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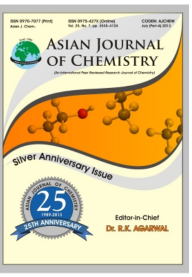

Studies on Magnesium-Based Wet Flue Gas Desulphurization Process with a Spray Scrubber

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This paper presents a gas-liquid absorption reaction between $Mg(OH)_2$ slurry and SO_2 for magnesium-based wet flue gas desulphurization (FGD) with a spray scrubber. Experimental investigations on effects of different operating variables show the reasonable process parameters such as the gas flow rate, the SO_2 inlet concentration in the gas, the liquid-gas ratio, the byproduct concentration in the slurry and the pH value of the slurry. An interesting phenomenon is also found under different byproduct concentrations. A desulphurization process model is established by the double-film theory of mass transfer. The calculated mass transfer coefficients are in reasonable agreement with the experimental values and the absorption process is found to be controlled by gas-film diffusion. These results would be useful for the design or process optimization of the magnesium-based wet FGD system.

Key Words: Wet flue gas desulphurization, Magnesium oxide, Spray scrubber, Mass transfer model.

INTRODUCTION

As China became the largest SO_2 emission country in the world after 1999, the State Environmental Protection Administration has targeted to reduce the national SO_2 emission level since 2000, which required that all new thermal power units as well as the most existing ones must have flue gas desulphurization (FGD) systems installed[1]. The domestic research on flue gas desulphurization is becoming the hottest topic in environmental science and engineering over the years[2].

The sulphur dioxide removal was first applied in 1930s, nearly 100 different FGD processes and process variations have been studied. However, only a few results have been applied in the industry[3]. Among these techniques, wet FGD processes are the most widely used in most countries because of their high desulphurization efficiency. The magnesium-based wet FGD process is one of the techniques to meet these requirements, which has been increasingly used on the industrial boiler in China because of its low investment, compact flow sheet, less land occupied, high efficiency, reliable operation, rare fouling and low price of the magnesium-based absorbent[4-12]. $MgSO_4$ is the byproduct from magnesium-based wet FGD process and can be sold as fertilizer.

Though the FGD processes have been studied by a number of researchers, it should be noted that the magnesium-based wet FGD process has not been the subject of many studies. The present work was undertaken to make a systematic study of the magnesium-based wet FGD process with a spray scrubber.

EXPERIMENTAL

A schematic diagram of the experimental apparatus is shown in Fig. 1. The spray scrubber is an organic glass cylindrical tower with 150 mm in internal diameter and 800 mm in gas-liquid contact height. The simulated flue gas in the experiments was prepared by mixing compressed air and gaseous SO_2 reagent. The absorption slurry [converted into $Mg(OH)_2$, 1 wt %] was prepared by mixing industrial grade MgO and tap water, which was stored in a constant-temperature slurry tank for 10 L under 25 ºC.

Air and SO_2 were introduced through the gas flowmeters as well as the gas mixing and heating chamber, which then flowed into the bottom of the spray scrubber and countercurrently contacted with the absorption slurry in the spray scrubber. The SO_2 concentration of the outlet gas was determined and the outlet gas was further absorbed by NaOH solution. A diaphragm pump was used to deliver the absorption slurry to a spiral nozzle, then the absorbed slurry returned to the slurry tank through the pipes on the bottom of the spray scrubber. The absorption processes were used with full absorbent cycle (discontinuous until the absorbent was used up) and without refreshing of the absorbent. The pH value of the absorption slurry was determined during the desulphurization process.

