

Enhancement performance of ozone mass transfer by high gravity technology

Weizhou Jiao*, Yuejiao Qin, Yonghong Wang, Liang Guo, Youzhi Liu

*Shanxi Province Key Laboratory of Hige-Oriented Chemical Engineering, North University of China, Taiyuan 030051, China
Tel. +86-351-3921986, Fax +86-351-3921497, email: jwz0306@126.com (W. Jiao), 476796841@qq.com (Y. Qin),
langya78@qq.com (Y. Wang), zbdxgl@126.com (L. Guo), zbdxlyz@126.com (Y. Liu)*

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ABSTRACT

This paper firstly proposed the method that provided enhanced performance of ozone mass transfer by high gravity. The experiment results showed that, in strong acid environment, with increasing high gravity factor, gas-phase ozone concentration and liquid flow-rate are beneficial to the improvement of ozone mass transfer rate. With the suitable operational condition, the ozone concentration in water can reach 5.0 mg/L. Under these situations, the mean liquid-phase ozone mass transfer coefficient is $2.145 \times 10^{-2} \text{ s}^{-1}$ which is about by 5.6 times compared with the traditional aeration device, proving that high gravity technology can be utilized to effectively improve the ozone mass transfer rate.

Keywords: Ozone; Gas-liquid mass transfer; High gravity; Rotating packed bed; Mass transfer coefficient

1. Introduction

The ozone-based advanced oxidation technology has been widely used in drinking water sterilization, wastewater treatment and other aspects [1]. In recent years, with advantages of the rapid reaction rate, high treatment efficiency and no secondary pollution, this technology has become the research hotspot in poisonous and non-biodegradable organic wastewater treatment fields [1–3]. In aqueous solution, ozone acts on various compounds by direct ozonation and radical reaction. Molecular ozone, a powerful oxidant agent, can react selectively with organic compounds in water. The ozonation system with a high oxidation efficiency should be able to form highly reactive radical species from the decomposition of molecular ozone in water so as to destruct the persistent organic pollutants via high-activity and strong oxidizing hydroxyl radical ($\cdot\text{OH}$) [4,5]. Because of the low solubility of ozone in water, the ozonation reactions of most organic matters are controlled by the ozone mass transfer process and the oxidation reaction simultaneously [6–9]. Therefore, how to effectively

improve the contact area of gas-liquid and speed up the interphase update rate are of great academic value and application significance to enhanced ozone mass transfer.

Rotating packed bed (RPB), a new type device with a high rotating speed, is used to generate high centrifugal force (high gravity field) to enhance gas-liquid mass transfer rate and then enhance the transfer reaction process [10]. In the high gravity field, the liquid is cut into small drops, wires and films by the high-speed rotating packing, which increases the contact area of gas-liquid. Under the high shear force in a RPB, tiny liquid droplets and thin liquid films are generated and flow chaotically through the packing with substantial turbulence in the rotor. As a result, RPB with rapidly renewed surfaces, which enhance the gas-liquid phase contact area and mass transfer rate [11,12]. Combining the high gravity technology and ozone advanced oxidation technology as a process intensification has a wide application prospect in the field of wastewater treatment.

Chiu et al. [13] adopted the RPB with ozone for the removal of naphthalene (NAP) in the $\text{C}_{12}\text{H}_{25}\text{O}(\text{C}_2\text{H}_4\text{O})_n$ (Brij 30)-containing solution. The presence of Brij 30 would significantly reduce volatilization of NAP from the solution.

*Corresponding author.

The oxidation treatment of phenol by O_3/H_2O_2 advanced oxidation processes (AOPs) was studied by Zeng et al. [14] in a RPB. Phenol degradation ratio reached 100% at an initial phenol concentration of 40 mg/L in the O_3/H_2O_2 AOPs. Our team [15] presented an integrated high gravity-ultrasonic/ozonation/electrolysis technology to degrade the wastewater containing nitrobenzene. Compared with traditional aeration ozone contactors, a novel RPB with high mass transfer rate was used to increase the ozonation treatment efficiency. Chang et al. [16] used RPB as a catalytic ozonation reactor to decompose phenol. The total organic carbon (TOC) removal efficiency of phenol employing the high gravity ultra violet ozonation (HG-UV-OZ) process is 94% at the reaction time 80 min.

