Removal of methylene blue from aqueous solution by ozone microbubbles

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Abstract— In this work, ozone microbubbles (OMBs) technique was used to remove methylene blue dye (MB) from water in a semi-batch reactor. The removal efficiency of methylene blue dye were investigated under various reaction conditions such as effect of initial solution pH, ozone generation rate, initial methylene blue dye concentration and determination of mass transfer coefficient. The removal of methylene blue by Ozonation microbubbles were very high at the acidic media and upon increasing ozone generation rate from 0.498 to 0.83 mg s⁻¹, the removal efficiency dramatically increased from 8 to 98%. The overall rate of the oxidation reaction fitted well a second order kinetic model. The results demonstrated that ozone microbubbles were effective in terms of the elimination of methylene blue concentration and its complete mineralization.

Keywords— Methylene Blue, Ozone Microbubble, Removal Rate.

1. Introduction

Industrial wastewaters are becoming more and more complex and that was owing to the attendance of emerging impurities. Water is essentially contaminated by the wastes that discharge from several industrial performances. The random discharge of these effluents into the environment without a proper remediation can cause major environmental problems. Among these contaminants, organic pollutants such as dyes, are the most important ones as they pose serious threat to the ecosystem [13]. Dyes are toxic contaminants in water and wastewater which causes remarkable threat to the environment if their concentration exceeds the acceptable limits [23]. They can remain in the environment for a long period of time if they are not addressed with an adequate treatment [20]. In fact, the direct dye discharge that containing effluents into water is unfavorable, and that’s not related to their color only, but also because various of these released dyes and their breakdown products be toxic, mutagenic or carcinogenic to life forms mainly because of the presence of carcinogens, such as naphthalene, benzidine and other aromatic components [2]. Over the years, numerous technologies have been investigated for treating dyed wastewater. These include chemical, physical and biological methods. Chemical oxidation is the generally used technique for the elimination of organics contaminated wastewater. This technique is more efficient than other methods and can be used for the treatment of some persistent pollutants. The organic contaminants can efficiently break down into smaller compounds or even convert into carbon dioxide and water by oxidants [21]. Ozonation process is a typical example of the chemical oxidation method in which the reaction rate is impacted by both the reaction kinetics and mass transfer [5]. Ozone is a strong oxidation and disinfection agent that has been widely applied in the field of water and wastewater treatment [29]. However, its use is limited by the conventional high energy consumption. In conventional reactors, dosing of ozone is achieved by inserting a gas jet into the water stream. The mass transfer is improved by turbulent flow of the gas stream, however this procedure is troubled by low mass transfer rates [19]. In addition, substantial amount of ozone is lost, which resulted in a considerable loss of energy. So that, a better gas–liquid system is required that can achieve quick oxidation of the organic compounds, and also mitigate the ozone loss. The rate of ozone mass transfer can be greatly enhanced by microbubble dispersal [19]. Microbubble technology has been widely applied for improving ozone-based oxidation processes for wastewater treatment owing to their important physicochemical properties, such as: low terminal velocity through water, large gas–liquid interfacial area,
and electrically charged gas–liquid interface [18]. These properties are of highly importance and effective in the enhancement of the rate of transfer of ozone into water and the generation of hydroxyl radical (OH) which is a strong oxidant [16]. Hence, ozone microbubbles may be an efficient alternative option for the complete elimination of MB from water.

The aim of the present work is to investigate the removal efficiency of Methylene Blue dye (MB), as an important basic dye, by ozone microbubbles using a commercial microbubble generator (microbubble diffuser). This work was undertaken to investigate the capability of ozone microbubbles in the rate treatment of wastewater contaminated with methylene blue dyes (MB). In addition, the effect of various operating conditions on the degradation of MB such as the initial concentration of MB, the pH of the reaction medium and the ozone generation rate were investigated.

