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journal or publication title	Science reports of the Research Institutes, Tohoku University. Ser. A, Physics, chemistry and metallurgy
volume	19
page range	254-269
year	1967
URL	<a href="http://hdl.handle.net/10097/27386">http://hdl.handle.net/10097/27386</a>

# Absorption of Gas in Liquid with Corona Discharge\*

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(Received November 1, 1967)

## Synopsis

The absorption rate of gases in liquid in wetted-wall columns is strongly accelerated by a corona discharge. The phenomenon occurs when the main resistance is in the gas phase. It is recognized that the acceleration of the absorption rate is caused by the turbulence of the gas stream, in wetted-wall columns, produced by the corona discharge.

## I. Introduction

The increasing amount of energy consumed has made it more and more necessary to remove sulfur dioxide from the gaseous effluent of chemical factories, refineries, electric power stations, etc. These factories are coming under increasing public pressure to reduce this atmospheric pollutant.

The following data report studies dealing with the absorption of gases in liquids with a corona discharge which is a method of removing sulfur dioxide from effluent gases. Furthermore, the mechanism of the acceleration of the absorption rate with a corona discharge was discussed. When the gas phase resistance controlled the absorption rate, experimentations were carried out in the following systems, SO<sub>2</sub>-H<sub>2</sub>O, NH<sub>3</sub>-H<sub>2</sub>O, SO<sub>2</sub>-NaOH, H<sub>2</sub>S-NaOH, Cl<sub>2</sub>-NaOH and CO<sub>2</sub>-NaOH. The systems in which the liquid phase resistance was controlling the absorption rate were absorption of H<sub>2</sub>S and Cl<sub>2</sub> in water.

## II. Apparatus and Procedure

The wetted-wall columns, one of them is shown in Fig. 1 are made of poly-metacrylic resin, and their inner diameters and lengths of columns are as follows:  $d=2.5$  cm,  $l=30.3$ , 60.3 and 120.3 cm;  $d=5.0$  cm,  $l=30.3$  cm. The liquid is fed by overflow from the top of a column along the wall uniformly, and the gas and the liquid are separated at the bottom of the column. A corona discharge occurs around a Pt wire electrode, diam. 0.5 mm being set as a high potential electrode along the axis of the column and the falling water along the wall is grounded. The gas is exhausted from the top of the tower after passing through a calming section and is absorbed in the wetted-wall column. The flow rates of the gas and the liquid were

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\* Reprinted in Japanese in Chem. Eng., Japan, **31** (1967), 878.

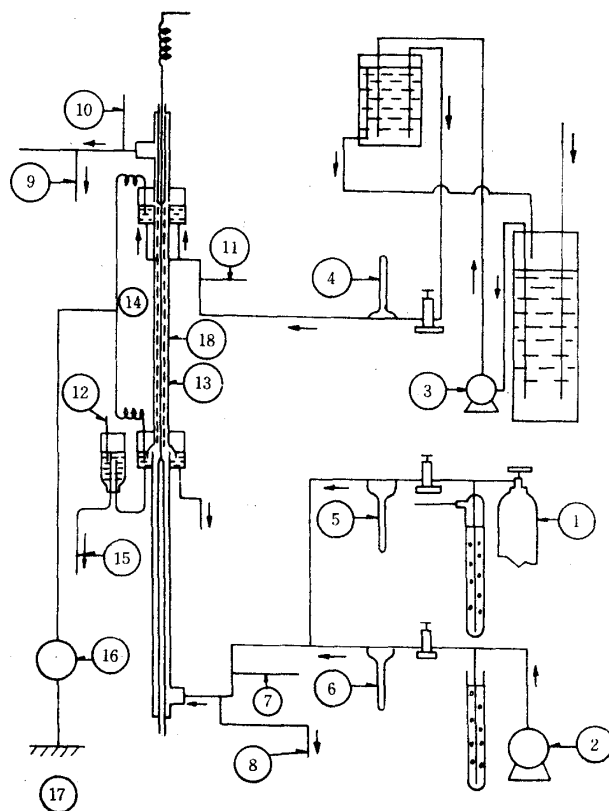


Fig. 1. Schematic diagram of apparatus.

1. SO<sub>2</sub> cylinder 2. air-blower 3. water pump 4.~6. orifice flow meter  
 7, 10.~12. thermometer 8. inlet gas sampling pipe 9. outlet gas sampling  
 pipe 13. wetted-wall tower 14. central wire electrode 15. outlet liquid  
 sampling tube 16. milli-ampere meter 17. earth 18. cylindrical net

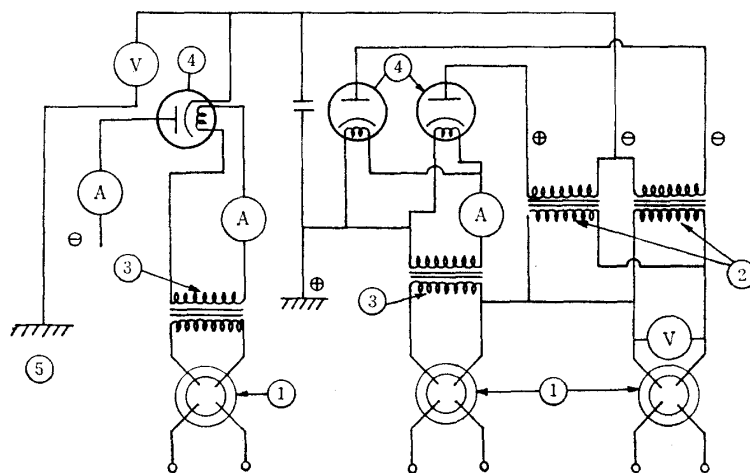


Fig. 2. High voltage apparatus.

1. aut-transformer 2. high voltage transformer 3. filament transformer  
 4. rectifying tube 5. earth

measured by glass orifice meters. A corona discharge was energized by a high tension taken from a wall outlet which was transformed and rectified by electric valves. The schematic wiring diagram is shown in Fig. 2. The Pt wire should be set in the center of the tower, and be tightened slightly to avoid the vibration of the Pt electrode and break of the water film inside of the column.

In another series of experiments, a control grid which captured ions was used to determine the mechanism of acceleration of the rate of absorption by a corona discharge. A control grid is a stainless steel cylindrical wire net, and a mesh of the net is  $1 \times 1$  mm. It was set concentrically in the wetted-wall column, diam. 5.0 cm, length 30.3 cm. The potential was applied to the Pt wire electrode and the cylindrical net and water electrode were grounded.