Current studies on high gravity ozone oxidation technology mainly focus on the influences of different factors on the degradation of organic matters in wastewater. Theoretical researches on how the ozone mass transfer can be influenced are seldom reported. Taking RPB as reactor, this paper studied the enhancing mechanism of high gravity technology on ozone mass transfer process and established mass transfer model. The influences of water quality, high gravity factor, initial pH value, liquid flow-rate, gas phase ozone concentration and some other factors on the ozone mass transfer process were studied, aiming at providing experimental data and theoretical basis for ozone degradation of wastewater.

2. Theoretical basis of ozone dissolution

Ozone dissolution in water is a complex process, which is relevant to the ozone reaction rate, thermodynamic distribution, catalytic decomposition and other parameters. Under normal conditions, ozone dissolution in water follows Henry's law Eq. (1).

$$P_G = H_G c_L^* \quad (1)$$

where c_L^* represents the ozone mass concentration in water at equilibrium state; P_G represents the gas-phase ozone partial pressure; and H_G represents the Henry's constant.

During the dissolution process of ozone in water, some dissolved ozone in water will decompose by itself. Thus, the dissolution process of ozone can be regarded as a gas-liquid reaction process. The chemical reaction of ozone in water is thought as an irreversible first-order reaction or quasi first-order reaction. For a batch bubble reactor, ozone mass transfer rate equation can be expressed as Eq. (2).

$$\frac{dc_L}{dt} = K_L \cdot a \cdot (c_L^* - c_L) - K_d c_L \quad (2)$$

where K_L represents liquid phase mass transfer coefficient; a represents the gas-liquid contact area in unit volume; $K_L a$ represents the ozone volumetric mass transfer coefficient; K_d represents ozone decomposition rate constant; c_L^* and c_L represents the ozone balance concentration and the actual concentration at the time of t , respectively.

When pH is below 7.0, the ozone decomposition reaction rate becomes very small, and its self-decomposition reaction can be neglected. The ozone dissolution process can be approximately seen as a physical process.

As seen from the ozone dissolution rate Eq. (2) that the expanding of contact area of gas-liquid and the decreasing of ozone self-decomposition would improve the ozone dissolution rate at a certain temperature and pressure. At the same time, the mass transfer coefficient for a liquid side resistance controlling system (absorption of O_3 in water) can be sped up by improving the liquid turbulence degree.

3. Calculation model of liquid-phase volumetric mass transfer coefficient

In the cross-flow RPB, the gas-liquid concentrations with cross flow contact in radial direction and axial direction are changeable. Thus, it is difficult to directly obtain the volumetric mass transfer coefficient expression. Usually, the packing is classified into infinitesimal rings, namely, m homocentric cylinders along the axis and n equal parts in radial are divided. Then, the recursion method is used to get the analytic expression of volumetric mass transfer coefficient.

3.1. Assumptions

For the convenience of building the model, the computation has the following assumptions. (1) In strong acid water solution, ozone dissolution is regarded as a physical absorption process. (2) The plug-flow condition is applied in both gas and liquid phases. (3) $G, L, K_L a$ are constants. (4) End effect is not considered.

3.2. Liquid-phase volumetric mass transfer coefficient equation

The infinitesimal rings at the packing with height of dz and radius of dr in RPB are studied, shown as Fig. 1.

