

## Research Article

# Heterogeneous Condensation for Abatement of Sulfuric Acid Aerosol and Water Saving

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Based on the fundamentals of heterogeneous nucleation, a method to eliminate sulfuric acid aerosol associated with water recycling in the process of limestone-gypsum desulfurization was investigated. The supersaturated environment was achieved in a heat exchanger. Numerical calculation shows that high temperature drop and relative humidity are conducive to the formation of supersaturated vapor environment, and vapor heterogeneous condensation can improve the removal efficiency of sulfuric acid aerosol. Experimental results indicate that the critical supersaturation degree of the sulfuric acid aerosol is found in inverse proportion to their sizes and the removal efficiency of sulfuric acid aerosol could be increased by about 20%. The theoretical and the actual condensable water mass values have been also studied in detail. The mass of condensed water produced by the experiment system is 0.0440 kg/(Nm<sup>3</sup>·h) as the temperature drop is 5°C, and the most suitable temperature drop is about 1~2°C for water scarce area. High temperature and humidity reveal a huge potential to recycling water.

## 1. Introduction

Air pollution has drawn human increasing attention all over the world. It has been reported that the pollution from coal combustion in the coal-fired power plant is one of the prime reasons [1–3]. In the coal-fired power plant, the emission of NO<sub>x</sub>, SO<sub>2</sub>, and dust is under the effective control in the development of pollutant control technology. However, the emission of sulfuric acid aerosol in the discharged flue gas is still a problem, which should be further investigated [4]. During the combustion of coal in boilers, most sulfur in coal is oxidized to SO<sub>2</sub> while partial SO<sub>2</sub> (0.5%~1.5%) would be oxidized to SO<sub>3</sub> at high temperature condition [5]. In addition, 0.5~2.0% SO<sub>2</sub> is oxidized to SO<sub>3</sub> by SCR catalyzer [6, 7]. Therefore, the concentration of SO<sub>3</sub> is comparatively high at the outlet of SCR. Meanwhile, with the decreasing of flue gas temperature, most SO<sub>3</sub> would convert into gaseous H<sub>2</sub>SO<sub>4</sub> at the outlet of air preheat (APH) [8]. When the temperature of flue gas is lower than 200°C and water content is 8% [9], 99% SO<sub>3</sub> would convert into gaseous

H<sub>2</sub>SO<sub>4</sub>. Hence, SO<sub>3</sub> exists as gaseous H<sub>2</sub>SO<sub>4</sub> in the flue gas at the inlet of the desulfurization tower. In the desulfurization process, the gaseous H<sub>2</sub>SO<sub>4</sub> condenses when the flue gas temperature decreases to the sulfuric acid dew point, resulting in the formation of sulfuric acid mist [10, 11].

A great number of methods have been investigated and developed to control the emission of SO<sub>2</sub> in coal-fired power stations [12–14]. The wet flue gas desulfurization (WFGD) is widely applied in the past 30 years due to high efficiency, low operating cost, and reliability in operation [15]. However, some new problems have been generated with the application of WFGD, such as gypsum rain and the generation of secondary particle [16]. Simultaneously, the water consumption amount of WFGD is huge because of the high relative humidity, low temperature desulfurized flue gas, and the water entrained by desulfurization slurry. The flue gas is nearly saturated and contains a lot of water after the desulfurization process, leading to the nucleation of fine aerosol after discharging into the atmosphere. The moisture loss brought away by the discharged flue gas would be huge,

and thus, efficiency of a boiler would decrease without effective measures [17]. In addition, the removal effect of sulfur trioxide by WFGD is not ideal even though the removal of sulfur dioxide may be effective. Therefore, developing an innovative approach contributed to preventing the formation of the sulfuric acid mist and reducing the water consumption of desulfurization cooperatively should be performed.

Combining pretreatment method with traditional dust removal equipment is more effective and efficient to control the emission of aerosol particles [18]. Heterogeneous vapor condensation is considered a promising pretreatment technology to eliminate the emission of aerosol in the coal-fired power plant [19, 20]. The mass and size of particles can be sufficiently enlarged via heterogeneous vapor condensation and then separated by inertial impaction. Researches have attempted to separate fine particles based on heterogeneous condensation [21–23]. Heidenreich and Ebert [24] investigated the fundamentals of droplet growth theory and found that submicron particles could be enlarged to droplets with a high growth rate at the effect of water vapor condensation. Chen et al. [25] found that the  $\text{SiO}_2$  and  $\text{TiO}_2$  particles could be activated and grown up in the supersaturated water vapor environment. However, these results are not suitable for coal-fired flue gas. Bao et al. [26] indicated that the removal of fine particles could be promoted by adding stream during wet flue gas desulfurization process. Wu et al. [27] proposed that the fine particles were separated efficiently with heterogeneous vapor condensation. However, they did not focus on the promotion of sulfur trioxide removal and water conservation.

The desulfurized flue gas is nearly supersaturated, allowing the supersaturation environment to form more easily [28]. In this work, the removal of sulfuric acid aerosol and water conservation based on heterogeneous condensation was investigated. The supersaturated vapor environment was achieved by a fluorine plastic heat exchanger installed at the outlet of the desulfurization tower. Additionally, the critical supersaturation degree of sulfuric acid aerosol was studied numerically. Sulfuric acid aerosol further nucleates and grows in this environment, and then the sulfuric acid aerosol is removed by the gravity and collision of droplets. Theoretical and actual values of condensate water mass were also studied. Meanwhile, effect of the characteristic parameters (temperature drop, initial temperature, and relative humidity) of the desulfurization flue gas on the removal efficiency of sulfuric acid aerosol was also discussed in detail. As a result, the most suitable parameters for the sulfuric acid aerosol removal and water saving were proposed.

