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Modelling and experimental study of mass transfer characteristics of SO₂ in sieve tray WFGD absorber

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In this paper, a fundamental model on the mass transfer characteristics of the sieve tray and spray absorber was developed, the model parameters and measured desulphurisation efficiency of the wet flue gas desulphurisation (WFGD) absorber with a sieve tray and no sieve tray were obtained by laboratory scale absorber with diameter of 500 mm using limestone water system, and the desulphurisation efficiency of sieve tray WFGD absorber in a 300 MW power plant was predicted under various operational conditions. It is shown that the predicted values are in good agreement with the measured results from a laboratory scale WFGD apparatus, and the average difference between predicted results and measurement results equals 0.73%, and the models are suitable for use in the design of the sieve tray WFGD absorber. Finally, the desulphurisation efficiency of sieve tray absorber with different sprayer combinations and a sieve tray were predicted, this can provide guidance for the operation of sieve tray absorber. The environmental requirement can be met when two sprayers runs when SO₂ concentration in flue gas is <4000 mg Nm⁻³, and when SO₂ concentration in flue gas is >8000 mg Nm⁻³, four sprayers should be run.

Keywords: Sieve tray, Desulphurisation efficiency, Mass transfer, Absorber

Introduction

Up to the time of 2010, China’s installed power generation capacity has exceeded 962 × 10⁶ kW, and thermal power installed capacity is 706-63 × 10⁶ kW, which takes ~ 73% in total capacity.¹ Coal fired power generation provides the power and heat, but also causes serious pollution of sulphur dioxide (SO₂), which forms a very serious danger for the human and environmental health.²,³ Depending on the SO₂ concentration it causes some diseases that may result even death.⁴ Therefore, its abatement is necessary.

Among the various physicochemical wet and dry methods, wet method is considered to be the simplest and the most economical method for gas scrubbing with very high removal efficiency,⁵ and limestone-gypsum wet flue gas desulphurisation (WFGD) technology is the most commonly used technology for controlling the emission of SO₂ in the world.⁶–⁸

To date, more than 90% thermal power plant have installed WFGD facility in China, and spray absorber is used dominantly, which process consists in spraying slurry drops containing suspended limestone in counter-current to the upward flowing flue gas. The liquid phase is injected into the absorber through nozzles, in form of fine droplets, located at different heights, namely several sprayers. The liquid is then collected in the bottom of the absorber, the so-called reaction tank, and is subsequently pumped again to the sprayers. The high interface area of the liquid particles and the intimate contact between the two phases promote the mass transfer of SO₂ from the gas phase to the liquid phase.⁹

A large number of high sulphur coal are used in thermal power plant in China, the desulphurisation efficiency and economy of the spray absorber decrease significantly in this time. On the contrary, the sieve tray absorber has obvious advantages when high sulphur coal is burned. The sieve tray absorber is that one or several sieve trays are installed in the existing spray absorber. The sieve tray, in which liquid and flue gas flow countercurrently through the same tray holes, can make more uniform flow field and significantly improve the efficiency of desulphurisation, and the sieve tray is a more economical and efficient device suitable for WFGD upgrading retrofit and treating high sulphur coal.

Literature indicates that commercial spray absorber in various forms have been investigated for the absorption of SO₂ in limestone slurry over the decades. Zhao et al.¹⁰ studied the degree of desulphurisation with response surface methodology, and the evolutive response surface model is helpful to describe the degree of desulphurisation of the limestone/gypsum wet FGD spray tower. Dou et al.¹¹ used an electrostatic spraying absorber as the reactor, and studied the FGD process. Ahlbeck

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developed a simple method suitable for industrial utilisation using polydisperse absorbents of technical quality. With such a method, parameters like drop size distribution, PH and reaction time should be considered as manipulative design and process variables for a full scale FGD plant.

Many researchers have adopted different gas–liquid mass transfer theory to build different WFGD models, and the desulphurisation characteristic of the spray absorber is researched by CFD software. Gerbec et al.13 introduced the unsteady state theory to model of pilot scale WFGD system. Based on penetration theory, Brogen et al.14,15 built model of spray absorber of WFGD system and calculated the absorption rate of SO2 into a drop of limestone slurry by acid base reaction, the main equation is as follows:

\[
\text{SO}_2(aq) + \text{OH}^- = \text{HSO}_3^-
\]

The OH\(^-\) ion produced from the limestone dissolution:

\[
\text{CaCO}_3(aq) + \text{H}^+ \rightleftharpoons \text{Ca}^{2+} + \text{OH}^- + \text{CO}_2\uparrow
\]

