Packed bed reactor for efficient dye removal using ozone and hydrogen peroxide

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ABSTRACT

This study aimed to investigate the degradation of Congo red and Rhodamine B dyes in synthetic wastewater by ozonation and study the parameters that influence the efficiencies of color removal such as concentration of pollutants, gas flow rate, time of reaction, and the pH of solution. Congo red dye and Rhodamine B dye were selected as model pollutants. ozonation process of these dyes was carried out in a continuous reactor with a diameter of 8 cm and length of 25 cm. It was observed that over 85% of Congo red and 95% of Rhodamine B could be removed at 90 min for both dyes. and when using the H_2O_2 (0.5, 1, 1.5) ml for Rhodamine B and Cong Red at 90 min the decolorization efficiency in natural pH was 93%, 95%, 96%, 84%, 89%, and 94% respectively. When used the packing the decolorization efficiency changed from 77% to 82% for Rhodamine B and from 53% to 63% for Congo Red. Higher color removal could be achieved when ozonation was carried out in an alkaline condition (pH 10) for Congo Red and an acidic condition (pH 1.5) for Rhodamine B. The ozonation of dyes was dominant at room temperature and atmospheric pressure.

Keywords: wastewater, ozonation, dye, Congo Red, Rhodamine B, decolorization, H_2O_2, packing.

INTRODUCTION

The contamination of water bodies with pollutants has shown a concerning upward trend in various regions worldwide, especially in areas at risk of high contamination, such as across Africa, the Americas Europe, and Asia (Mojiri et al., 2019). Recently, there has been concern about the presence of several organic micropollutant (OMP) residues at deficient concentrations (ng L^{-1}) in the environment, including soil, surface, and groundwater, and the possible impact on water-dwelling animals and the uncertain effect on human health (Rozas et al., 2017). Aquatic organic matter may be divided into three categories: a) particulate organic matter, which is the fraction that is kept in an arbitrarily specified 0.45 µm filter pore; b) dissolved organic matter (DOM), which is the fraction that flows through this 0.45 µm filter; and c) colloidal organic matter, which has diameters ranging from 1 nm to 1 µm (Artifon et al., 2019). Their elevated concentrations can potentially yield deleterious ramifications on

potable water quality (Loganathan et al., 2022). NOM's different components, including polysaccharides, hydrocarbons, amino acids, fatty acids, carbohydrates, proteins, and other compounds like zooplankton and humic acid complexes—mold aquatic environments with certain features (Ghernaout et al., 2010). Continuous uncontrolled release of pollutants and waste materials into the aquatic environment from various sources, including industrial processes, agricultural runoff, and household sewage, accounts for their widespread dispersal (Hakeem et al., 2020). These contaminants in natural water sources such as surface or groundwater constitute a significant risk to human health and ecological integrity (Machado et al., 2016). They may include organic compounds, fertilizers, pesticides, and heavy metals. If this problem isn't properly investigated and solved, then the quality of drinking water will continue to deteriorate even more (Lu et al., 2020). Different human activities practiced in residential households, manufacturing, and

production facilities result in a large volume of wastewater with varying chemical compositions; its management and treatment are indispensable if our environment is to be saved (Oller et al., 2011). The textile industry is using an increasing amount of pigments and dyes to color its products. It is believed that over 100,000 dyes have become commercially available in recent years (Cuiping et al., 2011). Wastewater from the textile sector, which sometimes contains high levels of complex organic dyes, is a big concern for the environment (Faryadi et al., 2016). Dyeing is the stage in the processing of precious stones with the greatest environmental impact because it changes the color of water (turbidity), its natural pH, its conductivity, the chemical oxygen demand (COD), the biochemical oxygen demand (BOD), solubilized nutrients (e.g., nitrogen and phosphorus), and also contaminates and pollutes the water with heavy metals (Machado et al., 2012). There are several processes to remove organic pollutants from wastewater.

Biological treatment

Degradation of a chemical by living organisms, typically microbes like bacteria and fungi found in soil and water, is known as biological degradation. These organisms eliminate the pollutants through metabolic processes (Oller et al., 2011). Biological treatment includes constructed wetlands (Cuiping et al., 2011), bioremediation (Faryadi et al., 2016), and activated sludge process (Modin et al., 2016). Traditional biological treatment techniques for dye-contaminated wastewater have limited efficiency since most dyes are chemically resistant and show poor biodegradability (Aksu, 2005).

Physical and chemical treatment

Physical water treatment techniques rely on physical forces to remove contaminants without altering their chemical composition. These methods are different from chemical treatment, which involves chemical reactions to transform or eliminate contaminants (Hanafi and Sapawe, 2020). Examples of the physical water treatment process include:

- chemical precipitation: in this process, the chemicals used form insoluble precipitates of the contaminants, which can then be removed by filtration or sedimentation (Reyes-Serrano et al., 2020);

- adsorption: it is the process of attachment of contaminants to the surface of a solid material (adsorbent) (Khudair and Abdulaziz, 2024);
- flotation: in this process, air bubbles are used and attach themselves to contaminants which makes them float on top for removal (Peleka et al., 2018);
- biosorption: this process involves adsorption of contaminants using microorganisms or their byproducts (Argun et al., 2016);
- ultrafiltration: a membrane is used with very small pores that filter out contaminants based on their size (Awad et al., 2021);
- electrochemical techniques: they are those processes using electrical currents that disinfect through electrolysis or electrocoagulation (Davarnejad and Sahraei, 2016);
- coagulation: this is a process in which chemicals, and coagulants, are introduced into a solution to destabilize the suspended particles so that they clump into larger particles, which are then easier to remove (Sukmana et al., 2021).

Ozonation process

One of the dye waste treatments is the ozonation method (Aziz et al., 2018). Ozone plays a crucial role as an oxidant in drinking water treatment due to its powerful oxidation and disinfection (Tripathi and Hussain, 2021). When ozone is used in water treatment, it works to eliminate highly reactive free radicals that quickly combine with other molecules. This reactivity makes them important for breaking down pollutants in the air and decomposing natural materials (Brodowska et al., 2018). They are also crucial for advanced water treatment and environmental cleanup processes (Gogate and Pandit, 2004). The recent increase in the use of ozone applications can be attributed to significant developments. First, Over the past decade, there has been a substantial reduction in the costs linked to ozone production. Second, the environmental benefits of using ozone over chlorine have become more apparent. Ozone has emerged as an efficient alternative for treating wastewater that has complex metals of organic origin (Altmann et al., 2014).

