

Article

Study on the Emission Characteristics of Fine Particulate Matter in the White Mud Desulfurization Process

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Abstract

White mud is a promising desulfurizing agent, but the risk of fine particulate emissions exists during its application. This study investigated the fine particulate emissions in the white mud desulfurization process and analyzed the effects of process parameters, including gas-to-liquid ratio, empty tower gas velocity, and slurry concentration, on particulate emissions. The results showed that white mud desulfurization achieved effective SO₂ removal, with a removal efficiency ranging from 93.5% to 95.8%. However, the emission of fine particulates was found to be a significant environmental concern. At a slurry concentration of 15%, the fine particulate number concentration was found to be 5.9×10^6 particles/cm³, with a mass concentration of approximately 43.2 mg/m³. The study further revealed that increasing the empty tower gas velocity from 2.5 m/s to 4.5 m/s also significantly increased particulate emissions. Similarly, increasing the gas-to-liquid ratio from 10 L/m³ to 15 L/m³ led to a 25.5% increase in the fine particulate number concentration. These changes were attributed to the increased atomization of fine droplets and the enhanced gas–liquid relative movement, which facilitated the entrainment of more fine particulates into the flue gas. While improving the slurry concentration led to better desulfurization efficiency, these adjustments also resulted in higher fine particulate emissions. Therefore, optimizing process parameters to balance desulfurization efficiency and fine particulate emission control was crucial for practical applications.

Keywords: white mud; desulfurization; fine particulate matter; pollutant control



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1. Introduction

With the ongoing global industrialization, environmental pollution has become an increasingly severe issue, particularly the impact of SO₂ emissions on the atmospheric environment and human health, which has been highlighted in recent years [1–3]. Wet flue gas desulfurization (WFGD) technology, as the most widely applied flue gas desulfurization method, has been extensively used due to its high efficiency and economic advantages [4–6]. White mud desulfurization, a novel wet desulfurization technology, has gained attention recently. White mud, a byproduct of the paper industry, is primarily composed of CaCO₃, Ca(OH)₂, and other mineral components, which have been found to possess high reactivity and good adsorption properties [7,8]. The principle of white mud desulfurization has been found to be similar to that of limestone desulfurization, where white mud slurry is

sprayed into the flue gas to react with SO_2 , producing bisulfites that remove SO_2 from the flue gas [9,10]. The bisulfites are then oxidized by O_2 from the air to form gypsum and other sulfates. Compared to traditional limestone desulfurization, white mud has been shown to offer lower raw material costs, and the gypsum produced during desulfurization has a higher market value. Therefore, as a new desulfurization technology, white mud desulfurization technology can effectively reduce SO_2 emissions while providing a new approach for the reutilization of white mud resources.

However, despite the significant advantages of white mud desulfurization in removing SO_2 , the issue of fine particulate emissions during its application has been recognized as an environmental concern. In the WFGD process, due to factors such as nozzle atomization and gas–liquid interactions, some slurry droplets and solid particles are carried out of the desulfurization tower by the gas flow. These fine particulates, due to their small particle size, are difficult to capture by conventional mist eliminators and are thus emitted into the atmosphere [11–13]. As the desulfurization efficiency has been improved and the gas velocity has been increased, the concentration of fine particulates has been found to rise, particularly in the white mud desulfurization process, where the generation and emission of fine particulates have been observed to be more pronounced. Therefore, the control of fine particulate emissions while improving desulfurization efficiency has become a major challenge faced by white mud desulfurization technology.

Numerous studies have been conducted to investigate the fine particulate emissions during the desulfurization process. It has been shown that the emission of fine particulates is closely related not only to the composition of the desulfurization slurry and gas–liquid contact efficiency, but also to operational conditions such as gas flow velocity, gas-to-liquid ratio, and slurry concentration [14,15]. Many studies have focused on the effects of factors such as gas-to-liquid ratio, empty tower gas velocity, and slurry concentration on fine particulate emissions, and strategies have been proposed to optimize these emissions by adjusting process parameters [16–18]. For instance, the increase in gas-to-liquid ratio or the enhancement of empty tower gas velocity has been shown to improve desulfurization efficiency, but it also leads to an increase in fine particulate generation and emissions. As a result, minimizing the emission of fine particulates while improving desulfurization efficiency has become a key research focus. Existing studies have provided theoretical support for controlling fine particulate emissions during desulfurization; however, balancing efficiency and emissions in practical applications remains an urgent issue.

