Proceedings of the International Conference on Industrial Engineering and Operations Management

Publisher: IEOM Society International, USA DOI: 10.46254/EU08.20250247
Published: July 2, 2025

Dry Flue Gas Desulphurization using $NaHCO_3$ and Na_2CO_3 Sorbents in a Fixed Bed Reactor

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Abstract

This study investigated sulphur dioxide (SO_2) removal efficiency of sodium bicarbonate ($NaHCO_3$) and sodium carbonate (Na_2CO_3) sorbents using a fixed-bed reactor, relevant to dry flue gas desulphurization (FGD) technology. A Box-Behnken experimental design was used to optimize key operational parameters, including sorbent bed mass (3 - 7g), flue gas flow rate (0.6 - 1.0 l/min), and inlet flue gas temperature (140 - 180°C), to maximize desulphurization efficiency. A simulated flue gas mixture (21% CO_2, 4% O_2, 0.03% SO_2, 74.97% N_2) was heated before passing through the reactor containing the sorbent. $NaHCO_3$ sorbent achieved a higher SO_2 removal efficiency of up to 81%, compared to 66% for Na_2CO_3. $NaHCO_3$ maintained consistent performance across varying conditions, especially with changes in flue gas flow rates and temperature levels. Na_2CO_3 exhibited sensitivity to these variables, with reduced efficiency under low flue gas temperature or high flow rates. Based on the experimental findings, a predictive model was developed correlating the operational variables and SO_2 removal efficiency. The model validation indicated a strong fit to the experimental data with R^2 values exceeding 0.96 for both sorbents. The analysis of variance (ANOVA) identified flue gas temperature as the most significant factor influencing SO_2 removal, followed by bed mass capacity, while flue gas flow rate had minimal impact for both sorbent materials. These findings highlight the potential of $NaHCO_3$ and Na_2CO_3 as reliable sorbent materials for low-temperature dry FGD applications.

Keywords
Dry FGD, fixed bed reactor, SO_2, Na_2CO_3, $NaHCO_3$.

1 Introduction
Flue gas desulphurization (FGD) is a critical process for controlling sulphur dioxide (SO_2) emissions from industrial sources such as power plants, cement kilns, and waste incinerators. SO_2 is a major air pollutant responsible for acid rain formation and significant environmental and human health hazards (Saxena, 2025). Traditionally, wet FGD systems utilizing calcium-based sorbents, particularly limestone ($CaCO_3$) and hydrated lime ($Ca(OH)_2$), have been widely used due to their high efficiency in SO_2 removal (Koech et al., 2021; Kumar and Jana, 2022). However, these systems have several drawbacks, including high water consumption, generation of large amounts of wastewater requiring treatment, and complex maintenance demands (Córdoba, 2015; Ma et al., 2019). As a result, dry and semi-dry FGD technologies have gained interest as alternative solutions, offering operational simplicity, lower capital costs, and reduced secondary waste production.

Dry FGD involves injecting a dry or semi-dry sorbent into the flue gas stream, where it reacts with SO_2 to form a solid product that can be easily removed by particulate control devices such as bag filters or electrostatic precipitators (Dai et al., 2022; Ning et al., 2025). Although dry FGD process have lower SO_2 removal efficiencies (50 – 80%) compared to wet the FGD (95 – 99%), they use less water (typically 60% less) and do not require extensive wastewater handling, making them particularly suitable for small-to medium-scale industrial facilities and retrofitting applications (Carpenter, 2012; Koech et al., 2021). Among the dry sorbents used for SO_2 capture, calcium-based materials such as hydrated lime have been the most studied (Chang et al., 2023; Han et al., 2005; Renedo and Fernández-Ferreras, 2016; Xing et al., 2023). However, sodium-based sorbents, particularly sodium bicarbonate ($NaHCO_3$) and sodium carbonate (Na_2CO_3), have emerged as viable alternatives due to their superior reactivity and higher SO_2 capture capacities at moderate temperatures (Ma et al., 2024; Makomere et al., 2024a; Mchabe et al., 2024; Zhang et al., 2023).