Ozone has a strong ability to break down organic pollutants. It does this in two ways: (1) by directly attacking with ozone molecules, and (2) by indirectly attacking with OH• radicals produced during the ozone decomposition process (Psaltou and Zouboulis, 2020). The oxidation potential of

ozone is 2.07 V (Lv et al., 2021). Ozone, with an oxidation potential of E^0 -2.07 V, surpasses other well-known oxidants such as hydrogen peroxide (H_2O_2) due to its superior ability to undergo oxidation reactions (Neamţu et al., 2013). This higher oxidation potential makes ozone a highly effective oxidizing agent in various applications. As a result, highly reactive species such as hydroxyl radicals (·OH), superoxide radicals (·O_2), and others can effectively expedite the decomposition and mineralization processes of toxic materials. This property makes ozone a potent agent for facilitating the breakdown of harmful substances in various applications (Rekhate and Srivastava, 2020).Ozone selectively interacts with unsaturated bonds present in organic molecules, forming saturated compounds as by-products. Ozone is used to remove the color from effluents by targeting the double bonds of functional groups (Ghuge and Saroha, 2018).

A thorough examination of existing literature revealed that the majority of research on dye removal by ozone has concentrated on acidic conditions and employed batch reactors. This fact motivated researchers to investigate the impact of altering these variables on the efficacy of the removal process, This research employs a novel strategy to address this knowledge gap. Experiments are structured to encompass a broad spectrum of pH values, from acidic to alkaline, to ascertain the best range for the oxidation process. A continuous reactor was employed to more accurately simulate the process conditions and enhance processing efficiency, To improve the efficacy of the removal procedure, hydrogen peroxide was amalgamated with ozone in certain tests to leverage their synergistic interactions. Gaskets were utilized within the reactor to augment the surface area for reaction and enhance material transmission. This research aims to enhance comprehension of ozone's mechanism in dye removal and to furnish critical design data for the extensive deployment of this technology in industrial water treatment, And also the effect of hydrogen peroxide on decolorizaion dyes.

MATERIAL AND METHODS

Table 1 illustrates the chemicals used in the experiment, while Figure 1 schematic diagram of the experimental setup of the ozonation process. The equipment used in this research is a continuous reactor (diameter 8 cm and length 25 cm), a beaker (ISO LAB, Germany) with 5 L volume, a bubble gas diffuser (Liay Iize aquarium, China), and a UV visible spectrophotometer (Model 6800, Japan). The ozonation investigations were conducted using a continuous-mode reactor. The primary variables that altered were pH (1.5, 3, 7, 10), dye concentration (10, 25, 50, 75 mg/L for Rhodamine B and Congo Red), ozone flow rate (0.5, 1, 1.5, 2 L/min), hydrogen peroxide (0.5, 1, 1.5) ml, and reaction time (90 minutes). Samples were collected at regular intervals to monitor the decline in dye concentration while ozone was introduced into the reactor's bottom. All experiments were carried out at atmospheric pressure and ambient temperature. By measuring the solution's absorption intensity, the decolorization efficiency was determined.

The following is the definition of dye decolorization efficiency (%):

$$Decolorization\ efficiency\ (\%) = (1 - C_i/C_o) \times 100 \quad (1)$$

where: C_0 and C_i are concentrations of dyes in an aqueous solution at the initial condition and the outlet stream, respectively.

RESULT AND DISCUSSION

Removing Rhodamine B dye in various conditions

Effect of initial pH on Rhodamine B dye

The pH of the solution is one of the most critical variables in ozonation since it influences the process's mechanism and rate (Von Sonntag and Von Gunten, 2012). The initial pH's impact on the decolorization of Rhodamine B is a significant factor in the ozonation
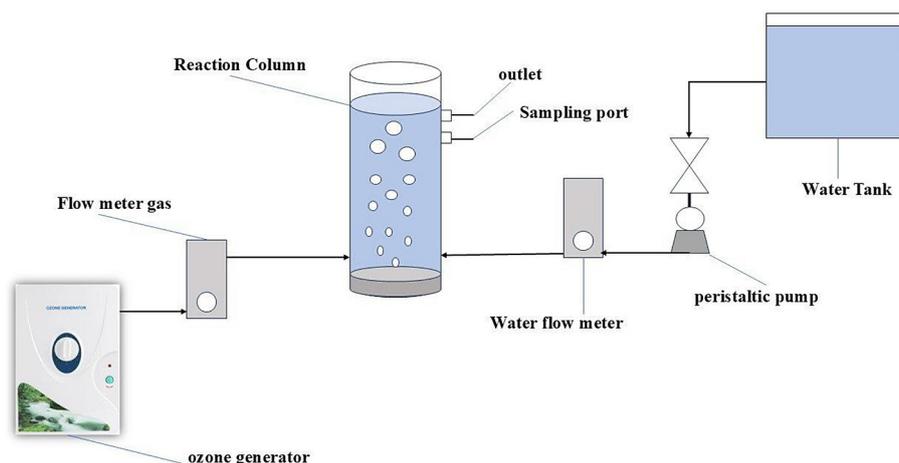
Table 1. The chemicals used in the experiment

Chemicals	Chemical formula	Purity	Origin
Congo Red	$C_{32}H_{22}N_6Na_2O_6S_2$	99%	India
Rhodamine B	$C_{28}H_{31}ClN_2O_3$	97%	Standard Fluka, Sigma-Aldrich, USA
Sodium hydroxide	NaOH	97%	Germany
Hydrochloric acid	HCl	35-38%	Germany
Distilled water	H_2O	99% purity	Iraq
Hydrogen peroxide	H_2O_2	50%	Spain