2. Mechanisim of Ozonation

The presence of dyes is a major threat to the environment, and that owes to their properties which is physical (color) and chemical property (toxicity). The diversity and intensity of dyes color is a potential problem when they are discharged into water streams. Generally, the color must be decolorized previously to the subsequently step of the treatment[24]. Moreover, numerous dyes are toxic and may possibly a reason for genetic mutations. As well, the natural environment does not recognize them and does not degrade their toxicity easily due to their synthetic origin [7]. Oxidation of dye particles in general, includes the oxidant assault on the sulfenic group (methylene blue) or assault on the binary bond between N molecules (azo dyes). Despite the fact that the break of dye atoms is adequate to wipe out undesirable shading, different mixes are delivered during oxidation, which is once in a while hard to change over [9]. By far, color degradation by ozonation process is a promising choice as it provides several advantages. It has absence of chemical sludge residue afterward the process, has the capability to degrade both organic pollutant and color in single step as well as having less danger than conventional methods because no stock of any chemical is needed and the residual ozone can be easily decomposed to oxygen in water, [4]. The direct and indirect reactions of ozonation process depend significantly on the pH of the solution. At acidic solution pH, the rate of ozone decomposition was very slow and negligible; therefore, as indicated by the responses given below [30], atomic ozone responds straightforwardly with organics or inorganics and oxidizes them:

\[ O_3 + \text{Organics} \rightarrow \text{By products} + H_2O + CO_2 \]  
(1)

\[ O_3 + \text{Organics} \rightarrow HO + \text{By products} + H_2O + CO_2 \]  
(2)

When the dye molecules were oxidized by ozone, first of all it assaults the dye chromophore group as well as break down its construction [25]. In accordance with the literature, molecular ozone reacts with organics such as phenols, aromatic compounds and amines easily; however, alternatively, the reaction of aldehydes, carboxylic acids and alcohols occurs more slowly with ozone, [11]. Observably, it is able to conclude that the direct and indirect ozone reactions are selective and depends on the target compound. When pH exceeds 7, Ozone decomposition starts to occur by the reaction of hydroxyl ions (OH−) with O3. Ozone decomposition provides the formation of hydroxyl radicals and enhances the hydroxyl radical reactions with organics [22]. Chu et al. (1999) [4], have stated that molecular ozone reacts with organics at acidic pH, however, at pH values more than 7, the indirect reactions between hydroxyl radicals and organics take place because of the high rate of ozone decomposition.

Staehelin et al. (1985) [28] suggested the potential decomposition reaction of ozone as follows:

\[ O_3 + OH^- \rightarrow HO_2^- + O_2^- \]  
(3)

After the decomposition of ozone, the following reactions occur in aqueous phase:

\[ O_2^- + O_3 \rightarrow -O_3 + O_2 \]  
(4)

\[ \text{Organics} + (HO \cdot, O_2^{-}, O_3) \rightarrow \text{By products} + CO_2 + H_2O \]  
(7)

Therefore, both direct and indirect oxidation by ozone and hydroxyl radicals is important in the ozonation process for the removal of dye, TOC and COD.

3. Experimental section

3.1 Materials

The chemicals used in the current work were methylene blue dye MB (0.1M) sodium hydroxide and (0.1M) hydrochloric acid. All chemicals were of high purity and analytical grade. Stock solutions were prepared and diluted as the requirements. The chemical structure and properties of MB is shown in Figure 1.

![Figure 1: Methylene blue structure](image-url)
3.2 Experimental methods

The bench scale of ozone microbubble schematic diagram unit is shown in Figure 2. The setup comprises a column reactor, an oxygen source, an ozone generator (Shanghai ENALY M&E; model: OZX05K), and a gas distribution system (i.e. diffuser) (A CO2 Bazooka Diffuser) to generate microbubbles. As illustrated in the figure, Oxygen with high-purity was fed to a laboratory ozone generator in which oxygen was converted to ozone by the corona-discharge method. The ozone generation rates that applied in this work were studied in the range of 0.498–0.83 mg s⁻¹. Oxygen and ozone gas mixture coming out of the ozone generator was then entered a flow meter to measure its flow rate before entering the diffusers which located at the base of the column reactor where the microbubbles were generated. In the microbubble ozone generator, the dissolution of gas in water was attained by applying an oxygen flow rate of 1 L/min, so microbubbles were continuously generated by the release of pressure. All experiments were performed at room temperature (25±1°C), the initial concentration of MB in the aqueous solutions was varied in the range of 5–25 ppm, the pH of the medium was maintained by using 0.1M HCl and 0.1M NaOH solutions. The samples from the reactor were collected initially every 60 s and then after 5 min they were taken every 300 s to measure their concentrations.