This work investigated the effects of different process conditions (such as gas-to-liquid ratio, empty tower gas velocity, and slurry concentration) on fine particulate emissions during the white mud desulfurization process. The number and mass concentrations of fine particulates were measured and analyzed under various process parameters. This study aimed to reveal the mechanisms behind the generation of fine particulates in white mud desulfurization and to propose effective optimization strategies for reducing particulate emissions. Through this research, a theoretical basis for the further optimization and promotion of white mud desulfurization technology was expected to be provided, while effective reference methods for controlling fine particulate emissions in industrial desulfurization processes are also anticipated.

2. Materials and Methods

2.1. Formation of White Mud

White mud was a typical byproduct of the alkali recovery process in the paper industry. Its formation involved multiple stages, including fiber extraction and alkali recovery, as shown in Figure 1 [8]. Raw materials such as straw, wood, and bamboo were chopped, screened, and washed during the fiber extraction stage. They were then reacted with NaOH

and Na_2S . The black liquor obtained from digestion was extracted and concentrated by evaporation, then sent to the alkali recovery furnace for calcination. White mud primarily originated from the causticizing process in the alkali recovery stage. The purpose of this process was to convert Na_2CO_3 in the green liquor to NaOH . In the specific operation, CaO was added to the clarified green liquor, and a digestion and causticizing reaction occurred. After clarification, the upper liquid, containing NaOH , was recovered and could be reused in the digestion process, while the white precipitate formed in the lower layer was dewatered to obtain white mud. Due to the addition of NaOH and Na_2S during digestion, the entire solution system was alkaline. Additionally, the addition of lime during the causticizing reaction resulted in a high content of CaCO_3 in white mud. Previous studies have shown that the CaCO_3 content in white mud typically ranged from 86.5% [19]. This characteristic provided the conditions for its use as a low-cost and efficient desulfurizing agent in the wet flue gas desulfurization process.

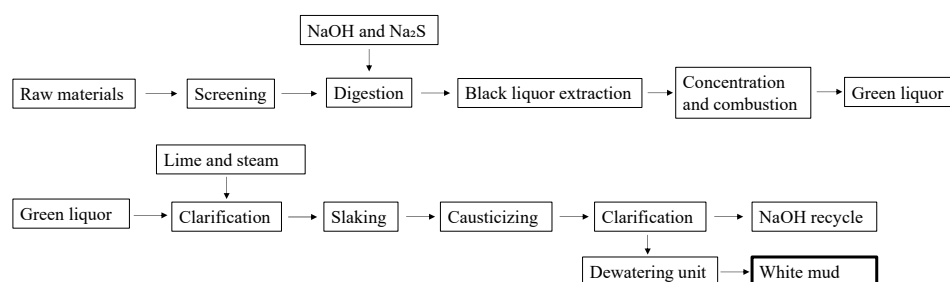


Figure 1. Formation of White Mud.

2.2. Physicochemical Characterization of White Mud

The microscopic morphology of white mud and the desulfurization byproduct was observed using a Field Emission Scanning Electron Microscope (FE-SEM, Zeiss Gemini Sigma 300, Oberkochen, Germany). Before testing, the samples were thoroughly dried at $90\text{ }^{\circ}\text{C}$ and gold-coated to improve conductivity. The crystal structure of white mud before and after desulfurization was analyzed using an X-ray diffractometer (XRD, Rigaku SmartLab SE, Tokyo, Japan). The testing conditions were set as follows: $\text{Cu K}\alpha$ radiation, tube voltage of 40 kV , tube current of 30 mA , scanning range of $2\theta = 5\text{--}90^{\circ}$, step size of 0.02° , and scanning speed of $5^{\circ}/\text{min}$. The resulting diffraction patterns were analyzed using the PDF card database for phase identification.

2.3. Experimental System

Based on the white mud desulfurization process, a simulated flue gas wet desulfurization system was designed and constructed, as shown in Figure 2a. The flue gas velocity investigated ranged from 2.5 to 4.5 m/s , while the liquid-to-gas ratio was varied between 10 and 15 L/m^3 . The system mainly consisted of a desulfurization tower, a gas mixing system, a simulated slurry preparation and transport system, and a sampling and analysis system. Air and SO_2 were thoroughly mixed in a gas mixer and then introduced into the desulfurization tower, where they were in countercurrent contact with the desulfurization slurry inside the tower. The desulfurization tower was of a spray tower type and is made of heat-resistant polycarbonate pipes. Each level was equipped with an array of full-cone nozzles uniformly distributed to ensure sufficient gas–liquid contact. The operating pressure of the nozzles was maintained at $0.1\text{--}0.2\text{ MPa}$, corresponding to an individual nozzle flow rate of $12\text{--}15\text{ L/min}$. A mist eliminator with baffles was placed above the spray layers. The simulated flue gas was discharged into the atmosphere after mist elimination. The slurry was sprayed and returned to the slurry tank for recycling.