Figure 1. Schematic diagram of the experimental setup of the ozonation process

process. Ozone oxidation pathways typically encompass direct oxidation by ozone as well as radical oxidation facilitated by the HO· radical. Direct oxidation exhibits greater selectivity and is more prevalent in acidic conditions (Ku et al., 2000). To study the effect of pH on the degradation rate of Rhodamine B, the initial pH of the solution was varied in the range of (1.5, 3, 7, and 10).The removal efficiencies at various pH values as a function of ozonation time are presented in Figure 2, which shows the maximum removal rate for Rhodamine was achieved at acidic conditions with a pH value of (1.5) at 90 min. The decolorization rate under acidic conditions surpasses that of alkaline conditions due to the elevated pH in the solution, which generates a greater number of free

radical scavengers (such as CO_3^{2-}, HCO_3^-, etc.) from the mineralization of organic material. This process ultimately leads to a reduction in the concentration of HO· radicals and these results are very similar to the study on the degradation of Rh B (Song et al., 2009).

Effect of inlet concentration of Rhodamine B dye

The inlet dye concentration has a significant impact on the cost-effectiveness of ozone decolorization. Several investigations have revealed that the time required for the decolorization of the dye solution depends on both inlet dye concentration and ozone consumption (Hsu et al., 2001). The impact of the initial concentration of rhodamine B on the
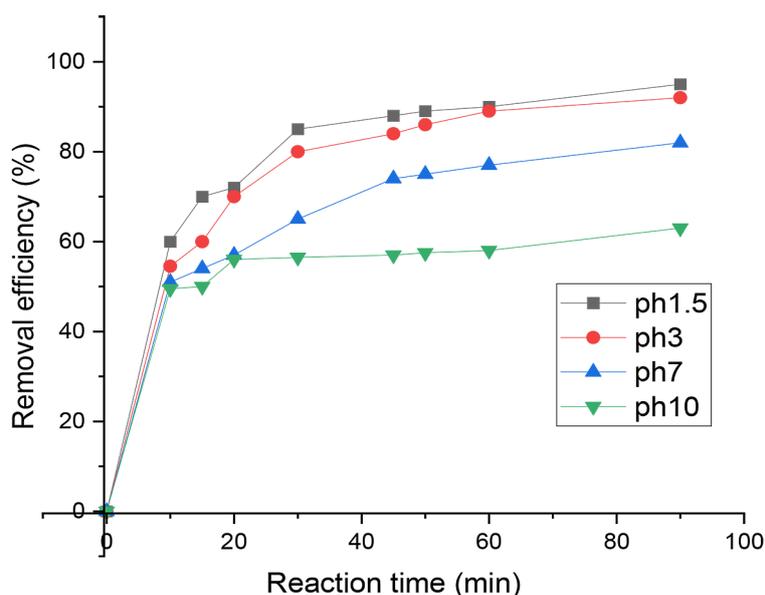

Figure 2. Effect of pH on the dye decolorization of Rhodamine B. 2 L/min gas flow rate, 10 mg/L, 90 min

decolorization of the dye by ozone was examined within the range of 10 to 75 mg/l. The results are presented in Figure 3. The decolorization rate of rhodamine B decreased as the initial concentration increased during the treatment process. The decolorization rate was observed to decrease from 95% to 13.54% as the initial concentration of rhodamine B increased from 10 mg/l to 75 mg/l. The quantity of HO· radicals in the system would remain constant under the same conditions, as the ozone flow rate in effluent is constant at a specific temperature (Bouasla et al., 2010).

Higher concentrations of rhodamine B consume more HO· radical, reducing the impact as the original concentration increases (Cuiping et al., 2011). The observed results can be attributed to the elevated levels of dye oxidation by-products that occur with increasing dye concentration, which in turn enhances ozone consumption. Ozone has demonstrated effectiveness in the treatment of highly concentrated dye solutions; however, to attain greater decolorization efficiencies, increased ozone doses will be necessary and these results are similar to this study (Zawadzki and Deska, 2021)

Effect of operating time for Rhodamine B dye removal

Figure 4 clearly depicts this relationship, as the depigmentation curve begins with a dramatic spike and then gradually begins to stabilise until it reaches an equilibrium state after 90 minutes. This motor behaviour implies that the elimination process occurs in stages, with the initial phase being quick due to a high dye concentration and then gradually slowing down as the dye concentration declines. Based on these findings, it can be inferred that process time control is an important aspect in increasing the efficiency of coloring processes. However, it should be noted that the ideal time may vary based on the type of dye and other experimental settings. (Zawadzki and Deska, 2021).

Effect gas flow rate on Rhodamine B

Different gas flow rates (0.5 to 2 L/min), a dye concentration of 10 mg/l, a reaction time of 90 minutes, and a pH of 1.5 were used in the experiments. The experiment kept these values the same, and the results are shown in Figure 5. The data shows that the dye degradation rate increased as the gas flow rate increased. At a gas flow rate of 0.5 L/min, the removal efficiency for Rhodamine B was 58.8%. Increasing the gas flow rate to 1.5 L/min resulted in an enhanced removal rate of 89%, while a flow rate of 2 l/min achieved a removal rate of 93.5%. Dye compounds react with gaseous ozone that has been introduced through microbubbles into the aqueous phase. It is well-known that the quantity of ozone present on the liquid side directly affects the amount of
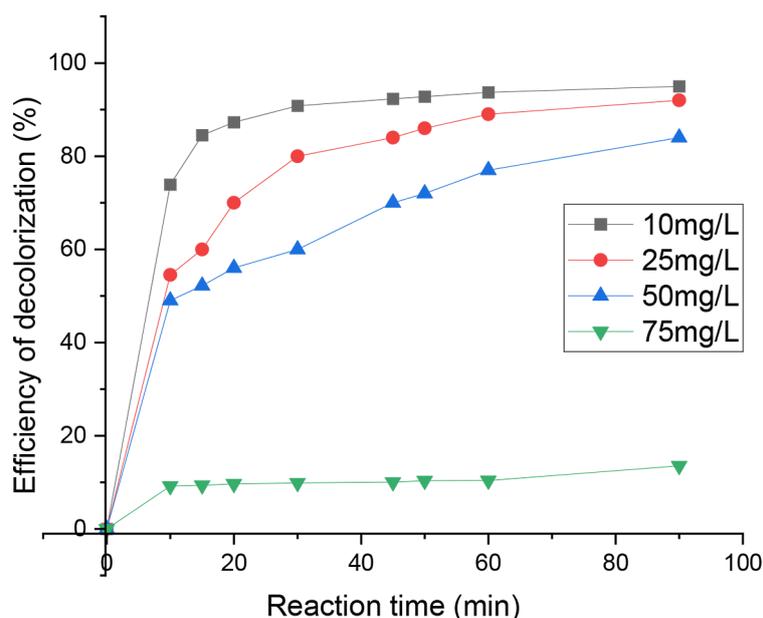

