

Experimental investigations on pilot plant SO₂ emission control system

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This paper provides experimental investigations for optimizing the parameters to work with pilot plant experimental system for achieving maximum SO₂ removal efficiency. It is analysed with different absorbers such as, water, NaOH, H₂O₂ and H₂SO₄ with different combinations and concentrations. The results reveal that, the removal efficiency acquired is 98.4% by adding 0.1M hydrogen peroxide (H₂O₂) externally on 0.01M sulphuric acid (H₂SO₄) solution. From the analysis, polypropylene pall rings with diameter of 15 mm, liquid flow rate of 150 lph, gas flow rate of 40 m³/hr and packed height of 1M are chosen based on their performance in increasing the SO₂ absorption rate.

Keywords: SO₂ absorption, SO₂ removal efficiency, packed column, hydrogen Peroxide, sulphuric acid.

Introduction

The presence of sulphur dioxide in flue gas occurs in many common processes such as combustion of coal and oil, chemical and metallurgical furnaces, sulphuric acid manufacturing plants and smelting processes. Flue Gas Desulphurization Technology (FGD) is the current state-of-the-art technology used for removing SO₂ from exhaust gases (Soud, 2000)⁸. It is classified into wet, semi dry and dry techniques. Cofala *et al.* (2004) ensures that the removal efficiency of SO₂ is up to 90% in case of wet FGD technique as compared to dry and semi dry techniques. Different authors used different absorbents for wet FGD technique such as, water with NH₃ (Ipek *et al.* 2008), sulphuric acid with hydrogen peroxide (Colle *et al.* 2004, 2005a, 2005b), and NH₃ Schultes (1998). For the present study, wet FGD process is selected with different absorbents are used as a reactant. The most commonly used device for absorption is packed column (Gomez *et al.* 2007 and Colle *et al.* 2004). The detailed column parameters used by several authors is given in Table 1. All the column parameters specified in Table 1 are taken as the reference for designing the pilot packed column.

Materials and Methods

Design of Packed column

Packed column should be designed in such a way that it must produce maximum absorption as possible

as it can. The performance of the column is depends on the maintenance of good liquid and gas distribution throughout the surface area. Better performance of the column is obtained by properly designing the parameters of the column such as absorbent used, diameter, total height, packing material, type of packing material used, packing height and Liquid/Gas (L/G) ratio. Based on the existing column parameters given in Table 1, the design specification of the packed column are as follows:

Liquid flow rate to the packed column - 27 to 150 litre/hr, Gas flow rate to the packed column - 13 to 40 m³/hr, Packed area- 0.6 to 1.5 m, Packing materials - Ceramic rings with 10 mm diameter, polypropylene pall rings with 10 mm diameter, and polypropylene pall rings with 15 mm diameter, Absorbents - NaOH, H₂O₂, H₂SO₄, H₂SO₄ with mixed H₂O₂, H₂SO₄ with added H₂O₂ with different concentrations, Inlet concentration of SO₂ -1000 to 500 ppm, Inlet air pressure -0.5 to 1.5 bar, Inlet SO₂ pressure - 0 to 0.25 bar, and Inlet SO₂ gas flow rate - 0 to 0.2 lph (For 100% pure SO₂). The efficiency of an absorption process is depends on the solubility of the gaseous contaminant. For most gases, only absorbents provide a high degree of turbulent contact and a long residence. The packed column is used for liquid-gas absorption process designed with vertical tubes filled with suitable packing materials. Packing materials used for this analysis are ceramic rings, polypropylene pall rings with 10 mm diameter and polypropylene pall

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Table 1—Existing column parameters

S.No	Author	SO ₂ concentration at the inlet of the packed column	Flow rate of gas at the inlet	Flow rate of liquid at the inlet	Absorbent Used	Size of the packed column
1	Fang Li-jun <i>et al.</i> (2010)	1799 - 3062 ppm	2.31m/s to 4.63m/s	18 L/m ³ to 11.25L/m ³	Two types of lime stones	Area 0.49×0.49 m ²
2	Daine Thomas <i>et al.</i> (2003)	200-5500 ppm	0.20–0.41 m/s	0.2–0.33 cm/s	H ₂ SO ₄ with added H ₂ O ₂	Diameter 0.045m, height 0.54 m.
3		0-5000 ppm	1 m ³ /hr –for contactor A 1.88 m ³ /hr –for contactor B	24 litre/hr	H ₂ SO ₄ with added H ₂ O ₂	Height 0.455 m
4	Colle <i>et al.</i> (2005)	0-5000 ppm	38-50 m ³ /h	0.12 -0.21m ³ /h	H ₂ SO ₄ with added H ₂ O ₂	Diameter 0.15 m, packed height 0.6 m
5	Yuegui <i>et al.</i> (2009)	0–1000 ppm	--	--	H ₂ O ₂ with Ca(OH) ₂	diameter 0.2 m Packed height of 1.0 m.

rings with 15 mm diameter. The process consists of spraying liquid, counter current to the gas at the top of the packed column. The liquid phase is injected through spray towers as fine droplets. The liquid is collected at the bottom of the column called recirculation tank and it is pumped again to the spray tower. The interfacial area between gas and liquid promotes the mass transfer.