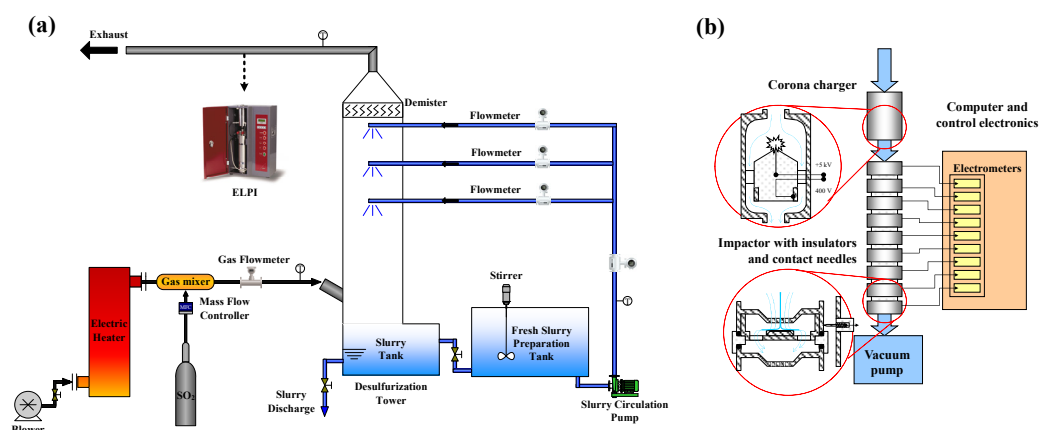


Figure 2. The simulated white mud desulfurization system. (a) flue gas wet desulfurization system, (b) schematic figure of ELPI.

The concentration and particle size distribution of fine particulate matter in the flue gas were measured in real-time using an Electrical Low Pressure Impactor (ELPI) produced by Detaki (Kangasala, Finland). The ELPI could classify particles smaller than $10\ \mu\text{m}$ into 12 levels for online measurement, as illustrated in Figure 2b. During the testing, the flue gas first passed through a cyclone dust collector to remove particles larger than $10\ \mu\text{m}$, and then was diluted using a diluter before entering the ELPI for analysis. The diluter consisted of two stages: in the first stage, purified dry heated air was used to dilute the gas, with the dilution air temperature maintained at the same level as the flue gas temperature. In the second stage, purified clean air was used for further dilution, ensuring no condensation occurred from water vapor or other substances in the flue gas during the sampling process. After dilution, the fine particles were charged and collected in a series of impactors according to their aerodynamic particle size distribution.

3. Results and Discussion

3.1. Desulfurization Performance of White Mud Slurry

Desulfurization slurry was prepared using white mud from a paper mill, with a slurry concentration of 15%. The experimental conditions were set: empty tower gas velocity of $3.5\ \text{m/s}$, gas-to-liquid ratio of $15\ \text{L/m}^3$, and SO_2 inlet concentrations of 1000, 1500, and $2000\ \text{mg/Nm}^3$. The absorption tower was equipped with a three-level spray system to ensure sufficient gas–liquid contact. To compare the desulfurization performance of white mud with traditional limestone, a slurry with the same concentration (15%) of limestone was also prepared and tested under the same operational conditions. The results showed that the SO_2 removal efficiency of the white mud slurry reached 93.5% to 95.8%, which was slightly higher than that of the limestone slurry, which had an SO_2 removal efficiency of 91.3% to 92.4% (Figure 3). The main reasons for this difference are as follows: First, in addition to CaCO_3 , white mud also contains some active Ca(OH)_2 and porous residual structures, which result in a larger specific surface area and more surface reaction sites, making its reactivity superior to that of pure limestone. Second, the density of the white mud slurry is lower than that of the limestone slurry, which improves the slurry’s flowability and enhances the mass transfer and diffusion processes within the absorption tower. Third, the impurities in white mud (such as Na, Mg, etc.) could promote the Ca–S reaction, further improving SO_2 absorption efficiency.