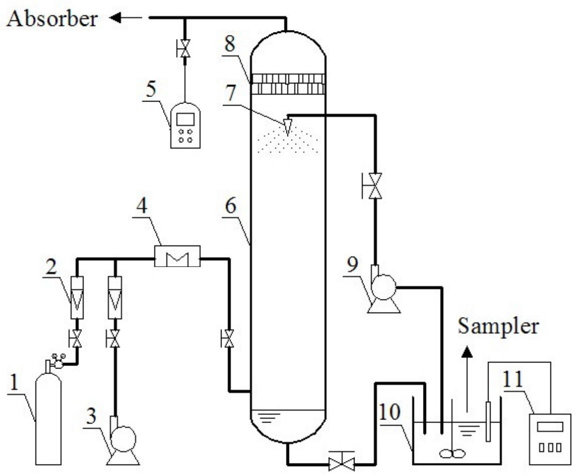

Fig. 1. Schematic diagram of the experimental apparatus (1- SO_2 cylinder; 2- gas flowmeter; 3- air compressor; 4- gas mixing and heating chamber; 5- flue gas analyser; 6- spray scrubber; 7- spiral nozzle; 8- demister; 9- slurry pump; 10- constant- temperature slurry tank; 11- pH meter)

The simulated flue gas flow rate (Q_G), the SO_2 concentration in the inlet flue gas (y_i), the ratio of the absorption slurry to the simulated flue gas flow (R_{LG}) and the byproduct concentration in the absorption slurry (C_{MgSO_4}) were changed in the experiment and the above procedure was repeated.

The gaseous SO_2 reagent (purity ≥ 99.9 %) was purchased from Shanghai Shenkai Gases Technology Co., Ltd. The composition of the industrial grade MgO was shown in Table-1, which was measured by XRF. The concentration of SO_2 in the gas was analyzed by the flue gas analyser (testo 335, testo AG). The pH value of the slurry was measured by the pH meter (DELTA 320, Mettler-Toledo International Inc.). In the following experiments, η_A is defined as the desulphurization efficiency to evaluate the absorption effects:

$$\eta_A = \frac{y_i - y_o}{y_i} \times 100\ \% \qquad (1)$$

where y_i and y_o are the inlet and outlet concentrations of SO_2 in the gas phase, respectively.

RESULTS AND DISCUSSION

Desulphurization experiments

Effect of the gas flow rate: Fig. 2 shows the effect of the gas flow rate on the desulphurization efficiency. Generally, the reduction of the gas flow rate will extend its residence time in the scrubber, which will also extend the gas-liquid contacting time and cause a higher absorption efficiency. Thus the absorption efficiency should be gradually reduced with the increase of the gas flow rate. However, the liquid-gas ratio was a constant in this section and caused the corresponding change of the slurry flow rate with the gas flow rate. When the gas flow rate was 15 Nm^3/h, the nozzle pressure could not reach the rated value and caused a poor atomization effect, resulting in a lower desulphurization efficiency of the system. When the gas flow rate increased to 25 Nm^3/h, the atomization effect was stable at the normal level while the gas-liquid contacting time was shortened, which caused the poor desulphurization efficiency. As the fresh absorbent would not be added during the absorption process, the desulphurization efficiency decreased tardily and maintained at the higher level for a long time with the consumption of the absorbent and then decreased rapidly with the empty of the absorbent at the end of absorption. In order to improve the desulphurization efficiency, good atomization effect of the nozzle and an appropriate extension of the gas-liquid contacting time are necessary in the actual industrial running.

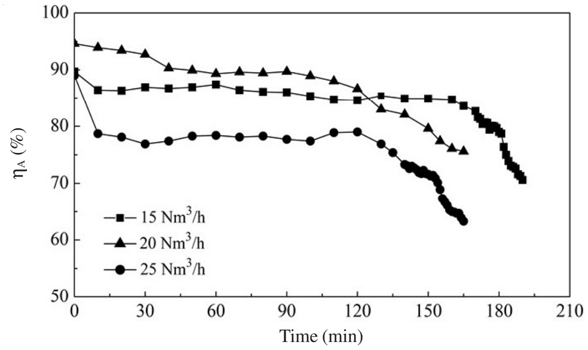

Fig. 2. Effect of the gas flow rate on the desulfurization efficiency (y_i = 2000 mg/Nm^3; R_{LG} = 8 L/Nm^3; C_{MgSO_4} = 0 g/L; T = 25 ºC)

Effect of the SO_2 inlet concentration: Fig. 3 shows that the desulphurization efficiency decreases with the SO_2 inlet concentration increases. A high SO_2 inlet concentration can enhance the driving force of the SO_2 mass transfer from the gas into the slurry, however, the total SO_2 load also increases at the same time. Thus, with the increase of the SO_2 inlet concentration, the absorbed SO_2 in the slurry raises but the desulphurization efficiency still goes down. The experimental results show that the magnesium-based wet FGD process is more suitable for the low SO_2 concentration flue gas desulphurization and the impact of SO_2 load should be fully considered for the design and operation of the desulphurization system.

