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Experimental and Modeling Study on CO_2 Absorption in a Cyclone Scrubber by Phenomenological Model and Neural Networks

Mariana, Ken-ichi Sumida, Toshihoko Satake, Akinori Maezawa*, Takenari Takeshita* and Shigeo Uchida*,†

Graduate School of Science and Engineering, *Department of Materials Science and Chemical Engineering,
Shizuoka University, 3-5-1 Johoku, Hamamatsu 432-8561, Japan
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Abstract−Experimental and modeling studies have been conducted on CO_2 absorption in a cyclone scrubber operated at room temperature. The effects of parameters such as the initial concentration of alkali in the solution and the liquid - gas ratio on the CO_2 absorbed flux were experimentally and theoretically investigated. A phenomenological model and three-layer feed-forward neural networks have been applied to estimate the CO_2 absorbed flux in the cyclone scrubber. It was shown that the neural networks' values agreed well with the experimental data, while the values by phenomenological model partly agreed with the experimental data around the initial concentration of alkali in the solution, C_{Bo}≤0.001 kmol/m^3 (pH≤11).

Key words: Gas Absorption, Cyclone Scrubber, Phenomenological Models, Neural Networks

INTRODUCTION

The development of a low cost process for the removal of acid gases and dusts in flue gases of incinerators is desirable. A cyclone scrubber is considered to be one of the processes that can absorb gases, separate particles, and decrease the gas temperature simultaneously. It is very important to understand the mechanism of gas absorption and particle separation in a cyclone scrubber for the proper design and the optimum operation of the process. Despite the relatively simple design and the broad use of these types of scrubbers, the fluid dynamics are quite complex and give rise to rather complicated problems. Consequently, its modeling is a complex task since a system of nonlinear differential equations with many transport and chemical parameters must be solved.

A few phenomenological models for cyclones have been developed to understand the mechanism of particle separation and gas absorption in the cyclone [Johnstone and Silcox, 1947; Schrauwen and Thoenes, 1988; Mothes and Loffler, 1988; Patterson and Munz, 1996]. The phenomenological models of gas absorption with chemical reaction in various types of absorbers have also been reported in many studies [Uchida and Wen, 1973; Row and Lee, 1984; Asai et al., 1997; Park et al., 1999, 2002; Oh et al., 1999]. However, due to the complexity of the process in the cyclone scrubber system, it is very difficult to obtain accurate phenomenological models. Even if such models are obtained, they may be highly complicated and require simplifying assumptions for their solution.

Another method for practical process modeling is the black box approach, where models are obtained exclusively from experimental plant data. Such models do not provide a detailed knowledge of the underlying physics of the problem, but they do provide a description of the dynamic relationship between input and output variables. The statistical model based on the regression analysis is an example of such a black box approach that commonly relies on linear system identification models. Unfortunately, the majority of processes found in the chemical industries are non-linear and the performances of the linear models cannot be adequate in those cases.

The neural network has been found in numerous applications in representing the non-linear functional relationship among variables and has been applied in several complex chemical engineering processes [Galvan et al., 1996; Chouai et al., 2001; Himmelblau, 2000; Sohn et al., 1999; Roj and Wilk, 1998]. Parisi and Labored [2001] used three layer feed-forward neural networks to evaluate global reaction rate for heterogeneous gas-solid reactors and compared the results with the results from physical model. Iliuta and Lavric [1999] used the neural network for studying hydrodynamic parameters in a two-phase flow fixed-bed reactor and compared the results with the results from reported correlations. Although the neural network has been applied in several complex chemical engineering processes, it has not yet been applied in the cyclone scrubber system.

In the present work, the phenomenological model used for simulating a cyclone scrubber system at room temperature is described by physical principles. In order to calculate overall CO_2 absorbed flux in this system, a mass balance inside the liquid droplets and liquid film on the cyclone wall must be performed. This procedure may be time consuming. A model using three-layer feed-forward neural networks (3-FFNN) to estimate the overall CO_2 absorbed flux is proposed. Both methods are applied, compared and discussed to the cyclone scrubber system.

