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# Study on the mass transfer of SO<sub>2</sub> in ammonia-based desulfurization process

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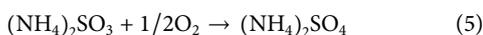
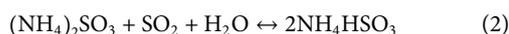
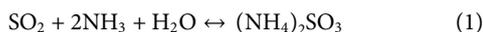
The coal hydrogen production process requires high-temperature steam as a reactant. Coal-fired boilers are usually used to provide high-temperature steam. The ammonia-based desulphurization process is an effective technology to remove SO<sub>2</sub> in the coal-fired flue gas. However, the mass transfer coefficient of SO<sub>2</sub> in such an engineered process is not readily available in the literature. In this paper, experiments were carried out to investigate the mass transfer process of SO<sub>2</sub> absorption. Combining calculating the air and liquid contact area of the droplet and liquid membrane on the tower wall, the mass transfer coefficient contain parameters pH, gas flue rate and liquid to gas ratio was proposed. The comparison results show that the calculated SO<sub>2</sub> mass transfer rate agrees well with the measured data. The relative error of the calculated and experimental values was less than ±20%. This proposed mass transfer coefficient appears to be beneficial for the design and operation of ammonia-based desulfurization FGD system.

## KEYWORDS

ammonia, desulphurization, mass transfer coefficient, absorption, modeling

## 1 Introduction

Sulfur dioxide (SO<sub>2</sub>) from coal fired power plants is known as a major air pollutant (Sun et al., 2010; Valle-Zermeño et al., 2014). The emission standard of SO<sub>2</sub> is increasingly strict in many countries and the flue gas is required to be treated before being discharged into the atmosphere. Among several desulfurization technologies, ammonia absorption based treatment process appears to be very attractive, as the resulting ammonium sulfate is a valuable by-product while meeting the emission standard of SO<sub>2</sub> (Gao et al., 2010; Jia et al., 2010; Wang et al., 2015). Compared with the widely used limestone-gypsum desulfurization method, the investment of ammonia desulfurization technology is about 23% lower and the operating cost is about 25% lower. The chemical reactions involved in the ammonia-based desulfurization processes are as following (He et al., 2003; Long et al., 2006; Jia et al., 2015):



In ammonia-based desulfurization process, removal of  $\text{SO}_2$  from flue gas by absorption is a major procedure (Ryuichi and Yukio, 1985). Spray scrubber is considered to be among the simplest, most economical and efficient absorption device for air pollution treatment (Javed et al., 2006; Zhong et al., 2008). In ammonia-based desulfurization process, the spray scrubber is also widely employed for removing  $\text{SO}_2$  from flue gas. The mass transfer rate of  $\text{SO}_2$  across the gas-liquid interface is an important system design parameter (Ryuichi and Yukio, 1985). The mass transfer coefficient can reflect the influence of operational condition on  $\text{SO}_2$  absorption rate, and theoretically guide the design and operation of spray scrubber, so as to make the design and operation of ammonia desulfurization system more accurate, and reduce the investment and operation cost. Accordingly, the mass transfer coefficient of  $\text{SO}_2$  absorption has attracted much attention of researchers.

By correlating with falling velocity of droplet, Henry constant and physical properties of solution *etc.*, Ryuichi developed a liquid-film mass transfer coefficient of the  $\text{SO}_2$  absorption by water droplets. The results of the research show that experimental values are smaller than the theoretical ones at contact times longer than 0.4 s (Ryuichi and Yukio, 1985). Hu also investigated the mass transfer coefficient by correlating with dimensionless number  $Re$ ,  $Sc$  and  $We$  (Hu et al., 2008). Marion deduced a gas-film mass transfer coefficient of the  $\text{SO}_2$  absorption by water drop, and the mass transfer coefficient was a function of the Sherwood number, molecular diffusivity and droplet diameter (Marion et al., 2006). Hixon and Scott investigated the gas phase volumetric mass transfer coefficient of  $\text{SO}_2$  absorption by sodium hydroxide solution, and the empirical equation contains parameters of gas flow rate, liquid flow rate and tower height was proposed (Hixon and Scott, 1935). Schmidt and Stichlmair conducted a research to investigate  $\text{SO}_2$  absorption in a spray scrubber and the results show that gas velocity has little influence on the mass transfer rate (Schmidt and Stichlmair, 1991). Milene investigated the absorption of  $\text{SO}_2$  by sodium hydroxide solution in a lab-pilot spray tower. The volumetric mass transfer coefficient expression has also been proposed, including three parameters: diameter of the spray nozzle orifice and exit velocity of liquid from the orifice, superficial gas and liquid velocities (Milene and Waldir, 2013).

Dimensionless numbers including Schmidt number  $Sc$ , Reynolds number  $Re$  and Sherwood number  $Sh$  are quantitatively correlated among physicochemical properties of the fluids such as density, viscosity and kinematic viscosity, *etc.* These physicochemical properties of the fluids are variable along