Based on the kinetic theory of gas–liquid reaction, 19 the mass transfer rate of SO2 in the slurry drop can be expressed by the following equation (first order reaction is assumed)

\[
\eta_{\text{SO}_2} = k c_{\text{SO}_2} c_{\text{OH}} = k x_{\text{SO}_2} x_{\text{OH}} c_{\text{lit}}^2
\]

The mass transfer rate of SO2 in the three zones mentioned above should be equality in steady state, so the total mass transfer rate of SO2 can be expressed as

\[
\eta_{\text{SO}_2} = P_{\text{YSO}_2} \left( \frac{RT}{K_{G} d} + \frac{H}{K_{Lac} c_{\text{tot}}} + \frac{H}{k x_{\text{OH}} c_{\text{lit}}^2} \right)
\]

According to the two film theory and Murphree efficiency, equation (4) can be deduced as

\[
\eta_1 = \frac{1}{\exp \left( \frac{RT}{K_{G} d} + \frac{H}{K_{Lac} c_{\text{tot}}} + \frac{H}{k x_{\text{OH}} c_{\text{lit}}^2} \right)} \frac{P}{n_{\text{SO}_2}}
\]

where \(n_{\text{SO}_2}\) is the mass transfer rate of SO2 (kmol m\(^{-2}\) s\(^{-1}\)); \(K_{G}\) and \(K_{L}\) are the gas and liquid transmission coefficients (m\(^2\)/m\(^2\)s); \(a\) is the interfacial area (m\(^2\)/m\(^3\)); \(P\) is the inlet pressure of absorber (bar); \(c_{\text{YSO}_2}\) and \(x_{\text{YSO}_2}\) are mole fractions of SO2 in the gas and liquid; \(\gamma_3\) and \(\gamma_1\) are mole fractions of SO2 at the interface; \(c_{\text{tot}}\) is the total concentration of gas and liquid (kmol m\(^{-3}\)); \(k\) is reaction rate constant; \(x_{\text{OH}}\) is mole fraction of OH\(^-\) ion in the liquid; \(H\) is Henry constant (bar); and \(\eta_1\) is desulphurisation efficiency of SO2 in the spray absorber with no sieve tray.

**Mathematical models**

**Gas–liquid mass transfer in spray absorber**

According to Lewis two film theory, there are three resistances in the process of SO2 absorption by limestone slurry in the spray absorber: gas film resistance, liquid film resistance and the reaction in the slurry drop. The mass transfer equation of SO2 in the three reaction zone can be expressed as follows:

(i) gas film zone

\[
n_{\text{SO}_2} = K_G d (p_{\text{SO}_2} - p) = K_G d \frac{P}{RT} (y_{\text{SO}_2} - y_1)
\]

(ii) liquid film zone

\[
n_{\text{SO}_2} = K_L d (c_1 - c_{\text{SO}_2}) = K_L d c_{\text{tot}} (x_1 - x_{\text{SO}_2})
\]

(iii) reaction in the slurry drop.

In the spray absorber, SO2 is absorbed into the limestone slurry by acid base reaction, the main equation is as follows:

\[
\text{SO}_2(aq) + \text{OH}^- = \text{HSO}_3^-
\]

The OH\(^-\) ion produced from the limestone dissolution:

\[
\text{CaCO}_3(aq) + \text{H}^+ \rightleftharpoons \text{Ca}^{2+} + \text{OH}^- + \text{CO}_2\uparrow
\]

Substituting equation (7) in equation (6), then

\[
\frac{dY}{Y - Y^*} = \frac{K_G d}{G} dh
\]

Integral is performed on both sides of equation (8), and equation (8) becomes
Mass transfer characteristics of SO₂ in sieve tray WFGD absorber

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According to the definition of a mass transfer unit and Murphree tray efficiency, following expressions can be obtained:

\[ N_{OG} = \int_{Y_{1}}^{Y_{2}} \frac{dY}{Y - Y^*} = \frac{K_{OG}a_{HI}}{G} \]  

(9)

where

\[ N_{OG} \] is the overall vapour phase mass transfer unit; \( G \) is the gas flowrate (kmol m⁻² s⁻¹); \( a_{HI} \) is the mole fraction of SO₂ in vapour phase at the import and export of the sieve tray; \( Y \) is the desulphurisation efficiency of sieve tray; \( Y_i \) is the desorption coefficient, which is the equilibrium line and operating line slope of the ratio of the slope (\( S = HLI/PG \)); \( t_i \) is the residence time of the liquid on the sieve tray; \( e \) is the porosity of vapour–liquid layer on the sieve tray; \( C_1, C_2, C_3 \) are the constants, which can be obtained by correlating the experimental data.