EXPERIMENTAL METHOD

The experimental apparatus is shown in Fig. 1. The cyclone consists of a cylindrical section with 11.6 cm in height joined to a conical section with 26.1 cm in height. Other dimensions are 5.9 cm in cyclone diameter, 2.7 cm in outlet duct diameter, 1 cm in apex diameter, 6.8 cm of gas outlet duct height, 1.4 cm in inlet duct width, 2.9 cm in inlet duct height, and 10 cm in inlet duct length. The liquid is injected by spraying through the nozzle (Full Cone Spray Noz-

†To whom correspondence should be addressed.
E-mail: tcsuchi@ipc.shizuoka.ac.jp

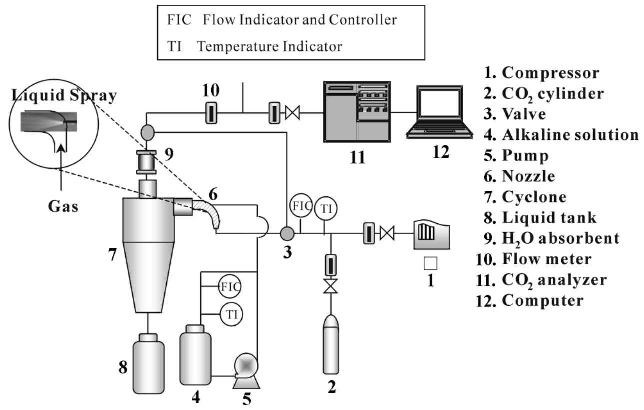

Fig. 1. Experimental apparatus.

zle, orifice diameter of 1 mm, pressure of 2 kg/m^2, and spray angle of 70^o, Ikeuchi, Co. Japan) into the gas stream in the inlet duct of the cyclone. The outlet concentration of CO_2 was continuously monitored by a CO_2 analyzer (URA-107 Shimadzu, Co. Japan). The experimental conditions are as follows: gas flow rate=5±0.2 liter/min, liquid rate=0.35-0.8 liter/min; liquid and gas temperatures=292±1 K; inlet CO_2 concentration=17±0.1%; and initial concentration of alkali in solution, $C_{B0}=1\times10^{-8}$-0.1 kmol/m^3.

PHENOMENOLOGICAL MODEL

The absorption in the cyclone occurs in two places: in the inlet duct of the cyclone and in the cyclone itself. To simplify the model, several assumptions are made as follows. (a) The liquid droplet to be sprayed is assumed to be spherical shape; (b) The reaction is instantaneous; (c) All the physical properties of gas and liquid are constant throughout; (d) When the absorption occurs at room temperature, the liquid and gas temperatures are constant during the absorption and there is no evaporation of liquid to the gas phase; and (e) The gas velocity in the cyclone was determined by the assumption of Mothes and Loffler [1988] and Paterson and Munz [1996].

1. Absorption in Inlet Duct

The equation of motion of a liquid droplet derived by Uchida and Wen [1973] in a venturi scrubber is applied to the present case in the inlet duct of the cyclone.

$$\frac{dv_d}{dz}=\frac{3}{4}\frac{C_D\rho_G}{d_P\rho_L}\frac{(v_G-v_d)|v_G-v_d|}{v_d} \tag{1}$$

The relationships for mass balance between gas and liquid in the inlet duct are used to derive the following set of differential equations:

$$\frac{dC_A}{dz}=\left[\frac{a\rho_m k_L}{L_m/S}\right]\Delta C_A \tag{2}$$

$$\frac{dP_A}{dz}=-\left[\frac{aPk_L}{G_m/S}\right]\Delta C_A \tag{3}$$

$$\frac{dt_i}{dz}=\frac{1}{v_d} \tag{4}$$

This set of differential equations is numerically solved by using given initial conditions at the nozzle point.