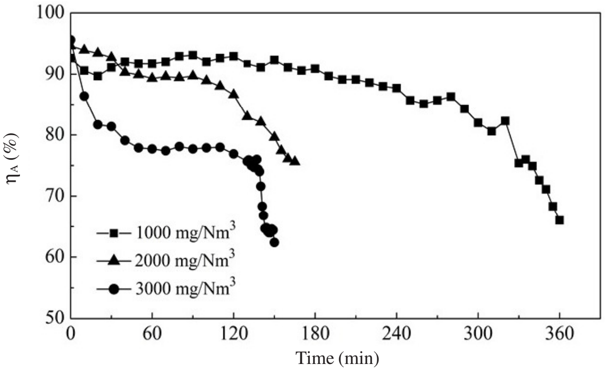

Fig. 3. Effect of the SO_2 inlet concentration on the desulfurization efficiency (Q_G = 20 Nm^3/h; R_{LG} = 8 L/Nm^3; C_{MgSO_4} = 0 g/L; T = 25 ºC)

TABLE-1
COMPOSITION OF THE INDUSTRIAL GRADE MgO

Ingredient	MgO	SiO_2	CaO	Al_2O_3	Fe_2O_3	SO_3	Cl	K_2O
wt (%)	90.66	6.32	1.88	0.415	0.397	0.134	0.0679	0.0341

Effect of the liquid-gas ratio: Fig. 4 shows the effect of the liquid-gas ratio R_{LG} on the desulphurization efficiency. The liquid-gas ratio is one of the most important parameters in desulphurization process. The increase of the liquid-gas ratio will directly raise the gas-liquid contacting area, greatly accelerate the mass transfer and improve the desulphurization efficiency. However, the increase of the liquid-gas ratio will also cause the increase in energy consumption in actual industrial running. Thus, the liquid-gas ratio should be controlled at a moderate level in order to minimize the running cost and to reach the acceptable desulphurization efficiency.

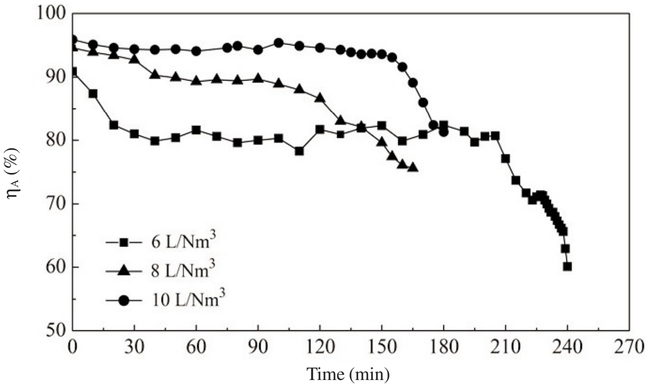

Fig. 4. Effect of the liquid-gas ratio on the desulfurization efficiency (Q_G = 20 Nm^3/h; y_i = 2000 mg/Nm^3; C_{MgSO_4} = 0 g/L; T = 25 ºC)

Effect of the $MgSO_4$ concentration: Fig. 5 shows the effect of the initial $MgSO_4$ concentration on the desulphurization efficiency. The $MgSO_4$ concentration has little effect on the desulphurization efficiency when it is under a low level (≤ 130 g/L), while the desulphurization efficiency decreases rapidly with the $MgSO_4$ concentration increases to 390 g/L. When the $MgSO_4$ concentration is above 520 g/L, the desulphurization efficiency gradually increases with the reaction and reaches a high level of about 88 %, which also facilitates the separation of the byproduct. The results indicate that the concentration of $MgSO_4$ has a great influence on the desulphurization efficiency, which has not been reported in related literatures yet.