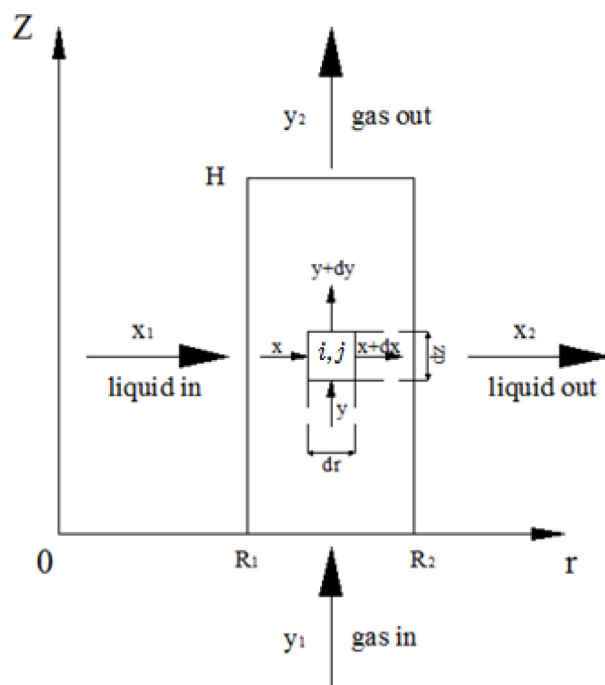


Fig. 1. Sketch of material balance.

Material balance equations of dz infinitesimal rings at the packing layer can be expressed as Eq. (3)

$$N_A \cdot a \cdot 2\pi r dz dr = \frac{L}{H} dz dx = -\frac{G}{\pi(R_2^2 - R_1^2)} \cdot 2\pi r dr dy \quad (3)$$

Namely, O_3 molar mass transferred between gas-liquid = added O_3 molar mass in liquid phase = decreased O_3 molar mass in gas phase. Where, G and L (mol/s) represent the molar flow rates of gas-phase and liquid-phase, respectively; R_1 and R_2 (m) represent the inner and outer radius of packing layer, respectively; H (m) represents the axial length of packing; a (m^2/m^3) represents the total specific surface area of mass transfer; N_A ($k \text{ mol}/(m^2 \cdot s)$) represents the mass transfer rate.

$$N_A = K_x(x^* - x) \quad (4)$$

For liquid-phase, Eq. (4) is substituted into Eq. (3), there is Eq. (5),

$$H = \frac{L}{\pi K_x a (R_2^2 - R_1^2)} \int_{x_0}^{x_{0,j}} \frac{dx}{x^* - x} = HTU \cdot NTU \quad (5)$$

where

$$HTU = \frac{L}{\pi K_x a (R_2^2 - R_1^2)} \quad (6)$$

is defined as the height of transfer unit of the j th section in the axial direction,

$$NTU = \int_{x_0}^{x_{0,j}} \frac{dx}{x^* - x} \quad (7)$$

is defined as the number of transfer unit of the j th section.

The liquid concentration of Eq. (7) and $K_L a$ can be obtained according to the algorithm proposed by Guo et al. [17] and Lin et al. [18].

$$K_L a = K_x a / C_L = \frac{L}{C_L \pi H (R_2^2 - R_1^2)} \int_{x_0}^{x_{0,j}} \frac{dx}{x^* - x} \quad (8)$$

3.3. Liquid-phase volumetric mass transfer coefficient equation in traditional device

For a batch bubble reactor, O_3/O_2 mixed gas is introduced in the ozone-free water. According to ozone dissolution mass balance Eq. (2), ozone self-decomposition can be neglected ($k_d = 0$). Mean liquid-phase volumetric mass transfer coefficient equation [19] can be converted into Eq. (9),

$$\frac{dC_L}{(c_L^* - c_L)} = K_L \cdot a \cdot dt \quad (9)$$

Therefore,

$$K_L a = \frac{\ln \frac{c_L^* - c_L^0}{c_L^* - c_L}}{t} \quad (10)$$

4. Experiments

4.1. Experimental facility

High gravity experimental facility adopts homemade cross-flow RPB. The packing is stainless-steel corrugated wire gauze and the characteristic parameters of the packing are shown in Table 1.

In the traditional aeration reaction device (Diameter, 56 mm; Height, 500 mm), ozone forms into fine bubble through aerator to contact with water. In this way, mass transfer process is completed.

4.2. Experimental method

The experiment is carried out at the temperature of $18 \pm 1^\circ\text{C}$ and the experimental process is shown as Fig. 2. The deionized water and running water were used in this experiment. Ozone was generated from pure oxygen by using an ozone generator, and the ozone-containing gas containing O_3 and O_2 via a gas rotameter was introduced into the bottom of RPB and passes vertically the packing. The liquid from liquid inlet is introduced from the tank into the inner edge of the packing through a liquid distributor and under the action of centrifugal force it is thrown away along the packing in radial direction. At the same time, it contacts with O_3 cross flow which enters from the bottom to complete the mass transfer process. After this, it enters into the reservoir for cycle. The unreacted O_3 is absorbed by KI solution and let out.

