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Study on the Efficiency of Fine Particle Removal in a Single-Tower Dual-Cycle Desulfurization Process Utilizing Heterogeneous Condensation

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Abstract: This study investigated a new method for controlling the emission of fine particles through heterogeneous condensation. Specifically, the research focuses on the application of single-tower double-cycle desulfurization technology in the wet flue gas desulfurization process. The establishment of a supersaturation environment necessary for heterogeneous condensation was achieved by reducing the temperature of desulfurization slurry in the oxidation zone. Numerical simulations were used to study the distribution of the supersaturation degree and fluid dynamics characteristics in the desulfurization tower after the cooling of desulfurization slurry. Furthermore, the impact of single-tower double-cycle technology on the removal efficiency of fine particles was examined. The results of the numerical simulations indicate that cooling the desulfurization slurry in the absorption zone could establish a supersaturated vapor environment, with the supersaturation degree and region increasing as the slurry temperature decreases. Under typical operating conditions, a temperature drop of approximately 8~10 °C was found to be most suitable for the desulfurization slurry. Moreover, lowering the temperature of the desulfurization slurry in the absorption zone increases the supersaturation degree from 0.93 to 1.85. Additionally, the use of single-tower double-cycle desulfurization technology is shown to significantly enhance the removal efficiency of fine particles, particularly those within the particle size range of 0.1~1 μm. Ultimately, this method could increase the removal efficiency of fine particles from 39.9% to 57.9%.

Keywords: fine particles; wet flue desulfurization; heterogeneous condensation; removal

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1. Introduction

Fine particles are the primary pollutants that impact the atmospheric environment and human health, a concern that has gradually gained attention [1]. According to statistics, coal-fired power plants are the main sources of emissions for fine particles and SO_2 [2]. The aerodynamic diameter that is less than 10 μm is referred to as fine particles, and the aerodynamic diameter that is more than 10 μm is referred to as coarse particles [3]. Fine particles pollution is an important factor leading to reduced atmospheric visibility, haze weather, acid rain, climate change, and other major problems, causing serious harm to the environment and human health [4,5]. Fine particles are much more harmful to the human body than coarse particles. The nasal cavity can basically block particles with an aerodynamic diameter greater than 10 μm, while the particles less than 10 μm will be deposited into the throat, bronchial tubes, and alveoli, and fine particles less than 2.5 μm will be permanently deposited on the bronchial tubes and alveoli and enter the blood circulation [6]. Many power plants have installed equipment such as electrostatic precipitators and wet flue gas desulfurization systems to eliminate dust and SO_2 [7,8]. While these systems have higher removal efficiency for larger particle sizes, their efficiency for smaller particle sizes is suboptimal. The removal efficiency can exceed 99.9% for particles larger than 10 μm, but it is relatively low for particles smaller than 2.5 μm [9–11]. Although wet desulfurization systems

have relatively high efficiency in removing sulfur dioxide, they also generate secondary pollution, such as sulfur trioxide [12–14]. Additionally, while the wet desulfurization system has some impact on removing fine particles, the removal efficiency for smaller particles is very low due to the Greenfield Gap effect [15,16]. Consequently, many researchers have studied and proposed new methods to enhance the removal of fine particles by increasing their size.

Heterogeneous condensation is one of the most effective and promising methods for enhancing the removal of fine particles [17]. In this technology, fine particles are activated to a nucleation center in a supersaturated vapor environment, where they form embryonic droplets [18]. Subsequently, water vapor condenses on the surface of the droplets, increasing the size and mass of the fine particlesleading to the formation of larger droplets due to the condensation of water vapor on the fine particles [19–21]. As a result, the enlarged fine particles are more easily removed in traditional dust removal equipment. The primary processes for improving the removal of fine particles through heterogeneous condensation include establishing a supersaturated vapor environment in flue gas, nucleating fine particles into condensation nuclei, forming embryonic droplets from these nuclei, and growing the droplets through condensation [22]. The most critical step is the establishment of a supersaturated vapor environment. The method of combining heterogeneous condensation with traditional dust removal equipment to enhance the removal of fine particles has been widely applied [23,24].

The most crucial step in the process of heterogeneous condensation is establishing a supersaturated vapor environment. Numerous studies have been conducted to promote the removal of fine particles based on the theory of heterogeneous condensation [25,26]. Fan et al. [27] conducted numerical calculations to assess the influence of various factors on the distribution of the supersaturated vapor degree. The results indicate that it is practical and feasible to combine wet flue gas desulfurization and heterogeneous condensation to reduce the emission of fine particles. Bao et al. [28] reported that the supersaturation environment can be achieved by adding steam to the desulfurized flue gas, resulting in a 40% to 50% improvement in the removal efficiency of fine particles. Wu et al. [29] investigated a new method to create a supersaturated environment by increasing the humidity of desulfurization flue gas, leading to a higher removal efficiency of fine particles. It is well known that the removal efficiency of fine particles during desulfurization is suboptimal without heterogeneous condensation. Nevertheless, significant improvement is achievable.