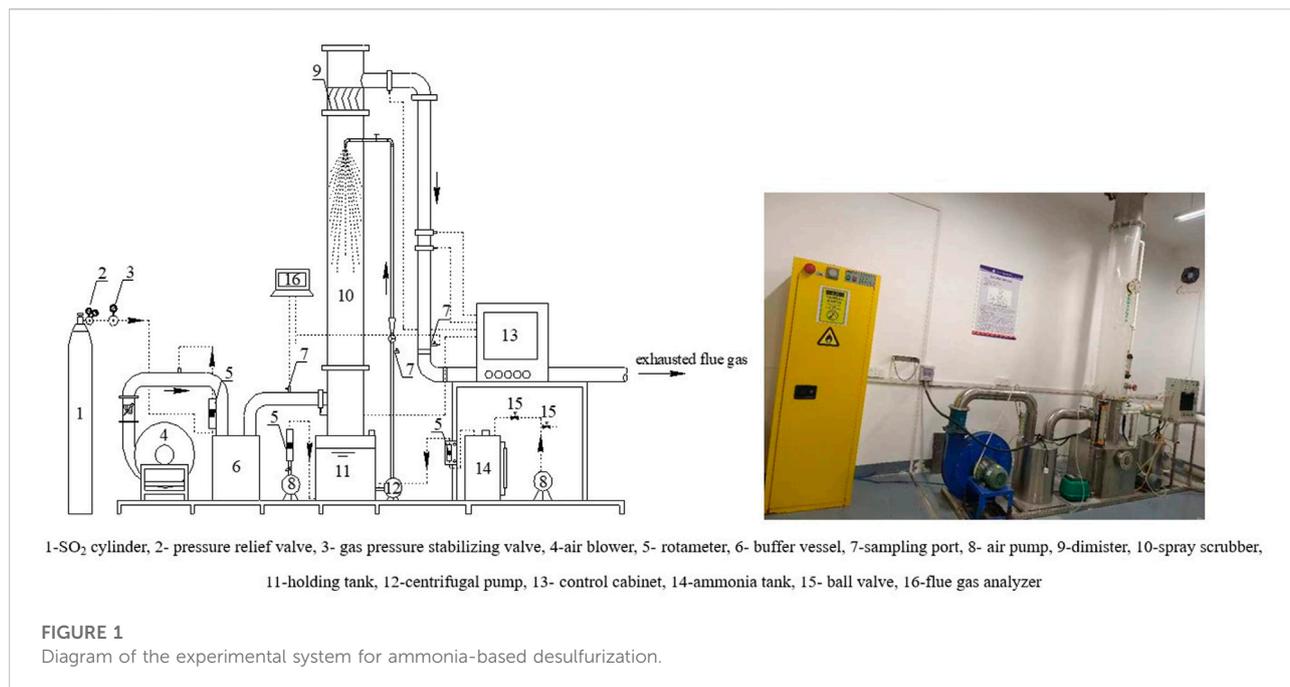
the height of absorption tower during the  $\text{SO}_2$  absorption process (Ryuichi and Yukio, 1985). Moreover, the gas-liquid interfacial area in the absorption tower is difficult to measure and the mass transfer coefficient of absorption is generally represented by overall volumetric gas phase mass transfer coefficient ( $K_G a$ ,  $\text{kmol}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{Pa}^{-1}$ ) (Hixon and Scott, 1935; Milene and Waldir, 2013; Bandyopadhyay and Biswas, 1998). However, the specific gas-liquid interfacial area per unit volume of absorption tower is highly depended upon the device and operating conditions. The specific gas-liquid interfacial area per unit volume of absorption tower is diverse in different systems. Although the mass transfer coefficient obtained by correlating with dimensionless number and the volumetric gas phase mass transfer coefficient are important parameters to evaluate the mass transfer process, they could not properly predict mass transfer rate of  $\text{SO}_2$  across the gas-liquid interface. On the other hand, extensive studies have been made on the mass transfer coefficient of absorption of  $\text{SO}_2$  by water,  $\text{NaOH}$  or  $\text{Na}_2\text{CO}_3$  solution, *etc.*, however, there are few investigations about mass transfer coefficient of  $\text{SO}_2$  absorption by ammonia solution. Accordingly, an independent study is needed.

Under operating conditions relevant to ammonia-based FGD plants, experiments were conducted to investigate the mass transfer rate of  $\text{SO}_2$  absorption by ammonia in a lab-scale spray scrubber in this work. Basing on experiments, a mass transfer coefficient of  $\text{SO}_2$  absorption by ammonia was also proposed. This mass transfer coefficient could predict the mass transfer rate of  $\text{SO}_2$  across the gas-liquid interface.

## 2 Experimental

Experiments were performed in a lab-scale spray scrubber. Figure 1 shows the schematic diagram of experimental setup. The spray scrubber was built of plexiglass and stainless steel, with an effective height of absorption zone 2.0 m and inner diameter of 0.3 m. A solid cone spray nozzle with 15° angle opening is installed at the top of the absorption zone. In the buffer vessel,  $\text{SO}_2$  was mixed with air to form simulated flue gas.  $\text{SO}_2$  (99.99 vol%) was supplied from a gas cylinder and the flow rate was controlled by a pressure stabilizing valve and a rotameter to ensure the desired  $\text{SO}_2$  concentration. The simulated flue gas was subsequently introduced into spray scrubber and countercurrently contacted with the descending liquid drops. The liquid drops and its absorbed  $\text{SO}_2$  fall into the holding tank located at the bottom of spray scrubber. The absorption liquid in the holding tank was circulated into the spray scrubber by a centrifugal pump. The amount of ammonia (17–20 wt%) was added into the solution at the front of centrifugal pump depended on the pH of solution.

In this work, experiments were conducted at solution pH level of 5.0–6.0, the gas flow rate ranging from 1.92 to 2.6  $\text{m}^3\cdot\text{s}^{-1}$ , and the liquid-to-gas ratio was between 3 and



5.5 L m<sup>-3</sup>. The pH of liquid sampled from the sampling port was monitored by pH meter (HANNA, HI198128). The inlet and outlet SO<sub>2</sub> concentrations were monitored using a flue gas analyzer (MRU Vario Plus, Accuracy is ±5 ppm). The simulated flue gas consisted of SO<sub>2</sub> (SO<sub>2</sub> mole fraction is 210 × 10<sup>-6</sup>) was stabilized for 1.5 h before measuring the inlet and outlet SO<sub>2</sub> concentration. In order to keep the sulfite concentration constant, a part of the solution was discharged according to the amount of SO<sub>2</sub> absorbed per hour. The total sulfite concentration (c<sub>S<sup>3+</sup></sub>) in the solution was about 0.05 mol L<sup>-1</sup>.