The reaction mechanisms for sodium-based sorbents in dry FGD are well documented. $NaHCO_3$ undergoes thermal decomposition at around 120°C to form Na_2CO_3, CO_2, and H_2O (Lee et al., 2023; Makomere et al., 2024b). Thermal decomposition step increases the reactive surface area which facilitates SO_2 capture (Zhang et al., 2023). The resultant Na_2CO_3 then reacts with SO_2 to form sodium sulphate (Na_2SO_4). Similarly, Na_2CO_3 directly reacts with SO_2 to form Na_2SO_4 (Lee et al., 2023; Walawska et al., 2014). Previous studies have shown that sodium-based sorbents exhibit higher SO_2 removal efficiencies than calcium-based sorbents at comparable operating conditions, with the additional advantage of producing less solid waste per unit of SO_2 removed (Mchabe et al., 2024; Ning et al., 2025).

Several studies have explored the performance of sodium-based sorbents in various FGD configurations, including spray-drying and circulating dry scrubbers (Bahrabadi-Jovein et al., 2017; Keener and Davis, 1984; Koech et al., 2023; Omidi Bibalani and Ale Ebrahim, 2022a; Omidi Bibalani and Ale Ebrahim, 2022b). However, limited research has focused on their application in fixed-bed reactors, where contact time, sorbent utilization, and operational conditions can be optimized for enhanced performance. Fixed-bed systems provide a controlled environment that allows detailed investigation of reaction kinetics and parameter influences, which are critical for understanding sorbent behaviour under specific conditions.

The commercial grade $NaHCO_3$ and Na_2CO_3 sourced from Botswana Ash (Pty) Ltd have not previously been tested for SO_2 capture. This study therefore aims to evaluate their SO_2 removal performance in a fixed-bed reactor. Using a Box-Behnken statistical experimental design, key operational parameters such as sorbent bed mass, flue gas flow rate, and flue gas temperature were optimized to maximize SO_2 capture. The study focussed on direct comparison of $NaHCO_3$ and Na_2CO_3 under identical conditions, using statistical analysis to identify the most influential parameters. The findings of this study show the potential of commercial grade Na_2CO_3 and $NaHCO_3$ from Botswana Ash (Pty) Ltd as sorbents for low-temperature dry SO_2 removal applications.

2 Methods
2.1 Materials and Preparation
The materials utilized in this study included a flue gas mixture with an SO_2 concentration of 3800 ppm, obtained from Afrox, South Africa. The sorbent materials, commercial grade sodium bicarbonate ($NaHCO_3$) and sodium carbonate (Na_2CO_3), in powder form, were supplied by Botswana Ash (Pty) Ltd. The chemical composition of the sorbents, determined by XRD analysis, showed that Na_2CO_3 consisted of 87.1% thermonatrite, 2.8% natrite, with the remainder being amorphous material. $NaHCO_3$ was composed of 2.8% thermonatrite, 93.6% natrite, with the remainder also being amorphous.

The sorbents were received in a coarse powder form, necessitating milling to achieve the desired particle size. A ball mill was used to reduce the particle size, ensuring that the sorbents passed through a 45 µm sieve. The milled samples were then stored in airtight containers to preserve their quality and prevent contamination before desulphurization experiments.

2.2 Experimental Setup and Procedure
The equipment employed during the experiments comprised a fixed-bed reactor, air heater, flue gas analyser, and associated connections. The fixed-bed reactor was designed and fabricated to facilitate interaction between the flue gas and the sorbent under varying experimental conditions. The reactor, with an internal diameter of 5.27 cm and a height of 7.62 cm, was coupled with a flow meter to regulate the flow of flue gas entering the reactor. The reactor base contained perforations to ensure even distribution of gas as it entered the reactor. A schematic of the experimental setup is illustrated in Figure 1. The air heater was equipped with a temperature PID controller, allowing precise control of the inlet gas temperature. The setup also included a Testo 340 flue gas analyser, which continuously monitored the SO_2 concentration in the gas exiting the fixed-bed reactor.

Each experiment involved weighing a known mass of the milled sorbent and placing it inside the reactor. The reactor was thoroughly cleaned before each run to eliminate potential carryover from previous experiments. The synthetic flue gas was passed through the gas heater where it was heated to the desired temperature. Once thermal equilibrium was achieved, the inlet valve was opened, allowing the gas to enter the fixed-bed reactor, where it interacted with the sorbent material. The flow rate of the inlet gas was regulated using a flow meter.

The exit flue gas SO_2 concentration was continuously analysed using the Testo 340 gas analyser, and data was recorded. Each experiment was conducted for 5 minutes, although equilibrium readings were typically achieved within 3 minutes. All experiments were conducted in triplicates and average values were recorded. After each run, the spent sorbent was collected and stored for further analysis.