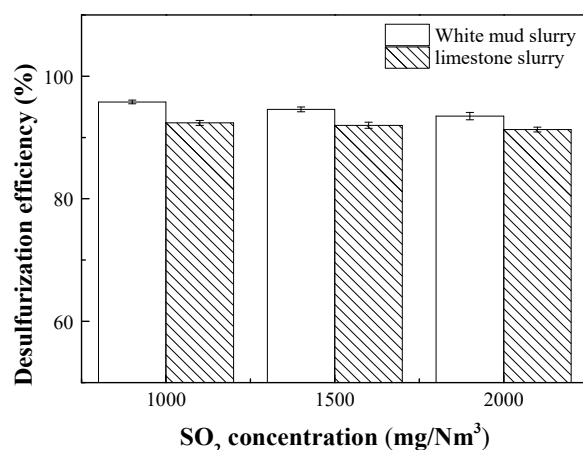


Figure 3. The desulfurization efficiency of white mud and limestone slurry.

According to the SEM results shown in Figure 4a, the white mud before desulfurization was found to form loose agglomerates composed of fine primary particles, with a rough surface rich in micropores and cracks. In contrast, after desulfurization, the morphology of the byproduct gypsum was found to change significantly, transforming into more regular crystalline structures, predominantly in plate-like and short columnar forms, as shown in Figure 4b,c. The crystal faces were dense, with clear contours, and the particle size was larger, with reduced agglomeration compared to the earlier agglomerates. In comparison, gypsum crystals generated during traditional wet flue gas desulfurization processes using limestone slurry as an absorbent were predominantly needle-like in structure [20].

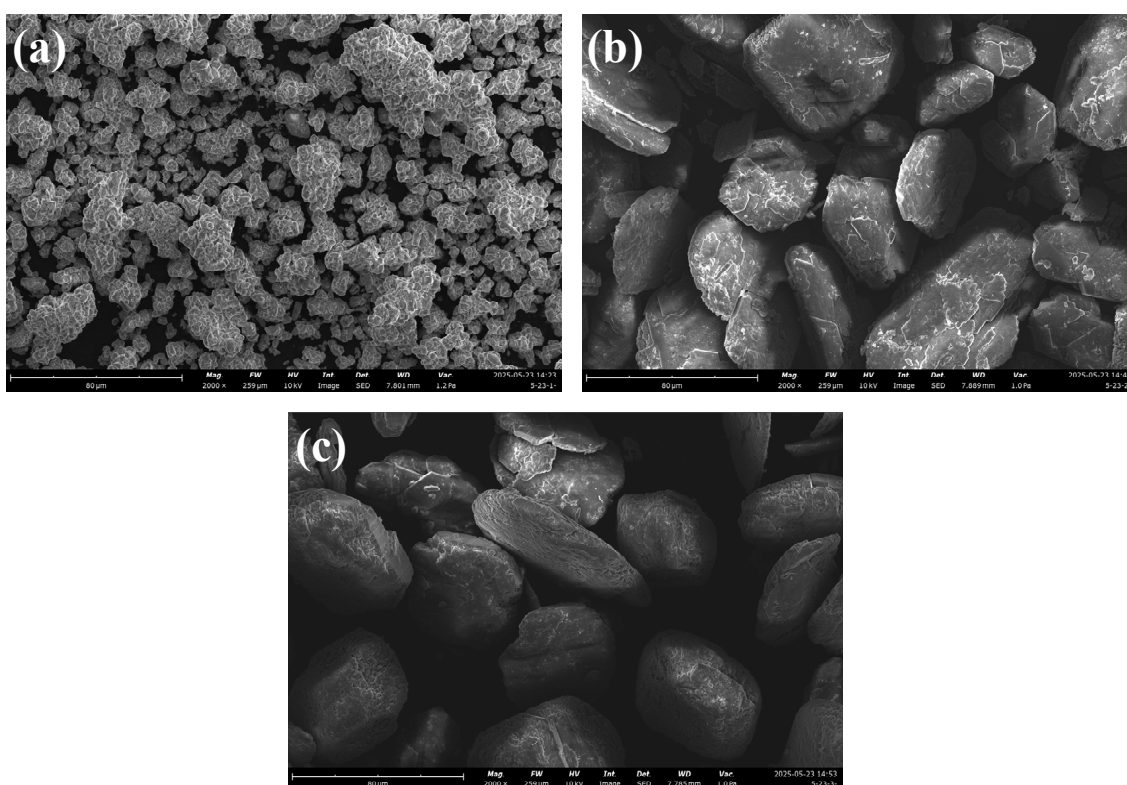


Figure 4. Microscopic morphology of white mud and the desulfurization byproduct. (a) white mud; (b,c) gypsum.

