

original article

# Degradation of reactive red 198 from aqueous solutions by advanced oxidation process: $O_3$ , $O_3/H_2O_2$ , and persulfate

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## ABSTRACT

**Aim:** The aim of this study was to investigate the degradation of an azo dye, reactive red 198 (RR198), by ozone,  $H_2O_2/O_3$ , and persulfate from aqueous solutions.

**Materials and Methods:** The application of several advanced oxidation processes including single ozonation,  $O_3/H_2O_2$ , and persulfate for degradation of RR198 at concentration of 200 mg/L was investigated. The effect of various parameters including pH,  $H_2O_2$ , persulfate doses, and temperature was studied on the degradation of RR198 from aqueous solutions. In addition, the effects of these processes on biodegradability of RR198 were evaluated.

**Results:** The results showed that the degradation rate was increased by increasing  $H_2O_2$  concentration. The optimum  $H_2O_2$  concentration was obtained in the range of 0.03 mol/L at dye concentration of 200 mg/L. Alkaline pH, higher temperature, and persulfate dose (12 mM) were favored in dye and COD removal. Moreover, compared with ozone and peroxone, the persulfate oxidation could achieve a higher color and COD removal at the same reaction time. Persulfate has greater potential to improve the biodegradability of RR198 solution than ozone and ozone/ $H_2O_2$  process. Biochemical oxygen demand/COD ratio of the dye solution treated by persulfate, ozone/ $H_2O_2$ , and ozone at reaction time 40 min was 0.73, 0.63, and 0.59, respectively.

**Conclusion:** These findings show that oxidation by persulfate is a promising alternative for the treatment of RR198 containing solution as a recalcitrant pollutant.

**Key words:** Degradation, dye, hydrogen peroxide, ozone, persulfate

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## INTRODUCTION

Wastewaters containing dyes are one of the most important problems that are produced by many industries including textile, dying, pulp, and paper.<sup>[1]</sup> Dyes have a great concern to the environment due to the toxicity on the aquatic organisms and soil and also disruption in conventional wastewater treatment system performance.<sup>[1-3]</sup> Textile wastewaters, depending on the forms of dyes, are strongly colored and contain high amounts of organic matter.<sup>[4]</sup> Among the organic dyes, azo dyes due to their complex structures are resistant to biodegradation and are often toxic, carcinogenic, and mutagenic.<sup>[2,5,6]</sup> Thus, it is important to treat azo dyes containing wastewaters with appropriate methods before discharging into the environment.<sup>[3,7]</sup> Various treatment methods such as coagulation/flocculation,<sup>[8]</sup> biological treatment,<sup>[9]</sup> electrochemical oxidation,<sup>[10]</sup> and adsorption<sup>[8,11,12]</sup> have been proved to be technically feasible to remove dye from industrial wastewaters. Treatment of textile wastewaters by conventional methods such as biological,<sup>[13]</sup> physical, and chemical processes or a combination of each is inefficient for color removal.<sup>[14]</sup> The mentioned methods let to the production of sludge<sup>[7]</sup> and only changes pollutants phase from wastewater to biomass or sludge<sup>[9]</sup> which subsequently create other environmental problems. Advanced oxidation processes (AOPs) are another effective treatment method for treating persistent compounds from aqueous media.<sup>[15]</sup> High efficacy, low cost, and simplicity are the main characteristics of AOPs, which has caused to be accepted as a new technique for treating the dye-containing wastewater.<sup>[16]</sup> Furthermore, the AOPs can completely mineralize the dye compounds.<sup>[17]</sup> This methodology is based on the generation of highly reactive radicals such as  $\cdot\text{OH}$  that produces *in situ*. These molecules are nonselective oxidants and afterward that cause a sequence of reactions to destroy the organic compounds into inorganic, final products and/or in most of the cases to less harmful substances.<sup>[9,18,19]</sup> Ultrasonic irradiation, ultraviolet (UV)/H<sub>2</sub>O<sub>2</sub>, Fenton oxidation, O<sub>3</sub>/H<sub>2</sub>O<sub>2</sub>, and ozonation can be also mentioned as the hydroxyl radical-based AOPs (HR-AOPs).<sup>[20]</sup> Ozone is a strong oxidant ( $E_0 = 2.07 \text{ V}$ ) that has been used as a principal component in most of the AOPs.<sup>[9,21]</sup> In recent years, persulfate ( $\text{S}_2\text{O}_8^{2-}$ ) has been considered as the newest oxidant in AOPs in degradation of persistent organic compounds.<sup>[20,22]</sup> Activation of persulfate anions with heat, UV light, or metal ions such as  $\text{Fe}^{2+}$  can generate  $\text{SO}_4^{\cdot-}$  which is a powerful oxidizing agent ( $E_0 = 2.6 \text{ V}$ ).<sup>[23,24]</sup> Some of the AOPs such as Fenton's reagent and UV/H<sub>2</sub>O<sub>2</sub> are H<sub>2</sub>O<sub>2</sub> based that has nonselective reactivity for the most recalcitrant organic compound and high oxidation potential ( $E_0 = 1.78 \text{ V}$ ).<sup>[22]</sup> Activation of H<sub>2</sub>O<sub>2</sub> can be occurred using iron or UV to produce HRs ( $\cdot\text{OH}$ ).<sup>[25]</sup> Furthermore, sonication of hydrogen peroxide can form HRs in aqueous solutions.<sup>[26]</sup> Various researchers showed the feasibility of the ultrasonic-assisted hydrogen peroxide oxidation process for decolonization of malachite green<sup>[27]</sup> and rhodamine B from aqueous solutions.<sup>[26]</sup> Several studies on the application of AOP in degradation of RR 198 have been reported. For

