Published by Avanti Publishers

Journal of Chemical Engineering Research Updates

ISSN (online): 2409-983X

Process Simulation of Wet Flue Gas Desulfurization

Changhong Li, Baiqiang Zhang*, Junhui Li and Yanhao Hu

Henan International Joint Laboratory of Energy, Efficient Conversion and Utilization, School of Energy, and Power Engineering, Zhengzhou University of Light, Industry, PR China

ARTICLE INFO

Article Type: Research Article

Academic Editor: Wujie Zhang

Keywords:
Simulation
Aspen plus
Calcium-sulfur ratio
Optimization analysis
Wet flue gas desulfurization

Timeline:
Received: October 30, 2024
Accepted: December 03, 2024
Published: December 19, 2024

Citation: Li C, Zhang B, Li J, Hu Y. Process simulation of wet flue gas desulfurization. J Chem Eng Res Updates. 2024; 11: 80-90.

DOI: https://doi.org/10.15377/2409-983X.2024.11.5

ABSTRACT

During combustion in power plants, sulfur in coal forms SO_2, a key air pollutant causing acid rain. Denitrification of SO_2 in exhaust gases is crucial, and simulation is a practical research approach. This article applies Aspen Plus software to simulate and optimize the limestone-gypsum wet flue gas desulfurization process. The results show that the established model can effectively reduce SO_2 content, achieving a desulfurization rate of 95.9%, which verifies the feasibility of the process flow. Through sensitivity analysis and orthogonal experiments, it is found that the inlet temperature of flue gas, calcium-sulfur ratio, and water content in limestone slurry are the key factors affecting the desulfurization efficiency. The optimal operating parameter combination is an inlet temperature of flue gas of 80°C, a calcium-sulfur ratio of 1.03, and water content in limestone slurry of 35 kmol/hr, with the calcium-sulfur ratio having the most significant impact on desulfurization efficiency. The study indicates that the combination of this software and the process has good application prospects.

*Corresponding Author
Email: zhangbq@zzuli.edu.cn
Tel: +(86) 15298391587

1. Introduction

During combustion in thermal power plants, most of the combustible sulfur in coal is oxidized to SO_2 at high temperatures [1]. The main components of untreated flue gas from thermal power plants typically include hydrogen halides, water vapor, N_2, CO, SO_2, hydrocarbons, CO_2, O_2, HCN, H_2S, NH_3, solids, as well as oxides of nitrogen, dust, and soot [2]. SO_2 is a major air pollutant that can cause acid rain, which can cause serious damage to the environment, including acidification of forests, lakes, and soils. It is harmful to human health and can cause respiratory problems such as bronchitis and asthma, and prolonged exposure may also increase the risk of heart disease [3]. Therefore, flue gas desulfurization (FGD) in coal-fired power plants is particularly important. Different countries have varying emission requirements for flue gas exports. For example, China requires coal-fired units to emit sulfur dioxide concentration of no more than 35mg/Nm^3, to achieve ultra-low emissions [4]; and the U.S. federal CAA through the National Ambient Air Quality Standards, SO_2 national ambient air quality standards for 75 ppb [5]. Flue gas desulfurization technology in large-scale thermal power plants is diverse. According to the forms of FGD products in power plants, FGD can be classified into three major categories: wet, dry, and semi-dry [6]. Dry FGD technology has relatively low desulphurization efficiency and may not be able to achieve the desired desulphurization effect, especially when dealing with flue gases with high sulphur content [7], semi-dry FD has lower removal efficiency and higher operating costs [8]. Wet Flue Gas Desulfurization (WFGD) technology, a vapor-liquid reaction, is widely used internationally in large-scale thermal power plants due to its fast reaction speed, high desulfurization efficiency, simple operation, and relatively mature technology [9]. Limestone–gypsum WFGD technology removes sulfur from the flue gas and produces gypsum for resource recovery [10], the first choice for the fossil fuel power generation industry [11]. WFGD technology has become the mainstream choice due to its high efficiency and maturity[12], but the model simplification under the complex reaction mechanism still needs further validation.