## 2. Experiment System

**2.1. Experimental Setup.** The experimental apparatus, depicted in Figure 1, consists of a flue gas generator system, a buffer tank, an ESP, a WFGD system, a fluorine plastic heat exchanger, and a measurement system. The flue gas was supplied by an automatic coal-fired boiler with the volume of  $350 \text{ Nm}^3\text{h}^{-1}$ . The  $\text{SO}_3$  was provided by  $\text{SO}_3$  generator and

then injected into the flue gas to simulate the actual condition of coal-fired power plants. A buffer tank was used to maintain the uniform of the mixed flue gas. Then, the flue gas went through the ESP, desulfurization tower, and heat exchanger and subsequently discharged into the atmosphere. The coarse aerosol particle was separated by ESP. The desulfurization tower adapted spray tower and 3 levers' spray. Thus, the flue gas and the desulfurization slurry were countercurrent contact. The medium of the heat exchanger was cold water, and the temperature drop of desulfurized flue gas was controlled by regulating the flow rate of cold water. The bottom of the heat exchanger is a water tank, which was used to collect condensate water during the heterogeneous vapor condensation. Then, the mass of the condensation water was obtained and reused in desulfurization process. The  $\text{SO}_3$  generator is shown in Figure 2, where  $\text{SO}_3$  was produced by means of oxidation of  $\text{SO}_2$  by  $\text{O}_3$ . The  $\text{O}_3$  and  $\text{SO}_2$  were provided by  $\text{O}_3$  generator and  $\text{SO}_2$  cylinder gas. The amounts of  $\text{SO}_2$  and  $\text{O}_3$  were accurately controlled through the mass flow meter.

**2.2. Measurement Methods.** In the experiment, isokinetic sampling was adopted in the gas sampling and the  $\text{SO}_3$  concentration was measured based on controlled condensation method according to the national standard [29, 30]. As shown in Figure 3, the sampling system includes a sampling probe, a sampling gun, a filter cartridge, a serpentine glass collection tube, a thermostatic water bath, and a dust parallel sampler. In actual power plants, the typically sulfuric acid dew point ranges from 95 to  $160^\circ\text{C}$  [31]. Therefore, the flue gas sample was heated to  $180^\circ\text{C}$  to prevent the condensation of sulfuric acid in the sampling gun and then cooled down in the serpentine glass collection tube. The temperature of circulating cooling water was set at  $60^\circ\text{C}$ . The particles in the flue gas could be separated by heating quartz filter before water bath. After sampling, the serpentine glass collection tube was flushed with deionized water. Then, the concentration of  $\text{SO}_4^{2-}$  in the collected condensed fluid was measured by an ion chromatographic analyzer. As a result, the concentration of the sulfur trioxide was obtained. In addition, a humidity transmitter was used to measure the temperature and the humidity of the flue gas. The flue gas was sampled at the inlet and outlet of the heat exchanger, respectively.

The removal efficiency of sulfuric acid aerosol is determined by

$$\eta_s = \frac{C_0 - C_t}{C_0} \times 100\%, \quad (1)$$

where  $\eta_s$  is the removal efficiency of sulfuric acid aerosol (100%) and  $C_0$  and  $C_t$  are the concentration of sulfuric acid aerosol without and with a heat exchanger, respectively ( $\text{mg}/\text{m}^3$ ).

## 3. Numerical Study

**3.1. Critical Supersaturation of the Sulfuric Acid Aerosol.** Two basic conditions need to be fulfilled for the occurrence of nucleation. One is the formation of a supersaturation

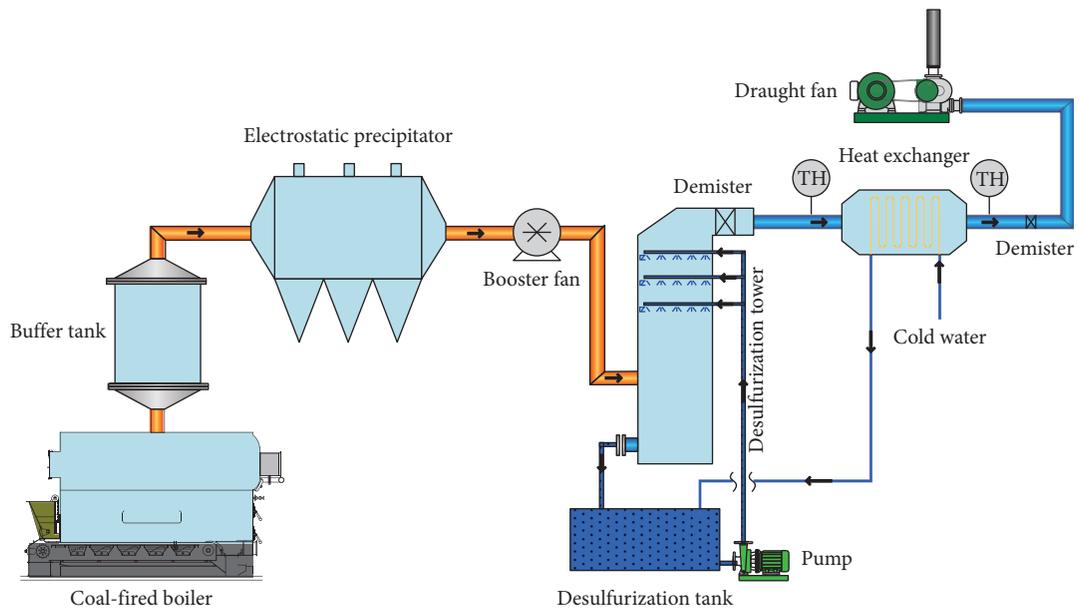


FIGURE 1: Schematic diagram of experimental setup.