example, Moussavi and Mahmoudi evaluate degradation and biodegradability improvement of the reactive red 198 (RR198) azo dye using catalytic ozonation with MgO nanocrystals<sup>[16]</sup> or Tabrizi *et al.* studied the ozonation of textile RR198 dye in a CSTR.<sup>[28]</sup> Nonetheless, to the best of our knowledge, there was no literature on the application of O<sub>3</sub>/H<sub>2</sub>O<sub>2</sub> and persulfate for the degradation of RR 198 dye-containing wastewaters. Therefore, the application of several AOPs including single ozonation, O<sub>3</sub>/H<sub>2</sub>O<sub>2</sub>, and persulfate for degradation of RR198 was investigated in the present work. The effect of various parameters including pH, H<sub>2</sub>O<sub>2</sub>, persulfate doses, and temperature was studied on the degradation of RR198 from aqueous solutions. In addition, the effects of these processes on biodegradability of RR198 were evaluated.

## MATERIALS AND METHODS

### Chemicals

RR198 was obtained from Alvan Sabet Co. (Iran). The chemical structure and characteristics of the dye are presented in Figure 1 and Table 1, respectively. NaOH and H<sub>2</sub>SO<sub>4</sub> (analytical grade), hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>, 30%), potassium iodide (KI, 99.0%), manganese dioxide (MnO<sub>2</sub>), sodium thiosulfate (Na<sub>2</sub>S<sub>2</sub>O<sub>3</sub>, 99.0%), sodium persulfate (Na<sub>2</sub>S<sub>2</sub>O<sub>8</sub>, 99.0%), HgSO<sub>4</sub>, and AgSO<sub>4</sub> were purchased from Merck Co (Germany). The stock dye solution (1000 mg/L) was prepared by dissolving 1 g of RR198 in 1 L of distilled water. The desired dye concentration was provided with dilution of stock solution by distilled water.