Aspen Plus software, known for its powerful and complete standard large-scale process simulation capabilities, is the world’s most widely used chemical simulation software. The application of Aspen Plus to the simulation of the overall wet flue gas process has been reported. Aspen Plus can model material and energy flows in unit operations such as reactors, separators, and heat exchangers, assisting engineers in analyzing process performance under varying conditions and optimizing energy consumption and cost efficiency [13]. Anyu *et al*. [14] evaluated the performance of various WFGD configurations by integrating advanced computational technologies with WFGD processes. Their study demonstrated that a four-layer spray system achieves significantly higher particulate capture efficiency than a two-layer configuration. These findings hold substantial implications for optimizing WFGD technology in coal-fired power plants to reduce emissions and support sustainable industrial development. Jiang *et al*. [15] developed a method for simulating a magnesium-based WFGD system with circulating water using Aspen Plus and validated this method. The validation results show that for an 80-ton/hour coal-fired boiler, the maximum recoverable waste heat from the WFGD system ranges from 1.41 to 2.27 megawatts, equivalent to saving 1385 to 2230 tons of standard coal annually. After recovering the waste heat, the temperatures of all flow streams decreased, with different streams showing varying trends. Additionally, water consumption and desulfurization efficiency improved. Zhang *et al*. [16] developed a cryogenic desulfurization process flow based on Aspen Plus. Using the corresponding property methods in Aspen Plus, they simulated the steady-state process and established a two-stage cryogenic desulfurization experimental platform to conduct process trials. The study systematically examined the effects of temperature, pressure, SO_2 concentration, and water vapor volume fraction on the desulfurization behavior. Through a combination of simulation results and practical engineering considerations, they determined the optimal operating conditions for the process.

Due to Aspen Plus limitation in modeling the desulfurization towers, which are central to FGD processes [17], there is limited literature available on the simulation of desulfurization towers. In this paper, Aspen Plus is used as the foundation for modeling the limestone-gypsum method of the WFGD process. By simplifying the process flow of the SO_2 absorber tower section and employing the software's MIXER model to simulate the desulfurization tower, the study provides a more intuitive analysis of the impact of various factors on the limestone-gypsum WFGD process. The aim is to contribute valuable insights for the future development of WFGD technologies, focusing on improving the WFGD process.

2. Experimental and Methods

2.1. Overall Process Flow Modeling

The limestone-gypsum WFGD process was simulated using Aspen Plus software. As shown in Fig. (**1**), the hot flue gas from the gas boiler enters the flue gas heat exchanger, which is cooled before being directed into the absorption section of the desulfurization tower. The flue gas ascends from the base of the absorption tower, countering the downward flow of the limestone slurry, which is sprayed from the upper layers to effectuate the cleaning process in a countercurrent mode to remove the solid particulate matter in the flue gas. Within the spray absorption tower, SO_2 in the flue gas reacts with water to form sulfurous acid, which reacts with calcium hydroxide in limestone to form calcium sulfite and water, followed by further oxidation of the calcium sulfite to calcium sulfate (gypsum) [18]. The reaction equations are shown in i. to iv.

i. SO_2 dissolution:

$$SO_2 + H_2O \rightarrow H_2SO_3 \rightarrow 2H^+ + SO_3^{2-}$$

ii. Neutralization with calcium carbonate:

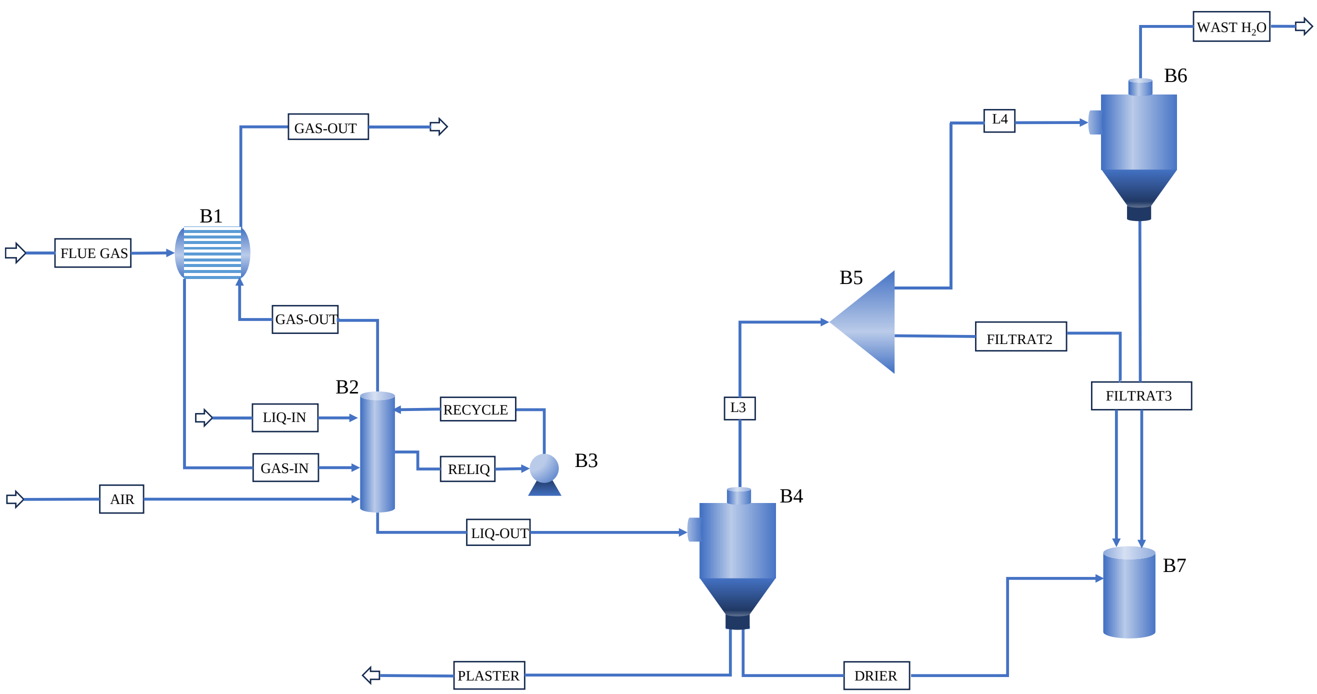
$$CaCO_{3(s)} + 2H^+ \rightarrow Ca^{2+} + CO_{2(g)} + H_2O$$

iii. Formation of calcium sulfite:

$$Ca^{2+} + SO_3^{2-} + 2H_2O \rightarrow CaSO_3 \bullet 2H_2O_{(s)}$$

iv. Oxidation to gypsum:

$$2CaSO_3 \bullet 2H_2O + O_2 \rightarrow 2CaSO_4 \bullet 2H_2O_{(s)}$$



B1: Flue gas heat exchanger B2: Absorption tower B3: Gypsum pumps B4: Gypsum cyclone
B5: Mixing separator B6: Industrial wastewater cyclone B7: Storage pool

Figure 1: Limestone-gypsum method FGD system overall process flow diagram.

After this reaction, the gypsum-laden slurry is extracted and conveyed to a cyclone for dewatering. A preliminary wastewater separation is conducted via a mixing separator, which streamlines the segregation process before the effluent is channeled into an industrial wastewater cyclone for additional separation. The refined water is then purified in specialized units and stored in a dedicated process reservoir, ensuring its utility for reuse. Gypsum gradually deposits in the desulfurization tower and is periodically removed through liquid level control and slag discharge systems. It can be further processed and utilized as a byproduct. The desulfurized flue gas undergoes treatment via a demister to remove moisture before being chilled at the tower's outlet, where it becomes imbued with steam. After that, the flue gas is reheated to a temperature exceeding 80°C through a heat exchanger. Ultimately, the treated flue gas is emitted into the atmosphere via the flue gas conduit, which leads to the boiler chimney.

