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DECOLORIZATION OF ACID YELLOW 17 BY OZONATION AND PEROXONE (O_3/H_2O_2) PROCESS

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Abstract: In this study, the decolorization of Acid Yellow 17, a mono azo dye with a wide range of applications such as in food, textiles, personal care products, and household cleaning products, was investigated in aqueous solutions using ozonation and peroxone processes. The effects of ozone gas flow rate (150, 200, and 250 L/h), ozone gas concentration (5.5, 11, and 16.5 g/m^3), initial dye concentration (100, 200, and 300 mg/L), and hydrogen peroxide concentration (25, 50 and 62.5 mg/L) on decolorization in the batch bubble reactor were investigated. When the ozone gas flow rate was increased from 150 L/h to 200 L/h in the ozonation process, the removal efficiency increased from 70% to 80.2%. At gas flow rates above 200 L/h, removal was negatively affected. The removal efficiency increased with increasing ozone gas concentration, and at the end of the 45-minute reaction time, a removal efficiency of 98% was achieved at an ozone gas concentration of 16 g/m^3. The increase in initial dye concentrations decreases the removal efficiency due to the increase in the amount of pollutant per unit ozone molecule. In the peroxane process, the effect of hydrogen peroxide on color removal was limited. It was determined that the ozonation process was more effective for the removal of Acid Yellow 17 from aqueous solutions.

Keywords: Acid yellow 17, Ozonation, Decolorization, Peroxane

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1. Introduction

Nowadays, when environmental problems threaten humanity, wastewater treatment is essential in terms of water pollution, protection, and sustainability of water resources. The need for innovative treatment processes is increasing due to the inadequacy of conventional wastewater treatment methods. Research in this area is essential to eliminate pollution and ensure economic, flexible, and sustainable processes. With the increase in the amount and types of pollutants in wastewater, especially with the increase in discharge standards, there is a need for research and development of treatment processes. Among the treatment methods investigated, advanced oxidation processes are interesting in the removal of resistant pollutants (Gągol et al., 2018; Değermenci et al., 2019; Gautam et al., 2019; Iqbal et al., 2023). Advanced oxidation processes are oxidation processes that convert organic compounds in wastewater into water and carbon dioxide through the production of free radicals such as hydroxyl radicals ($^{\bullet}OH$), hydroperoxyl radicals ($HO_2^{\bullet}$), sulfate radicals ($SO^{\bullet}_4^-$) and superoxide anion radicals ($O^{\bullet}_2^-$) by various methods (chemical, electrochemical, radiation-induced and cavitation) (Loures et al., 2013; Boczkaj and Fernandes, 2017; Değermenci, 2021; Çobanoğlu and Değermenci, 2022; Priyadarshini et al., 2022; Iqbal et al., 2023).

Ozone is an oxidizing gas consisting of three oxygen atoms. It is unstable, has a short half-life, and is unstorable. Therefore, it is used where it is produced. It is usually generated from dry or pure oxygen using commercial generators by passing it between electrodes with a high potential difference. In ozonation processes, organic and inorganic compounds are oxidized: i. directly by ozone molecules (dominant mechanism in acidic conditions), ii. indirectly by hydroxyl radicals (dominant mechanism in alkaline conditions) (Loures et al., 2013; Boczkaj and Fernandes, 2017; Chen and Wang, 2021; Iqbal et al., 2023). Ozonation has been widely investigated in drinking water and wastewater treatment processes. As pretreatment, the ozonation process enhances biodegradation and can be used to reduce toxic components for biological treatment. As post-treatment, ozonation can improve water quality by removing pollution before discharge. Ozone can be used as O_3/US, O_3/UV, O_3/Catalyst, O_3/H_2O_2, O_3/Cavitation, or combinations thereof (O_3/H_2O_2/US, O_3/H_2O_2/UV, O_3/Catalyst/UV, etc.) to increase hydroxyl radical concentration (Loures et al., 2013; Gągol et al., 2018; Chen and Wang, 2021). O_3/H_2O_2 (peroxane) is one of the advanced oxidation processes based on the development of the ozonation process. H_2O_2 is a strong oxidant that can decompose to HO_2^- and the addition of hydrogen peroxide to the ozonation process leads to ozone decomposition and the formation of the hydroxyl radical.