### Analytical methods

The concentration of RR 198 was determined by UV/visible spectrophotometer (DR 5000 Hach) at the maximum absorbance wavelength, 518 nm. The measurement of chemical oxygen demand (COD) was carried out with

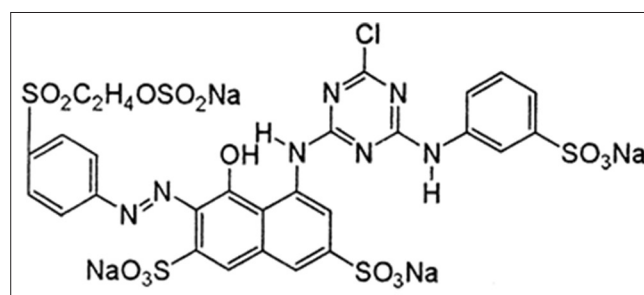


Figure 1: The chemical structure of reactive red 198

Table 1: General characteristics of reactive red 198

Characteristic	RR198
Type	Reactive dye
Commercial name	Remazol red 133
Chemical formula	C <sub>27</sub> H <sub>18</sub> ClN <sub>7</sub> Na <sub>4</sub> O <sub>16</sub> S <sub>5</sub>
Molecular weight (g/mol)	984.21
$\lambda_{\text{max}}$ (nm)	518
Appearance	Red powder

RR198: Reactive red 198

a standard potassium dichromate oxidation method.<sup>[29]</sup> Because the hydrogen peroxide interferes with the accuracy of COD test, the excess of hydrogen peroxide was removed by adding  $MnO_2$  into the solution.<sup>[30]</sup> The pH measurement was determined using electrode method (SensION 378, Hach).

## Experiments

### Ozonation experiment

Ozonation process was conducted with 200 mL of dye solution (200 mg/L) into a plexiglass cylindrical container with total volume of 500 mL. The solution pH was first regulated by 0.1 M NaOH and 0.1 M  $H_2SO_4$  to the desired values (6–10). The ozonation was then performed by ozone dose of 0.25 g/h (Ozone generator, OWA 1000 Japan) for 40 min at 25°C. The samples were finally taken during the process in time intervals of 10 min and were analyzed.

### $O_3/H_2O_2$ experiment

The  $O_3/H_2O_2$  process, such as the above experiments, was carried out by 200 mL of dye solution (200 mg/L) at 25°C. First, after the adjustment of the solution pH to the above-mentioned values, various dosages of  $H_2O_2$  (0.01, 0.02, 0.03, and 0.04 mM) were then added to the solution. Afterward, the ozonation was performed with dose of 0.25 g/h for 40 min and the sampling was finally taken during the process in time intervals of 10 min.

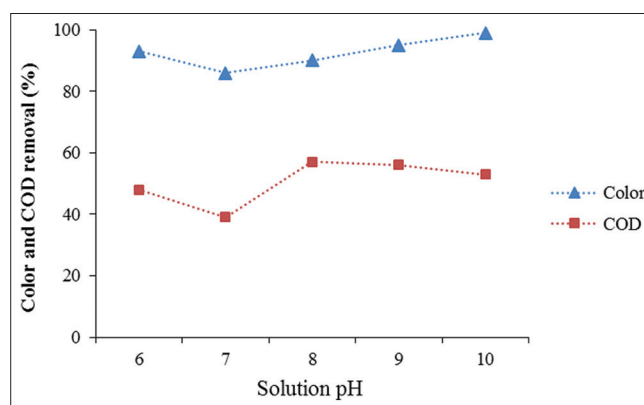
### Persulfate experiment

The decolorization experiments by persulfate radicals were done through addition of sodium persulfate to 200 mL of the dye solution (200 mg/L) into a cylindrical batch reactor with a magnetic stirrer. Various doses of persulfate (0, 4, 6, 8, 12, 14 mmol) were poured into the reactor after solution pH adjustment (3–10). To activate the persulfate radicals of the solutions, the solutions were raised to the desired temperatures (25°C, 60°C, and 70°C) by an experimental hot plate stirrer. The samples were finally taken and analyzed such as the above.

