

## Critical flow atomizer in SO<sub>2</sub> spray scrubbing

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### Abstract

Owing to its various deleterious effects, abatement of SO<sub>2</sub> from point sources assumes significant importance over the years. Spray towers offer great advantages over other gas cleaning devices. The performance of a counter-current spray tower for SO<sub>2</sub> scrubbing using water and dilute NaOH deploying an energy efficient critical flow atomizer is reported. The atomizer is capable of generating finer drops at very high velocity with high degree of spray uniformity. Preliminary hydrodynamic studies indicated that droplet diameter and velocity were strongly dependent on atomizing air pressure and liquid flow rate. Experimentation revealed that SO<sub>2</sub> removal efficiency increased with the increase in liquid flow rate, liquid-to-gas flow rate ratio, atomizing air pressure, droplet velocity and pH of the scrubbing liquor while it decreased with the increase in droplet diameter and gas flow rate. Inlet SO<sub>2</sub> concentration, however, has no noticeable effect on the removal efficiency. Very encouraging results were obtained for removal efficiency (~100%) and critical design parameters. Results also indicated that the present system is energy wise and efficiency wise much better than the existing systems. Empirical and semi-empirical correlations were developed for predicting the scrubbing performances as a function of pertinent variables studied in water and alkali, respectively. The water scrubbing efficiency and the mass transfer enhancement factor were combined while predicting the performance in alkaline scrubbing. The predicted values fitted excellently well with the experimental values. That the enhancement factor of mass transfer in alkaline scrubbing could never be expected to increase indefinitely with the concentration ratio of alkali to SO<sub>2</sub> at the interface is a striking feature of this study verified by experimentation. This finding does not seem to be detailed in the available literature of gas–liquid mass transfer as well as gas scrubbing. Determination of sizing parameters is also described for the purpose of designing. © 2007 Elsevier B.V. All rights reserved.

**Keywords:** Spray tower (scrubber); Two-phase critical flow atomizer; Air pollution control; SO<sub>2</sub> scrubbing

### 1. Introduction

Abatement of SO<sub>2</sub> assumes significant importance due to its deleterious effects on living systems. Legion of SO<sub>2</sub> scrubbing technologies have been evolved from the requirements of a number of industries stemming from their definite process and regulatory mandates. Scrubbing of SO<sub>2</sub> is common in petroleum industry, power generation, incineration, pulp and paper, metallurgical installations so on and forth. Each of these processes has specific scrubbing need. Amongst various wet scrubbers studied so far for SO<sub>2</sub> scrubbing, literature [1] revealed that wet scrubbers with column internals were replaced with spray towers especially due to its ability to treat large volume of gas in

addition to its other advantages like offering least pressure drop (compared to any other gas–liquid contacting devices) and no scaling problem.

Information available in the literature in the last decade is sparse on the subject although a flurry of research was undertaken over the past few decades. Some of the studies that were undertaken in the recent past are briefly highlighted here. Schmidt and Stichlmair [2] reported on the modeling of SO<sub>2</sub> absorption in co-current spray scrubbers using NaOH as one of the reagents for studying the effects of the different operating variables on the number of transfer units (NTU) for the purpose of designing. Experimental values were well predicted through the model developed. Brogren and Hans [3] developed a model based on the penetration theory to calculate the dynamic absorption rate of SO<sub>2</sub> into a droplet of limestone slurry. The model includes both instantaneous equilibrium reactions and reactions with finite rates; limestone dissolution, sulfite oxidation, gypsum crystallization and the hydrolysis reaction of CO<sub>2</sub>. The model elucidated various facets of reactive mass transfer; importantly,

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### Nomenclature

$C''$	initial NaOH concentration (kg/m <sup>3</sup> )
$C_{\text{SO}_2,i}$	inlet SO <sub>2</sub> concentration (ppm)
$C_{\text{SO}_2,\text{int}}$	interfacial SO <sub>2</sub> concentration (kmol/m <sup>3</sup> )
$C_{\text{SO}_2,o}$	outlet SO <sub>2</sub> concentration (ppm)
$d_i$	droplet diameter in the $i$ -th group (m)
$D$	diffusivity of SO <sub>2</sub> in air (m <sup>2</sup> /s)
$D'$	diffusivity of SO <sub>2</sub> in water (m <sup>2</sup> /s)
$D''$	diffusivity of NaOH in water (m <sup>2</sup> /s)
$D_C$	diameter of the spray tower (m)
$D_d$	droplet Sauter mean diameter (SMD) (m)
$D_i$	dispersion number $\{D/(v_L H_T)\}$ , dimensionless
$f$	functions of variables, dimensionless
$F_i$	number of droplets in the $i$ -th group, dimensionless
$H_T$	height of spray tower (m)
$H_\alpha$	thermodynamic equilibrium solubility constant (kmol/m <sup>3</sup> s atm)
$i$	mole ratio of SO <sub>2</sub> to NaOH, dimensionless
$N_d$	number of droplets generated (homogeneous flow) (s <sup>-1</sup> )
ppm	parts per million (v/v), dimensionless
$P_a$	atomizing air pressure (absolute) (N/m <sup>2</sup> )
$P_{\text{SO}_2}$	partial pressure of SO <sub>2</sub> (atm)
$Q_G$	gas flow rate (m <sup>3</sup> /s)
$Q_L$	liquid flow rate (m <sup>3</sup> /s)
$Re_D$	modified droplet Reynold's number ( $D_d v_d \rho_L / \mu_L$ ), dimensionless
$S$	single droplet surface area (m <sup>2</sup> )
$S_T$	total droplet surface area generated (homogeneous flow) (m <sup>2</sup> /s)
$Sc$	modified Schmidt number $\{\mu_g / (D' C_{\text{SO}_2,i})\}$ , dimensionless
$T$	Temperature (K)
$v_d$	droplet velocity (m/s)
$v_g$	superficial gas velocity to the spray tower (m/s)
$V_{Gi}$	inlet gas velocity to the nozzle (m/s)
$V_{Li}$	inlet liquid velocity to the nozzle (m/s)
$v_L$	superficial liquid velocity to the spray tower (m/s)
$We_D$	droplet Weber number $[\rho_g (V_{Gi} - V_{Li})^2 D_d / \sigma_L]$ , dimensionless
<i>Greek letters</i>	
$\eta$	removal efficiency of SO <sub>2</sub> , percentage
$\eta_C$	removal efficiency of SO <sub>2</sub> in alkaline scrubbing, percentage
$\eta_{\text{SO}_2}$	removal efficiency of SO <sub>2</sub> in water scrubbing, percentage
$\phi$	mass transfer enhancement factor dimensionless
$\mu_g$	viscosity of gas (kg/m s)
$\mu_L$	viscosity of liquid (kg/m s)
$\rho_g$	gas density (kg/m <sup>3</sup> )
$\rho_L$	liquid density (kg/m <sup>3</sup> )
$\sigma_L$	liquid surface tension (N/m)

it was used to quantify the mass transfer within a spray scrubber and to estimate the impact of the reactions with finite rate of the SO<sub>2</sub> absorption. Pettersson et al. [4] developed a simplified model for a wet flue gas cleaning system that was verified with a SO<sub>2</sub> spray scrubber at I/S Faelles Forbraending's waste incinerator plant in Hobro, Denmark. SO<sub>2</sub> removal efficiency of around 90% was achieved using 2.5–20 kmol/m<sup>3</sup> NaOH as the scrubbing liquor for inlet SO<sub>2</sub> concentration varied from 100 to 500 ppm. Experimentation and theoretical analysis revealed that the initial SO<sub>2</sub> concentration did not influence the removal efficiency. Chen [5,6] theoretically investigated on the unsteady absorption of SO<sub>2</sub> by single atmospheric water droplet in motion and the dynamics of SO<sub>2</sub> absorption in a raindrop falling at terminal velocity. Hay et al. [7] reported on the various operating features on an once through caustic spray scrubber treating the acid plant tail gas containing ~500 ppm SO<sub>2</sub> of the Port Kembla Copper (PKC) smelter in Australia. A very high removal efficiency of about 95%+ was achieved at a pH of 11. Limestone/gypsum wet flue gas desulfurization (WFGD) process was reported [8] for controlling SO<sub>2</sub> emissions from coal-fired power plants achieving very high removal efficiencies of SO<sub>2</sub>, SO<sub>3</sub> and particulate matter. Huang [9] reported a novel theoretical model to determine the SO<sub>2</sub> removal efficiency using fine water spray. The performance of the system was shown to generally improve by reducing the droplet diameter or the initial S(IV) concentration, or by increasing the inlet SO<sub>2</sub> concentration, the droplet pH or the liquid-to-gas flow rate ratio. In one of our previous studies reported [10] on the performance of a spray-cum-bubble column for SO<sub>2</sub> removal using water and dilute NaOH solution indicated very high removal efficiency. The prediction of removal efficiency of SO<sub>2</sub> was made using experimental data as a function of gas–liquid flow rates and other physico-chemical properties of the system. The behavior of a hybrid scrubber dealing with a spray system, as one of the two stages studied, was reported and compared were the stage-wise removal efficiencies. The performance was, however, not reported in detail characterizing the parameters governing the process of atomization.