There are studies investigating the treatment of dye and textile wastewater with ozone and peroxane processes in the literature (Sun et al., 2020; Pham et al., 2022; Shikha Agrawal et al., 2023).

Wastewater containing dyestuffs has reached enormous amounts today, and it is essential to treat them effectively to protect water resources. Acid Yellow 17 (AY 17) is a mono azo dye with a wide range of applications. AY 17 has many uses in textiles, paper, leather, detergents, soaps, cosmetics, and personal care products and is very soluble in water. It damages humans and other living organisms' nervous, respiratory, and cardiovascular systems. Like most dyestuffs, it has carcinogenic, mutagenic, and toxic effects. It has the potential to form toxic intermediates when appropriate treatment alternatives are not used (Alemu and Kerie, 2022; Kannaujiya et al., 2023; Muhammad et al., 2024). Generally, the visible color of dyes in the aquatic environment is not aesthetically appropriate. They also disrupt gas transfer and inhibit photosynthesis by reducing light transmission (Khan et al., 2018; Teli and Nadathur, 2018).

When the literature is examined, it is observed that various treatment methods have been studied for AY 17 removal. In a study using non-living aerobic granular sludge as biosorbent for AY 17 removal, the effects of pH, biosorbent dosage, initial AY 17 concentration, NaCl concentration, and temperature were investigated. It has been reported that the biosorption process is highly dependent on pH, and the optimum pH value is 2. It was stated that the biosorption capacity increased with increasing dye concentration and decreased with biosorption dosage and salt concentration. It was noted that the biosorption process obeyed the Temkin isotherm and pseudo-second-order kinetics. It was stated that non-living aerobic granular sludge could be a low-cost and alternative biosorbent for removing AY 17 (Gao et al., 2010). In a study where activated carbon/α-Fe_2O_3 nanocomposite was used for the adsorption of AY 17 dye, it was reported that the removal was in good agreement with Langmuir. It was also reported that the adsorption data were quite good with Freundlich and Temkin isotherms at higher concentrations of the dye (40–100 mg/L). The removal of AY 17 was shown to fit the pseudo-second-order kinetic model. AC/α-Fe_2O_3 nanocomposite material was reported to be an excellent magnetic adsorbent for removing acid dyes (Ranjithkumar et al., 2014).

Various advanced oxidation processes for AY 17 removal have been investigated in the literature. In a study examining the removal of AY 17 by the Fenton process; pH, temperature, Fe^{2+} concentration, H_2O_2 concentration, dyestuff concentration, and scavenger ion effects were investigated. It was reported that increasing scavenger anions decreased the reaction and removal rates. It was reported that 89% removal efficiency was obtained under optimum conditions (pH 3.0, [AY 17] = [Fe^{2+}] = 0.06 mM, and [H_2O_2] = 0.9 mM) in 60 minutes of reaction time (Khan et al., 2018).

It has been observed that advanced oxidation studies for AY 17 removal are limited in the literature. This study investigated the effects of ozone gas flow rate, ozone gas concentration, H_2O_2 concentration, and initial AY 17 concentration for ozonation and peroxane processes in a batch-operated bubble reactor for color removal from synthetic wastewater containing AY 17.

2. Materials and Methods

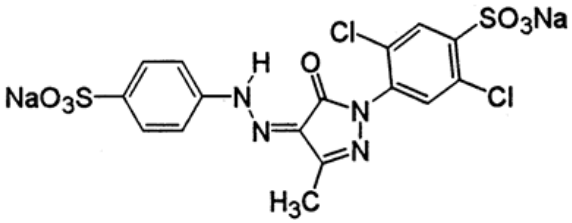
2.1. Chemicals and Analysis

The solid form of AY 17 dye used in the experimental studies was obtained commercially. The chemical structure and chemical properties of AY 17 with CAS registration number 6359-98-4 are given in Table 1. AY 17 was determined colorimetrically using a UV-VIS spectrophotometer (SpectroFlex 6600, WTW) at 403 nm wavelength. In the experimental study, 35% hydrogen peroxide was used. The pH measurements were measured using the WTW (3400i) brand multi-parameter; the experiments were carried out at the natural pH value of 5.7 and were measured and recorded throughout the experimental period. Color removal was calculated using equation 1, as shown below;

