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Effect of Gas Flow Rate on the Solubility and Lifetime of Liquid-Rotated Aerator Generated-Microbubbles

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Abstract

Ozone was adopted to be an alternative technology for the elimination of various kinds of micropollutants. However, ozonation is still limited by poor ozone dissolution in water. Microbubble technology was proposed to overcome this limitation. This paper was aimed to investigate the effect of inlet gas flow rate on the production and solubility of ozone in the liquid phase. A double dielectric barrier discharge (DDBD) -based ozone generator was used and connected to a microbubble aerator. Ozone concentration was measured both in outlet points and in water under various inlet gas flow rates. In addition, the lifetime of best-ozonated micro-bubbles (OMB) was investigated by storing them at two different temperatures. The application of a lower inlet gas flow rate increased ozone concentration, which can increase the mass of ozone input in the water resulted in better ozone solubility. Furthermore, the size of generated bubbles was known to be finer as the decrease of inlet gas flow rates. Lastly, we got that OMB water stored at lower temperatures resulted in longer retention time than the warmer one.

Keywords: Ozone, gas flow rate, solubility, ozone lifetime, microbubble
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1. Introduction

The removal of micropollutants in wastewater becomes increasingly important. The secondary effluent from the wastewater treatment plant usually discharged into urban rivers contains a lot of refractory organic pollutants that are inefficiently removed during conventional wastewater treatment methods. Ozone was known as a powerful oxidant, that widely used as an advanced wastewater treatment process. Ozone shows great performance in eliminating various persistent micropollutants through a process called ozonation [1].

Ozone can be generated by exciting oxygen molecules in a double dielectric barrier discharge (DDBD) which was known to produce higher purity of ozone compared to single dielectric barrier discharge (SDBD), this was due to the structure of DDBD itself which enable it to prevent inlet gas to be contaminated by its electrodes. Unfortunately, the ozonation performance is severely limited by its poor solubility in water, which is 1.0×10^{-6} mol/m^{3}Pa to 1.3×10^{-4} mol/m^{3}Pa [2]. In addition, the half-life of ozone in distilled water is known to be quite short, about 20-30 minutes at 20 ℃ and will decrease with increasing temperature [3].

Micro-bubbles technology was proposed to overcome these problems. Micro-bubbles are defined as bubbles having a diameter in a range of $1\ \mu m - 100\ \mu m$ (ISO 2017) [4], which can enhance the gas mass transfer efficiency in a liquid phase. Bubbles having a diameter of 110 μm are capable to rise up volumetric mass transfers coefficient until 100 times higher than macro bubbles [5]. Moreover, hydroxyl radicals are produced when the microbubble interface suddenly disappears. This condition can be an added advantage for eliminating recalcitrant pollutants in water [6].

Various techniques were developed to efficiently generating-bubbles such as rotary ventury type, electrostatic spraying, ultrasonic system, liquid flow type, etc [7]. Thus, In this work, a DDBD ozone generator has been investigated by characterizing the effects of inlet gas flow rates toward ozone concentration and ozone yield, also their correlation with ozone solubility under a liquid flow type based-aerator system. Furthermore, the OMB retention time in the water was also studied.

2. Methods

2.1. Treatment and Sampling Technique

In this study, ozone was generated by a double dielectric barrier discharge (DDBD). Oxygen was used as the input gas. The voltage was maintained at 2.6 kV and the oxygen flow rate was varied from 0.1 L/min to 0.5 L/min. The produced ozone by the DDBD generator was injected continuously into 10 L of distilled water for 90 minutes using different inlet oxygen flow rates. The water temperature was monitored between 25 to $27\ °C$

during the experiments. Figure 1 shows a schematic representation of this experimental setup. 10 mL samples of dissolved ozone measurement were taken every 15 minutes, meanwhile, 150 mL samples of stored ozone were collected post-treatment only.

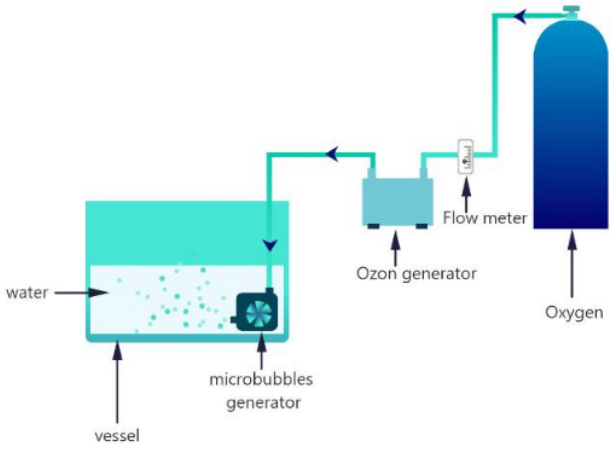

Figure 1. Experimental setup design

2.2 The Measurement of Produced Ozone

Ozone concentration (C_{O_3}) was calculated using the iodometry method and follows the below equation [8] (Nur et al. 2019), and then the ozone capacity was calculated by multiplying C_{O_3} with the corresponding flow rates [9].