Critical analysis of the available literature revealed that the spray tower has been used for scrubbing of SO<sub>2</sub> by either water or alkalis but the performance reported so far seldom takes into account the detailed studies on the droplet size and droplet velocity in tandem augmented with the atomizing air pressure as also the liquid flow rate that are essentially desired for the operation of the spray tower. On the other hand, characterization of the spray tower during the prediction of the performance in alkali scrubbing by combining the mass transfer enhancement factor with the performance in water scrubbing does not seem to be detailed in the available literature. An attempt has been made in this article to report on the performance of a counter-current spray tower for SO<sub>2</sub> scrubbing using water and dilute NaOH deploying an energy efficient critical flow atomizer [11] that is capable of producing finer droplets at very high relative velocity without sacrificing the spray uniformity. Attempts have also been made to develop empirical and semi-empirical correlations with the help of the experimental data that would take into account the various hydro-dynamic parameters including the atomizing parameters and the mass transfer enhancement

factor for predicting the performance of the spray tower in water and in alkali scrubbing respectively.

## 2. Development of critical flow atomizer

Central to any spray operation, is the atomization or the disintegration of the liquid stream into droplets or sprays. For spray scrubbing, a good atomizer should produce a fairly uniform spray with drop diameters small enough to generate large interfacial area of contact at the same time large enough to prevent excessive entrainment. Uniformity of spray, small drop size with high velocity and low energy of atomization are the desired criteria for the atomizer. Existing commercial atomizers are requiring very high energies for desired spray hydrodynamics. Therefore, an energy efficient and cost-effective critical flow atomizer [11] satisfying the above desired criteria is used in the present study.

In the present atomizer (Fig. 1) liquid and gas are brought together in a dispersing chamber at relatively low velocity in comparison to the critical velocity of individual phases because the critical velocity of a two-phase mixture is lower than that of the individual critical velocities. This concept exploits the shock mechanism arising in critical flow of any liquid–gas or liquid–liquid mixture. Since both frictional and acceleration pressure losses are strong functions of the phase velocities, the low operating critical velocities practicable in two-phase flow limit the overall system pressure drop. At the end of the atomizer, the two-phase mixture is expanded due to a pressure jump so that critical flow occurs. At the critical flow point, a sudden pressure difference, owing to this pressure jump, at the atomizer exit, between the upstream and downstream creates an instantaneous shock in the fluid stream which results in intense mixing of the phases as well as blasting and the liquid leaves the atomizer in the form of uniformly dispersed fine sprays. The advantages of this design consist in lower frictional acceleration losses compared to the external mixing atomizer. As can be seen in Fig. 1 that the gas–liquid mixing area can be

adjusted so that sprays with different droplet sizes and different initial droplet velocities can be generated. It can also be seen from the figure that the gas–liquid mixing volume and mixing area can be adjusted so that either sprays with different droplet Sauter Mean Diameters (SMDs) at a fixed mean droplet velocity, or a fixed droplet SMD with different mean velocities can be generated.

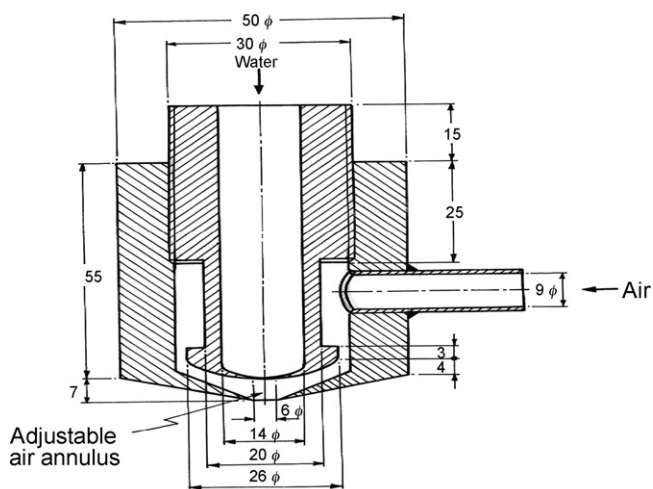
## 3. Selection of absorbent

The selection of a suitable absorbent or scrubbing liquid poses a very complex problem for removing  $\text{SO}_2$  from waste gas stream and is of significant importance for process design as well. Many of the problems experienced at various facilities are the result of the inappropriate selection of a scrubbing process [7]. For instance, a scrubber using limestone slurry that works well on the steady and weak  $\text{SO}_2$  generated in a coal-fired boiler will not be suitable for the stronger and fluctuating  $\text{SO}_2$  produced by the metallurgical processes. Common commercial scrubbing processes utilized lime/limestone and dual alkali. Lime slurry and limestone scrubbing is suitable for relatively low concentrations of  $\text{SO}_2$  [ $<100\text{--}5000$  ppm] and moderate collection efficiencies [90–95%]. When concentration of  $\text{SO}_2$  absorbed became sufficiently large [ $<100\text{--}150,000$  ppm] to make the economics of a simple throwaway process uneconomical, the waste solutions can be regenerated in the dual alkali process by reacting it with lime i.e.,  $\text{Ca}(\text{OH})_2$  outside the scrubber circuit. This approach permits the gas to be contacted with a clear solution of highly soluble scrubbing agent (usually sodium or aluminum based), thereby minimizing scaling, plugging and erosion problems in the absorbent circuit. The removal efficiency that can be offered by the dual alkali process is also very high [99%+]. Furthermore, it has been pointed out that the neutralization capacity of NaOH for  $\text{SO}_2$  is extremely high [12]. The reduction of  $\text{SO}_2$  concentration below 5 ppm (threshold limit value of  $\text{SO}_2$ ) can best be achieved with appropriate controls on pH, ionic strength and liquid recirculation rate with sodium alkalis.

Considering the foregoing discussion, water has been selected as a scrubbing liquid in our prelude study to characterize the overall performance of the spray tower and later dilute NaOH has been selected as a reagent for attaining very high removal efficiency under the optimum scrubber operating conditions that followed from water scrubbing. Furthermore, such a selection of dilute NaOH will help in determining the critical design parameters: the volumetric gas-side mass transfer coefficient and number of transfer unit [13] for spray tower and investigating into the definite insight of the process. The detailed reaction schemes for  $\text{SO}_2\text{--H}_2\text{O}$  and  $\text{SO}_2\text{--NaOH}$  considered in the present article is discussed in the reported literature [10,14].

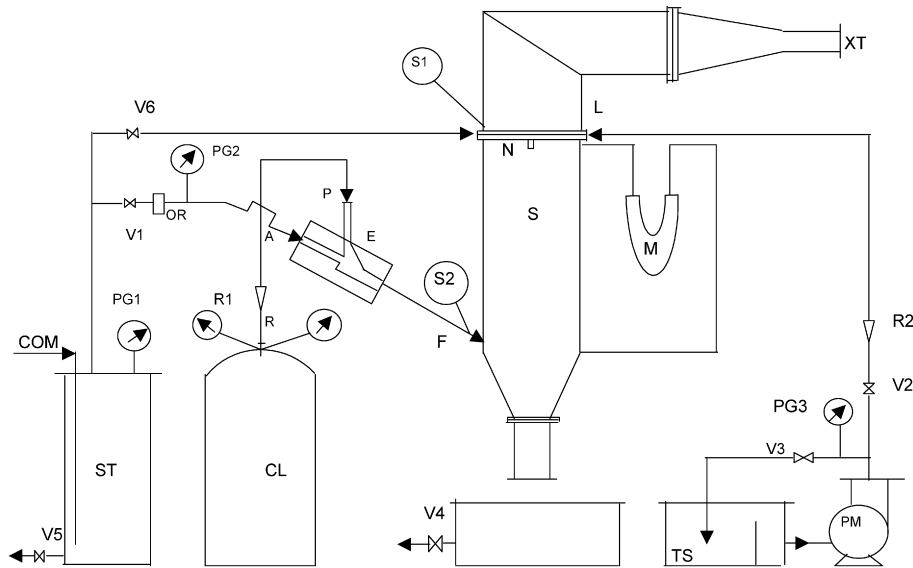
## 4. Experimental methods

The experimental column is a vertical cylindrical Perspex column, 0.1905 m in diameter and 2.0 m long. At the top of the cylindrical column the energy efficient two-phase critical flow



All dimensions are in mm.

Fig. 1. Schematic diagram of the two-phase critical flow atomizer.



A: Primary air line; CL: SO<sub>2</sub> Cylinder; COM: Air from compressor; E: Air jet ejector; F: Air diluted SO<sub>2</sub> entry; L: Liquid entry; M: Manometer; N: Two-phase critical flow atomizer; OR: Orifice meter; P: SO<sub>2</sub> entry into the ejector; PG1 – PG3: Pressure gauges; PM: Pump; R: SO<sub>2</sub> regulator; R1 – R2: Rotameters; S: Spray tower; S1-S2: Sampling points; ST: Surge tank; TS: Scrubbing liquid tank; V1 – V6: Gate valves; XT: Exhaust

Fig. 2. Schematic diagram of the experimental setup.

atomizer was provided for generating sprays. The simulated SO<sub>2</sub> concentration in the range of 500–2000 ppm was generated by mixing air and SO<sub>2</sub> in an air-jet ejector (E) assembly for intense mixing of the components. Compressed air at the desired motive pressure and flow rate was forced through the air nozzle and simultaneously the SO<sub>2</sub> from the SO<sub>2</sub> cylinder (CL), was routed through SO<sub>2</sub> regulator (R), into the ejector at point P. The air and SO<sub>2</sub> mixed intensely in the mixing throat of the ejector and the mixture was allowed to feed into the spray tower at point F. SO<sub>2</sub> concentration was varied by changing the flow rate of SO<sub>2</sub> with the help of rotameter (R<sub>1</sub>). The detailed experimental setup is schematically shown in Fig. 2 [15].