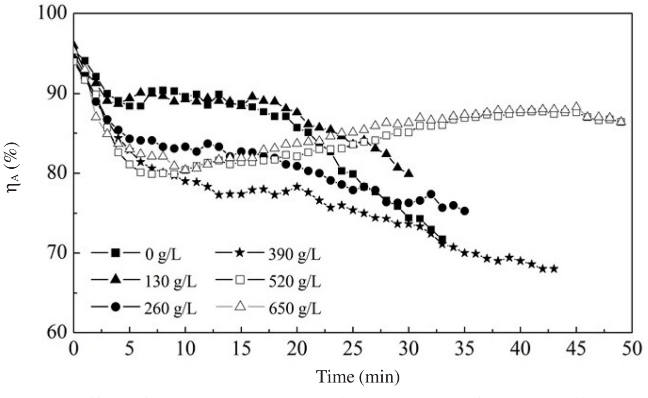

Fig. 5. Effect of the $MgSO_4$ concentration on the desulfurization efficiency (Q_G = 20 Nm^3/h; y_i = 2000 mg/Nm^3; R_{LG} = 8 L/Nm^3; T = 25 ºC)

Effect of the pH value: The pH value of the slurry under different $MgSO_4$ byproduct concentration is shown in Fig. 6. When the $MgSO_4$ concentration was lower than 390 g/L, the pH value of slurry declined gradually with the reaction time and presented two stages, which gently decreased above pH 7.5 and then decreased significantly. When the pH value was higher than 7.5, the desulphurization efficiency maintained at a higher level and then decreased rapidly with the decline of the pH value. When the $MgSO_4$ concentration was higher than 520 g/L, the slurry pH declined gradually in the whole reaction process. The desulphurization efficiency increased to the highest level when the pH value was *ca.* 5.5-6.0 and the efficient desulphurization stage could be maintained for a long time. In the actual industrial running, the slurry pH value should be maintained at *ca.* 5.5-6.0. Thus, it can not only ensure the desulphurization efficiency, but can also improve the utilization of the absorbent and cut down the running cost.

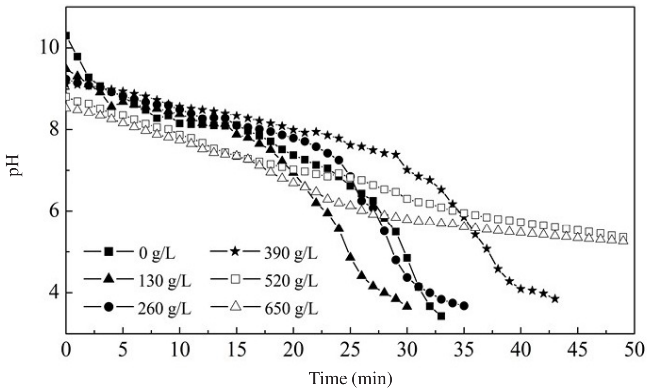

Fig. 6. Effect of the initial $MgSO_4$ concentration on the slurry pH value (Q_G = 20 Nm^3/h; y_i = 2000 mg/Nm^3; R_{LG} = 8 L/Nm^3; T = 25 ºC)

Mass transfer coefficients

SO_2 is easily dissolved in water and can converted into H_2SO_3. A number of models have been proposed to describe this transfer in wet FGD process across a phase boundary. A double resistance theory described the transfer being confined to two thin stagnant films on either side of the gas-liquid interface and the mass transfer of SO_2 can be divided into the gas side mass transfer and the liquid side mass transfer.