3.3 Analytical method

All aqueous solutions pH was measured by the use of a pH meter (ISOLAB.). Quantitative analysis of the MB dye concentration was made by an ultraviolet UV–visible spectrophotometer (Model: Cary-100 conc., Varian, USA). The wavelength of MB dye was detected at 665 nm. The analysis were repeated at least three times.

4. Results and discussion

4.1 De-colorization of MB using ozone microbubbles

The UV spectrum of MB was determined at 665 nm as shown in Figure 3. The absorbance as well as the spectrum was rapidly changed with time owing to the fast de-colorization of the solution and the formation of the intermediates. It was observed that more than 90% of the dye was removed within 15 min of the ozonation time. The fast decrease in absorbance of MB from the maximum peak (i.e. 665 nm) indicates the quick decomposition of the dye. The strong oxidizing power of ozone as well as the much higher surface contact area available by microbubbles for the same volumetric flow rate, were effective in the fast degradation of MB from the aqueous solution. About 99% of MB was oxidized in 20 min and that agree with what was obtained by previous studies [12].

![Figure 3: Absorption spectrum of methylene blue (MB)](image)

The rapid de-colorization of MB was also visible within 10 min for all the applied MB concentration which range between 5–25 mg/L. The spectral band changed (after ~5 min) due to the destruction of the structure of MB by ozone as shown in Figure 4.

![Figure 4: MB samples before and after 20 min of Ozonation by microbubbles (MB concentration =25mg/l, pH =3.7 at room temperature)](image)

4.2 Effect of the initial concentration of MB

Dye concentrations in water and wastewater change over a wide range. Therefore, it is essential to study the effect of the concentration of MB on its degradation rate by ozone microbubbles. Experiments were carried out using
prepared model wastewater solutions with different concentration of MB (5, 10, 15, 20, and 25ppm) at room temperature. The pH of the solution and the ozone generation rate were kept constant during these experiments at 3.7 and 0.83mg/s respectively and the results are presented in Figure 5.

It can be seen from the results that the concentration profiles followed an alike trend for all concentrations of MB applied in the current study. Upon increasing the initial concentration of MB from 5ppm to 25ppm, the removal efficiency increased from about 95% to 98% within 20 min of ozonation time, indicating that more MB is oxidized by ozone at higher initial concentration. This can be understood by considering that the reaction follows second-order kinetics (first-order with respect to both MB and ozonation). Therefore, the rate of reaction increased with increasing the concentration of MB and this is accepted with other previous work [12].

4.3 Effect of the initial pH of the feed solution

It is important to investigate the effect of the solution pH on the oxidation of methylene blue (MB), since pH of the aqueous medium controls both the mechanism and the ozonation process rate [1,5]. The ions present in the ozonation system (i.e. H⁺ or OH⁻) may promote the generation of free radicals which play a vital role in the oxidation of contaminants. The ozone decomposition mechanisms in alkaline and acidic conditions have been proposed previously by several researchers and summarized in the work of Khuntia et al., 2015[18].