To enhance the mass transfer, packed bed is placed in the reaction area of the packed column. In the packed column, random packing is chosen to increase the mass transfer rate. This provides more liquid-gas mass transfer area than the structured packing.

Chemical Analysis

Absorption with chemical reaction involves, removing the impurities from gas phase and dissolved in to a liquid phase by making a chemical reaction between them. The most commonly used model for describing the absorption process is the two-film theory (Whitman, 1923). This absorption mechanism starts with three assumptions such as, the rate of mass transfer takes place in each gas phase with the liquid phase, the two-film model assuming that the gas and liquid phases are contact with each other, and separated by an interface area where they meet, complete mixing takes place in gas and liquid phases and the interface is at equilibrium with respect to pollutant molecules transferring in or out of the interface. In SO₂ emission control process, gas phase (SO₂) is controlled by regulating the liquid phase (H₂O₂) to achieve high degree of solubility.

Experimental Setup

A 100% pure SO₂ gas form the gas cylinder is mixed with compressor air to maintain the

concentration of SO₂ at the inlet at 5000 ppm in the mixing chamber. SO₂ and air are supplied from the respective SO₂ gas cylinder and air compressor. From the mixing chamber, mixed gas is entered at the bottom of the packed column through air dryer. This mixed solution in the recirculation tank is pumped to the top of the packed column with the flow rate of 0-150 litre/hr. In the packed column, based on the mass transfer between gas and the mixed solution, SO₂ gets absorbed. The untreated gas exits through the outlet of the packed column. The concentration of SO₂ at the inlet and outlet of the packed column is measured by two SO₂ gas sensors and transmit equivalent current signals to the computer through VMAT DAQ card. Based on the SO₂ concentration in the untreated gas, flow rate of H₂O₂ is regulated through the peristaltic pump.

Results and Discussions

The packed column parameters are optimized for achieving the maximum SO₂ removal efficiency. For this optimization, the experimental runs are carried out in a pilot plant by changing the column parameters such as absorbent with various concentrations and combinations and L/G ratio. Experimental results are presented in this section to study the effect of these operating conditions on SO₂ removal efficiency.

Effect of absorbents on SO₂ removal efficiency

The effect of absorbents is analysed by conducting the experiments in the pilot plant continuously for 8 hours with each absorbent. The effect of absorbents with respect to removal efficiency for different absorbents such as water, hydrogen peroxide (H₂O₂,

0.1 M), sulphuric acid (H_2SO_4 , 0.01M), sodium hydroxide (NaOH , 0.1 M) and mixed hydrogen peroxide (0.1M) with sulphuric acid (0.01M) are recorded and shown in Figure 1. From the figure, it is observed that the removal efficiency obtained from water absorbent is only between 50 to 65% at various SO_2 inlet concentrations. Though the water is most used absorbent in industrial practice, the fractional absorption is occurred during continuous runs. Hence it is not considered for present study. During the reaction between SO_2 and sulphuric acid, the equilibrium between gas and liquid is maintained as constant but it is not so for increased sulphuric acid concentrations during continuous runs. Thus the removal efficiency is reduced during continuous absorption as compared to hydrogen peroxide absorption as shown in Figure 1. Similarly, during the experimental study using NaOH as an absorbent, the reaction kinetics and equilibrium between SO_2 with NaOH vary rapidly. Since NaOH solution does not produce resistance during gas and liquid reaction, 0.1M NaOH produces around 90% for various inlet SO_2 concentrations and the graph shows removal efficiency gradient is more than hydrogen peroxide at different SO_2 inlet concentrations as an absorbent. The figure also reveals that the hydrogen peroxide with 0.1M provides around 87% SO_2 removal efficiency than the sulphuric acid. Though NaOH solution provides better removal efficiency, hydrogen peroxide is chosen as an absorbent for this study to produce a by product rather the effluent to the atmosphere. Also, hydrogen peroxide is less toxic, safe for storage, non pollutant and easily decomposes in to water and oxygen. The results obtained by the experiments provide a quite satisfactory agreement with the results provided by Colle *et al.* (2004).