Table 1
Packing characteristics in RPB

Item	Parameter
Outside diameter, mm	75
Inside diameter, mm	40
Height, mm	75
Specific surface area, $m^2 \cdot m^{-3}$	935.07
Density, $g \cdot cm^{-3}$	7.9
Porsity	0.74

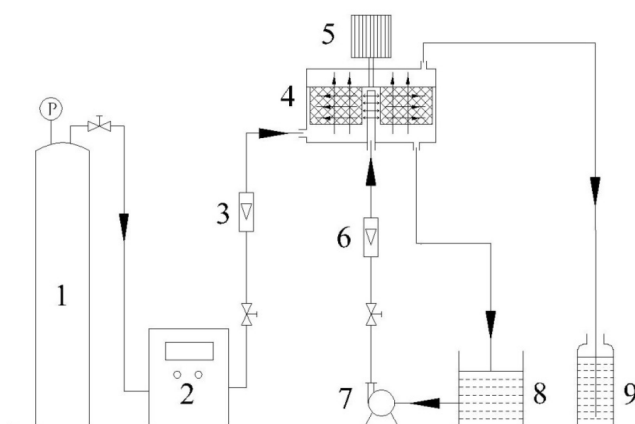


Fig. 2. Experiment process flow diagram: 1. Oxygen cylinder; 2. Ozone generator; 3,6. Rotameter; 4. Rotating packed bed reactor; 5. Motor; 7. Pump; 8. Wastewater reservoir; 9. Tail-gas absorption bottle.

4.3. Measurement system

Gas-phase ozone concentration was measured by ozone detector (Honghai, GT901, China). Ozone generator (LBC-50W, Shandong Lvbang Photoelectrical Equipment Co., Ltd, China), gas/liquid flow rate (ZML-2, Yuyao Yinhuang Flow Instrument Co., Ltd), and PHS-3C precision acidity meter. The measurement of liquid-phase ozone concentration adopts multi-parameter water quality analysis instrument for detection (SQ-MA, Full spectrum, Hebei Shangqing Electronic Technology Co., Ltd, China).

5. Results and discussions

5.1. Influences of high gravity factor

The high gravity factor [20] denotes the ratio of the average centrifugal acceleration versus the acceleration of gravity which can characterize the intensity of gravitational field. The rules relevant to the influence of high gravity factor on ozone solubility and liquid phase mass transfer coefficient $K_L a$ are presented in Fig. 3 and Fig. 4, respectively. The experiments were carried out at pH value of 3.5, liquid flow-rate of 90 L/h, gas flow-rate of 75 L/h and ozone concentration of 50 mg/L.

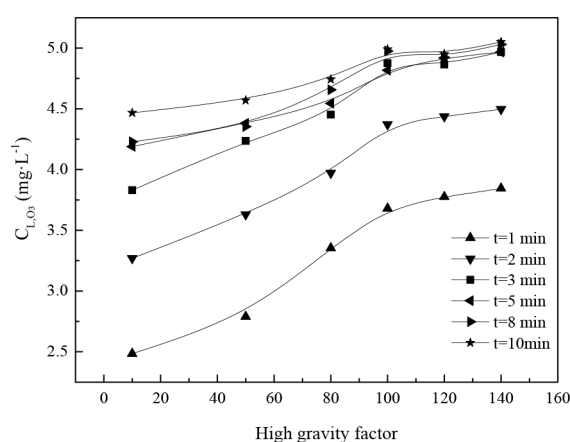


Fig. 3. Influence of high gravity factor on ozone solubility.

