



Continuous ozonation of methylene blue in water



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ARTICLE INFO

Article history:

Received 13 July 2015

Received in revised form

24 September 2015

Accepted 8 October 2015

Available online 21 October 2015

Keywords:

Wastewater treatment

Ozonation

Reaction engineering

Textile effluent

Model development

ABSTRACT

Dyes are the main pollutants in the textile industry effluents. To provide solutions to the environmental problem, a model dye, methylene blue (MB), was removed from aqueous solution by ozonation in a continuous reactor. The effect of liquid circulation on MB removal was investigated and an optimal circulation rate was found under given experimental conditions. Models based on film theory were developed to predict concentration profiles of MB, dissolved ozone and gaseous ozone for the continuous ozonation process. Validation of the models with experimental data indicates that the proposed models can successfully predict the concentration profiles. Evaluation of the models also reveals that the fast kinetic regime assumption was applicable for the mass balance model of MB, however, as evidenced by the presence of dissolved ozone shortly after the reaction started, the intermediate rate assumption better represented the variations of ozone concentration both in liquid phase and in gas phase.

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1. Introduction

Removal of dyes from the large amount of textile effluents has been a challenge for the textile industries. Due to the presence of up to 50% of the input dyes, the heavily colored effluents not only cause aesthetic problems, but also are considerable environmental concern for the toxicity and potential carcinogenicity of certain organic dyes [21,24,18]. Since dyes are intentionally designed to resist chemical and microbial attacks to keep the color stability, conventional biological wastewater treatment methods are ineffective in removing the color [8,24,20,25,12].

Ozone is a strong oxidant and ozonation is very effective in decolorizing textile wastewater. Ozonation can also convert biorefractory dyes in wastewater into biodegradable species so that biological treatment can follow [22,23,14,18,15,26]. Consequently, ozonation has been increasingly applied for textile wastewater treatment [16,11,26]. However, most of the researches were conducted in batch or semi-batch mode, little attention has been paid for the more practical continuous ozonation processes. In fact, continuous processes have been used more commonly in industries than the batch or semi-batch ones, therefore, our research focused on continuous ozonation processes.

In this study, the aqueous solution of a model dye, methylene blue (MB), was ozonated in a continuous reactor system. In addition to explore the effect of the liquid circulation on the dye removal,

models were developed to predict the concentration profiles of MB, dissolved ozone and gaseous ozone during the continuous process. The models were based on the film theory and took into account the influence of mass transfer and chemical reaction on the ozone absorption rate. The models were also validated against experimental data.

2. Experimental

2.1. Materials and methods

Reagent grade methylene blue (MB) was purchased from Sigma–Aldrich (Oakville, Ontario, Canada). All other chemicals used in this study were reagent grade and obtained either from Sigma–Aldrich or VWR (Mississauga, Ontario, Canada).

The concentration of MB was followed by measuring the solution absorbance at the wavelength of maximum absorbance of MB, 664 nm, using a Biochrom Ultrospec 50 UV Visible Spectrophotometer obtained from VWR [19,25]. The dissolved ozone concentration was determined by the standard indigo method [1].

2.2. Apparatus and procedure

Ozone was produced from pure oxygen gas by a model GL-1 ozone generator (PCI-WEDECO, Environmental Technologies, Charlotte, NC). The inlet oxygen flowrate to the generator was 9.4 L/min (20 SCFH). The ozone concentration in the output of the generator was readily adjustable by varying the power of the generator.

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Table 1
Experimental conditions used in this study.

Concentration of MB at the reactor inlet, C_{MB_i} , (M)	3.12×10^{-5}
Liquid flow rate, v_L , (L/min)	0.1
Concentration of gaseous ozone in feed gas, $C_{O_{3gt}}$, (M)	$(3.1-9.3) \times 10^{-4}$
Gas flow rate, v_G , (L/min)	1
Temperature, T , ($^{\circ}$ C)	15 and 25
pH	8.0
Liquid circulation rate, F_{cir} , (L/min)	1–3

Ozone concentration in the gas phase was measured by an OZOCAN ozone monitor (OZOCAN Corporation, Toronto, Canada).

The aqueous MB solutions were prepared in the feeding tank. The MB solutions were pH-buffered (5×10^{-3} M phosphate) with a MB concentration of 10 mg/L (3.12×10^{-5} M). The pH of the solution was 8.0.

Experiments were conducted in continuous mode. Fig. 1 shows the experimental setup. The reactor system consisted of a 3 L lower reservoir and a packed column. The packing of the column was 1 cm polyvinyl chloride (PVC) balls which were supported by a glass-filled TFE packing support disc. The total volume of the reservoir and the void volume of the column was 4.99 L. The ozone containing gas from ozone generator was bubbled into the reservoir by a 25×70 mm extra coarse gas diffuser. Excess ozone leaving the column was destroyed by a catalytic ozone-destruct unit filled with Carulite catalyst (Carus Chemical Company, Peru, IL). Initially, 2.5 L of MB solution was pumped from the feeding tank to the reservoir and circulated through the heat exchanger and the packed column to reach the desired temperature. Then 0.1 L/min of MB solution was fed to the reactor system continuously. Once the MB solution reached steady state flowrate, the reaction started by bubbling the ozone containing gas into the reservoir through the diffuser. When the experiment was conducted with liquid circulation, according to the set circulation rate, a specific portion of liquid would be pumped back to the reservoir through the heat exchanger and the column, with the remaining portion going to the receiving tank. During the continuous operation, samples were collected at specific intervals from the outlet liquid stream. The experimental conditions are listed in Table 1.

