

Chemical Engineering Journal 131 (2007) 263–271

Chemical Engineering Journal

www.elsevier.com/locate/cej

# Modeling of removal of sulfur dioxide from flue gases in a horizontal cocurrent gas–liquid scrubber

S. Sarkar<sup>a</sup>, B.C. Meikap<sup>a,∗</sup>, S.G. Chatterjee<sup>b</sup>

<sup>a</sup> *Department of Chemical Engineering, Indian Institute of Technology (IIT), Kharagpur, Dist. Midnapur (West), West Bengal 721302, India*

<sup>b</sup> *Faculty of Paper Science and Engineering, SUNY College of Environmental Science & Forestry, University of New York, 1 Forestry Drive, Syracuse, NY 13210, USA*

Received 26 September 2006; received in revised form 31 October 2006; accepted 7 December 2006

#### **Abstract**

Removal of sulfur dioxide and its accurate prediction from the flue gases emitted from various chemical industries in a techno-econo-enviro manner is of great importance. The concentration of sulfur dioxide in and around Indian chemical process plants dealing with sulfur compounds overshoots the danger point. Prediction of SO2 removal efficiency is very important for the selection of pollution control equipment. In the present investigation, a residence time distribution (RTD) approach has been used for developing a theoretical model for predicting the  $SO_2$  removal efficiency in a horizontal co-current gas–liquid scrubber by water spray. Experimental result shows that a very high percentage removal of  $SO_2$ could be achieved from air–SO<sub>2</sub> mixture without using any additives or pre-treatment. Experimental results agreed excellently with developed theoretical model and it shows that almost 75–99% removal efficiency could be achieved by this approach. © 2006 Elsevier B.V. All rights reserved.

*Keywords:* Air pollution; Wet scrubber; Sulfur dioxide; Modeling; Horizontal scrubber; Flue gas

# **1. Introduction**

A review of the literature on the removal of  $SO<sub>2</sub>$  from an air stream by water spray reveals that both cocurrent and countercurrent techniques have been practiced for the removal of  $SO<sub>2</sub>$ in an air stream by Meikap et al. [\[1–3\].](#page-8-0) However, it has been found that very few studies have been made using the residence time distribution (RTD) approach in the recent past. Higbie [\[4\]](#page-8-0) proposed the penetration theory based on Fick's onedimensional unsteady diffusion for unsteady mass transfer at the gas–liquid interface. Fogler [\[5\]](#page-8-0) gave an interesting discussion on the residence time distribution of particles in a stream. Chatterjee and Tien [\[6\]](#page-8-0) discussed the effects of RTD of adsorbents in continuous-flow well-mixed tanks.

Huckaby and Ray [\[7\]](#page-8-0) reported studies on the absorption of SO2 into growing or evaporating droplets of water. Han and Park [\[8\]](#page-8-0) reported studies on the absorption of a single bubble of  $SO<sub>2</sub>$ into pure water. Richard and Gregory [\[9\]](#page-8-0) extensively studied the

 $SO<sub>2</sub>$  removal by in-duct dry scrubbing using calcium hydroxide from flue gas.

Glomba Michal [\[10\]](#page-8-0) reported the removal of  $SO<sub>2</sub>$  from boiler flue gases. Meikap et al.[\[1,11\]](#page-8-0) developed a horizontal co-current ejector system for scrubbing of  $SO<sub>2</sub>$  by using water and alkaline solution. Their experimental results indicate that  $SO<sub>2</sub>$  can be removed from lean gas mixture by water with a removal efficiency exceeding 98.62%. In addition, they reported that 100% removal of  $SO<sub>2</sub>$  could be achieved by alkaline solution from rich gas mixtures, by using alkaline scrubber.

Tsai et al. [\[12\]](#page-8-0) developed an experimental set-up for the removing sulfur dioxide gas from the make-up air of a semiconductor plant by fine water spray and Meikap et al. [\[11,13\]](#page-8-0) reported a method for removing sulfur dioxide and fly ashes from boiler flue gases. Dohman et al. [\[14\]](#page-8-0) reported a process for the removal of pollutants and trace impurities from flue gas, especially incinerator flue gas. De-dusting and wet scrubber removed pollutants.

Critical analysis of the literature revealed that very few studies on the removal of  $SO<sub>2</sub>$  from an air stream by fine water spray have been made using RTD consideration. Of the entering water droplets all will not have the same residence time, so while developing a model this fact have to be kept in mind.