In the actual experiment, water and dilute NaOH solution (scrubbing liquid-phases studied) were pumped into the column through the atomizer routed through valve (V<sub>2</sub>) and rotameter (R<sub>2</sub>). Low pressure [ $1.19 \times 10^5$ – $1.68 \times 10^5$  N/m<sup>2</sup> (abs)] air was used to convert the liquid into fine sprays at high velocities. A simulated air borne SO<sub>2</sub> mixture was then introduced into the spray tower as discussed earlier. The scrubbing experiments were carried out without liquid recirculation. The experimental conditions were as follows:

#### 4.1. Spray hydrodynamics and water scrubbing

$$T = 306 \pm 1 \text{ K}; \quad Q_L = 0.4 \times 10^{-5} - 4.02 \times 10^{-5} \text{ m}^3/\text{s}; \quad Q_G = 3.75 \times 10^{-3} - 6.20 \times 10^{-3} \text{ m}^3/\text{s}; \quad P_a = 1.19 \times 10^5 - 1.68 \times 10^5 \text{ N/m}^2 \text{ (abs)}; \quad D_d = 33.6 \times 10^{-6} - 149 \times 10^{-6} \text{ m}; \quad v_d =$$

$21.06 - 35.33 \text{ m/s}; \quad C_{\text{SO}_2, i} = 500 - 2000 \text{ ppm};$  relative humidity of the ambient air =  $98.2\% \pm 2\%$ . The atomizing air introduced into the scrubber was about 1% of the total gas flow rate. The effects of  $C_{\text{SO}_2, i}$  as well as various flow and operating variables on the performance of the spray tower were studied at a fixed  $P_a$  of  $1.19 \times 10^5$  N/m<sup>2</sup> (abs), while the effect of  $D_d$  on the performance was separately conducted at different  $P_a$  ranging between  $1.23 \times 10^5$  and  $1.68 \times 10^5$  N/m<sup>2</sup> (abs).

#### 4.2. Alkali scrubbing

Alkaline scrubbing experiments were carried out under optimum hydro dynamical conditions in water scrubbing discussed later. The chemical variables studied were:  $C_{\text{SO}_2, i} = 500 - 2000 \text{ ppm}; \quad C'' = 2 \times 10^{-4} \text{ to } 50 \times 10^{-4} \text{ kmol/m}^3; \quad T = 306.7 \pm 1 \text{ K};$  relative humidity of the ambient air =  $97.7\% \pm 2\%$ .

The SO<sub>2</sub> concentrations (at S<sub>1</sub> and S<sub>2</sub>) were analyzed using aspirator bottles by the Iodine Method [16] under steady operating conditions as detailed in the literature [10,14]. The SO<sub>2</sub> concentrations were corrected for the dilute atomized air (~1%) that was introduced into the gas phase. The reproducibility of the measured SO<sub>2</sub> concentration using Iodine Method was 97%. The droplet size was measured with a Phase Doppler Analyzer (PDA). The PDA was programmed to evaluate the droplet Sauter Mean Diameter (SMD) in situ while measuring the size distribution. The SMD or the volume to surface mean diameter is



Table 1

Experimental data on liquid flow rate, droplet SMD and calculated values of number of droplets generated per second, single droplet surface area, total droplet surface area generated per second

Liquid flow rate (m <sup>3</sup> /s)	Atomizing air pressure, $P_a$ , (N/m <sup>2</sup> )	Droplet SMD $D_d \times 10^6$ (m)	Number of droplets $N_d$ (s <sup>-1</sup> )	Single droplet surface area, $S$ (m <sup>2</sup> )	Total droplet surface area generated $S_T = N_d \times S$ (m <sup>2</sup> /s)
(a) For SO <sub>2</sub> scrubbing at a constant atomizing air pressure					
5.56E-06	1.19E+05	4.16E-05	1.48E+08	5.44E-09	0.8019
1.11E-05		7.22E-05	5.63E+07	1.64E-08	0.9224
1.83E-05		1.02E-04	3.26E+07	3.29E-08	1.0733
2.50E-05		1.19E-04	2.84E+07	4.44E-08	1.2616
3.11E-05		1.40E-04	2.17E+07	6.14E-08	1.3348
(b) For SO <sub>2</sub> scrubbing at various atomizing air pressures					
4.10E-06	1.23E+05	5.00E-05	6.26E+07	7.85E-09	0.4920
	1.39E+05	3.38E-05	2.03E+08	3.59E-09	0.7278
	1.58E+05	3.57E-05	1.72E+08	4.00E-09	0.6891
	1.68E+05	3.33E-05	2.12E+08	3.48E-09	0.7387
1.04E-05	1.23E+05	7.22E-05	5.28E+07	1.64E-08	0.8643
	1.39E+05	5.06E-05	1.53E+08	8.04E-09	1.2332
	1.58E+05	4.11E-05	2.86E+08	5.31E-09	1.5182
	1.68E+05	3.86E-05	3.45E+08	4.68E-09	1.6166
2.02E-05	1.23E+05	9.43E-05	4.60E+07	2.79E-08	1.2853
	1.39E+05	7.90E-05	7.82E+07	1.96E-08	1.5342
	1.58E+05	5.90E-05	1.88E+08	1.09E-08	2.0542
	1.68E+05	5.35E-05	2.52E+08	8.99E-09	2.2654
3.01E-05	1.23E+05	1.40E-04	2.10E+07	6.14E-08	1.2918
	1.39E+05	1.07E-04	4.69E+07	3.60E-08	1.6879
	1.58E+05	7.93E-05	1.15E+08	1.98E-08	2.2774
	1.68E+05	6.99E-05	1.68E+08	1.53E-08	2.5837
4.02E-05	1.23E+05	1.49E-04	2.32E+07	6.97E-08	1.6188
	1.39E+05	1.27E-04	3.71E+07	5.10E-08	1.8932
	1.58E+05	1.03E-04	7.05E+07	3.33E-08	2.3440
	1.68E+05	8.06E-05	1.47E+08	2.04E-08	2.9926

defined as:

$$\text{SMD} = \sum \frac{F_i \times d_i^3}{F_i \times d_i^2} \quad (1)$$

where  $F_i$  is the number of droplets in the  $i$ -th group. SMD averages the diameter (first order of length) yet weighs according to its surface area (second order of length), which contributes to the total surface area of the droplet population. SMD is useful in the present case since gas-droplet mass transfer is influenced by the droplet surface amongst other parameters similar to other gas-liquid mass transfer applications.

The mean velocity and the root mean square (r.m.s.) velocity of the droplets were measured by Laser Doppler Velocimetry (LDV) method. Mean droplet velocities were measured 1 m downstream from the nozzle exit. The measured droplet SMDs along with the atomizing air pressure necessary to generate the droplets are presented in Table 1. The errors in the measured and reported flow rates and droplet sizes were within  $\pm 5\%$ .

## 5. Results and discussion

Experiments on the SO<sub>2</sub> scrubbing have been conducted at various process operating conditions as mentioned earlier. The percentage removal of SO<sub>2</sub> has been calculated in each run by

the formula:

$$\eta = \left[ \frac{(C_{\text{SO}_2,i} - C_{\text{SO}_2,o})}{C_{\text{SO}_2,i}} \right] \times 100 \quad (2)$$

The spray hydro dynamics are initially highlighted and then the trends of variation of percentage removal (performance analysis) are discussed in details in the respective sections.

### 5.1. Spray hydrodynamics

#### 5.1.1. Effect of atomizing air pressure on droplet SMD

The effect of atomizing air pressure (absolute) on droplet SMD at various liquid flow rates is shown in Fig. 3. It can be seen from the figure that the droplet SMD was reduced gradually with the increase in atomizing air pressure at a constant liquid flow rate. Atomizing air at higher pressure enabled the mass of definite amount of liquid to accelerate at a higher rate when the liquid was being disintegrated into smaller droplets. In the process, reduction of droplet SMD was achieved. It can also be seen from the figure that increase in liquid flow rate, in contrast, has an opposite effect on droplet SMD. The droplet SMD increased with the increase in the liquid flow rate at a constant atomizing air pressure. It may be attributed to the fact that increase in the definite mass of liquid at a constant atomizing air pressure retarded the disintegration process and larger droplets

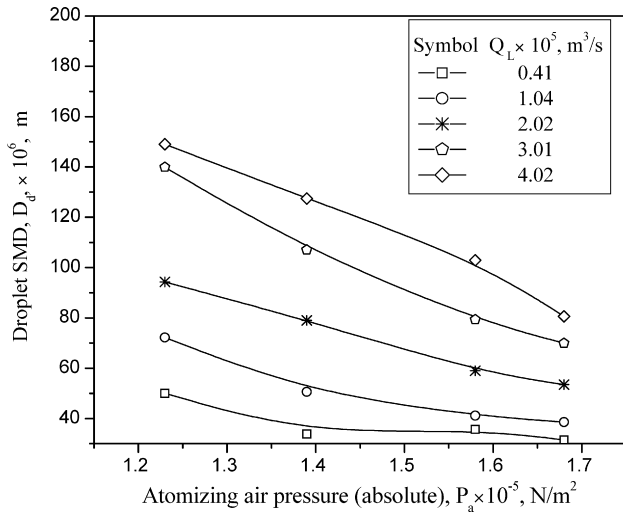


Fig. 3. Effect of atomizing air pressure on the droplet SMD at constant liquid flow rates.