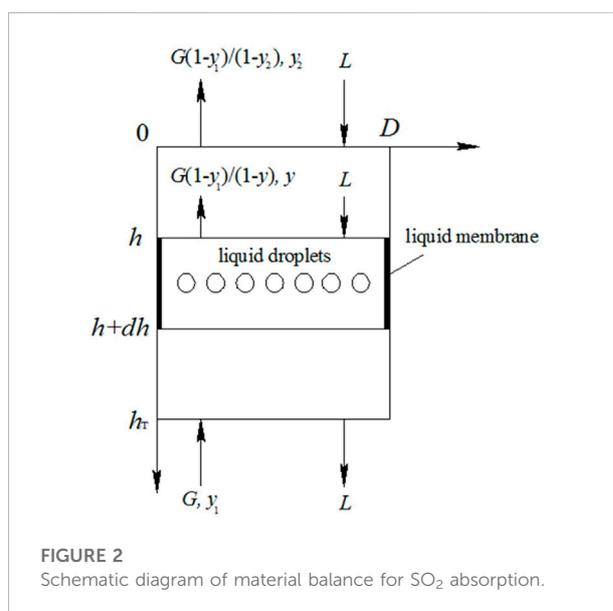
### 3 Theoretical

#### 3.1 Assumptions

- The falling drops are considered to be ball in shape and fall along vertical line.
- The collisions and coalescence between drops are neglected.
- The radial SO<sub>2</sub> concentration and velocity of flue gas are constant while the axial SO<sub>2</sub> concentration vary along the height of spray scrubber.
- Temperature is evenly distributed in the spray scrubber.

#### 3.2 Model

The physicochemical equilibrium is assumed at the liquid-gas interface and the mass transfer rate's equation can be

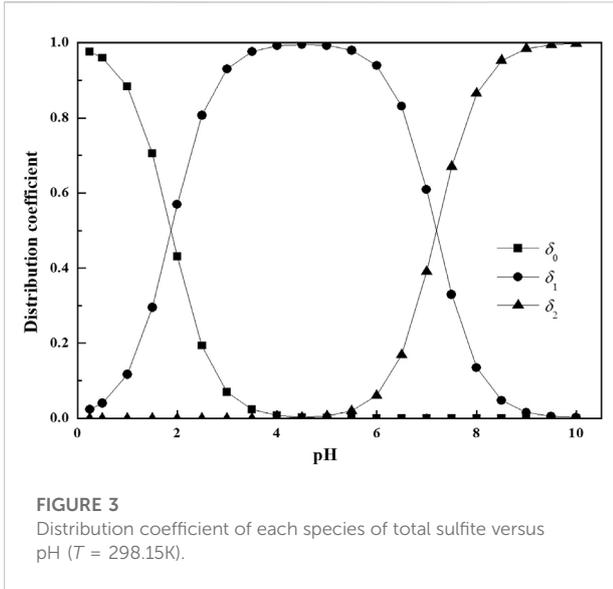


described as Eq. 6 in the process of SO<sub>2</sub> absorption (Colle et al., 2005).

$$N = K_G(p - p^*) \tag{6}$$

The overall gas-phase mass transfer coefficient  $K_G$  could be correlated by Eq. 7 (Liu et al., 2015):

$$\frac{1}{K_G} = \frac{1}{k_G} + \frac{1}{Hk_L} \tag{7}$$



Where,  $k_G$  is the individual gas phase mass transfer coefficient,  $k_L$  is the individual liquid phase mass transfer coefficient and  $H$  is the Henry coefficient.  $\text{SO}_2$  is easily soluble in water and the numerical value of Henry coefficient  $H$  is large. Accordingly, Eq. 7 can be simplified to Eq. 8:

$$\frac{1}{K_G} \approx \frac{1}{k_G} \quad (8)$$

Combining Eqs.6–8, the mass transfer rate’s equation of  $\text{SO}_2$  absorption can be transformed into:

$$N = k_G(p - p^*) = k_y(y - y^*) \quad (9)$$

Dividing the absorption zone of spray scrubber into small height incremental volumes (Figure 2), in each element, the material balance for  $\text{SO}_2$  absorption could be described as (Colle et al., 2005):

$$Na = k_y a \cdot (S \cdot dz) \cdot (y - y^*) = Q(1 - y_1) \frac{y}{1 - y} |z + dz - Q(1 - y_1) \frac{y}{1 - y} |z \quad (10)$$

Eq. 10 could be transformed into:

$$\frac{k_y a \cdot (S \cdot dz) \cdot (y - y^*)}{dz} = Q(1 - y_1) \frac{d}{dz} \left( \frac{y}{1 - y} \right) \quad (11)$$

In ammonia-based desulfurization process,  $\text{SO}_2$  is absorbed by dilute ammonia and the total sulfite ( $\text{SO}_2(\text{aq})$ ,  $\text{HSO}_3^-$  and  $\text{SO}_3^{2-}$ ) is formed in the liquid phase. The distribution coefficients of  $\text{SO}_2(\text{aq})$  ( $\delta_0$ ),  $\text{HSO}_3^-$  ( $\delta_1$ ) and  $\text{SO}_3^{2-}$  ( $\delta_2$ ) could be calculated and the distribution coefficients of each species versus pH are shown in Figure 3 (Jia et al., 2010, 2015).