$$Removal\% = ((C_0 - C_t)/C_0) * 100 \quad (1)$$

C_0 represents the initial concentration of AY 17, and C_t represents the remaining concentration of AY 17 at time t.

Table 1. Chemical structure and properties of Acid Yellow 17

Commercial Name	Acid Yellow 17
Molecular structure	
Molecular Formula	$C_{16}H_{10}Cl_2N_4Na_2O_7S_2$
Molecular Weight	551.29 g/mol
Max. Wavelength	403 nm

2.2. Experimental Procedure

A glass reactor with a total height of 16.0 cm and an inner diameter of 6.5 cm was used in the experimental studies. Ozone was introduced into the system through a diffuser placed at the bottom of the reactor. The studies were carried out at ambient temperature (20 °C) without temperature control. The volume of the solution AY 17 used in the reactor is 500 mL, and the natural pH of this solution is 5.7. The schematic representation of the experimental system is given in Figure 1. Ozone gas was produced using dry air by electrical discharge. The air from the compressor used as an air source was first passed through the dust and moisture trap and then given to the ozone generator (Anseros, COM AD-08). The solution placed in the glass reactor was subjected to ozonation under the conditions investigated for 45 minutes, and samples were collected over time and analyzed for AY 17.

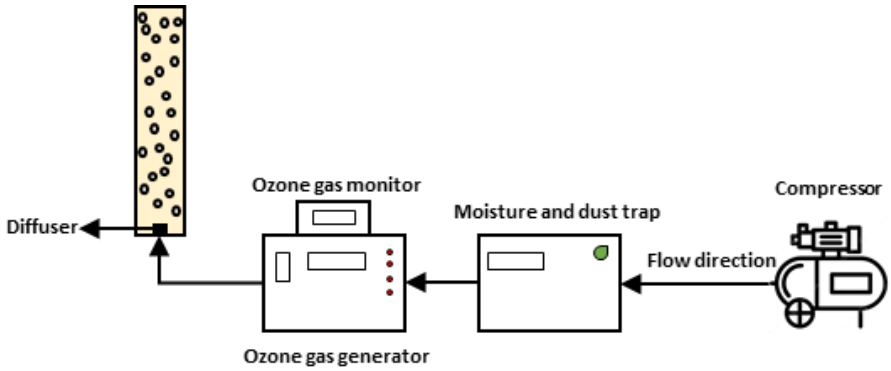


Figure 1. Schematic representation of the experimental system.

3. Results and Discussion

3.1. Effect of Gas Flow Rate on AY 17 Removal in Ozonation Process

To investigate the effect of ozone gas flow rate on AY 17 removal by ozonation process in a batch-operated bubble reactor, gas flow rates of 150, 200, and 250 L/h were tested. A 500 mL solution containing 200 mg/L AY 17 at natural pH (5.70) and ambient temperature (20 °C) was placed in the reactor, and 5.5 g/m^3 ozone gas was introduced through a diffuser at the bottom of the reactor. To examine the effect of gas flow rate, the color removal obtained as a result of ozonation for 45 minutes at different flow rates is given in Figure 2.

