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Mass Transfer Characteristics and Overall Mass Transfer Coefficient in the Ozone Contactor

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Abstract−The overall mass transfer coefficient $k_{La,F}$ in the flow characteristics was determined by the measurement of the diffusivity of ozone, density of aqueous solution, and viscosity. However, the measured values $k_{La,F}$ in the range of 0.0096-0.0622 min^{-1} show large changes in hydraulic retention time, and the dissolved ozone concentration $C_{L,F}$ presented under 0.1 mg/*l* is lower than the dissolved ozone observed. The overall mass transfer coefficient $k_{La,M}$ in the ozone decomposition was determined by measurement of the equilibrium dissolved ozone, overall decomposition rate constant, and overall Henry's law constant. The measured values $k_{La,M}$ are in the range of 0.0441-0.0749 min^{-1}, and they present small changes depending on the hydraulic retention time. Furthermore, the measured dissolved ozone concentration $C_{L,M}$ presents a larger value than the $C_{L,F}$. Then, the $k_{La,M}$ is selected as an input overall mass transfer coefficient to predict the dissolved ozone requirement in the ozone contactor.

Key words: Ozone Contactor, Overall Mass Transfer Coefficient, Henry's Law Constant, Decomposition Rate Constant, Gas Holdup

INTRODUCTION

Many researchers have measured and improved on the overall mass transfer coefficient of oxygen, acicular goethite, and ozone, using various reactors [Kang et al., 1986, 1990; Koh et al., 1989; Yoon, 1999]. A bubble column has been largely applied to the field of chemical engineering because it needs only small amounts of working expenses and shows a big mass transfer coefficient between the phases [Kang et al., 1990]. The purpose of this paper is to determine the overall mass transfer coefficient using the methods that consider reactor flow characteristics and ozone decomposition characteristics in the ozone contactor. The ozone contactor in this study is a bubble column that has a continuous liquid phase and dispersed gas phase where the factors of reactor performance are the size of the bubble, gas holdup, and flow characteristics [Kang et al., 1986, 1990; Koh et al., 1989; Choi and Lee, 1992; Choi, 2001; Lee and Lee, 2002; Lee et al., 2003]. The $k_{La,F}$ in the flow characteristics is measured by the liquid mass transfer coefficient and the specific interfacial area. The specific interfacial area can be determined by the bubble size and gas holdup, which depends on the gas velocity a major factor for the $k_{La,F}$. The values of $k_{La,M}$ in the ozone decomposition are estimated by using a trial and error method where the overall decomposition rate constant k_{OD} and Henry's law constant H_0 were determined by the multiple regression analysis empirical equation. The selected overall mass transfer coefficient $k_{La,M}$ was inputted to predict the dissolved ozone requirement in the ozone contactor.

EXPERIMENTAL METHODS

The ozone contactor used in the continuous experiment is shown in Fig. 1. The ozone generator is a silent electrical discharger that has a water cooling system at the outside of the discharge tube and filter, which is a moisture filter and is used to remove nitric acid caused by humidity. The ozone contactor is made of acryl and its height and diameter are 2,000 mm and 100 mm relatively. The measurements of the flow of water and air are done by a water meter and flow meter, Dwyer, USA, respectively. The model of the sample loading pump is PH-0430D and its total pump head is 4.5 m, rated power consumption 95 Watts. In addition, it has a drain valve to control the flow level. The ozone off gas can be measured in the absorption bottle with 400 ml of 2% potassium iodide solution [Clesceri et al., 1985; Yoon et al., 1999]. The pictures of the size and number of the air bubbles are taken by ASA 1600 high-resolution film with a shutter rate of 1/60 second. The flows in the ozone contactor are the plug flow of up-flow for the ozone and axial dispersion flow of

