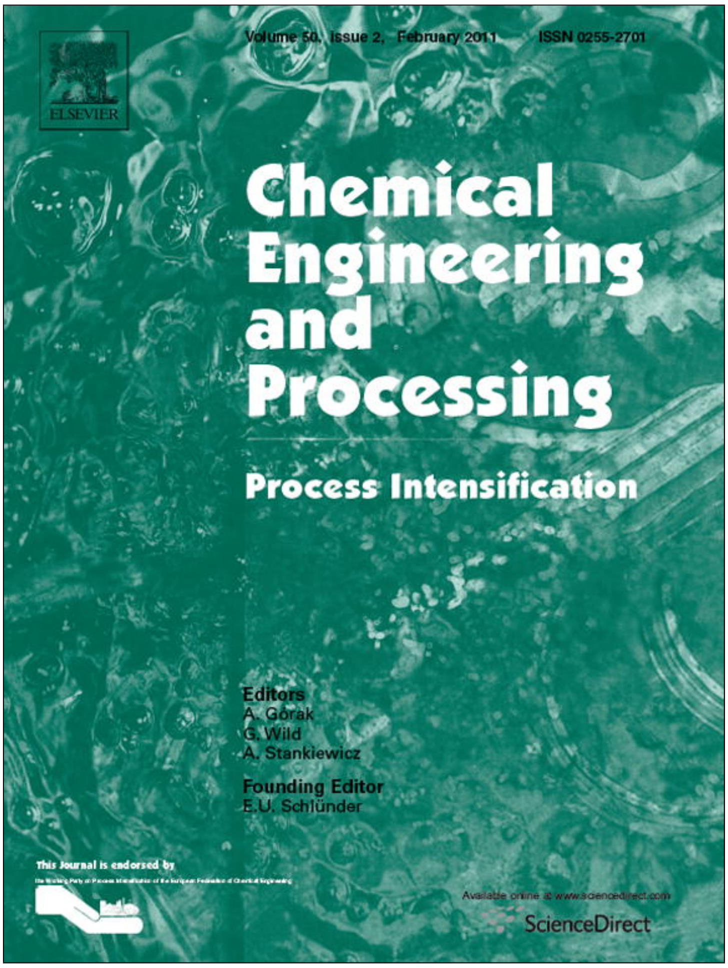

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Experimental study of wet flue gas desulphurization with a novel type PCF device

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ABSTRACT

Scrubbers are being widely used to remove the dust, sulphur dioxide and other harmful gases from coal-fired boilers. In this paper, a novel 'wet-type' desulphurization absorber, the PCF device (Chinese LOGO), was developed and studied through an experimental method. The mixture of air and SO_2 was used as simulated flue gas and $CaCO_3$-in-water suspension was used as absorbent. The results show that the PCF device has a good overall performance for FGD. Under moderate conditions employed, the content of SO_2 in outlet flue gas can achieve a level much lower than that permitted, while the pressure drop is very small due to co-flows in preliminarily treating chamber and no venturi structure in inlet tube. Guide plates and self-excitation chamber can improve the SO_2 removal efficiency by intensifying the mass-transfer and second purification. Some feasible process parameters are as follows: slurry pH value = 5.6–6.0, liquid–gas ratio = 8.7–10.4 L/m^3, superficial gas velocity in inner cylinder = 3.5–4.5 m/s, and addition of Cl^- (in the form of $CaCl_2$) to the slurry (25 g/L) decreased the degree of SO_2 removal about 13.12%.

1. Introduction

Air pollution caused by SO_2 from coal- and fuel oil-burning is a global issue and thus receives more and more attention [1]. Several schemes, such as fuel pretreatment, concurrent burning and adsorption, and flue gas post treatment, i.e., flue gas desulphurization (FGD), have been proposed [2,3]. Among those schemes, FGD is the most reasonable one from both technological and economic point of views. And different processes, such as dry-, semidry- and wet-processes, have been developed for FGD, while wet-processes have been earned widespread use due to lower operating cost and more stable operation [4].

Since the goal of wet FGD processes is only for environmental protection and there are no value-added products during the course, the cost and performance should be the most important factors to be considered. Coincidentally, both the cost and performance are related closely to the type of absorber [5]. The PCF device (Chinese LOGO) is newly developed for industrial application of wet FGD, deriving from the conventional granite water film dust collector (GWFDC). Compared with the original GWFDC, the novel wet-type PCF device possesses the following virtues: (a) Dewatering performance improves significantly and extra demister is out of consideration. No demister means lower energy-consumption, cost and maintenance. (b) Co-flows of gas–liquid in the preliminary treating chamber and no venturi structure in the inlet tube, therefore the pressure drop of the device is much lower than that of the original GWFDC. (c) The self-excitation chamber has second purification for the flue gas, which further improves the collection efficiency of the device. (d) Draft fans of the original GWFDC can be reused in the novel PCF device, reducing the investment cost of the PCF desulphurization technology.