### III. Numerical Data

#### 1. Partial pressure and solubility coefficient

Partial pressures and solubility coefficients of many sorts of gases in dilute aqueous solutions are obtained by extrapolation from the following data:  $\text{SO}_2\text{-H}_2\text{O}$  system by Yui and Saito<sup>(1)</sup>,  $\text{NH}_3\text{-H}_2\text{O}$  system by Sherwood<sup>(2)</sup>,  $\text{H}_2\text{S-H}_2\text{O}$  system by Wright<sup>(3)</sup>,  $\text{Cl}_2\text{-H}_2\text{O}$  system by Whitney and Vivian<sup>(4)</sup>. Solubility coefficient of  $\text{CO}_2$  in NaOH solution was calculated using the equation by Nijsing and Kramers<sup>(5)</sup>. Partial pressures of  $\text{SO}_2$ ,  $\text{Cl}_2$ ,  $\text{H}_2\text{S}$  and  $\text{CO}_2$  in the gas phase dissolved in NaOH solution were neglected, when the values of  $K_{OG}$  were calculated.

#### 2. Mass transfer coefficient of gas and liquid phase

Mass transfer coefficient in the gas phase was calculated by Ramm's equation<sup>(6)</sup> in order to compare the experimental results of gas absorption in water with the calculated values and  $k_l$  mass transfer coefficient for liquid phase, was calculated by Hikita's equation<sup>(7)</sup>.

$$k_g = 0.027 \times \frac{D}{d} \left( \frac{d u \rho}{\mu} \right)^{0.8} \left( \frac{\mu}{\rho D} \right)^{0.33} \times \frac{1}{RT} \quad (1)$$

$$H_L \left( \frac{\rho^2 g}{\mu^2} \right)^{1/3} = 2.36 \times Re^{1.0} \left( \frac{\mu}{\rho D_l} \right)^{1/2} \quad (2)$$

#### 3. Diffusion coefficient of gas

The diffusion coefficients of gas were calculated by Arnold's equation.

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- (1) S. Saito and N. Yui *J. Soc. Jap., Pure Chem. Sec.*, **80** (1958), 139.
  - (2) T.K. Sherwood, *Ind. Eng. Chem.*, **17** (1925), 745.
  - (3) R.H. Wright and O. Mass, *Canad. J. Res.*, **6** (1932), 94.
  - (4) Roy P. Whitney and J.E. Vivian, *Ind. Eng. Chem.*, **33** (1941), 741.
  - (5) R.A.T.O. Nijsing and H. Kramers, *Chem. Reaction Eng., Meeting Europ. Federation Chem. Eng. 12th Amsterdam* (1957), 81.
  - (6) W.M. Ramm, *Absorptionprozesse in der chemischen Technik* p. 197 VEB Verlag Technik (1953).
  - (7) H. Hikita, H. Nakanishi and K. Katoka, *Chem. Eng. Japan*, **23** (1959), 459.

Diffusion coefficients in a liquid for SO<sub>2</sub> and Cl<sub>2</sub> by Peaceman<sup>(8)</sup>, for H<sub>2</sub>S by Wilke<sup>(9)</sup>, for CO<sub>2</sub> by Nijsing<sup>(10)</sup> and for NH<sub>3</sub> by Arnold's equation<sup>(11)</sup> were used. Diffusion coefficient for CO<sub>2</sub> in NaOH solution was calculated by following equations  $D_l = D_{H_2O}/\eta$ ,  $\eta = \mu_{NaOH}/\mu_{H_2O}$

#### 4. Reaction coefficient

The reaction coefficient for CO<sub>2</sub> in NaOH solution was calculated by Hatta's equation<sup>(12)</sup>.

$$\beta = \gamma / \tanh \gamma \quad (3)$$

where

$$\gamma = \sqrt{k [\text{OH}^-] D_l} / k_l \quad (4)$$

The second order rate constant,  $k$ , for CO<sub>2</sub> + OH<sup>-</sup> → HCO<sub>3</sub><sup>-</sup> was obtained by interpolating Pinsent's results<sup>(13)</sup>. The values of  $\beta$  are given in Table 1.

Table 1. Calculated value of  $\beta$ .

Run No.	Temp. of liquid [°C]	$k_g$ Calc. by Eq. (9)	$k_l$ Calc. by Eq. (10)	$H$ [kg mole / m <sup>3</sup> · atm]	$\beta$ [-]
1	25.0	0.372	0.356	0.269	40.2
2	22.0	0.375	0.338	0.293	36.3
3	24.5	—	0.353	0.273	39.3
4	24.8	—	0.355	0.270	39.8

## IV. Results

There is a following relation between the rate of absorption and the overall coefficient of mass transfer based on the pressure.

$$w = K_{OG} (p_g - p^*)_{lm} \quad (5)$$

where  $w$  is an absorption velocity per unit area.  $p_g$  and  $p^*$  are the partial pressure of bulk of gas and the equilibrium partial pressure with the solution with the concentration of  $C_l$ .  $(p_g - p^*)_{lm}$  is a logarithmic mean value between the inlet and the outlet.

#### 1. Effect of electric current

The effect of the current density,  $i$ , on  $K_{OG}$  with the constant flow rates of the gas and the liquid is shown in Figs. 3, 4 and Table 2.  $K_{OG}$  increases with  $i$  and are

(8) D.W. Peaceman, Sc. D. thesis, Mass. Inst. Tech. (1951).

(9) C.R. Wilke, Chem. Eng. Progress, **45** (1949), 218.

(10) R.A.T.O. Nijsing, R.H. Hendriksz and H. Kramers, Chem. Eng. Sci., **10** (1959), 88.

(11) J.H. Arnold, Ind. Eng. Chem. **22** (1930), 1091.

(12) S. Hatta, J. Chem. Soc. Japan, Ind. Chem. Sec., **35** (1932), 1389.