Figure 3. Effect of initial concentration of Rhodamine B on the dye decolorization. gas flow rate = 2 L/min, pH = 1.5, and 90 min
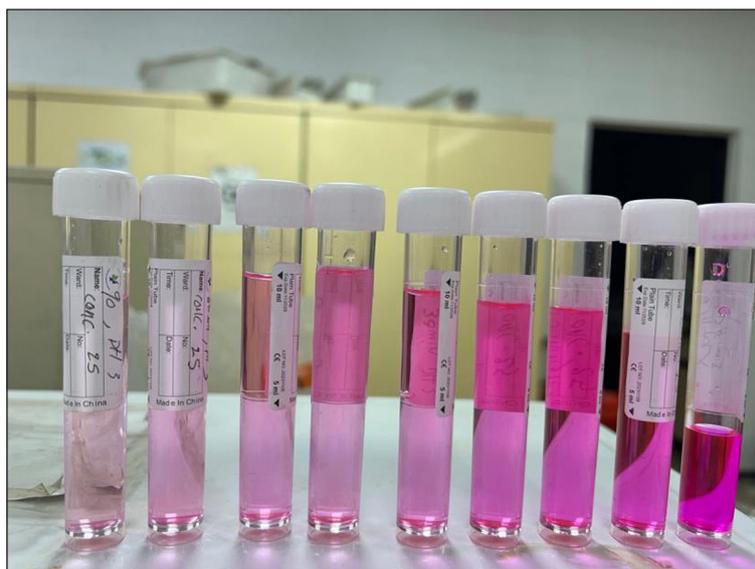
Figure 4. Samples of Rhodamine B concentration with time. pH = 1.5, dye concentration = 10 mg/L, gas flow rate was 2 L/min, and the reaction time was 90 min
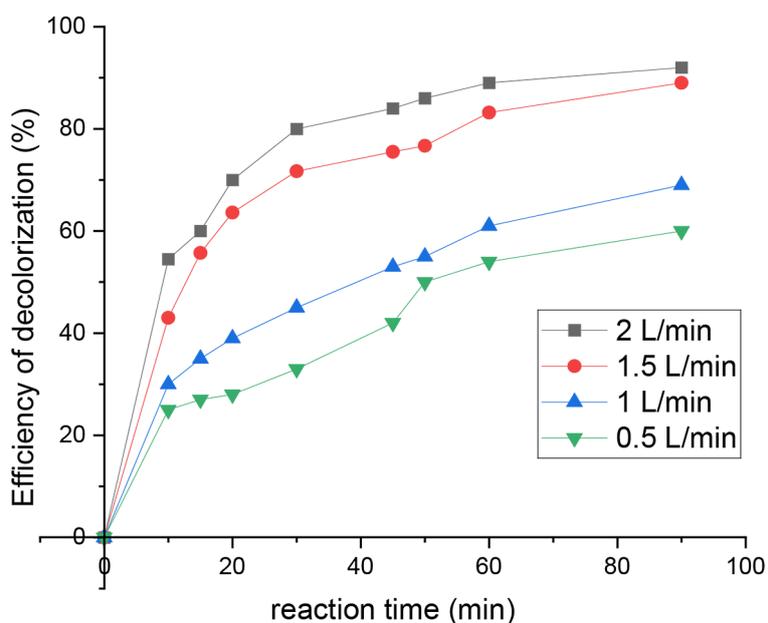

Figure 5. Effect of gas flow rate on dye decolorization of Rhodamine B. 10 mg/L, pH = 1.5, 90 min

the oxidized pollutant. In addition, a larger net surface area was available for the mass transfer of ozone to the aqueous phase due to an improved volumetric mass transfer coefficient of ozone with increasing gas flow rate (Wang et al., 2009)

Effect of the addition of H_2O_2 on Rhodamine B dye

Hydrogen peroxide is effective in improving the removal of color from dyes. It was used in different quantities to improve the efficiency of removing Rhodamine B color from wastewater,

where (0.5,1, 1.5) ml was applied. We noticed that with 0.5 ml of H_2O_2, the removal was 93 percent. When 1.5 ml of H_2O_2 was used, the removal reached 96 percent, which means that when the percentage of hydrogen peroxide increased, the removal rate increased. In contrast, the removal was 82 percent before using peroxide when Ozone was used with the packing only. As observed in Figure 6 higher removal efficiencies were achieved at the higher H_2O_2 concentration.

The degradation ratios of rhodamine B were 93%, 95%, and 96%, with initial H2O$_2$ amounts

of 0.5 ml, 1 ml, and 1.5 ml respectively. The improvement of H_2O_2 in the degradation of rhodamine B can primarily be attributed to the generation of additional •OH radicals resulting from the dissociation of H_2O_2. A greater production of •OH radicals was observed, resulting in higher degradation ratios with increased amounts of H2O2, The result is very similar to those studies on the decolorization of Rhodamine B by also using ozone and H_2O_2 (Wang et al., 2009).