The time dependence of the effect of pH on the oxidation of MB for the current work is shown in Figure 6. The experiments were accomplished at an ozone generation rate of 0.83 mg/s and an initial MB concentration of 15ppm. The pH of the solution was varied from 3.7 to 9.5 to consider both the acidic and alkaline regions. It can be noticed from the results that the acidic medium, (i.e. pH 3.7) was more suitable for the oxidation of MB by ozone microbubbles, however with increasing the pH , the removal of MB decreased significantly. For 20 min ozonation time, the removals of MB were 95% and 20% at pH 3.7 and 7.1, respectively. More increase in the pH had a significant influence on the MB removal rate. Thus, it is apparent that the concentration of OH⁻, which favored the indirect reaction, increased with increasing pH [11]. In fact, Ozone reacts with organic compounds through a direct attack to the molecule and through free radicals attacks. These radicals, especially hydroxyl radicals, are generated from the ozone decomposition in aqueous solutions. However, the constant stoichiometry obtained with pH seems to indicate that in the present case, the reaction mainly occurs via the direct attack at low pH (i.e. pH 3.7) and this agree with the previous findings of Grabowsk et al. (2007)[12].

The solution pH profile with different initial pH values was also investigated and the results are plotted in Figure 7. From this figure it can be seen that the extent of pH of the solution was strongly decreased with time for all initial pH solutions considered in this study. This is expected due to the creation of organic acid intermediates during the oxidation of MB which dropped the pH of the medium [12].

![Figure 5: Effect of the initial concentration MB on the removal efficiency (pH=3.7, ozone generation rate =0.83mg/sec).](image1)

![Figure 6: The effect of pH with time on the removal efficiency of MB (T=25°C, MB concentration =15mg/l, ozone generation rate= 0.83 mg/sec).](image2)

![Figure 7: Change of pH solution with time (T=25°C, MB concentration =15mg/l, ozone generation rate= 0.83 mg/sec).](image3)
4.4 Effect of the ozone concentration:

Since, the concentration of ozone in the aqueous phase has a great influence on the MB oxidation rate, the next series of experiments were focused on studying its effect on the degradation of MB with ozone microbubbles. In the present study, OMBs experiments were investigated with different ozone generation rates ranging from 0.498 to 0.83 mg/s. The experimental data are plotted in Figure 8 which shows that, when the Ozonation rate increased, the MB removal rate increased. This is expected because increasing ozone concentration in the aqueous phase enhances the direct reaction of ozone with MB and causes to increase the rate of reaction. In addition, increasing the ozone generation rate causes an increase in the partial pressure of ozone in the gas mixture unconfined from the ozonator and these results in an augmentation in the ozone concentration in the aqueous phase, as predicted by Henry’s law.

From the data in Figure 8, it can be clearly seen that raising generation rate of ozone from 0.498 to 0.83 mg/s significantly enhanced the removal rate of MB from 37% to 98% within 20 min of ozonation time. Though, much longer time of ozonation was needed to attain similar removal level when the ozone generation rate was 0.581 mg/s. Also, it was found that increasing the ozone generation rate further (i.e. beyond 0.83 mg/s) had a small effect on the oxidation of MB.

The change in the pH solution at different Ozonation rates with time was also studied at different initial pH values and the results are plotted in Figures [9-13]. From the results presented in these figures, it is evident that with increasing Ozonation rate, the drop in the solution pH increased with time and this is due to the increase in the transfer of ozone from gas phase to liquid phase [12] which leads, in turn, to increase the decolorization rate and decrease the pH solution through time as observed in Figure 9.

Figure 9: Variation of pH with Ozonation rate at different time intervals (MB concentration =15mg/l, pH = 3.7).

Figure 10: Variation of pH with Ozonation rate at different time intervals (MB concentration =15mg/l, pH = 5.3).

Figure 11: Variation of pH with Ozonation rate at different time intervals (MB concentration =15mg/l, pH = 6.2).
be determined through kinetic studies on our semi-batch reactor. The dissolved ozone concentration in water has been calculated as a function of time and continue until it was reached saturation, which referred to the ozone concentration steady state, \([C_{AL}]_{ss}\). After that, the generation of ozone was stopped and the reduce in the ozone concentration in the reactor was determined during the experiment time as shown in Fig. 11. The time taken to achieve steady state ozone concentration decreased when the pH of the medium increased. Ozone decomposition in water depends on the pH and it was followed a pseudo first-order kinetics and that reported in in Khuntia et al., (2013) [17]; Sotelo et al., (1989) [26]; and Sotelo et al., (1987) [27]. The self-decomposition reaction rate of ozone equation can be

\[
\frac{d[C_{AL}]}{dt} = k_d[C_{AL}]
\]  