This paper established a supersaturated vapor environment in the desulfurized flue gas using a new method called single-tower double-circulation technology. Numerical simulations were conducted to investigate the effect of desulfurization slurry temperature on the supersaturation degree distribution and the fluid dynamics characteristics inside the desulfurization tower. Additionally, experimental studies were performed to assess the impact of the single-tower double-circulation technology on the removal performance of fine particles. Concurrently, the influence of the slurry temperature, liquid to gas ratio, and flue gas temperature at the inlet of the desulfurization tower on the removal efficiency of fine particles was examined. Finally, the paper discussed the effect of single-tower double-circulation technology on desulfurization water consumption.

2. Experiment System and Numerical Calculation

2.1. Experimental System

The experimental setup, as depicted in Figure 1, comprises a fully automated coal-fired boiler, a buffer tank, an ESP system, and a single tower with double-circulation desulfurization. The rated flue gas volume of the boiler is 350 $Nm^3\ h^{-1}$. The flue gas produced by the boiler was introduced into the buffer tank for thorough mixing before passing through the ESP, where larger particles (the particle size is above 10 μm) could be removed. Subsequently, the flue gas entered the desulfurization tower. The spray layer of the desulfurization tower consists of two distinct zones, namely the oxidation zone and absorption zone. These zones were controlled separately, unlike traditional wet flue

gas desulfurization systems. The desulfurization slurry in the oxidation zone was cooled by a heat exchanger, leading to the creation of a supersaturated vapor environment that enhanced the removal efficiency of fine particles (the particle size is less than 10 μm). The desulphurization slurry in the oxidation zone is recycled to the slurry tank in the oxidation zone after spraying. The desulphurization slurry in the absorption zone is collected by the tray after spraying, and then circulates into the slurry tank in the oxidation zone. The systems are independent of each other. The pH of the desulfurization slurry in the oxidation zone typically ranged between 4.0 and 5.0, facilitating the absorption of some SO_2 from the flue gas. In contrast, the pH of the desulfurization slurry in the absorption zone generally fell between 5.6 and 6.2, enabling the deep removal of SO_2 from the flue gas. Furthermore, the desulfurization tower was equipped with a highly efficient demister at the top to eliminate larger condensable droplets. The average flow rate of flue gas inside the tower was controlled at 3.5 m/s. The measurement position is the inlet and outlet position of the desulfurization tower, and the measurement is continuous. It was tested three times for each measuring point under each working condition, and the average value was finally taken.

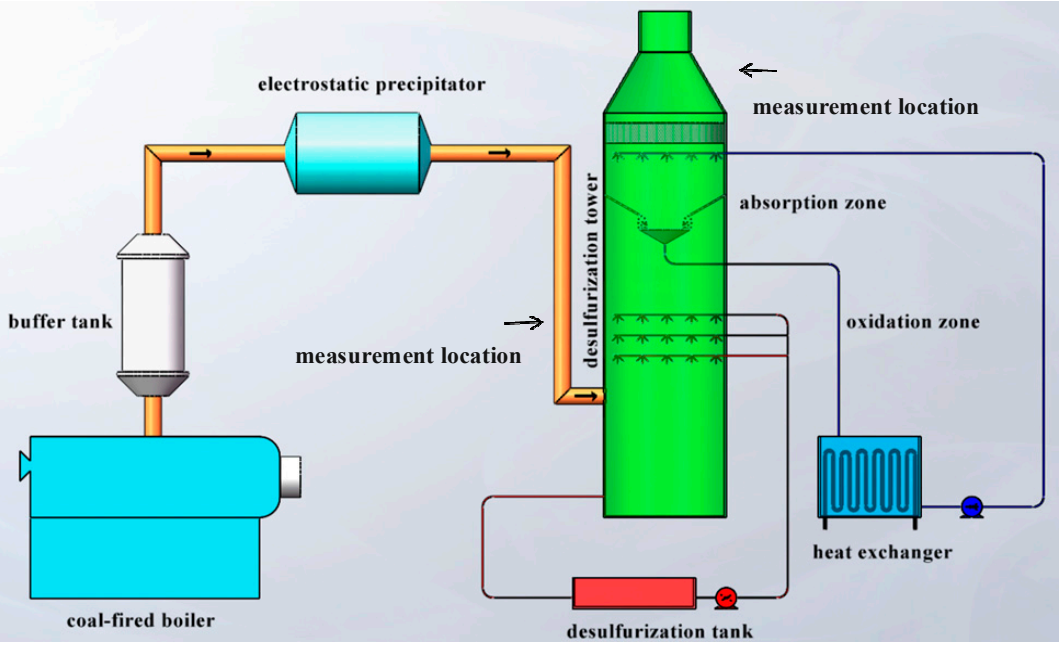

Figure 1. Scheme of the experimental system.