Fundamental studies have focused on the spray scrubber [6,7], packed tower [8,9], jet bubbling reactor (JBR) [10], and so on [11,12]. However, as far as the PCF device is concerned, very little work has been reported on its performance. Therefore, the objective of this work was to perform an experimental study on the flue gas desulphurization process in the PCF device. During the course, the mixture of air and SO_2 was used as simulated flue gas and $CaCO_3$-in-water suspension was used as absorbent.

2. Design mechanism of the PCF device

The schematic diagram of a full-scale PCF device is shown in Fig. 1. It is a sleeve structure, by building an outer cylinder around the original granite water film dust collector (GWFDC). The outer cylinder is lower than inner cylinder (original GWFDC), and between them is preliminary treating chamber where gas–liquid contact and are in co-flows. The nozzles are distributed at the top

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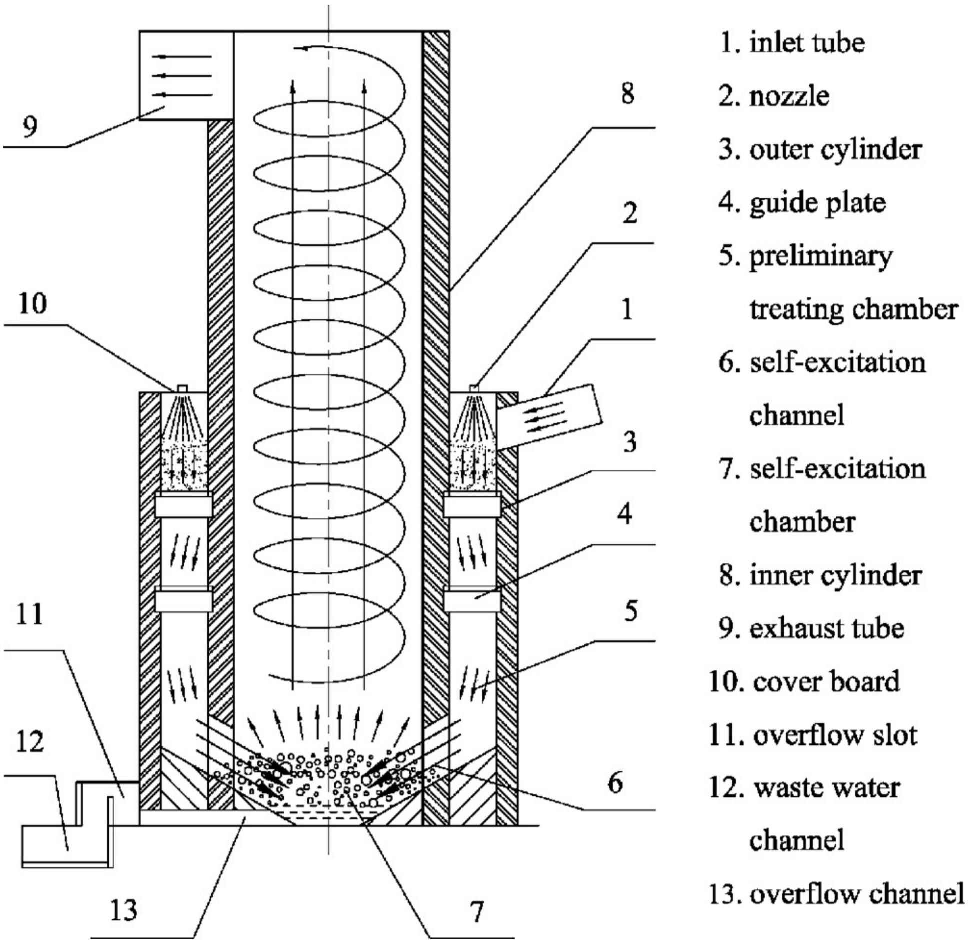

Fig. 1. Schematic illustration of a full-scale PCF device for FGD.

of preliminary treating-chamber and generate atomization. Two layers of guide plates are equipped in the preliminary treating-chamber, promoting mass-transfer. Self-excitation channels lying in the wall of inner cylinder are employed to connect the preliminary treating chamber and inner cylinder, and simultaneously make the gas rotate into the inner cylinder. At the bottom of inner cylinder is a self-excitation chamber which has second purification for the flue gas. The whole inner cylinder is used to remove water from air. Detailed information about some key components of the device is described in the following.

2.1. Guide plates

The schematic diagram of guide plate distribution is shown in Fig. 2. The guide plates have some angle with horizontal direction. This angle can make the gas flow forms spin-flow. Water film or curtain can be generated at or between guide plates. As gas–liquid–solid passes through them, retention time becomes longer and mass-transfer effect gets improved. And plus, because the two layers of guide plates are titled in opposite directions, swirl flows of gas–liquid–solid mixture from upper plates can collide with lower plates directly. As a result, the renewal period of water film on the plates becomes shorter and mass-transfer effect gets further improved.