2.2. Methodology

In the FGD system, the absorption tower serves as the core of the process, with all reactions occurring within the tower. Therefore, the design of the absorption tower' is central to optimizing desulfurization technology. To simplify the overall process into the model shown in Fig. (**2**), the following assumptions are made:

(1) It is assumed that the desulfurization process is operating under steady-state conditions.

(2) It is assumed that the flue gas composition consists of CO_2, SO_2, H_2O, N_2, and O_2, with only CO_2 and SO_2 participating in the reactions and the other components having no impact on the desulfurization process.

(3) It is assumed that there is no pressure loss within the absorption tower.

(4) It is assumed that the absorption section only involves absorption without oxidation, and the oxidation section involves only oxidation without absorption.

Based on these assumptions, the limestone-gypsum wet flue gas desulfurization process is simplified, and the absorption section of the process flow is modeled using the MIXER module in Aspen Plus. The MIXER module in Aspen Plus is a simplified unit operation model that assumes the complete mixing of input fluids under steady-state conditions with no pressure drop or heat exchange. It is commonly used to simulate fast mixing processes such as absorption or neutralization reactions. The absorption process involves gas-liquid contact and rapid chemical reactions between sulfur dioxide and limestone slurry. The MIXER module captures instantaneous mixing and reaction kinetics under steady-state assumptions.

The simulated inlet flue gases and contents are based on typical coal combustion exhaust gases, as shown in Table **1**. At the same time, the non-reactive components (nitrogen and oxygen) have been excluded from the reaction calculations. The mixer model represents the absorption tower (B1). The process flow is simplified as shown in Fig. (**2**). The limestone slurry enters the absorption tower from the top in Fig.(**1**), simplified as the LIQ-IN pipeline in Fig. (**2**). The coal-fired flue gas enters the absorption section of the tower from the bottom, simplified as the GAS-IN pipeline in Fig. (**2**). The slurry and flue gas react in the mixer to form $CaSO_3{\cdot}2H_2O$. The reacted slurry flows into the oxidation zone of the absorption tower, where it is oxidized and converted to gypsum.

Table 1: Flue gas composition and substance content [19].

	Component	Quantity	Unit (of Measure)
Flue Gas Composition	Flue gas quantity	760000	Nm^3/h
	N_2	74.07	Vol%
	O_2	1.79	Vol%
	CO_2	15.49	Vol%
	H_2O	8.58	Vol%
	SO_2	1546	Mg/Nm^3
	NO_X	0.013	Vol%

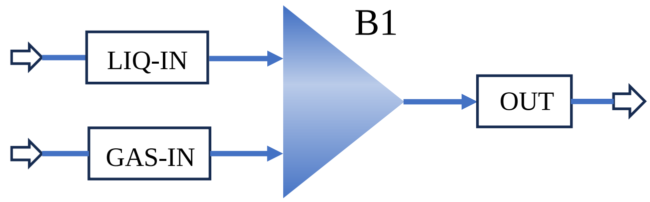


Figure 2: Simplified absorption model. B1 represents the mixer (which substitutes the absorption tower to simulate the absorption process).

In the SO_2 absorption process, it involves ion reactions and the formation of salts [20]. The electrolyte NRTL activity coefficient model (ELECNRTL model) is selected as the overall physical property model based on specific thermodynamic parameters to simulate the system [21]. This study primarily investigates the effects of the inlet flue gas temperature, ratio of calcium to sulfur (Ca/S), slurry inlet free water flow rate, and the sulfur dioxide content at the desulfurization tower inlet on the sulfur dioxide absorption process. The following parameters are set for the simulation: the flue gas inlet temperature is 80°C, pressure is 1 bar, and molar flow rate is 18.359 kmol/hr. The circulating slurry inlet parameters are set to a temperature of 20°C, pressure of 1 bar, and molar flow rate of 52.77 kmol/hr.

3. Results and Discussion

Based on the simplified model of the absorption tower outlined above, simulations were conducted under diverse assumptions to explore various scenarios. The operation and results are shown in Table **2**.

