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APPLYING MASS TRANSFER MODELS FOR CONTROLLING ORGANIC COMPOUNDS IN OZONATION PROCESS

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Mass transfer plays a significant role in the ozonation process. The prediction models associated with the volumetric overall mass transfer coefficient ($K_{L,a}$) and initial fractional ozone absorption (FOA_0) during the ozonation process were developed through the use of dimensional analysis. It was found that the volumetric overall mass transfer coefficient is the function of diffusivity, agitation speed, and gas flow rate, and the parameters in the $K_{L,a}$ equation are determined. Application of the prediction models for $K_{L,a}$ and FOA_0 would yield information to choose the most practically feasible operating parameters. The removability of total organic carbon (TOC) can be estimated based on the mass balance relationship and kinetic expression of TOC oxidation, during continuous laboratory ozonation of humic acid solution. The reaction rate constant averaged 0.0291 L/mg·min. The developed model in combination with the mass transfer and reaction kinetics can be used successfully in forecasting the most efficient agitation speed to control the formation of organic compounds. Also, the critical value of ozone partial pressure to achieve the highest TOC removability can be determined through the use of the above developed model. ©1997 Elsevier Science Ltd

INTRODUCTION

In Taiwan, chlorination is the most extensively used technique for disinfecting drinking water in the water treatment process. However, disinfection of water by chlorine can lead to the formation of organic halides and disinfection by-products, some of which have adverse toxicological effects on human health. There is an increasing interest in the use of ozone because it has a higher oxidation potential for reduction of dissolved organic substances than chlorine. During the ozonation process, ozone gas is usually brought into continuous

contact with water under dynamic conditions. Since mass transfer of ozone is generally the rate-determining-step concerning the mechanisms of ozone reaction, factors including water qualities and operational parameters affecting mass transfer should be examined.

The mass transfer rate is influenced by operating variables such as pH, temperature, agitation speed, and gas flow rate (Sotelo et al. 1989). According to Sheffer and Esterson (1982), $K_{L,a}$, the volumetric overall mass transfer coefficient of ozone in tap water, was about 1.14 min⁻¹ and reached a higher value at a higher water flow rate. There was a large decrease in $K_{L,a}$ to 0.250 min⁻¹ without agitation. Sheffer and Esterson (1982) reported that there was a linear relationship.

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 NOMENCLATURE

a	Specific interfacial surface area (cm^{-1})
b,c,d,e,f,g	Constants in Eq. 3 (dimensionless)
d_i	Diameter of agitation blades (cm)
Dt	Tank diameter (cm)
D_v	Diffusivity (cm^2s^{-1})
E	Height of impeller above vessel floor (cm)
E	Enhancement factor (dimensionless)
G	Gas flow rate ($\text{L}\cdot\text{min}^{-1}$, except for Eq. 1)
H	Depth of liquid in vessel (cm)
H	Henry's law constant ($\text{kPa}\cdot\text{L}\cdot\text{mg}^{-1}$)
h, x	Constants in Eq. 6 (dimensionless)
J	Width of baffles (cm)
K	Reaction rate constant of TOC with O_3 ($\text{L}\cdot\text{mg}^{-1}\text{min}^{-1}$)
K_L	Overall mass transfer coefficient ($\text{cm}\cdot\text{min}^{-1}$)
k_L	Liquid-phase mass transfer coefficient ($\text{cm}\cdot\text{min}^{-1}$)
$K_L a$	Volumetric overall mass transfer coefficient (min^{-1} , except for Eq. 1)
$k_L a$	Volumetric liquid-phase mass transfer coefficient (min^{-1})
L	Length of impeller blades (cm)
N_p	Power number (dimensionless)
n	Agitation speed (s^{-1})
P_0	Power consumption in ungasged liquid (watt)
P_g	Power consumption with gas dispersion (watt)
P_{O_3}	Ozone partial pressure (kPa)
Q	Liquid flow rate ($\text{L}\cdot\text{min}^{-1}$)
r_{O_3}	Reaction rate of ozone ($\text{mg}\cdot\text{L}^{-1}\text{min}^{-1}$)
$r_{\text{O}_3,\text{sd}}$	Self-decomposition rate of ozone ($\text{mg}\cdot\text{L}^{-1}\text{min}^{-1}$)
T	Temperature (K)
t	Time (min)
t_R	Hydraulic retention time (min)
V	Volume of liquid phase (L)
W	Impeller width (cm)
[TOC]	Concentration of TOC (mg/L)
$[\text{O}_3]$	Ozone concentration in liquid phase (mg/L)
$[\text{TOC}]_{\text{in}}$	Influent TOC concentration (mg/L)
$[\text{TOC}]_{\text{eff}}$	Effluent TOC concentration (mg/L)
$[\text{O}_3]^*$	Equilibrium dissolved ozone concentration (mg/L)