Effect of packing and ozone on Rhodamine B decolorization

Packing was used to improve the decolorization of Rhodamine B dye in the natural pH to obtain a better color removal rate. The dye concentration was 10 mg/L and the packing height was 5 cm, As shown in Figure 7 the decolorization rate increased from 77% to 82% when using packing. The limited solubility of ozone in water is why one uses packaging. Low mass transfer of ozone from the gas to the liquid phase and the great ozone demand for pollutants breakdown during ozonation result from this low solubility. To boost ozone utilization and raise the removal efficiency of Rhodamine B degradation, packing material can be added to the bubble column reactor to extend the residence duration. This leads to the accelerated breakdown of ozone gas in an aqueous solution, These results are in line with Le Sang and his group's studies (Sang et al., 2020).

Removing Congo Red dye in various conditions

Effect of initial pH on Congo Red dye

Ozonation therapy efficacy is sensitive to a number of variables, one of the most important of which is pH. Figure 8 displays the decline in Congo red dye removal efficiency as a function of time. As shown in Figure 8 the maximum removal efficiencies were achieved in the alkali conditions with a pH value of (10) We note that the removal rate was 86%, while this percentage decreases as we move towards the acidic pH. which means the decolorization rate increases with increasing pH value. Reactive •OH radicals help explain pH's favorable effect on color elimination. While the molecular ozone stays the primary oxidant at low pH levels, the hydroxyl radicals are produced from ozone via breakdown at high pH values. (Soares et al., 2006). The initial stage of decolorization takes place via the reaction of ozone with the azo chromophoric group –N=N– or with the double bond of the –C=C– that links aromatic rings. The decolorization process utilizing ozone typically occurs swiftly, attributed to the rapid breakdown of the conjugated chains within the dye molecules that are responsible for the coloration(Tizaoui and Grima, 2011).
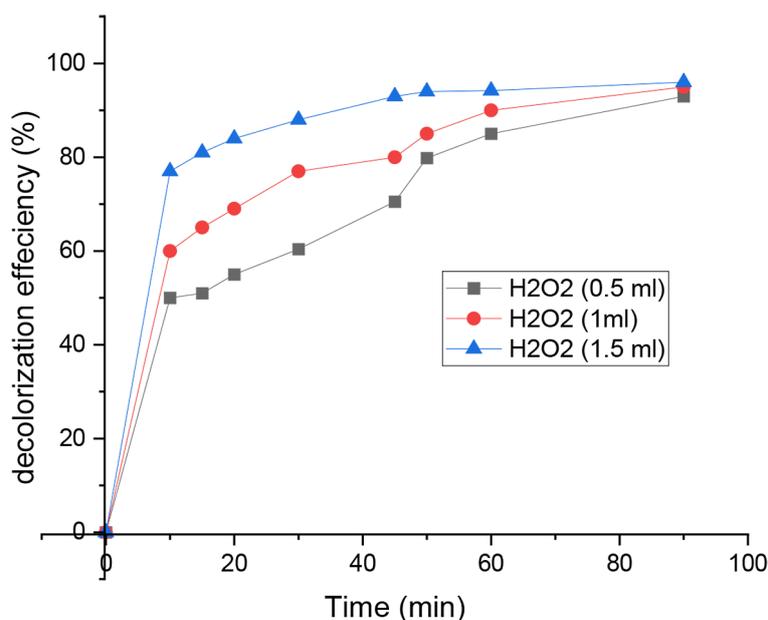

Figure 6. Effect H_2O_2 on decolorization of Rhodamine B. pH = natural, the concentration of Rhodamine B = 10 mg/L, gas flow rate = 2 L/min, time = 90 min
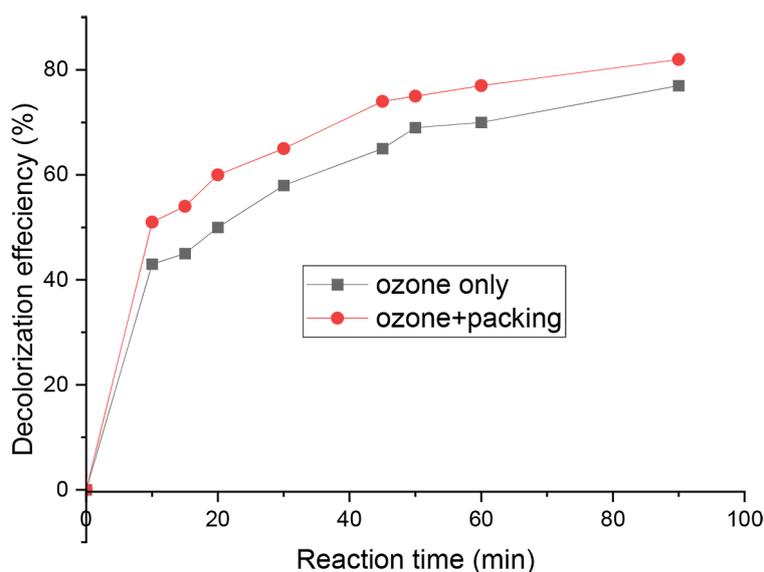
Figure 7. Effect of packing on decolorization of Rhodamine B dye. pH = natural, gas flow rate = 2 L/min, dye concentration = 10 mg/L, time = 90 min
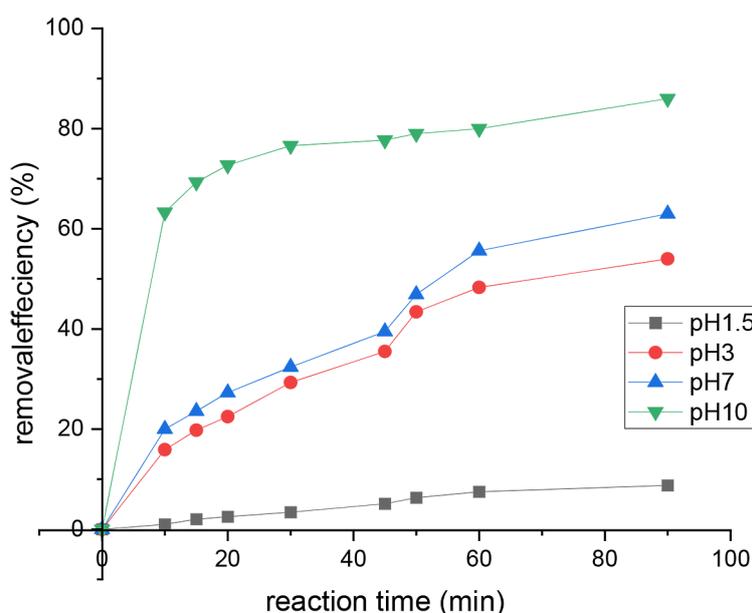