To clarify the phase composition of the samples, XRD analysis was performed, and the results were shown in Figure 5. The characteristic diffraction peaks of the

white mud sample were clearly identified as corresponding to CaCO_3 , with JCPDS card number 47-1743, which was consistent with previously reported results. It was noted that Ca(OH)_2 , which might remain in the white mud, was not detected by XRD. This could be due to the fact that, on one hand, it existed in a microcrystalline or amorphous form under the reaction conditions, with its diffraction peaks being obscured by the intense peaks of CaCO_3 . On the other hand, if its mass fraction were below the XRD detection limit ($<5\%$), it would also have been difficult to detect. The diffraction peaks of the desulfurized white mud gypsum sample matched the JCPDS card number 33-0311, confirming that the primary desulfurization product was $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$.

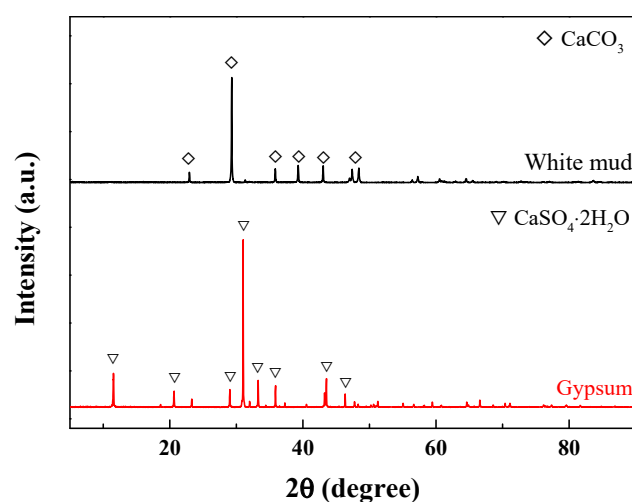


Figure 5. The phase composition of white mud and gypsum.

3.2. Study on Fine Particulate Emissions in the White Mud Desulfurization Process

In the wet desulfurization process, the slurry droplets atomized by the nozzles, when in countercurrent contact with the high-temperature flue gas, may be carried out into the atmosphere due to evaporation and entrainment. Because the desulfurization slurry droplets inherently contained gypsum particles and soluble salts, a significant amount of residual fly ash and secondary sulfate fine particles were carried with the flue gas into the atmosphere, increasing the concentration of fine particulate emissions at the desulfurization tower outlet [21,22]. The experimental conditions were set with a slurry concentration of 15%, a gas-to-liquid ratio of 15 L/m^3 , and a SO_2 inlet concentration of 1500 mg/Nm^3 . The fine particulate emissions concentration in the desulfurized flue gas was shown in Figure 6. After the white mud slurry wet desulfurization system, the fine particulate number concentration reached $5.9 \times 10^6 \text{ particles/cm}^3$, with a mass concentration of approximately 43.2 mg/m^3 . This indicated that a large amount of fine particulate matter could be generated during the white mud desulfurization process. The fine particulate emissions during limestone slurry desulfurization were also tested to compare the fine particulate emissions. The results showed that during limestone slurry desulfurization, fine particulate matter of about $1.2 \times 10^7 \text{ particles/cm}^3$ was generated, with a mass concentration of approximately 90.8 mg/m^3 , which was higher than that of the white mud slurry.