In ammonia-based desulfurization process,  $\text{SO}_2$  is commonly absorbed in the pH range from 5.0 to 6.0. It can be observed from

Figure 3 that the distribution coefficient of  $\text{SO}_2(\text{aq})$  is nearly zero and the  $\text{SO}_2(\text{aq})$  concentration in solution could be neglected over the pH range of 5.0–6.0. According to Henry’s Law, it can be concluded that the mole fraction of  $\text{SO}_2$  in gas phase is much bigger than the equilibrium mole fraction of  $\text{SO}_2$  at gas-liquid interface ( $y \gg y_i$ ). Moreover, the mole fraction of  $\text{SO}_2$  in gas phase is much smaller than 1. Therefore, Eq. 11 can be transformed into:

$$G \frac{1}{y} dy = k_y a dh \quad (12)$$

Integrating Eq. 12 from top to bottom of the absorption zone along the vertical direction, yields:

$$\int_{y_2}^{y_1} G \frac{1}{y} dy = \int_0^{h_T} k_y a dh \quad (13)$$

The numerical integration of Eq. 13 allowed to compute the mass transfer rate:

$$k_y = \frac{G}{ah_T} \ln \left( \frac{y_1}{y_2} \right) \quad (14)$$

Where,  $a$  is the specific surface area which contains surface area of liquid droplet ( $A_d$ ) and liquid film ( $A_m$ ). The specific surface area ( $a$ ) could be calculated by the following equation:

$$a = (A_d + A_m) / \pi R^2 h_T \quad (15)$$

The surface area of liquid droplet ( $A_d$ ) in the element can be expressed as:

$$A_d = \frac{1}{3600} \times \frac{L_d}{\pi d^3} \times \pi d^2 \times \frac{dh}{u_d} \quad (16)$$

In Eq. 16,  $d$  is the Sauter mean diameter of droplets and it can be calculated by Eq. 17; (Wu et al., 1995):

$$d = 133.0 \cdot (D/2) \cdot We^{-0.74} \quad (17)$$

where:

$$We = \frac{\rho(u_d^0)^2 (D/2)}{\sigma} \quad (18)$$

In Eq. 18, the initial velocity of liquid droplet out of the nozzle  $u_d^0$  could be calculated according to the law of conservation of momentum:  $m_l u_d^0 = P_l \pi d_k^2 / 4$ . The equation of motion for describing the movement of falling liquid droplets in the absorption zone could be determined as Eqs.19–22 (Zhong et al., 2008; Michalski, 2000):

$$\frac{du_d}{dt} = g \left( \frac{\rho_d - \rho_g}{\rho_d} \right) - \frac{3}{4} \left( \frac{\rho_g (u_d - u_g)^2 C_d}{\rho_d g} \right) \quad (19)$$

$$\int_0^{t_z} u_d dt = dh \quad (20)$$

In Eq. 18, the drag coefficient could be calculated by Eq. 20 (Zhong et al., 2008; Michalski, 2000):

$$C_d = \frac{24}{Re_d} (1 + 0.125Re_d^{0.72}) \quad (21)$$

where:

$$Re_d = \frac{d|u_d - u_g|\rho_g}{\mu_g} \quad (22)$$

The surface area of liquid film ( $A_m$ ) in the element could be expressed as:

$$A_m = \pi(D - 2h_m) \cdot dh \quad (23)$$

The flow of the liquid wall film is assumed to be in the laminar and the thickness of the liquid wall film can be determined as (Dai and Chen, 1988):

$$h_m = 0.369 \left( \frac{3\nu^2}{g} \right)^{\frac{1}{3}} Re_m^{0.5} \quad (24)$$

Combining Eqs.14–16 and Eq. 23 yields:

$$k_y = \frac{G}{(A_d + A_m)h_T} \ln\left(\frac{y_1}{y_2}\right) \quad (25)$$

## 4 Results and discussion

### 4.1 Determination of the mass transfer coefficient

Frequently, the  $K_Ga$  was correlated in the dependency on the flow rate of gas and liquid in literature (Milene and Waldir, 2013). The mass transfer rate of SO<sub>2</sub> absorption by ammonia is influenced by several factors, such as pH of solution (pH), gas flow rate ( $\nu$ ) and liquid-to-gas ratio ( $m$ ). The pH of solution and the gas- and liquid-phase mixing have great influences on the mass transfer rate of SO<sub>2</sub> absorption by ammonia. On basis of experiments, the mass transfer coefficient  $k_y$  could be related to pH,  $\nu$  and  $m$  by means of multivariable nonlinear regression and the expression is in the following form:

$$k_y = k \cdot pH^a \nu^b m^c \quad (26)$$

where  $k$ ,  $a$ ,  $b$  and  $c$  are the coefficients for the respective parameters modeled in the proposed equation mentioned above.

In this work, the inlet and outlet concentration of SO<sub>2</sub> were measured at various operating conditions of pH,  $\nu$  and liquid-to-gas ratio  $m$ . Subsequently, the mass transfer rate of SO<sub>2</sub> absorption at different operating conditions were calculated according to Eqs.16, 23 and 25. The experimental conditions and experimental mass transfer rate obtained are presented in Table 1.

TABLE 1 Experimental mass transfer rate of SO<sub>2</sub> absorption.