Figure 8. Effect of pH solution on decolorization of Congo Red dye. gas flow rate = 2 L/min, 25 mg/L, 90 min

Effect of inlet concentration of Congo Red dye

To investigate the viability of the ozonation process for varied dye concentrations, experiments were performed at pH 1.5, room temperature, and an ozone gas flow rate of 2 L/min while altering the initial dye concentration (25, 50, and 75 mg/L). Figure 9 depicts the effect of Congo Red starting concentration on decolorization efficiency. Figure 7 shows that raising the dye concentration reduces the rate of decolorization and causes total decolorization to take longer. The

cause could be that, under the given conditions, the ratio of ozone molecules to dye molecules in the solution falls as the dye concentration increases. This conclusion is consistent with research that studied the removal of Reactive Blue 19 (RB19) for the same media conditions (Tehrani-Bagha and Amini, 2010). In advanced oxidation processes, an increase in dye concentration can lead to the formation of various intermediates during the degradation of the parent dye, which may interfere with the oxidation process. The suppression would be more significant when
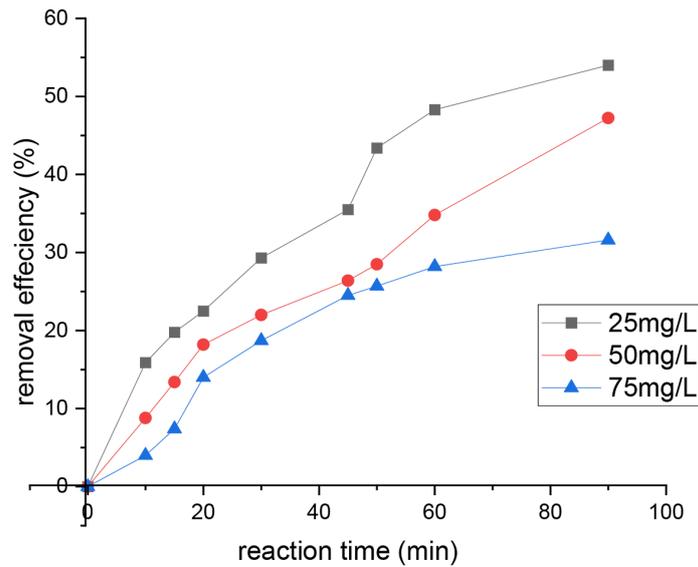
Figure 9. Effect of initial concentration of Congo Red on dye decolorization by ozonation process. Gas flow rate 2 L/min, pH = 1.5, 90 min

there is a higher concentration of degradation intermediates resulting from an increased dye concentration (Song et al., 2008).

Effect of operating time for Congo Red dye removal

Venkatesh and colleagues' 2014 work demonstrated that increasing the contact duration between Congo red dye and ozone resulted in a considerable boost in decolorization efficiency. The color removal rate was first fast, but eventually stabilized over time. However, increasing the contact duration to 90 minutes resulted in an overall

color removal percentage of 86%. This suggests that increasing the contact duration considerably improves the efficiency of the ozone-based oxidation process (Venkatesh et al., 2014) (Figure 10).

Effect of gas flow rate on Congo Red

This study looked at how the flow rate of ozone gas affected the elimination of colors. Experiments were conducted with varying gas flow rates (0.5–2 l/min), dye concentrations (25 mg/l), reaction times (90 min), and pH values (1.5), as illustrated in Figure 11. For this experiment, we kept these values constant, and you
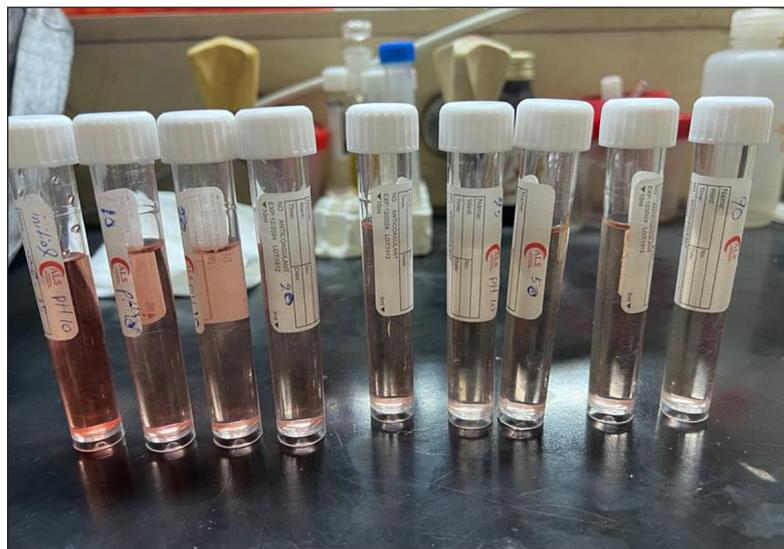

Figure 10. Samples of Congo Red concentration with time. pH = 1.5 ,Dye concentration = 25 mg/L, Gas flow rate = 2 L/min, Reaction time = 90 min

can see the results in the figure below. Figure 11 shows that when the gas flow rate dropped, the dye degradation rate also dropped. The Congo Red elimination rate was 2.68% when the gas flow rate was 0.5 l/min.

However, raising the gas flow rate to 1.5 l/min resulted in a removal rate of (45%), and reaching 2 l/min resulted in a removal rate of (54%). It is the highest dye removal rate achieved with pH 10 and a gas flow rate of 2 L/min at 25 ppm. As the gas flow rate in the zone increases, the size of the generated bubbles also increases. This process results in a reduction of the total surface area between the bubble and the liquid. At lower gas flow rates, the bubbles were observed to be smaller in size compared to those at higher rates. Consequently, at a specified dose of ozone, increased ozone gas flow rates enhance the elimination of organics this can seen similarly to this study (Chung and Kim, 2011).