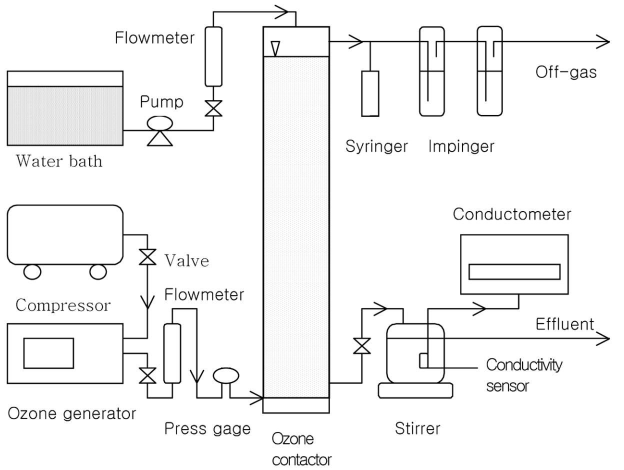

Fig. 1. The schematic diagram of the experimental apparatus.

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Table 1. Conditions of the determination of dispersion number, dispersion intensity, and the $k_{La,F}$

Item / Run	Water flowrate (L/hr)	Water velocity (m/hr)	HRT (min)	Gas flowrate (L/hr)	Gas velocity (m/hr)	Gas flowrate/Water flowrate (G/L)
I	200.0	40.0	3	10.0	1.8	0.05
				20.0	3.6	0.10
				30.0	5.4	0.15
				40.0	7.2	0.20
				50.0	9.0	0.25
II	120.0	24.0	5	6.0	1.2	0.05
				12.0	2.4	0.10
				18.0	3.6	0.15
				24.0	4.8	0.20
				30.0	6.0	0.25
III	80.0	17.0	7	4.0	0.8	0.05
				8.0	1.6	0.10
				12.0	2.4	0.15
				16.0	3.2	0.20
				20.0	4.0	0.25
IV	70.0	13.0	9	3.5	0.7	0.05
				7.0	1.4	0.10
				10.5	2.1	0.15
				14.0	2.8	0.20
				17.5	3.5	0.25
V	50.0	10.0	12	2.5	0.5	0.05
				5.0	1.0	0.10
				7.5	1.5	0.15
				10.0	2.0	0.20
				12.5	2.5	0.25

Table 2. Conditions of the determination of the dissolved ozone concentrations and the $k_{La,M}$

Item / Run	Gas ozone conc. (mg/L)	Ozone dose (mgO_3/L)	HRT (min)	Gas flowrate/ Water flowrate (G/L)
VI	2.3	0.5	9	0.2
	3.5	0.7		
	4.5	0.9		
	6.0	1.1		
VII	4.5	0.9	3	0.2
			5	
			7	
			9	
			12	
VIII	7.3	1.1	9	0.15
	5.5			0.20
	4.4			0.25
	3.7			0.30

down-flow for the aqueous solution. The dispersion numbers (tracer: NaCl, 1 M) were measured by the following method. The characteristics of the ozone contactor as a function of G/L ratio for a fixed HRT were measured, and then the dispersion number depending on the hydraulic retention time was determined from the FORTRAN program by a trial and error method. The operating conditions of the ozone contactor to determine the $k_{La,F}$ are shown in Table 1. The dissolved ozone concentration depending on the height of the ozone contactor is presented in Table 2. Run VII is the operating condition to determine the $k_{La,M}$.

DETERMINATION OF THE OVERALL MASS TRANSFER COEFFICIENT

The overall mass transfer coefficient in the ozone contactor is a steady state laminar flow as an axial dispersion, and it has assumptions as follows.

(a) Pressures are linearly changed by the height of contactor, and the gas holdup and interface has a constant value.
(b) The resistance of mass transfer on the ozone absorption is limited to the axis of liquid phase and is not increased by the ozone depletion. The rate of the ozone depletion is the first order reaction in liquid phase but is to be neglected in the gaseous phase.
(c) Henry's law is applied.