were produced. The dependence of the droplet SMD on atomizing air pressure (abs) and liquid flow rate in the present system is found to be of the following form (correlation coefficient of 0.97527)

$$D_d = 3.78934 \times 10^7 (P_a)^{-1.82414} (Q_L)^{0.48657} \quad (3)$$

#### 5.1.2. Effect of atomizing air pressure on droplet velocity

The effect of atomizing air pressure on the droplet velocity at various liquid flow rates is shown in Fig. 4. It can be seen from the figure that the droplet velocity was increased sharply with the increase in the atomizing air pressure for all the liquid flow rates and finally reached almost a constant value of  $1.68 \times 10^5 \text{ N/m}^2$  (abs). Beyond this atomizing pressure no significant change in droplet velocity was observed perhaps due to attaining the critical velocity of the two-phase mixture at about 35 m/s. It can also be seen from the figure that the increase in the liquid flow rate, reduced the droplet velocity for a fixed atom-

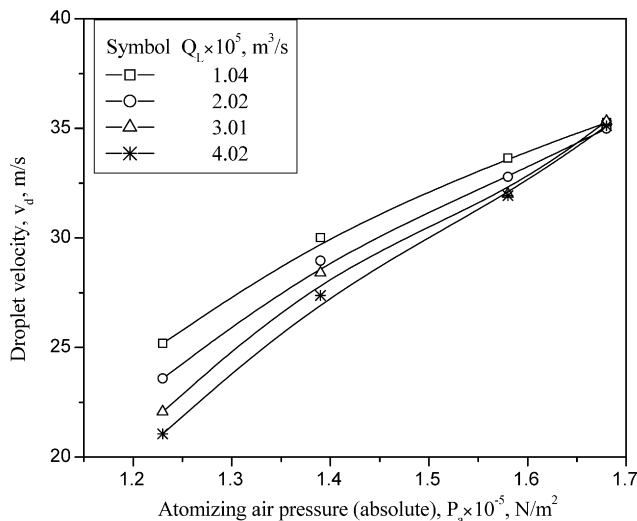


Fig. 4. Effect of atomizing air pressure on the droplet velocity at constant liquid flow rate.

izing air pressure, the reason for such variation is discussed earlier. The dependence of the droplet velocity on atomizing air pressure (abs) and liquid flow rate in the present system is found to be of the following form (correlation coefficient of 0.96689)

$$v_d = 6.73225 \times 10^{-6} (P_a)^{1.2608} (Q_L)^{-0.02754} \quad (4)$$

On combining Eq. (3) and (4) the relationship of  $D_d$  and  $v_d$  with  $P_a$  and  $Q_L$  can be comprehensively represented as:

$$[D_d \times v_d] = 2.55 \times 10^2 (P_a)^{-0.56334} (Q_L)^{0.45903} \quad (5)$$

Eq. (5) will be useful for selecting the droplet SMD and droplet velocity based on the flow regime available for a system.

#### 5.1.3. Effect of atomizing air pressure and water flow rate on fineness of droplet generation

The operating limits of the 6 mm atomizer for generating sprays of various finenesses is shown in Fig. 5 as a function of water flow rates and atomizing air pressure. In these sets of experiments the air entry area was adjusted such that the velocity of air at the entry corresponded to the velocity of sound in the two-phase mixture or was within 80% of it. It is seen from the figure that the maximum operating mass flow rate of water should be below 7.5 kg/h for generating sprays with droplet of SMD of  $10 \mu\text{m}$ , whereas the water capacity as high as 32.5 kg/h may be required for generating sprays with droplet of SMD of  $40 \mu\text{m}$ . These maximum rates corresponded to the critical mass flux as discussed earlier. Experiments with 4, 6, 8 and 10 mm atomizer have shown that the liquid capacities of the 4 and 6 mm atomizers were very low. However, for smaller atomizers, somewhat finer atomization was produced with identical flow conditions. On the other hand, it was observed necessary to use atomizers with larger diameters for generating finer droplets with higher liquid flow rates.

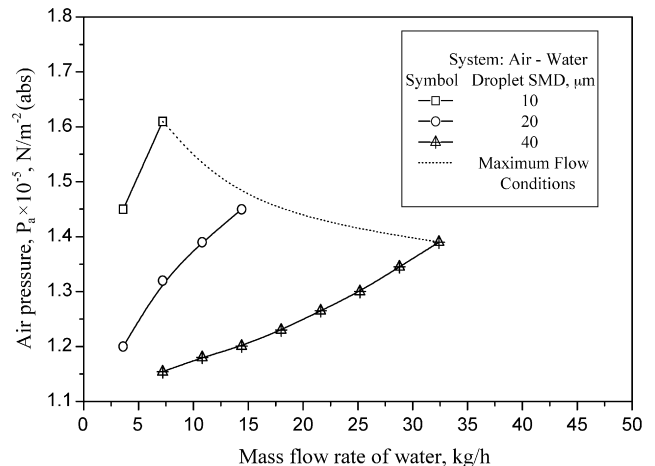


Fig. 5. Spray characteristics relating to atomizer operating conditions for 6 mm atomizer.

## 5.2. Water scrubbing of SO<sub>2</sub>

### 5.2.1. Effect of inlet concentration of SO<sub>2</sub>

The experimentation revealed that the variation of inlet SO<sub>2</sub> concentration in the range of 500–2000 ppm had insignificant effect on the percentage removal. In fact, the amount of SO<sub>2</sub> removed was increased with the increase in initial concentration of SO<sub>2</sub> since the concentration driving force was also increased in such a situation.

Furthermore, the removal efficiency is the ratio of the amount absorbed to the initial concentration and with the rise in the initial concentration of SO<sub>2</sub> both the numerator and the denominator were increased almost to the same extent in the present case that resulted in the removal efficiency almost unaffected with the increase in initial concentration of SO<sub>2</sub>. However, the removal efficiency could have increased with the rise in initial concentration of SO<sub>2</sub> if the amount absorbed at (a) lower initial concentration of SO<sub>2</sub> would have relatively lower than at the higher initial concentration of SO<sub>2</sub> and (b) higher initial concentration of SO<sub>2</sub> would have relatively higher than that absorbed actually. These could not have happened perhaps due to the fact that the tower was charged with very high velocity droplets that were capable of momentarily absorbing the gas and the droplet surface was reaching the equilibrium concentration of SO<sub>2</sub> very fast.

That the inlet SO<sub>2</sub> concentration did not have any influence on the percentage removal was theoretically investigated and validated with experiments by Pettersson et al. [4]. Huang [9] theoretically investigated that the percentage removal of SO<sub>2</sub> increased very sharply with the inlet concentration of SO<sub>2</sub> up to 10<sup>-2</sup> ppb beyond which the percentage removal reached a constant value. Therefore, the observed effect of inlet concentration of SO<sub>2</sub> on the percentage removal of SO<sub>2</sub> in the present study agreed excellently well with the findings reported in the literature [4,9]. The maximum removal efficiency achieved in water scrubbing was 73.53%.

### 5.2.2. Effect of liquid flow rate, gas flow rate and liquid-to-gas flow rate ratio

From the economic point of view, the liquid-to-gas flow rate ratio in terms of m<sup>3</sup>/1000 ACM (Actual Cubic Meter) has been found to be one of the most important criteria for reporting the scrubbing performance. In the light of this observation, the effect of  $Q_L/Q_G$  ratio on the percentage removal of SO<sub>2</sub> is shown in Fig. 6 for different gas flow rates at a constant inlet SO<sub>2</sub> loading of 2000 ppm. It can be seen from the figure that the percentage removal of SO<sub>2</sub> increased initially very sharply with the increase in  $Q_L/Q_G$  ratio and thereafter reaches almost a constant value beyond a  $Q_L/Q_G$  ratio of 3.0 m<sup>3</sup>/1000 ACM [10]. The reason for such observation may be explained as follows. With the increase in  $Q_L/Q_G$  ratio the hydraulic loading, i.e.,  $Q_L$  was increased in the scrubber. As the liquid flow rate was increased, the drop diameter and the total droplet surface area were also increased (Table 1(a)). This phenomena coupled with enhanced droplet oscillation at higher liquid flow rate might have caused the droplet-gas collision more effective and hence the percentage removal was increased with the increase in  $Q_L$  as well as

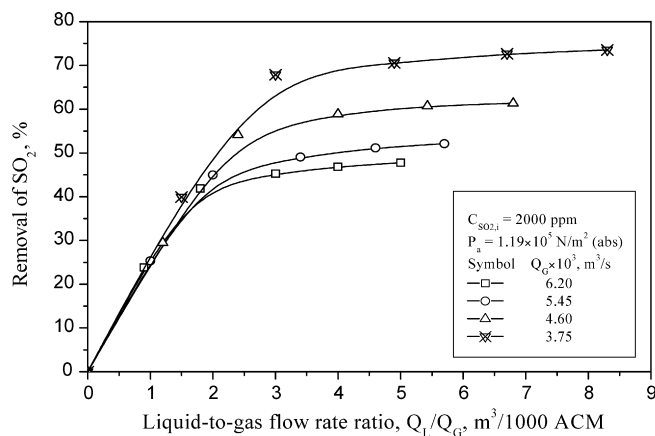


Fig. 6. Effect of liquid-to-gas flow rate ratio on the percentage removal of SO<sub>2</sub>.