## RESULTS

### Effect of solution pH in ozonation process

Solution pH is an important factor in the degradation of dyes from wastewaters. The effect of initial solution pH on the degradation and COD removal of RR198 was studied at pH values ranging from 6 to 10, under constant reaction time and ozone dosage in single ozonation process. The effect of solution pH on dye and COD removal in single ozonation is shown in Figure 2. As shown in Figure 2, the percentage of color removal in the ozonation process was decreased from 93% to 86%, with increasing solution pH from 6 to 7 and then it was gradually increased to 99% at solution pH of 10.



**Figure 2:** Effect of solution pH on the color and chemical oxygen demand removal in the ozonation process (Reaction time: 40 min and dye concentration: 200 mg/L)

### Effect of $H_2O_2$ concentration

Figure 3 shows the performance of  $O_3/H_2O_2$  process on the color and COD removal at different doses of hydrogen peroxide under pH 8 and contact time 40 min at 25°C. As shown, the color and COD removal were slightly enhanced with increasing  $H_2O_2$  dosage. Maximum color (100%) and COD (55%) removal were achieved in 0.03 mmol  $H_2O_2$  dosage. Hence, in this study, the optimum concentration of  $H_2O_2$  was found to be 0.03 mmol in  $O_3/H_2O_2$  experiments. This removal efficiency can be explained as the production of more oxidizing radicals in the presence of a higher dosage of  $H_2O_2$ .<sup>[31]</sup> As shown in Figure 3, further doses of  $H_2O_2$  led to a decrease in COD removal efficiency.

### Effect of initial dye concentration

The effect of dye concentration (200–1000 mg/L) was investigated on the color and COD removal in  $H_2O_2/O_3$  process at 25°C. Other parameters including pH 8, contact time 40 min, and  $H_2O_2$  concentration 0.03 mmol were constant during the process. As presented in Figure 4, the color and COD removal efficiency are inversely proportional to the dye concentration used in the experiment. The removal efficiency of COD and color showed a similar reduction trend. By increasing RR198 concentration from 200 mg/L to 1000 mg/L, COD and color removal efficiency were decreased from 69% to 43% and from 100% to 88%, respectively. Figure 4 also shows that the value of COD removal is lower than color removal. Thus, longer time and higher ozonation dose would be required for significant COD removal.

### Effect of initial pH on persulfate oxidation

The effect of solution pH on the color and COD removal using persulfate oxidation at 25°C is shown in Figure 5. As shown, the color removal was increased with increasing the initial solution pH. The color removal percentage at pH 3 was 90% and reaching a maximum removal of 100% when pH was increased to 10. Further, the maximum COD removal was observed at pH 9. Therefore, pH 9 was considered as the optimum condition for persulfate oxidation.

### Effect of temperature on persulfate oxidation

Figure 6 illustrates the effect of temperature (25°C, 60°C, and 70°C) on the color and COD removal of RR198 by persulfate process. The experiments in this phase were carried out in reaction time 40 min, RR198 concentration 200 mg/L, the optimum persulfate dosage 12 mM, and pH of 9. As shown in Figure 6, the color removal by persulfate was significantly increased as the temperature was increased. Approximately 98% of the color removal was achieved by persulfate oxidation in 40 min at 70°C. The effect of temperature on the COD removal through persulfate oxidation such as color removal showed a similar trend but in slower rate. Lower COD removal than that of color removal may be due to short reaction time. The maximum COD removal of 64% was achieved in 40 min at 70°C.

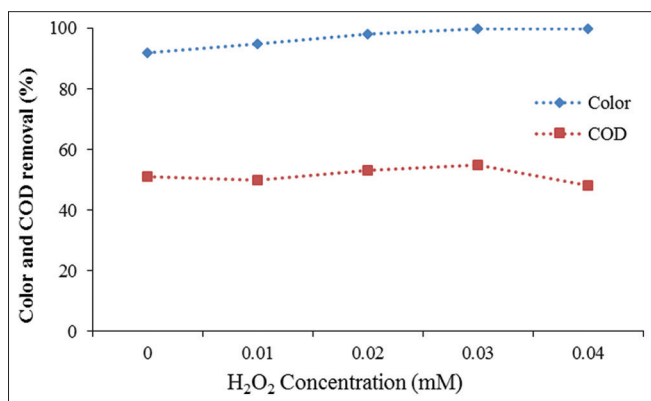
### Effect of persulfate dosage

The influence of persulfate dose on the color and COD removal at the temperature of 70°C is shown in Figure 7. As seen, by increasing the persulfate dosage from 4 mmol to 12 mmol, the color and COD removal were increased from 55%–97% to 30%–67%, respectively, within 40 min of reaction