Experiments were also conducted to determine the residence time distribution by pulse injecting 3 mL of 100 mg/L (3.12×10^{-4} M) MB at the inlet liquid stream to the reactor system with steady liquid flow rate. The concentration of MB was monitored at the reactor outlet stream. The injections were performed at 0, 1, 2 and 3 L/min of liquid circulation rates, respectively.

3. Results and discussion

3.1. Effect of liquid circulation rate

Ozonation of MB solutions was conducted at different liquid circulation rate to investigate its effects on the removal of MB and on the concentration of dissolved ozone, respectively. Circulation rates of 1, 2 and 3 L/min were tested.

Fig. 2 shows the effect of liquid circulation rate on the removal of MB. CMB is the concentration of MB in the solution at time t . It can be observed from Fig. 2 that the removal of MB increased with increase in the liquid circulation rate until the circulation rate reached 2 L/min. The liquid circulation rate played an important role on ozone removal of MB. For example, when liquid circulation rate was 2 L/min, ozonation for 4 min removed 95.3% of MB whereas only 28.2% of MB were removed without liquid circulation. It can also be found from Fig. 2 that further increase in the liquid circulation rate beyond 2 L/min only resulted in negligible improvement in MB removal. Therefore, 2 L/min was considered the optimum liquid circulation rate and was used for all the subsequent experiments.

The increase in MB removal with the increase in liquid circulation rate should be the results of two factors. Firstly, the increase in liquid circulation rate resulted in the increase in reactants' resident time, which in turn resulted in longer contact time between the reactants, i.e., MB and dissolved ozone, consequently, more MB was removed. Secondly, the increased liquid circulation rate also improved the mixing and eliminated the dead zones in the continuous reaction system [6] so that MB removal was enhanced. However, when the removal of MB by ozone reached its maximum after certain contact time under the given reaction conditions and the mixing could not be improved further, a limiting liquid circulation rate was reached, which is 2 L/min in this case.

The dissolved ozone concentration (C_{O_3}) was also measured at different liquid circulation rate. It has to be mentioned that there was no dissolved ozone detected when the liquid circulation rate was zero, but at the same time, MB was still present as shown in Fig. 2. This is probably because the reaction between ozone and MB was fast and mass transfer limited, and occurred within the film. Therefore, ozone was consumed in the film instead of being transferred to the bulk of the liquid [13,9].

Fig. 3 shows the effect of liquid circulation rate on dissolved ozone concentration. As can be observed from Fig. 3, the dissolved ozone concentration was lower at the liquid circulation rate of 1 L/min than those at 2 and 3 L/min. A comparison of Fig. 3 with Fig. 2 reveals that the build-up concentration of dissolved ozone started approximately at the time when MB concentration became very low. With more liquid back to the reactor system, the concentration of dissolved ozone was getting higher until the ozone equilibrium concentration under the experimental conditions was reached. The presence of dissolved ozone in the bulk liquid may indicate the slowdown of the reaction.

3.2. Distribution of residence time of the reactor system

Generally, the real reactor system, as the one under consideration, does not behave as an ideal reactor with perfect mixing. It is necessary to determine the residence time distribution and the mean residence time for such a system [6]. In addition, determination of the mean residence time is also necessary in modeling the process so that the model developed could represent the real situation.

The residence time distribution, $E(t)$ can be determined by [6]:

$$E(t) = \frac{C_{MB}}{\int_0^{\infty} C_{MB} dt} \quad (1)$$

and the mean residence time, t_m , was defined as:

$$t_m = \int_0^{\infty} tE(t) dt \quad (2)$$

Results of $E(t)$ are shown in Fig. 4. It is observed from Fig. 4 that the residence time distribution has the sharpest peak shortly after the MB injection when there was no liquid circulation. As the liquid circulation rate increased, the peak started to decrease. For circulation rate of 2 L/min, the peak reached the minimum. Negligible change in $E(t)$ profile was observed when the circulation rate increased from 2 to 3 L/min. Since $E(t)$ is the function that describes in a quantitative manner how much time different fluid elements have spent in the reactor [6], results shown in Fig. 4 indicate that with liquid circulation, most of the fluid elements stayed longer in the reactor system than those without circulation, and the higher the circulation rate, the more liquid elements stayed longer until the circulation rate reached 2 L/min. On the other hand, the mean residence time, t_m , corresponding to these $E(t)$ profiles were found to be 15.4, 18.4, 27, and 26.2 min for the liquid circu-

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