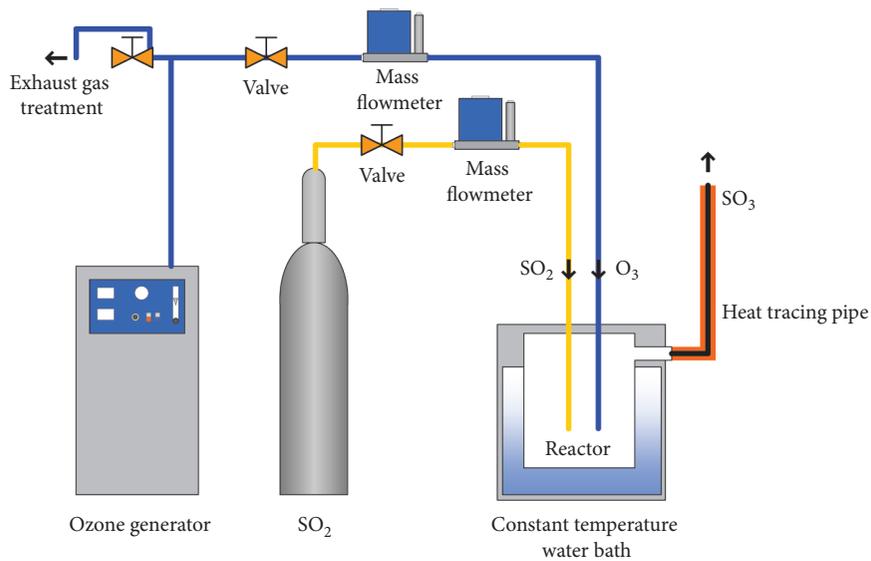
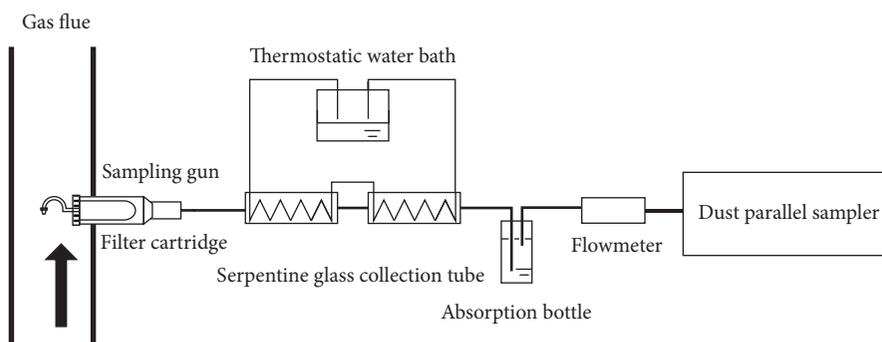
FIGURE 2: SO<sub>3</sub> acid mist generating device.

FIGURE 3: The sampling system of sulfuric acid aerosol.

vapor environment and the other is that the supersaturation degree of environment is higher than the critical supersaturation degree of the aerosol particles [32]. The physicochemical characteristics of the aerosol particles, such as the size, the surface structure, and the wetting properties with regard to the liquids, determine the critical supersaturation degree of the aerosol particles. Generally, the critical supersaturation degree for hydrophobic aerosol particles would be relatively lower when hydrophobic aerosol particles are covered by a liquid film. The critical supersaturation degree of sulfuric acid aerosol is low because of the hydrophilic property, proving that the sulfuric acid aerosol could nucleate and enlarge more easily compared with the hydrophobic particles in the same size. The critical supersaturation degree of the sulfuric acid aerosol  $S_{cr}$  can be defined as

$$S_{cr} = \frac{P_v}{P_s(T)} = \exp\left[\frac{2\sigma M_w}{R_G T \rho_w r}\right], \quad (2)$$

where  $P_v$  is the surface vapor pressure of the sulfuric acid aerosol (Pa),  $T$  is the temperature of flue gas (K),  $P_s(T)$  is the saturation vapor at temperature  $T$  (Pa),  $\sigma$  is the surface tension (N/m),  $M_w$  is the molar mass (kg/mol),  $R_G$  is the gas constant (J/(mol·K)),  $\rho_w$  is the density of the liquid (kg/m<sup>3</sup>), and  $r$  is the radius of the sulfuric acid aerosol (m).

Figure 4 presents the critical supersaturation degree as a function of the sizes of the sulfuric acid aerosol. The temperature of the flue gas is 55°C during the numerical calculation based on equation (2). As seen in Figure 4, the critical supersaturation degree of the sulfuric acid aerosol is in inverse proportion to its size. The bigger the size of the sulfuric acid aerosol, the lower the critical supersaturation degree of the sulfuric acid aerosol. Thereby, sulfuric acid aerosols with larger size (more than 0.1 μm) are easier to be nucleated and enlarged, whereas the sizes in the range of 0.01~0.1 μm require higher vapor supersaturation for the heterogeneous condensation. It can also be seen that the highest critical supersaturation degree is 1.19 for the sulfuric acid aerosol with 0.01 μm.

**3.2. Establishment of the Supersaturation Environment of the Desulfurized Flue Gas.** The supersaturation degree  $S$  required for vapor heterogeneous condensation is given by

$$S = \frac{P(T, x)}{P_x(T, x)}, \quad (3)$$

where  $P(T, x)$  is the actual vapor partial pressure at temperature  $T$  (Pa) and  $P_x(T, x)$  is the saturated vapor partial pressure at temperature  $T$  (Pa).