$\nu$ (m•s <sup>-1</sup> )	pH	$m$ (L•m <sup>-3</sup> )	T (K)	$k_y$ (kmol•m <sup>-2</sup> •h <sup>-1</sup> )
2.4	5.51	2	298.15	6.12
2.4	5.51	2.5	298.15	6.85
2.4	5.51	3	298.15	7.52
2.4	5.51	3.5	298.15	7.69
2.4	5.51	4	298.15	8.52
2.4	5.51	4.5	298.15	9.13
2.4	5.51	5	298.15	10.05
2.4	5.51	5.5	298.15	10.82
2.4	5.51	6	298.15	11.33
2.4	5.02	3	298.15	6.2
2.4	5.2	3	298.15	6.56
2.4	5.35	3	298.15	6.92
2.4	5.38	3	298.15	7.06
2.4	5.51	3	298.15	7.65
2.4	5.58	3	298.15	8.04
2.4	5.79	3	298.15	8.49
2.4	5.88	3	298.15	8.71
2.4	5.92	3	298.15	9.01
1.92	5.51	3	298.15	7.29
2.21	5.51	3	298.15	7.37
2.4	5.51	3	298.15	7.43
2.6	5.51	3	298.15	7.48
2.21	5.02	3	298.15	5.97
2.21	5.07	3	298.15	6.05
2.21	5.08	3	298.15	6.07
2.21	5.47	3	298.15	7.35
2.21	5.67	3	298.15	7.95
2.21	5.85	3	298.15	8.42
2.21	5.97	3	298.15	8.56
2.21	5.51	2	298.15	5.71
2.21	5.51	2.5	298.15	6.44
2.21	5.51	3	298.15	7.37
2.21	5.51	3.5	298.15	7.88
2.21	5.51	4	298.15	8.49
2.21	5.51	4.5	298.15	8.92
2.21	5.51	5	298.15	9.52
2.21	5.51	5.5	298.15	10.42
2.21	5.51	6	298.15	11.56
1.92	5.51	2	298.15	5.01
2.21	5.51	2	298.15	5.82
2.4	5.51	2	298.15	6.03
2.6	5.51	2	298.15	6.53

Eq. 27 is obtained by taking natural logarithm on both sides of Eq. 26:

$$\ln k_y = \ln k + a \ln pH + b \ln \nu + c \ln m \quad (27)$$

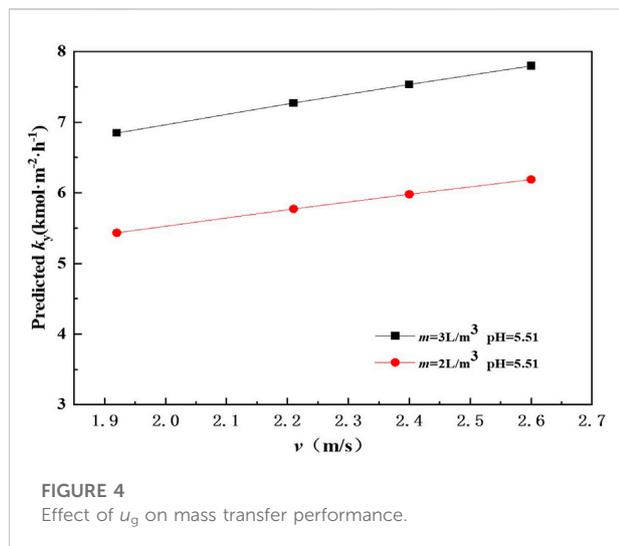


FIGURE 4 Effect of  $u_g$  on mass transfer performance.

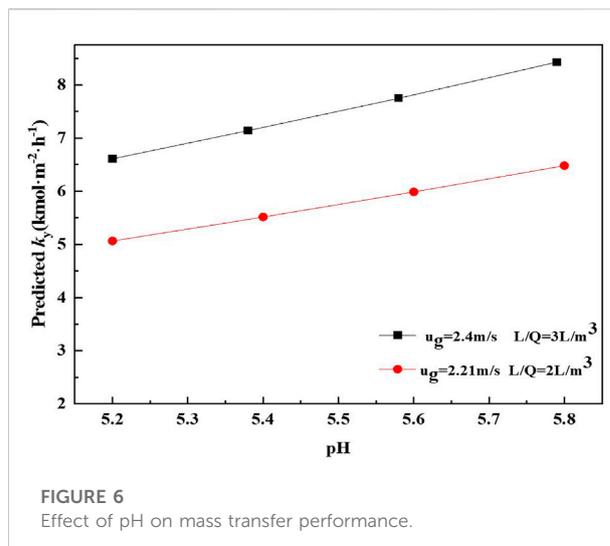


FIGURE 6 Effect of pH on mass transfer performance.

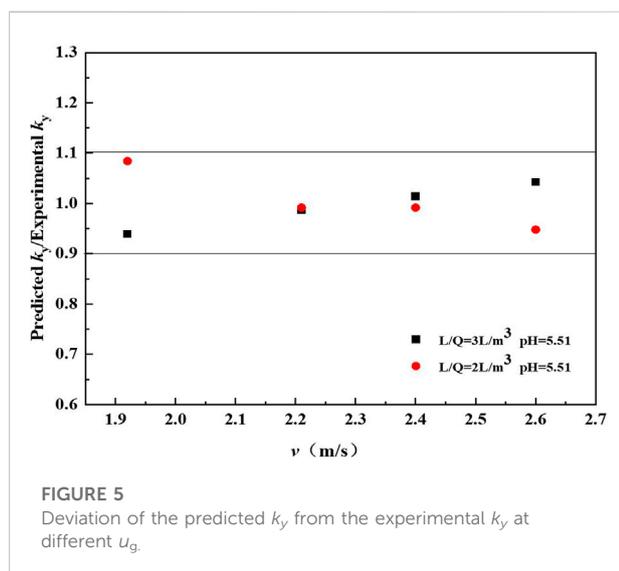


FIGURE 5 Deviation of the predicted  $k_y$  from the experimental  $k_y$  at different  $u_g$ .