Gas side mass transfer: The absorption of SO_2 in the slurry is considered to be a rapid chemical reaction and the absorption rate through the gas film can be written as[13,14]:

$$N_{SO_2} = k_G a (P_{SO_2} - P^*_{SO_2}) \tag{2}$$

where k_G is the gas side mass transfer coefficient, a is the gas-liquid interfacial area per unit volume of liquid, P_{SO_2} and $P^*_{SO_2}$ are the partial pressure of SO_2 in the gas phase and the interface, respectively. The k_G in eqn. 2 can be obtained by the Frossling correlation[13-16]:

$$Sh = \frac{k_G d_s RT}{D_G} = 2 + 0.6\,Re^{1/2}\,Sc^{1/3} \tag{3}$$

where

$$Re = \frac{\rho_G d_s u_G}{\mu_G} \tag{4}$$

$$Sc = \frac{\rho_G}{\mu_G D_G} \tag{5}$$

where Sh is the Sherwood number, Re is the Reynolds number of the gas, Sc is the Schmidt number, d_s is the average size of slurry droplets, D_G is the diffusion coefficient of SO_2 in gas, R is the gas constant, ρ_G is the gas density, u_G is the gas velocity and μ_G is the gas viscosity.

The a in eqn. 2 can be obtained by the relation[13,17]:

$$a = \frac{6}{d_s} \quad (6)$$

where d_s can be obtained by the relation[18]:

$$d_s = 0.013 d_0 We^{-0.50598} Rn^{0.279575} \quad (7)$$

where

$$We = \frac{\rho_G u_s^2 d_0}{\sigma} \quad (8)$$

$$Rn = \frac{\rho_L u_s d_0}{\mu_L} \quad (9)$$

where We is the Weber number, Rn is the Reynolds number of the liquid, d_0 is the inlet diameter of the nozzle, u_s is the slurry dropt velocity, σ is the surface tension of the liquid, ρ_L is the liquid density and μ_L is the liquid viscosity.

Liquid side mass transfer: The absorption rate of SO_2 through the liquid film can be written as[13,14]:

$$N_{SO_2} = E k_L a (C^*_{SO_2} - C_{SO_2}) \quad (10)$$

where E is the mass transfer enhancement factor, k_L is the liquid side mass transfer coefficient, $C^*_{SO_2}$ and C_{SO_2} are the concentration of SO_2 in the interface and the liquid phase, respectively. The E in eqn. 10 can be written as[13,16]:

$$E = 1 + \frac{D_M C_M}{2 D_{SO_2} C^*_{SO_2}} \quad (11)$$

where D_M is the diffusion coefficient of $Mg(OH)_2$ in water, C_M is the concentration of $Mg(OH)_2$ in water and D_{SO_2} is the diffusion coefficient of SO_2 in water.

The k_L in eqn. 10 can be calculated by the relation[13-16]:

$$k_L = 0.88\sqrt{f D_{SO_2}} \quad (12)$$

where

$$f = \sqrt{\frac{8\sigma}{3\pi m_s}} \quad (13)$$

where m_s is the mass of a slurry droplet.

The interfacial concentration of SO_2 can be obtained by[19]:

$$C^*_{SO_2} = H P^*_{SO_2} \quad (14)$$

where H is the thermodynamic equilibrium constant and the value can be obtained by[20,21]:

$$H = \exp[32143.3/T + 198.14 \ln T - 0.3384T - 1135.62] \quad (15)$$

The optimal slurry pH value for the magnesium-based wet FGD process is *ca.* 5.5-7.5, which means that the SO_2 concentration in the slurry is close to zero and eqn. 10 can be simplified as:

$$N_{SO_2} = E k_L a C^*_{SO_2} \quad (16)$$

External mass transfer of SO_2: According to the equations above, the absorption rate of SO_2 also can be written as[13,14]:

$$N_{SO_2} = K_G a P_{SO_2} \quad (17)$$

where K_G is the overall gas side mass transfer coefficient and can be written as:

$$\frac{1}{K_G} = \frac{1}{EHk_L} + \frac{1}{k_G} \quad (18)$$

The absorption rate of SO_2 in the scrubber can be written as[22]:

$$N_{SO_2} = \frac{Q_G}{Q_L RT}\left(\frac{-dP_{SO_2}}{dt}\right) \quad (19)$$

where Q_L is the liquid flow rate and t is the time.