<sup>∗</sup> Corresponding author. Tel.: +91 32 22283958; fax: +91 32 22282250. *E-mail addresses:* [bcmeikap@che.iitkgp.ernet.in](mailto:bcmeikap@che.iitkgp.ernet.in) (B.C. Meikap),

[schatterjee@esf.edu](mailto:schatterjee@esf.edu) (S.G. Chatterjee).

<sup>1385-8947/\$ –</sup> see front matter © 2006 Elsevier B.V. All rights reserved. doi[:10.1016/j.cej.2006.12.013](dx.doi.org/10.1016/j.cej.2006.12.013)

<span id="page-1-0"></span>



 $\beta$  *U*<sub>ter</sub>/*H*  $\gamma$  a function of the pH of the liquid  $\gamma$ m  $\frac{1}{(1/\gamma)-\beta(3d_p/6k_g)}$ δ 6*k*g/3*d*pγ<sup>m</sup>

- $\eta$  SO<sub>2</sub> removal efficiency of the scrubber
- $\phi$  overall efficiency of the scrubber for liquid drops

### *Subscripts and superscripts*



- + cation
- g,s gas phase just adjacent to the surface of a liquid drop
- l liquid phase just adjacent to the surface of a liquid drop (quantities in square brackets denote concentrations of the different species)  $(g \text{ mol/cm}^3)$

In the present investigation, therefore, an attempt has been made to develop a generalized theoretical model using RTD approach, for predicting the removal efficiency in a co-current gas–liquid scrubber with a view to attain definite insight into the process of absorption of  $SO_2$  in water. The proposed model takes into consideration, the concentration distribution of  $SO<sub>2</sub>$ , both in liquid phase and gas phase as a function of droplet diameter, pH level of the liquid, ppm level of the inlet gas, gas and liquid flow rates and height of the scrubber.

# **2. The theoretical model**

Consider the schematic of a horizontal co-current gas–liquid scrubber (Fig. 1) where an air stream containing a trace amount of  $SO_2$  enters horizontally at the left face  $(x=0)$  with a velocity  $U_{\text{gas}}$  (cm/s) along with a spray of water droplets whose number concentration at the inlet is  $N_{\text{in}}$  (g/cm<sup>3</sup>). SO<sub>2</sub> is progressively removed from the gas stream by absorption in the drops as both of them travel through the chamber from left to right. We will assume that any individual drop moves horizontally with a velocity equal to *U*gas and falls vertically (due to gravity) at its terminal settling velocity  $U_{\text{ter}}$  (cm/s). The SO<sub>2</sub> concentration of the inlet air stream is  $C_{g,in}$  (g mol/cm<sup>3</sup>) and the S(IV) (or total sulfur) concentration of the inlet water is  $C_{\text{lin}}$  (g mol/cm<sup>3</sup>). As the gas and liquid travel through the chamber, the  $SO<sub>2</sub>$  concentration of the air falls while the sulfur concentration of the liquid rises, and gas and liquid exit the chamber at  $x = L$  (cm) with concentrations of  $C_{g, \text{out}}$  and  $C_{1, \text{out}}$ , respectively. The problem is to predict these outlet concentrations as a function of operating conditions and chamber dimensions (*W* and *H* are the width and height of the chamber in cm) so that we can calculate the  $SO<sub>2</sub>$  removal efficiency of the scrubber.

The problem is complicated by the following phenomenon. Of the entering droplets a fraction will pass through the chamber



Fig. 1. Schematic diagram of horizontal gas–liquid scrubber.

<span id="page-2-0"></span>and leave at its exit. This fraction will absorb  $SO<sub>2</sub>$  (whose concentration in the air decreases with distance into the chamber, *x*, cm) during a residence time of  $t_c = L/U_{gas}$  (s) in the chamber. However, the other fraction will have a distribution of residence times in the chamber ranging from 0 to  $t_c$  since they will settle out (i.e., reach the floor of the scrubber) at various times as they pass through the chamber. Thus, drops in this fraction will be exposed to  $SO<sub>2</sub>$  for different times, and therefore, the residence time distribution (RTD) of this fraction has to be taken into account in the theoretical analysis.