Desulphurisation efficiency of sieve tray WFGD absorber

According to equations (5) and (13), the desulphurisation efficiency of the sieve tray WFGD absorber can be deduced as:

\[ \eta = \eta_1 + \eta_2 - \eta_1 \eta_2 \]  

(17)

where \( \eta \) is the total desulphurisation efficiency in the sieve tray WFGD absorber.

Experimental

Experimental apparatus

The experiments were conducted on the laboratory scale WFGD apparatus, whose configuration is shown in Fig. 2. The apparatus consisted of the absorber, the flue gas system and the slurry system. The absorber was manufactured according on a 1 : 10 scale model of a full scale FGD for a 300 MW unit. The absorber diameter was 500 mm, the height 4000 mm, two sprayers and a mist eliminator were installed in the absorber, a 3 mm thick steel sieve tray was installed between the absorber entrance and the first sprayer (the sieve tray porosity range is 30–45% and the pore diameter range is 25–45 mm), its pores distribution in the form of triangle. The flue gas system consisted of the fan, sulphur dioxide and gas mixing box. The slurry system consisted of the slurry circulating pump and a slurry tank, which size is φ 1500 × 1400 mm. The experimental apparatus design parameters: flue gas flux, 2000–3000 m³ h⁻¹; mass concentration of SO₂ in flue gas, 3000–5000 mg Nm⁻³; liquid gas ratio, 12–18 L m⁻³; the slurry PH value, 5.2–5.8. The SO₂ concentration inlet and outlet the absorber is measured by the flue gas analyser, the average gas velocity inlet the absorber was measured with a pilot tube and the flue gas flux can be calculated. The sampling sites of the SO₂ concentration and the gas velocity are shown in Fig. 2.

Experimental description

In the experiment process, the gas mixture of the air and sulphur dioxide entered the bottom of the absorber, passed through the sieve tray, the sprayer and the mist eliminator, and then entered the atmosphere from the top of the absorber. The prepared limestone slurry in the slurry tank was injected into absorber through the nozzle under the circulating pump pressure, the sulphur dioxide were absorbed by the slurry in the absorber, and then the slurry returned the tank from the bottom of the absorber.

In the course of experiments, the desulphurisation efficiency in the absorber with a sieve tray and no sieve tray was obtained by measuring the SO₂ concentration inlet and outlet the absorber by the flue gas analyser at the difference inlet SO₂ concentration (3000–5000 mg Nm⁻³), flue gas flux (2000–3000 m³ h⁻¹), PH value (5.2–5.8), the sieve tray porosity (30–45%) and pore diameter (25–45 mm). Then the desulphurisation efficiency of the sieve tray was obtained by equation (17).
Results and discussion

Sieve tray desulphurisation efficiency

The porosity and pore diameter are the most important parameter in the sieve tray design, which are directly related to the sieve tray pressure drop and efficiency. Previous studies show that the larger the porosity and pore diameter, the lower the desulphurisation efficiency, but the lower the sieve tray pressure drop. The effect of the sieve tray porosity and pore diameter, flue gas velocity and liquid gas ratio on the desulphurisation efficiency is shown in Fig. 3. It can be seen that the sieve tray desulphurisation efficiency decreases smoothly with the increasing porosity, and the desulphurisation efficiency is the most sensitive to the pore diameter, the sieve tray desulphurisation efficiency decreases steeply with the increasing pore diameter. The sieve tray desulphurisation efficiency decreases by 59.8% when the porosity increasing from 30 to 45%. The sieve tray desulphurisation efficiency decreases exponentially from 47-04 to 23-96%, when the pore diameter increases from 30 to 35 mm. The probable cause is that with increasing the porosity, the flue gas velocity through the hole decreases, the gas–liquid disturbance and fluctuation on the sieve tray decreases, and in turn, the slurry froth height decreases, therefore, the sieve tray desulphurisation efficiency decreases. Meanwhile, the pore diameter should be $<$35 mm when high desulphurisation efficiency was required, but at this time the sieve tray pressure drop and energy consumption should be taken into account.

Figure 3c and d shows the effect of flue gas velocity and liquid gas ratio on the sieve tray desulphurisation efficiency (the sieve tray porosity is 40% and pore diameter is 35 mm). It can be seen that the desulphurisation efficiency increases linearly with the flue gas velocity and liquid gas ratio, and the desulphurisation efficiency increases by 3-05% when the flue gas velocity increases from 3-5 to 3-9 m s$^{-1}$, the desulphurisation efficiency increases by 20-8% when the liquid gas ratio increases from 12 to 18 L m$^{-3}$.