The liquid-phase mass transfer coefficient with an instantaneous reaction is given as follows if both the diffusivity of the gas and that of the reactant in the liquid are nearly the same [Brunson and Welek, 1970]:

$$k_L=[1+(C_{B0}/C_{Ai})]k_{LP} \tag{5}$$

where k_{LP} is the mass transfer coefficient for the physical absorption into the droplet, C_{Ai} is the interfacial concentration of CO_2 and can be defined as [Uchida and Wen, 1973]

$$k_{LP}=\frac{2D_A}{r_0}\sum_{m=1}^{\infty}\exp\left(-\frac{D_A m^2\pi^2 t_i}{r_o^2}\right) \tag{6}$$

$$C_{Ai}=(C_A^*-C_{BO}R_{g-L})/(1+R_{g-L}) \tag{7}$$

where, $R_{g-L}=\frac{k_{LP}H}{k_G}$, $C_A^*=HP_A$

The solubility, H, and the diffusivity of CO_2 in alkaline solution, D_A, are estimated by using the methods presented by Schumpe [1993] and Hikita et al. [1976], respectively. The gas-phase mass transfer coefficient of an individual droplet is calculated by Steinberger and Treybal's correlation [1960]:

$$N_{Sh}=2+0.347(N_{Re}N_{Sc}^{1/2})^{0.62} \tag{8}$$

where $N_{Sh}=\frac{RT_G k_G}{D_{AG}}$, $N_{Re}=\frac{d_p v_s\rho_G}{\mu_G}$, $N_{Sc}=\frac{\mu_G}{\rho_G D_{AG}}$

The applicable ranges of this correlation are $1<N_{Re}<30{,}000$ and $0.6<N_{Sc}<30{,}000$, which are always satisfied in the experimental conditions investigated here.

Mass mean diameter of droplet, d_p, is estimated by Kim and Marshall's correlation [1971] and the slip velocity, v_s, is given by $v_s=v_G-v_d$.

2. Absorption in Cyclone

The mass transfer in the cyclone scrubber occurs in two places: in the liquid droplets and in the liquid film on the cyclone wall [Johnstone and Silcox, 1947]. By assuming that the absorption by the liquid phase is predominant in the cyclone, the mass transfer in the cyclone can be expressed as

$$\int_{P_{A1}}^{P_{A2}}\frac{dP_A}{P^*-P_A}=\frac{PH}{SG_{mf}}(k_LA+k_{Lw}A_w) \tag{9}$$

The integral represents the total number of transfer units and the terms on the right represent the number of transfer units resulting from the absorption in the liquid droplets and in the liquid film on the cyclone wall, respectively.

The liquid-phase mass transfer coefficient with an instantaneous reaction, k_L, into the droplet is estimated by using the same correlations as for the inlet duct [Eq. (5)]. The effective value of k_LA (cm^3/s) is estimated from the value of k_L into the droplet and the interfacial area of droplets, A. The interfacial area of droplets is the product of the number of droplets moving in the cyclone and the droplet surface area. The number of droplets is determined from the liquid supply, the droplet volume and the flight time. The gas-phase mass transfer coefficient around the droplets, k_G, in the cyclone is estimated from the average value of Sherwood numbers over the velocity calculated at each position in the cyclone which is calculated according to Steinberger and Treybal's correlation [1960]. Analogous with slip velocity in the inlet duct, slip velocity in the cyclone was defined by

$$v_s = v_{G(resultant)} - v_{d(resultant)} \tag{10}$$

where $v_{G(resultant)} = \sqrt{v_{rG}^2 + v_{\theta G}^2 + v_{zG}^2}$, $v_{d(resultant)} = \sqrt{v_{rd}^2 + v_{\theta d}^2 + v_{zd}^2}$

The gas velocities in the cyclone for the cylindrical section are modeled according to (cylindrical coordinates) [Patterson and Munz, 1996]:

$$v_{rG}(r_c) = 0, \quad v_{rG}(r_e) = \frac{G}{2\pi r_e (h_t - s)} \tag{11}$$

$$v_{zG} = \frac{G(h_t - h)}{\pi (r_c^2 - r_e^2)(h_t - s)} \tag{12}$$

$$v_{\theta G} = \frac{v_{\theta w}}{\frac{r}{r_c}\left[1 + D\left(1 - \frac{r}{r_c}\right)\right]} \tag{13}$$