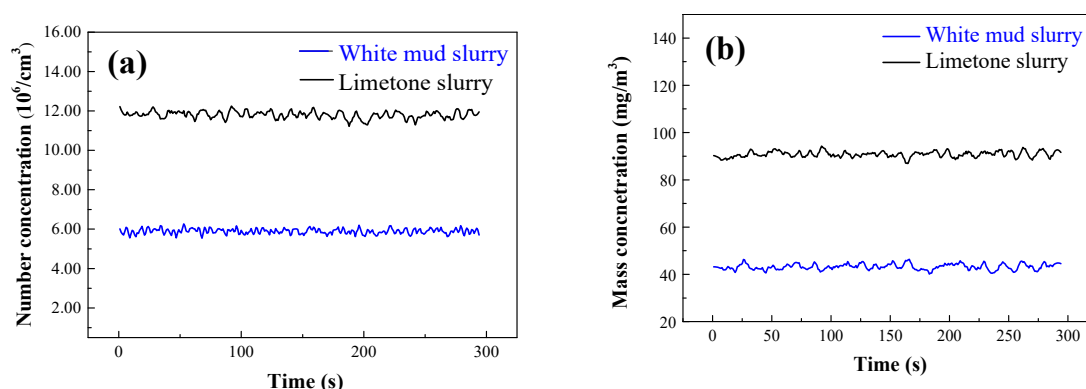


Figure 6. Fine particulate emissions using white mud and limestone slurry. (a) Number concentration; (b) Mass concentration.

From the perspective of the fine particle formation mechanism in the desulfurization process, during limestone desulfurization, coarse CaCO_3 particles were dissolved and formed $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$, due to the relatively high oversaturation of the slurry, resulted in gypsum crystals of various sizes and uneven distribution. Some unreacted CaCO_3 remained in the form of fine particles, and when the slurry droplets carried by the high-temperature flue gas evaporated, the residual solute further formed submicron-sized fine particles, causing a large number of fine particles to escape [23]. In contrast, in the white mud desulfurization system, impurities such as Na_2O and MgO in white mud were found to effectively reduce the slurry oversaturation, suppress the precipitation of fine particles, and promote the formation of more regular and larger $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ crystals [7]. From the SEM images, the gypsum generated in the white mud desulfurization process was found to be regular short columns, with a narrow particle size distribution. Even when soluble salts were present in the slurry, they were mainly found to be attached to the surface of the gypsum crystals, rather than forming a large number of fine particles. As a result, the fine particulate emissions in the white mud desulfurization process were lower than those in the limestone desulfurization process.

3.3. Study on the Factors Affecting Fine Particulate Emissions

3.3.1. Effect of Empty Tower Gas Velocity

Gas velocity was an important factor influencing slurry carryover in industrial wet desulfurization systems [24]. In general, the gas velocity was maintained between 2.5 m/s and 4 m/s. When the gas velocity was too high, droplets condensed on the tower walls were easily re-entrained by the gas flow, leading to secondary droplet carryover. In this experiment, to investigate the effect of empty tower gas velocity on fine particulate emissions, gas velocities of 2.5, 3.5, and 4.5 m/s were selected. The fine particulate concentration was shown in Figure 7. Under higher gas velocities, the concentration of fine particulate matter was found to increase from 4.2×10^6 particles/ cm^3 to 6.8×10^6 particles/ cm^3 , and the mass concentration of particulate matter increased from 32.9 to 55.3 mg/m^3 . A higher empty tower gas velocity enhanced the relative movement between the gas and liquid phases within the tower, shortening the residence time of the gas in the tower. This process significantly affected the fine particulate matter of different particle sizes. First, as the empty tower gas velocity was increased, the relative movement between the gas flow and desulfurization slurry was intensified, which increased the probability of droplets being carried over by the gas flow [25]. Higher gas velocities accelerated the gas flow, promoting the entrainment of smaller droplets into the flue gas. These submicron fine particles, due to their smaller size, had shorter residence times and were difficult to capture, and thus tended to escape into the desulfurized flue gas. Second, as the empty tower gas

velocity was increased, the gas–liquid contact area in the tower was expanded, enhancing the gas–liquid contact effect, which made it easier for solutes in the desulfurization slurry to be carried over by the gas flow. From the above analysis, it was observed that increasing the empty tower gas velocity not only intensified the carryover of fine droplets from the white mud desulfurization slurry, leading to an increase in the number of fine droplets in the desulfurized flue gas, but also accelerated the relative movement between the gas and liquid phases and shortened the residence time, increasing the generation and emission of fine particulate matter.