2.2. Measure Methods

During the experiment, an electrical low-pressure impactor (ELPI, Dekati Ltd., Kangasala, Finland) was utilized to measure the real-time size distribution and concentration of fine particles. The ELPI is highly precise and can measure particles within the range of 0.023 μm to 9.314 μm. However, due to the high humidity of the flue gas, vapor could condense in the sampling pipelines and on the impact plate of the ELPI, resulting in inaccurate measurements. Therefore, pretreatment is necessary before the flue gas enters the ELPI. A humiture transmitter (HMT337, Vaisala Ltd., Vantaa, Finland) was employed to measure the temperature and humidity of the flue gas.

2.3. Numerical Simulation

In this paper, Ansys-CFD 17.0 software is mainly used for simulation. Figure 2 shows the physical model of a single-tower double-circulation desulfurization tower. In the simulation, the temperature of the desulfurization slurry in the oxidation zone (first to third spray layers) is constant, while the temperature of the desulfurization slurry in the absorption zone (fourth spray layer) is varied. The total height of the desulfurization tower

simulation test bench is 1300 mm, with a tower diameter of 120 mm and an inlet pipeline diameter of 60 mm, and the length of the inlet pipeline is 300 mm. Unidirectional atomization nozzles are positioned at heights of 300 mm, 500 mm, 700 mm, and 900 mm within the desulfurization tower to analyze the fluid dynamics characteristics and supersaturation distribution. In the simulation, the effect of the slurry temperature in the absorption zone on the distribution of the supersaturation degree in the desulfurization tower is mainly simulated, and the process of mass and heat transfer is regarded as a stable process.

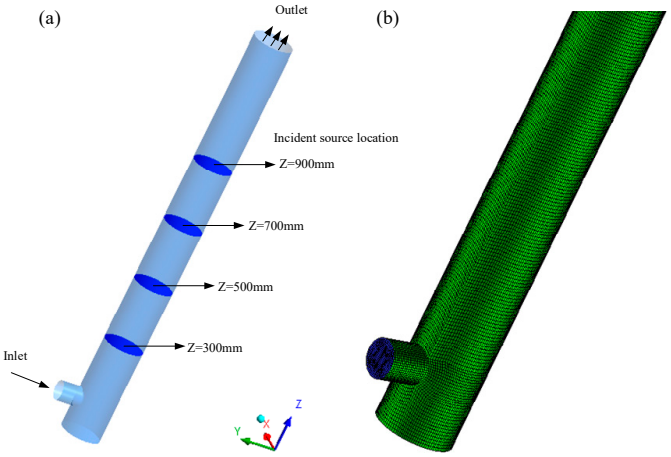

Figure 2. (**a**) Schematic diagram of desulfurization tower structure, (**b**) grid division of desulfurization tower.

During the cooling process of desulfurization slurry, intricate mass and heat transfer processes occur. It is essential to account for the interaction between flue gas and slurry droplets, as well as the heat transfer and phase change processes between gas and droplets, in order to establish a coupling process between continuous and discrete phases. The volume fraction of droplets within the tower is much smaller than that of flue gas. Thus, the Discrete Phase Model (DPM) [30] can predict the distribution of supersaturation inside the desulfurization tower after the desulfurization slurry is cooled. The gas phase can be simulated as continuous using the Realizable k-ε Turbulence model and incompressible gas model [31]. The continuity equation is as follows:

The mass-conservation equation [32]

$$\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x_i}(\rho u_i) = S_m \tag{1}$$

The momentum conservation equation [33]

$$\frac{\partial \rho}{\partial t}(\rho u_i) + \frac{\partial}{\partial x_i}(\rho u_i u_j) = \frac{\partial p}{\partial x_i} + \frac{\partial \tau_{ij}}{\partial x_j} + \rho g_i + F_i \tag{2}$$

The energy conservation equation [34]

$$\frac{\partial(\rho T)}{\partial t} + div(\rho u T) = div\left(\frac{k}{c_p} grad T\right) + S_T \tag{3}$$

where ρ is the fluid density (kg/m^3), t is the time (s), u is the fluid velocity (m/s), S_m is the source term, representing the mass added to the continuous phase due to droplet evaporation, and p is the static pressure (Pa), τ_{ij} is the stress tensor (Pa), g_i and F_i are the weight volume force and external volume force (N/kg), c_p is the specific heat capacity (J/(kg·K)), T is the temperature (K), k is the heat transfer coefficient (W/(m·2K)), and S_T is the viscous dissipation term, representing the heat source inside the fluid or the partial heat energy converted from mechanical energy.