The overall mass transfer coefficient was determined by the mass transfer coefficient of the liquid phase in the flow characteristics of reactor and mass balance in the decomposition of the aqueous solution. The overall mass transfer coefficient $k_{La,F}$ can be produced by multiplying the liquid mass transfer coefficient and the specific interfacial area. The liquid mass transfer coefficient and viscosity can be calculated by Eqs. (3) and (4), respectively [Bingham, 1922; Danckwerts, 1970].

$$k_{La}=k_L\times a \tag{1}$$

$$a=\frac{6\varepsilon_g}{d_b(1-\varepsilon_g)} \tag{2}$$

$$\frac{k_L\times L}{D}=N_{Sh}=0.323N_{Re}^{1/2}N_{Sc}^{1/3} \tag{3}$$

$$\frac{1}{\mu_L}=2.15[(T-8.44)+\sqrt{8078.4+(T-8.44)^2}]-120 \tag{4}$$

where d_b is the average diameter of the bubble.

The method that is used to produce the overall mass transfer coefficients is a trial and error method, which are determined by the changing rates of concentration depending on the hydraulic retention time for the dissolved ozone [Carphentier, 1981; Sotelo et al., 1989; Yoon, 1999; Zhou and Smith, 2000; Bewtra and Nicholas, 1970].

$$\frac{dC_L}{dt}=k_{La,M}(C_L^*-C_L)-k_{OD}C_L \tag{5}$$

$$C_L^*-C_L=C_L^*e^{-k_{La,M}t}+\frac{k_{OD}C_L}{k_{La,M}}(e^{-k_{La,M}t}-1) \tag{6}$$

$$k_{La(T)}=k_{La(20)}\times 1.024^{T-20} \tag{7}$$

FLOW PATTERN AND DISSOLVED OZONE

The dispersion number presents the flow characteristics of the reactor, and the large value shows a completely mixed flow. If the value is small, it becomes a plug flow that improves the efficiency of the reactor. Therefore, the dispersion number will affect the efficiency for the treatment of an objective. That is, if the optimum ratio of G/L is determined for the small value of dispersion number, the efficiency of the reactor can be increased. The experimental result of the dispersion number in the ozone contactor is presented in Table 3. The dispersion number according to the hydraulic retention time (HRT) in the ozone contactor as the function of the ratio of G/L is presented in Fig. 2. The dispersion number is directly proportional to the increment of HRT and G/L ratio, and it is 0.03-0.13. The intensity of dispersion can be expressed as the parameter which measures the extent of axial dispersion.

Table 3. Dispersion number in the ozone contactor

Run \ Parameters	G/L	HRT (min)	θ	σ^2	σ_θ^2	$E(\theta)_{max}$	D_L/U_LL
I	0.0	3.0	1.0	0.33	0.076	0.54	0.04
	0.05		0.8	0.52	0.103	0.43	0.05
	0.10		0.7	0.88	0.175	0.35	0.10
	0.15		0.7	1.04	0.182	0.34	0.10
	0.20		0.7	1.19	0.205	0.42	0.12
	0.25		0.5	1.70	0.278	0.30	0.17
II	0.0	5.0	0.9	1.20	0.09	0.30	0.05
	0.05		0.6	2.35	0.17	0.24	0.09
	0.10		0.6	2.95	0.21	0.22	0.12
	0.15		0.6	3.70	0.23	0.21	0.14
	0.20		0.5	3.85	0.24	0.21	0.14
	0.25		0.5	4.75	0.30	0.20	0.18
III	0.0	7.0	0.7	2.01	0.08	0.27	0.04
	0.05		0.6	4.81	0.17	0.19	0.10
	0.10		0.5	5.21	0.19	0.19	0.11
	0.15		0.4	7.27	0.24	0.17	0.14
	0.20		0.4	7.30	0.25	0.19	0.12
	0.25		0.4	7.38	0.25	0.18	0.15
IV	0.0	9.0	0.9	3.95	0.09	0.24	0.05
	0.05		0.6	7.00	0.15	0.14	0.08
	0.10		0.5	9.26	0.18	0.15	0.10
	0.15		0.4	9.35	0.19	0.17	0.11
	0.20		0.4	11.3	0.23	0.15	0.14
	0.25		0.4	11.4	0.24	0.15	0.14
V	0.0	12.0	0.7	8.8	0.11	0.24	0.06
	0.05		0.7	11.5	0.15	0.14	0.08
	0.10		0.5	16.9	0.20	0.13	0.11
	0.15		0.4	18.5	0.22	0.13	0.12
	0.20		0.4	20.0	0.24	0.13	0.14
	0.25		0.4	21.7	0.25	0.15	0.15