Table 2: Operation and results.

Item/(Units)		GAS-IN	LIQ-IN	OUT
Temperature/(℃)		80	20	81.45
Pressure/(bar)		1	1	1
Mole Flows/(kmol/hr)		18.36	52.77	62.34
Mass Flows(kg/hr)		1176.17	2394.31	3570.48
Volume Flow(cum/hr)		534.95	1.28	1085.91
Enthalpy/(Gcal/hr)		-1.29	-7.48	-8.77
Mole Flow / (kmol/hr)	SO_2	18.36	0	0.76
	Water	0	35.18	26.38
	CO_2	0	1.06e-08	17.59
Mass Flow / (kmol/hr)	SO_2	1176.17	0	48.99
	Water	0	633.78	475.29
	CO_2	0	4.66e-07	774.13

Based on the operational data presented in Table **2**, it can be observed that under the specified conditions, the flue gas outlet temperature is 81.45°C. Moreover, the pressure remains constant both before and after the mixing reaction. The molar flow rate of H_2O diminishes, while that of CO_2 experiences a significant increase. This indicates that SO_2 in the slurry is being absorbed, and CO_2 gas is released as a byproduct. The gas volume flow rate rises from 534.95 cum/hr to 1085.91 cum/hr. The sulfur dioxide content is reduced from 18.36 kmol/hr at the inlet to 0.76 kmol/hr at the outlet, achieving a desulfurization efficiency of 95.9%. The simulated process flow model exhibits excellent performance in absorbing sulfur dioxide, successfully achieving the desired outcome. The process conditions and flow scheme are indeed feasible and highly effective for desulfurization, ensuring a robust and efficient system operation.

3.1. Sensitivity Analysis of Key Parameters

The Sensitivity Analysis module in Aspen Plus is a powerful tool for determining the effect of altering operating conditions on the process [22]. One or multiple of the process conditions can be chosen to modify and analyze the effect on the other process conditions. The altered condition is referred to as the manipulated variable, while the process variable affected by this manipulation is known as the target variable. The influence of each parameter on the target variable can be determined efficiently, and the process operating conditions can be optimized using sensitivity analysis. The effect of different operating conditions on the system desulfurization efficiency was examined by changing the operating variables, and the appropriate operating conditions were analyzed for each condition. The final results are plotted on a graph, which can make the relationship between the different variables more transparent (as presented in Fig. **3**).

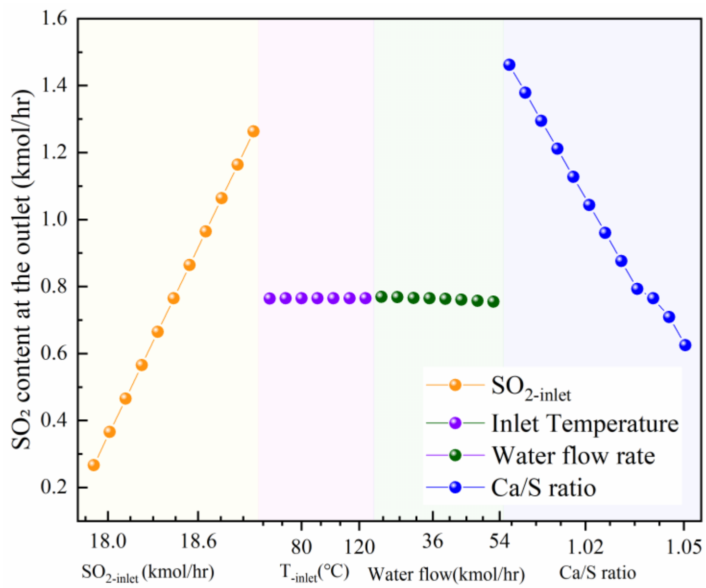


Figure 3: The influence of various elements on the concentration of SO_2 at the outlet.