Hexahedral structured mesh was utilized to segment the geometric model. The operating condition parameters are presented in Table 1, with the average flow rate of flue gas inside the tower controlled at 3.5 m/s. The Realizable k-ε Turbulence model and incompressible gas model are employed for continuous phase calculations. The inlet boundary condition was set as the velocity inlet, while the outlet boundary condition was set as the free outflow. The SIMPLEC algorithm is utilized to handle the coupling of pressure and velocity [35]. For discrete phases, a Pressure Swirl Atomizer is used as the nozzle type, with the wall and inlet boundary conditions set to reflect, and the outlet boundary conditions set to escape. The simulation calculation results directly yield the velocity field, temperature field, and water vapor distribution based on the specified model and boundary conditions. The flue gas temperature and actual water pressure at different positions of the desulfurization tower are obtained from the simulation results. Additionally, the distribution of supersaturation inside the desulfurization tower can be determined based on a specific formula.

Table 1. Typical conditions for simulated calculation.

Operating Parameters	Numerical Value
Temperature of desulfurization slurry (°C)	40, 35, 32, 30, 20
Temperature of flue gas (°C)	100, 110, 120, 130, 140
Liquid to gas ratio (L/Nm^3)	5, 10, 15, 20

The level of supersaturation in desulfurized flue gas determines the quantity of fine particulate matter nucleation centers, expressed as [36]

$$S = \frac{P(T, x)}{P_S(T, x)} \tag{4}$$

where $P(T, x)$ is the actual vapor pressure and $P_S(T, x)$ is the saturated vapor pressure.

The heterogeneous condensation will occur with the fine particles as condensation nucleus when the supersaturation degree of flue gas reaches the critical supersaturation degree of fine particles. Studies indicate that the critical degree of supersaturation required for the nucleation of fine particles during coal burning is approximately 1.15–1.20 [37], and when the supersaturation reaches 1.15 or more, heterogeneous condensation will occur. Therefore, the heterogeneous condensation process will occur when the supersaturation in the desulfurization tower is higher than 1.15.

3. Results and Discussion

3.1. The Effect of Heterogeneous Condensation on the Removal Performance of Fine Particles under Typical Condition

Figure 3 illustrates the distribution of water vapor and temperature inside the desulfurization tower under typical conditions. These include Condition 1 (inlet flue gas temperature of 120 °C, desulfurization slurry temperature in the oxidation zone of 40 °C, and desulfurization slurry temperature in the absorption zone of 32 °C) and Condition 2 (inlet flue gas temperature of 120 °C, desulfurization slurry temperature of 40 °C). Figure 3a,b depict how temperature varies with the height of the desulfurization tower, while Figure 3c,d display the distribution of the water mass fraction in the tower. The results indicate that heterogeneous condensation significantly affects the distribution of water vapor and temperature in the desulfurization tower. The temperature gradient remains relatively constant between the heights of 300 mm and 700 mm in the tower, but fluctuates significantly at the 900 mm mark, resulting in a sharp decrease in water vapor content. Simulation results reveal that the supersaturation levels at heights of 700 mm, 900 mm, and the tower outlet are 0.8839, 0.9318, and 0.9334, respectively, under Condition 2, and 1.0762, 1.3393, and 1.3431, respectively, under Condition 1. It is evident that a supersaturated vapor environment cannot exist in the desulfurization tower without external conditions. The

supersaturated vapor environment can be achieved by reducing the temperature of the desulfurization slurry. It can be concluded that under the condition where the inlet flue gas temperature of the desulfurization tower is 120 °C, the desulfurization slurry temperature in the oxidation zone is 40 °C, and the liquid–gas ratio is 15 L/Nm^3, the critical condition for heterogeneous condensation occurs when the temperature of the desulfurization slurry in the absorption zone drops by 8 °C.