(13) B.R.W. Pinsent, L. Pearson and F.J.W. Roughton, Trans. Faraday Soc., **52** (1956), 263.

saturated gradually in the following systems,  $\text{SO}_2\text{-H}_2\text{O}$ ,  $\text{NH}_3\text{-H}_2\text{O}$ ,  $\text{SO}_2\text{-NaOH}$ ,  $\text{Cl}_2\text{-NaOH}$ ,  $\text{H}_2\text{S-NaOH}$  and  $\text{CO}_2\text{-NaOH}$ . In the system  $\text{SO}_2\text{-H}_2\text{O}$ ,  $K_{OG}$  has the maximum value with the increase of  $i$ , when the concentration is large. Fig. 5 shows the corona discharge in air- $\text{SO}_2$  mixture in a wetted-wall column. On the contrary, it seems that there is no acceleration of the absorption rate in the systems  $\text{H}_2\text{S-H}_2\text{O}$  and  $\text{Cl}_2\text{-H}_2\text{O}$  with a corona discharge. There appears to be a trend of apparent increase of  $K_{OG}$  with  $i$  in the absorption of  $\text{H}_2\text{S}$  in water, probably because of the decomposition of  $\text{H}_2\text{S}$  to  $\text{S}$  at a negative electrode.

## 2. Effect of concentration of gas

$K_{OG}$  decreases with the increase of the concentration of  $\text{SO}_2$  in a corona discharge, as shown in Fig. 6. Fig. 7 shows the characteristic current vs. voltage curves, which has the definite relation between voltage and current at the constant composition. Thus, the higher voltage is needed to maintain the same current density, in accordance with the increase of concentration of  $\text{SO}_2$  in air- $\text{SO}_2$  and  $\text{N}_2\text{-SO}_2$  mixture as shown in Fig. 7.

## 3. Effects of voltage and polarity of an electrode

Fig. 8 shows the correlations between  $K_{OG}$  and voltage.  $K_{OG}$  with a corona (her-

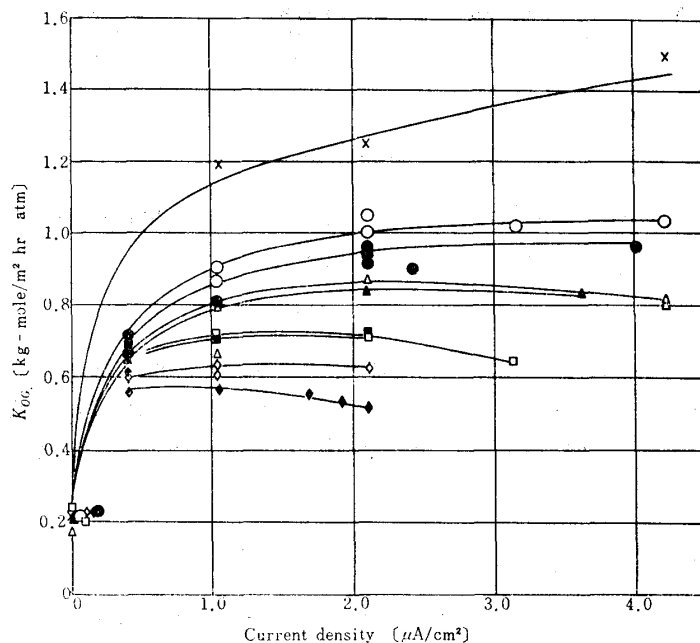


Fig. 3. Correlation of  $K_{OG}$  with current density.

$\text{N}_2\text{-SO}_2$					
$\text{SO}_2$ %	• 3.1	▲ 5.7	■ 11.6	◆ 20.4	
$Re_g$	500~550	460~570	480~580	520~710	
$Re_L$	770~720	540~670	560~680	550~670	
$\text{Air-SO}_2$					
$\text{SO}_2$ %	× 1.7	○ 3.2	△ 5.6	◻ 11.9	◇ 23.4
$Re_g$	480~500	520~540	540~550	590~600	720~730
$Re_L$	490~570	510~580	510~540	520~540	540~570

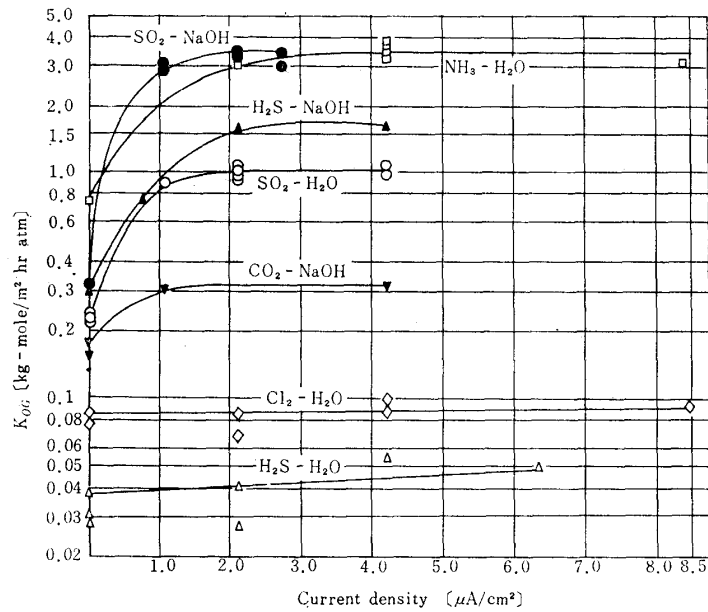


Fig. 4. Absorption of gas in water and NaOH solution with corona discharge.

Absorption by H<sub>2</sub>O

	conc. of gas	$Re_g$	$Re_L$
□	NH <sub>3</sub> 4.4~5.7%	680~760	490~710
○	SO <sub>2</sub> 2.9~3.4%	490~540	440~610
△	H <sub>2</sub> S 2.6~2.7%	490~500	620~640
◇	Cl <sub>2</sub> 3.7~4.1%	580~590	510~540

## Absorption by NaOH (0.95~1.07 N)

	conc. of gas	$Re_g$	$Re_L$
▽	CO <sub>2</sub> 2.4~2.6%	490	370~470
●	SO <sub>2</sub> 2.7~2.9%	500~510	380~390
▲	H <sub>2</sub> S 2.6~2.8%	490~500	350~390

Table 2. Absorption of Cl<sub>2</sub> in NaOH solution with corona discharge.