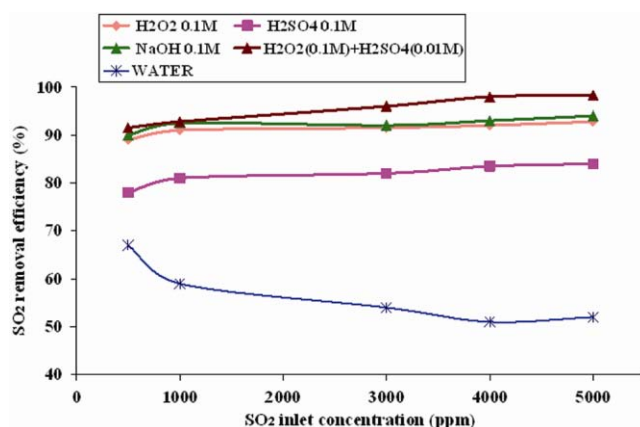


Fig. 1— SO_2 removal efficiency for various absorbents

Effect of H_2O_2 with added H_2SO_4 on SO_2 absorption

Though pure hydrogen peroxide and mixed solution including $\text{H}_2\text{SO}_4 + \text{H}_2\text{O}_2$ produces better removal efficiency (85-90%), quite interesting results are obtained for H_2SO_4 with externally added H_2O_2 . H_2O_2 with 0.1 M is added externally with various concentrations of H_2SO_4 and its effect on SO_2 removal efficiency is recorded and shown in Figure 2. From the figure, it is observed that the flow rate of H_2O_2 has the direct influence on the SO_2 removal efficiency. In this work, removal efficiency obtained for externally added H_2O_2 provides 98.4% SO_2 removal efficiency at the maximum. The SO_2 removal efficiency for different concentrations of H_2SO_4 is shown in the figure, it is clear that the increase in sulphuric acid concentrations reduces the SO_2 removal efficiency. The similar result is ensured from the results obtained by Colle *et al.* (2004).

Effect of L/G ratio on SO_2 absorption

The experimentation is carried out to optimize the L/G ratio for the absorbent that contains H_2SO_4 with externally added H_2O_2 solutions. The experiments are conducted by varying liquid and gas flow rate with different L/G ratio for the following 4 cases, Case 1:1.27litre/hr /7m³/hr, Case 2:50 litre/hr /13m³/hr, Case 3:75 litre/hr /21m³/hr, and Case 4:150 litre/hr /40m³/hr. The increase in L/G ratio increases the absorption rate for the 4 test cases. The absorption study is limited to an L/G ratio of 150 lph/ 40m³/hr since further increase in L/G ratio results flooding over the column. Hence the L/G ratio is optimized at 150 lph/40 m³/hr for further experimentation.

Effect of packing material and packed height on SO_2 removal efficiency

Selection of packing material plays a key role in determining absorption rate in the packed column.

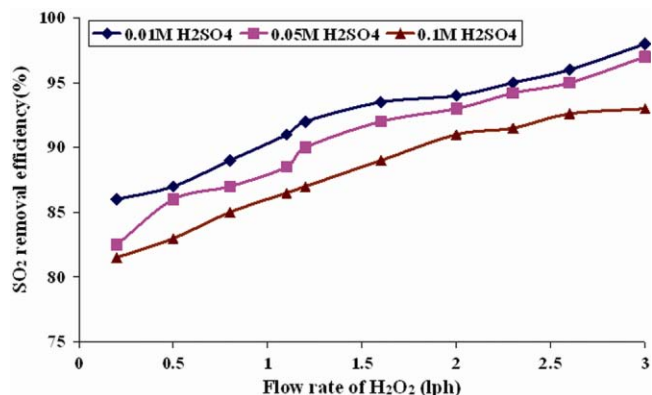


Fig. 2—Effect of H_2O_2 0.1M + H_2SO_4 various concentrations on SO_2 removal efficiency

Different types of packing material used as pointed out in Table 1, among which ceramic rings and polypropylene pall rings are considered for this experimental study. Ceramic ring with 10 mm diameter, polypropylene pall rings with 10 mm and 15 mm diameter are chosen for experimental studies. Though the ceramic ring is still used in industrial packed column, Polypropylene pall ring provides increased liquid distribution characteristics since it is designed with opening stripes of the surfaces. Hence the polypropylene pall ring with 15 mm diameter is considered for further studies. For the present study, packed height is optimized as 1 m since the packed height increases beyond 1m, more liquid is accumulated at the top of the packed column.

Conclusion

For flue gas desulphurization process, sulphuric acid with externally added hydrogen peroxide is an attractive alternative to bring down the solid and other secondary waste produced by other FGD techniques. Sulphuric acid with 0.01M with 0.1M externally added hydrogen peroxide produces 98.5% SO₂ removal efficiency as compared to the other combinations. A better control action can be provided by regulating the flow rate of hydrogen peroxide, since it has a direct influence on SO₂ removal efficiency. This pilot plant packed column developed with 150 mm inner diameter can be used to receive a maximum of 150 litre/hr liquid flow rate and 40 m³/hr gas flow rate. Packed height is selected as 1m and polypropylene pall rings with 15 mm diameter are chosen for analysis because they produced better SO₂ removal efficiency. With these optimized parameters, a suitable controller will be developed to reduce the SO₂ emission, by regulating the flow rate of H₂O₂.

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Nomenclature:

H ₂ O ₂	:	Hydrogen peroxide
H ₂ SO ₄	:	Sulphuric acid
L	:	Liquid inlet flow rate to Gas-Liquid absorption column(litre/hr)
G	:	Gas inlet flow rate to Gas-Liquid absorption column(m ³ /hr)
SO ₂	:	Sulphur dioxide
NaOH	:	Sodium Hydroxide
ACF	:	Activated Carbon Fibre
NH ₃	:	Ammonia
DAQ	:	Data Acquisition
L/G ratio	:	Liquid to Gas ratio

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