Desulphurisation efficiency of sieve tray WFGD absorber

Figure 4 shows the comparison of measured desulphurisation efficiency of the laboratory scale sieve tray WFGD absorber with predicted by the equation (19), the root mean square error between predicted results and measurement results equals 0.36%. The predicted results agree with the measured results. The comparison reveals that the model can describe the mass transfer characteristics of the sieve tray WFGD absorber, and can be used to calculate the desulphurisation efficiency of the sieve tray WFGD absorber.

In order to provide a reference for the design and operation of sieve tray WFGD absorber, the desulphurisation efficiency of sieve tray WFGD absorber in a 300 MW power plant was simulated by the models.
Simulation was made in the following conditions: gas flux, \(9.5 \times 10^7\)–\(1.1 \times 10^6\) Nm\(^3\) h\(^{-1}\); mass concentration of SO\(_2\) in flue gas, 5000–10000 mg Nm\(^{-3}\); sieve tray amount, one layer; sieve tray diameter, 35 mm; sieve tray porosity, 40%. Figure 5 shows the desulphurisation efficiency of the sieve tray WFGD absorber at different liquid gas ratio. It can be seen that the desulphurisation efficiency of the sieve tray WFGD absorber increases with increasing the liquid gas ratio, the desulphurisation efficiency decreases with increasing the concentration of SO\(_2\) in flue gas. The desulphurisation efficiency increases by 12.6 and 14.8% when the liquid gas ratio increases from 12 to 24 L m\(^{-3}\) at the different SO\(_2\) concentration (8000 and 10000 mg Nm\(^{-3}\) respectively). Figure 5 also shows that the desulphurisation efficiency increases by 1.2–5.2% when a sieve tray was installed in the absorber at different SO\(_2\) concentration and liquid gas ratio.

Figure 6 shows the liquid gas ratio in absorber with a sieve tray and no sieve tray at different desulphurisation efficiency. It can be seen that the liquid gas ratio increases with increasing the desulphurisation efficiency in the absorber with a sieve tray and no sieve tray at the different SO\(_2\) concentration, and the higher SO\(_2\) concentration, the higher liquid gas ratio at the same desulphurisation efficiency. The liquid gas ratio decreases by 25.4–33.8%, when a sieve tray was installed in the absorber at different SO\(_2\) concentration and desulphurisation efficiency.

The operation mode of boilers of power plant in China is different to that of other countries since variable coal property and unstable loads of boilers. The differences of operation mode lead to the variation of process parameters of WFGD system. For optimisation of performance and cost, the combination of different sprayers and a sieve tray are calculated in above mentioned operation conditions. The combinations of different sprayers are shown in Table 1. The calculation results of the desulphurisation efficiency of sieve tray absorber with different sprayer combination were shown in Fig. 7. It can be seen that the SO\(_2\) concentration at outlet of absorber meet the environmental requirement (\(<400\) mg m\(^{-3}\)) when SO\(_2\) concentration in flue gas is 4000 mg Nm\(^{-3}\) at all cases, i.e. the environmental requirement can be met when two sprayers runs at this time. Similarly, when SO\(_2\) concentration in flue gas is 6000 mg Nm\(^{-3}\), three sprayers can be run, and when SO\(_2\) concentration in flue gas is more than 8000 mg Nm\(^{-3}\), four sprayers should be run. This can provide guidance for the operation cost optimisation of sieve tray WFGD absorber.

**Conclusions**

A fundamental model to predict the desulphurisation efficiency of the sieve tray absorber was developed for studying the mass transfer characteristics on the sieve tray WFGD absorber. The predicted values are in good agreement with the measured results from a laboratory scale WFGD apparatus, and the average difference between predicted results and measurement results equals 0.73%.

<table>
<thead>
<tr>
<th>Case</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
</tr>
</thead>
<tbody>
<tr>
<td>1st sprayer (lowest)</td>
<td>+</td>
<td>−</td>
<td>+</td>
<td>−</td>
<td>−</td>
<td>+</td>
</tr>
<tr>
<td>2nd sprayer</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>−</td>
<td>+</td>
<td>+</td>
</tr>
<tr>
<td>3rd sprayer</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
</tr>
<tr>
<td>4th sprayer (highest)</td>
<td>+</td>
<td>+</td>
<td>−</td>
<td>−</td>
<td>−</td>
<td>−</td>
</tr>
</tbody>
</table>

* + represents the sprayers are in operation; − represents the sprayers are out of operation.
The model has been successful to predict the desulphurisation efficiency of the sieve tray WFGD absorber in a 300 MW power plant under various operational conditions. The satisfactory results obtained could provide a reference for the operation cost optimisation of sieve tray WFGD absorber.

Reference
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