The experiments were conducted under Conditions 1 and 2. Figures 4–6 illustrate the impact of heterogeneous condensation on the removal efficiency of fine particles. Figure 3 shows that the number concentration of fine particles at the exit of the desulfurization tower is approximately 4.39 × 10^6 1/cm^3 and 3.07 × 10^6 1/cm^3 under Conditions 1 and 2, respectively. It is evident that heterogeneous condensation can greatly reduce the emission of fine particles. Figure 4 displays the size distribution of fine particles at the outlet of the desulfurization tower under two different working conditions. It is apparent from the figure that the number concentration of fine particles in each size segment decreases to some extent under both conditions. Figure 6 presents the fractional removal efficiency of fine particles under conditions with and without heterogeneous condensation. It can be observed from Figure 6 that wet desulfurization affects the removal of fine particles without heterogeneous condensation, and the removal efficiency of fine particles in the size range of 0.1 μm to 1 μm is lower than that of other size segments. This phenomenon is attributed to the penetration window, known as the Greenfield Gap [38]. During wet desulfurization, fine particles in the flue gas are primarily removed by diffusion and inertia forces. Particles smaller than 0.1 μm are mainly affected by the Brownian diffusion force during washing, which increases as the particle size decreases, and have weak inertia forces [39]. Particles larger than 1 μm are mainly impacted by inertial force, which increases with particle size, while having weaker diffusion forces. However, particles between 0.1 μm and 1 μm have weak inertia and diffusion forces [40], resulting in low removal efficiency in wet desulfurization. Heterogeneous condensation in the desulfurization tower improves the removal efficiency of fine particles in each size segment, especially particles between 0.1 μm and 1 μm.

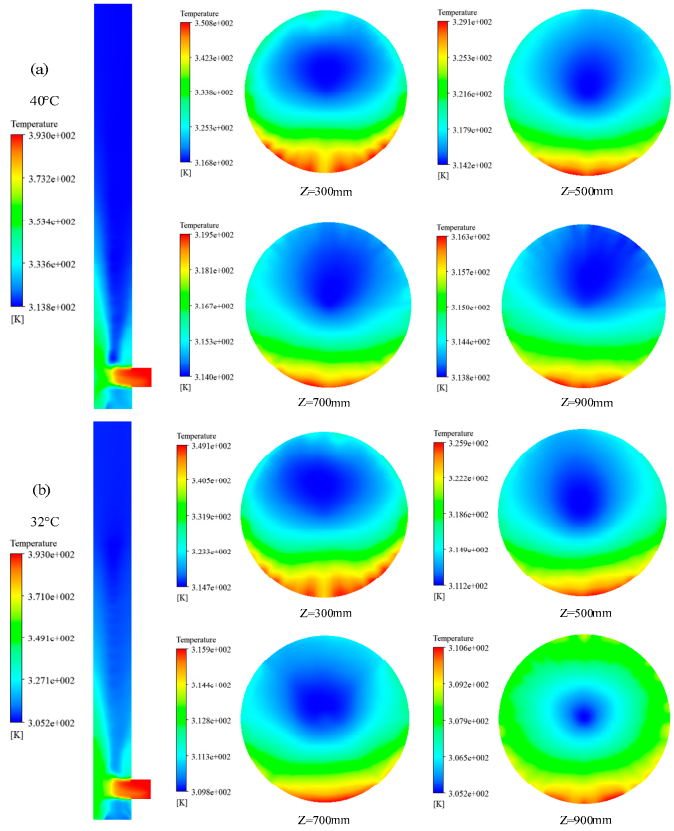

Figure 3. *Cont.*

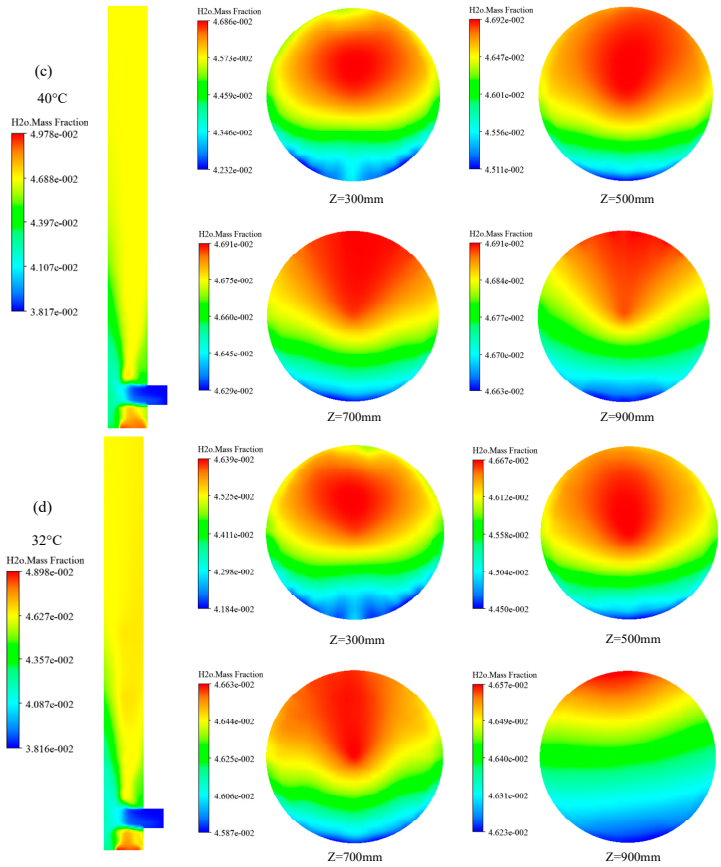

Figure 3. The distribution of temperature and water mass fraction with and without heterogeneous condensation: (**a**) temperature distribution without heterogeneous condensation; (**b**) temperature distribution with heterogeneous condensation; (**c**) water mass distribution without heterogeneous condensation; and (**d**) water mass distribution with heterogeneous condensation.