As seen in Figure 2, increasing the gas flow rate from 150 L/h to 200 L/h increased the removal efficiency. The increase in removal efficiency with increasing gas flow rate can be explained in two ways; i. the ozone gas mass supplied to the system will increase (W_{ozone}= C_{ozone}*Q_{ozone}) and ii. increasing the gas flow rate will increase the mass transfer rate. Increasing the flow rate will increase turbulence and mass transfer surface area. However, as seen in Figure 2, the removal efficiency has a negative effect when the gas flow rate exceeds 200 L/h. This effect can be attributed to two reasons. Firstly, an excessive gas flow rate will cause bubbles to coalesce, and mass transfer will be negatively affected. Secondly, when the increased gas flow rate increases the gas bubble velocity, it will cause the ozone molecules in the gas bubbles to leave the system without transfer. In other words, ozone molecules will leave the system without reacting with the dyestuff (Duong et al., 2022; M. Gao et al., 2012; Konsowa, 2003; Turhan et al., 2012).

3.2. Effect of Gas Concentration on the Removal of AY 17 in the Ozonation Process

Ozone gas concentrations of 5.5, 11, and 16.5 g/m^3 were used to study the effect of gas ozone concentration on the decolorization of the AY 17 aqueous solution. A 500 mL volume solution containing 200 mg/L AY 17 at natural pH (5.70) and ambient temperature was placed in the reactor, and ozone gas at a flow rate of 200 L/h was introduced through a diffuser at the bottom of the reactor. During the 45 minute reaction, samples taken over time were analyzed, and removal efficiencies were calculated and given in Figure 3.

As seen in Figure 3, increasing gas ozone concentrations increased the removal of AY 17. At the end of 45 minutes of reaction time, AY 17 removal efficiencies for ozone gas concentrations of 5.5, 11, and 16.5 g/m^3 were 80.5%, 89%, and 98%, respectively. Since increasing ozone gas concentrations cause an increase in ozone molecules given to the system per unit time, the removal efficiencies also increase. In addition, increasing ozone concentration in the gas phase will increase the ozone gas transfer as it will increase the concentration gradient, which is the driving force in mass transfer. This will reflect positively on dyestuff removal. However, this increase is not unlimited for process efficiency. Because after a specific value, the ozone molecule given to the system will leave the system without reacting and will have a negative effect on cost-effectiveness (Tehrani-Bagha et al., 2010; Tizaoui and Grima, 2011; Turhan et al., 2012).

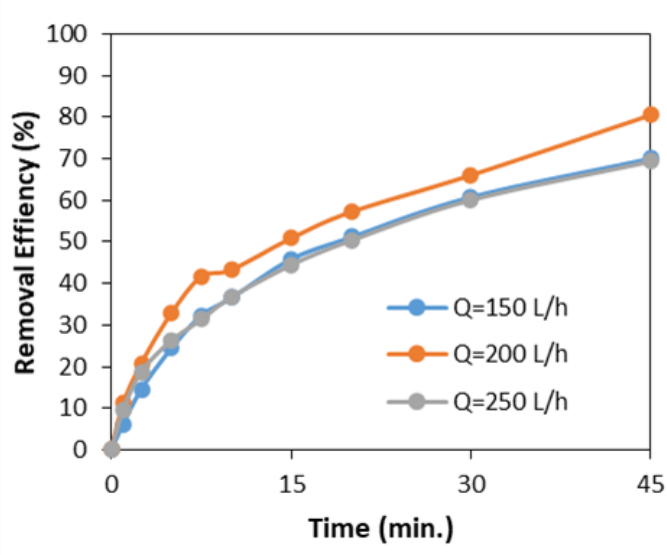


Figure 2. Effect of gas ozone flow rate on AY 17 removal.

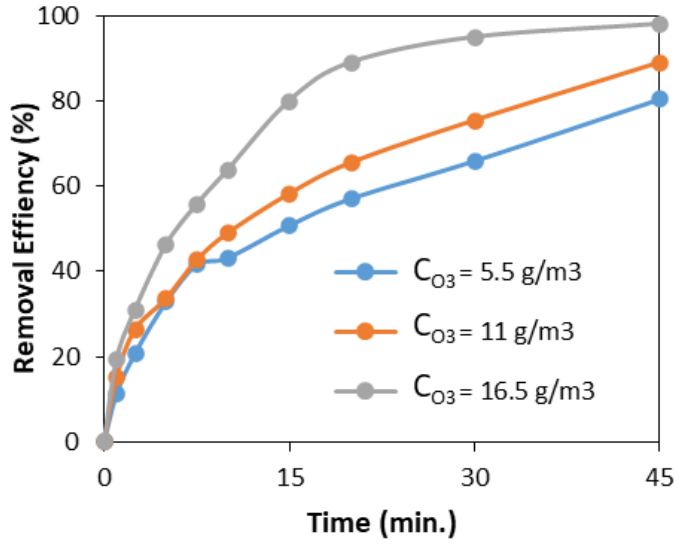


Figure 3. Effect of gas ozone concentration on AY 17 removal.