The condensable water vapor amount  $M$  is defined by

$$M = m_s - m, \quad (4)$$

where  $m_s$  is the moisture content at the ideal supersaturation degree  $S$  (g/Nm<sup>3</sup>) and  $m$  is the moisture content at the saturation degree (g/Nm<sup>3</sup>).

Figure 5 presents the effect of three main parameters (the relative humidity, temperature drop, and initial temperature

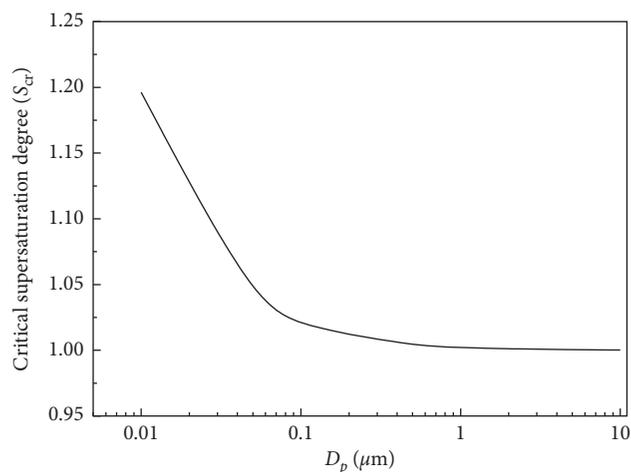


FIGURE 4: The critical supersaturation degree of sulfuric acid aerosol with different sizes.

of desulfurized flue gas) on the formation of supersaturation environment in desulfurized flue gas. Figure 5(a) is under the condition that the initial temperature was 55°C. It can be seen that the supersaturation degree increases rapidly with the increase of temperature drop, when the initial relative humidity is constant. Specifically, the supersaturation degree increases from 0.90 to 1.5 as the temperature drop increases from 0°C to 10°C at the relative humidity of 95%. Meanwhile, the supersaturation degree shifts to higher values as the relative humidity increases, when the temperature drop is constant. The supersaturation degree increases from 1.14 to 1.27 with the relative humidity shifting from 90% to 100%, when the temperature drop is 5°C. Thus, it can be concluded that the relative humidity and temperature drop contribute significantly to the establishment of the supersaturation degree, and the temperature drop of desulfurized flue gas has a greater influence in this work. Figure 5(b) shows that the supersaturation degree value increases with the increase of initial temperature at constant temperature drop. However, the increase amplitude of supersaturation degree is smaller. For the temperature drop 5°C, the supersaturation degree varies from 1.20 to 1.24 as the initial temperature decreases from 60°C to 40°C. Clearly, the amplitude variation of supersaturation degree is smaller. Therefore, the decrease of initial temperature has a little influence on the formation of supersaturation environment.

To identify the most beneficial parameters in the process of establishing supersaturation environment, a prediction equation fitted by multivariate nonlinear regression could be derived [33]. The calculated results, under the typical conditions in Table 1, were used to fit the equation. Therefore, the prediction equation is only suitable for these typical conditions, which can be described as

$$S = 0.01471 \cdot \Delta t^{0.22341} \cdot \phi^{1.00009} \cdot t^{-0.11397}. \quad (5)$$