$$C_{O_3} = \frac{R \times V_t \times N_t}{V_{inlet\ oxygen}} \qquad 3.2\ (1)$$

C_{O_3} is the concentration of produced ozone $(gram/L)$, V_t is the titrant ($Na_2S_2O_3$) volume (L), N_t is the normality of titrant ($Na_2S_2O_3$) 0.4 M, $V_{inlet\ oxygen}$ is the total volume of injected oxygen (L) into KI 0,2 M solution.

2.3 The Measurement of Ozone Dissolution

Micro-bubble was generated using a self-made liquid-rotating micro-bubble aerator. The micro-bubble generator consisting of a propeller to rotate inlet water at high speed around its axis and was placed in the bottom of a container. The inlet ozone was positioned perpendicularly to the axis of rotating inlet water. Such type of aerator produces micro-bubbles by pressuring air-saturated water toward a modified nozzle, and then release it into the container [10]. Dissolved ozone (D_{O_3}) was measured using *Spectroquant® Move DC* (173635) - *ozone test kit* (Merck KGaA, Germany).

2.4 The study of ozone decomposition in distilled water

Ozonated micro-bubbles-water was stored in two different temperatures, which were room temperature $(26 - 27°C)$ and cooler temperature $(3 - 4° C)$ in a plastic bottles. Both of the two groups had the same initial ozone concentrations (0.36 ppm). The decomposition of ozone was examined in distilled water each day for three days.

3. Results

3.1 The effect of Flow rate in Ozone Concentration

The characterization of the ozone generator was carried out using a high voltage AC source of 2.6 kV. In the ozone formation process, this high power is needed to facilitate the dissociation process (R-1) to produce O atoms. Then with the help of neutral atoms, these O atoms are recombined in a three-body process (R-2) to produce ozone [11,12].

$$e^- + O_2 \rightarrow 2O + e^- \quad \text{(R-1)}$$

$$O + O_2 + O_2 \rightarrow O_3 + O_2 \quad \text{(R-2)}$$

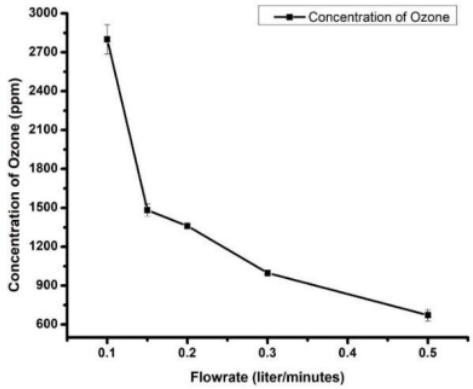

Figure 2. Ozone concentration as a function of flow rate

The effect of oxygen flow rate on ozone concentration is given by Figure 2. The maximum ozone concentration of 2800 ppm was obtained when the oxygen flow rate was 0.1 L/minute while the lowest value of 812.8 ppm was obtained when the oxygen flow rate of 0.5 L/minute. A decrease in the input gas flow rate results in a higher concentration of ozone. On the other hand, an increase in the gas flow rate results in lower ozone production.

The high gas flow rate causes the residence time of the oxygen in the DDBD gap to shorten, which means the contact time between oxygen and electrons decreases. The O atoms resulting from the dissociation process also come out faster along with a high gas flow rate. As a result, the number of O atoms available in the reactor is small, while these O atoms are needed for the three-body process to take place. This is what causes relatively small ozone production at high gas flow rates [9,13].

Ozone capacity is defined as the amount of produced ozone in a given time. Figure 3 shows ozone capacity that is proportional to the input gas flow rate. This chart shows an uptrend, but at the point of 0.15 L/min the corresponding ozone capacity drops. The maximum ozone capacity was 24.38 g/h when the input gas flow was 0.5 L/min. Wei et al [14] found that the higher flow rate leading to the decrement of input gas temperature enabling the generator to achieve maximum capacity.

