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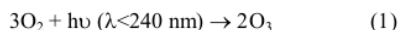
Mass Transfer Coefficient of Ozone in a Bubble Column

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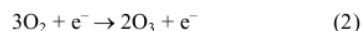
Abstract.

1 Introduction

Ozone (O₃) is naturally formed in the stratosphere by chemical reactions involving oxygen with the aid of solar ultraviolet radiation (sunlight) of wavelengths below 240 nm [1]. The reaction can be simplified as Equation (1).



Ozone can be generated using non-thermal plasma corona discharges at atmospheric pressure and room temperature where an extra-high voltage electrical current is passed from an electrode through a neutral fluid, which is usually air [2]. The current is able to ionize the fluid creating plasma around the electrode. Oxygen experiences ionization and the overall reaction that may occur is written as Equation (2) [3].



Ozone is a strong oxidant with a very high redox potential, i.e. 2.07 V [4]. Ozone is highly reactive that it readily reacts with various organic and inorganic substances [5]. It has been utilized in advanced oxidation process (AOP), without or with H₂O₂ or UV to disinfect microorganism [5], to eliminate pollutants [6, 7], and to control odor problem [8].

To implement AOP in a liquid phase, the ozone must be solubilized in the liquid. The solubility of ozone in water is very low [9]. The solubilization of ozone in water is influenced by mass transfer coefficient (k_L) of ozone in water. The $k_L a$ is affected by the ozone concentration in the gas phase, the gas flow rate, temperature, and pH of the liquid phase [10 – 12]. The objective of this work is to study the effect of pH of the

water and the flow rate of the gas on the mass transfer coefficient of ozone in water.

2 Materials and Method

2.1. Materials

The materials used in this study included potassium iodide (Merck, Cat. No. 1.05043.0250), sulphuric acid (Merck, Cat. No. 1.12080.1000), sodium thiosulfate (Merck, Cat. No. 1.93248.0521), sodium hydroxide (Merck, Cat. No. 1.06498.050), and hydrochloric acid (Merck, Cat. No. 1.93401.0521). The materials were utilized without any treatment.

2.2 Method

The experiment was conducted by using equipment as depicted in Fig.1, which consisted of a bubble column (a glass cylinder of 10 cm diameter and 50 cm height) equipped with a bubble diffuser, a corona discharge (40 kV) ozone generator (Dipo Technology Indonesia), and a compressor.

Thirty grams of potassium iodide was dissolved in distilled water to form 1500 mL solution with a concentration of 2% (w/w). The pH of the solution was varied (3, 4, 5, 6, 7, 8, 9, and 10) and was adjusted using HCl or NaOH solution. The solution was then placed in the column of which the temperature was kept at 28 ± 1°C. Air was passed the corona discharge ozone generator using the compressor at a certain flow rate (2, 2.5, 3, 3.5, 4, 4.5, and 5 L min⁻¹), then it was bubbled

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into the column. The soluble ozone readily reacted with KI to form I₂ according to Equation (3).



A sample was withdrawn every 5 minutes and was analyzed for concentration of ozone by KI titrimetry. Ozone reacts with KI to form I₂ by a titrimetric method using sodium thiosulfate as the titrant. The concentration of I₂ is equivalent to that of ozone.

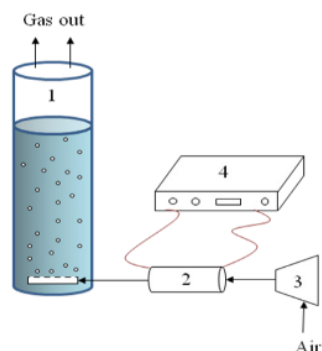


Fig.1. Schematic of equipment for experiment: bubble gas reactor (1), corona discharge ozone generator (2), compressor (3), and power supply (4)

2.3. Mathematical model for mass transfer

According to two-film theory, to be solubilized in liquid water, ozone molecules must diffuse from the gas phase (gas bubble) to the bulk liquid through gas and liquid films [13]. However, as ozone has a very low solubility on water. The absorption of ozone through the liquid film results in a very small concentration difference across the film. The absorption of ozone through the liquid film proceeds very slowly. It is reasonable to assume that resistance to mass transfer through the liquid film is much higher than that through the gas film. The gas-phase resistance of mass transfer is therefore negligible. The mathematical model to describe the mass transfer of ozone from the gas phase to the liquid water can be represented by equation (4) [10],

$$\frac{dC}{dt} = k_L a (C^* - C) \quad (4)$$

where $k_L a$ is the volumetric mass transfer coefficient of ozone, C and C^* are the concentration ozone at time t and at saturated condition, respectively. Equation (4) is integrated to give Equation (5).

$$\ln(1 - C/C^*) = -k_L a t \quad (5)$$

3 Results and Discussion

3.1. Influence of time on the concentration of dissolved ozone

The concentration of ozone at various pH and time is depicted in Fig. 2. The results show that the

concentration of ozone increases rapidly in the first 10 minutes and slows down afterward. Finally, it reaches a steady-state value after 30 minutes.