3.1.1. Effect of Inlet SO_2 Content of Flue Gas

Since the actual operation of the equipment, changes in the operating environment and conditions will affect the SO_2 content in the flue gas. To explore the impact of changes in the SO_2 content in the imported flue gas on the effect of desulfurization, a sensitivity analysis of the changes in the inlet SO_2 content of the flue gas was established using Aspen Plus. The inlet flue gas parameters were kept unchanged except for the SO_2 content, and then manipulated project parameter settings were adjusted. The inlet content of SO_2 in the flue gas was changed in the range of 16~19 kmol/hr and the change of SO_2 content at the outlet of the absorber tower were analyzed to explore the desulphurization efficiency of the limestone-gypsum wet flue gas desulphurization method. As analyzed in Fig. (**3**), when the flue gas inlet molar flow rate increases, the exit SO_2 content increases, which indicates a decrease in the flue gas desulfurization efficiency. This is because when the SO_2 content increases, the absorber saturates its absorption capacity, causing SO_2 to flow out with the flue gas, decreasing the efficiency of flue gas desulfurization.

3.1.2. Effect of Calcium-Sulfur Ratio

The calcium-sulfur ratio (Ca/S) measures the molar ratio of desulfurizer to SO_2 removed in limestone desulfurization. It indicates the excess calcium needed for a target FGD efficiency and calcium utilization. To study its effect on WFGD using the limestone-gypsum method, a sensitivity analysis was conducted in Aspen Plus, with the ratio ranging from 1.02 to 1.05. According to Fig. (**3**), a distinct negative linear correlation is evident between the calcium-sulfur ratio in the feed and the SO_2 content at the outlet. This phenomenon can be attributed to the fact that an increase in the quantity of desulfurization agents leads to a rise in the pH value of the slurry, thereby enhancing the chemical rate of the absorption reaction. Consequently, the SO_2 content continues to decline, resulting in improved flue gas desulfurization efficiency. In practical production, although increasing the calcium-sulfur ratio indeed augments the rate of flue gas desulfurization, it is imperative to maintain this ratio within reasonable limits. Exceedingly high ratios can lead to the wastage of raw materials and fail to further enhance flue gas desulfurization efficiency.

3.1.3. Effect of Flue Gas Inlet Temperature

The inlet temperature of flue gas in limestone-gypsum desulfurization affects efficiency and equipment health. An Aspen Plus study analyzed the impact of inlet temperatures from 60 to 120℃ at 10℃ intervals on SO2 absorption to optimize operations. The goal was to identify an ideal temperature for effective desulfurization without compromising equipment performance. From Fig. (**3**), it can be seen that as the temperature changes, the overall outlet concentration tends to stabilize. In order to see the changing trend in detail, this part of the data has been enlarged and plotted, as shown in Fig. (**4**). The inlet temperature of flue gas significantly impacts the efficiency of limestone-gypsum desulfurization. High temperatures increase heat loss and corrosion risks, while low temperatures reduce SO_2 absorption. Aspen Plus simulations revealed that as the inlet temperature increases from 60 to 120°C, the outlet SO_2 concentration rises less, indicating lower desulfurization efficiency. Considering equipment performance and resource utilization, the optimal operating temperature is around 80°C. Adjusting the slurry flow helps maintain the flue gas temperature within this optimal range for improved absorption and high desulfurization efficiency.