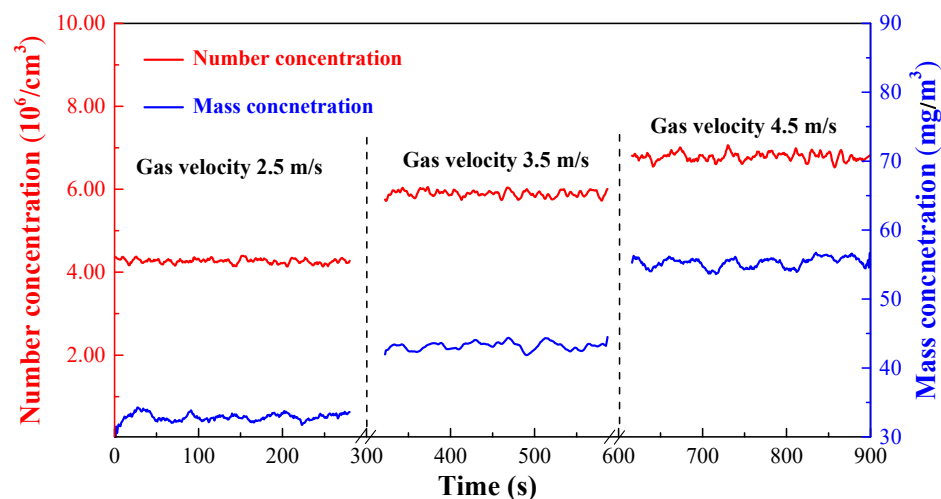


Figure 7. Effect of empty tower gas velocity on fine particulate emissions.

3.3.2. Effect of Gas-to-Liquid Ratio

The gas-to-liquid ratio was one of the key parameters influencing the gas–liquid mass transfer performance in the white flue gas desulfurization process. Studies have shown that desulfurization efficiency was generally increased significantly with the rise in gas-to-liquid ratio. This was because the increase in gas-to-liquid ratio enhanced the relative movement between the gas and liquid phases, which helped improve the efficiency of the desulfurization reaction. However, changes in the gas-to-liquid ratio also had a significant impact on the fine particulate emissions [26]. Figure 8 showed the test results for the droplet and fine particulate emissions under different gas-to-liquid ratios. With the increase in gas-to-liquid ratio, the number concentration of fine particulates in the desulfurized flue gas was found to increase significantly, from 4.7×10^6 particles/ cm^3 to 5.9×10^6 particles/ cm^3 , a growth of approximately 25.5%. As the gas-to-liquid ratio was increased, the enhanced flow caused more droplets to be atomized into smaller particle sizes. These fine droplets were more difficult to capture by the mist eliminator and were more easily carried by the gas flow, eventually released into the atmosphere, which led to an increase in fine particulate concentration [27]. Lowering the gas-to-liquid ratio was considered an effective control measure to reduce fine particulate emissions while improving desulfurization efficiency. By controlling the gas-to-liquid ratio, the number of fine droplets generated by atomization could be reduced, thereby decreasing the concentration of fine particulates in the desulfurized flue gas.

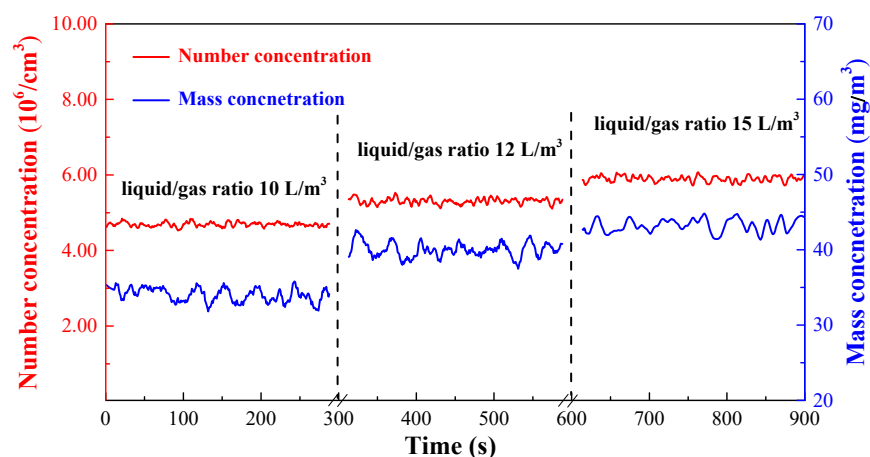


Figure 8. Effect of gas-to-liquid ratio on fine particulate emissions.