#### **3. Mass balance of liquid drops**

Let  $Q_{\text{gas}}$  (assumed constant in the scrubber) and  $Q_{\text{liq,in}}$  be the inlet volumetric gas and liquid flow rates  $\text{cm}^3\text{/s}$ ). Also, let  $N_{\text{in}}$ ,  $N_x$ , and  $N_{x + \Delta x}$  be the number concentration of water drops (#/cm<sup>3</sup>) at the inlet ( $x = 0$ ) and at locations  $x$  and  $x + \Delta x$ , respectively. We will also assume mixed (turbulent) flow in chamber, i.e., there is complete mixing in the vertical direction while no mixing in the horizontal direction. Consider the slice or control volume of thickness  $\Delta x$ . There will be a certain height *h* along the vertical direction in this slice below which all drops will reach the floor of the chamber, i.e., all such drops will be collected. (We will assume that after a drop is collected, it no longer absorbs  $SO_2$ .) Denoting the residence time of the gas (and droplets) in this slice as  $\Delta t$ , we have

$$
\Delta t = \frac{h}{U_{\text{ter}}} = \frac{\Delta x}{U_{\text{gas}}} \tag{1}
$$

Let  $\Delta N$  be the number of drops per unit volume ( $g/cm<sup>3</sup>$ ) that settle in the slice; then the fraction of drops entering the slice that are collected (i.e., reach the floor of the scrubber) is given by

$$
\frac{WHU_{\text{gas}}(N_x - N_{x + \Delta x})}{WHU_{\text{gas}}N_x} = -\frac{\Delta N}{N_x} = \frac{h}{H} = \frac{U_{\text{ter}}}{HU_{\text{gas}}}\Delta x
$$
 (2)

which upon letting  $\Delta x \rightarrow 0$ , yields the mass balance of liquid drops in a differential form, i.e.,

$$
\frac{dN}{N} = -\frac{U_{\text{ter}}}{HU_{\text{gas}}} dx
$$
\n(3)

Integrating Eq.  $(3)$  from the inlet to any location *x* gives

$$
N = N_{\rm in} \exp\left(\frac{-U_{\rm ter}x}{HU_{\rm gas}}\right) \tag{4}
$$

Thus, the overall collection efficiency of the scrubber for liquid drops  $\phi$  is given by

$$
\phi = \frac{N_{\text{in}} - N_{\text{out}}}{N_{\text{in}}} = 1 - \exp\left(-\frac{U_{\text{ter}}L}{HU_{\text{gas}}}\right)
$$
(5)

where  $N_{\text{out}}$  (g/cm<sup>3</sup>) is the number concentration of liquid drops at the outlet  $(x = L)$  of the scrubber.

The above derivation for  $\phi$  has also been given by Nevers [\[15\].](#page-8-0) Since we have assumed that a liquid drop moves horizontally with a velocity equal to  $U_{\text{gas}}$ , the inlet liquid volumetric flow rate ( $Q_{\text{liq,in}}$ ) and gas volumetric flow rate ( $Q_{\text{gas}}$ ) in the scrubber are related by

$$
N_{\rm in} = \frac{Q_{\rm liq,in}}{Q_{\rm gas}(\pi d_{\rm p}^{\rm 3}/6)}\tag{6}
$$

where  $d_p$  is the diameter of a liquid droplet (cm).

#### **4. Mass balance of sulfur**

We make an overall mass balance (g/s) of sulfur species over the slice of length  $\Delta x$  shown in [Fig. 1](#page-1-0) and make use of Eq. (3) to get

$$
Q_{\text{gas}}C_{\text{g,x}} \times 32 + N_x Q_{\text{gas}} \frac{\pi d_p^3}{6} C_{1,x} \times 3 \times 32
$$
  
=  $Q_{\text{gas}}C_{\text{g,x}+\Delta x} \times 32 + N_{x+\Delta x} Q_{\text{gas}} \frac{\pi d_p^3}{6} C_{1,x+\Delta x} \times 3 \times 32$   
+  $N_x \frac{U_{\text{ter}}}{HU_{\text{gas}}} \Delta x Q_{\text{gas}} \frac{\pi d_p^3}{6} C_{1,x} \times 3 \times 32$  (7)

where  $C_g$  and  $C_l$  are the local concentrations of SO<sub>2</sub> and S(IV) in the gas and liquid phases  $(g \text{ mol/cm}^3)$ , respectively. The atomic weight of sulfur is 32, and there is 1 g-atom of sulfur per gmol of  $SO_2$  and 3 g-atoms of sulfur per g-mol of  $S(IV)$ —see Eq. [\(19\). I](#page-3-0)t has also been assumed in Eq. (7) that (a)  $SO_2$  dissolves in water and immediately transforms into S(IV) species and (b) the droplets that settle out in the slice have an average concentration of  $C_{1,x}$ . The second assumption becomes exact in the limit as  $\Delta x \rightarrow 0$  and using Eqs. (4) and (6), Eq. (7) becomes