(8)

Where \([C_{AL}]\) is the ozone concentration in the aqueous solution and \(k_d\) is the rate constant of ozone self-decomposition. Equation (8) integrated with the boundary condition that at \( t = 0, [C_{AL}] = [C_{AL}]_{ss}\) and this gives the following equation:

\[
\ln \frac{[C_{AL}]_{ss}}{[C_{AL}]} = k_d t
\]

(9)

Eq. (9) predicts that \(k_d\) would be the slope of a straight line passing through the origin and this line is a plot of \(\ln \frac{[C_{AL}]_{ss}}{[C_{AL}]}\) versus time \(t\). Table 1 summarizes the experimental values of \(k_d\) at various pH.

| pH  | 5    | 7    | 9    |
|-----|------|------|------|
| \(k_d \text{ (ms}^{-1})\) | 0.039 | 0.03 | 0.025 |

\(k_d\) is a basic kinetic parameter, and it should be independent on the reactor type. On the other hand, the values investigated in the literature [27,10, 19, 15] show extensive differences from one experiment to another. For example, \(k_d\) values of 0.04 m/s at pH 7 was found by (Gao M-T et al., 2005)[10], \(k_d\) values of 0.21–0.42 /s at pH 2.5–9 was measured by (Sotelo et al., 1987)[27], 0.21–0.54 m/s at pH 2.6–7 by (Hsu et al., 2002)[15], and 0.42 m/s (pH unspecified) by (Kukuzaki et al., 2010)[19]. The values of \(k_d\) found in this study are similar to these values.
One of the parameters that have a significant effect on ozone decay is pH. So as to be aware of the influence of pH on ozone decay, a number of experiments were conducted at various pH values of 5, 7, and 9. Figure 12 illustrated the pH effect on the decay rate of ozone when the experiments were carried out in the absence of any dye. It was observed that at low pH the decay rate of is slow and it was increases when the pH increased. Moreover, that’s means; the half-life time $t_{1/2}$ of ozone was pH dependent, for that reason, any increase in the pH leads to a decrease in the $t_{1/2}$.

4.6 Determination of volumetric mass transfer coefficient $k_{La}$:

As stated before, the absorption of ozone start to increase gradually with time until it reaches to the saturation value $C_{AL}^*$. The results in Figure 15 reveal that after 30 min at any pH value, the equilibrium concentration of ozone was reached. The saturation concentration of the ozone was (0.04, 0.025 and 0.018) mol/m$^3$ at pH of 9, 7 and 5 respectively.

The kinetic calculation will be performed at various pH values to investigate the influence of pH on the volumetric mass transfer coefficient for absorption process.

- For pH = 9: $K = 0.081259$ ms$^{-1}$ and $k_{La} = 0.056259$ ms$^{-1}$. Figure 16 show the experimental and the theoretical data that obtained from equation 10 [13]:

$$C_{AL} = \frac{k_{La}}{K} C_{AL}^* (1 - \exp(-Kt))$$  \hspace{1cm} (10)

Where: ($K = k_{La} + k_d$). $k_d$ was determined from a separate experimental study as shown in Figure 14; $C_{AL}^*$ was determined from the Figure 15.

For pH = 7: $K = 0.080366$ ms$^{-1}$ and $k_{La} = 0.050366$ ms$^{-1}$. Figure 17 shows the experimental and theoretical data.

For pH = 5: $K = 0.0912$ ms$^{-1}$ and $k_{La} = 0.0522$ ms$^{-1}$. Figure 18 show the experimental and theoretical data for this case.
The results in Table (2) show the effect of pH on the constant rate of ozone absorption. When the pH increases to 9, the constant rate of absorption also increases. pH was taken as a vital parameter in ozone reaction pathway. Molecular ozone oxidation has a tendency to dominate in an acidic condition whereas, free-radical oxidation is dominated at higher pH. This owes to the generation of hydroxyl radicals is improved in an alkaline condition. So, these radicals have an oxidation potential further more than ozone molecular [14], then the removal efficiencies of MB are expected to increase as alkalinity increases.