Eqs. (11) to (13) can also be used for conical section by changing the cylindrical section radius, r_c, with the conical section radius, r_c^*. The conical section radius is a function of the conical height as follows [Mothes and Loffler, 1988]:

$$r_c^* = \sqrt{\frac{V_{con.}}{\pi z_{con.}}} \tag{14}$$

The liquid drops trajectories are estimated by using differential equations of force balances with cylindrical coordinates [Schrauwen and Thoenes, 1988].

$$\rho_L V_d \frac{dv_{rd}}{dt_c} = C_D(v_s) A_d \frac{1}{2} \rho_G |v_s|^2 \frac{(v_{rG} - v_{rd})}{|v_s|} + (\rho_L - \rho_G) \frac{v_{\theta d}^2}{r_d} V_d \tag{15}$$

$$\rho_L V_d \frac{dv_{\theta d}}{dt_c} = C_D(v_s) A_d \frac{1}{2} \rho_G |v_s|^2 \frac{(v_{\theta G} - v_{\theta d})}{|v_s|} - (\rho_L - \rho_G) \frac{v_{rd} v_{\theta d}^2}{r_d} V_d \tag{16}$$

$$\rho_L V_d \frac{dv_{zd}}{dt_c} = C_D(v_s) A_d \frac{1}{2} \rho_G |v_s|^2 \frac{(v_{zG} - v_{zd})}{|v_s|} + (\rho_L - \rho_G)|g| V_d \tag{17}$$

This set of differential equations is numerically solved by using given initial conditions at the end of the inlet duct of the cyclone as shown in Fig. 2.

The mass transfer coefficient in the liquid film on the cyclone wall, $k_{Lw}A_w$, is predicted by a correlation based on the measurements of Johnstone and Silcox [1947] in a cyclone spray tower as quoted by Schrauwen [1988] as follows:

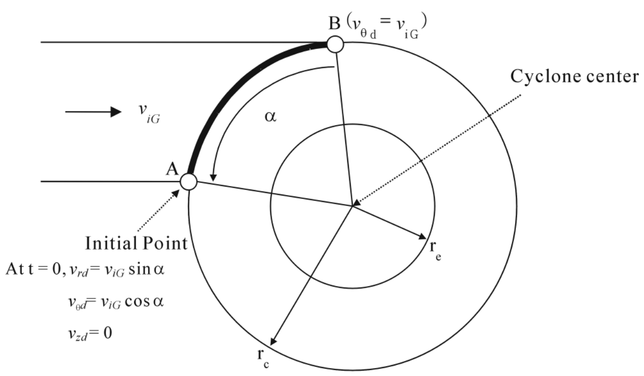

Fig. 2. Initial conditions of differential equations inside cyclone.

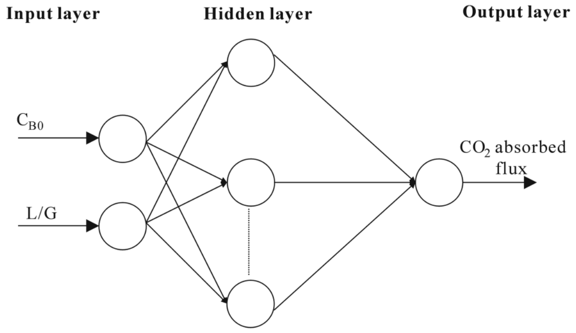

Fig. 3. The structure of three layer feed-forward neural network (3-FFNN).

$$k_{Lw} A_w = 0.0049 (v_{\theta G}^2(r_c) + v_{zG}^2(r_c))^{0.37} N_{Sc}^{-2/3} 2\pi r_c z_w \tag{18}$$

NEURAL NETWORK MODEL

Many artificial neural network architectures have been proposed. In this study, the three-layer feed-forward neural network (3-FFNN) is used as shown in Fig. 3. A more detailed description of 3-FFNN will be found in many textbooks that have been published on neural networks [Freeman and Skapura, 1991].