$Q_L/Q_G$  ratio. It can also be seen from the figure that the increase in  $Q_G$  reduced the percentage removal. It might be due to the reduction in gas–liquid contact time i.e., the reduction in the overall mass transfer. Also the SO<sub>2</sub> accumulated at the surface of the droplet phase did not get enough time to diffuse into the bulk liquid phase and this phenomenon hindered the absorption rate of SO<sub>2</sub> at higher gas flow rate, which resulted in the reduction in the percentage removal. On the other hand, at higher gas flow rate with relatively lower liquid flow rate, the rate of flow of SO<sub>2</sub> molecules into the scrubber was also higher and there might have been an over-crowding of SO<sub>2</sub> molecules in the bulk liquid phase causing a hindrance to the overall diffusion of SO<sub>2</sub> which resulted in reduction in the percentage removal at higher gas flow rate. That the percentage removal remained almost unaltered beyond a  $Q_L/Q_G$  ratio of 3.0 m<sup>3</sup>/1000 ACM, might be due to the fact that SO<sub>2</sub> concentration in the liquid was reaching its equilibrium value in the gas phase. It could also be due to the result of increased rate of coalescence of the droplets, whereby both the number and the total droplet surface area might decrease, and the percentage removal might level off. This trend of variation of the performance of the spray tower also supports the observation made by Huang [9].

### 5.2.3. Effect of droplet SMD, atomizing air pressure and droplet velocity

In the previous discussion, the effect of droplet SMD could not be studied since the liquid flow rate was varied at a fixed atomizing air pressure and in the process the droplet SMD was also changed with the liquid flow rate. In fact, the performance of the spray tower was shown as a function of  $Q_L/Q_G$  ratio in Fig. 6. To study the effect of the droplet SMD either at constant  $Q_L$  or at fixed  $P_a$  on the performance of the spray tower a few experiments were conducted as discussed earlier. The effect of droplet SMD on the percentage removal of SO<sub>2</sub> under these circumstances is shown in Fig. 7. In general, it can be seen from the figure that the percentage removal of SO<sub>2</sub> was sharply increased with the increase in the atomizing air pressure while slowly decreased with the increase in droplet SMD for all liquid flow rates. This behavior can be explained with the variation of the droplet SMD based on the hydrodynamic parameters that

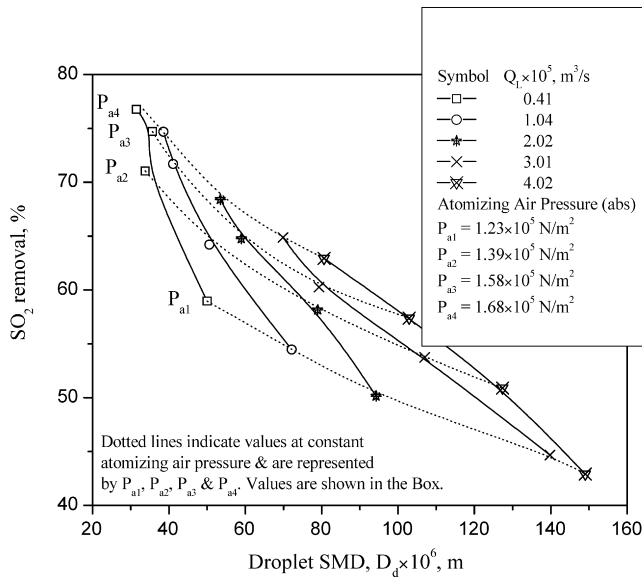


Fig. 7. Effect of droplet SMD and atomizing air pressure on the percentage removal of  $\text{SO}_2$ .

are affecting the atomization process. On increasing  $P_a$  at constant  $Q_L$ , the droplet SMD was generally observed to decrease and the droplet loading introduced into the scrubber was enormously increased (Table 1(b)) and thereby the interfacial area of contact was also increased which resulted in the sharp increase in the percentage removal of  $\text{SO}_2$ . This phenomenon expectedly resulted in the higher  $\text{SO}_2$  removal for smaller droplets than larger ones at a constant  $Q_L$ . Furthermore, on decreasing  $Q_L$  at a constant  $P_a$  the droplet SMD was reduced and the number of droplets was increased slowly effecting the total droplet surface available for mass transfer increased (Table 1(b)). This resulted in the increase in the percentage removal of  $\text{SO}_2$  slowly with the increase in the  $Q_L$ .

The effect of droplet velocity on the percentage removal of  $\text{SO}_2$  is shown in Fig. 8 for different liquid flow rates. It can be seen from the figure that the percentage removal of  $\text{SO}_2$  was

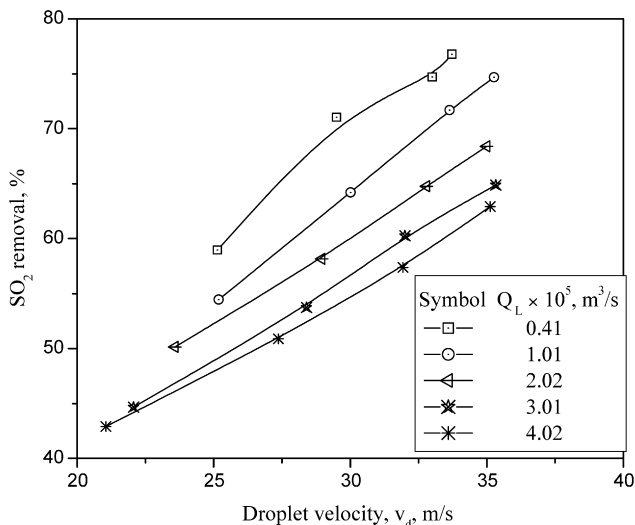


Fig. 8. Effect of droplet velocity on the percentage removal of  $\text{SO}_2$ .

increased sharply with the increase in droplet velocity. This can be explained as follows. The droplet velocity, a manifestation of the atomizing air pressure ( $P_a$ ), was increased with the increase in  $P_a$  (Eq. (3)) and hence the percentage removal of  $\text{SO}_2$  was increased as discussed earlier. Thus for an assigned droplet SMD or  $v_d$ , achieving a desired removal will require selecting  $Q_L$  and  $P_a$ . These findings are very useful for scrubber modeling as also for its design.

The comparison of energy efficiencies of the present atomizer with the commercially available Caldyn nozzle revealed that the present atomizer required energy of 2.5 kJ/kg  $\text{H}_2\text{O}/\text{h}$  at a liquid loading of 20 kg/h to generate droplets with an SMD of 40  $\mu\text{m}$ , compared with 15 kJ/kg  $\text{H}_2\text{O}/\text{h}$  for Caldyn nozzle under similar hydro dynamical conditions [11]. Thus the performance of the present atomizer, i.e., the operation of the spray tower, indicates that it is energetically better than the existing systems.

#### 5.2.4. Predicting $\text{SO}_2$ scrubbing efficiency with the help of empirical correlation

The performance of the present system takes into account of a wide range of variables of the system in addition to the physico-chemical properties of the gas–liquid system. These variables were  $C_{\text{SO}_2,i}$ ,  $C''$ ,  $Q_L$ ,  $Q_G$ ,  $D_d$ ,  $v_d$ ,  $P_a$ ,  $V_{\text{Gi}}$ ,  $V_{\text{Li}}$ ,  $D_C$  and  $H_T$ . Therefore, the performance can be presented as a function of these variables and the physico-chemical properties of the gas–liquid system. Variables considered in the available literature require analysis for the purpose of comparing the performances of the present system with the system reported. Therefore, relevant investigations are discussed here. Pettersson et al. [4] reported on  $C_{\text{SO}_2,i}$  (100–500 ppm), NaOH concentration (2.5–20 kmol/ $\text{m}^3$ ), scrubber bleed pH (3.75–5.5) for validating their model and obtained maximum removal efficiency of around 90%. Chen [5,6] developed theoretical models for  $\text{SO}_2$  absorption using single water droplets in atmospheric process falling under terminal settling velocities. Hay et al. [7] investigated on the experimental removal of  $\text{SO}_2$  (95%+) using the variables: NaOH concentration ( $\sim 5.0$  kmol/ $\text{m}^3$ ),  $C_{\text{SO}_2,i}$  ( $\sim 500$  ppm), scrubber exit  $\text{SO}_2$  concentration ( $\sim 50$  ppm),  $Q_G$  (130,000  $\text{Nm}^3/\text{h}$ ) and  $T$  (80  $^\circ\text{C}$ ). None of these investigations reported on the complete set of variables comparable to the present study especially on the parameters that govern the process of atomization, one of our prime objectives in the present case for the purpose of design and analysis of the spraying devices used commercially. Huang [9] developed a theoretical model for predicting  $\text{SO}_2$  removal efficiencies in water scrubbing and validated the model considering  $D_d$  (30–150  $\mu\text{m}$ ), droplet pH (6–9), droplet phase S(IV) concentration (0.10–10 M),  $C_{\text{SO}_2,i}$  ( $10^{-5}$ – $10^2$  ppb) and  $Q_L/Q_G$  ratio (0.025–0.00575  $\text{m}^3/1000\text{ACM}$ ). Removal efficiency of  $\sim 100\%$  was reported theoretically. Though this investigation resembles to our study, its use is limited here due to the use of very low values of  $C_{\text{SO}_2,i}$  and absence of parameters governing the process atomization. Critical appraisal of the available literature, therefore, indicates that an exact comparison of the performance achieved in the present study with the available data in the literature is not possible.