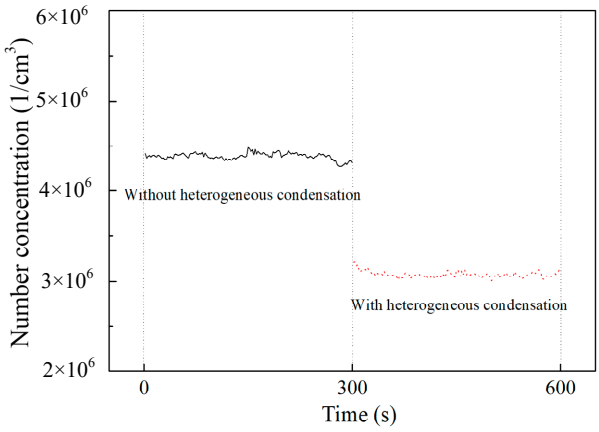

Figure 4. The effect of heterogeneous condensation on number concentration of fine particles.

Figure 7 illustrates the impact of heterogeneous condensation on the efficiency of fine particle removal. The removal efficiency for fine particles is 39.9% and 57.9% under Conditions 1 and 2, respectively, showing a significant increase. Heterogeneous condensation promotes the removal of fine particles by providing an environment in which the particles can nucleate, grow, and condense in supersaturated vapor, leading to more efficient removal.

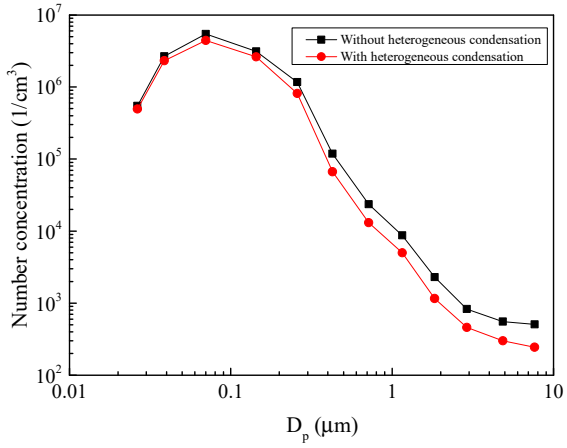

Figure 5. Effect of heterogeneous condensation on the size distribution of fine particles.

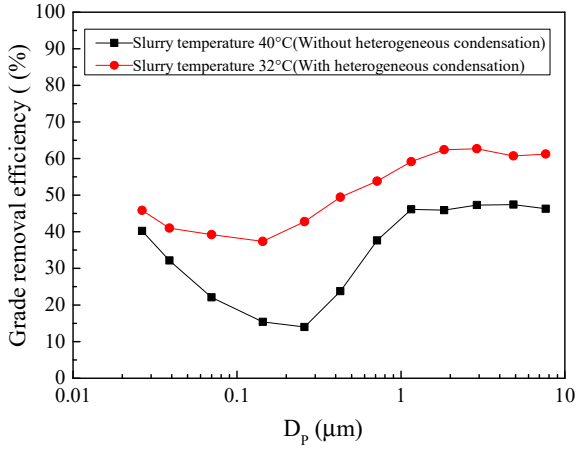

Figure 6. The effect of heterogeneous condensation on grade removal efficiency of fine particles.

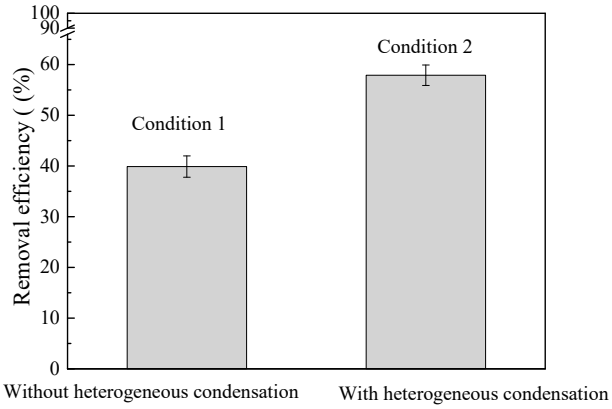

Figure 7. The removal efficiency of fine particles with and without heterogeneous condensation.