Many studies have proved the ability of 3-FFNN to approximate any nonlinear relationship between a set of input and output. In the present case, we are interested in finding the CO_2 absorbed flux (output of the network) as a function of state variables (input of the network). In our system the inputs will be the initial concentration of alkali in the solution, C_{B0}, and the liquid-gas ratio, L/G. The correlation between input and output can be expressed as the following equation:

$$\left(C_{B0}, \frac{L}{G}\right) = f(CO_2 \text{ absorbed flux}) \tag{19}$$

To achieve a proper neural network system, which may give an adequate result for Eq. (19), two parameters, namely, the speed of the network and the computation time should be kept in mind. To determine the speed of the network and to improve the convergence rate, both learning and momentum rates were employed. By trial and error of the neural network program, the relative minimum error and computation time to be used in the neural network parameters are given as in Table 1. These parameters are used in the further computation.

Table 1. Parameters for neural network calculation

Learning rate	0.12
Momentum rate	0.12
Iteration cycles	2000
Mean squared error	1.00E-05
Structure:	
Input neurons	3
Hidden neurons	6
Output neurons	1
Transfer function:	
Hidden layer	Sigmoid
Output layer	Linear

The facilities of the Neural Network Toolbox of the MATLAB software (MATLAB V5.3, 1999) have been used in the present computation. All the data as shown in Figs. 4 and 5, were used in the calculation.

RESULTS AND DISCUSSION

Fig. 4 shows the effect of the initial concentration of alkali, C_{B0}, on CO_2 absorbed flux for two absorbents at L/G of 0.16. The increase in C_{B0} slightly increased the CO_2 absorbed flux. The flux increased significantly from the initial concentration of alkali in solution, C_{B0}=0.001 kmol/m^3. It indicates that the reaction between CO_2 and the solutions can take place in a basic solution of C_{B0}=0.001 kmol/m^3 (pH≥11). Although the reaction also occurs at pH lower than 11, it can be considered negligible. In all cases, CO_2 was absorbed significantly when the initial concentration was increased [Camacho et al., 2000]. Fig. 4 also shows that the experimental data agreed well with the simulation result by the neural network. The phenomenological model gives partly agreeable results with the experimental data around C_{B0}≤0.001 kmol/m^3 (pH≤11). The deviation becomes significant with C_{B0}.

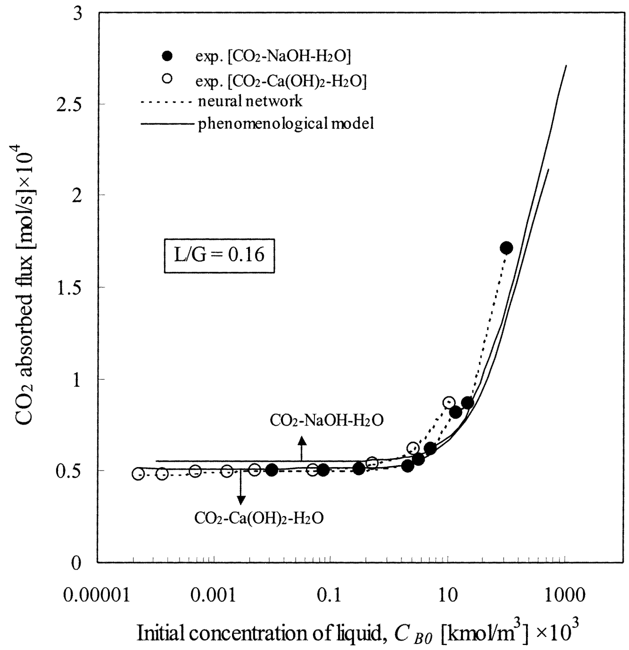

Fig. 4. Effect of initial concentration of alkali in solution, C_{B0}, on CO_2 absorbed flux.