2.2. Self-excitation channels and self-excitation chamber

The self-excitation channel is a venturi structure. Two or more ones are uniformly distributed along the inner cylinder circular (as shown in Fig. 3) so that gas flow rate of each channel is basically equal. The section area of self-excitation channel depends on the gas flow rate, which makes sure that the gas has some velocity and impact momentum. And meanwhile, each of channels has a horizontal angle γ and a radial angle β. γ makes gas flow produce a slant impact on the liquid in self-excitation chamber, and β makes the mixture of gas–liquid–solid rotate and consequently form centrifugal separation for dust removal and dewatering in inner cylinder. Mention that theradial angle β is an angle formed by self-excitation channels and inner cylinder, in corresponding with the rotating direction of gas from lower guide plates. The self-excitation chamber is an obconical structure (see Fig. 1).

As the primarily purified gas goes through the self-excitation channels, it is accelerated and then decelerated. This effect strengthens the gas–liquid–solid contact. The particles with larger surface and unreacted sulphur-oxides are collected. Meanwhile, the absorption liquid at the surface of self-excitation chamber was excited into bubbles by the mighty gas–flow. The bubbles have many advantages, such as conglutination, interception, collision, and absorption for sulphur dioxide. These functions further improve the collection performance of the PCF device.

3. Experiment

3.1. Experimental setup and analytical methods

A schematic diagram of the experimental setup is shown in Fig. 4, of which the essential element is the lab-scale PCF device where SO_2 removal occurs. The inner cylinder is a cylinder with

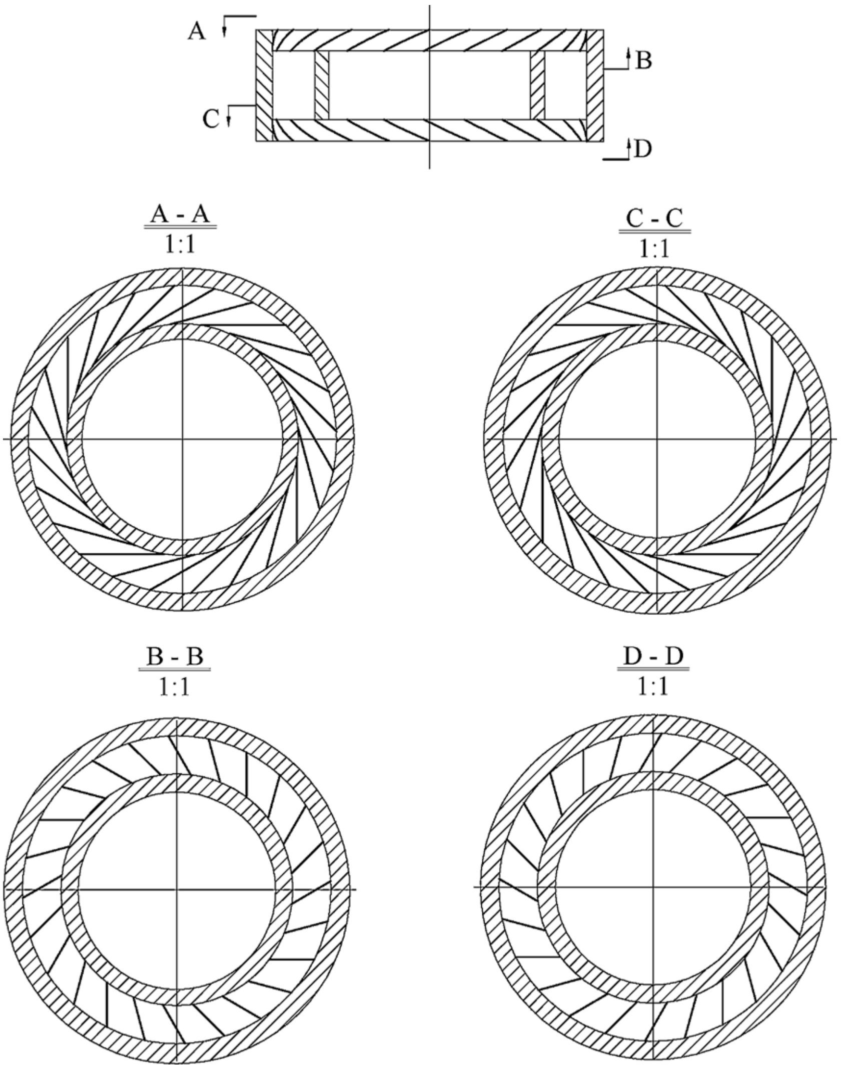

Fig. 2. Schematic diagram of guide plate distribution.