Equation (5) can be locally calculated on a log-log scale as

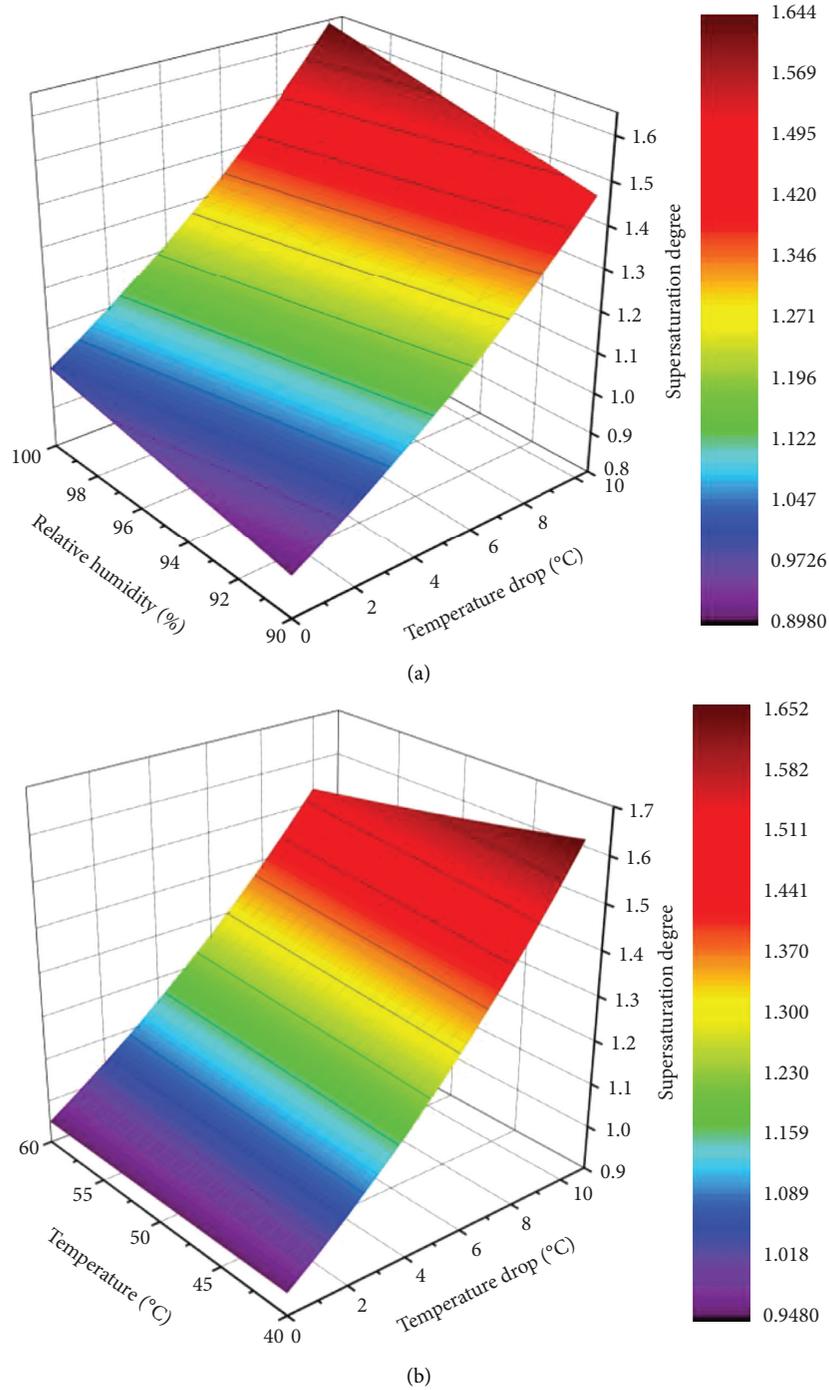


FIGURE 5: Numerical calculation results of supersaturation degree. (a) Influence of desulfurized flue gas relative humidity and temperature drop. (b) Influence of desulfurized flue gas temperature and temperature drop.

$$\ln S = \ln 0.01471 + 0.2234 \ln \Delta t + 1.00009 \ln \varphi - 0.11397 \ln t, \quad (6)$$

where  $\Delta t$  is the temperature drop of desulfurized flue gas (°C),  $\varphi$  is the relative humidity (%), and  $t$  is the temperature of desulfurized flue gas (°C).

We deduced the derivative of  $S$  with respect to  $\Delta t$ ,  $\varphi$ , and  $t$ , respectively. The result reveals that the influence of

TABLE 1: Typical conditions of thermodynamic calculation.

Operating parameters	Value of number (interval 1)
Desulfurized flue gas temperature	40°C~60°C
Desulfurized flue gas relative humidity	90%~100%
Desulfurized flue gas temperature drop	1~10°C

parameters on the establishment of supersaturation environment is  $\Delta t > \varphi > t$ . Obviously, higher temperature drop and relative humidity of the desulfurized flue gas are favorable to form the supersaturation environment, while the initial temperature of desulfurized flue gas is detrimental.

## 4. Results and Discussion

**4.1. Influencing Factors on the Removal Performance of Sulfuric Acid Aerosol.** To study the effect of temperature drop on the removal efficiency of sulfuric acid aerosol, the experiment was carried out at the temperature of 55°C and relative humidity of 95%. Figure 6 describes the relationship between the removal efficiency of sulfuric acid aerosol and the temperature drop. The removal efficiency increases with the increase of temperature drop. As the temperature drop increases from 2°C to 10°C, the removal efficiency shifts from 59.4% to 74.2%. This is mainly because the degree of vapor supersaturation, necessary for the nucleation of sulfuric acid aerosol, increases with the decrease of temperature. It can be seen from Figure 5(a) that the supersaturation degree increases to 1.5 from 0.9, as the temperature drop increases from 0°C to 10°C with instant initial temperature and relative humidity. As shown in Figure 4, the highest critical supersaturation degree for the sulfuric acid aerosol is 1.19. Thus, more submicron sulfuric acid aerosol would be activated and nucleated and then grown up into larger droplets at the effect of vapor heterogeneous condensation. The condensable water amount also increases with the increase of supersaturation degree, leading to higher condensable water amount divided into each droplet. The enlarged droplets are removed more effectively and easily at the effect of the high efficient demister and the gravity of sulfuric acid aerosol. Correspondingly, the temperature drop of desulfurized flue gas is controlled by the flow rate of the cold water in the fluorine plastic exchanger. As a result, the thermophoretic force and diffusiophoresis force are strengthened due to the increase of temperature gradient around the heat exchanger capillary, while the function of Brownian motion decreases [34–36]. Additionally, lower temperature enhances both the nucleation and growth rate of sulfuric acid aerosol [37]. Subsequently, sulfuric acid aerosols are removed to the surface of the heat exchanger capillary and captured by the condensate liquid film. Consequently, the removal efficiency is greatly enhanced due to the temperature drop of desulfurized flue gas.

Figure 7 presents the influence of initial temperature of desulfurized flue gas on the removal efficiency of sulfuric acid aerosol. The relative humidity and temperature drop were set at 95% and 5°C, respectively. It can be seen the tendency of the removal efficiency of sulfuric acid aerosol increases when the initial temperature rises. However, the variation is not significant. As depicted in Figure 5(b), the supersaturation degree reduces to 1.20 from 1.22 with the increase of initial temperature. The highest critical supersaturation degree of sulfuric acid

aerosol is 1.19, as shown in Figure 4. Therefore, the decrease of the vapor supersaturation degree has no or only minor effects on the removal of sulfuric acid aerosol. Nevertheless, the thermophoretic force and Brownian movement are enhanced during the increase of initial temperature. This is beneficial to the movement of sulfuric acid aerosol with small size, resulting in more sulfuric acid aerosol captured by the film. Hence, although the supersaturation degree decreases when the initial temperature increases, the removal efficiency of sulfuric acid aerosol tends to be at a higher value. This illustrates that the method has a good adaptability to the different initial temperature of desulfurized flue gas in this work, and the removal efficiency is almost unchanged with varying initial temperature.