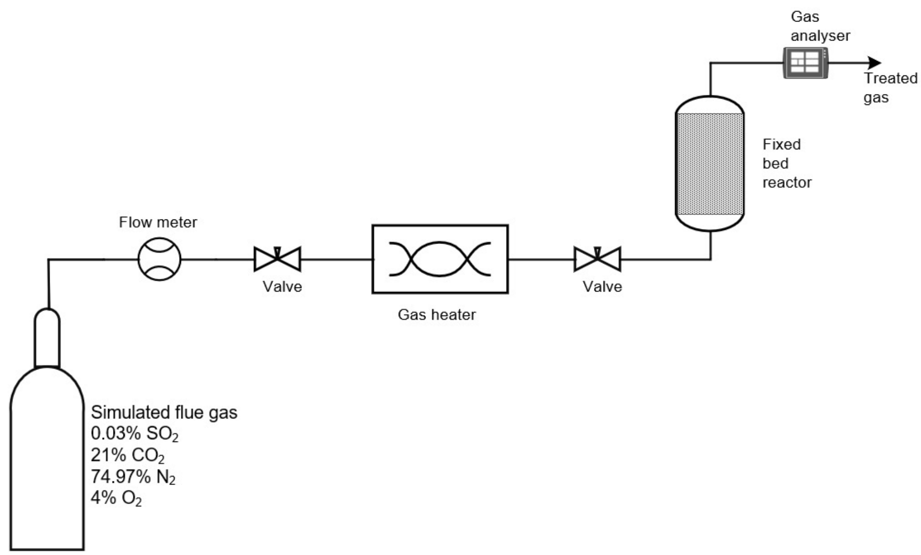

Figure 1. Schematic of the experimental setup

2.3 Design of Experiment

A Box-Behnken experimental design (BBD), a subset of response surface methodology (RSM), was employed to optimize and analyse the effects of key independent variables on the SO_2 removal efficiency of the sorbent materials. The experimental design was implemented using *Design Expert* software (Version 13.0.01.0). Three factors (flue gas temperature, x_1; sorbent bed mass, x_2; and flue gas flow rate, x_3) were selected as independent variables, each evaluated at three levels to determine their influence on the response variable (SO_2 removal efficiency). The selection of parameter levels was guided by preliminary experimental data to ensure meaningful variable interactions.

Table 1 presents the independent variables and their respective levels, while Table 2 outlines the full experimental design matrix for both sorbents, which consists of 15 runs based on three factors at three levels each. The experimental results for the respective sorbent were used to develop a second-order polynomial model for predicting desulphurization efficiency. The response surface model follows the quadratic equation:

$$\mathrm{Y} = b_0 + \sum_{i=1}^{n} b_i x_i + \sum_{i=1}^{n} b_{ii} x_i^{\,2} + \sum_{i=1}^{n-1} \sum_{j=i+1}^{n} b_{ij} x_i x_j \tag{1}$$

Where Y represents the predicted desulphurization efficiency (%), b_0 is the intercept, b_i are the linear coefficients, b_{ii} are the quadratic coefficients, and b_{ij} are the interaction coefficients of the independent variables.

The statistical significance of the polynomial model was evaluated through analysis of variance (ANOVA) and a lack-of-fit test. Model adequacy was assessed using the coefficient of determination (R^2), ensuring that the model accurately represents the experimental data.

Table 1. Independent variables and respective levels

Variable name	**Code**	$\alpha = -1$	$\alpha = 0$	$\alpha = 1$
Flue gas temperature (°C)	x_1	140	160	180
Sorbent bed mass (g)	x_2	3	5	7
Flue gas flow rate (l/min)	x_3	0.6	0.8	1

Table 2. Matrix of experiments for $NaHCO_3$ and Na_2CO_3

Run	x_1	x_2	x_3	**SO_2 removal efficiency (%)**	
				Na_2CO_3	**$NaHCO_3$**
1	160	7	0.6	56	69
2	180	5	0.6	56	72
3	140	7	0.8	47	61
4	140	5	1.0	40	51
5	160	5	0.8	54	63
6	180	7	0.8	66	81
7	140	5	0.6	36	49
8	180	3	0.8	54	65
9	180	5	1.0	55	67
10	160	5	0.8	48	63
11	160	3	1.0	40	53
12	160	3	0.6	45	59
13	160	5	0.8	53	63
14	160	7	1.0	53	62
15	140	3	0.8	30	41