The partial pressure of SO_2 in the gas phase at the inlet and outlet are defined as P_i and P_o, respectively. The residence time of the gas in the scrubber τ is obtained by[13,14]:

$$\tau = \frac{V_R}{Q_G} \quad (20)$$

where V_R is the effective volume of the scrubber.

The integral amount of SO_2 absorbed within the residence time of gas in the effective volume of the scrubber can be written as:

$$\int_{P_i}^{P_o} \frac{1}{P_{SO_2}} dP_{SO_2} = -\int_0^{\tau} \frac{K_G a Q_L RT}{Q_G} dt \quad (21)$$

Eqn. 21 can be written as:

$$\frac{P_o}{P_i} = \exp\left(-\frac{K_G a Q_L RT V_R}{Q_G^2}\right) \quad (22)$$

The desulphurization efficiency can be expressed as[13,14]:

$$\eta_A = \left(1 - \frac{P_o}{P_i}\right) \times 100\ \% \quad (23)$$

Finally, the desulphurization efficiency can be written as:

$$\eta_A = \left[1 - \exp\left(-\frac{K_G a Q_L RT V_R}{Q_G^2}\right)\right] \times 100\ \% \quad (24)$$

Analysis of mass transfer coefficients: The parameter values for the calculations are shown in Table-2 and the results under different experimental conditions are shown in Table-3[13,19,23].

The overall gas side mass transfer coefficients K_{G_1} and K_{G_2} are obtained by theoretical calculation and experimental results, respectively. According to Table-3, when the gas flow rate was too low or the SO_2 inlet concentration was too high (No. 1 and No. 5), the relative error of K_{G_1} and K_{G_2} was more than 20 %, while it was *ca.* 6.3 % under the optimal conditions (No. 4). Therefore, the desulphurization process model can be considered to be suitable for this system under normal operating conditions.

From the coefficients, the gas and liquid side mass transfer resistances, defined as $\frac{1}{k_G}$ and $\frac{1}{EHk_L}$, respectively, can

TABLE-2
PARAMETERS FOR THE CALCULATIONS

Parameter	Value	Unit
D_G	1.4×10^{-5}	m^2/s
D_{SO_2}	1.8×10^{-9}	m^2/s
D_M	1.67×10^{-9}	m^2/s
T	298.15	K
V_R	1.41×10^{-2}	m^3
ρ_G	1.185	kg/m^3
μ_G	17.81	kg/(m s)
σ	7.2×10^{-2}	N/m
ρ_L	1003	kg/m^3
μ_L	1.005×10^{-3}	$N/(m^2\ s)$
d_0	4	mm
d_s	900	μm
E	1.05	–
H	1.24	$kmol/(m^3\ atm)$

be calculated. If $\frac{1}{k_G}$ is close to $\frac{1}{K_G}$, the mass transfer is controlled by the gas phase and the ratio of $\frac{1/k_G}{1/K_G} \leq 0.1$ is the criterion for liquid phase control[13,14]. Table-3 shows that $\frac{1}{k_G}$ is very close to $\frac{1}{K_G}$, indicating that the absorption of SO_2 in magnesium-based wet FGD process is controlled by gas-film diffusion.

Conclusion

This paper presents a magnesium-based wet flue gas desulphurization (FGD) process with a spray scrubber. The results show that the magnesium-based wet FGD process is more suitable for the low SO_2 concentration flue gas and the atomization effect of the nozzle plays an important role in the desulphurization process. As an important parameter in the desulphurization process, the liquid-gas ratio should be controlled at an appropriate level to reduce the energy consumption for slurry cycling and to reach the required desulphurization efficiency. The concentration of $MgSO_4$ has a great influence on the desulphurization efficiency, which has not been reported in related literatures yet. When the $MgSO_4$ concentration is above 520 g/L, the desulphurization efficiency can reach a high level of *ca.* 88.0 % with the pH value is about 5.5-6.0. The high concentration of the byproduct will also facilitate the separation of the byproduct. The optimal conditions for desulphurization under the laboratory scale are fixed as follows: the gas flow rate Q_G is 20 Nm^3/h, the SO_2 inlet concentration y_i is less than 2000 mg/Nm^3, the liquid-gas ratio R_{LG} is 8 L/Nm^3, the $MgSO_4$ concentration C_{MgSO_4} is 650 g/L and the slurry pH value is 5.5-6.0.