3.2. Influence of the Temperature Drop of the Desulfurized Slurry

Figure 8 illustrates the distribution of supersaturation in the desulfurization tower. The operating conditions include an unchanged desulfurization slurry temperature in the oxidation zone and a decreased temperature in the absorption zone. The inlet flue gas temperature is 120 °C, the desulfurization liquid–gas ratio is 15 L/Nm^3, and the desulfurization slurry temperature in the oxidation zone is 40 °C. The figure shows that as the relative height of the desulfurization tower increases, the supersaturation degree of the flue gas gradually increases. When the slurry temperature in the absorption zone is not cooled down, the supersaturation degree of flue gas is always less than 1, indicating that a supersaturated vapor environment

cannot be established in the desulfurization tower. However, when the desulfurization slurry in the absorption zone is cooled down by 8 °C, the flue gas begins to establish a supersaturated vapor environment at the position of 0.58 m/m relative to the tower height, and the supersaturation degree reaches 1.3393 and 1.3431 at this position (absorption zone). However, in the area from 0.25 m/m to 0.42 m/m (the oxidation zone) relative to the height of the tower, a supersaturated vapor environment still cannot be established. The supersaturated vapor environment can be achieved throughout the entire desulfurization tower by reducing the slurry temperature by 10 °C or 20 °C. Studies indicate that the critical degree of supersaturation required for the nucleation of fine particles during coal burning is approximately 1.15–1.2 [37]. Therefore, the majority of fine particles can nucleate and then grow into larger dust-containing droplets through heterogeneous condensation when the degree of supersaturation exceeds 1.2. In general, the critical conditions for establishing a supersaturated vapor environment in the desulfurization tower are as follows: the flue gas temperature at the inlet of the desulfurization tower is 120 °C, the desulfurization liquid–gas ratio is 15 L/Nm^3, the desulfurization slurry temperature in the oxidation zone is 40 °C, and the slurry temperature in the absorption zone is 32 °C (cooled down by 8 °C).

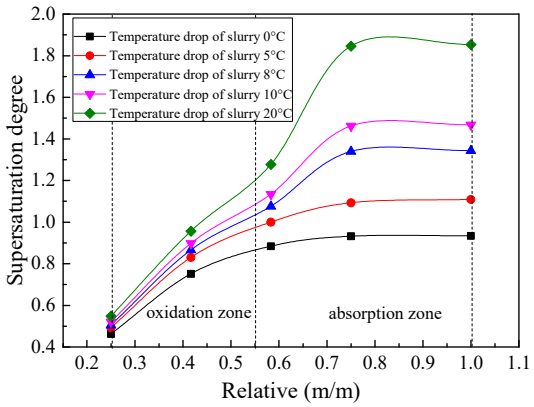

Figure 8. The effect of temperature drop of desulfurization slurry in absorption zone on supersaturation distribution in desulfurization tower.

Figure 9 illustrates the variation in fine particle number concentration at the exit of the desulfurization tower under the constant temperature of the slurry in the oxidation zone and varied temperatures of the slurry in the absorption zone. The fine particle concentration decreased from 7.32 × 10^6 1/cm^3 to 3.07 × 10^6 1/cm^3 with the temperature drop of the desulfurization slurry in the absorption zone increasing from 0 °C to 20 °C.

In Figure 10, the impact of the temperature drop of desulfurization slurry on the removal of fine particles is illustrated. During the experiment, the flue gas temperature at the inlet of the desulfurization tower was maintained at 120 °C, with a desulfurization liquid–gas ratio of 15 L/Nm^3. The desulfurization slurry temperature in the oxidation zone was 40 °C, and the temperature drop of desulfurization slurry in the absorption zone ranged from 0 °C to 20 °C. It is evident from Figure 10 that the removal efficiency of fine particles increases as the temperature drop of desulfurization slurry increases. Specifically, when the temperature drop of desulfurization slurry increases from 0 °C to 20 °C, the removal efficiency of fine particles increases from 39.9% to 78.7%. This can be attributed to the increased temperature difference between the desulfurization flue gas and the desulfurization slurry, which promotes the evaporation of the desulfurization slurry droplets into the gas phase, leading to an increase in the moisture content of the flue gas. Simultaneously, droplet evaporation absorbs heat, reducing the temperature of the flue gas, increasing the actual water pressure, decreasing the supersaturated vapor pressure, and ultimately resulting in an increase in supersaturation. Fine particles nucleate and grow in a supersaturated vapor environment when the flue gas in the desulfurization tower reaches a supersaturation higher than the critical supersaturation of fine particles, and are ultimately removed by an efficient defogger. From Figure 10, it is evident that the removal efficiency