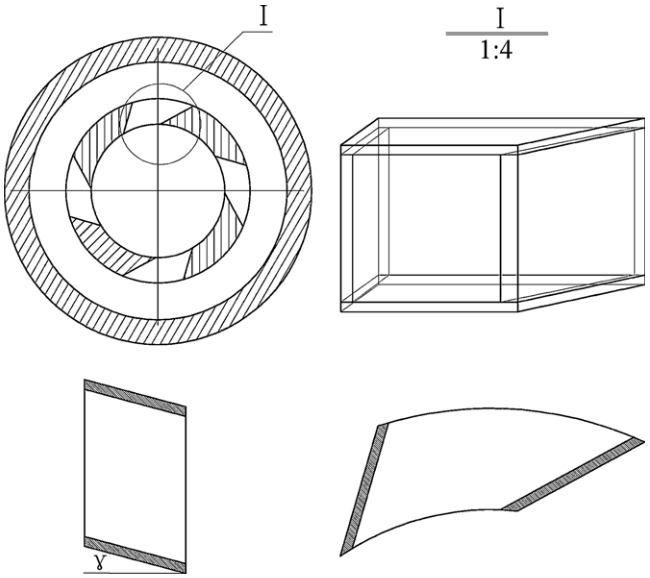

Fig. 3. Schematic diagram of self-excitation channels.

an inner diameter of 0.3 m and a height of 1.4 m. The preliminary treating-chamber is an annular configuration with a width of 0.08 m and a height of 0.68 m. The recycle slurry tanker lies at the bottom of the device. All pipes and vessels are made of PVC or organic glasses to avoid corrosion.

The desired amount of SO_2 in air was prepared by mixing bottled SO_2 with air drafted by exhaust fan. Volumetric flow rates of SO_2 and air were adjusted by the pressure reduction valve and volume damper, respectively. SO_2 was absorbed by limestone slurry. Limestone slurry prepared by mixing $CaCO_3$ of 25 μm with tap water was stored in slurry tank. The circulating pump was provided for the recirculation of the limestone slurry, and the quantity of slurry pumped into the absorber was adjusted by means of a valve. During the experiment, a digital pH meter (Model: HI 8424) was employed to measure the pH value of slurry by inserting a pH probe into the liquid phase. Pitot tube (Model: Y25-150) was used to measure the gas flow rate, and two micro-computer smoke test instruments (Model: Leibo3020) were employed for on-line measurement of SO_2 concentration in gas phase by putting sensors at the test cross sections of inlet tube and outlet tube simultaneously.

3.2. Experimental mechanism

Firstly, the air–SO_2 mixture enters the preliminary treating chamber through the inlet tube at the side-top of the absorber.

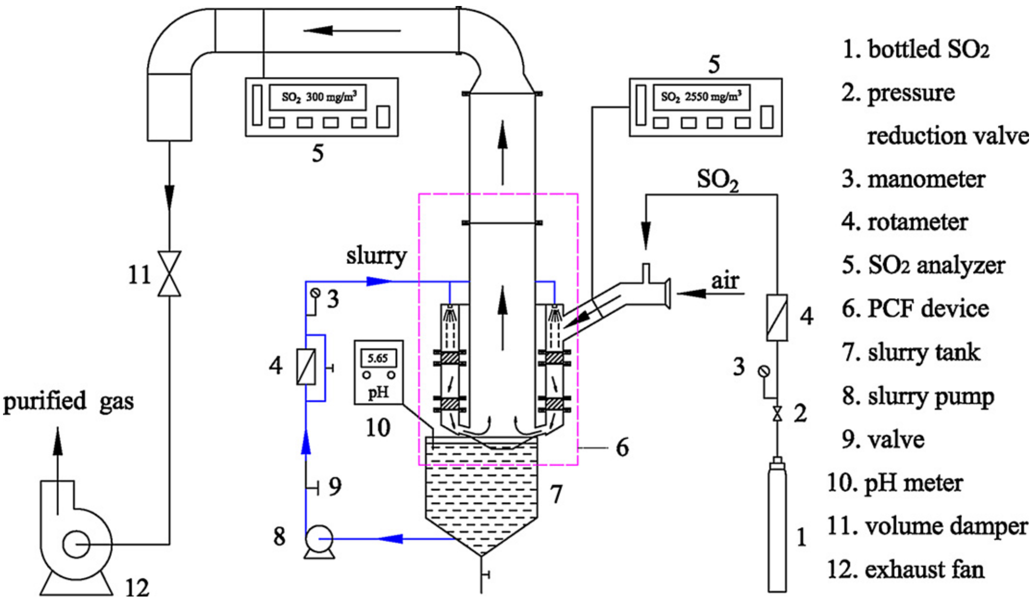

Fig. 4. Flow diagram of experimental setup for absorption of SO_2.