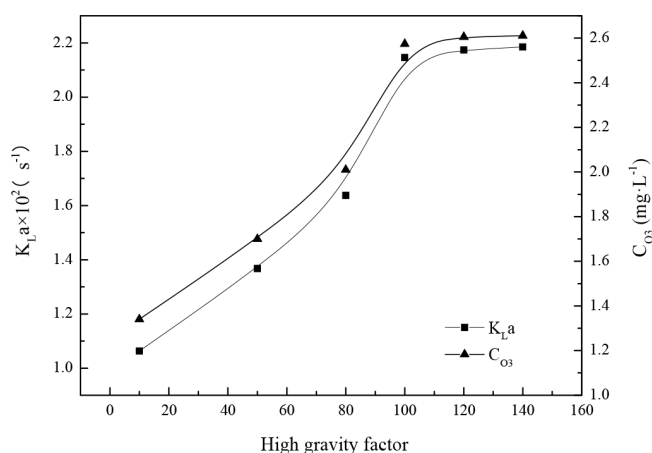


Fig. 4. Influence of high gravity factor on $K_L a$ and ozone solubility.

It can be seen from Fig. 3 that when the high gravity factor increases from 10 to 140, the ozone dissolution rate in water obviously speeds up. The ozone concentration in water increases from 3.83 mg/L to 4.97 mg/L when wastewater treatment was recycled for 3 min. As seen from Fig. 4, $K_L a$ and ozone solubility increase with the high gravity factor under single absorption condition and both of them present an increasing trend. When high gravity is over 100, they tend to be flat. This is because the mass transfer process from gas phase to liquid phase belongs to liquid film control process. In premise of the same treated water yield, water temperature and ozone concentration, the higher the mixing efficiency is, the faster the ozone dissolution rate is. With the increase of high gravity factor, the liquid in the rotator is cut into fine drops, liquid wire and liquid film by packing, further increases the mass transfer area and decreases the liquid film resistance. With the drive of rotator, the turbulence level of the ozone gas and liquid increases, the ozone mass transfer process is enhanced [21]. At the same time, the high gravity technology cuts wastewater into thinner liquid films and smaller droplets to effectively increase the contact surface area of gas-liquid and mass transfer efficiency that can provide ozone with an easy access to the liquid phase so as to enhance the ozonation efficiency [22]. When the high gravity is over 100, the acceleration on ozone mass transfer is not obvious and the increase of rotating speed will result in the increase of power consumption. Therefore, an appropriate high gravity factor β specified was 100.

5.2. Influence of liquid flow-rate

It can be seen from Fig. 5 that the relationship between ozone solubility and times at different liquid flow-rates under the following conditions that initial pH value of 3.5, gas flow-rate of 75 L/h, high gravity of 100 and ozone concentration of 50 mg/L. Effects of liquid flow-rate (No liquid circulation) on ozone solubility and liquid phase mass transfer coefficient $K_L a$ are presented in Fig. 6. It can be seen from Fig. 6 that $K_L a$ increases with increasing liquid flow rate. The possible reason for this phenomenon was that a higher liquid flow rate provided a greater liquid-side mass transfer, which was caused by more liquid spreading over the packing. With an increase of liquid flow, the liquid film flow speed and wet-

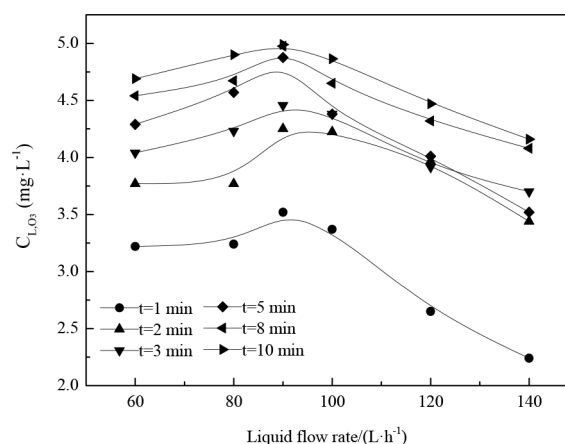


Fig. 5. Influence of liquid flow rate on ozone solubility.