Figure 8 shows the impact of relative humidity on the removal efficiency. The temperature drop was set at 5°C and the initial temperature was set at 55°C. It can be seen that the removal efficiency of sulfuric acid aerosol increases when the relative humidity increases. The removal efficiency increases from 64.3% to 70.1% with the relative humidity shifting from 90% to 100%. The range of relative humidity is less than 100%, while the desulfurized flue gas will reach supersaturation level as there is a 5°C drop of desulfurized flue gas. As seen in Figure 5(a), a higher supersaturation environment is formed with an increase in relative humidity, which plays a decisive role in the nucleation, activation, and growth of sulfuric acid aerosol. Then, more sulfuric acid aerosols with smaller particle size are activated and nucleated. The enlarged sulfuric acid aerosol would be removed more efficiently due to the collision and gravity of the sulfuric acid aerosol. Simultaneously, the sulfuric acid aerosols are easily captured by the liquid film on the surface of the heat exchange tube due to the strong hydrophilicity, inducing a higher removal efficiency of sulfuric acid aerosol.

**4.2. Water Saving Coupled the Removal of Sulfuric Acid Aerosol.** The relationships of the mass of condensable water changes with the initial temperature, temperature drop, and the relative humidity were calculated, as shown in Figures 9–11. Figure 9 shows the effect of initial temperature on the mass of condensable water. The temperature drop was set at 5°C and the relative humidity was set at 95%. The mass of condensable water increases with the increase of initial temperature of flue gas. This is because the water content is higher as the flue gas temperature is high under the same humidity. Figure 10 describes the relationship between the mass of condensable water and the temperature drop. The higher the temperature drop, the higher the mass of condensable water. Figure 11 presents the influence of the relative humidity on the mass of condensable water. The initial temperature was set at 55°C and the temperature drop was set at 5°C. The mass of condensable water increases when the relative humidity increases. This is because the water content of the flue gas is high when the relative humidity is high.

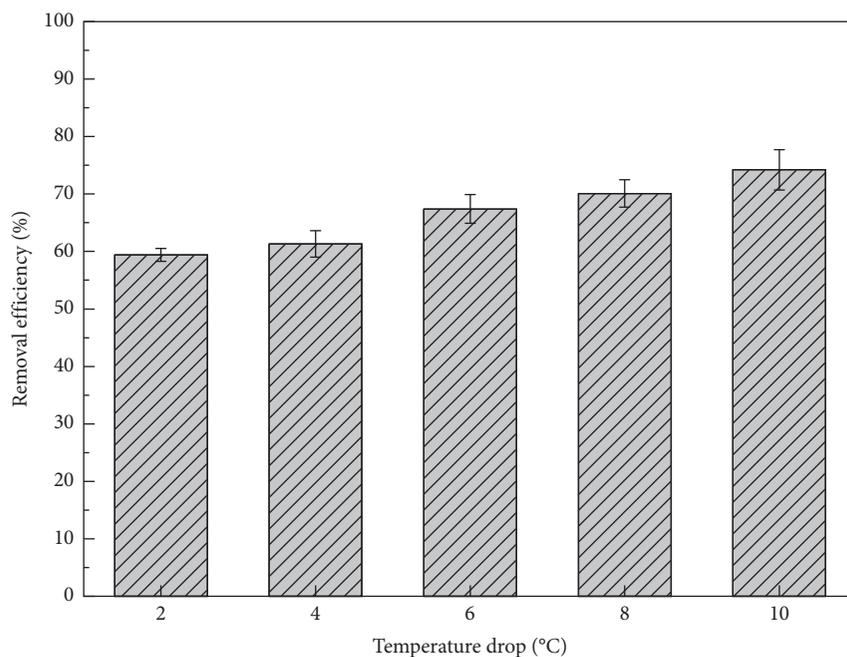


FIGURE 6: Effect of temperature drop on the removal efficiency of sulfuric acid aerosol.

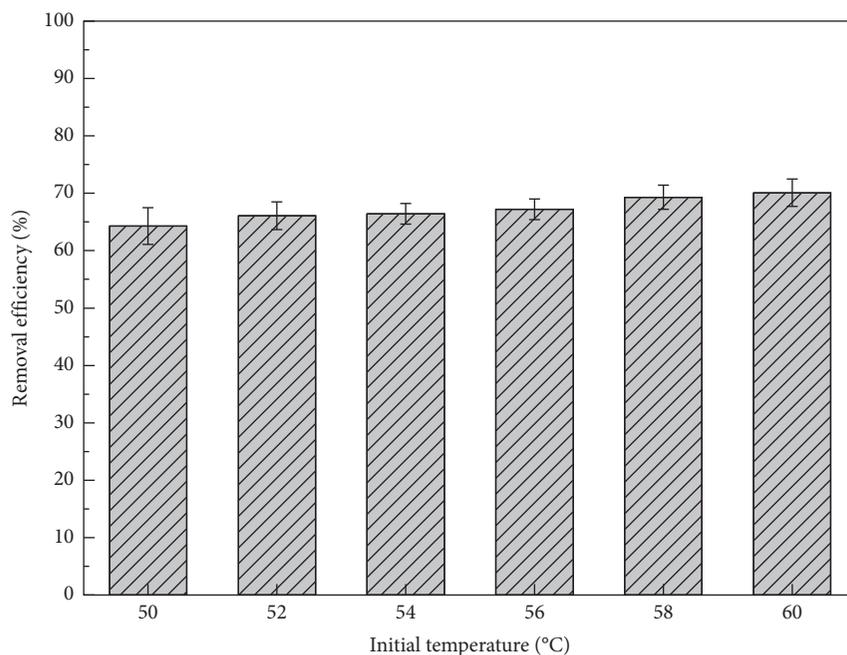


FIGURE 7: Effect of initial temperature on the removal efficiency of sulfuric acid aerosol.