$$
-\frac{dC_g}{dx} = \frac{3Q_{\text{liq,in}}}{Q_{\text{gas}}} \exp\left(-\frac{U_{\text{ter}}x}{HU_{\text{gas}}}\right) \frac{dC_1}{dx}
$$
(8)

which relates the gas-phase and liquid-phase concentration gradients. Integrating Eq. (8) from the inlet to any horizontal location *x* in the scrubber, yields

$$
C_{g}(t) = C_{g,in} - R\left(e^{-\beta t}C_{l}(t) - C_{l,in} + \beta \int_{0}^{t} e^{-\beta \lambda} C_{l}(\lambda) d\lambda\right)
$$
\n(9)

where

$$
R = \frac{3Q_{\text{liq,in}}}{Q_{\text{gas}}}
$$
 (10)

$$
t = \frac{x}{U_{\text{gas}}} \tag{11}
$$

$$
\beta = \frac{U_{\text{ter}}}{H} \tag{12}
$$

The SO<sub>2</sub> removal efficiency of the scrubber  $\eta$  is given by

$$
\eta = 1 - \frac{Q_{\text{gas}} C_{\text{g,out}}}{Q_{\text{gas}} C_{\text{g,in}}}
$$
  
= 
$$
\frac{R}{C_{\text{g,in}}} \left( e^{-\beta t_c} C_1(t_c) - C_{1,\text{in}} + \beta \int_0^{t_c} e^{-\beta \lambda} C_1(\lambda) d\lambda \right)
$$
 (13)

<span id="page-3-0"></span>where  $t_c$  is the residence time of the gas (and of the liquid drops that leave with the gas at the exit) in the scrubber given by

$$
t_{\rm c} = \frac{L}{U_{\rm gas}}\tag{14}
$$

From Eqs. [\(3\)](#page-2-0) and (4) it can be seen that the quantity  $\beta e^{-\beta \lambda} d\lambda$ in Eqs. [\(9\)](#page-2-0) and [\(13\)](#page-2-0) is the fraction of inlet droplets that have a residence time in the scrubber between  $\lambda$  and  $\lambda + d\lambda$  where  $\lambda < t_c$ .

#### **5. Determination of**  $C_1(t)$

Since  $SO<sub>2</sub>$  is present in trace amounts in the air stream and due to internal circulation inside the water drops, we will assume that the major resistance to mass transfer of  $SO<sub>2</sub>$  from gas to a liquid drop resides in the gas phase, i.e., the concentration of sulfur in the drop  $C_1$  (g mol/cm<sup>3</sup>) is uniform. The dissolution and subsequent dissociation of  $SO<sub>2</sub>$  in water (assumed to be instantaneous) are expressed by the relations Tsai et al. [\[12\]:](#page-8-0)

$$
SO2(g) + H2O \xrightarrow{k_1} SO2H2O
$$

$$
SO2H2O \xrightarrow{k_1} H+ + HSO3-
$$

 $HSO_3^- \xrightarrow{k_2} H^+ + SO_3^{2-}$ 

Here, *H* is Henry's law constant while  $k_1$  and  $k_2$  are dissociation constants (g mol/cm<sup>3</sup>), i.e.,

$$
H = \frac{[SO_2H_2O]_1}{[SO_2]_{g,s}}
$$
(15)

$$
k_1 = \frac{[H^+]_1[HSO_3^-]}{[SO_2H_2O]_1}
$$
\n(16)

$$
k_2 = \frac{[H^+]_1[SO_3^{2-}]}{[HSO_3^-]}_1\tag{17}
$$

Also

$$
C_{g,s} = [SO_2]_{g,s} \tag{18}
$$

$$
C_1 = [S(IV)]_1 = [SO_2H_2O]_1 + [HSO_3^-]_1 + [SO_3^{2-}]_1 \tag{19}
$$

The subscripts l and g,s denote the liquid phase and the gas phase just adjacent to the surface of a liquid drop while quantities in square brackets denote concentrations of the different species  $(g \text{ mol/cm}^3)$ . [Note that we have not considered the formation of S(VI) species earlier. since we assume that the water is free of hydrogen peroxide  $(H_2O_2)$ . If a portion of S(IV) transforms into S(VI) at a finite rate (see Eq. (14) in Huang's manuscript), this will complicate the analysis.]