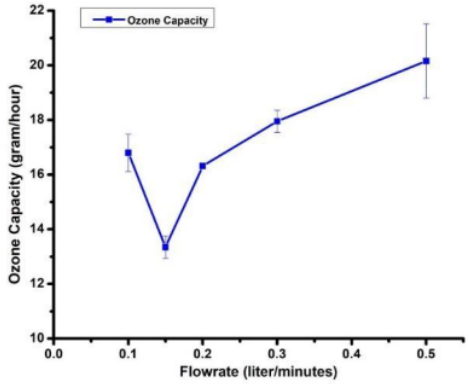

Figure 3. ozone capacity as a function of flow rate

3.2 Effect of Oxygen Flow Rate On The Dissolved Ozone

The concentrations of the dissolved ozone in water were plotted against time as seen in Figure. 4. Solid lines are the Boltzmann fit results. For a given constant gas flow rate, the dissolved ozone increase with aeration times goes by until reaching the equilibrium state. The equilibrium of ozone concentration in water increase with the decreasing gas flow rate from 0.5 L/min to 0.1 L/min. This is due to higher ozone production at lower gas streams which increases the ozone mass input for aeration. This result is similar to the previous study by Wei et al., (2017) that the dissolved ozone concentration was found to be proportional to the incoming ozone concentration.

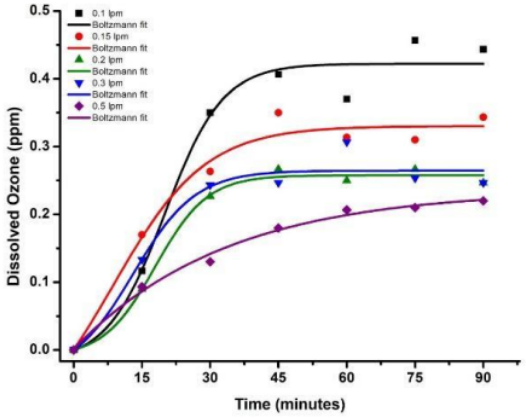

Figure 4. The effect of gas flow rate on the dissolved ozone concentration (pH 7.0; temp. $25 \pm 1\ °C$)

The gas flow rate also affects the size of the bubbles in the water. Higher gas flow rates produce larger bubbles. The larger the bubble size, the smaller the total surface area and bubble density, as a result the gas-liquid mass transfer decreases [7]. While [5] found that the average bubble size radius of the different air streams, between 1 L/min and 0.25 L/min, was the same. However, the number of microbubbles is greater at lower gas flow rates [5].

In addition to the input ozone concentration and bubble diameter, the solubility of ozone is also significantly affected by other factors such as temperature, pH, the electrical conductivity of the medium, and the presence of impurities [15].

3.3 Kinetic Decomposition

The OMB decomposition in water was plotted against time for two different storage conditions, which were the cold temperature $(3 - 4°C)$ and room temperature $(26 - 27°C)$ as illustrated by Figure 5. The fitted results are based on exponential equation for fitting (cooler temperature $(R^2 = 0.999)$ and room temperature $(R^2 = 1)$).

After three days, the ozone concentration left was 0.067 ppm and 0.06 ppm at cold temperature and room temperature respectively The ozonated water exhibited faster decay mechanism when stored at room temperature than the other group. The ozone lifetime in water could be prolonged by increasing the acidity of the water (Egorova et al. 2015). From these results, there is the potential for Micro Bubbles to prolong the binding time of ozone in the water. Although it still needs to be improved again to be used for many applications.

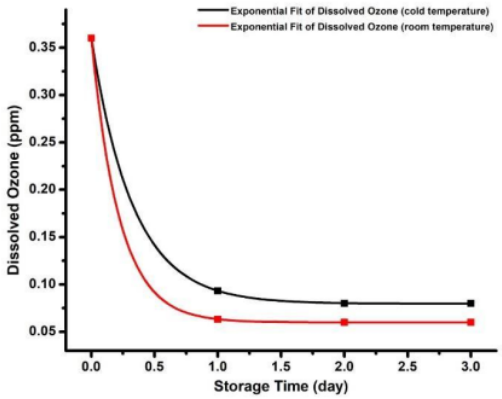

Figure 5. Kinetics of ozone decomposition in cooler temperature $(3-4°C)$ and room temperature $(26-27°C)$.

4.Conclusion

The lower flow rate of input gas led to the higher ozone concentration. This phenomenon was in reverse with ozone capacity that tends to be smaller with the decrement of input gas flow rate. In the case of dissolved ozone in water, the lower oxygen flow rate might reduce the size of the bubbles, giving better mass transfers. Increasing ozone concentration injected into water might rise ozone mass input and lead to scaling up total dissolved ozone. The ozonated micro-bubbles stored in colder temperatures were known to stay longer than those stored in room temperatures.

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