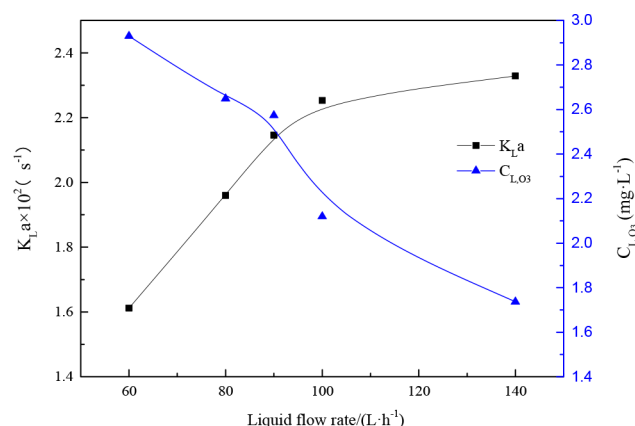


Fig. 6. Influence of liquid flow rate on K_La and ozone solubility (No liquid circulation)

ting degree on the packing surface were increased to boost the liquid film mass transfer coefficient and effective specific surface area that could facilitate the gas liquid mass transfer rate, resulting in a continuously increased liquid phase mass transfer coefficient K_La . Meanwhile, the total zone mass flow of Fig. 6 is from 175.8 mg/h to 242.3 mg/h when the liquid flow increases from 60 L/h to 140 L/h, although the ozone concentration in water decreases from 2.93 mg/L to 1.73 mg/L. However, when the gas amount is certain, it is not necessary that the more the liquid amount is, the better it is. The increase of liquid flow can increase the liquid cycle times in the same time, but it also may increase the thickness of liquid film and the size of liquid drops and obviously decreases the contact area of gas-liquid [23], which do no good to the ozone mass transfer. The ozone concentration in water decreases from 4.87 mg/L to 3.52 mg/L for 5 min cycle absorption when the liquid flow rate increases from 90 L/h to 140 L/h (Fig. 5). Therefore, in given gas amount, the proper liquid flow-rate in this paper is 90 L/h.

5.3. Influence of initial pH

Under the conditions of gas flow-rate of 75 L/h, ozone concentration of 50 mg/L, liquid flow-rate of 90 L/h and high gravity of 100, the effects of initial pH value on ozone solubility and liquid phase mass transfer coefficient K_La are presented in Figs. 7 and 8 (No liquid circulation), respectively.

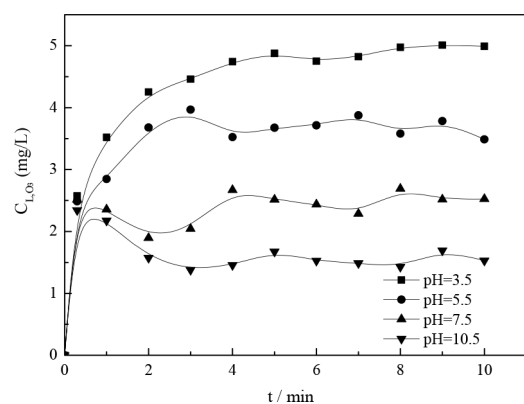


Fig. 7. Influence of initial pH value on ozone solubility.

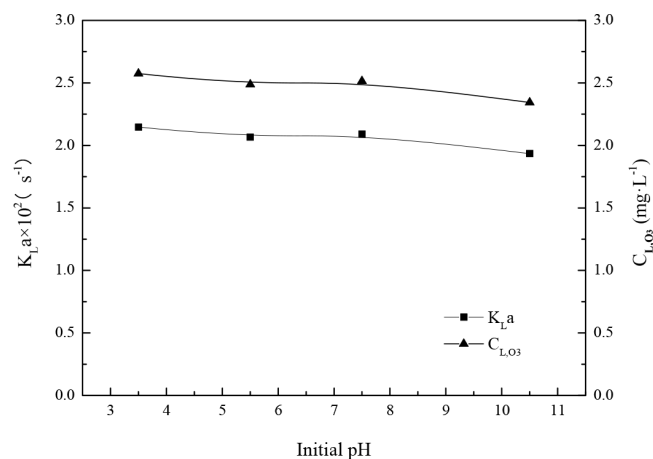


Fig. 8. Influence of initial pH value on K_La and ozone solubility (No liquid circulation).

As seen from Fig. 7, the ozone concentration in water gradually increases with the contact time in the acid environment with pH between 3.5 and 5.5. However, when pH is over 7, the ozone concentration increases firstly and then decreases and stabilizes. Gurol [24] pointed out that when pH is between 2 and 4, ozone decomposition rate basically is not relevant to pH. Martins [25] found that the ozone decomposed quickly in alkaline solution. Through the studies on ozone self-decomposition process, many scholars [26–28] have proposed various reaction mechanisms and dynamics expressions. The major reactions are shown as Eqs. (11) and (12).