3.3.3. Effect of Slurry Concentration

The slurry concentration significantly influenced several key factors in the wet desulfurization process, including the viscosity of the liquid, the atomization efficiency of the nozzles, and the entrainment of solid particles in the slurry droplets, all of which affected the fine particulate emission characteristics [28]. In desulfurization processes, slurry concentration was typically controlled within the range of 5% to 20% to ensure high desulfurization efficiency. To investigate the relationship between slurry concentration and the concentrations of droplets and fine particulates in the desulfurized flue gas, tests were conducted at slurry concentrations of 10%, 15%, and 20%. The variation in fine particulate number concentration in the desulfurized flue gas under different slurry concentrations was shown in Figure 9. The results indicated that as the slurry concentration increased from 10% to 20%, the fine particulate number concentration in the desulfurized flue gas increased significantly, from 5.1×10^6 particles/ cm^3 to 6.6×10^6 particles/ cm^3 . The relationship between slurry concentration and particulate emissions was complex [29]. On one hand, the higher slurry concentration resulted in a greater mass of solid particles in the slurry, leading to the generation of more fine particulates during the desulfurization process. On the other hand, while the increased slurry concentration led to larger droplet sizes, which could theoretically reduce the entrainment of fine particles, the overall effect was an increase in fine particulate emissions due to the higher solid content in the slurry, which increased the potential for fine particle generation. Therefore, an optimal slurry concentration was required to balance both desulfurization efficiency and fine particulate emissions, particularly in industrial settings with stringent emission standards.

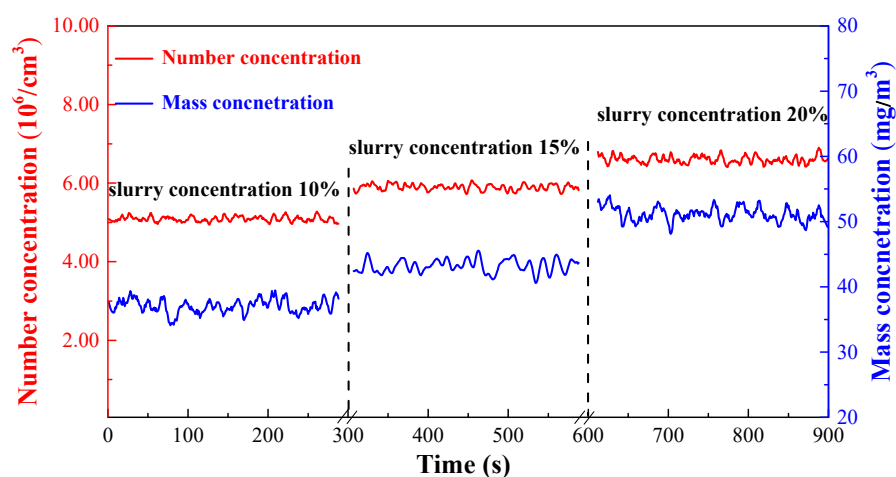


Figure 9. Effect of slurry concentration on fine particulate emissions.

4. Conclusions

This work investigated the fine particulate emissions in the white mud desulfurization process and analyzed the effects of process parameters such as empty tower gas velocity, gas-to-liquid ratio, and slurry concentration. The main conclusions are summarized as follows.

(1) The white mud desulfurization achieved good SO₂ removal efficiency, ranging from 93.5% to 95.8%. However, fine particulate emissions remained a significant environmental concern during the process, which were difficult to capture by conventional mist eliminators.

(2) The experimental results indicated that gas-to-liquid ratio, empty tower gas velocity, and slurry concentration significantly impacted the fine particulate emissions. When the empty tower gas velocity was increased from 2.5 m/s to 4.5 m/s, the particulate concentration increased from 4.2×10^6 particles/cm³ to 6.8×10^6 particles/cm³. As the gas-to-liquid ratio increased from 10 L/m³ to 15 L/m³, the fine particulate number concentration increased from 4.7×10^6 particles/cm³ to 5.9×10^6 particles/cm³, a rise of about 25.5%. Additionally, at a slurry concentration of 20%, the fine particulate number concentration increased to 6.6×10^6 particles/cm³, with a mass concentration of approximately 51.2 mg/m³.

(3) These findings suggested that increasing the gas-to-liquid ratio, empty tower gas velocity, and slurry concentration improved desulfurization efficiency but led to higher particulate emissions. Therefore, balancing desulfurization efficiency and fine particulate emissions in practical applications requires careful optimization of these parameters.

Based on the above research conclusions, future research could further explore the mechanisms of fine particulate emissions and provide more precise optimization strategies and technical support by combining experimental data and numerical simulations. In addition, advanced hybrid configurations may be considered, in which the conventional spray tower is complemented by wet electrostatic precipitator or electrified spray column. These systems have shown great potential for enhancing the simultaneous removal of SO₂ and ultrafine particles, and thus represent a promising direction for the further development of desulfurization technologies.

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