From Eqs. (15)–(19), it can be easily shown that

$$
C_1 = \gamma C_{\rm g,s} \tag{20}
$$

where

$$
\gamma = \left(\frac{\left[H^{+}\right]_{1}^{2} + k_{1}\left[H^{+}\right]_{1} + k_{1}k_{2}}{\left[H^{+}\right]_{1}^{2}}\right)H
$$
\n(21)

From Eq. (21), it can be observed that  $\gamma$  is a function of the pH  $(=-\log_{10}[H^+])$  of the liquid and *H*,  $k_1$  and  $k_2$ . For simplicity, we will assume that the pH of the liquid is constant in the scrubber and equal to that of the entering liquid. Thus,  $\nu$  is a constant assuming isothermal conditions in the scrubber.

As mentioned earlier, we will assume that the mass transfer resistance resides only in the gas phase (i.e., concentration of S(IV) in a liquid drop is uniform). Assuming further that the transfer of  $SO<sub>2</sub>$  from the gas phase to a liquid droplet occurs over its entire surface area  $(\pi d_p^2)$  and making a liquid-phase mass balance (g/s) for sulfur over the slice of length  $\Delta x$  in [Fig. 1,](#page-1-0) it can be shown that

$$
\frac{d}{dt}(NC_1) = \frac{6k_g}{3d_p}(C_g - C_{g,s})N
$$
\n(22)

where  $k_g$  is the gas-phase mass transfer coefficient (cm/s) and  $d_p$  is the droplet diameter (cm). The factor 3 is the ratio of the number of g-atoms of sulfur per g-mol of S(IV) to that per g-mol of SO<sub>2</sub>. *N* is given by Eq. [\(4\)](#page-2-0) with  $x = U_{\text{gas}}t$  from Eq. [\(11\). \[](#page-2-0)Note that if there was no settling of the drops in the chamber then *N* would be a constant, and Eq. (22) reduces to the familiar form.] The initial condition for Eq. (22) is

$$
C_l = C_{l, \text{in}}, \quad \text{at } t = 0 \tag{23}
$$

Utilizing Eqs.  $(4)$ ,  $(9)$ ,  $(11)$  and  $(20)$ , Eq.  $(22)$  transforms into

$$
\frac{dC_1}{dt} = \frac{6k_g}{3d_p} \left[ C_{g,\text{in}} + RC_{1,\text{in}} - \left( \frac{1}{\gamma_m} + Re^{-\beta t} \right) C_1 - R\beta \int_0^t e^{-\beta \lambda} C_1(\lambda) d\lambda \right]
$$
\n(24)

The solution of Eq. (24) subject to Eq. (23) can be obtained via the Laplace transformation. It is given by

$$
C_1(t) = C_{1,\text{in}} e^{-\delta t} + \gamma_{\text{m}} (C_{g,\text{in}} + RC_{1,\text{in}})(1 - e^{-\delta t})
$$

$$
- R \int_0^t \left[ \left( 1 - \frac{\beta}{\delta} \right) e^{-\delta(t - \lambda)} + \frac{\beta}{\delta} \right] e^{-\beta \lambda} C_1(\lambda) d\lambda
$$
(25)

where

$$
\gamma_{\rm m} = \frac{1}{(1/\gamma) - \beta(3d_{\rm p}/6k_{\rm g})}
$$
 (26a)

$$
\delta = \frac{6k_{\rm g}}{3d_{\rm p}\gamma_{\rm m}}\tag{26b}
$$

Eq. (25) is an integral equation, which can be solved for  $C_1(t)$ by the numerical procedure outlined below. The gas-phase mass transfer coefficient  $k_g$  (with any necessary corrections to it) and the terminal velocity of a liquid drop  $U_{\text{ter}}$  can be obtained from appropriate correlations.