Run No.	Conc. of NaOH solution [N]	Conc. of Cl <sub>2</sub> [mole fraction]		Temp. [°C]		$Re_g$ [-]	$Re_L$ [-]	Voltage [kV]	Electric current density [ $\mu$ A/cm <sup>2</sup> ]	$K_{OG}$ [kg mole/m <sup>2</sup> ·hr·atm]
		Inlet	Outlet	Gas	Liquid					
1	1.05	0.0274	0.0165	21.3	20.5	515	353	0	0	0.235
2	1.05	0.0285	0.0014	21.0	20.6	518	355	8.8	2.11	1.37

eafter abbr. to  $K_{OG}$ , c.d.) has the same value as  $K_{OG}$  without a corona (hereafter abbr. to  $K_{OG}$ , n.) until 7.5 and 9.5 kV with a positive and a negative corona, respectively.  $K_{OG}$  increased rapidly beyond 10kV with a negative corona, which produces a corona current of 0.42  $\mu$ A/cm<sup>2</sup>, and then the increase of  $K_{OG}$  continues until 11kV, but  $K_{OG}$  does not increase beyond 11kV, and current density increases. It is assumed that there will be the same trend as a negative corona in the case of positive corona. Experimentation is not carried out for the higher voltage, because the characteristic relation between electric potential and current can not be maintained by unstable electric field which is produced by dropping water along the electrode.

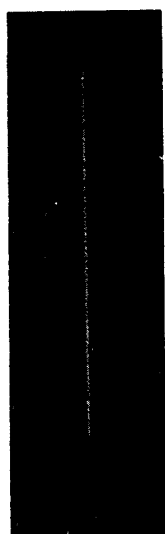


Fig. 5. Corona discharge in air-SO<sub>2</sub> mixture, in a wetted-wall column.  
Diameter of column 2.5 cm; height of column 60.3 cm; air-SO<sub>2</sub> mixture, SO<sub>2</sub> 2.85%;  $Re_g$  500; the flow rate of water 800cc/min; current density 2.11 $\mu$ A/cm<sup>2</sup>; voltage 10.8kV; F2, time 1 sec.

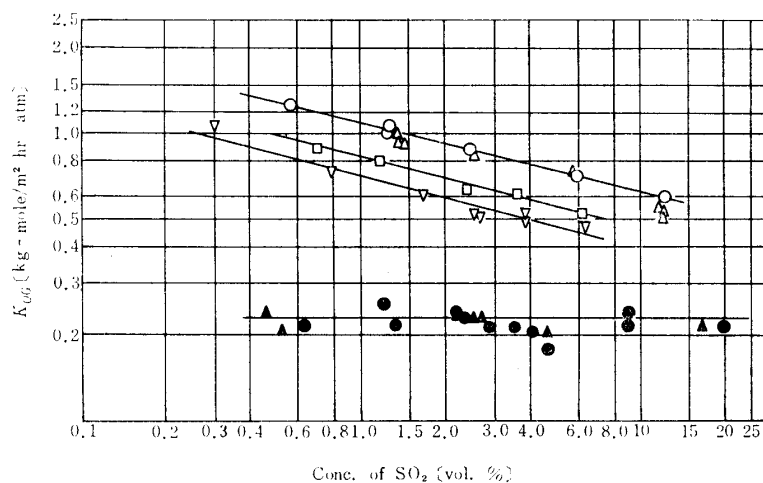


Fig. 6. Correlation of  $K_{OG}$  with SO<sub>2</sub> concentration, with corona;  $Re_L$  470~680:  
negative corona, ○ air-SO<sub>2</sub>, 2.11 $\mu$ A/cm<sup>2</sup>,  $Re_g$  490~730,  
▽ air-SO<sub>2</sub>, 0.42 $\mu$ A/cm<sup>2</sup>,  $Re_g$  490~530,  
△ N<sub>2</sub>-SO<sub>2</sub>, 2.11 $\mu$ A/cm<sup>2</sup>,  $Re_g$  540~690,  
□ air-SO<sub>2</sub>, 0.42 $\mu$ A/cm<sup>2</sup>,  $Re_g$  500~530, A. C. corona  
without corona  
● air-SO<sub>2</sub>,  $Re_g$  500~740,  $Re_L$  500~600  
▲ N<sub>2</sub>-SO<sub>2</sub>,  $Re_g$  460~520,  $Re_L$  610~720

Therefore it is considered that the acceleration of the absorption rate depends rather on the electric current than on the voltage because the acceleration begins with the flow of electric current. Fig. 6 shows that an A.C. corona has the same trend as the positive and the negative corona.

#### 4. Effect of column length

SO<sub>2</sub> was absorbed by water, using wetted-wall columns diam. 2.5 cm and lengths 30.3, 60.3 and 120.3 cm.  $K_{OG}$  decreases with the increase of column length



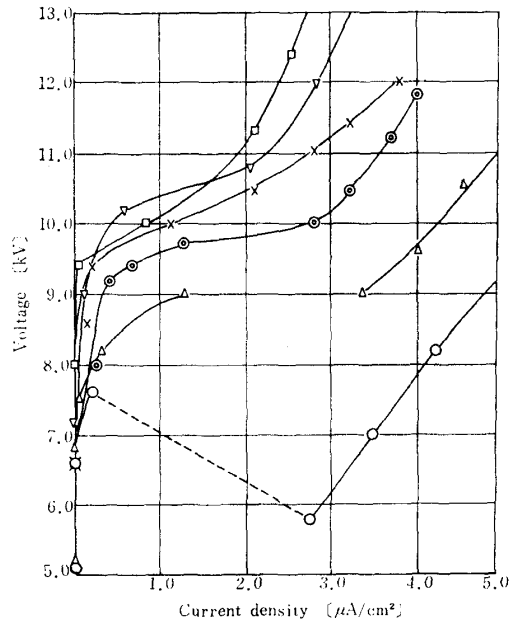


Fig. 7.

Fig. 7. Characteristics between current density and voltage.

$Re_g$  500,  $Re_L$  500~570

○ pure  $N_2$ ;  $N_2$ - $SO_2$ ,      △  $SO_2$  1%,      ▽  $SO_2$  3%;  
 ● air;      air- $SO_2$ ,      ×  $SO_2$  1%,      ◻  $SO_2$  3%

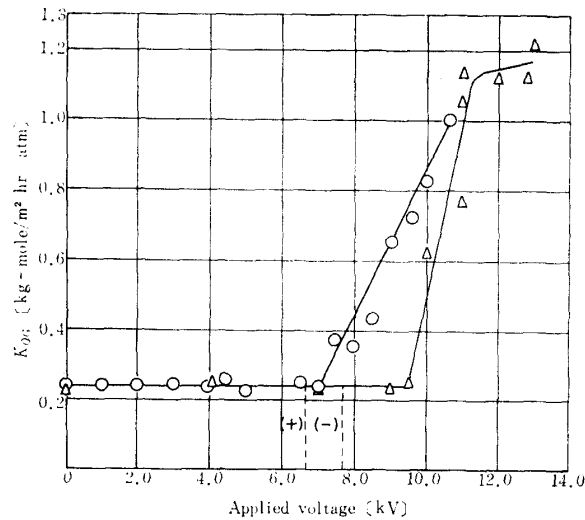


Fig. 8.