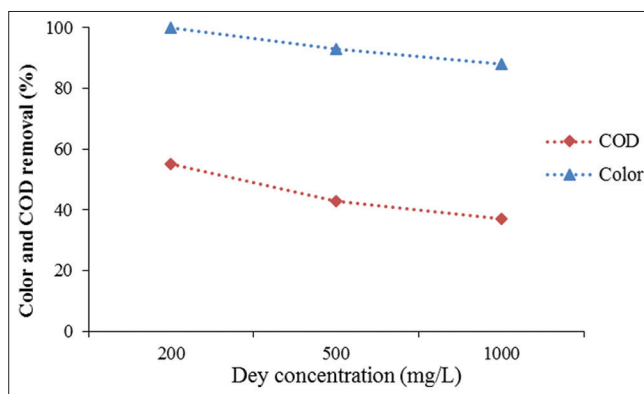
time. However, further increasing in persulfate dosage from 12 mmol to 14 mmol resulted in negligible improvement in the color and COD removal.

### Biodegradability analysis for different advanced oxidation processes

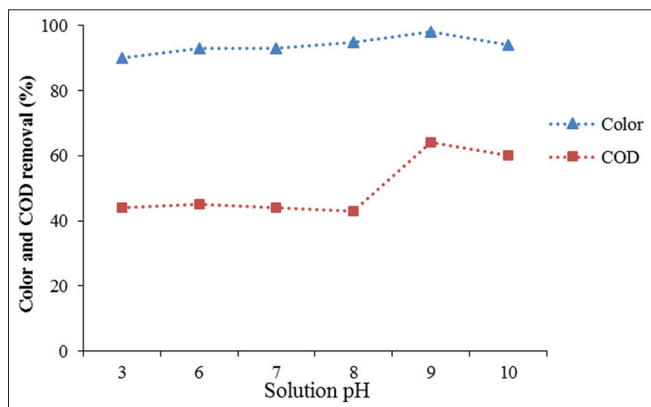
Wastewaters that have a biochemical oxygen demand (BOD<sub>5</sub>)/COD ratio between 0.4 and 0.8 can be significantly considered as biodegradable.<sup>[32]</sup> To enhance the wastewater biodegradability, first, different AOPs (ozone, ozone/H<sub>2</sub>O<sub>2</sub>, and persulfate) under the optimum condition were conducted on the dye solution. Then, BOD<sub>5</sub>/COD ratio was determined for each dye solution. Figure 8 shows the effect of these processes on the biodegradability of RR198 containing solutions. As seen, the BOD<sub>5</sub>/COD ratio for the raw dye solution (before using AOPs) was 0.18. The lower BOD<sub>5</sub>/COD ratio of dye solution shows that the solution is not readily biodegradable. As shown in Figure 8, the BOD<sub>5</sub>/COD ratio (biodegradability rate) for the treated solutions was increased with the reaction time. As illustrated in the figure, persulfate has greater potential to improve the biodegradability of RR198 solution