#### **6. Numerical solution**

The integral in Eq. (25) can be approximated by a procedure suggested by Chatterjee and Tien [\[6\].](#page-8-0)

Let

$$
I_{\rm A} = \int_0^t e^{(\delta - \beta)\lambda} C_{\rm I}(\lambda) d\lambda \tag{27}
$$

$$
I_{\rm B} = \int_0^t e^{-\beta \lambda} C_{\rm I}(\lambda) d\lambda \tag{28}
$$

We wish to find the values of these integrals at a time  $t_n$  $(n=1, 2, 3,$  etc., denotes the time level) given their values at the previous time  $t_{n-1}$ . Denoting the time step by  $\Delta t$ , we then have

$$
t_n = t_{n-1} + \Delta t \tag{29}
$$

Eqs. (27) and (28) can be approximated as

$$
I_{A}(t_{n}) = \int_{0}^{t_{n}} e^{(\delta - \beta)\lambda} C_{1}(\lambda) d\lambda
$$
  
\n
$$
= \int_{0}^{t_{n-1}} e^{(\delta - \beta)\lambda} C_{1}(\lambda) d\lambda + \int_{t_{n-1}}^{t_{n}} e^{(\delta - \beta)\lambda} C_{1}(\lambda) d\lambda
$$
  
\n
$$
= I_{A}(t_{n-1}) + \frac{C_{1}(t_{n-1}) + C_{1}(t_{n})}{2(\delta - \beta)} (e^{(\delta - \beta)t_{n}} - e^{(\delta - \beta)t_{n-1}})
$$
\n(30)

$$
I_{\mathcal{B}}(t_n) = \int_0^{t_n} e^{-\beta \lambda} C_1(\lambda) d\lambda
$$
  
=  $\int_0^{t_{n-1}} e^{-\beta \lambda} C_1(\lambda) d\lambda + \int_{t_{n-1}}^{t_n} e^{-\beta \lambda} C_1(\lambda) d\lambda$   
=  $I_{\mathcal{B}}(t_{n-1}) + \frac{C_1(t_{n-1}) + C_1(t_n)}{2\beta} (e^{-\beta t_{n-1}} - e^{-\beta t_n})$  (31)

Substituting Eqs. (30) and (31) into Eq. [\(25\)](#page-3-0) yields

$$
C_1(t_n) = \frac{f_1 - f_2 - f_3 C_1(t_{n-1})}{1 + f_3} \tag{32}
$$

where

$$
f_1 = C_{1, \text{in}} e^{-\delta t_n} + \gamma_m (C_{g, \text{in}} + RC_{1, \text{in}}) (1 - e^{-\delta t_n})
$$
 (33)

$$
f_2 = R\left[\left(1 - \frac{\beta}{\delta}\right) e^{-\delta t_n} I_{A}(t_{n-1}) + \frac{\beta}{\delta} I_{B}(t_{n-1})\right]
$$
(34)

$$
f_3 = \frac{R}{2\delta} [e^{-\delta t_n} (e^{(\delta - \beta)t_n} - e^{(\delta - \beta)t_{n-1}}) + (e^{-\beta t_{n-1}} - e^{-\beta t_n})] \quad (35)
$$

Eq. (32) can be used to calculate the value of  $C_1$  at time  $t_n$ given its value at time  $t_{n-1}$ . To start the calculation process, we note that

$$
C_1(t_0) = C_{1, \text{in}}, \quad I_A(t_0) = 0, \quad I_B(t_0) = 0; \quad t_0 = 0
$$
 (36)

The calculations for  $C_1(t)$  are to be continued until  $t = t_c$  (residence time of gas in scrubber). The gas phase concentration profile of  $SO_2$  in the scrubber can then be determined from Eqs. [\(9\),](#page-2-0) [\(11\)](#page-2-0) and (31). The exit gas concentration of SO<sub>2</sub>  $C_{\text{g,out}}$  [ $C_{\text{g}}$  $(t = t_c)$ ] and exit liquid concentration of S(IV)  $C_{\text{Lout}}$  [ $C_1$  ( $t = t_c$ )] from the scrubber can thus be found. The  $SO<sub>2</sub>$  removal efficiency of the scrubber  $\eta$  can be calculated from Eqs. [\(13\)](#page-2-0) and (31).

#### **7. Experimental technique**

Experiments have been conducted by passing an air stream containing a trace amount of  $SO_2$  horizontally along with a spray of water droplets.  $SO_2$  is progressively removed from the gas stream by absorption in the drops as both of them travel through the chamber and elaborately discussed elsewhere by Meikap et al. [\[2\].](#page-8-0) The concentration of  $SO<sub>2</sub>$  was measured by using this model; the outlet concentration is predicted as a function of operating conditions and chamber dimensions from which we can calculate the  $SO_2$  removal efficiency of the scrubber. The  $SO_2$  in gas phase were analyzed by the "Tetrachloro Mercurate Method" (IS: 5182 (Part-VI) [\[12\]\).](#page-8-0) The method consisted of passing a portion of the air– $SO_2$  sample, through a solution of absorbing medium (sodium tetrachloro mercurate) and analyzing the resulting solution spectrophotometrically (UV–visible recording spectrophotometer, Model No. UV-2100, Shimadzu, Japan). The concentration in liquid phase was estimated by volumetric titration method.