3 Results and Discussion

3.1 Reaction Mechanisms

In this study, the reaction mechanisms for $NaHCO_3$ and Na_2CO_3 in a fixed bed reactor is schematized to follow the equations (2-4). At temperatures above 120°C, $NaHCO_3$ decomposes into Na_2CO_3, releasing CO_2 and H_2O (Keener and Davis, 1984; Shu et al., 2023).

$$2NaHCO_3 \rightarrow Na_2CO_3 + CO_2 + H_2O \tag{2}$$

Na_2CO_3 then reacts with SO_2 and O_2 to produce sodium sulphate (Na_2SO_4).

$$Na_2CO_3 + SO_2 + 1/2\,O_2 \rightarrow Na_2SO_4 + CO_2 \tag{3}$$

For Na_2CO_3, SO_2 removal occurs through direct reaction with SO_2, bypassing the decomposition step required for $NaHCO_3$.

$$Na_2CO_3 + SO_2 + 1/2\,O_2 \rightarrow Na_2SO_4 + CO_2 \tag{4}$$

3.2 Statistical Analysis and Model Evaluation and Validation

Following the analysis of experimental data presented in Table 2, two predictive quadratic models (Equations 5 and 6 for $NaHCO_3$ and Na_2CO_3, respectively) were developed using BBD to describe the relationship between the coded independent variables (x_1, x_2, x_3) and SO_2 removal efficiency (Y_1 and Y_2).

$$Y_1 = 63.00 + 10.37x_1 + 6.87x_2 - 2x_3 - x_1x_2 - 1.75x_1x_3 - 0.25x_2x_3 - x_1^2 - 2.25x_3^2 \quad (5)$$

$$Y_2 = 51.67 + 9.75x_1 + 6.62x_2 - 0.63x_3 - 1.25x_1x_2 - 1.25x_1x_3 + 0.5x_2x_3 - 2.08x_1^2 - 0.33x_2^2 - 2.83x_3^2 \quad (6)$$

Y_1 and Y_2 are the SO_2 removal efficiencies for $NaHCO_3$ and Na_2CO_3, respectively.
To evaluate the effects of these independent variables on SO_2 removal efficiency, the model coefficients for Equations 5 and 6 were compared. The statistical significance of the predictive models and their individual terms was assessed through ANOVA. Table 3 and Table 4 present the ANOVA results, including the significance of model terms and the lack-of-fit analysis for both $NaHCO_3$ and Na_2CO_3 models. The ANOVA was conducted to assess the statistical significance of factors influencing SO_2 removal efficiency for $NaHCO_3$ and Na_2CO_3 sorbents. The results indicate that flue gas temperature (x_1) and sorbent bed mass (x_2) are the most significant factors affecting SO_2 removal efficiency, with p-values well below 0.05, confirming their strong influence on the model. The effect of flue gas flow rate (x_3) varies between the two sorbents. x_3 is statistically insignificant ($p > 0.05$) for both $NaHCO_3$ and Na_2CO_3, suggesting a minimal impact on SO_2 removal efficiency. Interaction terms, such as x_1x_2 and x_1x_3, are generally insignificant for both sorbents, indicating that the main effects dominate over interactive contributions. Comparatively, $NaHCO_3$ demonstrates slightly better statistical performance, with higher sums of squares for its significant factors under the studied conditions.

Table 3. ANOVA for $NaHCO_3$

Source	Sum of Squares	Mean Square	F-value	p-value
Model	1309.18	145.46	14.05	0.0048
x_1	861.12	861.12	83.20	0.0003
x_2	378.12	378.12	36.53	0.0018
x_3	32.00	32.00	3.09	0.1390
x_1x_2	4.00	4.00	0.39	0.5614
x_1x_3	12.25	12.25	1.18	0.3263
x_2x_3	0.25	0.25	0.02	0.8826
x_1^2	3.69	3.69	0.36	0.5763
x_2^2	0.00	0.00	0.00	1.0000
x_3^2	18.69	18.69	1.81	0.2367

Table 4. ANOVA for Na_2CO_3

Source	Sum of Squares	Mean Square	F-value	p-value
Model	1170.82	130.09	13.86	0.0049
x_1	760.50	760.50	81.05	0.0003
x_2	351.12	351.12	37.42	0.0017
x_3	3.13	3.13	0.33	0.5889
x_1x_2	6.25	6.25	0.67	0.4515
x_1x_3	6.25	6.25	0.67	0.4515
x_2x_3	1.00	1.00	0.11	0.7573
${x_1}^2$	16.03	16.03	1.71	0.2481
${x_2}^2$	0.41	0.41	0.04	0.8426
${x_3}^2$	29.64	29.64	3.16	0.1357

The accuracy of the predictive models (equations 5 and 6) was evaluated by comparing the model-predicted values with the corresponding experimental data. Figure 2 presents the regression plots for Na_2CO_3 (Figure 2a) and $NaHCO_3$ (Figure 2b), where the predicted SO_2 removal efficiencies are plotted against the observed experimental values. The strong alignment of data points along the 45-degree reference line indicates high predictive reliability. Both models exhibit a high coefficient of determination (above $R^2 = 0.96$), confirming that the models account for nearly all variability in SO_2 removal efficiency.