of fine particles decreases gradually as the temperature drop of the desulfurization slurry increases from 0 °C to 7 °C, but increases significantly when the temperature drop increases from 8 °C to 10 °C. However, when the temperature drop of the desulfurization slurry increases to 20 °C, the increasing trend of fine particulate removal efficiency becomes slower. This phenomenon is mainly attributed to the establishment of a supersaturated vapor environment in the desulfurization tower and the range of this environment. As depicted in Figure 10, the degree of supersaturation formed in the desulfurization tower is lower than 1.15 when the temperature drop of the desulfurization slurry is less than 7 °C, and heterogeneous condensation cannot occur efficiently. Consequently, the removal efficiency of fine particles changes minimally when the temperature drop of the desulfurization slurry increases from 0 °C to 7 °C. A supersaturated vapor environment was created in the absorption zone of the tower when the temperature of the desulfurization slurry dropped to 8 °C, leading to heterogeneous condensation. As a result, the removal efficiency of fine particles increases significantly as the temperature drop increases from 8 °C to 10 °C. However, the removal efficiency of fine particles plateaued as the temperature drop increased from 10 °C to 20 °C. Despite a significant increase in the supersaturation degree due to the temperature drop of the desulfurization slurry, the regional variation of the established supersaturated vapor environment is not evident. Based on the nucleation rate formula for fine particles, the final size of grown particles does not increase linearly with the increase in the supersaturation degree. Thus, the particle size of fine particles is limited. Finally, the removal efficiency of fine particles increases slowly as the temperature drop of the desulfurization slurry increases from 10 °C to 20 °C.

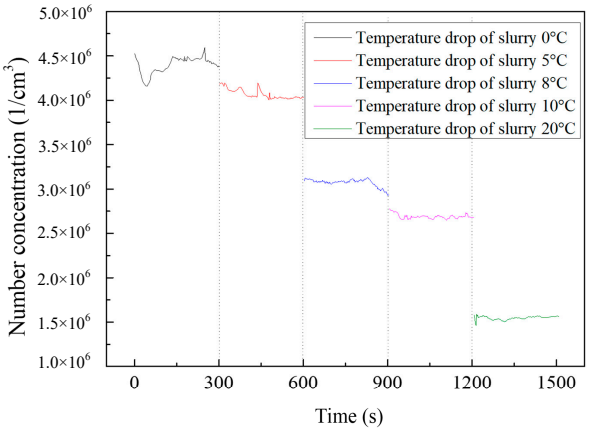

Figure 9. The effect of temperature drop of desulfurization slurry on the removal performance of fine particles.

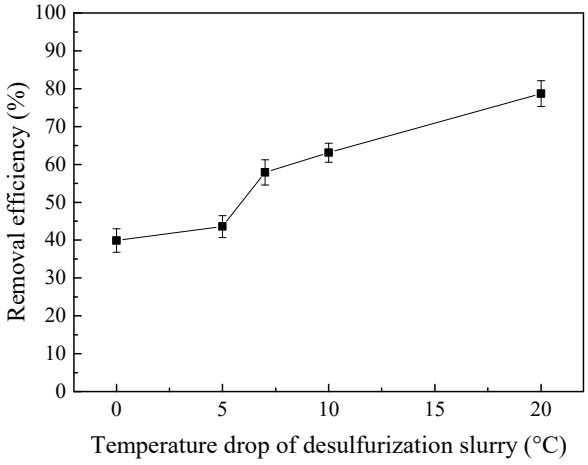

Figure 10. Relationship between temperature drop of desulfurization slurry and removal efficiency of fine particles.

4. Conclusions

An investigation was conducted to improve the efficiency of removing fine particles through heterogeneous condensation. The experiment established the necessary supersaturated vapor environment for heterogeneous condensation using single-tower double-cycle desulfurization technology. Numerical simulation was used to study the fluid dynamics characteristics in the desulfurization tower, and the impact of the temperature drop of the desulfurization slurry on the distribution of supersaturation in the tower was examined. Additionally, the study evaluated the effect of heterogeneous condensation based on single-tower double-cycle desulfurization technology on the removal efficiency of fine particles. The results indicate that by reducing the temperature of the desulfurization slurry in the absorption zone, a supersaturated vapor environment can be achieved, resulting in an increase in the supersaturation degree and region as the temperature drop of the slurry increases. Under typical working conditions, the most suitable temperature drop for the desulfurization slurry is approximately 8~10 °C. Furthermore, the supersaturation degree in the absorption zone increased from 0.93 to 1.85 with an increased temperature drop of the slurry. The use of single-tower double-cycle desulfurization technology can notably enhance the removal efficiency of fine particles, particularly for those with a particle size range of 0.1~1 μm. Ultimately, this method can elevate the removal efficiency of fine particles from 39.9% to 57.9%.

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Data Availability Statement: The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest: The authors declare no conflict of interest.

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