An attempt has therefore, been made to develop a correlation by dimensional analysis in order to predict the  $\text{SO}_2$  removal effi-



ciency from the directly measurable parameters of the system. The pertinent variables that could possibly influence the performance i.e., the removal efficiency,  $\eta_{\text{SO}_2}$ , of the spray tower are as:

- physical parameters: inlet  $\text{SO}_2$  concentration ( $C_{\text{SO}_2,i}$ ), gas density ( $\rho_g$ ), liquid density ( $\rho_L$ ), gas viscosity ( $\mu_g$ ), liquid viscosity ( $\mu_L$ ), liquid surface tension ( $\sigma_L$ ), diffusivity of  $\text{SO}_2$  in air ( $D$ ), diffusivity of  $\text{SO}_2$  in water ( $D'$ ).
- geometrical parameters: droplet SMD ( $D_d$ ), column height ( $H_T$ ), column diameter ( $D_C$ );
- flow parameters: droplet velocity ( $v_d$ ), superficial gas velocity ( $v_g$ ), air velocity at atomizer entry ( $V_{Gi}$ ), superficial liquid velocity ( $v_L$ ), liquid velocity at atomizer entry ( $V_{Li}$ );

Dimensional analysis was carried out with the formation of relevant dimensionless groups leading to the following equation:

$$\eta_{\text{SO}_2} = f \left[ \frac{D_d v_d \rho_L}{\mu_L} \right]^a \times \left[ \frac{\rho_g (V_{Gi} - V_{Li})^2 D_d}{\sigma_L} \right]^b \times \left[ \frac{\mu_g}{(D' C_{\text{SO}_2,i})} \right]^c \times \left[ \frac{D}{(v_L H_T)} \right]^d \quad (6)$$

$$= f [Re_D]^a \times [We_D]^b \times [Sc]^c \times [D_i]^d \quad (7)$$

The inlet concentration of  $\text{SO}_2$  is expressed in  $\text{kg/m}^3$  for the purpose of the dimensional analysis. In order to establish the functional relationship between the percentage removal of  $\text{SO}_2$  from a mixture of air– $\text{SO}_2$ , and the various dimensionless groups in Eq. (7), multiple linear regression analysis has been used to evaluate the constant and coefficients of the equation. It can be seen that the following equation, which yields the minimum percentage error (0.907%) and the minimum standard deviation of percentage error (0.971%), presents the best possible correlation with a correlation coefficient of 0.9979

$$\eta_{\text{SO}_2} = 1 - 0.74557 [(Re_D)^{-0.1636} \times (We_D)^{-1.34853} \times (Sc)^{-0.349} \times (D_i)^{-0.77869}] \quad (8)$$

Eq. (8) describes the percentage removal of  $\text{SO}_2$  from an air borne  $\text{SO}_2$  stream in the spray tower, which is an important parameter for assessing the performance of the spray tower from the standpoint of air pollution control. The above equation is obtained for the modified droplet Reynold's number in the range 1160–3420, droplet Weber number in the range of 0.061–0.199, modified Schmidt number in the range of  $2.7 \times 10^6$  to  $11.5 \times 10^6$  and dispersion number in the range of  $5.96 \times 10^{-3}$  to  $33.3 \times 10^{-3}$ . The values of percentage removal of  $\text{SO}_2$  ( $\eta_{\text{SO}_2}$ ) predicted by Eq. (8) have been plotted against the experimental values of percentage removal of  $\text{SO}_2$  in Fig. 9. It can be seen from the figure that the predicted values fitted excellently well with the experimental values (well within  $\pm 10\%$  deviation). Furthermore, to test the acceptability of the correlation, various statistical tests ( $t$ -test and  $F$ -test) have been carried out and determined at 99.1% confidence level, which reveals that

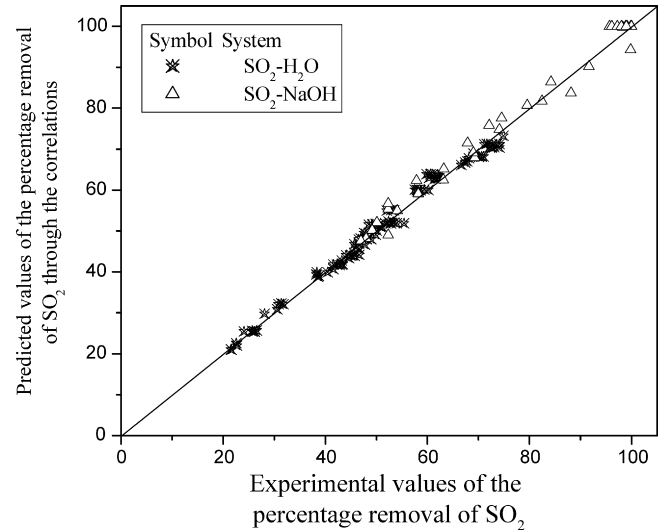


Fig. 9. Comparison of predicted and experimental percentage removal of  $\text{SO}_2$  in water and alkali scrubbing.

the correlation is highly functional. In our previous study [10] discussed earlier, the correlation was developed without considering the dimensionless droplet Weber number,  $We_D$ , and was unable to take in to account the intrinsic parameters that governed the atomization process for the purpose of predicting the performance of the critical flow atomizer in  $\text{SO}_2$  spray scrubbing.

### 5.3. Alkaline scrubbing of $\text{SO}_2$

In general, the overall rate of mass transfer influences the removal efficiency of a pollutant gas during its scrubbing using a liquid. In such a situation, the two-film model [17] can be used for studying the gas–liquid mass transfer operation having gas-side as well as liquid-side resistances [18]. Furthermore, the mass transfer in alkaline scrubbing is characterized by the enhancement factor, which is defined as follows:

mass transfer enhancement factor,  $\phi$

$$= \frac{\text{rate of mass transfer due to chemical reaction}}{\text{rate of mass transfer due to physical absorption}} \quad (9)$$

while the enhancement factor following the two-film model for  $\text{SO}_2$ – $\text{NaOH}$  system can be given by [19]:

$$\phi = \left( 1 + i \frac{D''}{D'} \frac{C''}{C_{\text{SO}_2,\text{int}}} \right) \quad (10)$$

The interfacial concentration of  $\text{SO}_2$  was obtained from the following relationship:

$$C_{\text{SO}_2,\text{int}} = H_\infty P_{\text{SO}_2} \quad (11)$$

where  $H_\infty$  is the thermodynamic equilibrium-distribution constant given by [20]:

$$H_\infty = \exp \left[ \left( \frac{32, 143.3}{T} \right) + 198.14 \ln T - 0.3384T - 1135.62 \right] \quad (12)$$

The removal efficiency in alkaline scrubbing can, therefore, be determined from the enhancement factor and the water scrubbing efficiency considering the fact that the ratio of mass transfer rates of  $\text{SO}_2$  with and without chemical reaction occurring inside a drop is proportional to the ratio of the overall spray tower removal efficiencies of  $\text{SO}_2$  with and without chemical reaction [15]. Thus the removal efficiency of  $\text{SO}_2$  in alkaline scrubbing in aqueous  $\text{NaOH}$  solution can be given by the following generalized equation:

$$\begin{aligned} &\text{alkaline removal efficiency of } \text{SO}_2 \\ &= (\text{removal efficiency of } \text{SO}_2 \text{ in water scrubbing}) \\ &\quad (\text{mass transfer enhancement factor}) \end{aligned}$$

$$\text{or, } \eta_C = \eta_{\text{SO}_2} \phi \quad (13)$$

Eq. (13) can be applicable to any gas–liquid absorption process in reactive systems based on the assumptions and conditions specified by the physico-chemical hydrodynamics. It is evident from Eq. (13) that the alkaline removal efficiency of any pollutant gas (for instance,  $\text{SO}_2$ ) is strongly dependent on the initial gas phase and the initial reagent concentrations. It can also be seen from Eq. (13) that the removal efficiency would decrease with the increase in the initial gas phase concentration while it would increase with the increase in the initial reagent concentration. The removal efficiency using Eq. (13) may sometimes yield values greater than 100% and in that case it should be considered 100%, since such a situation does not practically possible [10,14].

The behavior of the spraying device in alkaline scrubbing using dilute aqueous  $\text{NaOH}$  solution has been studied by carrying out a few experiments choosing the combinations of flow variables ( $Q_L$  and  $Q_G$ ) based on the following consideration [10]:

- Intermediate  $Q_L$ , amongst the range of liquid flow rates studied, which was  $1.83 \times 10^{-5} \text{ m}^3/\text{s}$ .
- Optimum  $Q_L/Q_G$  ratio, above which the variation of percentage removal of the scrubber in water scrubbing was insignificant, which was  $3.0 \text{ m}^3/1000 \text{ ACM}$ .
- $Q_G$  arrived, at using  $Q_L = 1.83 \times 10^{-5} \text{ m}^3/\text{s}$  and  $Q_L/Q_G$  ratio =  $3.0 \text{ m}^3/1000 \text{ ACM}$ , was  $6.20 \times 10^{-3} \text{ m}^3/\text{s}$ .