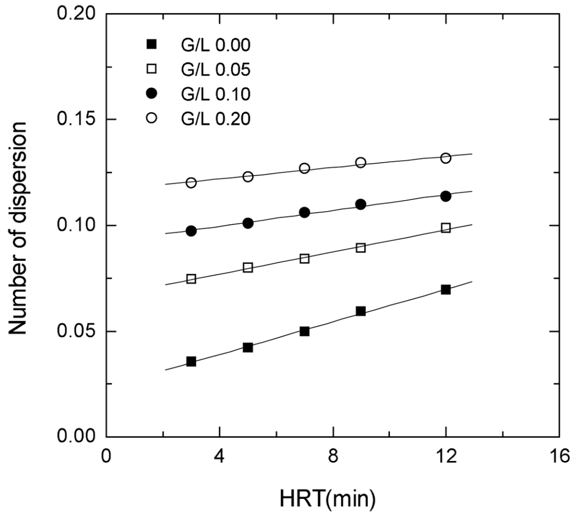

Fig. 2. Dispersion number according to the HRT (tracer: NaCl, 1 M).

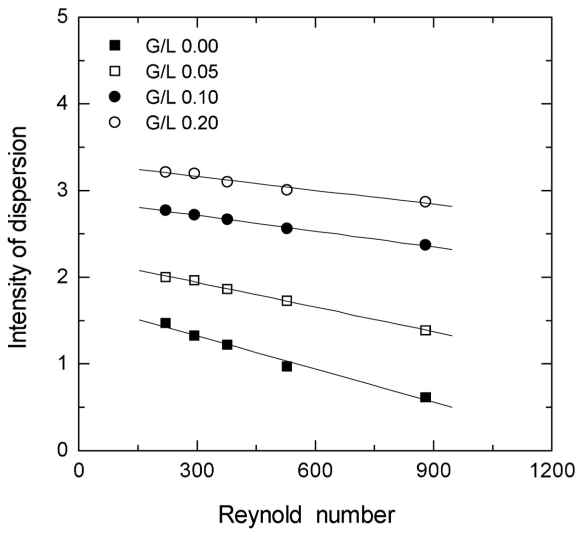

Fig. 3. Dispersion intensity according to the Reynolds number.

$$\text{Number of dispersion} = \frac{D_L}{U_L L} \quad (8)$$

The dispersion number was from 0.025 to 0.13. Then the flow pattern might be close to the plug flow pattern with an intermediate amount of dispersion. The dispersion number shows a smooth value after HRT 9 minutes and 0.2 for the ratio of G/L. The ratio of G/L and HRT is determined by the minimum value of the effective operation of the reactor, and then the conditions for the optimum operation of the ozone contactor are determined as HRT 9 minutes and 0.2 for the ratio of G/L.

The dispersion intensity of the fluid using the function of the Reynolds number and the ratio of G/L under the conditions of Run from to in Table 1 is shown in Fig. 3. The intensity of dispersion can be expressed as follows:

$$\text{Intensity of dispersion} = \frac{D_L}{U_L d_R} \quad (9)$$

where D_L is the axial dispersion coefficient of ozone, U_L is the superficial liquid velocity and d_R is the diameter of the ozone contactor. The dispersion intensity is decreased by the increment of the Reynolds number, and it is increased by the increment of the ratio of G/L. The dispersion intensity is 0.6-3.2 which is affected by the ratio of G/L rather than that of the Reynolds number. Because the dispersion intensity is more affected by dispersion coefficient than by