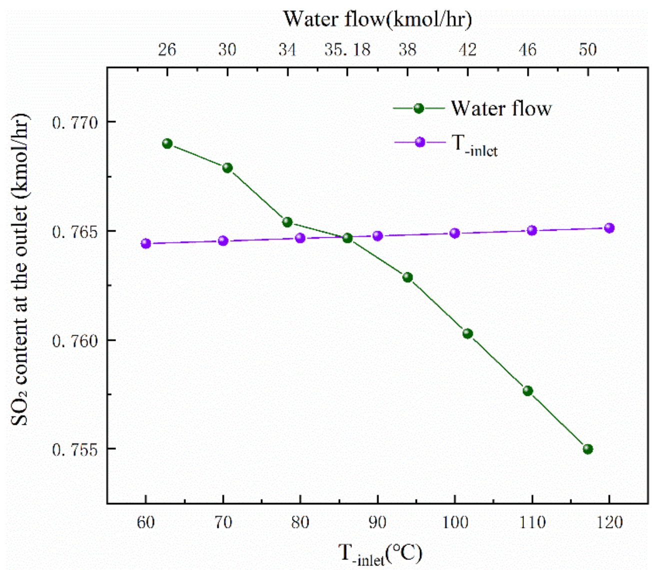


Figure 4: Effect of flue gas inlet temperature and water flow rate on SO_2 content at flue gas outlet.

3.1.4. Effect of Water Content in Limestone Slurry

In a sensitivity study with constant process parameters, the water content of the limestone slurry was varied from 25 to 50 kmol/hr. This adjustment aimed to observe the impact on SO_2 outlet levels, thereby assessing the absorber's efficiency. Free water in slurries refers to unbound moisture capable of dissolving SO_2 to drive ionic reactions during desulfurization, excluding chemically bound water in calcium carbonate ($CaCO_3$) or calcium sulfate dihydrate ($CaSO_4 \cdot 2H_2O$). This mobile water phase is essential for facilitating sulfur removal processes. The results, plotted in Fig. (**4**), reveal the influence of slurry water content on absorption effectiveness. From Fig. (**4**), it can be seen that the water content in the limestone slurry is inversely proportional to the SO_2 content at the outlet, i.e., the water content in the slurry is directly proportional to the flue gas desulfurization efficiency. This is because in an actual flue gas desulfurization system, the percentage of water carried by the slurry is directly related to the concentration value of the slurry in the desulfurization tower. The main chemical composition of the slurry consists of calcium carbonate, calcium sulfite, and insoluble hydrochloric acid. The concentration value of the slurry is too high, which will lead to a smaller area of contact between calcium sulfite and oxygen, and when the concentration of the slurry is increased to saturation, resulting in the saturation of the absorber, which will prevent the contact between SO_2 and calcium carbonate in the limestone slurry, and then lead to a decrease in the pH value of the slurry when the SO_2 accumulates to a particular concentration [23], which will reduce the SO_2 absorption rate in the desulphurization tower and reduce the desulphurization efficiency.

3.2. Orthogonal Experiment Results and Analysis

In the sensitivity analysis of Aspen Plus, the effect of each factor on the desulfurization efficiency was further investigated by referring to the working design of Yang *et al*. [24] and conducting orthogonal tests. In this orthogonal experiment, there are 3 levels and 3 factors, and only the effect of the three factors with an apparent linear relationship on the SO_2 content of the flue gas outlet is investigated, without considering the interaction effect. The inlet flue gas temperature (A), Ca/s (B), and water content in limestone slurry (C) are arranged on columns 1, 2, and 3 of L9 (33) in order, and the results are shown in Table **3**.

Table 3: Orthogonal experiment design and overall experimental results.

Experiment Number	Experiment Factors			Experiment Results
	A	B	C	SO_2 Content at Flue Gas Outlet（kmol/hr）
1	1(60)	1(1.01)	1(25)	1.301
2	2	1	2	1.295
3	3	1	3	1.287
4	1(80)	2(1.03)	2(35)	0.960
5	2	2	3	0.953
6	3	2	1	0.965
7	1(100)	3(1.05)	3(45)	0.619
8	2	3	1	0.629
9	3	3	2	1.625
K_1	2.880	3.883	2.895	-
K_2	2.876	2.884	2.880	-
K_3	2.878	1.873	2.865	-
k_1	0.960	1.294	0.965	-
k_2	0.961	0.961	0.960	-
k_3	0.959	0.625	0.955	-
T1	2.880	3.883	2.895	-
T2	2.883	2.884	2.880	-
T3	2.878	1.874	2.865	-
$T1^2$	8.294	15.078	8.381	-
$T2^2$	8.313	8.318	8.296	-
$T3^2$	8.280	3.5103	8.211	-
Range R	0.001081	0.333	0.005	-
SS	0	0.673	0	-
n	2	2	2	-
DF	2.67157E-06	0.336491456	7.29643E-05	-
F-value	0.32	61084.27	13.25	-
P-value	0.76	0	0.02	-
Assessment	Not Significant	Highly Significant	Significant	-