Greek letters

α	Ratio of P_g over P_0 (dimensionless)
μ	Viscosity ($\text{g}\cdot\text{cm}^{-1}\text{s}^{-1}$)
ρ	Density ($\text{g}\cdot\text{cm}^{-3}$)

between the ozone feed rate and ozone residual. Water flow rate, gas flow rate, and temperature determined the slope and the intercept of the linear function.

Some semi-empirical regressions of $K_L a$ have been reported (Grasso 1987; Ouederni et al. 1987; Stankovic

1988). Ouederni et al. (1987) proposed the following equation to relate the ozone transfer in stirred reactors:

$$K_L a = 2.57 \times 10^{-2} \times (G \times n)^{0.67} \quad (1)$$

where,

$K_L a$ is expressed in s^{-1} ;

G is the gas flow rate expressed in $10^{-3} m^3/s$; and,

n is the number of rotations per second.

Previous investigators (Roth et al. 1982; Gurol 1985; Benitez et al. 1991) incorporated mass transfer and kinetics with operating parameters for scale-up of the ozonation process and to study the removal efficiency of the target compound. The resulting model proposed by Roth et al. (1982) included the mass transfer intrusion originating from the diffusional resistance of ozone and the complex kinetics encountered during ozonation, and can be applied to both semi-batch mode and CSTR mode operations. Benitez et al. (1991) studied the reaction between ozone and MCPA, a pesticide included in the group of the phenoxyalkyl acid derivatives, and concluded that both the gas flow rate and the agitation speed had a positive effect on the MCPA degradation.

The main objective of this study was to develop the prediction models associated with the mass transfer characteristic of the ozonation system, and to choose the most practically feasible operating parameters to reduce organic compounds. First, the prediction equation of $K_L a$ was derived through the use of dimensional analysis. Then, the parameters in the equation were determined using the experimental data. The removability of total organic carbon (TOC) was estimated based on the mass balance relationship and kinetic expression of TOC oxidation, during the continuous laboratory ozonation of humic acid solution. Also, the reaction rate constant was determined from the experimental data. In conjunction with the semi-empirical equation of $K_L a$ and TOC reaction kinetics, this ozone mass transfer model can provide the most efficient agitation speed required to control the formation of organic compounds.

MATERIALS AND METHODS

Ozone contacting system

As shown in Fig. 1, a laboratory-scale ozonation system, including a 15 L stainless steel reactor, a cylindrical vessel with a built-in diffuser for ozone introduction, a flat-bladed turbine with six blades, and four internal baffles to facilitate mixing was used in this investigation. The design of reactor was based on the criteria of shape factors for a standard six-bladed turbine (McCabe et al. 1985): $d_i/D_t=1/3$, $E/d_i=1$, $L/d_i=1/4$, $W/d_i=1/5$, $J/D_t=1/12$, and $H/D_t=1$, as shown in Fig. 1. Ozone generated from pure oxygen, along

with the carrier gas, was admitted to the reactor in a semi-batch or continuous mode. All fittings, valves, and tubing were of stainless steel, glass, or Teflon.

Analytical methods

TOC analysis was performed using a TOC analyzer (Model 700, O.I. Corp.) and calibrating with potassium hydrogen phthalate. Dissolved ozone concentration was determined by the indigo method (Bader and Hoigne 1981), while the gaseous ozone concentration was quantified by absorbing in potassium iodide traps and subsequently titrating with sodium thiosulfate solution.

Experimental methods

Before starting absorption and/or reaction, the ozone-oxygen stream was directed into a KI solution to analyse gaseous ozone concentration. When the desired ozone concentration was reached, the gas stream was fed into the reactor. In the physical absorption experiments, the phosphate salts (disodium hydrogen phosphate and potassium dihydrogen phosphate) were added into deionized water (Milli-Q SP) to maintain a pH of 5 or 7 and ionic strength of 0.015 M. The absorption process was carried out, in a semi-batch mode, until a steady state concentration of dissolved ozone was reached. The synthetic samples prepared by humic acids (Wako Pure Chemical Industries) and phosphate salts (the same as the physical absorption experiments) were introduced to run the TOC reduction tests, which were conducted in a continuous mode.