# **8. Results and discussions**

In the scrubbing experiments, studies were conducted to determine the effect of gas and liquid flow rates, pH of the inlet liquid, ppm level of the inlet air, height of the scrubber and diameter of the water droplets on the SO<sub>2</sub> removal efficiency.

#### *8.1. Effect of gas flow rate and SO2 loading on the percentage removal of SO<sub>2</sub>*

The percentage removal efficiency of  $SO<sub>2</sub>$  at different inlet SO2 loading and for constant height of the scrubber, have been plotted against gas flow rates and presented in Fig. 2. It can be seen that as the gas flow rate increases the removal efficiency decreases, and so is the case with increase in inlet  $SO<sub>2</sub>$ loading. This fact can be explained by the consideration that as

Fig. 2. Effect of gas flow rate on efficiency.





Fig. 3. Effect of liquid flow rate on efficiency at various concentration level.

the gas flow rate increases the time of contact between the gas and the liquid droplets become shorter, and this decreases the efficiency.

# *8.2. Effect of liquid flow rate and SO2 loading on the percentage removal of SO2*

The effect of liquid flow rate on the percentage removal efficiency is presented in Fig. 3 at various inlet  $SO<sub>2</sub>$  concentrations and for constant gas flow rates. It can be seen from the figure that the removal efficiency increases as the liquid flow rate increases. In the present investigation, as the liquid flow rate is increased. the bubble–water interfacial contact area increases. As a result of this, the percentage removal increases with increase in liquid flow rate.

# *8.3. Effect of scrubber height on outlet loading and removal efficiency of SO2*

The effect of height (height refers to the distance from the inlet of the scrubber to the outlet, along the flow) on outlet concentrations and efficiency has been shown in Figs. 4–6. As the height of the scrubber increases, the outlet concentration of  $SO<sub>2</sub>$ in the gas phase decreases but the outlet concentration of  $SO<sub>2</sub>$ in the liquid phase increases. As the tower height increases, the bubble–water interfacial contact area increases and so is the time of contact, this causes the gas phase concentration to decrease and the liquid phase concentration to increase, enhancing the SO2 removal efficiency.

# *8.4. Effect of pH of the inlet liquid on outlet loading and efficiency of the scrubber*

[Figs. 7 and 8](#page-6-0) show the effect of pH on the efficiency and outlet loading of SO2. As the pH of the inlet liquid increases the removal efficiency increases and the outlet loading decreases. This means that the efficiency increases as the liquid becomes



Fig. 4. Effect of scrubber height on concentration at various ppm level.



Fig. 5. Effect of scrubber height on efficiency.



Fig. 6. Effect of tower height on liquid phase concentration.

<span id="page-6-0"></span>

Fig. 7. Effect of pH on efficiency of scrubber.

more basic. This is due to the affinity of the acidic  $SO<sub>2</sub>$  towards a basic solution.

# *8.5. Effect of droplet diameter on efficiency and outlet loading of SO2*

The effect of droplet diameter has been shown in Figs. 9 and 10. As the diameter of the liquid droplets increases, the gas phase concentration increases while the percentage removal efficiency decreases. This can be attributed to the fact that with finer droplets we will have more bubble–liquid interfacial contact area. This in turn enhances the mass transfer with finer water droplets. Hence as the droplet diameter becomes smaller we will have better removal efficiency.

# *8.6. Effect of time*

The gas phase concentration and the liquid phase concentration at any instant has been plotted as a function of time in



Fig. 8. Effect of pH on gas phase outlet concentration.



Fig. 9. Effect of droplet diameter on gas phase efficiency.



Fig. 10. Effect of droplet diameter on gas phase outlet concentration.



Fig. 11. Effect of time on liquid concentration at various pH level.



Fig. 12. Effect of time on gas phase concentration at various pH.



Fig. 13. Plot of gas concentration vs. time at different ppm level.



Fig. 14. Plot of efficiency vs. time at different ppm level.



Fig. 15. Effect of time on removal efficiency at various inlet concentration.