The impacts of different parameters studied on the performance of the spraying device are presented in this section within the framework of the present investigation.

### 5.3.1. Effect of scrubbing liquid pH and inlet concentration of $\text{SO}_2$

The effect of scrubbing liquid pH at the scrubber entry and inlet concentration of  $\text{SO}_2$  on the percentage removal is shown in Fig. 10. Expectedly, the figure indicates that the percentage removal increased with the increase in pH of the scrubbing liquid. It can also be seen from the figure that higher removal efficiency was obtained at lower initial concentration of  $\text{SO}_2$ , while it was necessary to increase the pH of the scrubbing liquid

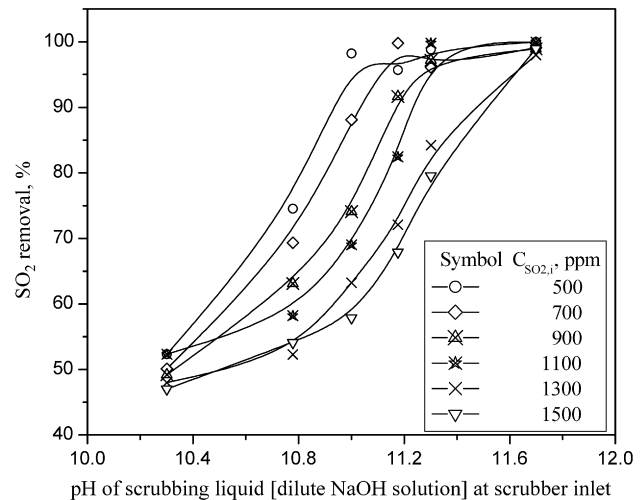


Fig. 10. Effect of scrubbing liquid pH at scrubber inlet on the percentage removal of  $\text{SO}_2$ .

to achieve higher removal efficiency at higher inlet  $\text{SO}_2$  concentration and this observation is in good agreement with Eq. (13) discussed earlier. Almost 100% removal of  $\text{SO}_2$  was achieved at a pH of 11.0 for inlet  $\text{SO}_2$  concentration of 500 ppm and at a pH of 11.7 for all initial concentrations of  $\text{SO}_2$ . In general, the spray tower performed well beyond pH of about 11.3. Hay et al. [7] reported removal efficiency of about 95%+ at a pH of 11.0 for inlet  $\text{SO}_2$  concentration of 500 ppm. On comparison of the present findings with the result reported by Hay et al. [7] indicates that the present system is efficiency wise much better than the existing system. Thus for a desired performance in alkali scrubbing, pH of the scrubbing liquid should be selected based on the initial  $\text{SO}_2$  concentration besides knowing the effects of other pertinent variables on water scrubbing.

### 5.3.2. Effect of ratio of initial concentration of $\text{NaOH}$ to interfacial concentration of $\text{SO}_2$ on enhancement factor of mass transfer and percentage removal of $\text{SO}_2$

The effect of concentration ratio on the experimental enhancement factor at  $Q_L = 1.83 \times 10^{-5} \text{ m}^3/\text{s}$ ,  $Q_G = 6.20 \times 10^{-3} \text{ m}^3/\text{s}$  and  $Q_L/Q_G$  ratio =  $3.0 \text{ m}^3/1000 \text{ ACM}$  is compared with theoretical values as shown in Fig. 11. The experimental values of the removal efficiencies are also shown in the same figure (see right vertical axis). It can be seen from the figure that the theoretical enhancement factor increased linearly and sharply with the increase in concentration ratio as expected from the linear nature of Eq. (9). The experimental values were also increased sharply with the concentration ratio up to a value of 2.0 kmol/kmol. In this range, the predicted values are found to be in excellent agreement with the experimental values. Beyond a concentration ratio of 2.0 kmol/kmol, a very striking feature was, however, observed from the figure. The enhancement factor theoretically increased with the concentration ratio linearly and indefinitely but the experimentally determined values did not follow the trend in the same manner. In the present case for  $\text{SO}_2$ – $\text{NaOH}$  system, the enhancement factor after reaching a maximum value of 2.10 at a concentration

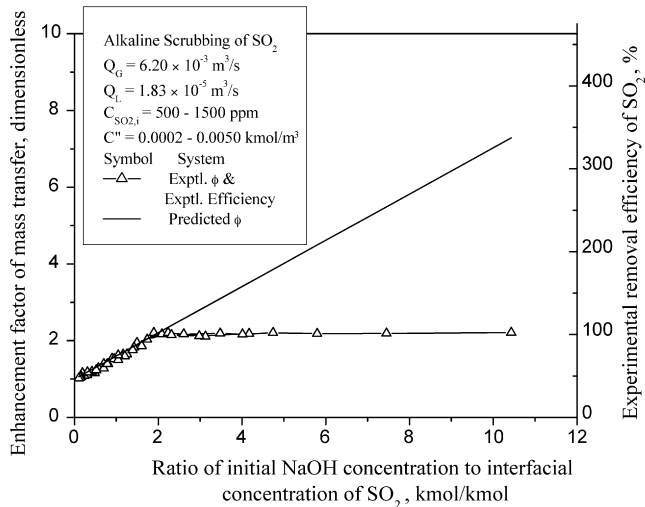


Fig. 11. Effect of concentration ratio on the mass transfer enhancement factor and on the percentage removal of  $\text{SO}_2$  in alkaline scrubbing at a  $Q_L/Q_G$  ratio of  $3.0 \text{ m}^3/1000 \text{ ACM}$ .

ratio of about 2.0 kmol/kmol, saturates and moves along the concentration ratio axis. It is attributed to the fact that the maximum enhancement could only be achieved at maximum removal efficiency of  $\sim 100\%$  as can be seen from the right vertical axis and beyond which there was no scope of any mass transfer and hence no scope of any enhancement of mass transfer further, which resulted in such behavior of the enhancement factor in alkaline scrubbing. Clearly, it demonstrates that the enhancement factor of mass transfer in alkaline scrubbing can never be expected to increase indefinitely with the increase in concentration ratio. This feature does not seem to be detailed in the available literature of gas–liquid mass transfer as well as gas scrubbing and is an extremely important finding for designing the scrubber operation. The application of enhancement factor reported in the present article may also be extended to other available gas–liquid mass transfer models.

It can also be seen from the figure that the percentage removal was increased with the increase in the concentration ratio. The maximum removal of  $\text{SO}_2$  obtained was almost 100% at a concentration ratio of 2.0 kmol/kmol in the studied spraying device within the framework of the investigation. Available literature [1] on the other hand, indicated that the removal efficiency of  $\text{SO}_2$  over 95% was readily attainable in a double alkali scrubbing process at a  $Q_L/Q_G$  ratio of  $4.35 \text{ m}^3/1000 \text{ ACM}$  [ $3.35 \text{ m}^3/1000 \text{ ACM}$  in the venturi (Stage-I) and  $1.0 \text{ m}^3/1000 \text{ ACM}$  in the tray/spray tower (Stage-II)].  $\text{SO}_2$  removal efficiency of around 90% was achieved by Pettersson et al. [4] using  $2.5\text{--}20 \text{ kmol/m}^3$  of NaOH solution and 95%+ by Hay et al. [7] using  $\sim 2.5 \text{ kmol/m}^3$  of NaOH solution. But in the present study maximum concentration of NaOH used was  $5 \times 10^{-3} \text{ kmol/m}^3$ . Clearly, the comparison of the performance of the present spraying device with the systems reported in the literature indicates that the present system had performed efficiently at significantly lower values of  $Q_L/Q_G$  ratio and NaOH concentration than the existing systems. Furthermore, the atomizer generated droplets at lower energy than the existing systems. Thus the system devel-

oped is efficiency wise and energy wise much better than the existing systems. Here is the novelty of the system developed.

### 5.3.3. Prediction of removal efficiency of $\text{SO}_2$ in alkaline scrubbing

The percentage removal of  $\text{SO}_2$  in alkaline scrubbing can be predicted for the spraying device with the help of Eqs. (8), (10) and (13). The final form can be given as:

$$\eta_C = \eta_{\text{SO}_2} \phi = [1 - 0.74557[(Re_D)^{-0.1636} \times (We_D)^{-1.34853} \times (Sc)^{-0.349} \times (Di)^{-0.77869}]] \times \left(1 + i \frac{D''}{D'} \frac{C''}{C_{\text{SO}_2, \text{int}}}\right) \quad (14)$$

Diffusivities of  $\text{SO}_2$  in water and NaOH were taken from previous workers [21,22] for the purpose of calculating the percentage removal. It can be seen from Eq. (14) that the percentage removal of  $\text{SO}_2$  would increase with the decrease in droplet SMD and with the increase in droplet velocity as discussed earlier. Thus either reducing the droplet SMD or increasing the droplet velocity would reduce the consumption of the reagent for  $\text{SO}_2$  scrubbing. The predicted values have been plotted against the experimental values of percentage removal of  $\text{SO}_2$  in Fig. 9. It can be seen from the figure that the predicted values fitted excellently well with the experimental values (well within  $\pm 10\%$  deviation).