RESULTS AND DISCUSSION

Model development

Semi-empirical equation of $K_L a$. The following assumptions were made to derive the appropriate model for prediction of $K_L a$:

- 1) Higbie's penetration theory is applied to correlate the relationship between the overall mass transfer coefficient, K_L , and diffusivity.
- 2) Liquid-phase mass transfer coefficient, k_L , is equal to overall mass transfer coefficient, K_L , when the gas-side resistance is comparatively neglected.
- 3) Influent gas flow rate is irrelevant to the overall mass transfer coefficient, K_L .
- 4) Temperature and agitation speed are irrelevant to a, the specific interfacial surface area.

Although the agitation speed and temperature may influence the gas bubble diameter, and therefore the specific interfacial area, the dependence of the specific inter-

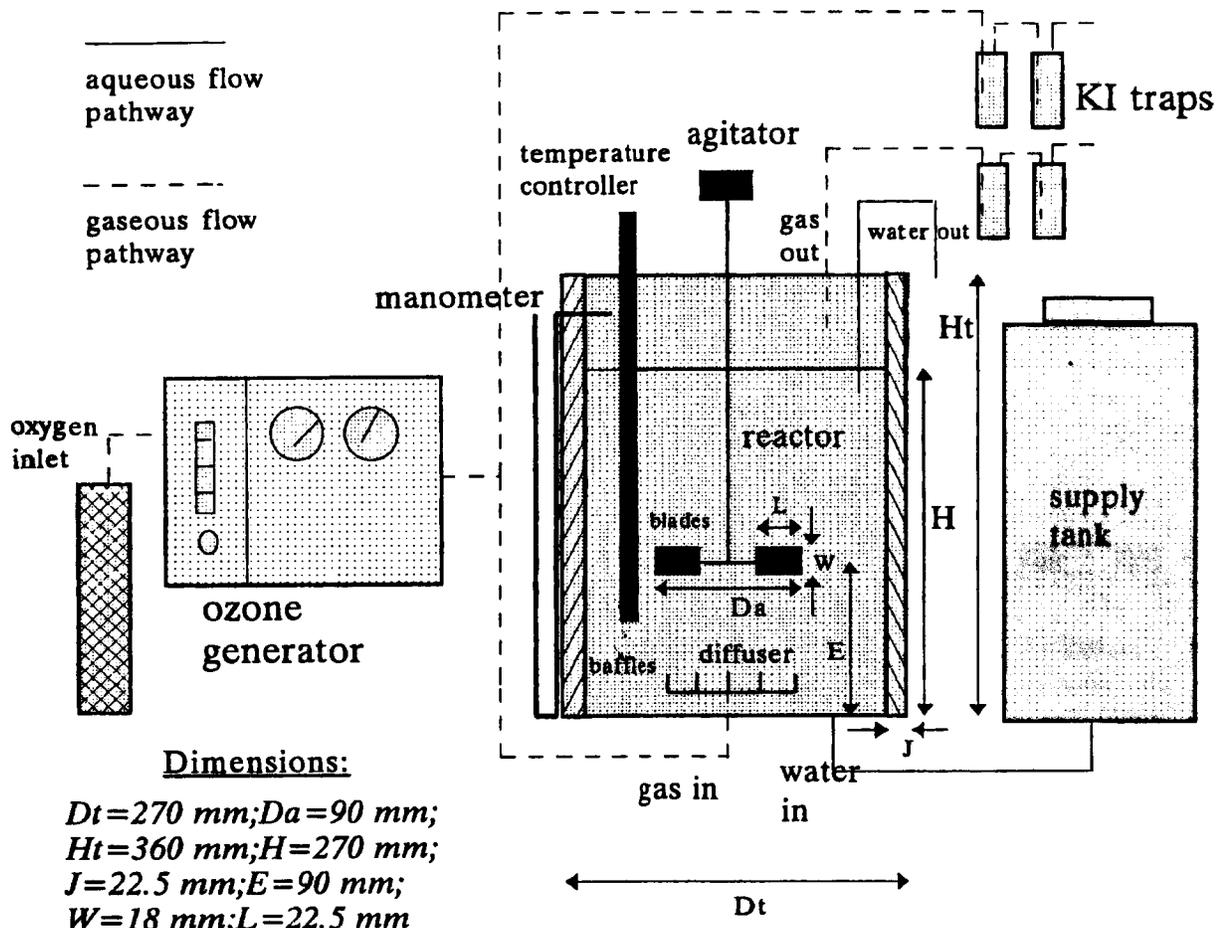


Fig. 1. Schematic diagram of ozone experimental system.

facial area on the gas flow rate is much stronger. Therefore, hypothesis (4) is a reasonable assumption, which greatly simplifies the model development process.