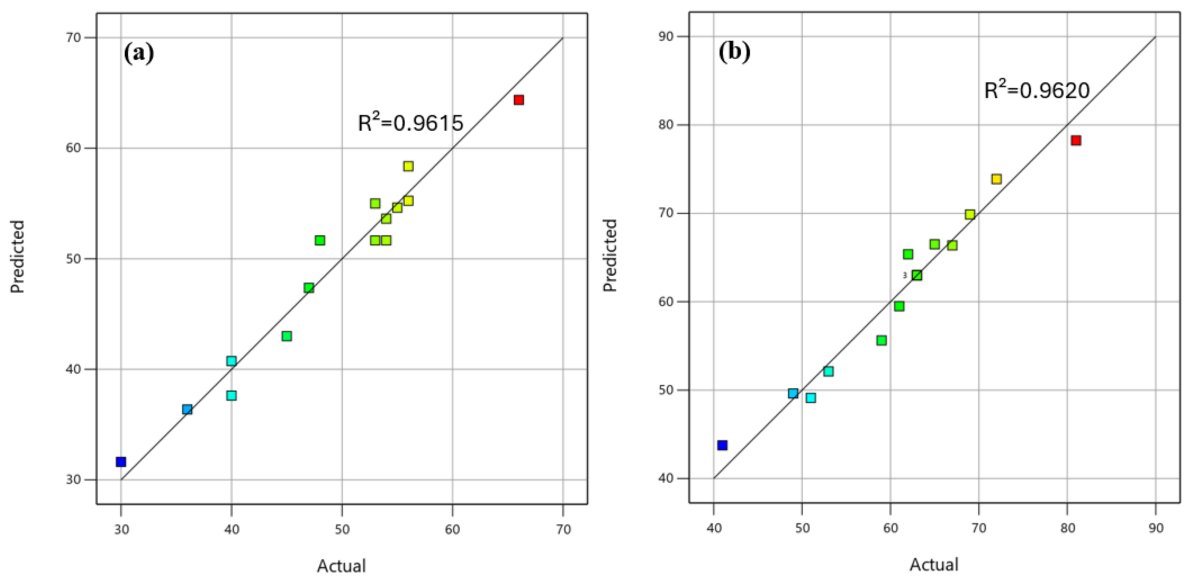

Figure 2. Plot of predicted vs experimental data for (a) Na_2CO_3 and (b) $NaHCO_3$.

3.3 Effect of Experimental Variables

The 3D surface plots in Figure 3 illustrate the interactive effects of flue gas temperature (x_1) and bed mass capacity (x_2) on SO_2 removal efficiency for $NaHCO_3$ and Na_2CO_3. For $NaHCO_3$ (Figure 3a), SO_2 removal efficiency reaches approximately 78% at a bed mass capacity of 7g and flue gas temperature of 180°C. The observed increase in efficiency with flue gas temperature and bed mass capacity is attributed to the thermal decomposition of $NaHCO_3$ into Na_2CO_3, CO_2, and H_2O, which enhances the reactive surface area for SO_2 capture (Zhang et al., 2023). The released water vapor and CO_2 induce a popcorn effect, creating a porous structure within the particle. This results in a substantial increase in surface area, typically ranging from 5 to 20 times the original value. (Carpenter, 2012).

For Na_2CO_3 (Figure 3b), a similar trend is observed, with efficiency reaching approximately 65% under the same conditions. The higher SO_2 removal efficiency of Na_2CO_3 is attributed to its direct reaction with SO_2, without requiring an initial decomposition step. The decline in removal efficiency at lower flue gas temperature, evident from the downward slope of both plots, is due to poor conversion at lower temperatures. Studies show that the conversion of $NaHCO_3$ and Na_2CO_3 increases with temperature, reaching a peak around 232°C before declining (Keener and Davis, 1984). Both plots show that increasing bed mass capacity results in higher SO_2 removal efficiency. In both cases, lower bed mass capacity leads to rapid sorbent depletion, reducing desulphurization efficiency. The observed trend suggests that the optimal bed capacity was not achieved within the tested range, as indicated by the near-linear increase in SO_2 removal efficiency with increasing bed mass capacity.