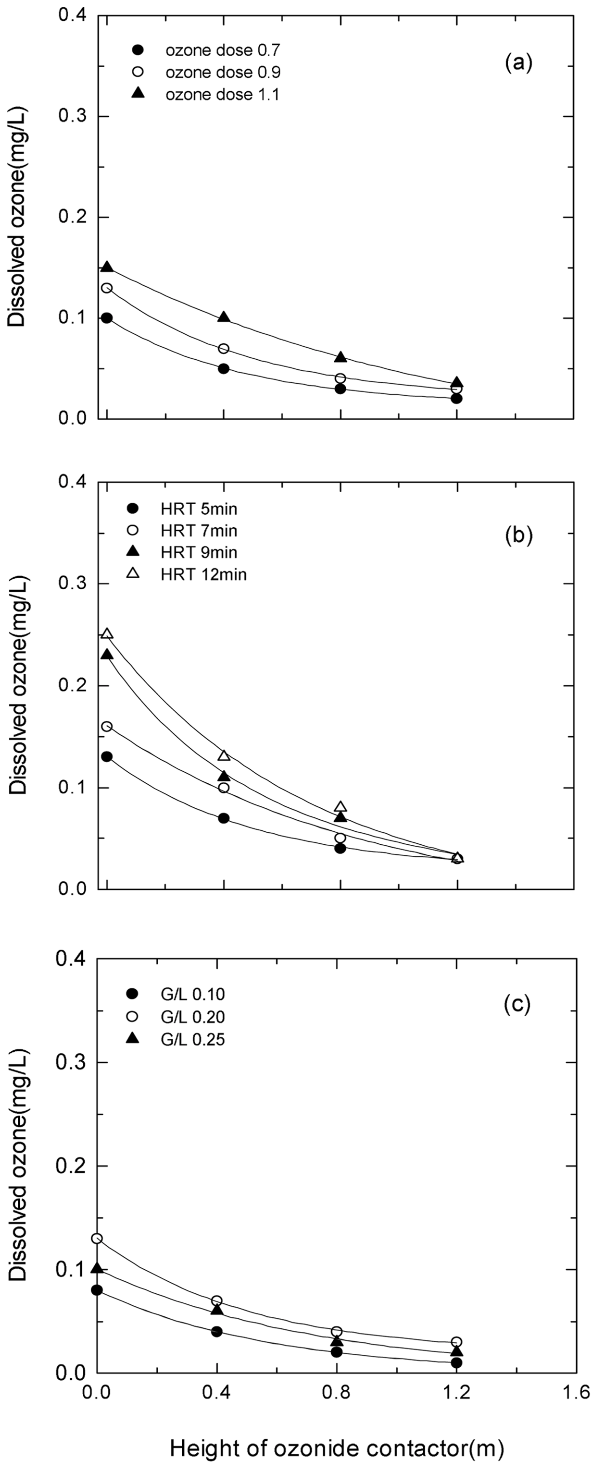

Fig. 4. Dissolved ozone according to the ozone contactor (a) ozone dose (b) HRT (c) G/L ratio.

water velocity, the dispersion coefficient is affected by advection owing to the G/L ratio rather than Reynolds number. The dispersion intensity that has the ratio of G/L by 0.05 is 0.6-1.5, and it is mainly transferred by the molecular diffusion. In the case of the ratio of G/L by 0.1, the dispersion intensity is 2.4-2.8, which is mainly transferred by the molecular diffusion and advection [Yoon, 1999].

The dissolved ozone is an input parameter to estimate the value of $k_{La,M}$ in the equation of mass balance. The measured values for the height of the contactor using the function of the ozone dose, HRT and ratio of G/L under the conditions of Run from VI to VIII in Table 2 are presented in Fig. 4. The pH is 7.0, water temperature is 20 °C, TOC is 0.04 mg/L, Alkalinity is 0mg $CaCO_3$/L, and ionic strength is zero. The dissolved ozone (mg/*l*) is increased by the conditions of the parameters, such as the lower height of the contactor, large of ozone dose (mg/*l*), longer HRT, and the increment of the ratio of G/L [Yoon, 1999; Park et al., 2001; Rhim, 2003]. The rate of increase for the dissolved ozone is significantly increased by the ozone dose of 0.90 mg/*l*, 9 minutes of HRT, and 0.2 of the ratio of G/L, and then it is almost constant after the conditions are met. However, the dissolved ozone is decreased at 0.25 of the ratio of G/L. The dissolved ozone is increased less than 0.2 of the ratio of G/L because the mass transfer is increased by the effect of the size and numbers of the ozone bubble depending on the increment of the ratio of G/L. But it is decreased more than 0.25 of the ratio of G/L because the mass transfer is reduced by such factors as the lower concentration of ozone in the bubble, decrement of the concentration gradient for the water solution under the constant ozone dose even though the specific interfacial area is increased by the increment of the number of bubbles. Therefore, the optimum conditions to preserve the dissolved ozone are assumed to be HRT 9 minutes, 0.20 of the ratio of G/L [Yoon, 1999].