The effect of the liquid - gas ratio on absorption is presented in Fig. 5 with the constant gas flow rate. The increase in L/G obviously increased the CO_2 absorbed flux. The consequence of a higher L/G ratio is a higher surface area. According to Jorg and Buttner [1994], the number of droplets increases and thus the mean distance between droplets is reduced and the surface area of droplets significantly increase at a higher L/G ratio. Fig. 5 also shows that the experimental data agrees well with calculated values by the neural network. The phenomenological values also show in good agreement with the experimental data.

Fig. 6 shows the comparison of the experimental results of CO_2 absorbed flux with those calculated by Eq. (20) using neural network and by the phenomenological model, while the absolute mean relative errors are summarized in Table 2.

$$\text{Relative error} = \left(\frac{\text{calculated } CO_2 \text{ abs. flux}}{\text{experimental } CO_2 \text{ abs. flux}} - 1\right) \times 100\% \qquad (20)$$

The neural network gives much better results than the phenomenological model. The largest absolute mean relative error was found to be 0.725% by neural network and 8.965% by phenomenological model. It might be concluded that the 3-FFNN has enough accuracy to obtain the CO_2 absorbed flux in the cyclone scrubber under

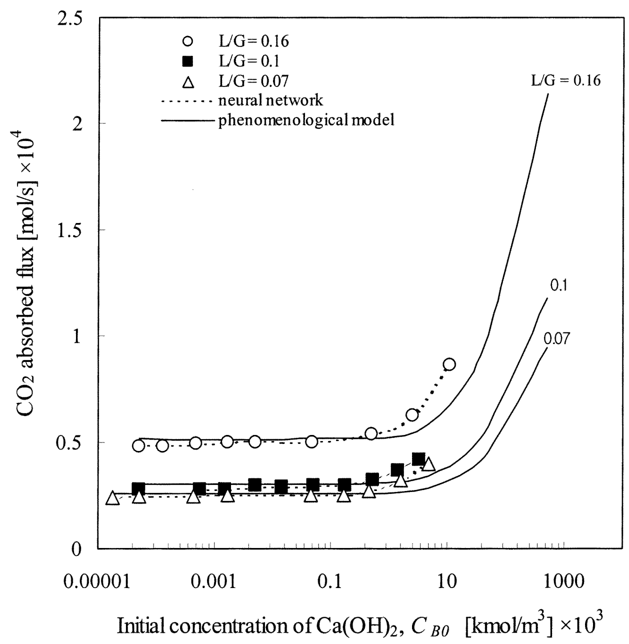

Fig. 5. Effect of liquid-gas ratio, L/G, on CO_2 absorbed flux.

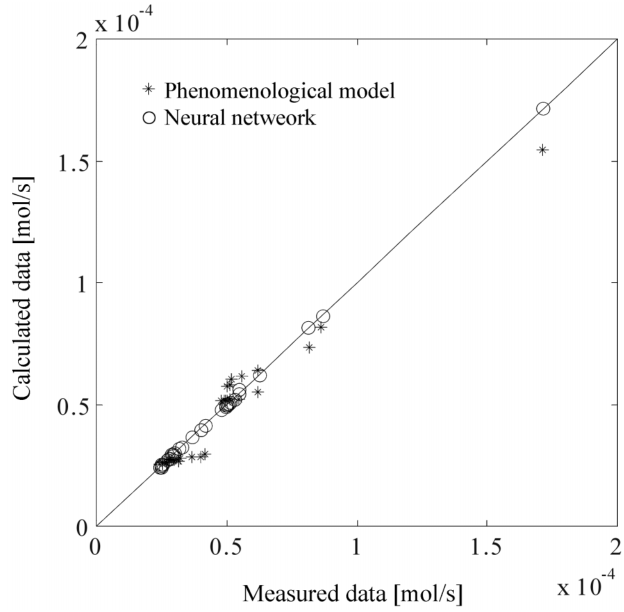

Fig. 6. Parity plot of experimental CO_2 absorbed flux *vs.* calculated values.