27 at  $v$  range from 1.92 to 2.6 m/s. It can be seen from Figure 4 that the predicted mass transfer rate of  $\text{SO}_2$  absorption increases as  $v$  increases. When pH is 5.51 and  $m$  is  $2 \text{ L m}^{-3}$ , the predicted mass transfer rate of  $\text{SO}_2$  absorption by ammonia solution is about  $5.43 \text{ kmol m}^{-2} \text{ h}^{-1}$  at  $v$   $1.92 \text{ m s}^{-1}$ . The predicted mass transfer rate reaches  $6.19 \text{ kmol m}^{-2} \text{ h}^{-1}$  when  $u_g$  increases to  $2.6 \text{ m s}^{-1}$  at  $m$   $2 \text{ L m}^{-3}$ . A similar tendency has also been observed by Milene and Waldir (Milene and Waldir, 2013). It could be explained as that raising  $v$  results in an increase of gas-liquid mixing. Meanwhile, the residence time of liquid drops in the spray scrubber increase as  $v$  increases. An examination of Figure 4 also shows that the predicted mass transfer rate of  $\text{SO}_2$  absorption by ammonia at  $m$   $3 \text{ L m}^{-3}$  is larger than that at  $m$   $2 \text{ L m}^{-3}$ .

Figure 5 compares the mass transfer rate predicted by the proposed correlation Eq. 27 and the experimental data. Approximately 100% of the predicted points lie within an uncertainty of  $\pm 10\%$  of the values obtained experimentally. The predicted values agree well with the measured data.

$\ln(v)$ ,  $\ln(\text{pH})$  and  $\ln(m)$  are independent variables in the three-variable linear Eq. 27. Basing on data listed in Table 1, the mass transfer coefficient ( $a$ ,  $b$  and  $c$ ) of  $\text{SO}_2$  absorption is related to pH,  $v$  and  $m$  by means of multivariable linear regression and the expression of Eq. 26 is as follows:

$$k_y = 5.8 \times 10^{-2} \times v^{0.43} \text{pH}^{2.26} m^{0.57} \quad (28)$$

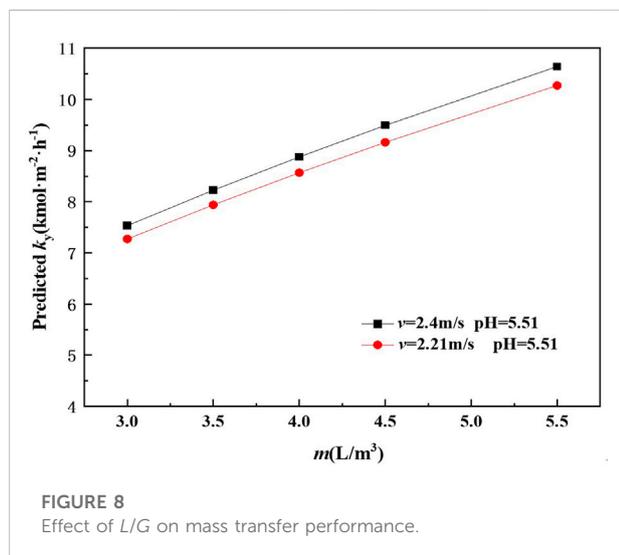
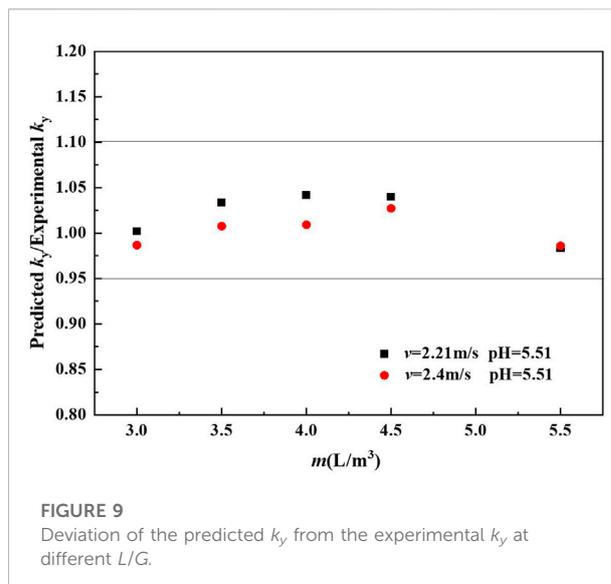
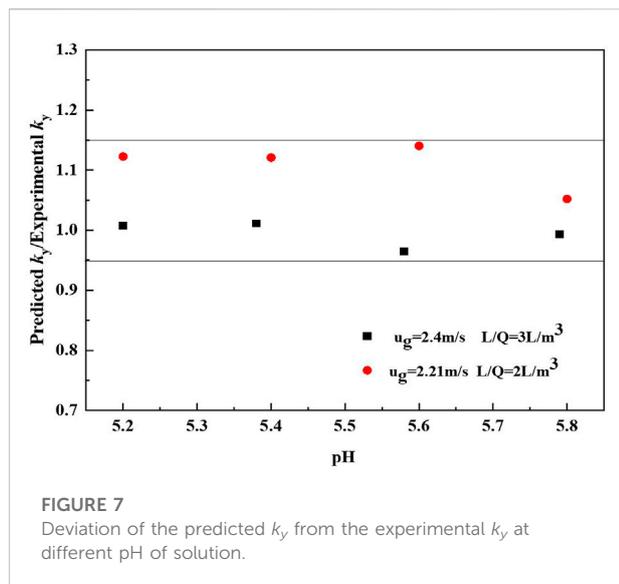
## 4.2 Effect of $v$ on mass transfer performance

In order to examine the effect of gas flow rate on mass transfer performance, the mass transfer rate was calculated by Eq.

## 4.3 Effect of pH on mass transfer performance

The effect of pH of solution on the mass transfer rate predicted by Eq. 27 is presented in Figure 6.