3.3. Effect of Initial Dye Concentration on AY 17 Removal

To investigate the effect of initial dye concentration on the decolorization of AY 17 aqueous solution, initial dye concentrations of 100, 200, and 300 mg/L were studied. At a gas ozone concentration of 16.5 g/m^3, natural pH (5.70), and ambient temperature (20 ^{o}C), 500 mL of solution was placed in the reactor, and ozone gas at a flow rate of 200 L/h was introduced through a diffuser at the bottom of the reactor. During the 45 minute reaction, samples taken over time were analyzed, and removal efficiencies were calculated and given in Figure 4.

As seen in Figure 4, it is clear that increasing dye concentrations decrease the removal efficiency and increase the decolorization time. Increasing dye concentration increases the dye molecule per unit ozone molecule. It increases the intermediate product concentration, which leads to a decrease in removal efficiency and an increase in decolorization time (Duong et al., 2022; Zhang et al., 2015). Tehrani-Bagha et al. reported that increasing initial dye concentrations decreased the color removal rate and increased the complete color removal time in RB 19 removal by the ozonation process. This is because the ratio of ozone molecules to dye molecules in wastewater will decrease with increasing initial concentration, and various intermediate products formed upon degradation of the leading dye will affect the color removal (Tehrani-Bagha et al., 2010). Similarly, there are studies suggesting that increasing dye concentrations will increase the color removal time (Konsowa, 2003; Turhan and Turgut, 2009).

3.4. Effect of Hydrogen Peroxide Concentration on AY 17 Removal

To investigate the effect of hydrogen peroxide concentration on the decolorization of solutions

containing AY 17, concentrations of 25, 50, and 62.5 mg/L H_2O_2 were studied. At a gas ozone concentration of 16.5 g/m^3, natural pH (5.70), and ambient temperature (20 ^oC), 500 mL of solution was placed in the reactor, and ozone gas at a flow rate of 200 L/h was introduced through a diffuser at the bottom of the reactor. During the 45 minute reaction, samples taken over time were analyzed, and removal efficiencies were calculated and given in Figure 5.

As seen in Figure 5, low hydrogen peroxide addition decreases the removal of AY 17 to a limited extent and does not affect the decolorization at increasing dosages. In the literature, although there is generally an improvement in dyestuff removal with the addition of hydrogen peroxide, the opposite is the case. The decrease or no effect on color removal with the addition of hydrogen peroxide can be explained in two ways. First, it may be due to the scavenging effect of $^{\bullet}OH$ radicals in an alkaline environment. Second, it can be said that the oxidation mechanism depends on the dye structure and is specific to each dye group (Bilińska et al., 2017; Muthukumar et al., 2005). Components in the dyestuff structure can affect the reaction rate with ozone or hydroxyl radicals. In a study investigating the removal of different dyes by various advanced oxidation processes (O_3, H_2O_2/O_3, and H_2O_2/O_3/UV), it is stated that the color removal performances are different for different mono azo dyes and that not only azo groups but also other structural properties of dyes are effective in the removal of dyes (Muthukumar et al., 2005).