**Table 2:** The volumetric mass transfer at different pH and ozonation rate of 0.83 mg/s:

| pH  | $k_{La}, \text{ms}^{-1}$ |
|-----|-----------------|
| 5   | 0.0522          |
| 7   | 0.050366        |
| 9   | 0.056259        |

4.7 **Removal of chemical oxygen demand (COD) from MB aqueous solution**

In general, several organic chemicals such as dyes, biocides, ionic and non-ionic surfactants, carriers, sizing agents are contained in industrial wastewaters. As a result, it was difficult to establish the degradation rate for each organic compound present in wastewater individually. Hence, some worldwide waste water parameters such as COD were used as an indication for the degradation kinetics of organic compounds by Ozonation. For this purpose, experiments were carried out in order to assess the effect of ozonation process on the COD removal efficiency.

Figure 19 indicates the change of COD values of MB model solution with concentration of 100 mg/l after treatment with ozone microbubbles. As can be seen from the results, the color removal rate was increased with time. Within the first thirty minutes of the experiment, the color removal rate was around 45%. Between 30 and 60 min, these rate of removal continued to increase rapidly, however, after 60 min a slowdown has been noticed.

The decrease in the COD concentration of MB aqueous solution might be caused by the decomposition of MB by Ozonation which leads in turn to the formation of sulfoxides (S=O) in high yield. JAVIER et al., (1993)[8] found that 1 mole of ozone is required in this step. Moreover, another molecule of ozone can react with the N atom to give a hydroxylamine derivative which can be attacked later by a second molecule of ozone to yield a nitro compound [8].

5. **Conclusions**

This study was focused on investigating the efficiency of using ozone microbubbles for the oxidation of methylene blue dye (MB) from aqueous solutions. The data obtained from this work showed that this technique was very effective in removing MB from liquid medium. From the experimental work, it was found that acidic condition enhances the separation efficiency of MB. Removal efficiencies of about 98% was obtained under conditions of pH 3.7 and ozone generation rate of 0.83 mg/s, it was found that the direct reaction of MB with molecular ozone was the dominant during the separation process, while the reaction between MB and hydroxyl radical was inactive and it cannot degrade more than 20% of the dye at alkaline condition. It was also found that increasing the ozone generation rate, increases the concentration of dissolved ozone in the aqueous phase, which enhanced the degradation of MB at pH 3.7. Complete mineralization was accomplished at low pH 3.7 and 0.83 mg s^{-1} ozone generation rate. The COD removal rate for wastewater with MB initial concentration of 100ppm was approximately 64% after 65 min of treatment. Also, the oxidation of MB with ozone followed second-order rate constant.
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azalatat siba3 at min halawat el-mahall el-mani bi wasta3a taghayvat el-auroz el-sawhirat

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الخلاصة – في هذا العمل، تم استخدام تقنية فقاعات الأوزون الصغيرة لإزالة صبغة الميثيلين الزرقاء. تم التأكد من كفاءة إزالة صبغة الميثيلين الزرقاء بواسطة هذه الفقاعات في ظروف التفاعل المختلفة مثل تأثير درجة الحموضة في محلول الأولي ومعادل توليد الأوزون، وتركيز الصبغة الأولي وتحديد معدل نقل الكتلة. كانت عملية إزالة الصبغة بواسطة هذه الفقاعات عالية جدا في الوسط الحامضي، وعند زيادة معدل توليد الأوزون من 0.421 إلى 0.13 ملغ في الثانية، زادت كفاءة الإزالة بشكل كبير من 8 إلى 98٪. أظهرت النتائج أن استخدام تقنية فقاعات الأوزون الصغيرة فعالة جدا في إزالة هذه الصبغة كليا.

الكلمات الرئيسية – الميثيلين الزرقاء، فقاعات الأوزون، معدل الإزالة.