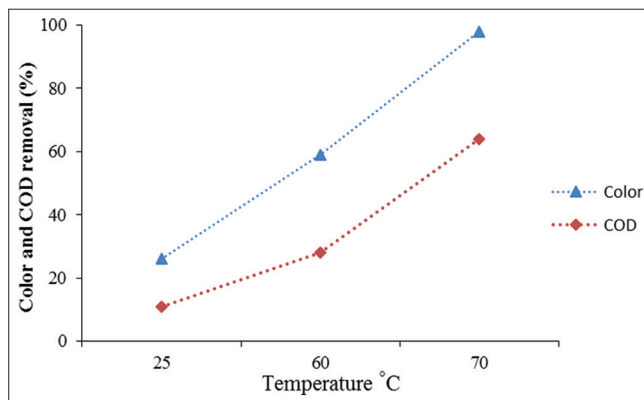
**Figure 3:** Effect of H<sub>2</sub>O<sub>2</sub> concentration on the color and chemical oxygen demand removal of reactive red 198 in the O<sub>3</sub>/H<sub>2</sub>O<sub>2</sub> process (Solution pH: 9, reaction time: 40 min and dye concentration: 200 mg/L)



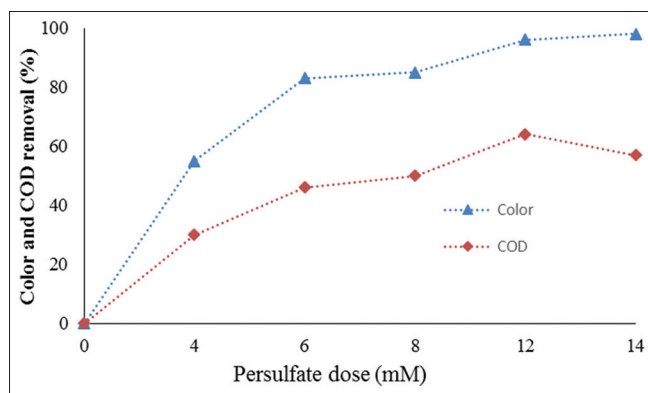
**Figure 4:** Effect of initial dye concentration on the color and chemical oxygen demand removal in the O<sub>3</sub>/H<sub>2</sub>O<sub>2</sub> process (Solution pH: 9, reaction time: 40 min and H<sub>2</sub>O<sub>2</sub> dose: 0.03 mM)



**Figure 5:** Effect of solution pH on the color and chemical oxygen demand removal by persulfate process (Reaction time: 40 min, persulfate dose: 12 mM and dye concentration: 200 mg/L)



**Figure 6:** Effect of temperature on the color and chemical oxygen demand removal from the solution (Solution pH: 9, reaction time: 30 min, dye concentration: 200 mg/L and persulfate dose: 12 mM)



**Figure 7:** Effect of initial concentration of persulfate on the color and chemical oxygen demand removal from the solution (Solution pH: 9, reaction time: 40 min, dye concentration: 200 mg/L and temperature: 70°C)

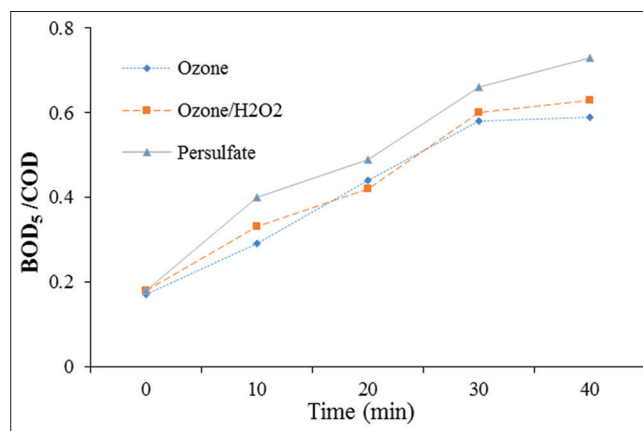
than ozone and ozone/H<sub>2</sub>O<sub>2</sub> process. BOD<sub>5</sub>/COD ratio of the dye solution treated by persulfate, ozone/H<sub>2</sub>O<sub>2</sub>, and ozone at reaction time 40 min was 0.73, 0.63, and 0.59, respectively.