Table 2. Mean absolute relative error between experimental and calculated CO_2 absorbed flux

Gas-liquid system	Mean absolute relative error, %	
	Phenomenological model	3-FFNN
CO_2-$Ca(OH)_2$-H_2O:		
L/G=0.16	4.373	0.385
L/G=0.1	7.944	0.540
L/G=0.07	8.184	0.366
CO_2-NaOH-H_2O:		
L/G=0.16	8.965	0.725

the current experimental conditions. Because of a narrow data range used in this study, more data are necessary to make the proposed model more confident. The absolute mean relative error between phenomenological model and experimental data is less than 10%, which it is reasonable prediction for a phenomenological model.

CONCLUSION

This paper presents modeling strategies by the phenomenological model and artificial neural networks for the non-linear dynamic processes of a cyclone scrubber. The phenomenological models were built on physical principles. The three layer feed-forward neural network (3-FFNN) has been chosen for neural network modeling.

The comparison of the simulation results of the neural network model and the nonlinear phenomenological model with experimental data has been discussed to show the validity of the proposed models. The comparison illustrates that the accuracy of 3-FFNN and the phenomenological model is satisfactory with experimental data. In conclusion, the highly non-linear behavior of cyclone scrubber can be modeled successfully by utilizing the 3-FFNN, and the phenomenological model can be the next best description of the performance of gas absorption in the cyclone scrubber.

NOMENCLATURE

a : contact area per unit volume [m^2/m^3]
A : interfacial area of droplet [m^2]
A_d : cross sectional area of droplet [m^2]
A_w : interfacial area of wetted wall [m^2]
C_A : concentration of CO_2 at time [$kmol/m^3$]
C_A^* : concentration of CO_2 in equilibrium with CO_2 in gas phase [$kmol/m^3$]
C_{B0} : initial concentration of alkaline solution [$kmol/m^3$]
C_D : drag coefficient [-]
d_p : diameter of droplet [m]
D : momentum exchange parameter [-]
D_{AG} : diffusivity of CO_2 in gas [m^2/s]
g : standard acceleration of gravity [m/s^2]
G : volumetric gas flow rate [m^3/s]
G_m : molar gas flow rate [kmol/s]
G_{mf} : molar gas flow through the inlet duct of the cyclone [kmol/($s\cdot m^2$)]
H : Henry's constant [kmol/($m^3\cdot$atm)]
h_t : total height of the cyclone [m]
h : height position in cyclone [m]
k_G : gas-phase mass transfer coefficient for CO_2 [kmol/($m^2\cdot s\cdot$atm)]
k_L : mass transfer coefficient for CO_2 with reaction for droplets [m/s]
k_{Lw} : mass transfer coefficient for CO_2 with reaction for wetted wall [m/s]
L_m : molar liquid flow rate [kmol/s]
L : volumetric liquid flow rate [m^3/s]
P : total pressure of CO_2 [atm]
P_A : partial pressure of CO_2 [atm]
P^* : equilibrium pressure of CO_2 [atm]
P_{A1} : partial pressure of CO_2 in inlet gas [atm]
P_{A2} : partial pressure of CO_2 in outlet gas [atm]
r_e : radius of gas outlet duct [m]
r_o : initial radius of droplet [m]
R : gas law constant [kmol/($m^3\cdot$K)]
R_{g-L} : ratio of gas-side resistance to liquid-side resistance [-]
s : gas outlet duct height [m]
S : cross-sectional area of tangential inlet duct [m^2]
t_i : contact time in tangential inlet duct of the cyclone [s]
t_c : contact time in cyclone [s]
v : velocity [m/s]
T : temperature [K]
$V_{con.}$: volume of conical section [m^3]
V_d : volume of droplet [m^3]
z : distance from nozzle point [m]
$z_{con.}$: height position in conical section [m]
z_w : wetted height of the cyclone [m]

Greek Letters

ΔC_A : driving force based on liquid concentration [kg/m^3]
ρ : density [kg/m^3]
ρ_m : molar density of liquid [$kmol/m^3$]
μ : viscosity [kg/($m\cdot s$)]

Subscripts

d : droplet
G : gas
L : liquid
r : radial
θ : tangential
w : cyclone wall
z : axial

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