According to the mechanism of mass transfer, K_L is a function of diffusivity (D_v), viscosity (μ), density (ρ), and characteristics of liquid flow, which can be given by the velocity of the tip of agitation blades, $\pi n d_i$, where d_i is the diameter of agitation blades and n is the agitation speed. Therefore, the relationship between the variables affecting K_L can be written as:

$$K_L = f(D_v, \mu, \rho, n d_i) \quad (2)$$

Equation 2, including six variables and three fundamental quantities (mass, length, and time), can be regarded as the sum of a number of terms each consisting of products of powers of the variables.

$$K_L = g(D_v)^f (\mu)^b (\rho)^c (n)^d (d_i)^e \quad (3)$$

where b, c, d, e, f , and g are constants.

These can be expressed in terms of mass, length, and time as follows:

$$K_L = L T^{-1}; D_v = L^2 T^{-1}; \mu = M L^{-1} T; \\ \rho = M L^{-3}; n = T^{-1}; d_i = L$$

Based on Buckingham's π Theorem, the number of dimensionless groups will be three. The conditions of dimensional consistency must be met for each of the fundamentals and the orders of each of these variables can be equated. After substitution and calculation, the following equation can be expressed:

$$\frac{K_L d_i}{D_v} = \left(\frac{n d_i^2 \rho}{\mu} \right)^{1-f-b} \left(\frac{\mu}{D_v \rho} \right)^{1-f} n^{2b} \quad (4)$$

Three dimensionless groups, $K_L d_f / D_v$, nd_i^2 / μ , $\mu / D_v \rho$, known as the Sherwood number, Reynolds number, and Schmidt number, respectively, were generated.

Higbie's penetration theory indicates that K_L is proportional to $D_v^{0.5}$. Substitution of this relationship and the condition of dimensional consistency in Eq. 4 resulted in the following:

$$K_L = D_v^{0.5} n^{0.5} \quad (5)$$

On the other hand, combining the empirical relationship reported by Stankovic (1988) with Eq. 5 yielded the following equation:

$$K_L a = h D_v^{0.5} n^{0.5} G^x \quad (6)$$

where,

h, x are constants;

a = specific interfacial area, cm^{-1} ; and,

G = influent gas flow rate, L/min.

$$\frac{d[TOC]}{dt} = -K[TOC][O_3] \quad (7)$$

Removal of TOC in ozone reaction. The reaction of ozone with organic compounds usually conforms to a first-order kinetic expression with respect to both the concentration of ozone and that of organic compounds. The humic acids solution in terms of TOC was chosen as the target compound to determine the removal efficiency of ozone. Thus, the following expression was obtained:

where,

$[TOC]$ = concentration of TOC, mg/L;

t = time, min;

K = reaction rate constant, L/mg·min; and,

$[O_3]$ = ozone concentration in liquid phase, mg/L.

The mass balance equation for TOC in continuous operation can be described as follows:

where,

$$Q[TOC]_{in} = V \left(-\frac{d[TOC]}{dt} \right) + Q[TOC]_{eff} \quad (8)$$

Q = liquid flow rate, L/min;

$[TOC]_{in}$ = influent TOC concentration, mg/L;

$[TOC]_{eff}$ = effluent TOC concentration, mg/L; and,

V = volume of liquid phase, L.

$$\frac{[TOC]_{eff}}{[TOC]_{in}} = \frac{1}{t_R K[O_3] + 1} \quad (9)$$

Substitution of Eq. 7 into Eq. 8 yielded the following equation:

where t_R = hydraulic retention time = V/Q , min.