After colliding with arc walls, it changes direction and flows up or down. The ups form into reverse flows because of the board, and the downs rapidly spread the whole preliminary treating-chamber cross. Thanks to the existence of guiding plates, the absorbing liquid can form liquid film or curtain at or between the guide plates, and gas–liquid turbulent intensity increases. When the gas with SO_2 passes through the liquid film or curtain, SO_2 gets preliminary purification by reacting with $CaCO_3$ in the absorbing liquid. And then, the gas goes through the self-excitation channels and enters inner cylinder where it can impinge the slurry in self-excitation chamber and is purified once more. As the gas swirls up, water is removed and the purified gas is released to the atmosphere through the exhaust tube. The scrubbed liquid flows into inner cylinder along with self-excitation channels and returns to the recycle slurry tank.

3.3. Experimental procedure and basic conditions

The experiments were carried out in a batch mode at ambient temperature. Before each run, the tank was refilled with fresh limestone slurry. The data were collected during the first 3–4 min. Because the slurry volume was about 450 L and the slurry pump capacity was equal to 100 L/min, the slurry was recycled only almost one time during 3–4 min runs. Thus SO_2 concentration in slurry did not increase significantly. The basic experimental conditions for the desulphurization system are listed in Table 1. The relative errors of experimental results were controlled below 1%. With neglecting the variation of gas volume due to absorption, the SO_2 removal efficiency (η) is defined as

$$\eta = \frac{C_{in} - C_{out}}{C_{in}} \times 100\% \qquad (1)$$

where C_{in} and C_{out} are the SO_2 inlet concentration and outlet concentration, respectively.

Table 1
Basic experimental conditions for the wet-type PCF desulphurization system.

Parameter	Value
Temperature of gas	Atmosphere (25 ± 1 °C)
Operating pressure	Atmosphere
Gas flow rate (m^3/s)	0.24
Liquid–gas ratio (L/m^3)	10
Droplet size (mm)	2.5
Limestone content in feed stream (wt%)	10
Limestone slurry pH	5.7 ± 0.1
SO_2 inlet concentration (mg/m^3)	2500

4. Results and discussion

4.1. Effects of guide plates and self-excitation chamber

For the novel type PCF device, some parts such as guide plates, self-excitation channel and self-excitation chamber can intensify the mass-transfer of gas–liquid–solid. To illustrate the effects, some related experiments were discussed under different gas flow rates in this section.

Fig. 5 shows the effect of guide plates on the relationship of SO_2 removal efficiency (η), pressure drop (ΔP) and gas flow rate. From the figure, it can be seen that the novel type PCF device has a satisfactory overall performance. Under moderate operating conditions, the content of SO_2 in outlet flue gas can achieve a level much

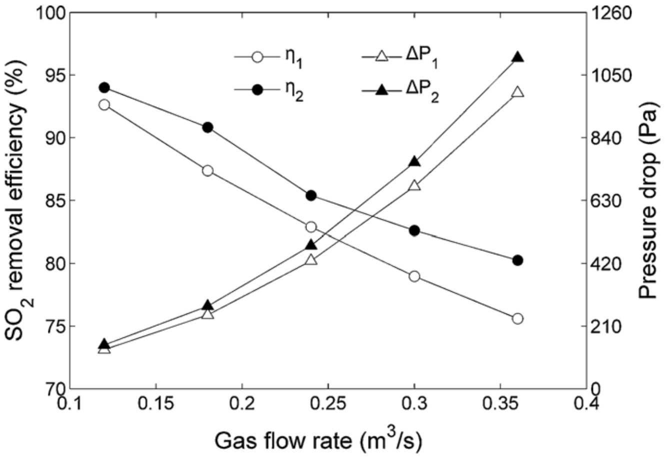

Fig. 5. Effect of guide plates on the relationship of SO_2 removal efficiency, pressure drop and gas flow rate. Solid points stand for the PCF device with two layers of guide plates and empty points stand for the PCF device with no guide plates.

Table 2
SO_2 removal efficiency in preliminary treating chamber and self-excitation chamber.

Gas flow rate (m^3/s)	0.12	0.18	0.24	0.3	0.36
η_{pre} (%)	93.56	88.36	83.94	80.16	76.93
η (%)	94.00	90.83	85.39	82.60	79.26

lower than that permitted (960 mg/m^3 in China), while the pressure drop is very small by comparison with that of the other scrubbers reported previously [13–15]. The reason for those is due to co-flows in preliminarily treating chamber and no venturi structure in inlet tube. The lower pressure drop means lower energy consumption. And hence, the PCF device is an energy-saving device. On the other hand, keep the liquid–gas ratio fixed, η decreases with gas flow rate increasing. When the gas flow rate is changed from 0.12 to 0.36 m^3/s, η decreases about 17%. Although it is so, larger gas flow rate means more flue gas volume to be treated one-time. In this experiment, when the gas flow rate was controlled in the range of 0.2–0.3 m^3/s (note the superficial gas velocity in inner cylinder is 2.831–4.246 m/s), SO_2 removal efficiency can reach above 80% and the pressure drop of the PCF device is only 336–756 Pa.