Effect of H_2O_2 adding on the Congo Red dye

The hydrogen peroxide compound is one of the strong oxidizing agents. The interaction between hydrogen peroxide and Congo red constitutes an oxidation-reduction reaction. Hydrogen peroxide undergoes reduction to form water, while Congo red is oxidized to yield ketones and aliphatic carboxylic acid. The increases were 84%, 89% and 94%. when

applied (0.5, 1, 1.5) ml of H_2O_2. And before using H_2O_2 the decolorization efficiency was 63% in natural conditions. Based on Figure 12 it is seen that the greater the amounts of H_2O_2 added, the Congo red degradation will increase as more OH is formed. We have seen that the removal efficiency increases when the amount of hydrogen peroxide increases. This is due to providing more hydroxyl radicals to oxidize the Congo Red dye molecules as we have seen in this study (Wulansarie, 2023).

Effect of packing and ozone on Congo Red decolorization

Figure 13 illustrates the relationship between removal efficiency and treatment time using ozone and a continuous packing reactor. It was also noted that the efficiency of the ozonation process, both with and without packing, substantially impacted the decolorization of Congo red in a continuously packed reactor. For example, the Congo Red removal increases by 53% to 63% when packing material is added for the same reaction time of 90 minutes and 25 mg/L of Congo Red concentration. Because a high mass transfer rate is caused by an increase in the contact area between the gas and liquid, which is improved with extra packing to the bubble column reactor, the removal efficiency of Congo Red is greatly enhanced (Hashim et al., 2019).
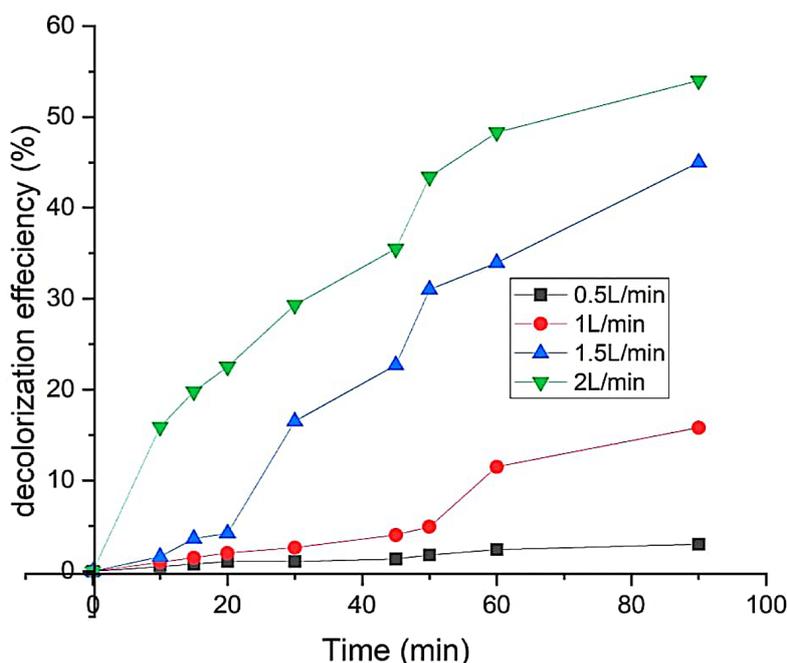

Figure 11. Effect of gas flow rate on dye decolorization of Congo Red. pH = 1.5, 25 mg/L, 90 min
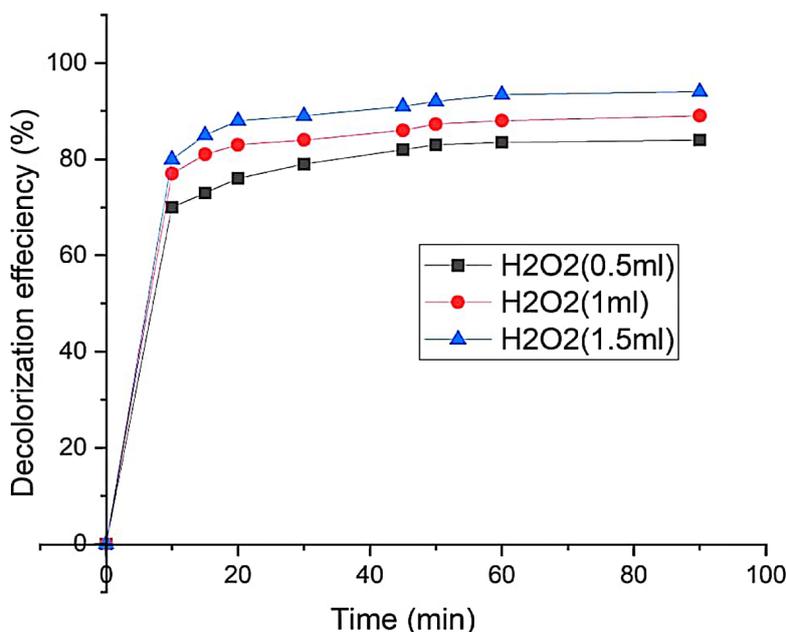
Figure 12. Effect H$_2$O$_2$ on Congo red decolorization. pH = natural, gas flow rate = 2 L/min, dye conc. = 10 mg/L, time = 90 min

Comparison of color removal of Congo Red dye and Rhodamine B dye in different media