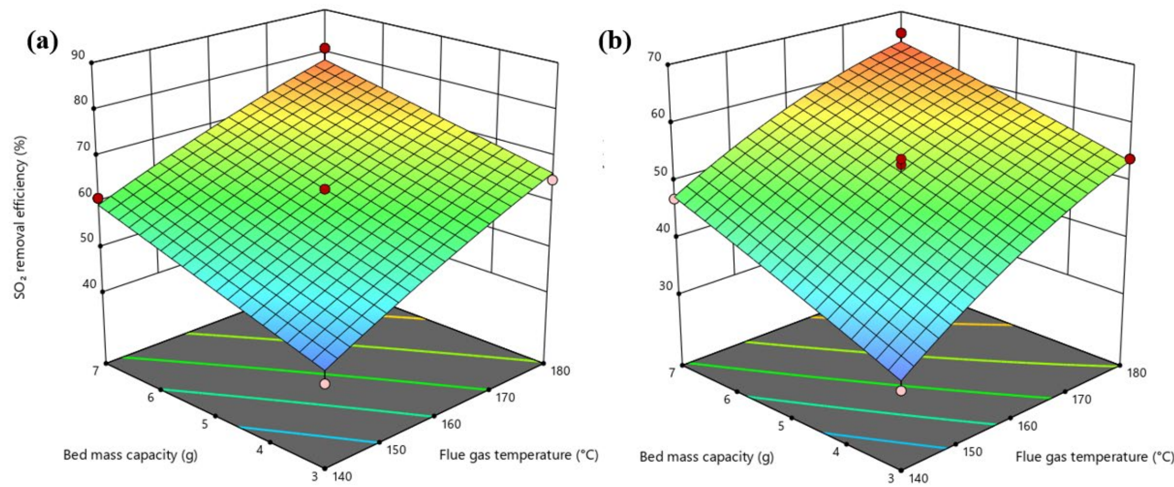

Figure 3. 3D surface plot illustrating the interactive effects of flue gas temperature and bed mass capacity on SO_2 removal efficiency for (a) $NaHCO_3$ and (b) Na_2CO_3.

The interactive effects of flue gas flow rate (x_3) and flue gas temperature (x_1) on SO_2 removal efficiency for $NaHCO_3$ and Na_2CO_3 is presented in Figure 4. $NaHCO_3$ achieved a maximum removal efficiency of approximately 72%, while Na_2CO_3 reaches about 58%, both at optimal conditions (0.6 l/min flow rate and 180°C). The minimum removal efficiency for $NaHCO_3$ is around 49%, observed at low flue gas temperature (140°C) and a higher flow rate (1 l/min). Similarly, Na_2CO_3 shows a minimum efficiency of about 38% under the same conditions. Increasing flue gas temperature enhances SO_2 removal for both sorbents by promoting conversion of $NaHCO_3$ to Na_2CO_3 which significantly improves the surface area of the sorbent material especially for $NaHCO_3$. However, higher flue gas flow rates reduce residence time, limiting gas - sorbent interaction and decreasing efficiency. It is evident from the plots that flue gas flowrate has minimal effect on the removal efficiency of SO_2. This confirms the statistical analysis which indicated flue gas flowrate as statistically insignificant.