The results show that the mass transfer performance in terms of  $k_y$  increases as the pH of solution increases. The similar tendency has also been reported by Mounsef (Mounsef et al., 2015). The possible reason is that the absorption of  $\text{SO}_2$  has faster reaction kinetics at higher pH than that at lower pH. The higher the pH of solution, the more sulfite ions could react with  $\text{SO}_2$  and hence a more intense mass transfer performance is observed.



### 4.4 Effect of $m$ on mass transfer performance

The mass transfer rate of  $\text{SO}_2$  absorption by ammonia was also calculated at different liquid-to-gas ratio ( $m$ ) and the results was present in Figure 8.

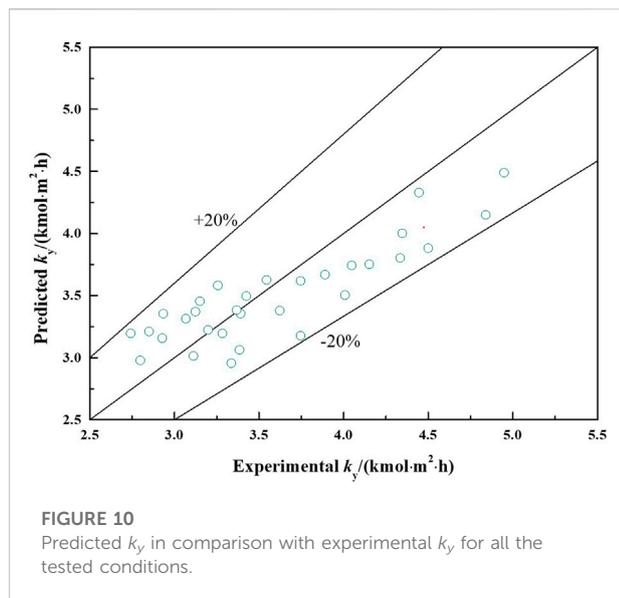
Figure 8 shows that the predicted mass transfer rate profile increases as  $m$  increases over the range of 3.0–5.5  $\text{L m}^{-3}$ . When the pH is 5.51 and  $v$  is 2.21  $\text{m s}^{-1}$ , the predicted mass transfer rate of absorption of  $\text{SO}_2$  by ammonia is about 7.53  $\text{kmol m}^{-2}\cdot\text{h}^{-1}$  at  $m$  3  $\text{L m}^{-3}$ . The predicted mass transfer rate is about 8.88  $\text{kmol m}^{-2}\cdot\text{h}^{-1}$  as  $m$  increases to 4  $\text{L m}^{-3}$ . The reason for such observation may be explained as, with the increase in the  $m$ , the total gas-liquid interfacial area also increased (Ma et al., 2013). However, the power consumption of the recirculating pump increased as  $m$  increases. Meanwhile, with the increase in the  $m$ , the coalescence of the droplets increased while the specific gas-liquid interfacial area increases slowly. Moreover, Figure 8 also shows that the mass transfer rate obtained at conditions of  $v$  2.21  $\text{m s}^{-1}$  and pH 5.51 is lower than that obtained at conditions of  $v$  2.4  $\text{m s}^{-1}$  and pH 5.51. The possible reason is similar as explained in Sections 4.2 and 4.3.

Figure 9 compares the mass transfer rate predicted by the proposed correlation Eq. 27 and the experimental values.

Approximately 100% of the predicted values lie within the uncertainty of  $\pm 5\%$  of the experimental results. Moreover, it can be seen from Figure 9 that the deviation between the predicted values and experimental values increases as  $m$  decreases over the range of 3.0–4.5  $\text{L m}^{-3}$ . It could be reasoned that the droplets are not evenly distributed in the spray scrubber at low  $m$  in this study.

Regard to the results observed in Figure 6 and Eq. 27, it can be inferred that pH has a significant effect on the mass transfer performance for  $\text{SO}_2$  absorption by ammonia. Obviously, it can also be seen from Figure 6 that the mass transfer rate obtained at conditions of  $v$  2.21  $\text{m s}^{-1}$  and  $m$  2  $\text{L m}^{-3}$  is lower than that obtained at conditions of  $v$  2.4  $\text{m s}^{-1}$  and  $m$  3  $\text{L m}^{-3}$ . It can be observed from Eq. 27 that the mass transfer rate is proportional to  $v$  and  $m$ . Meanwhile, the influence of  $v$  on mass transfer rate of  $\text{SO}_2$  absorption is more significant than  $m$ .

Figure 7 compares the values of predicted  $k_y$  and the experimental values. Obviously, nearly 100% of the predicted points lie within an uncertainty of  $\pm 15\%$  of the values obtained experimentally.



## 4.5 Validation of predicted $k_y$

The comparison between the experimental mass transfer rate and the predicted mass transfer rate was conducted to verify the reliability of this proposed mass transfer coefficient (Eq. 27). The inlet and outlet concentration of  $\text{SO}_2$  were measured at different operating conditions such as  $v$ , pH and  $m$ , then the mass transfer rate were calculated by Eq. 25. The corresponding predicted mass transfer rate at the same conditions were also calculated by the mass transfer coefficient in Eq. 28. The gas flow rate  $v$  ranged from 1.92 to 2.6 m/s, the pH of solution ranged from 5.2 to 5.8 and the liquid-to-gas ratio  $m$  ranged from 3 to 5.5 L/m<sup>3</sup>.

Figure 10 shows the predicted mass transfer rate *versus* the measured values of a lab-scale ammonia-based desulfurization system. The comparison reveals that the relative error of the predicted values and the measured data is almost within  $\pm 20\%$ . The mass transfer rate of absorption of  $\text{SO}_2$  by ammonia predicted by Eq. 28 agree well with the measured values. This proposed mass transfer coefficient appears to be beneficial for the design and operation of ammonia-based desulfurization FGD system.