Fig. 8. Correlation of applied voltage (potential difference between the electrodes) with  $K_{OG}$

$SO_2$ , 2.8~3.2%;  $Re_g$ , 500~540;  $Re_L$ , 530~640:

△ negative corona,      ○ positive corona

(+), (-) positive and negative corona onset voltage

with and without a corona. Fig. 9 shows the correlation between the number of transfer unit,  $N_{OG}$  and the column length. The relation is plotted as a straight line, but when it is extrapolated, it does not pass through the origin because of end effects. But these values need not be corrected considering that their deviations are small for the purpose of observing the effect of the corona discharge.

## 5. Effect of gas flow rate

The experimental results of the absorption of  $SO_2$  and  $NH_3$  in water in the range of  $Re_g=500\sim6000$ , are shown in Fig. 10. With the corona discharge,  $K_{OG}$  in  $SO_2$ - $H_2O$  system decreases gradually with  $Re_g$ , and  $K_{OG}$  becomes constant at the range from 2000 to 3000 in  $Re_g$ . However,  $K_{OG}$ , n. increases with  $Re_g$ , and coincides with  $K_{OG}$ , c.d. at  $Re_g=6000$ , on the absorption of  $SO_2$ . On the absorption of  $NH_3$  in water, the trend of curves is almost similar to that of the absorption of  $SO_2$ , and the curves of  $K_{OG}$ , n. and  $K_{OG}$ , c.d. coincide at  $Re_g=10000$ . The increase of  $K_{OG}$ , n. with  $Re_g$  is considered to be caused by the decrease of the gas phase resistance, and the increase of  $K_{OG}$ , c.d. is also considered to be caused by the decrease of the gas resistance which is given by the vortex of gas in the wetted-wall column.

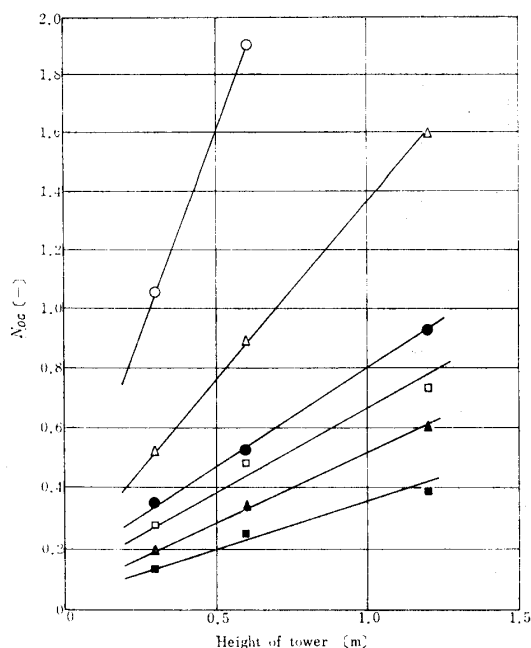


Fig. 9. Correlation of  $N_{OG}$  with height of tower.

SO<sub>2</sub> 3%,  $Re_L$  570~690;

with corona,  $2.11\mu\text{A}/\text{cm}^2$

$Re_g$ , ○ 500,

△ 1000,

□ 2000

SO<sub>2</sub> 3%,  $Re_L$  570~660;

without corona

$Re_g$  • 500,

▲ 1000,

■ 2000

## 6. Effect of liquid flow rate and temperature of liquid and gas

Fig. 11 shows the correlation of  $K_{OG}$  and  $Re_L$ .  $K_{OG}$ , c.d. decreases with the increase of  $Re_g$ . As shown in Fig. 12,  $K_{OG}$  decreases with the rise of the liquid temperature with and without the corona discharge, and the effect of the temperature on  $K_{OG}$ , c.d. is larger than  $K_{OG}$ , n. There is no effect of the gas temperature on  $K_{OG}$ , n. and  $K_{OG}$ , c.d. as shown by the data in Fig. 13.

## 7. Effect of the concentration of oxygen

The correlation between  $K_{OG}$  and the concentration of oxygen is shown in Fig. 14. When the concentration of SO<sub>2</sub> in nitrogen is very small, the state of the corona becomes unstable and the effectiveness of the corona is very slight. However, the corona discharge becomes stable and has the larger effect on the absorption, when the oxygen concentration in N<sub>2</sub>-O<sub>2</sub>-SO<sub>2</sub> mixture exceeds more than 5%, even if the SO<sub>2</sub> concentration is very dilute.

## 8. Effect of cylindrical wire net

Fig. 1 shows the apparatus which has a stainless steel cylindrical wire net between the wetted-wall column and the Pt electrode. Experimental results obtained by the above apparatus are presented in Table 3. The electric current

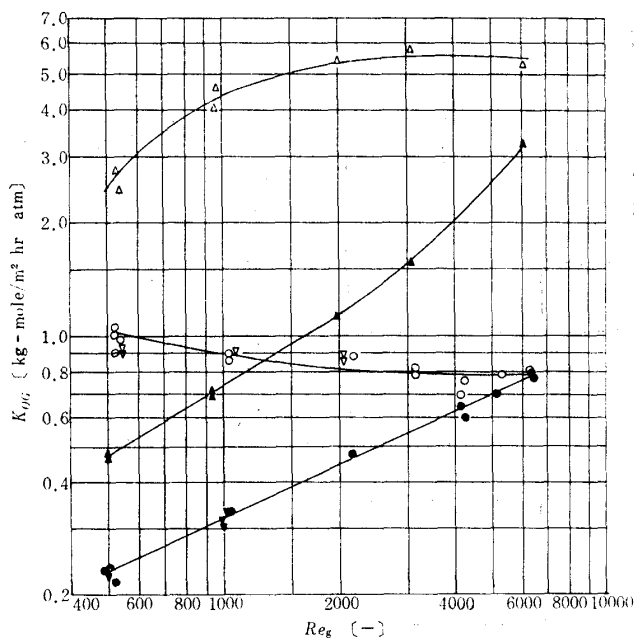


Fig. 10. Correlation of  $K_{OG}$  with  $Re_L$ .  
 NH<sub>3</sub>, 2.0~2.7%;  $Re_L$  520~630:  
 ▲ without corona    △ with corona 4.2μA/cm<sup>2</sup>  
 SO<sub>2</sub>, 2.83~3.38%:  
 without corona • air-SO<sub>2</sub>,  $Re_L$  490~610; ▼ N<sub>2</sub>-SO<sub>2</sub>,  $Re_L$  670~730:  
 with corona, 2.11μA/cm<sup>2</sup>,  
 ○ air-SO<sub>2</sub>,  $Re_L$  510~570; ▼ N<sub>2</sub>-SO<sub>2</sub>,  $Re_L$ , 540~580