It can be seen from the Eq. (11) that OH^- concentration in water increases with the increase of pH, which is easy to trigger free radical chain reaction and promote the ozone self-decomposition in water and further decreases the ozone concentration. When the initial pH of water solution decreases from 10.5 to 3.5, the ozone concentration in water improves from 1.5 mg/L to 5.0 mg/L at 8 min cycle absorption. Thus, the decreasing of solution pH can decrease the ozone self-decomposition and increase the ozone concentration. It can be seen from Fig. 8 that the increase of initial pH from 3.5 to 5.5 has few influence on the ozone solubility and K_La , which conforms to the former 20 s cycle absorption in Fig. 6. However, with the increase of cycle times and time, the ozone concentration in alkaline environment decreases obviously compared with that in the acid environment. The reason may be that in the RPB, the liquid has a very short residence time in rotator, approximately at millisecond-scale [29]. However, the ozone half-life period in water is about 30 min, so the influence of pH in extreme short time is small. However, with the extension of time, the decomposition amount of ozone in alkaline environment gradually increases and the concentration tends to decrease firstly and then stabilize. Therefore, to get a higher ozone

water solution, the pH of the solution shall be lowered in acid environment.

5.4. Influence of gas-phase ozone concentration

As shown from Figs. 9 and 10 (no liquid circulation), the effects of gas phase ozone concentration on the ozone concentration in water and liquid phase mass transfer coefficient $K_L a$ are presented under the operating conditions of initial pH value of 3.5, gas flow-rate of 75 L/h, liquid flow-rate of 90 L/h and high gravity of 100. When gas-phase ozone concentration increases from 20 mg/L to 60 mg/L, single absorption ozone concentration increases from 1.67 mg/L to 2.28 mg/L. The ozone concentration in water increases from 4.13 mg/L to 4.63 mg/L when wastewater treatment was recycled for 3 min. The equilibrium relationship between the gas-phase ozone and liquid-phase soluble ozone can be expressed as Henry's law, Eq. (1). Ozone solubility increases with the enhancement equilibrium pressure and concentration of gas ozone [30]. Therefore, increasing the gas-phase ozone concentration can speed up the ozone transfer rate and improve ozone concentration. However, the ozone concentration in water increases gradually and the mass transfer drive decreases with the extension of

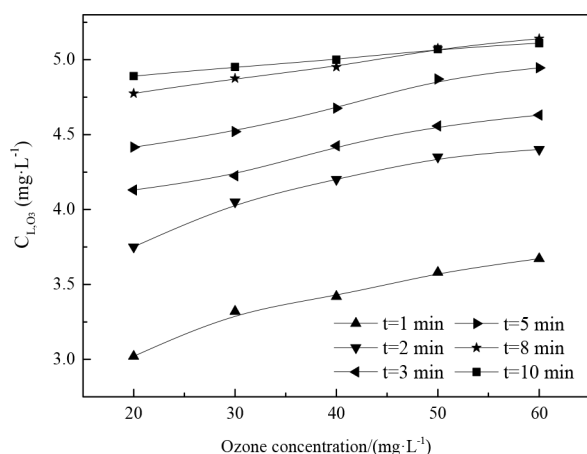


Fig. 9. Influence of ozone concentration on ozone solubility.

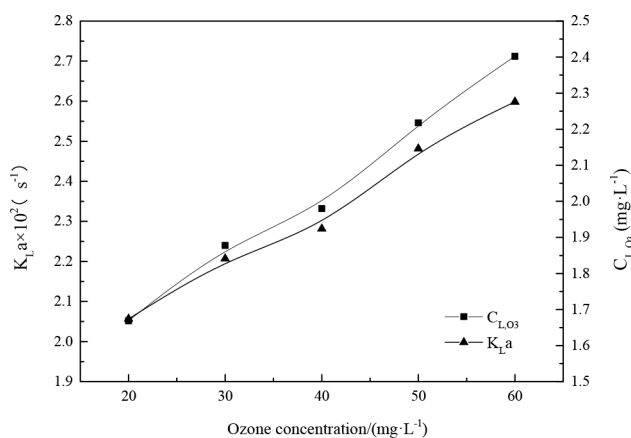


Fig. 10. Influence of ozone concentration on $K_L a$ and ozone solubility (No liquid circulation).

duration ventilation, resulting in the ozone concentration increase slowly. In a comprehensive consideration, this experiment selected the ozone concentration of 50 mg/L.