The desulphurization process model is established by the double-film theory of mass transfer, which involves gas side mass transfer, liquid side mass transfer and external mass transfer of SO_2. The results show that the calculated mass transfer coefficients are in reasonable agreement with the experimental values and indicate that the absorption process is controlled by gas-film diffusion.

Nomenclature

a = interfacial area per unit volume of liquid, m^2/m^3
C_M = concentration of $Mg(OH)_2$ in the liquid phase, $kmol/m^3$
C_{MgSO_4} = concentration of $MgSO_4$ in the absorption slurry, g/L
C_{SO_2} = concentration of SO_2 in the liquid phase, $kmol/m^3$
$C^*_{SO_2}$ = concentration of SO_2 in the interface, $kmol/m^3$
d_0 = inlet diameter of the nozzle, m
D_G = diffusion coefficient of SO_2 in gas, m^2/s
D_M = diffusion coefficient of $Mg(OH)_2$ in water, m^2/s
d_s = average size of slurry droplets, m
D_{SO_2} = diffusion coefficient of SO_2 in water, m^2/s
E = mass transfer enhancement factor
H = thermodynamic equilibrium constant, $kmol/(m^3\ atm)$
k_G = gas side mass transfer coefficient, $kmol/(m^2\ s\ atm)$
k_L = liquid side mass transfer coefficient, m/s
K_G = overall gas side mass transfer coefficient, $kmol/(m^2\ s\ atm)$
m_s = mass of a slurry droplet, kg
N_{SO_2} = absorption rate, $kmol/(m^3\ s)$
P_i = partial pressure of SO_2 in the gas phase at the inlet, atm
P_o = partial pressure of SO_2 in the gas phase at the outlet, atm
P_{SO_2} = partial pressure of SO_2 in the gas phase, atm
$P^*_{SO_2}$ = partial pressure of SO_2 in the interface, atm
Q_G = gas flow rate, Nm^3/s
Q_L = liquid flow rate, m^3/s
R = gas contant, 8.3145 J/(mol K)
Re = Reynolds number of the gas
R_{LG} = liquid-gas ratio, L/Nm^3

TABLE-3
CALCULATION RESULTS FOR MASS TRANSFER COEFFICIENTS

Parameter	No.							Calculation basis
	1	2	3	4	5	6	7	
Q_G (Nm^3/h)	15	20	20	20	20	20	25	Experimental parameter
R_{LG} (L/Nm^3)	8	6	8	8	8	10	8	Experimental parameter
y_i (mg/Nm^3)	2000	2000	1000	2000	3000	2000	2000	Experimental parameter
η_A (%)	87.4	81.0	91.1	89.7	81.7	94.1	78.9	Experimental result
$k_G \times 10^5$ ($kmol\ m^{-2}\ s^{-1}\ atm^{-1}$)	1.29	1.29	1.29	1.29	1.29	1.29	1.29	Eqn. 3
$k_L \times 10^4$ (m/s)	7.46	7.46	7.46	7.46	7.46	7.46	7.46	Eqn. 12
$K_{G_1} \times 10^5$ ($kmol\ m^{-2}\ s^{-1}\ atm^{-1}$)	1.27	1.27	1.27	1.27	1.27	1.27	1.27	Eqn. 18
$K_{G_2} \times 10^5$ ($kmol\ m^{-2}\ s^{-1}\ atm^{-1}$)	0.94	1.31	1.44	1.35	1.01	1.35	1.16	Eqn. 24
Relative error of K_{G_1} and K_{G_2} (%)	26.0	3.1	13.4	6.3	20.5	6.3	8.7	–