Parameters identification

Determination of parameters in the $K_L a$ prediction equation. To demonstrate the application of Higbie's penetration theory to this experimental system, the linear regression of $K_L a$ vs $D_v^{0.5}$ was examined and is shown in Fig. 2. In Fig. 2, D_v was calculated by Wilke and Chang's correlation; while $K_L a$ and equilibrium dissolved ozone concentration ($[O_3]^*$) were obtained from the following mass balance equation:

$$\frac{d[O_3]}{dt} + r_{O_3, sd} = K_L a ([O_3]^* - [O_3]) \quad (10)$$

where $r_{O_3, sd}$ = self-decomposition rate of ozone. This can be described by (Yang 1991):

$$r_{O_3, sd} = 5.117 \times 10^{10} \exp\left(-\frac{8835.25}{T}\right) [O_3] + 9.207 \times 10^{16} \exp\left(-\frac{8835.25}{T}\right) [OH^-]^{0.5} [O_3] \quad (11)$$

where T = temperature, K.

With the known $[O_3]$ and t , as well as $r_{O_3, sd}$, $K_L a$ and $[O_3]^*$ were determined through the use of a linear regression analysis under specific experimental conditions.

Substitution of 20 sets of experimental results into Eq. 6 led to the prediction model of $K_L a$, by the use of non-linear regression analysis:

$$K_L a = 1.20 \times D_v^{0.5} n^{0.5} G^{1.18} + 0.027, R^2 = 0.964 \quad (12)$$

where the unit of $K_L a$ is min^{-1} .

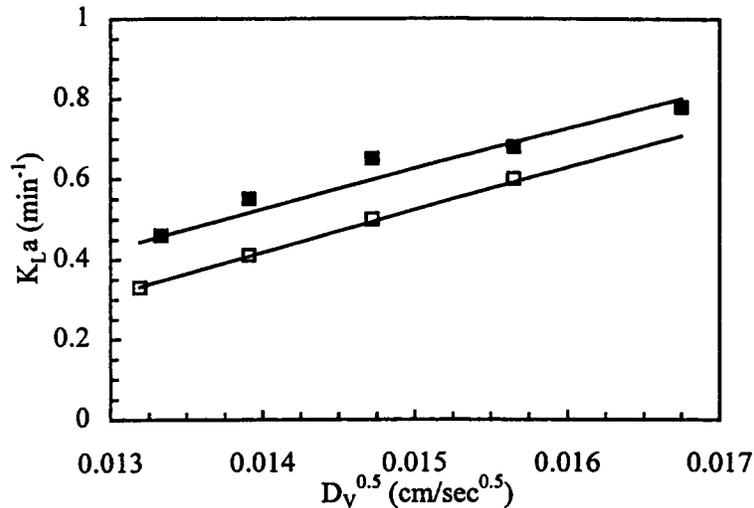


Fig. 2. Results of the linear regression of $K_L a$ vs $D_v^{0.5}$ at different pH. □ pH = 5.1, $R^2 = 0.997$; ■ pH = 7.2, $R^2 = 0.945$.

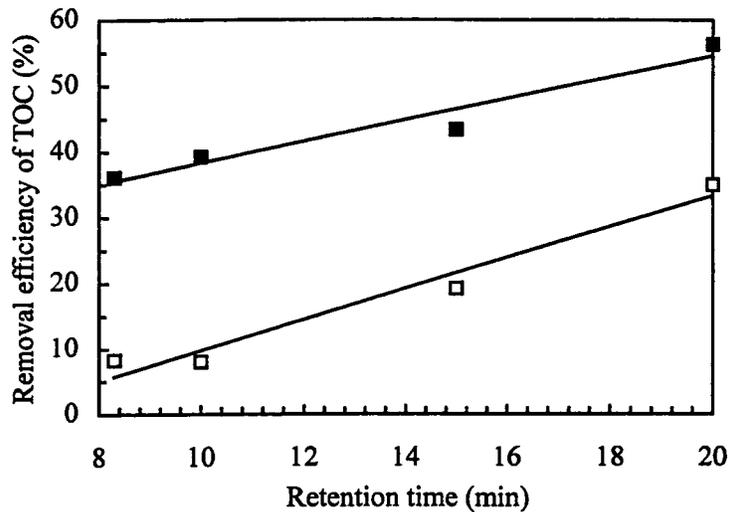


Fig. 3. Effects of retention time on TOC removal efficiency during continuous-type ozonation. □ agitation speed = 100 rpm; ■ agitation speed = 300 rpm.