THE $k_{La,F}$ IN THE FLOW CHARACTERISTICS

The mass transfer coefficient in the flow characteristics is determined by the liquid mass transfer coefficient and the specific interfacial area. The parameters and values required to calculate the $k_{La,F}$ are shown in Table 4. The diffusivity of ozone in water was determined by Eq. (10) of the Wilke-Chang formulas:

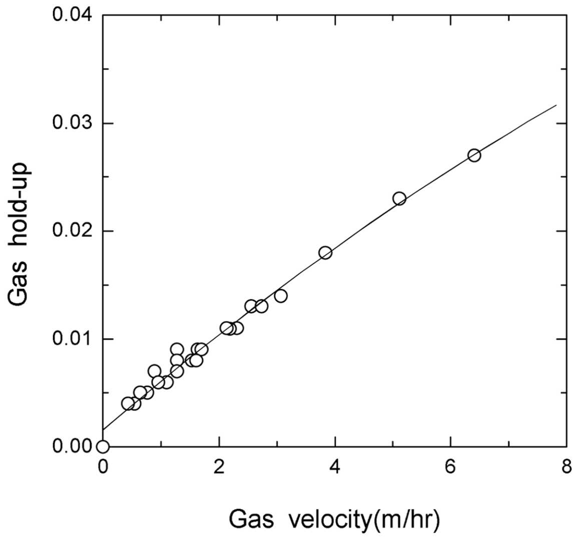

Fig. 5. Effect of gas velocity on gas holdup.

$$\frac{D\times\mu_L}{T}=\frac{7.4\times10^{-8}(\xi M_L)^{1/2}}{V^{0.5}} \tag{10}$$

where ξ is the association factor of ozone, M_L is the molecular weight of water, and V is the molar volume of ozone at its normal boiling temperature. The specific interfacial area can be determined by the bubble size and gas holdup, which depends on the gas velocity a major factor for the $k_{La,F}$ [Kang et al., 1986]. The effect of gas velocity on gas holdup under the conditions of Run from I to V in Table 1 is shown in Fig. 5. The coefficient of determination is 0.986, and the standard deviation is 0.000752. As a result, it is considered that a small eddy is increased by the increment of the energy distinction rate, and then the $k_{La,F}$ is increased by the increment of gas-liquid contact frequency [Kang et al., 1986; Yoon, 1999]. The size of the bubble is 0.1-0.2 cm, and the average size is 0.15 cm. The specific interfacial area is 0.4-1.3 cm^{-1}. The value of $k_{La,F}$ is 0.0096-0.0622 min^{-1}, and the $C_{L,F}$ verified by the mass balance is under

Table 4. Results of the overall mass transfer coefficient $k_{La,F}$

Parameters and units \ HRT (min)	3	5	7	9	12
gas velocity (U_g: cm/sec)	0.200	0.133	0.089	0.078	0.056
water velocity (U_L: cm/sec)	1.110	0.670	0.480	0.370	0.280
$N_{Re}\left(\frac{d_R\rho_L U_L}{\mu_L}\right)$	1.109	663.3	475.2	366.3	277.2
$N_{Sc}\left(\frac{\mu_L}{\rho_L D}\right)$	273.7	273.7	273.7	273.7	273.7
$N_{sh}(0.323N_{Re}^{1/2}N_{Sc}^{1/3})$	69.63	53.89	45.72	40.14	34.92
$k_L\left(\frac{D\cdot N_{Sh}}{L}: 10^{-3}\ \text{cm/sec}\right)$	1.040	0.545	0.320	0.249	0.160
$\varepsilon_g(0.00406U_g+0.00201)$	0.0312	0.0215	0.0150	0.0134	0.010
$a\left(\frac{6}{d_b}\frac{\varepsilon_g}{1-\varepsilon_g}: \text{cm}^{-1}\right)$	1.290	0.880	0.610	0.540	0.400
$k_{La,F}(k_L\times a: \text{min}^{-1})$	0.0622	0.0327	0.0192	0.0149	0.0096