Furthermore, the curves shown in Fig. 5 also illustrate that with the same operating conditions, the desulphurization efficiency of the PCF device with two layers guide plates is higher than that of the one with no guide plates. This result testifies the fact that guide plates can improve the mass-transfer and thereby in the desulphurization efficiency. While for the pressure drop, guide plates lead it to increase slightly.

Table 2 presents the SO_2 removal efficiency in preliminary treating chamber and self-excitation chamber under different gas flow rates. Generally speaking, most SO_2 is removed in the preliminary treating chamber, and the self-excitation chamber strengthens the removal effect to some extent. Gas flow rate has great effect on the SO_2 removal efficiency of preliminary treating chamber. However, according to Table 2, it can be calculated that with gas flow rate increasing, the desulphurization effect of self-excitation chamber almost remains unchanged. The reason may be that the residence time of gas becomes shorter at higher gas flow rate although the volume of bubble zone expands.

4.2. Effects of slurry pH

With C_{in} = 2500 mg/m^3, the desulphurization efficiency of PCF device at different pH values is shown in Fig. 6. As shown in the figure, η appears to increase almost linearly from 78.8% to 85.6% when the slurry pH in a range of 5.0–6.25. The results agree well with Zhong [16] and Zheng et al. [10]. The reason may be that at higher pH, the dissociation reaction of SO_2:

$$SO_2 + H_2O \rightleftharpoons H^+ + HSO_3^- \qquad (2)$$

is shifted to the right, leading to an increase in the enhancement factor and thereby in the degree of desulphurization [10]. Owing to different setups and experimental conditions, only the trend in the dependence of removal efficiency on pH is provided in Fig. 6.

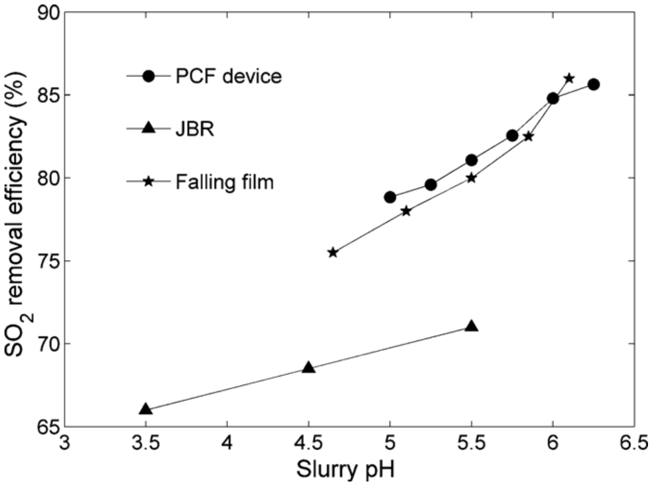

Fig. 6. Comparison of pH effect on η among PCF device, JBR and a falling film pilot plant. The PCF device of this work was run at ambient temperature with C_{in} = 2500 mg/m^3; the Zheng's JBR was run at 23 °C with C_{in} = 2831 mg/m^3; the falling film pilot plant was run at 50 °C with C_{in} = 2857 mg/m^3.

Although higher pH value can lead to a higher SO_2 removal efficiency, the reasonable pH value should be adopted by synthesizing three aspects in practice.

(1) The influence of pH value to the mass-transfer performance.

It is advantageous to run a wet FGD plant at higher pH value from SO_2 solubility and total mass-transfer coefficient point of views. However, the limestone dissolution rate decreases at higher pH value. And hence, the most optimal pH value should balance these two aspects, getting the biggest mass-transfer rate [17].

(2) The influence of pH value to the oxidization effect in slurry tank.

The related research indicated that the $HSO_3{}^-$ oxidization rate is highest at pH of 4.5 [18,19]. If the pH value is higher, $HSO_3{}^-$ will be difficult to be oxidized, extra air must be introduced into the slurry tank to force $HSO_3{}^-$ oxidized into $SO_4{}^{2-}$. Only in this way, the $SO_4{}^{2-}$ ions can combine with Ca^{2+} ions originating from the dissolved limestone and crystallize as gypsum consequently.

(3) The influence of pH value to the scale in scrubbers.