Firstly, ascertain the optimal levels for the experimental factors. Drawing on the concept of orthogonal experimentation for A1, A2, and A3, the three sets of experiments exhibit precisely identical testing conditions, facilitating direct comparison. As evident in Table **3**, the KA1, KA2, and KA3 values differ, implying that Factor A, namely the calcium-sulfur ratio, significantly impacts the experimental conditions. Given that the experimental influence index is centered around the SO_2 content at the flue gas outlet, a comparison of the three values reveals that KA2 is the largest, followed by KA1, with KA3 being the smallest. Hence, it is concluded that A2 represents the superior level for Factor A, with a calcium-sulfur ratio of 1.03. Employing a similar approach, the superior levels for Factors B and C are determined as B1 and C1, respectively. Consequently, the optimal combination of levels for the three factors is delineated as a flue gas inlet temperature of 80°C, a calcium-sulfur ratio of 1.03, and a water content in the limestone slurry of 35 kmol/hr.

Second, the order of priority of the influencing factors must be determined. The magnitude of the range R can be used to assess the significance of each factor's influence on the experimental indicators. As evident from Table **3**, the values of R are ranked as follows: RB > RC > RA. It can be concluded that the influence factor of the calcium-sulfur ratio has the most significant influence on the test results, followed by the water content of limestone slurry. In contrast, the inlet flue gas temperature has a smaller influence factor on the test. Finally, the experiment was analyzed using variance analysis (ANOVA), which can reduce the influence of experimental level and error on the experimental results. According to Table **3**, no additional error columns exist in this orthogonal experiment. Hence, the column with the least significant impact is selected for analysis as the error column. The error columns of calcium-sulfur ratio and water content in limestone slurry are designated as Factor I and Factor III, respectively. The outcomes of this analysis are presented in Table **3**. A detailed examination of Table **3** reveals that the calcium-sulfur ratio is the most significant factor influencing SO_2 absorption efficiency. Additionally, the water content in the limestone slurry exerts a moderate impact, while the flue gas inlet temperature demonstrates no substantial effect.

4. Conclusion

In summary, integrating Aspen Plus with the limestone-gypsum WFGD process is an optimal decision. Upon successfully modeling and executing the WFGD process flow, the findings of the analysis, along with the outcomes of the designed orthogonal experimental validation, can be distilled into the following key points:

(1) The model simulation results indicate that the SO_2 content in the flue gas decreased from 18.36 kmol/hr at the desulfurization tower inlet to 0.76 kmol/hr at the outlet, achieving a desulfurization rate of 95.9%. It confirms the feasibility of the process and operating conditions.

(2) Sensitivity analysis using Aspen Plus software shows that the inlet flue gas temperature moderately affects desulfurization efficiency. Therefore, to protect equipment and optimize resource use, the inlet temperature is reduced to around 80°C. Within the range of 1.00 to 1.05, increasing the Ca/S enhances desulfurization efficiency. The water content in the limestone slurry affects the concentration of the slurry in the desulfurization tower, indirectly impacting desulfurization efficiency. When the inlet SO_2 concentration is between 16 and 19 kmol/hr, a lower molar flow rate of SO_2 improves desulfurization efficiency.

(3) Based on the orthogonal experimental design and analysis of influencing factors, the optimal operating parameters for this model are a flue gas inlet temperature of 80°C, a Ca/S of 1.03, and water content in the limestone slurry of 35 kmol/hr. Among these factors, the Ca/S has the most significant impact on desulfurization efficiency.

Conflict of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Funding

The financial supports in the authors' laboratory were received from Doctoral Scientific Research Foundation of Zhengzhou University of Light Industry (No. NYY20200051).

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