5.5. Influence of water quality

With the above-mentioned proper operating conditions, deionized water and running water are used for contrast experiment, respectively, aiming at investigating the influence of water quality on ozone solubility in high gravity environment and conventional aeration device. Results are shown in Fig. 11.

It can be seen from Fig. 11 that whether it is in high gravity environment or normal-pressure aeration device, ozone solubility in deionized water is higher than that in running water. The ozone stability in water solution is relevant to the water quality. Namely, the higher the water purity is, the slower the ozone decomposes and the better the ozone stability is. Otherwise, the poorer the water quality is, the faster the ozone decomposes and the worse the ozone stability is. There are many materials in water consuming ozone, for example, common carbonate, so the less the ozone curing agent in water is, the higher the ozone concentration is.

6. Comparative analysis of $K_L a$ between RPB and batch reactor (BR)

It is clear to see from Table 2 that liquid-phase ozone volumetric mass transfer coefficient $K_L a$ in RPB equipment is significantly higher than that in traditional aeration reaction device. For deionized water, the former one is 5.6 times of the latter. For the running water, the former one is 4.9 times of the latter. This clearly reflects that high gravity technology can effectively enhance ozone mass transfer.

7. Conclusions

- (1) Ozone dissolution rate in high gravity field is obviously higher than that in traditional aeration reaction device. Analysis indicates that the major reason lies in the enhanced ozone mass transfer of high-speed RPB on liquid cutting and dispersive action.

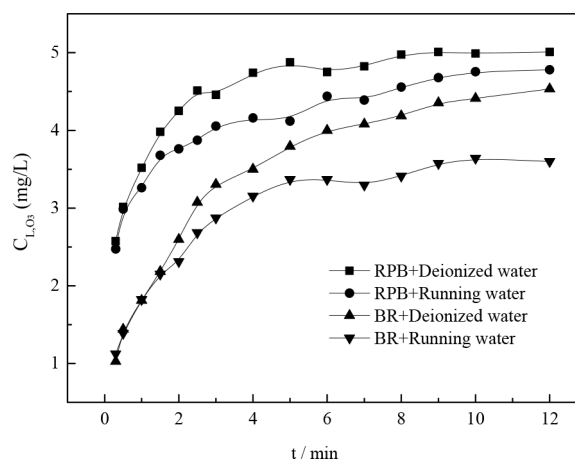


Fig. 11. Influence of water quality on ozone solubility.

Table 2
Ozone solubility and $K_L a$ in different water quality with single absorption

Water quality	C_{L,O_3} (mg/L)	$K_L a \times 10^2$ (s ⁻¹)	Liquid volume (ml)
Deionized water (RPB)	2.574	2.1454	700
Deionized water (BR)	1.025	0.3802	700
Running water (RPB)	2.473	2.0525	700
Running water (BR)	1.124	0.4185	700

- (2) In strong acid environment, the ozone concentration in water is higher due to the lower ozone decomposition. When the initial pH of deionized water is 3.5, the high gravity is 100, gas flow-rate is 75 L/h, liquid flow-rate is 90 L/h, and gas-phase ozone concentration is 50 mg/L, the single-absorption ozone concentration can reach 2.57 mg/L. The ozone concentration in water can reach 5.0 mg/L when wastewater treatment was recycled for 8 min.
- (3) Through the comparison of liquid-phase volumetric mass transfer coefficients, it can be found that $K_L a$ in RPB is significantly higher than that in traditional aeration reaction device. In case of deionized water, the former one is 5.6 times of the latter. In case of running water, the former one is 4.9 times of the latter. This clearly reflects that high gravity technology can effectively enhance ozone mass transfer.

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