According to Paul and Richard [20], if slurry pH value is lower, there will be lots of hard scales of $CaSO_4$; and, if slurry pH value is higher, there will be lots of soft dirt of $CaSO_3$. As we all know, both $CaSO_3$ and $CaSO_4$ are prone to block work system up. Therefore, some attention should be paid on this point in practice. Experiments around the world confirm that the soft dirt can be effectively avoided when the pH value is controlled below 6.2. But the pH value should not be too low; otherwise, there will be hard scales [21].

Combining the above analysis and experimental results, the slurry pH value of the PCF device was chosen as 5.5–6.0. Thus, η can reach 82.1–84.8%. And meanwhile, it can effectively keep scales out producing, only that the forced oxidization mode and sufficient limestone slurry must be employed in time.

4.3. Effects of liquid–gas ratio

According to Brogren and Karlsson [6], the absorption of SO_2 into a limestone spray scrubber to a large extent is liquid-side controlled, thus a large interface area is favorable; while the interface area mainly depends on the liquid–gas ratio, V_L/V_G. And however, increase in V_L/V_G will lead to more energy consumption and thus operation cost. Therefore, V_L/V_G needs to be optimized. Fig. 7 gives the results of SO_2 removal efficiency and pressure drop for different liquid–gas ratios. The experiments were carried out at a fixed flue gas rate (V_G), and liquid flow rate (V_L) is controlled according to the requested V_L/V_G.

As shown in the figure, η increases continuously with V_L/V_G increasing in the range of V_L/V_G < 11 L/m^3. However, when V_L/V_G is more than 11 L/m^3, η increases relaxed. It can be explained as follows. V_L/V_G influences the mass transfer performance by changing the gas–liquid interface area of the mass transfer equation [12]. With an increase in the amount of V_L/V_G delivered to the absorber, the gas–liquid mass–transfer area and total alkalinity for the absorption of SO_2 increase when the gas flow rate of flue gas is

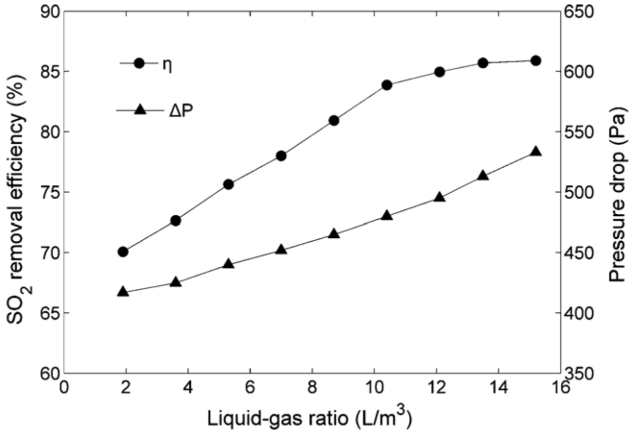

Fig. 7. Relationship of SO_2 removal efficiency, pressure drop and liquid–gas ratio.

fixed. Consequently, the SO_2 absorption rate increases and thereby in removal efficiency of SO_2. However, when V_L/V_G is too large, the cohesion of droplets will strengthen, and the effective gas–liquid interface area no longer increases but even decreases, resulting in smaller mass-transfer rate [5]. This moment, further increase in V_L/V_G becomes meaningless, and η increases relaxed. On the other hand, it can be seen that the range of variation of pressure drop is not significant when increasing the liquid–gas ratio. According the data of Fig. 7, the range of $V_L/V_G = (8.7–10.4)$ L/m^3 is considered the optimal one.

4.4. Effect of SO_2 inlet concentration

Fig. 8 gives the results of SO_2 removal efficiency at various gas flow rates for different SO_2 inlet concentrations. The curves in diagram show that for constant gas flow rate an increase of C_{in} leads to a decrease in η. As the inlet concentration of SO_2 varies from 1.254 to 5.634 g/m^3, each of η decreases about 13%. But this does not imply decrease in SO_2 absorption rate, and on the contrary, the absorption rate is a simple increasing function of C_{in}. The decreasing tendency of η is only due to faster increase in the amount of SO_2 than that needs to be absorbed [5]. And hence in the case of very high C_{in}, certain improvement of operating conditions, i.e., increasing liquid–gas ratio, is needed to achieve higher desulphurization efficiency. On the other hand, for constant SO_2 inlet concentration, η decreases as the gas flow rate increases. The possible reason is that the gas-side mass-transfer coefficient, residence time of gas in the absorber and liquid–gas ratio become shorter as the gas flow rate is enhanced [22], and meanwhile, larger gas flow rate means smaller molar ratio of Ca/S.