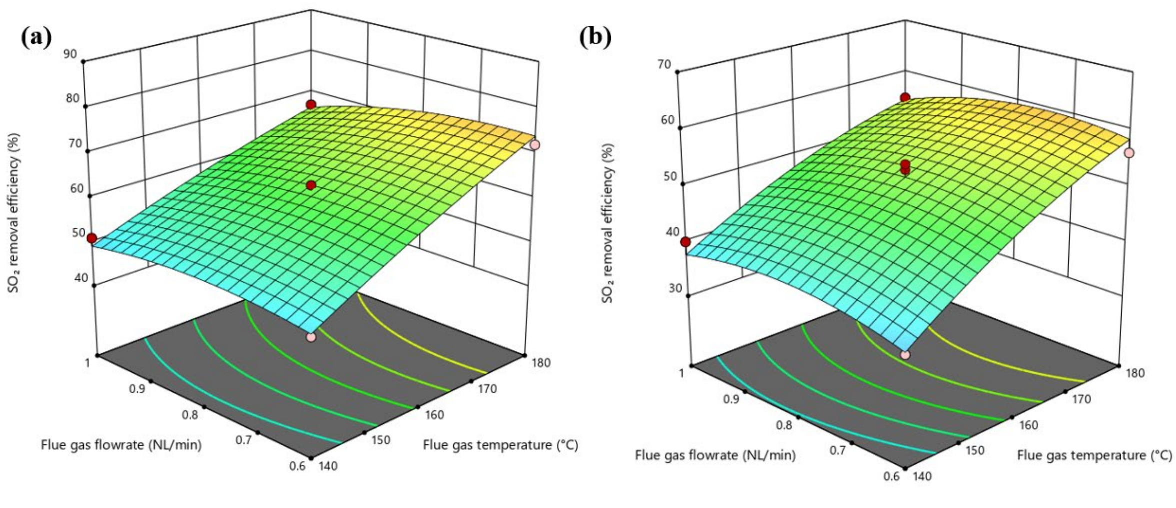

Figure 4. 3D surface plot illustrating the interactive effects of flue gas temperature and flue gas flowrate on SO_2 removal efficiency for (a) $NaHCO_3$ and (b) Na_2CO_3.

The 3D surface plots in Figure 5 illustrate the interactive effects of bed mass capacity (x_2) and flue gas flow rate (x_3) on SO_2 removal efficiency for $NaHCO_3$ and Na_2CO_3. In both cases, an increase in bed mass capacity enhances SO_2 removal efficiency due to the greater availability of active sorption sites. At the highest bed mass capacity (7 g), $NaHCO_3$ reaches a removal efficiency of approximately 70%, while Na_2CO_3 achieves about 55%. At the lowest bed mass capacity (3 g), efficiencies decrease to around 50% and 40%, respectively, as fewer reactive sites are available for SO_2 capture. Both variables x_2 and x_3, have minimal impact on SO_2 removal efficiency. This is evident from the slight increase in SO_2 removal efficiency with increasing bed mass capacity and the negligible change observed with variations in flue gas flow rate. These findings suggest that the interactive effects of these two independent variables are minimal.

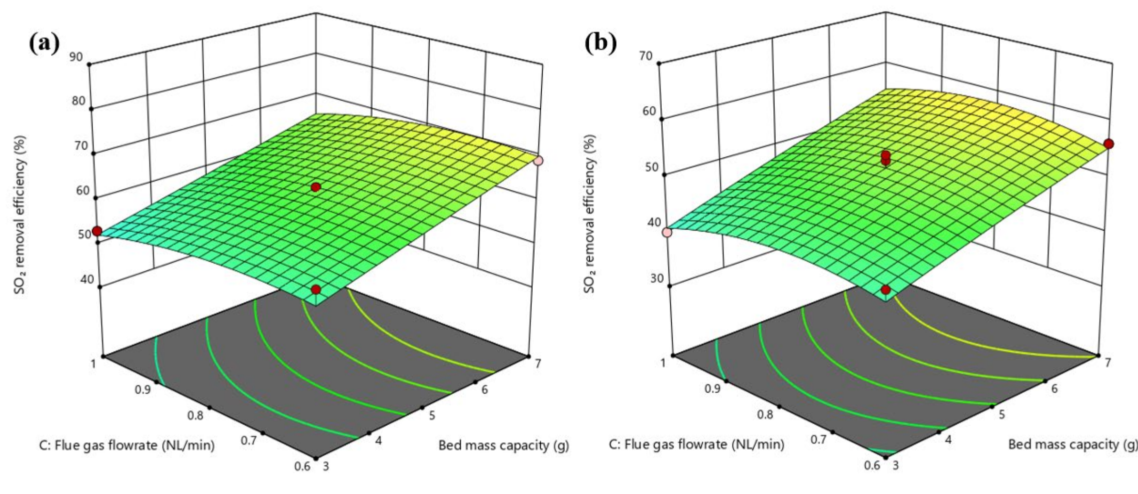

Figure 5. 3D surface plot illustrating the interactive effects of flue gas flowrate and bed mass capacity on SO_2 removal efficiency for (a) $NaHCO_3$ and (b) Na_2CO_3.

3.4 Perturbation Plots

The perturbation plots in Figure 6 illustrate the influence of the variables x_1, represented by A, x_2, represented by B, and x_3, represented by C on SO_2 removal efficiency for Na_2CO_3 (a) and $NaHCO_3$ (b). For Na_2CO_3, variables A and B exhibit a positive linear correlation with SO_2 removal efficiency, increasing from approximately 40% to 60%. In contrast, variable C follows a non-linear trend, peaking at around 50% near the reference point before declining.