0.1 mg/l and is lower than the observed dissolved ozone.

THE $k_{La,M}$ IN THE OZONE DECOMPOSITION

The value of $k_{La,M}$ is measured by the mass balance in the ozone decomposition:

(a) The overall decomposition rate constant and the overall Henry's law constant were predicted by the multiple regression analysis Eqs. (11) and (12) [Yoon, 1999].

(b) The equilibrium dissolved ozone concentration can be measured by Henry's law equation.

(c) The value of $k_{La,M}$ was determined by using a trial and error method by means of the substitution of the factors of (a), (b) and observed dissolved ozone into Eq. (6).

(d) The values of $k_{La,F}$ and $k_{La,M}$ verified.

To measure the value of k_{OD} and H_0, the test water is used by the prescribed water, that is, a mixture of pure water and humic acid. The pH is 7.0, temperature is 20 °C, TOC is 0.158 mg/L, alkalinity is 10 mg/l, and ionic strength is zero. The value of C_L^* is the equilibrium dissolved ozone concentration and is produced by the substitution of the partial pressure of the dissolved ozone (0.11 kPa, 1.086×10^{-3} atm) and Henry's law constant (496,021 kPa/mol$_f$). In addition, the value is converted into the unit of mg/l after calculating the mole (1.23×10^{-5}) of C_L^* [Yoon, 1999; Rhim, 2003].

$$\log k_{OD}=0.053[\mathrm{pH}]+0.018[\mathrm{Temp}]+0.20\log[\mathrm{I.S}]-0.33\log[\mathrm{Alk}]+0.43\log[\mathrm{TOC}]-2.6 \quad (11)$$

$$\ln H_0=0.23\ln[\mathrm{pH}]+0.57\ln[\mathrm{Temp}]+1.40[\mathrm{I.S}]+0.024[\mathrm{Alk}]+0.053[\mathrm{TOC}]+10.71 \quad (12)$$

$$p=H_0C_L^* \quad (13)$$

The overall mass transfer coefficient using the mass balance in the ozone decomposition is presented in Table 5. The values of $k_{La,M}$ (min^{-1}) are 0.0441-0.0749, and they present small changes depending on the hydraulic retention time.

The relationship of the dissolved ozone between the values of $k_{La,F}$ and the values of $k_{La,M}$ presented in Table 4 and 5 relatively is shown in Fig. 6. The values of $C_{L,F}$ are decreased depending on the hydraulic retention time, and the verified ozone concentration, which is under 0.1 mg/l, is lower than the observed values.

Therefore, the values of $k_{La,M}$, which are similar to the dissolved ozone verified by the mass balance, are selected as the inputted overall mass transfer coefficient to predict the requirements of the dissolved ozone in the ozone contactor [Yoon et al., 1999].

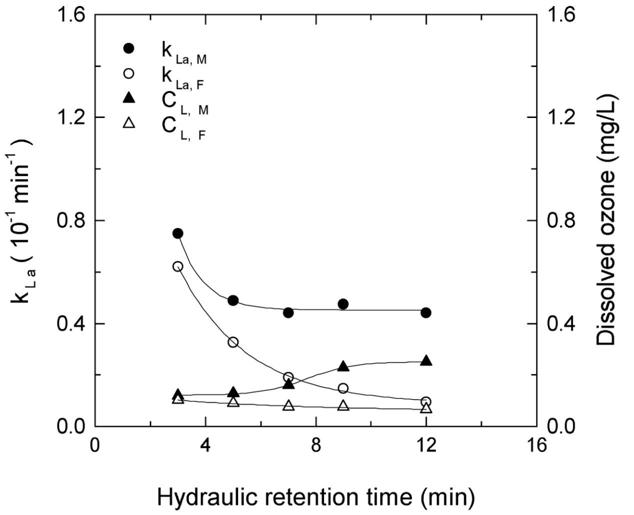

Fig. 6. The values of k_{La} and dissolved ozone depending on the hydraulic retention time.