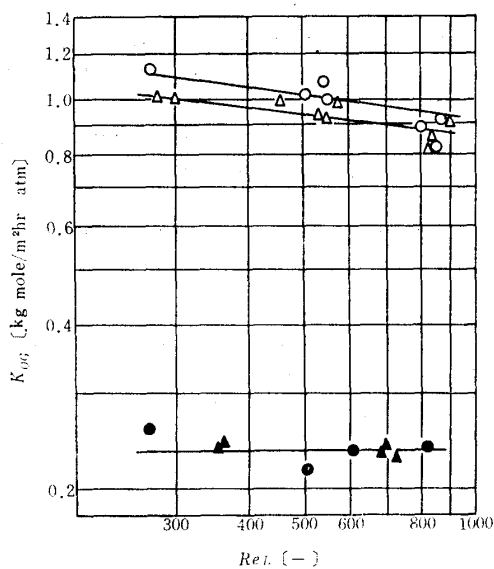


Fig. 11. Effect of the flow rate of water with corona, SO<sub>2</sub> 3.0~3.3%,  
 ○ air-SO<sub>2</sub>,  $Re_g$  540  
 ▲ N<sub>2</sub>-SO<sub>2</sub>,  $Re_g$  550  
 without corona SO<sub>2</sub> 2.8~3.4%,  
 • air-SO<sub>2</sub>,  $Re_g$  520  
 ▲ N<sub>2</sub>-SO<sub>2</sub>,  $Re_g$  500

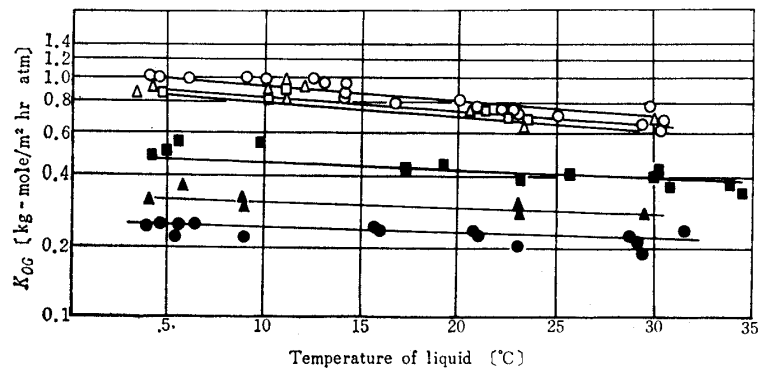


Fig. 12. Effect of liquid temperature.  
the flow rate of water 800cc/min;  $\text{SO}_2$ , 2.5~3.4%  
without corona  $Re_g$  • 500 ▲ 1000, ■ 2000  
with corona  $2.11 \mu\text{A}/\text{cm}^2$ ,  $Re_g$  ○ 500, △ 1000, □ 2000

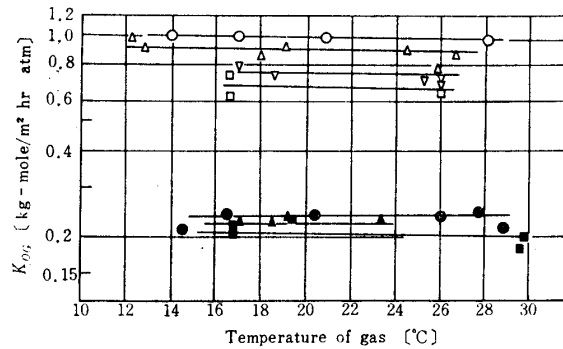


Fig. 13. Effect of gas temperature.  
with corona  $2.11 \mu\text{A}/\text{cm}^2$ ,  $\text{SO}_2$ , 2.5~3.3%,  $Re_g$  490~540, water 800cc/min; temp. of liquid  
○ 4.1~9.0°C ▲ 10.0~16.7°C, ▼ 20.0~22.8°C, □ 29.3~30.5°C.  
without corona,  $\text{SO}_2$ , 2.8~3.4%,  $Re_g$  490~520, water 800cc/min; temp. of liquid  
• 3.9~9.0°C, ▲ 15.8~21.5°C, ■ 23.0~29.4°C

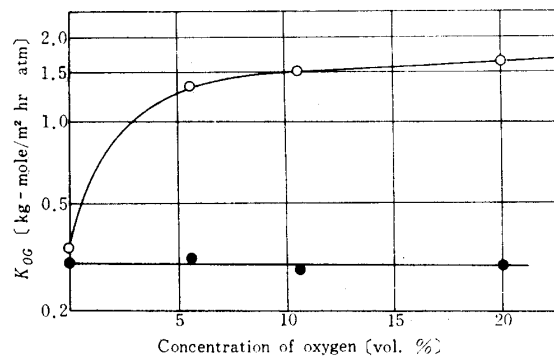


Fig. 14. Effects of concentration of oxygen.  
○ with corona,  $2.11 \mu\text{A}/\text{cm}^2$ ,  $\text{SO}_2$ , 0.66~0.76%,  $Re_g$  750  $Re_L$  480~530;  
• without corona  $\text{SO}_2$ , 0.66~0.79%,  $Re_g$  750.  $Re_L$  520~570,

Table 3. Absorption of SO<sub>2</sub> in a wetted-wall column with cylindrical net.

Run No.	Conc. of SO <sub>2</sub> [mole fraction]	Temp. of H <sub>2</sub> O [°C]	$Re_g$ [-]	$Re_L$ [-]	Voltage [kV]	Electric current		$K_{OG}$ [kg. mole /m <sup>2</sup> ·hr·atm]
						Betw. net and Pt-wire [mA/30.3cm]	Betw. net and water [ $\mu$ A/30.3cm]	
1	0.0280	14.8	502	297	0	0	0	0.228
2	0.0290	14.9	504	298	10.4	0.3	4	0.777
3	0.0263	15.1	500	298	10.7	0.5	4	0.793

Table 4. Absorption of SO<sub>2</sub> in wetted-wall columns without cylindrical net.