Rn = Reynolds number of the liquid
Sc = Schmidt number
Sh = Sherwood number
t = time, s
T = temperature, K
u_G = gas velocity, m/s
u_s = liquid velocity, m/s
V_R = the effective volume of the scrubber, m^3
We = Weber number
y_i = concentration of SO_2 in the gas phase at the inlet, mg/Nm^3
y_o = concentration of SO_2 in the gas phase at the outlet, mg/Nm^3

Greek symbols

ρ_G = gas density, kg/m^3
ρ_L = liquid density, kg/m^3
τ = residence time of the gas in the scrubber (s)
μ_G = gas viscosity, kg/(m s)
μ_L = liquid viscosity, $N/(m^2\ s)$
σ = surface tension of the liquid (N/m)
η_A = desulphurization efficiency (%)

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REFERENCES

1. S. Wang and J. Hao, *J. Environ. Sci*., **24**, 2 (2012).
2. Z.G. Tang, C.C. Zhou and C. Chen, *Ind. Eng. Chem. Res*., **43**, 6714 (2004).
3. N. Karatepe, *Energy Sources*, **22**, 197 (2000).
4. R. Bitsko and E. Sandell, *Elect. World*, **199**, 44 (1985).
5. M. Hartman, K. Svoboda, O. Trnka and V. Veselý, *Chem. Eng. Sci*., **43**, 2045 (1988).
6. R. Bitsko and R.W. Helt, In Proceedings of the 53rd Annual American Power Conference, USA, pp. 701-705 (1991).
7. J.H. Beeghly, M. Babu and K.J. Smith, In Proceedings of the 16th Annual International Pittsburgh Coal Conference, USA, pp. 462-467 (1999).
8. P.L. Bishop, Q. Wu, T. Keener, L.A. Zhuang, R. Gurusamy and S. Pehkonen, In Proceedings of the 16th Annual International Pittsburgh Coal Conference, USA, pp. 477-486 (1999).
9. G.Q. Dong and J. Wang, *Inorg. Chem. Ind. (China)*, **37**, 11 (2005).
10. S. Han, B.H. Song, S.Y. Lu, Y.J. Han, W.X. Wang and Z.Y. Yang, *Chin. Environ. Prot. Ind. (China)*, **6**, 56 (2008).
11. Y.J. Wang and W.Y. Wang, *Boiler Manuf. (China)*, **3**, 45 (2011).
12. Z. Shen, S. Guo, W. Kang, K. Zeng, M. Yin, J. Tian and J. Lu, *Ind. Eng. Chem. Res*., **51**, 4192 (2012).
13. B. Dou, Y.-C. Byun and J. Hwang, *Energy Fuels*, **22**, 1041 (2008).
14. B. Dou, W. Pan, Q. Jin, W. Wang and Y. Li, *Energy Convers. Manage*., **50**, 2547 (2009).
15. J. Warych and M. Szymanowski, *Ind. Eng. Chem. Res*., **40**, 2597 (2001).
16. T. Neveux and Y.L. Moullec, *Ind. Eng. Chem. Res*., **50**, 7579 (2011).
17. S. Liu and W. Xiao, *Chem. Eng. Technol*., **29**, 1167 (2006).
18. N.L. Liu and X. Zhang, *J. Exp. Fluid Mech. (China)*, **20**, 8 (2006).
19. J.M. Hao, S.X. Wang and Y.Q. Lu, Technical Handbook of SO_2 Emission Control from Coal, Chemical Engineer Press, Beijing (2001).
20. M. Eigen, K. Kustin and G. Maass, *Z. Phys. Chem.*, **30**, 130 (1961).
21. A. Bandyopadhyay and M.N. Biswas, *Chem. Eng. Technol*., **29**, 130 (2006).
22. M.V. Dagaonkar, A.A.C.M. Beenackers and V.G. Pangarkar, *Chem. Eng. Sci*., **56**, 1095 (2001).
23. T.K. Sherwood, R.L. Pigford and C.R. Wilke, Mass Transfer, McGraw-Hill Inc., New York (1975).