The mass of condensable water increases with the increase of the initial temperature, temperature drop, and relative humidity. It can be found that high temperature and high humidity flue gas have a great potential to recover water. Meanwhile, desulfurized flue gas is high humidity. Therefore, it may be advantageous for saving water by

installing a heat exchanger after desulfurization. The desulfurized flue gas temperature reduces after entering into the heat exchanger. Water vapor homogeneously condenses into water droplets or heterogeneously condenses on the tube wall or aerosol surface. Then the condensed water is collected in the sink at the bottom of the heat exchanger.

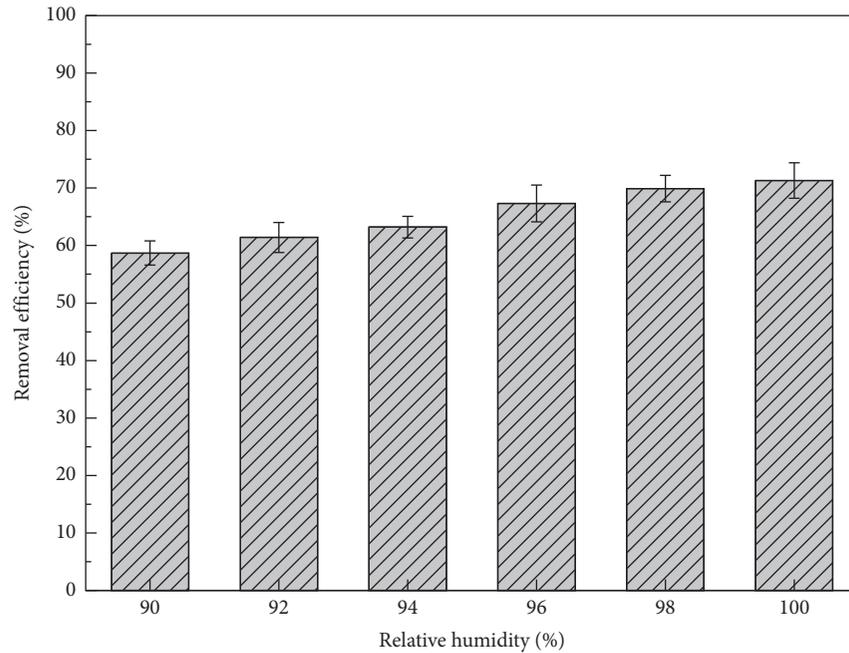


FIGURE 8: Effect of relative humidity on the removal efficiency of sulfuric acid aerosol.

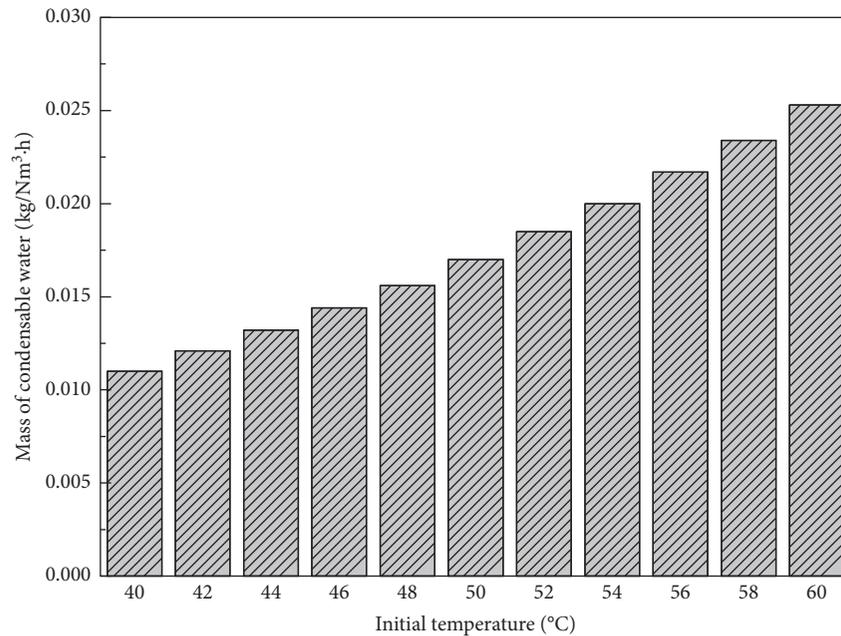


FIGURE 9: Effect of the initial temperature on the mass of condensable water.

Thus, the recovered condensation water volume could be obtained. Table 2 is the mass of condensed water with the heat exchanger under typical conditions. The initial temperature is 55°C and the relative humidity is 95%. The temperature drop increases from 1°C to 5°C. As seen in Table 1, the condensed water mass increases from 0.0046 kg/(Nm<sup>3</sup>·h) to 0.0349 kg/(Nm<sup>3</sup>·h). The higher temperature drop, the higher condensed water mass. When the

temperature is reduced by 1°C, the condensed water mass is 0.0046 kg/(Nm<sup>3</sup>·h). For a 300 MW system, it is approximately equal to 29.3714 kg/(Nm<sup>3</sup>·h), revealing the better water saving performance of the fluoroplastic heat exchanger. The results show that the more temperature drop, the more mass of condensed water. However, considering the cold source, the most suitable temperature drop is 1°C to 2°C for the arid area.