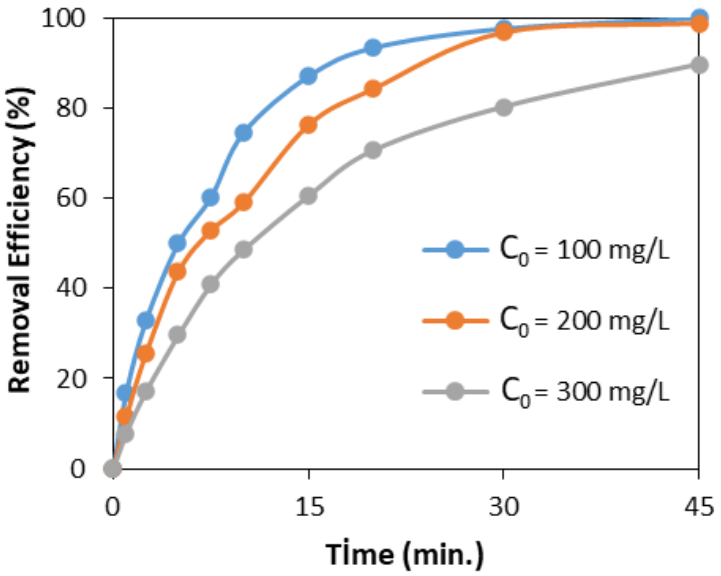


Figure 4. Effect of initial dye concentration on AY 17 removal.

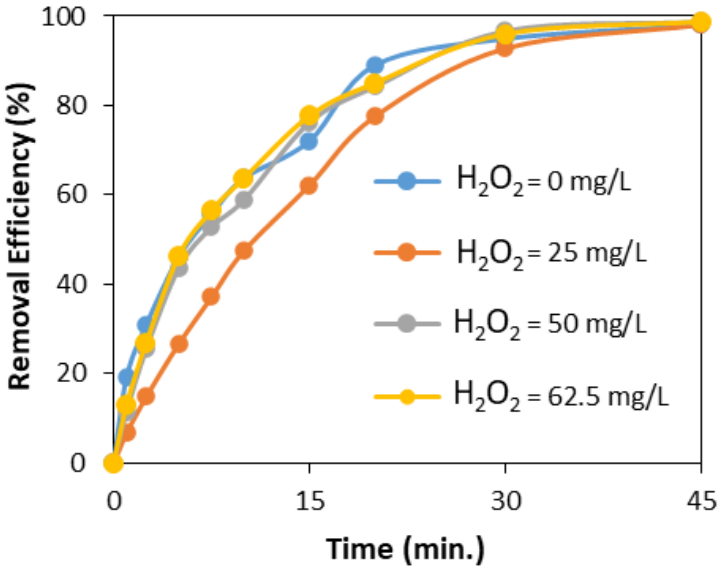


Figure 5. Effect of H_2O_2 concentration on AY 17 removal.

4. Conclusion

In this study, ozonation and peroxane processes were investigated in a batch-operated bubble reactor for decolorization in aqueous solutions containing AY 17 dye, and the parameters affecting the processes were investigated. The effects of ozone gas concentration, ozone gas flow rate, initial AY 17 concentration, and hydrogen peroxide concentration on color removal were investigated. Increasing gas flow rate and gas concentrations in the ozonation process increased color removal, but optimum values should be obtained for these parameters in terms of process efficiency and energy cost. The increase in gas flow rate will have a negative effect at a specific value depending on the system geometry and flow conditions. The increase in dye concentration decreased the removal efficiency. In real wastewater studies, the system can be operated effectively by changing gas ozone concentrations for changing dye concentrations. It is seen that the peroxane process is not effective for wastewater containing AY 17, while the ozonation process is quite effective.

Author Contributions

The percentage of the author contributions is presented below. The author reviewed and approved the final version of the manuscript.

	İ.C.
C	100
D	100
S	100
DCP	100
DAI	100
L	100
W	100
CR	100
SR	100
PM	100
FA	100

C=Concept, D= design, S= supervision, DCP= data collection and/or processing, DAI= data analysis and/or interpretation, L= literature search, W= writing, CR= critical review, SR= submission and revision, PM= project management, FA= funding acquisition.

Conflict of Interest

The author declared that there is no conflict of interest.

Ethical Consideration

Ethics committee approval was not required for this study because of there was no study on animals or humans.

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