## 5 Conclusion

The mass transfer performance of  $\text{SO}_2$  absorption by ammonia has been studied in a lab-scale spray scrubber. Experiments were conducted by varying operating conditions of  $v$ , pH and  $m$ . On basis of experimental results, the functional relationship between  $k_y$ ,  $v$  and  $m$  was deduced by

means of multivariable linear regression and the mass transfer coefficient of absorption of  $\text{SO}_2$  by ammonia was proposed as following:

$$k_y = 5.8 \times 10^{-2} \times v^{0.43} \text{pH}^{2.26} m^{0.57} \quad (29)$$

The mass transfer rate of  $\text{SO}_2$  absorption by ammonia at different operating conditions was calculated by the equation mentioned above. The mass transfer coefficient was verified against measured values of a lab-scale ammonia-based desulfurization system. The comparison results show that the predicted mass transfer rate agrees well with the measured data. The relative deviation between predicted mass transfer rate and measured data is almost within  $\pm 20\%$ . The mass transfer coefficient proposed could predict the mass transfer rate of  $\text{SO}_2$  across the gas-liquid interface. Accordingly, the mass transfer coefficient proposed in this work would be beneficial for the design and operation of ammonia-based desulfurization system.

## Data availability statement

The original contributions presented in the study are included in the article/supplementary material, further inquiries can be directed to the corresponding author.

## Author contributions

YJ: Conceptualization, Methodology, Investigation. GY: Validation, Formal analysis, Writing—original draft. DW: Validation, Formal analysis, Visualization, Writing—review and editing. YG and YD: Validation, Formal analysis, Visualization. JJ, YC, MZ, and CS: Resources, Writing—review and editing. LG: Validation, Formal analysis.

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## Conflict of interest

DW was employed by Xi'an Aerospace Yuan Dongli Engineering Co., Ltd.

The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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## Nomenclature

### Abbreviations

- $a$  Gas-liquid mass transfer contact area ( $\text{m}^2/\text{m}^3$ )
- $A_d$  The surface area of droplets in a spray scrubber ( $\text{m}^2$ )
- $A_m$  Liquid film surface area of spray tower ( $\text{m}^2$ )
- $C_d$  Drag coefficient
- $d$  Sauter mean diameter of droplet (m)
- $d_k$  Inner hole diameter of nozzle,  $4 \times 10^{-3}$  m
- $D$  The diameter of tower (m)
- $G$  Flue gas flow rate ( $\text{kmol}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ )
- $g$  Acceleration of gravity ( $\text{m}\cdot\text{s}^{-2}$ )
- $h_m$  The thickness of spray wall liquid film (m)
- $H$  Henry coefficient
- $k_0$  coefficient
- $K_G$  The overall volumetric mass transfer coefficients ( $\text{kmol}\cdot\text{m}^{-3}\cdot\text{h}^{-1}\cdot\text{Pa}^{-1}$ )
- $k_G$  The overall mass transfer coefficients in the gas phase ( $\text{kmol}\cdot\text{m}^{-3}\cdot\text{h}^{-1}\cdot\text{Pa}^{-1}$ )
- $k_L$  The mass transfer coefficients in the liquid phase ( $\text{kmol}\cdot\text{m}^{-3}\cdot\text{h}^{-1}\cdot\text{Pa}^{-1}$ )
- $k_y$  The mass transfer rate ( $\text{kmol}\cdot\text{m}^{-3}\cdot\text{h}^{-1}\cdot\text{Pa}^{-1}$ )
- $k'_y$  The mass transfer coefficient ( $\text{kmol}\cdot\text{m}^{-3}\cdot\text{h}^{-1}\cdot\text{Pa}^{-1}$ )
- $L$  Liquid flow rate ( $\text{L}\cdot\text{h}^{-1}$ )
- $L_d$  Slurry spray amount in the presence of liquid drop ( $\text{L}\cdot\text{h}^{-1}$ )
- $L_m$  Tower wall slurry flow ( $\text{kg}\cdot\text{s}^{-1}$ )
- $m_1$  mass flow rate of liquid from nozzle ( $\text{kg}\cdot\text{s}^{-1}$ )
- $N$  The mass transfer rate of  $\text{SO}_2$  ( $\text{kmol}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ )
- $p$  The pressure of  $\text{SO}_2$  in gas phase (Pa)
- $P_1$  Operating pressure of nozzle (MPa)
- $p^*$  Interfacial equilibrium partial pressure of  $\text{SO}_2$  in gas phase (Pa)
- $Re_d$  Reynolds number
- $Re_m$  The Reynolds number of Liquid film flow
- $u_d^0$  Initial velocity of droplet ( $\text{m}\cdot\text{s}^{-1}$ )
- $u_d$  The velocity of droplet ( $\text{m}\cdot\text{s}^{-1}$ )
- $u_g$  The velocity of gas ( $\text{m}\cdot\text{s}^{-1}$ )
- $We$  Weber number.
- $y_1$  Inlet mole fraction of  $\text{SO}_2$
- $y_2$  Outlet mole fraction of  $\text{SO}_2$
- $y^*$  Equilibrium mole fraction of  $\text{SO}_2$  at gas-liquid interface.
- $y$  Mole fraction of  $\text{SO}_2$  in gas phase
- $Z$  Height of absorption area of spray tower (m)
- $\eta$   $\text{SO}_2$  removal efficiency (%)
- $\rho$  Slurry density ( $\text{kg}\cdot\text{m}^{-3}$ )
- $\mu$  Viscosity of slurry (Pa·s)
- $\nu$  Kinematic viscosity of slurry ( $\text{m}^2\cdot\text{s}^{-1}$ )
- $\sigma$  Surface tension of slurry (N·m).