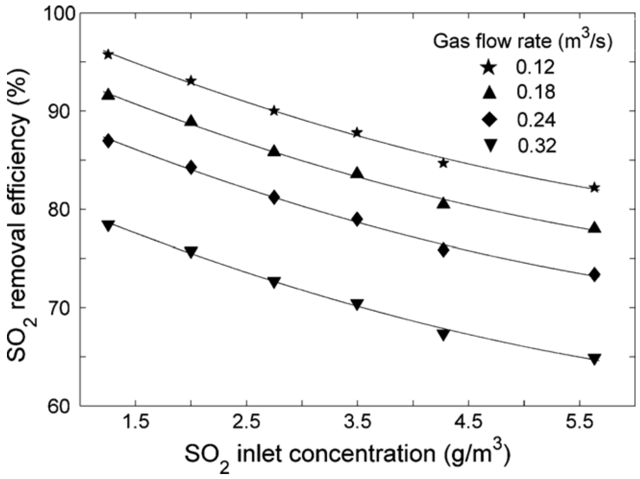

Fig. 8. SO_2 removal efficiency at various gas flow rates for different SO_2 inlet concentrations.

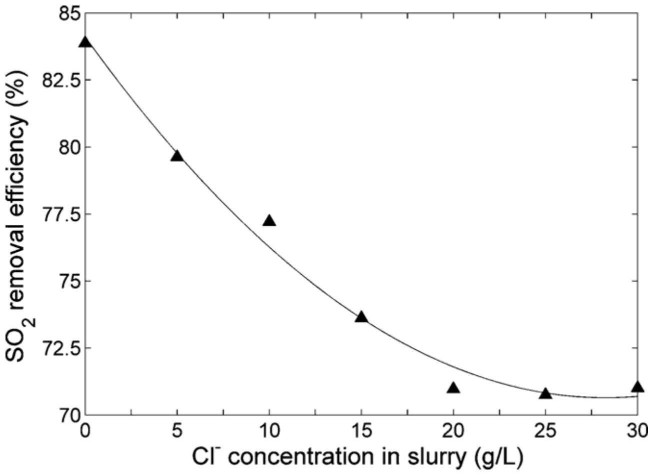

Fig. 9. SO_2 removal efficiency for different Cl^- concentrations.

4.5. Effects of Cl^-

The PCF process is a closed-loop desulphurization process. Cl is released during the coal combustion as HCl and can be subsequently absorbed in the slurry. According to Eden and Luckas [7] and Frandson et al. [23], the steady state slurry concentration of Cl^- should not exceed 25–30 g/L. And hence, this interval of concentration was studied in present study. Cl was added to the slurry tank in the form of $CaCl_2(s)$. As shown in Fig. 9, the addition of Cl^- can strongly affect the desulphurization process. When the Cl^- concentration in slurry reaches 25 g/L, SO_2 removal efficiency decreases from 83.87% down to 70.75%. The main reason for this effect is assumed to be a decrease in the dissolution rate of the limestone [24–26]. When the concentration of Cl^- is over 30 g/L, this variation trend becomes weak. Besides, the ion of Cl^- has many other side-effects on desulphurization system [9], such as influencing the dehydration of desulphurization gypsum, eroding equipments, and reducing the degree of $SO_4{}^{2-}$ removal. So some measures must be taken to get rid of the Cl^- in desulphurization process.

5. Conclusions

(1) The novel type PCF device has a good overall performance for FGD. Under moderate conditions employed, the content of SO_2 in outlet flue gas can achieve a level much lower than that permitted, while the pressure drop is very small due to co-flows in preliminarily treating chamber and no venturi structure in inlet tube. Lower pressure drop means less energy consumption, which is beneficial to the energy-saving.

(2) Guide plates and self-excitation chamber can improve the SO_2 removal efficiency by intensifying the mass-transfer and second purification. Gas flow rate has great effect on the pressure drop, while the effect of liquid–gas ratio on the pressure drop is not heavy.

(3) Some optimal or feasible conditions are obtained: slurry pH value = 5.6–6.0, liquid–gas ratio = 8.7–10.4 L/m^3 and superficial gas velocity in inner cylinder = 2.8–4.2 m/s. Compared with other absorber, the PCF device possesses large specific capacity for flue gas treatment.

(4) The addition of Cl^- decreases the SO_2 removal efficiency significantly. As the concentration of Cl^- in slurry reaches 25 g/L,

the desulphurization efficiency decreases from 83.87% down to 70.75%.

(5) Although there is no demister in the PCF device, no water was observed in out flue gas during the course of experiments, showing that the PCF device possesses a good dewatering performance. Simultaneously, no demister means lower energy-consumption, cost and maintenance.

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Appendix A. Nomenclature

C_{in}	inlet SO_2 concentration (mg/m^3)
C_{out}	outlet SO_2 concentration (mg/m^3)
Δp	pressure drop (Pa)
V_G	gas flow rate (m^3/s)
V_L	liquid flow rate (m^3/s)

Greek letters

β	radial angle of self-excitation channels
γ	horizontal angle of self-excitation channels
η_{pre}	SO_2 removal efficiency in preliminary treating chamber
η	SO_2 removal efficiency

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