Determination of TOC reaction rate constant. According to Eq. 9, one can regulate the liquid flow rate and ozone concentration to achieve the expected level of TOC removal efficiency, if the reaction rate constant, K , is known. Therefore, the lab-scale ozonation experiments in the continuous type operation were conducted to measure K . The results of TOC removal efficiency under various agitation speeds and retention time are shown in Fig. 3. This indicates that both the enhancement of retention time and agitation speed would significantly raise the TOC removal efficiency. The K value averaged 0.0291 L/mg·min, with a standard deviation of 0.0054 L/mg·min.

Applications

The optimum operation conditions for semi-batch experiments. First, the Henry's law constant was calculated from the available data. In Table 1, the ozone concentration in the gas phase was directly obtained by the titration method as stated in the Analytical methods section. Then, assuming the ideal gas behavior of ozone yielded the ozone partial pressure (P_{O_3}), the average Henry's law constant was determined to be 0.380 kPa·L/mg.

Fractional ozone absorption (FOA) is defined as the ratio of the ozone transferred to water to the ozone applied to the reactor (Guroi 1985). The following

Table 1. Effects of ozone partial pressures on $K_L a$, $[O_3]^*$ and Henry's law constant.

Influent gas temperature (°C)	Ozone partial pressure, P_{O_3} (kPa)	$K_L a$ (min^{-1})	$[O_3]^*$ (mg/L)	Henry's law constants (kPa·L/mg) [#]
27	1.517	0.487	4.078	0.372
22	2.212	0.472	5.712	0.387
27	2.680	0.466	7.024	0.382

[#]: $P_{O_3} = H \cdot [O_3]^*$, H = Henry's law constant (kPa·L/mg).
 Operating conditions: pH = 6.98; temperature = 25°C; gas flow rate = 10 L/min; and agitation speed = 200 rpm.

equation for FOA in semi-batch operation was derived:

$$FOA(\%) = \frac{V \times K_L a ([O_3]^* - [O_3])}{G \times P_{O_3} \times 1.98 \times 10^5} \times 100 \quad (13)$$

where P_{O_3} = ozone partial pressure, kPa.

The value of 1.98×10^5 is the factor which will transfer the term of denominator into the unit of mg/min. Substitution of Eq. 12 and the Henry's law relationship into Eq. 13 yielded the following equation:

$$FOA(\%) = \frac{V \times (1.20 \times D_v^{0.5} n^{0.5} G^{1.18} + 0.027) + \frac{V \times (P_{O_3} - [O_3])}{0.380}}{G \times P_{O_3} \times 1.98 \times 10^5} \times 100$$

Therefore, Eq. 14 can be applied to predict the initial ozone absorption efficiency (FOA_0) in this experimental system at $t = 0$, that is, $[O_3] = 0$.

$$FOA(\%) = \frac{V \times (1.20 \times D_v^{0.5} n^{0.5} G^{1.18} + 0.027)}{75240 \times G} \times 100 \quad (15)$$

The effects of temperature, agitation speed, and influent gas flow rate on FOA_0 and $K_L a$ can be examined based on Eqs. 15 and 12. An increase in temperature increased the diffusivity of ozone in water which led to the enhancement of K_L , because K_L is

directly proportional to $D_v^{0.5}$ based on Higbie's penetration theory (Danckwerts 1970). $K_L a$ and FOA_0 were increased with an increase in agitation speeds. This is attributed to the breakdown of liquid film resistance and the increase in surface renewal rates. As a result, $K_L a$ increased when the influent gas flow rate was increased, due to the increase of specific interfacial surface area, a (Stankovic 1988).

Figure 4 shows the relationships between the agitation speeds and influent gas flow rates under the specific conditions of $K_L a$ and FOA_0 . This indicates FOA_0 is mainly controlled by n , and not by G . Figure 4 also provides the baseline information for the engineers to choose the most practical alternative among the various arrangements of n , G , and $K_L a$ to achieve the same level of FOA_0 . For instance, one can find the agitation speed to obtain the desired FOA_0 in the water treatment plants, if the value of G is given.

Furthermore, power consumption (P_g) should be taken into consideration for the most efficient design and operating conditions in practice. P_g was calculated as follows (McCabe et al. 1985):

$$P_g = \alpha N_p n^3 d_i^5 \rho \quad (16)$$

where,

P_g = power consumption with gas dispersion, watt;
 α = ratio of P_g over P_0 , where P_0 is the power consumption in ungasged liquid; and,
 N_p = power number, $N_p = P_0/n^3 d_i^5 \rho$.