A similar result was reported by Egorova *et al.* [14] who passed a mixture of oxygen and ozone through water in a bubble reactor at 20°C. They reported after 10 minutes of bubbling, that concentration of ozone remained constant. The time needed to reach the saturation concentration in this work is much longer than that needed by Egorova *et al.* [14]. It is possibly because this work used air to be fed to the ozone generator, while Egorova *et al.* [14] utilized oxygen. The concentration of ozone in the gas phase in this work must be lower, so it took a longer time to reach the saturation concentration.

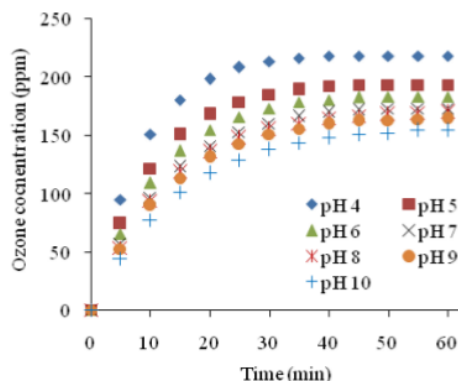


Fig. 2. Effect of time on the concentration of ozone in water at 28±1°C and various pH with a gas flow rate of 4 L min⁻¹

3.2. Influence of gas flow rate on the solubility of ozone

The effect of the flow rate of the ozone-containing gas on the concentration of dissolved ozone is depicted in Fig. 4. All of the data were obtained by bubbling the ozone-containing gas into the reactor at 28±1°C and pH 7 for 30 minutes.

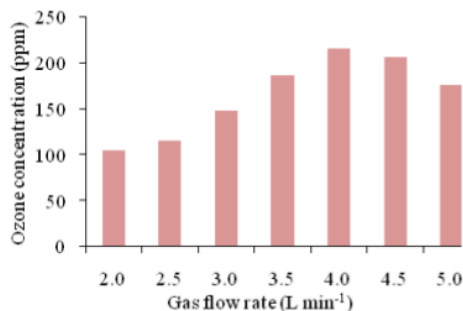


Fig. 4. Effect of gas flow rate on the concentration of dissolved ozone in water at 28±1°C and pH 7 after 30 minutes

Fig. 4 shows that the ozone concentration increases from 105 ppm to 216 ppm as the gas flow rate rises from

2 to 4 L min⁻¹. The ozone concentration then decreases at a flow rate higher than 4 L min⁻¹. As the gas flow rate is higher, the residence time of air in the ozone generator becomes shorter. With less reaction time, less ozone is formed [17]. The same trend was also reported by Boonduang *et al.* [16]. They reported that the optimum ozone concentration was obtained by using a gas flow rate of 2 L min⁻¹.

3.3. Influence of gas flow rate on the $k_L a$

To investigate the effect of gas flow rate on the mass transfer coefficient, a series of experiments at pH 7 with flow rate varied from 2 to 5 L min⁻¹ were conducted. The results are depicted in Fig 5. Fig. 5 shows how the $k_L a$ of ozone is influenced by the gas flow rate. The $k_L a$ increases from 0.4×10^{-3} s⁻¹ to 1.45×10^{-3} s⁻¹ as the gas flow rate rises from 2 L min⁻¹ to 4 L min⁻¹, after which the $k_L a$ drops with increasing gas flow rate. The maximum mass transfer coefficient is reached with the gas flow rate of 4 L min⁻¹. This flow rate was used to investigate the effect of pH on the mass transfer coefficient.

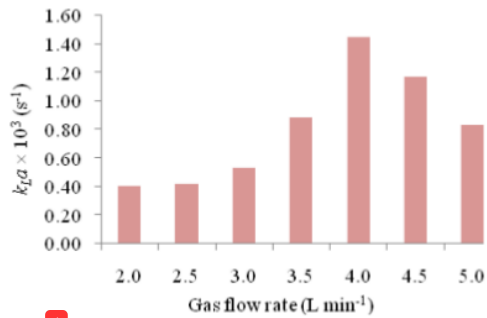


Fig. 5. Effect of gas flow rate on mass transfer coefficient of ozone in water at 28±1°C and pH 7 after 30 minutes

Other groups of researcher revealed similar results to this work, in which the $k_L a$ was influenced by the gas flow rate. Gao *et al.* [10] utilized Karman contactor to study the mass transfer of ozone in water. They found that the $k_L a$ increased with the gas flow rate. Grima reported that the $k_L a$ was 2.95×10^{-3} s⁻¹ when the gas flow rate was 100 mL min⁻¹, and it increased to 7.47×10^{-3} when the experiment was conducted at higher gas flow rate of 600 mL min⁻¹.