For $NaHCO_3$, the removal efficiency is generally higher across all variables, reaching approximately 73%, 70%, and 58% for A, B, and C, respectively. This indicates that $NaHCO_3$ achieved greater SO_2 removal under the tested conditions. The comparison suggests that Na_2CO_3 is more sensitive to variations in A and B, necessitating precise control for optimal performance.

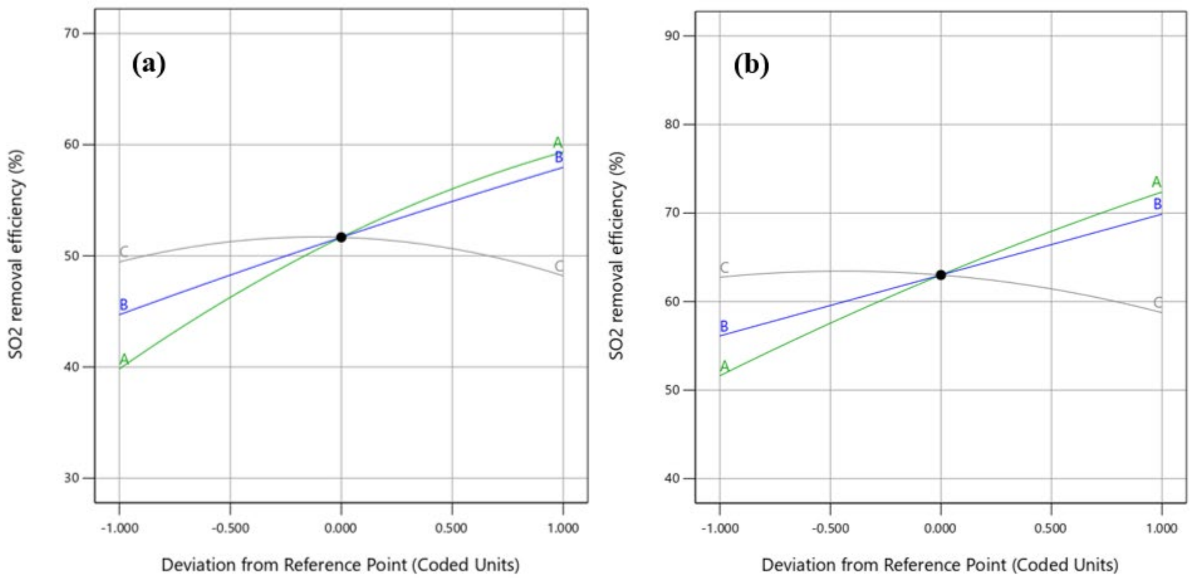

Figure 6. Perturbation plots for (a) Na_2CO_3 and (b) $NaHCO_3$.

4 Conclusions

This study investigated the SO_2 removal efficiency of sodium bicarbonate ($NaHCO_3$) and sodium carbonate (Na_2CO_3) in a fixed-bed reactor under varying operational conditions. The results showed that $NaHCO_3$ achieved a higher maximum removal efficiency of 81%, compared to 66% for Na_2CO_3, attributed to its thermal decomposition, which enhances reactivity. Statistical analysis using a Box-Behnken statistical experimental design revealed that flue gas temperature was the most significant factor influencing SO_2 removal, followed by bed mass capacity, while flue gas flow rate had minimal impact. The interactive effects of process variables indicated that increasing bed mass and flue gas temperature improved SO_2 removal efficiency due to increased reactive sites and enhanced sorbent activation. On the other hand, higher flue gas flow rates reduced residence time, leading to lower efficiency. Perturbation analysis further confirmed that $NaHCO_3$ exhibited stronger dependence on operational variables, requiring precise control for optimal performance, whereas Na_2CO_3 exhibited more stable but lower removal efficiency. These findings establish the potential of sodium-based sorbents for dry FGD applications, with $NaHCO_3$ emerging as the superior sorbent under the studied conditions i.e., sorbent bed mass (3 - 7g), flue gas flow rate (0.6 - 1.0 l/min), and inlet flue gas temperature (140 - 180°C). Future work is intended to focus on kinetic modelling, and the impact of flue gas composition on sorbent performance to further optimize their application in industrial desulphurization systems.

Funding

This work was supported by the National Research Foundation (NRF) – South Africa (Grant number: PSTD240502217083).

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