CONCLUSIONS

The following conclusions are reached on the basis of the results of this research.

The individual mass transfer coefficient k_L was deduced experimentally. The value obtained was 0.16×10^{-3}-1.04×10^{-3} cm/sec depending on the hydraulic retention time of 3-12 minutes. The specific interfacial area was determined by the bubble size and gas hold-up, and the value was 0.4-1.29 cm^{-1}. The values of $k_{La,F}$ and $C_{L,F}$ in the flow characteristics of ozone contactor were 0.0096-0.0622 min^{-1} and 0.06-0.10 mg/l, respectively. The overall decomposition rate constant k_{OD} and the overall Henry's law constant H_0 were determined by the multiple regression analysis empirical equation. The values of k_{OD} and H_0 were 2.86×10^{-2} min^{-1} and 496,021 kPa/mol$_f$, respectively. The $k_{La,M}$ and $C_{L,M}$ in the decomposition of ozone were 0.0441-0.0749 min^{-1} and 0.12-0.25 mg/l, respectively. The $C_{L,F}$ decreased depending on the hydraulic retention time, and the verified value was lower than the measured dissolved ozone. The values of $C_{L,M}$, were similar to the dissolved ozone verified by the mass balance. Therefore, the values $k_{La,M}$ were selected as the overall mass transfer coefficient to predict the dissolved ozone demand in the ozone contactor.

Table 5. Determination of the overall mass transfer coefficient in the ozone contactor

HRT (min) / Parameters and units	3	5	7	9	12
k_{OD}(min^{-1})			2.86×10^{-2}		
C_0(mg/L)			0.000		
C_L^*(mg/L)			0.592		
C_L(mg/L)	0.120	0.130	0.160	0.210	0.250
$k_{La,M}$(min^{-1})	0.0749	0.0489	0.0441	0.0476	0.0441

NOMENCLATURE

a : specific gas-liquid interfacial area [cm^{-1}]
C_0 : initial ozone concentration of liquid phase [mg/l]
C_L : ozone concentration of liquid phase [mg/l]
$C_{L,F}$: dissolved ozone concentration considered in flow characteristics [mg/l]
$C_{L,M}$: dissolved ozone concentration considered in decomposition characteristics [mg/l]
C_L^* : equilibrium ozone concentration of liquid phase [mg/l]
D : molecular diffusion coefficient of ozone [cm^2/sec]
D_L : axial dispersion coefficient of ozone [cm^2/sec]
d_b : average diameter of the bubble [cm]
d_R : diameter of the ozone contactor [cm]
H_0 : Henry's law constant [kPa/mol_f]
k_L : individual mass transfer coefficient of liquid phase [cm/sec]
k_{La} : overall volumetric mass transfer coefficient of liquid phase [sec^{-1}]
$k_{La(T)}$: overall volumetric mass transfer coefficient in T [sec^{-1}]
k_{OD} : overall decomposition rate constant [sec^{-1}]
L : column height [cm]
M_L : molecular weight of water [g]
N_{Re} : Reynolds number [-]
N_{Sc} : Schmidt number [-]
N_{Sh} : Sherwood number [-]
T : temperature [^oC or K]
t : hydraulic retention time of water [sec or min]
U_g : superfacial gas velocity [cm/sec]
U_L : superfacial liquid velocity [cm/sec]
V : molar volume of ozone at its normal boiling temperature [cm^3/mol]

Greek Letters

ε_g : gas holdup [-]
μ_L : viscosity [g/cm/sec]
ξ : association factor of ozone [-]
ρ_L : specific weight of the liquid [g/cm^3]

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