Run No.	Conc. of SO <sub>2</sub> [mole fraction]	Temp. of H <sub>2</sub> O [°C]	$Re_g$ [-]	$Re_L$ [-]	Voltage [kV]	Electric current [mA/30.3cm]	$K_{OG}$ [kg·mole/m <sup>2</sup> hr·atm]	Diam. of tower [m]
1	0.0232	12.5	498	278	0	0	0.207	0.05
2	0.0317	12.3	516	277	16.2	0.09	0.666	0.05
3	0.0242	13.0	501	282	16.2	0.30	0.965	0.05
4	0.0308	14.3	516	593	0	0	0.306	0.025
5	0.0425	15.0	542	595	10.5	0.2	0.777	0.025
6	0.0309	14.3	516	591	10.8	0.5	0.973	0.025
7	0.0296	17.0	502	627	12.0	1.0	0.986	0.025

Table 5. Amount of O<sub>3</sub> produced in the air with corona discharge.

Run No.	Flow rate of gas [l/min]	Flow rate of water [cc/min]	Voltage [kV]	Electric current density [ $\mu$ A/cm <sup>2</sup> ]	Temp. of gas [°C]	Temp. of liquid [°C]	Conc. of ozone produced in the air [%]
1	4	802	9.8	2.11	12.0	10.0	0.034
2	8	802	9.8	2.11	12.0	10.0	0.032
3	16	802	9.8	2.11	12.0	10.0	0.008

Table 6. Oxidized SO<sub>2</sub> in a wetted-wall column with corona discharge.

Run No.	289	287
Conc. of SO <sub>2</sub> in raw gas [mole fraction]	0.0307	0.0307
Voltage [kV]	10.8	12.0
Current density [ $\mu$ A/cm <sup>2</sup> ]	2.11	4.22
$Re_g$ [-]	515	527
$Re_L$ [-]	581	561
Temperature of gas [°C]	18.0	12.8
Temperature of water [°C]	14.0	13.0
SO <sub>4</sub> <sup>=</sup> in solution [H <sub>2</sub> SO <sub>4</sub> /total S in H <sub>2</sub> O 100mole%]	3.36	9.83
SO <sub>3</sub> in exhaust gas [SO <sub>3</sub> , H <sub>2</sub> SO <sub>4</sub> /total S in gas × 100mole%]	0	1.8

between the Pt electrode and the cylindrical net is 300 $\mu$ A and that between the cylindrical net and falling water is 4 $\mu$ A, and therefore most of the ions and the electrons which migrate to the water electrode are captured by the cylindrical net. Table 4 shows the experimental results of  $K_{OG}$  without the cylindrical net using wetted-wall columns diam. 5.0 and 2.5 cm. Comparing with the  $K_{OG}$  of Table 3,  $K_{OG}$  with the cylindrical net is a little smaller than the one without the net, but the effect of the corona is clearly confirmed.

### 9. Formation of ozone and sulfur trioxide

The amount of ozone which was produced by the electric discharge in the air is very slight as shown in Table 5. As shown in Table 6 the amount of  $\text{H}_2\text{SO}_4$  produced in water, when  $i$  is  $4.22 \mu\text{A}/\text{cm}^2$ , will be three times as great as when  $i$  is  $2.11 \mu\text{A}/\text{cm}^2$ . In the outlet gas, the oxidation efficiency of  $\text{SO}_2$  to  $\text{SO}_3$  is very small, and its values are zero and 1.8 mole%, when  $i$  is 2.11 and  $4.22 \mu\text{A}/\text{cm}^2$  respectively. Very small amount of  $\text{SO}_2$  is oxidized in higher current density.

### V. Mechanism of acceleration of the rate of absorption

The current of electricity is produced with the migration of electrons and ions which are produced by the corona discharge between two electrodes. Electrons and ions collide violently with neutral molecules, and are accelerated by the electric field at the interval of each collision. Therefore, it will be considered

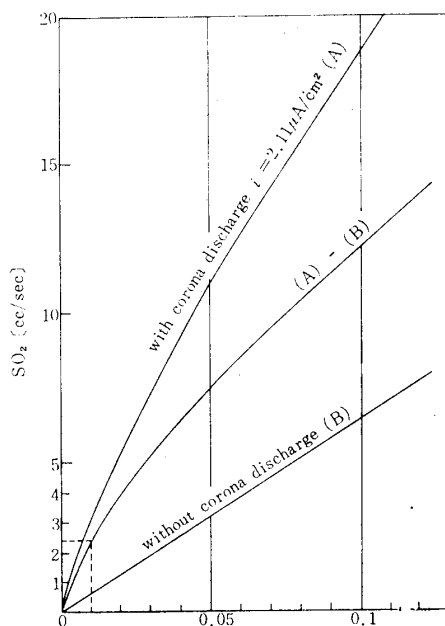


Fig. 15. Correlation of concentration of  $\text{SO}_2$  with absorbed  $\text{SO}_2$  without corona discharge and with corona discharge.

that the accelerated  $\text{SO}_2$  ions will reach rapidly to the water surface and the rate of absorption will be accelerated by the corona discharge. We compared the actual corona current with the ion current and found that all the  $\text{SO}_2$  molecules absorbed were ionized. The amount of absorbed  $\text{SO}_2$  by the corona discharge means the difference between the amount of absorbed  $\text{SO}_2$  with and without the corona discharges. When the concentration of  $\text{SO}_2$  is 1%, the rate of absorption of  $\text{SO}_2$  is 2.48 cc/min, as shown in Fig. 15. Assuming that the absorbed  $\text{SO}_2$  was transported from the Pt-wire electrode to the water electrode as ions, the numbers of  $\text{SO}_2$  ions

can be converted into the electric current equivalent, by the equation (5).

$$i = e N_{\text{SO}_2^-} \quad (5)$$

When the column diameter is 2.5 cm and the length is 60.3 cm, the current density is as follows.

$$i = e N_{\text{SO}_2^-} = 2.25 \times 10^{-2} \text{ A/cm}^2$$

The experimental current density is as follows.

$$i = 2.11 \times 10^{-6} \text{ A/cm}^2$$

The former is 10 thousand times as large as the measured one.

The effect of the corona on the absorption of  $\text{SO}_2$  is clearly recognized, even when the current of electricity between the cylindrical net and the water electrode hardly flows.

