ratio of 1.0 to a higher flue gas flowrate of 33 m^3/h with a stoichiometric ratio of 2.0. The graph highlights the pronounced impact of the stoichiometric ratio, as evidenced by the substantial increase in removal efficiency. Conversely, the effect of varying the flue gas flowrate from 27 m^3/h to 33 m^3/h on the removal efficiency is relatively minor. In general, the observed desulphurization limits in the spray dryer were within the South African regulatory limit of 382 ppm (Department of Environmental Affairs, 2023), which demonstrates the possibility of using limestone as a sorbent in spray drying scrubbing of SO_2 from industrial flue gases.

Desulphurization residue analysis

Figure 8 illustrates the SEM micrographs of both the raw sorbent and the desulphurization residue. The micrograph in Figure 8A displays the plain regular surfaces of the sorbent particles, which appear fine and scattered. In contrast, Figure 8B reveals rough-surfaced particles in the residue, indicating some degree of agglomeration. This agglomeration is believed to have hindered the accessibility of active sites within the sorbent, leading to suboptimal sorbent utilization. This is consistent with the XRD analysis (Table 4), which indicates the presence of CaCO_3 in the desulphurization residue. The presence of unreacted CaCO_3 can be attributed to the poor solubility of limestone in water and its limited affinity towards SO_2 (Gu *et al.* 2020; Haynes, 2014). Additionally, XRD analysis identifies the presence of hannebachite ($\text{Ca}_2\text{SO}_3 \cdot 0.5\text{H}_2\text{O}$) as a desulphurization product in the residue. The agglomeration of desulphurization products may have impeded the exposure of active sites within the particles to react with SO_2 , further contributing to the presence of unreacted CaCO_3 in the residue.)

Table 3. Desulphurization residue XRD analysis (normalized major minerals)

Mineral name	Chemical formula	Composition
Calcite	CaCO_3	61.41 wt.%
Hannebachite	$\text{CaSO}_3 \cdot 0.5\text{H}_2\text{O}$	30.21 wt %
Magnesium sulphite	MgSO_3	0.79 wt %
Silica	SiO_2	1.42 wt %

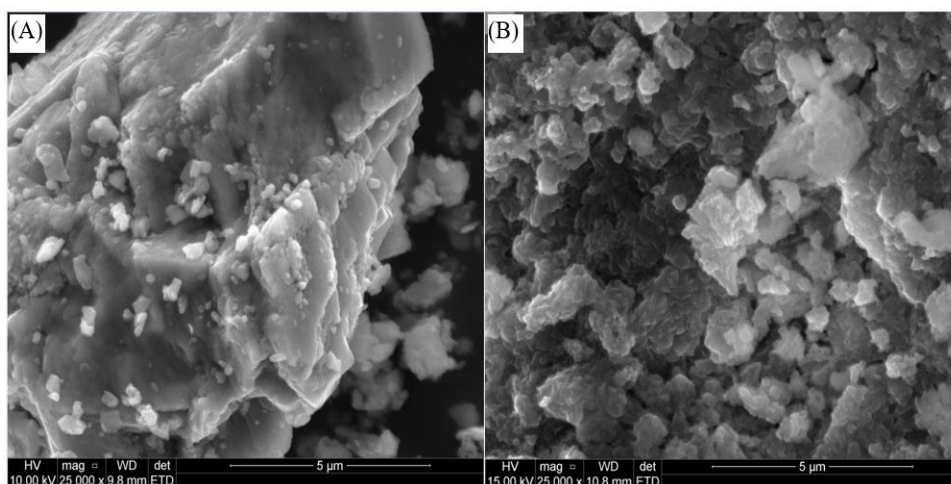


Figure 8
SEM micrographs of the raw sorbent (A) and the desulphurization residue (B)

Conclusions

This study presents an investigation into the interactive influence of spray drying parameters on desulphurization efficiency using limestone as the sorbent. Through the application of response surface methodology, the independent variables, including stoichiometric ratio, gas flowrate, and inlet flue gas temperature, were systematically varied and manipulated to assess their combined effects. The experimental data was regressed using Central Composite Design to establish a predictive polynomial model that correlates the input parameters with desulphurization efficiency. The model demonstrated a satisfactory fit, as indicated by a high R-squared value of 0.98. Analysis of variance (ANOVA) revealed that the stoichiometric ratio exerted a significant influence on desulphurization efficiency, while the impact of flue gas flowrate was minimal. Interactive effects analysis showed that high desulphurization efficiency above 70% was achieved at a high stoichiometric ratio of 2.0 and a relatively low temperature of 140 °C. Examination of the desulphurization residue confirmed the presence of desulphurization products and unreacted CaCO_3 . The obtained desulphurization rates using limestone in the spray dryer were relatively lower, reaching a maximum of approximately 74%. However, these levels complied with the regulatory limit in South Africa, suggesting that limestone can be effectively employed as a sorbent in spray drying flue gas desulphurization. Further research is recommended to compare different sorbents for spray drying and conduct a detailed analysis of the desulphurization residue to quantitatively determine sorbent conversion.

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