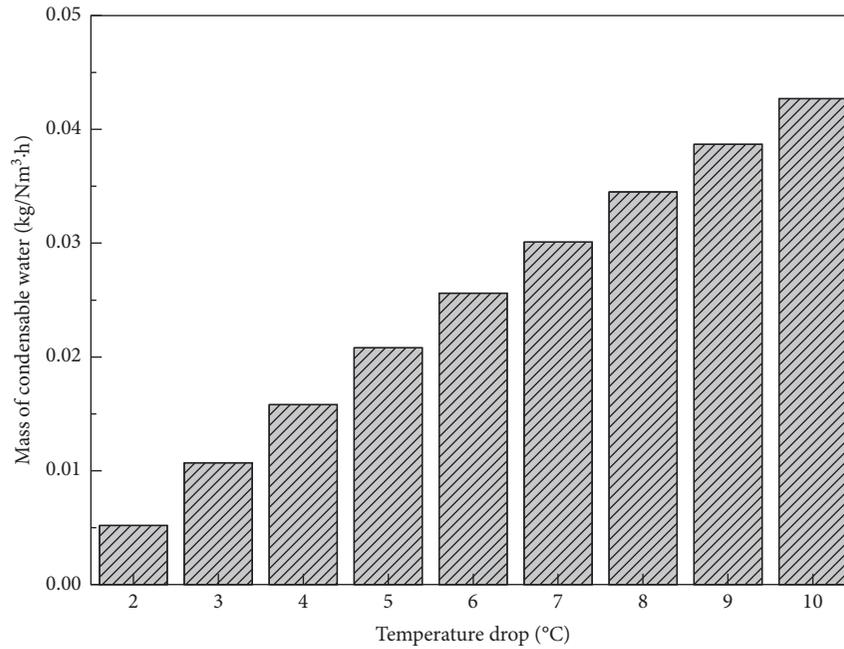


FIGURE 10: Effect of temperature drop on the mass of condensable water.

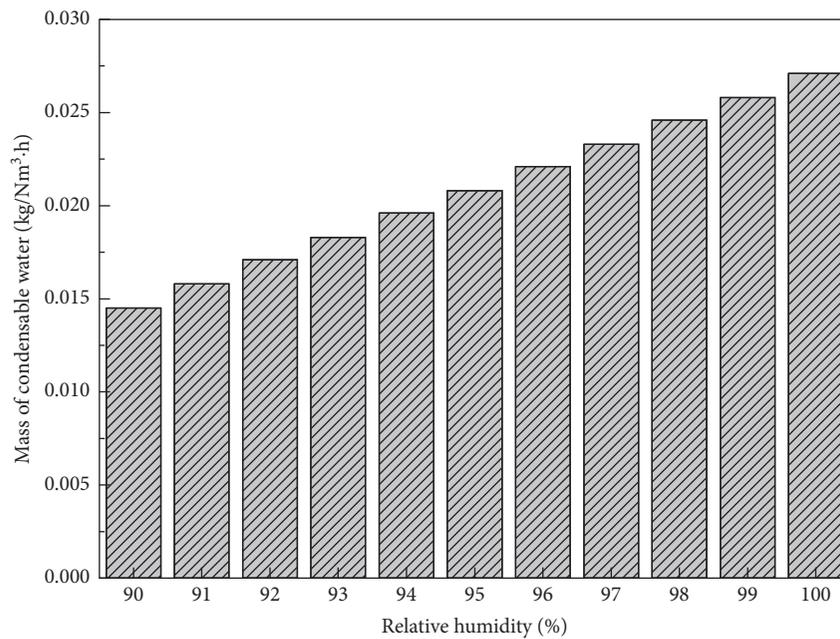


FIGURE 11: Effect of the relative humidity on the mass of condensable water.

TABLE 2: Recovered mass of condensed water with the heat exchanger on.

Number	Temperature drop (°C)	Condensed water mass (kg/(Nm <sup>3</sup> ·h))
1	2	0.0046
2	4	0.0160
3	6	0.0260
4	8	0.0349
5	10	0.0440

## 5. Conclusions

An approach to remove the sulfur acid aerosol coupling with water saving in the limestone-gypsum desulfurization system was investigated. The required supersaturation environment was achieved by the heat exchanger after the desulfurization. Numerical calculation of the supersaturation degree and condensable water mass was carried out. Attempts were made to probe the factors that affect the removal efficiency of sulfur trioxide. The results are discussed and summarized as follows:

- (1) The critical supersaturation degree of sulfuric acid aerosol decreases with the increasing size. The bigger the size, the lower the critical supersaturation degree. The highest critical supersaturation degree for sulfuric acid droplet is approximately equal to 1.2.
- (2) The removal of sulfuric acid aerosol is successfully achieved by the effect of heterogeneous vapor condensation. Correspondingly, the temperature drop has a strong impact on the separation of sulfuric acid aerosol. However, the removal efficiency of sulfuric acid aerosol changes little with the variation of the initial temperature. Therefore, this method is suitable for different initial temperature in the experimental conditions.
- (3) The relative humidity and temperature drop contribute significantly to supersaturation degree and the sulfuric acid aerosol removal efficiency, and the temperature drop of desulfurized flue gas has a greater influence.
- (4) When the temperature is reduced by 1°C, the condensed water mass is 0.0046 kg/(Nm<sup>3</sup>·h). This is equivalent to about 29.3714 kg/(Nm<sup>3</sup>·h) water saving for a 300 MW system. The more temperature drop, the more mass of condensed water. However, considering the cold source, the most suitable temperature drop is 1°C to 2°C for the arid area.

## Data Availability

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

## Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

## Acknowledgments

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