Fig. 16. Variation of percentage removal efficiency for different sample numbers.

[Figs. 11–15](#page-6-0) under various operating conditions. The efficiency is also plotted as a function of time at different inlet loading. It can be easily inferred that as time increases the gas–liquid contact time increases and this increases the liquid phase concentration and decreases the gas phase concentration, and hence the efficiency is also increased.

#### **9. Sample number**

In Fig. 16, the percentage removal efficiency has been plotted for different sample numbers. From the figure, it can be said that the efficiency lies in the range of 60–97%. But this may vary depending on the operating conditions.

# **10. Conclusions**

A simple but realistic model has been developed for the absorption of  $SO_2$  in a co-current horizontal gas-liquid scrubber. <span id="page-8-0"></span>Experimental results show that the theoretical equations based on physical mass transfer predict very closely the performance of the scrubber for scrubbing of  $SO<sub>2</sub>$  using water as the scrubbing medium. Removal efficiency is found to be a function of inlet  $SO_2$  loading, the air flow rate, the liquid flow rate, the pH of the inlet liquid, diameter of water droplets, and height of the tower. Experimental investigation shows that a very high percentage removal of  $SO<sub>2</sub>$  can be achieved from air– $SO<sub>2</sub>$  mixture without using any additives or pre-treatment. Results also indicate that a higher liquid flow rate results in higher  $SO<sub>2</sub>$  removal efficiency whereas a higher value of the gas flow rate decreases the removal efficiency. Thus, the present model fits very well for scrubbing of  $SO<sub>2</sub>$ , which is experimentally verified and found excellent agreement predicted from model.

#### **References**

- [1] B.C. Meikap, G. Kundu, M.N. Biswas, Modeling of a novel multi-stage bubble column scrubber for flue gas de-sulfurization, Chem. Eng. J. 86 (2002) 331–343.
- [2] B.C. Meikap, S. Satyanarayan, A. Nag, M.N. Biswas, Scrubbing of sulfur dioxide from waste gas stream by horizontal co-current flow ejector system, Indian J. Environ. Protect. 19 (1999) 523–532.
- [3] B.C. Meikap, G. Kundu, M.N. Biswas, Prediction of the interfacial area of contact in a variable area multi-stage bubble column, Ind. Eng. Chem. Res. 40 (2001) 6194–6199.
- [4] R. Higbie, Rate of absorption of a pure gas into a still liquid during short periods of exposure, Trans AIChE J. 31 (1935) 365–373.
- [5] H.S. Fogler, Elements of Chemical Reaction Engineering, Prentice Hall of India, 1992.
- [6] S.G. Chatterjee, C. Tien, Adsorption in continuous-flow well-mixed tanks—the effect of residence time distribution of adsorbents, Sep. Technol. 1 (1991) 79–86.
- [7] J.L. Huckaby, A.K. Ray, Absorption of sulfur dioxide by growing and evaporating water droplets, Chem. Eng. Sci. 44 (1989) 2797–2805.
- [8] S.B. Han, P.W. Park, Absorption from a rising bubble of sulfur dioxide in pure water, Int. Chem. Eng. 30 (1990) 308–341.
- [9] W.R. Richard, A.B. Gregory, Flue gas desulphurization by in-duct dry scrubbing using calcium hydroxide, AIChE J. 36 (1990) 473– 478.
- [10] Glomba Michal, Method for removing sulfur dioxide and fly ashes from boiler flue gases, Polish Patent PL-171523 (1997).
- [11] B.C. Meikap, G. Kundu, N.M. Biswas, A novel modified multi-stage bubble column scrubber for  $SO_2$  removal from industrial off gases, Sep. Sci. Technol. 37 (2002) 3421–3442.
- [12] C.J. Tsai, C.H. Huang, H.H. Lu, Removal of air pollutants in the make-up air of a semiconductor plant by fine water spray, Sep. Sci. Technol. 38 (2003) 1429–1434.
- [13] B.C. Meikap, G. Kundu, M.N. Biswas, Scrubbing of fly-ash laden SO<sub>2</sub> in a modified multi-stage bubble column scrubber, AIChE J. 48 (2002) 2074–2083.
- [14] J. Dohman, F.A. Mian, M. Iqbal, K. Keldenich, Process for removal of pollutants and trace impurities from flue gas, US Patent (1999).
- [15] N. Nevers, Air Pollution Control Engineering, McGraw-Hill, Inc., New York, 1995.