## 6. Scrubber design

In view of a large volume of dilute gas to be scrubbed in industrial gas cleaning processes, equipment which reduces energy consumption to a reasonable magnitude are preferable, as in the long run this leads to a substantial savings in the operating cost. The present system developed is a kind of device that can satisfy this condition. In this section the aspects of design is presented. The critical process design parameters for a gas–liquid mass transfer device are true volumetric gas side mass transfer coefficient and number of transfer unit. In order to compare the performances of the present spraying device with the existing devices, reporting on these two parameters are necessary.

The true volumetric gas side mass transfer coefficient was found to vary between  $3 \times 10^{-3}$  and  $40 \times 10^{-3} \text{ kmol/m s atm}$  [10] as compared to  $0.0754 \times 10^{-3}$  and  $0.2557 \times 10^{-3} \text{ kmol/m s atm}$  reported by Mehta and Sharma [23,24] under similar flow regimes. The maximum value of number of transfer unit of 9 obtained [10] was very high than that reported by Schmidt and Stichlmair [2] ranging between 0.2 and 2.0 for tower height of 0.2–4.2 m, tower diameter of 0.025–0.390 m and liquid-to-gas flow rate ratio of 1.0–5.0  $\text{m}^3/1000 \text{ ACM}$ . Thus the system is better than the existing systems from the standpoint of intrinsic parameters of process design.

The sizing analysis as a part of the process design has been shown in a flow chart (Fig. 12) that takes into account the empirical and semi-empirical correlations. The desired performance level coupled with the available physico-chemical properties is the feed data into this flow chart. The desired performance can

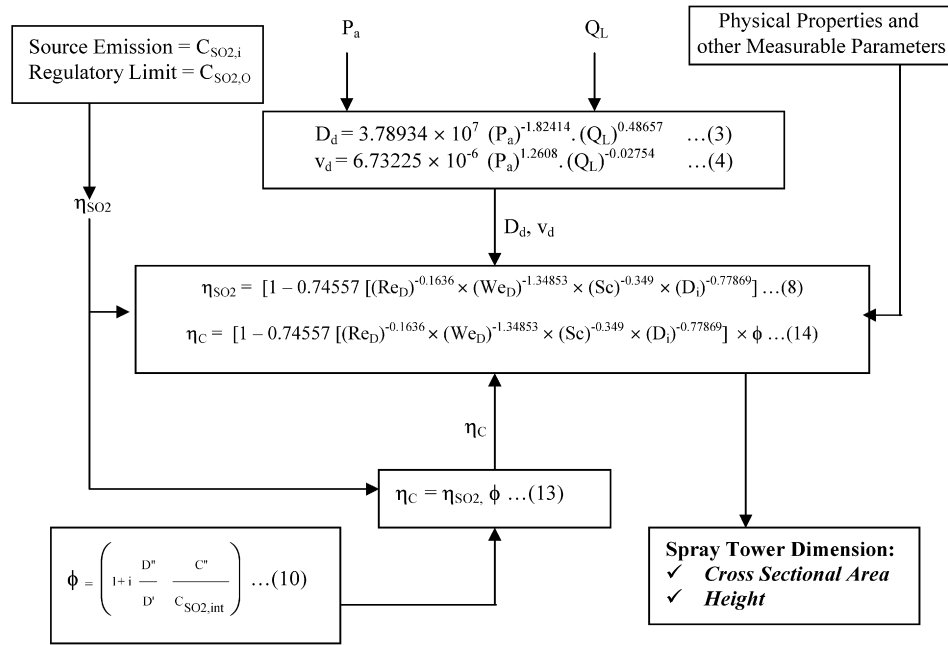


Fig. 12. Spray tower sizing analysis.

be obtained from the source emission and regulatory limit in tandem. Eq. (8) shall be rearranged as follows to generate data for the sizing of the spray tower for water scrubbing.

$$\begin{aligned} \eta_{SO_2} &= [1 - 0.74557 (Re_D)^{-0.1636} \times (We_D)^{-1.34853} \\ &\quad \times (Sc)^{-0.349} (D_i)^{-0.77869}] \dots (8) \\ &= [1 - 0.74557 [(D_d v_d \rho_L / \mu_L)^{-0.1636} \\ &\quad \times \{\rho_g (V_{Gi} - V_{Li})^2 D_d / \sigma_L\}^{-1.34853} \{\mu_g / D' C_{SO_2,i}\}^{-0.349} \\ &\quad \times \{D / v_L H_T\}^{-0.77869}] = f\{(D_d, v_d, C_{SO_2,i}, \rho_L, \rho_g, \mu_L, \\ &\quad \mu_g, \sigma_L, D', D, V_{Gi}, V_{Li}, Q_L)(D_T, H_T)\} \\ &[\text{substituting } v_L = 4 \times Q_L / (\pi D_T^2)] = f\{K, (D_T, H_T)\} \\ &\quad \times [\text{where, } K = D_d, v_d, C_{SO_2,i}, \rho_L, \rho_g, \mu_L, \mu_g, \sigma_L, D', \\ &\quad D, V_{Gi}, V_{Li}, Q_L] \end{aligned}$$

$K$  contains the pertinent parameters of the systems that are available. Therefore, knowing the desired performance level, cross-sectional area of the tower (hence the diameter) and the height of the tower can be determined using the correlation developed by a trial-error between the height and the diameter of the tower considering the fact that the ratio of height to diameter of the tower is ranging between 8.0 and 10.0 for gas scrubbing operations [2]. Similarly, Eq. (14) shall be rearranged to generate data for sizing the spray tower for alkaline scrubbing.

## 7. Conclusions

Abatement of SO<sub>2</sub> assumes significant importance due to its deleterious effects on living systems. Legion of SO<sub>2</sub> scrubbing technologies have evolved from the requirements of a number

of industries stemming from their definite process and regulatory mandates. The performance of an energy efficient critical flow atomizer deploying in a counter-current spray tower for SO<sub>2</sub> scrubbing using water and dilute NaOH was reported. The atomizer is capable of producing finer droplets at very high relative velocity without sacrificing the spray uniformity. Followings were the major findings of the present investigation:

### 7.1. Hydrodynamics of the spray tower

Preliminary hydrodynamic studies revealed that the droplet SMD was reduced with the increase in atomizing air pressure (absolute) and decrease in the liquid flow rate while the droplet velocity was increased with the increase in atomizing air pressure (absolute) and decrease in the liquid flow rate.

### 7.2. Performance in water and in alkaline scrubbing

Experimentation revealed that the SO<sub>2</sub> removal efficiency increased with the increase in liquid flow rate, liquid-to-gas flow rate ratio, atomizing air pressure and droplet velocity while it decreased with the increase in droplet SMD and gas flow rate. Inlet SO<sub>2</sub> concentration, however, did not have any significant effect on the SO<sub>2</sub> removal efficiency. Results also indicated that the percentage removal of SO<sub>2</sub> had almost reached a maximum value for a Q<sub>L</sub>/Q<sub>G</sub> ratio of 3.0 m<sup>3</sup>/1000 ACM and was considered as optimum for the scrubber operation. Results showed almost 100% removal efficiency at a Q<sub>L</sub>/Q<sub>G</sub> ratio of 3.0 m<sup>3</sup>/1000 ACM and at a scrubbing liquor pH of 11.7. Results also indicated that the present system is energy wise and efficiency wise much better than the existing systems.



### 7.3. Prediction of the performance

Critical appraisal of the available literature revealed that an exact comparison of the performance achieved in the present study with the available reported data is not possible. Therefore, empirical and semi-empirical correlations were developed for predicting the removal efficiencies of SO<sub>2</sub> in water and alkaline scrubbing respectively and the predicted values fitted excellently well with the experimental values. The water scrubbing efficiency and the mass transfer enhancement factor were combined while predicting the performance in alkaline scrubbing. It was further pointed out that the removal efficiency for any gas would inversely varied with initial gas phase concentration while directly varied with initial reagent concentration. It should be considered 100% in case it exceeds 100%. Experimentation coupled with theoretical analysis further revealed that the enhancement factor of mass transfer in alkaline scrubbing could never be expected to increase indefinitely with the increase in ratio of initial concentration of NaOH to the interfacial concentration of SO<sub>2</sub>. This feature does not seem to be detailed in the available literature of gas–liquid mass transfer as well as gas scrubbing and is an extremely important finding for designing the scrubber operation. The application of enhancement factor reported in the present article may also be extended to other available gas–liquid mass transfer models. Furthermore, the semi-empirical correlation developed for characterizing the performance of alkaline scrubbing of SO<sub>2</sub> revealed that either reducing the droplet SMD or increasing the droplet velocity would reduce the consumption of the reagent.

### 7.4. Process design and sizing analysis

Reportedly the values of true volumetric gas side mass transfer coefficient were very high and were better than the existing system. Knowing the desired performance level and physical properties pertinent to the system studied should be fed to the rearranged empirical and semi-empirical correlations developed so as to evaluate the tower sizing parameters, for instance, the tower cross-section (hence the tower diameter) and the tower height, in water and alkaline scrubbing respectively.

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