The removal of Congo Red and Rhodamine B dyes in different pH environments is primarily due to their chemical structures and interactions with the solution. Congo Red dye is acidic, meaning that it contains acidic functional groups such as sulfonic groups (-SO$_3$H) that lose hydrogen protons (H+) in the basic environment, which increases its negative charge. In the basic environment, the concentration of hydroxyl ions (OH-), which repel negative dye ions, increases. This repulsion increases the solubility of the dye
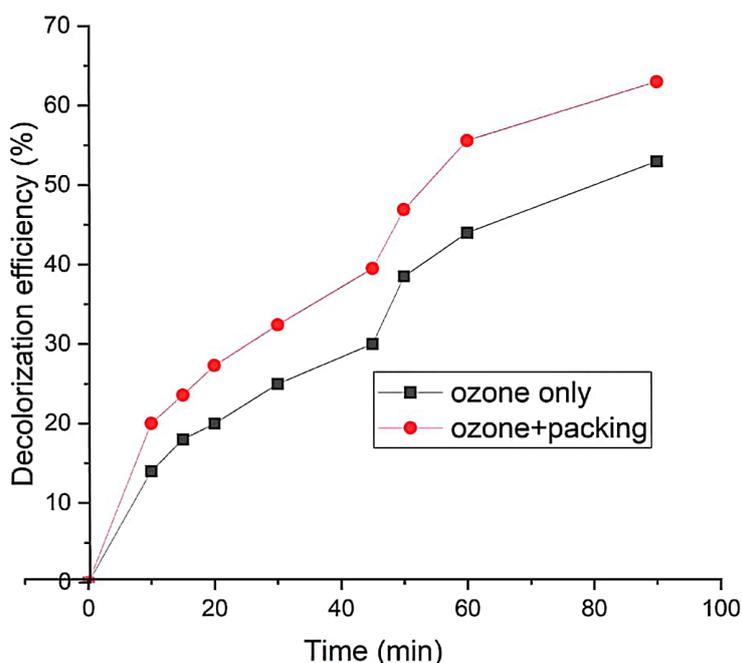

Figure 13. Effect packing on the decolorization of Congo Red.ph = natural, gas flow rate = 2 L/min, concentration of dye 25 mg/L, tim90

Table 2. Comparing the efficiency of the ozone oxidation process in removing a range of industrial dyes

Pollutants	Method	Experimental condition	Efficiency	Year	References
Acid red 27	O_3	Time: 20 min, pH: 3.4 Ozone dose: 0.80 mg/L, conc. Of dye: 42.30 mg/L	Color removal 100%	2009	(Beak, Ijagbemi and Kim, 2009)
Basic yellow	O_3	Time: 30 min, pH: 6.6,ozone dose: 500 ppm, dye conc. 216 ppm	Color removal: 99%	2014	(Zhu *et al.*, 2014)
Acid red 88	O_3	pH: 11, time: 20 min, ozone dose 2 g/L, dye conc.: 500 mg/L	Color removal: 100%	2009	(Avramescu *et al.*, 2009)
Azo dye, reactive red 198	O_3/H_2O_2	Sample volume: 200 ml, H_2O_2 dose: 0.03 mmol, ozone dose: 0.25 g/h, time: 40 min, pH: 10	Color removal: 100%	2016	(Karami *et al.*, 2016)
Textile wastewaters	O_3/H_2O_2	Sample volume: 75 ml, H_2O_2 dose0: 7.14 M H_2O corresponding to 0.5 ml H_2O_2 (30) %, ozone dose: 2.67 mg/L, pH: 5, time: 15 min	Decolorization efficiency: 63.5%	2009	(Zaharia *et al.*, 2009)
Azo dye reactive red 120	O_3	Time: 40 min, conc. Of dye 0.5 g/L, pH: Alkaline	Color removal:98%	2015	(Abidin *et al.*, 2015)
Congo Red	O_3	Dye con.: 15 mg/L, ozone rate: 20 L/min, pH: 11	Color removal: 95%	2021	(Gerulová *et al.*, 2021)
Rhodamine B	O_3	Dye conc.: 100 ppm, ozone dose: 30 ml/min, pH: 2	Color removal: 85.64%	2011	(Cuiping *et al.*, 2011)
Rhodamine B	O_3	Dye conc.: 10 mg/L, pH: 1.5, ozone flow rate: 2 L/min	Color removal: 95%	2024	This study
Congo Red	O_3	Dye conc: 25 mg/L, pH: 10, ozone flow rate: 2 L/min	Color removal: 86%		
Rhodamine B	O_3/H_2O_2	Dye conc.: 10, PH: natural, ozone flow rate: 2 L/min, H_2O_2: 1.5 ml	Color removal: 96%		
Congo red	O_3/H_2O_2	Dye conc.: 25, PH: natural, H_2O_2: 1.5 ml, ozone flow rate: 2 L/min	Color removal: 94%		

and reduces its attachment to surfaces or other materials. Rhodamine B is a positively charged (cationic) organic dye. This means that it attracts negative charges. In acidic media, hydrogen ions (H+) are present in high concentrations. These ions also carry a positive charge. When we put Rhodamine B dye in an acidic medium, the positive ions of the dye repel the positive hydrogen ions in the medium. This repulsion makes the dye less stable and it tends to aggregate or precipitate. The removal rate of Congo Red dye in the acidic medium was approximately 14 percent, while the removal of Rhodamine B dye in the acidic medium was much better, which was 95 percent. Hydrogen peroxide was used to improve the removal of the two dyes, and good results were obtained in removal after its application. The removal process was improved in the natural pH for the two dyes, as the removal rate increased from 63 percent to 94 percent for Congo Red dye when 1.5 ml of hydrogen peroxide was added. The removal rate of rhodamine B dye increased from 82 percent to 96 percent when the same amount of hydrogen peroxide was added, which is 1.5 ml. In summary, removing Congo Red and Rhodamine B dyes depends on their chemical structures and interactions with the solution. Congo Red is removed in a basic medium due to its negatively charged sulfonate ion, while Rhodamine B is removed in an acidic medium due to its hydrophobic character (Table 2).

CONCLUSIONS

It can be concluded that increasing the concentration of the pollutant, affects the efficiency of color removal inversely, which means that when the concentration of the pollutant increases, the removal efficiency decreases. Also, the ozone flow rate plays an important role, as increasing the gas flow rate leads to an increase in the efficiency of color removal. The effect of PG plays an important role in the removal process, as we note from the results that the Congo Red dye was removed in the basic medium, while the Rhodamine B dye was removed in the acidic medium. This is due to the chemical composition of the dye and also depending on its charge and the nature of the material. Excellent removal results were observed after they used hydrogen peroxide and packing for both dyes Congo red and Rhodamine B. Also, The role of hydrogen peroxide and packing was effective in improving the efficiency of color removal of the two dyes.

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