From these results, the absorption of  $\text{SO}_2$  by the corona is not only dependent on the acceleration of the velocity of  $\text{SO}_2$  ions. According to the experiment by Wilson<sup>(14)</sup>, the top of a flame of a coal gas or a candle was extremely pulled to a negative plate. According to the experiments by Payne<sup>(15)</sup> et al, the gas was burned with the corona discharge in the cylindrical metal jacket heat exchanger which had a wire electrode along its axis. In the column, it was observed that ions moved radially from the center, the flame was pushed to the wall and gas flowed upward from the bottom producing a vortex. It is assumed that the gas flows through the column in the same way as Payne's experiment, though it was not confirmed experimentally. From the correlation between  $Re_g$  and  $K_{OG}$  in the absorption of  $\text{SO}_2$  in water, the effect of the corona discharge is very large even in the range of laminar flow,  $Re_g$  from 500 to 2300. It will be considered that the turbulence of the gas stream occurs in the column, when those ions produced by the corona violently collide with the unionized neutral molecules and are drifted to the wetted-wall under the influence of the electric field. The gas flows passing through the column with turbulence at the range of  $Re_g$  from 2300 to 6000, without a corona. And the acceleration of the absorption rate by the corona is seen at that range. But the effect of the corona can not be seen at  $Re_g$  more than 6000, for the turbulence by the corona in a column becomes relatively small by decreasing a gas phase resistance by the turbulent flow of gas in a column. In spite of the violent turbulence of a gas flow by the corona, the increase of  $K_{OG}$ , c.d. is not seen when the absorption rate of resistance. This fact shows that the corona does not affect  $\text{Cl}_2$  and  $\text{H}_2\text{S}$  in water is mainly controlled by the liquid phase resistance. This fact shows that the corona does not affect practically on the liquid phase resistance. Though almost all ions are captured by the cylindrical net in the absorption of  $\text{SO}_2$  in water with the corona, the rate of absorption is accelerated by a violent turbulence of the gas which was transferred to the outside through the meshes of the net. The rate of

(14) H.A. Wilson, Rev. Mod. Phys., **3** (1931), 156.

(15) K.G. Payne and F.J. Weinberg, Proc. Roy. Soc., **A250** (1959), 316.

absorption is accelerated by a negative, a positive and an A.C. corona, in spite of their different polarities and characteristics of the corona discharges. All these results clearly show that the acceleration of the rate of absorption is dependent on the decrease of the gas phase resistance by the turbulence of the gas which is induced by collision of gas with the corona discharge.

### Summary

(1)  $K_{OG}$ , c.d. in different conditions were measured and examined. The values of  $K_{OG}$  were largely affected by the flow rate of the gas and the concentration of  $\text{SO}_2$ . And, the temperature of the liquid gave a little change to  $K_{OG}$ , c.d.

(2) On the absorption of the gas in the liquid, the rates of the absorption were accelerated by the corona discharge in the following systems, which had the main resistance in the gas phase,  $\text{SO}_2\text{-H}_2\text{O}$ ,  $\text{NH}_3\text{-H}_2\text{O}$ ,  $\text{H}_2\text{S-NaOH}$ ,  $\text{Cl}_2\text{-NaOH}$  and  $\text{CO}_2\text{-NaOH}$ . On the contrary, the rate of absorption of  $\text{H}_2\text{S}$  and  $\text{Cl}_2$  in water which was mainly controlled by the liquid phase resistance is not accelerated by the corona discharge.

(3) It was considered that the acceleration of the rate of the absorption with the corona was caused by decrease of the gas phase resistance. At that time the stream of the gas was turbulent, because ions produced by the corona discharge repeated violent collisions with neutral molecules and moved toward the wetted-wall, and the neutral molecules were pushed toward the direction of ion movement.

### Nomenclature

A:	electric current	[ampere]
$D, D_l$ :	diffusion coefficient for gas phase and for liquid phase, respectively	$[\text{m}^2/\text{hr}]$ or $[\text{cm}^2/\text{sec}]$
$d$ :	inside diameter of wetted-wall column	[m] or [cm]
$e$ :	electronic charge ( $=1.602 \times 10^{-19}$ )	[A·sec]
$g$ :	acceleration by gravity	$[\text{m}/\text{hr}^2]$
$H$ :	Henry's constant	$[\text{kg-mole}/\text{m}^3 \cdot \text{atm}]$
$H_L$ :	(H.T.U.) height of transfer unit in liquid phase	[m]
$i$ :	electric current density	$[\mu\text{A}/\text{cm}^2]$ or $[\text{A}/\text{cm}^2]$
$k$ :	second-order reaction rate constant	$[\text{l}/\text{mole} \cdot \text{sec}]$
$k_g$ :	mass transfer coefficient for gas phase	$[\text{kg-mole}/\text{m}^2 \cdot \text{hr} \cdot \text{atm}]$
$k_l$ :	mass transfer coefficient for liquid phase	[m/hr]
$K_{OG}$ :	overall mass transfer coefficient	$[\text{kg-mole}/\text{m}^2 \cdot \text{hr} \cdot \text{atm}]$
$L$ :	superficial molar velocity of liquid	$[\text{kg-mole}/\text{hr} \cdot \text{m}^2]$
$N_{OG}$ :	number of transfer unit	[—]
$N_{\text{SO}_2^-}$ :	number of $\text{SO}_2$ ions absorbed per unit area per unit time	$[\text{SO}_2^-/\text{cm}^2 \cdot \text{sec}]$
$p_g$ :	partial pressure of solute gas in inlet stream	[atm]



$p^*$ : partial pressure of solute gas in equilibrium with liquid phase concentration	[atm]
$R$ : gas constant (0.08024)	[m <sup>3</sup> atm/kg-mole·°C]
$Re_L$ : Reynolds number of liquid ( $4\Gamma/\mu$ )	[—]
$Re_g$ : Reynolds number of gas ( $du\rho/\mu$ )	[—]
$T$ : absolute temperature	[°K]
$u$ : volumetric flow rate of gas divided by the conduit cross section	[m/hr]
$w$ : mass transfer rate	[kg-mole/hr]
$\beta$ : Hatta Number ( $\gamma/\tanh \gamma$ )	[—]
$\gamma$ : dimensionless parameter ( $\sqrt{kD_e/k_e}$ )	[—]
$\Gamma$ : mass flow rate of liquid per unit perimeter	[kg/m·hr]
$\mu$ : viscosity of gas	[kg/m·hr] or [C.P.]
$\rho$ : gas density	[kg/m <sup>3</sup> ]