Since α is a function of superficial gas velocity which is directly proportional to G , P_g is influenced by both n and G . In addition, for a standard six-bladed turbine, $N_p = 5.2$ and α can be calculated corresponding to various G values (McCabe et al. 1985). The resulting P_g values under different n and G are shown in Fig. 4. This figure can be used to determine G and n for given requirements of FOA_0 and P_g . Also, under a

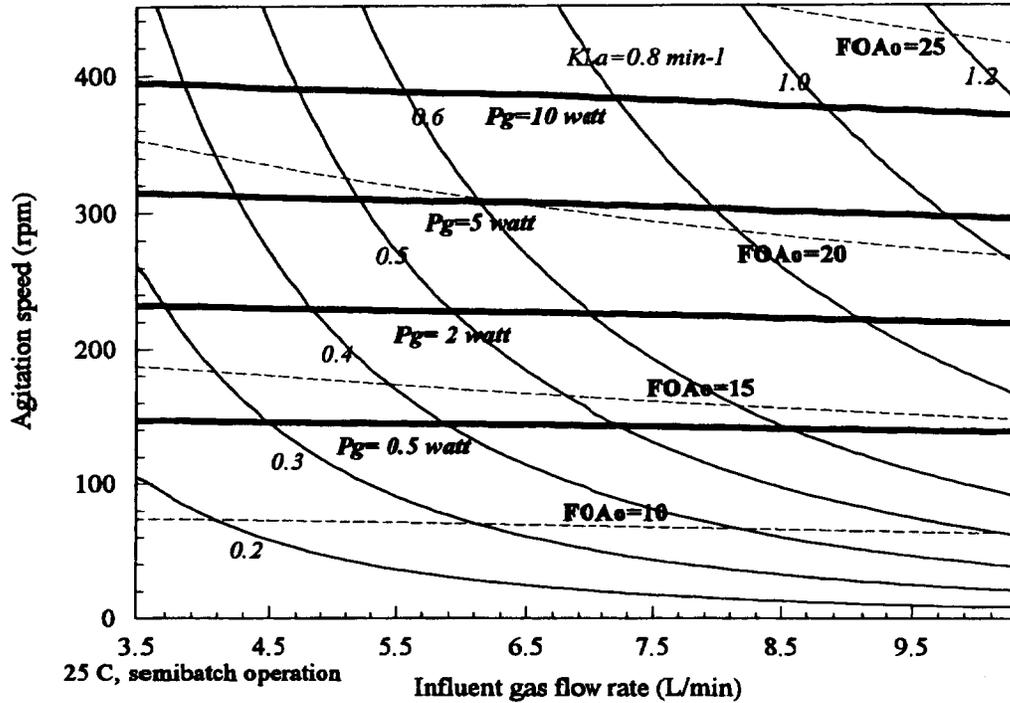


Fig. 4. The relationships between agitation speeds and influent gas flow rates under specific conditions of K_La , FOA_0 , and power consumption.

constant value of G , the optimum operation conditions for semi-batch operation can be determined through the use of Fig. 4.

The optimum agitation speed for TOC removal during continuous operations. In this section, the K_La prediction model is integrated with TOC removal efficiency relationship, to develop the ozone mass transfer model in view of controlling the formation of organic compounds. Based on mass balance of dissolved ozone concentration in reactor during continuous-type operation, the following equation can be derived:

$$V \frac{d[O_3]}{dt} = V \times K_La([O_3]^* - [O_3]) - Vr_{O_3} - Q \quad (17)$$

where r_{O_3} = reaction rate of ozone, mg/L·min, = $K[TOC][O_3]$, as shown in Eq. 7.

For steady state condition, Eq. 17 becomes:

$$Q[O_3] = V \times K_La([O_3]^* - [O_3]) - Vr_{O_3} \quad (18)$$

If $[O_3]^*$ is replaced by the ozone partial pressure, P_{O_3} , Eq. 18 can be modified as:

$$Q[O_3] = V \times K_La \left(\frac{P_{O_3}}{0.380} - [O_3] \right) - Vr_{O_3} \quad (19)$$

Since the prediction model of K_La (Eq. 12) is applicable only to the deionized water, it is necessary to introduce the enhancement factor, E , to account for the effect of organic compounds in the real water sample on K_La :

$$E = \frac{K_La \text{ with reaction}}{K_La \text{ without reaction}} \quad (20)$$

For the fixed level of water quality parameters, applied ozone concentration, and influent gas flow rate, the following equations were developed by combining Eqs. 12, 19, and 20:

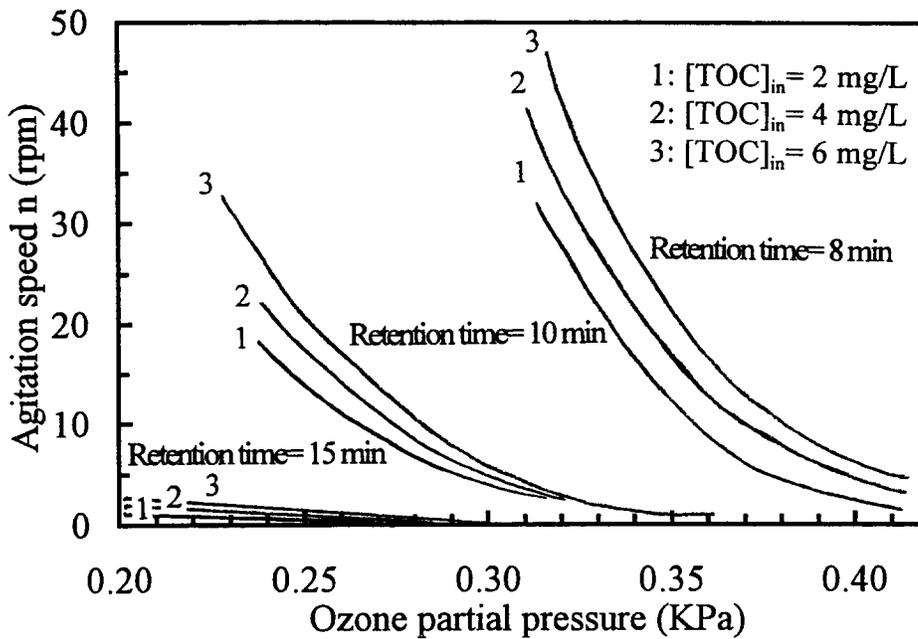


Fig. 5. Simulation results of the relationship between agitation speed and ozone partial pressure in the view of controlling the formation of TOC.

$$n = \left(\frac{Q[O_3] + Vr_{O_3}}{1.20 \times EV \left(\frac{P_{O_3}}{0.380} - [O_3] \right)} - 0.0225 \right)^2 D_v^{-1} G^{-2.36} \tag{21}$$

where the unit of n is rps.

Equation 21 includes the mass transfer and reaction kinetics characterizations and can be successfully used in predicting the most efficient agitation speed for controlling the formation of organic compounds in the ozonation process. Also, the marginal level of ozone partial pressure to achieve the highest TOC removability can be determined through the use of the above developed model (Eq. 21). For example, Fig. 5 gives the relationship between the agitation speed and applied ozone concentration under various retention times. In this simulation study, the temperature (25°C), pH (7), influent gas flow rate (10 L/min), the removal efficiency of TOC (15%), and enhancement factor ($E = 1$) were held constant. The higher the ozone partial pressure, the lower the agitation speed needed to achieve the same level of performance. In other words, it is only necessary to increase the ozone partial pressure slightly, instead of greatly enhancing the agitation power consumption, to obtain the same level of removal efficiency of TOC.

CONCLUSIONS

The prediction model of $K_L a$ was developed through the use of dimensional analysis. Substitution of laboratory data into the $K_L a$ prediction model showed that overall mass transfer coefficient is a function of diffusivity, agitation speed, and gas flow rate, and the parameters in the equation were determined.

Based on the mass balance relationship and kinetic expression of TOC oxidation, an equation governing TOC removal efficiency during the continuous experiments was developed. The K value averaged 0.0291 L/mg·min, with a standard deviation of 0.0054 L/mg·min.

The relationships between agitation speeds and influent gas flow rates under specific conditions of $K_L a$, FOA_0 , and power consumption were developed as shown in Fig. 4. Figure 4 can provide the baseline information for the engineers to choose the most practical alternative among the various combinations of n , G , $K_L a$, and P_g to achieve the same level of FOA_0 .

The developed model in combination with mass transfer and reaction kinetics can be used successfully in predicting the most efficient agitation speed for controlling the formation of organic compounds. Also, the critical value of ozone partial pressure to achieve the highest TOC removability can be determined

through the use of the above developed model. For example, it is only necessary to increase ozone partial pressure slightly, instead of greatly enhancing the agitation power consumption, to achieve the same level of removal efficiency of TOC.

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