When the gas flow rate increases, its superficial velocity (U_G) increases as well. Bouaifi *et al.* [18] explained that in a bubble column, the U_G is related to the $k_L a$ in which the $k_L a$ increases with increasing U_G . According to Akita and Fumitake [19], the superficial gas velocity of the gas in a bubble column is linearly correlated to the gas holdup (ε_G), which can be related to the mass transfer coefficient through Equation (6).

$$k_L a = c_2 D_L^{0.5} \nu_L^{-0.12} (\gamma/\rho_L)^{-0.62} D^{0.17} g^{0.98} \varepsilon_G^{-1.1} \quad (6)$$

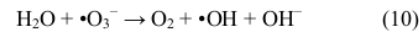
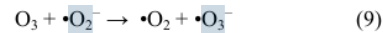
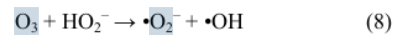
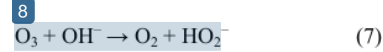
where c_2 is a constant, D is column diameter, g is gravitational constant, while D_L , ν_L , and ρ_L are liquid phase diffusivity, viscosity, and density, respectively.

However, as it has been previously explained, when the gas flow rate increases, the residence time of the air in the ozone generator is shorter, and less ozone is formed. It eventually decreases the mass transfer coefficient.

3.4. Influence of pH on the $k_L a$

It is shown in Fig. 2 that pH influences the ozone concentration in water. At the same bubbling time, the concentration of ozone increases as the pH decreases. Clever *et al.* [9] reported similar trend, in which at 35°C the solubility of ozone in water increased from 6.8 ppm to 39.6 ppm as the pH decreased from 6.8 to 2.7 when the ozone concentration in the gas was 200 ppm. Roth and Sullivan [20] found that the Henry's constant of ozone in water decreased as the pH dropped. A smaller value of Henry's constant leads to a higher value of solubility. Mizuno and Tsuno [21] found similar results as well.

Egorova *et al.* [13] explained that the solubility of ozone in water decreased with increasing pH. In alkaline solution, the hydroxyl ion (OH⁻) catalyzes the decomposition of ozone through the following chain reactions:



Using the data presented in Fig. 2, $\ln(1 - C/C^*)$ was plotted versus t for each pH. Only sets of data at pH 4, 7, and 10 are depicted in Fig. 7. Each set of data at respective pH can be approximated by linear lines with excellent correlation coefficients. The $k_L a$ can be concluded from the slope of the plot. The results are shown in Table 1, along with those of pH 5, 6, 8, and 9 and respective correlation coefficient. The coefficients of correlation (R^2) of the correlation for all pH are higher than 0.99. The values of $k_L a$ obtained by other researchers [12, 22] are presented in Table 1 as well.

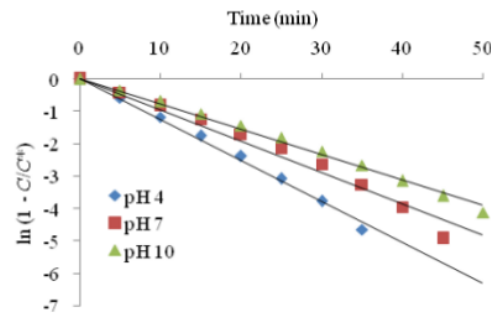


Fig. 7. Plot of $\ln(1 - C/C^*)$ versus time

The results presented in Table 1 show that the k_{La} of ozone drops from 2.10×10^{-3} to $1.22 \times 10^{-3} \text{ s}^{-1}$ as pH increases from 4 to 10. Flores-Payán *et al.* [12] revealed a similar trend, in which the mass transfer coefficient of ozone decreased from 6.90×10^{-3} to $4.30 \times 10^{-3} \text{ s}^{-1}$ as the pH increased from 1.5 to 11.

Table 1. Mass transfer coefficient at various pH.

pH	$k_{La} (\text{s}^{-1})$		
	This work	[12]	[23]
1.5		6.90×10^{-3}	
4	2.10×10^{-3}	6.30×10^{-3}	8.75×10^{-3}
5	1.75×10^{-3}		
6	1.63×10^{-3}		
7	1.45×10^{-3}	5.30×10^{-3}	7.61×10^{-3}
8	1.37×10^{-3}		
9	1.35×10^{-3}	4.30×10^{-3}	6.54×10^{-3}
10	1.22×10^{-3}		

The decrease of the k_{La} of ozone in water at alkaline condition is related to the existence of HO• radical. This radical catalyzes the self-decomposition of ozone according to equations (7) – (10) [12].

The values of k_{La} obtained in this work are lower than those of other groups of researcher [12, 23]. It could be because of different type of reactor, gas diffuser geometry, and gas flow rate. However, the values of k_{La} are of the same order of magnitude.

Conclusion

The results show that the concentration of ozone increases with time. The saturation concentration is attained after 30 minutes of ozonation. Both the gas flow rate and pH influence the concentration and the k_{La} . The highest value of k_{La} of $2.10 \times 10^{-3} \text{ min}^{-1}$ is obtained at a gas flow rate of 4 L min^{-1} and pH 4.

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