## DISCUSSION

The color degradation at acidic pH is related to the direct oxidation of ozone with the dye molecules, which is predominant reaction in acidic condition.<sup>[16]</sup> Higher COD removal at basic (alkaline) solution pH can be associated with the formation of HRs ( $\cdot\text{OH}$ ). In alkaline conditions, production of the reactive radicals is increased due to accelerating the rate of ozone decomposition.<sup>[33]</sup> The characteristics of the solution (e.g., pH, promoters, and scavengers in the reacting medium) determine the ozonation reaction pathway.<sup>[16,34]</sup> The degradation of aromatic compounds may be occurred by the direct oxidation and indirect radical chain type reaction.<sup>[34,35]</sup> Turhan and Turgut showed that decolorization of direct dye in textile wastewater were noticeable, due to the formation of HRs, at basic solution pH of 12.<sup>[35]</sup> At pH values above 10, the decomposition of ozone leads to generation of high concentration of HRs in solution. The maximum COD removal efficiency at pH 8 can be related to the formation of radical species other than hydroxyls.<sup>[36]</sup> Decrease in color removal in the ozonation process with increasing solution pH from 6 to 7 can be attributed to the radical scavengers including CO<sub>3</sub> and SO<sub>4</sub> that may exist in this condition. These compounds can lead to reduce  $\cdot\text{OH}$  concentration which results in decrease in COD removal at pH 10.<sup>[37,38]</sup>

Decrease in COD removal efficiency with increasing of H<sub>2</sub>O<sub>2</sub> dosage from 0.03 mm may be due to the consumption of HR via scavenging effects of H<sub>2</sub>O<sub>2</sub> and recombination of  $\cdot\text{OH}$  radicals. When the available H<sub>2</sub>O<sub>2</sub> exceeds a critical concentration,  $\cdot\text{OH}$  could be depleted through the scavenging of  $\cdot\text{OH}$  by excessive H<sub>2</sub>O<sub>2</sub>.<sup>[31]</sup>

Results of the previous studies confirm our result.<sup>[23]</sup> Generation of other radicals (e.g., hydroperoxyl) that less



**Figure 8:** Effect of three different AOPs on the biodegradability of reactive red 198 in the solution (Dye concentration: 200 mg/L)

reactive than hydroxyl may be the other reason for this observation.<sup>[18]</sup> As reported by others,<sup>[39]</sup> the presence of H<sub>2</sub>O<sub>2</sub> into the effluent because of overuse of hydrogen peroxide causes an increase in COD of the effluent. It is essential to optimize the applied H<sub>2</sub>O<sub>2</sub> concentration to maximize the treatment performance of the AOPs since the presence of excess H<sub>2</sub>O<sub>2</sub> can reduce the treatment efficiency of AOPs.<sup>[34]</sup> Furthermore, it has been known that in comparison with ozonation process, COD removal is slightly decreased in peroxone process (O<sub>3</sub>/H<sub>2</sub>O<sub>2</sub>). This can be related to reaction of H<sub>2</sub>O<sub>2</sub> with ozone to produce  $\cdot\text{OH}$  that results in reduction the effective concentration of ozone. The same results were also reported by others.<sup>[40]</sup> The abatement of color and COD removal efficiency with increasing the initial dye concentration can be resulted from the fact that higher dye concentration can increase the number of dye molecules and possibly inorganic anions which may compete for the reaction with  $\cdot\text{OH}$  radicals. Moussavi and Mahmoudi and Siddique *et al.* reported that RR198 and reactive blue 19 (RB19) removal efficiency were decreased by increasing the dye concentration during various AOPs.<sup>[16,41]</sup>

Increase in COD removal at alkaline pH in persulfate experiment can be attributed to the fact that at the alkaline pH, activation of S<sub>2</sub>O<sub>8</sub><sup>2-</sup> is occurred and more SO<sub>4</sub><sup>-</sup> is produced; therefore, the oxidation capacity was increased. Deng *et al.* reported that in alkaline pH, activation of persulfate can occur.<sup>[20]</sup> Superoxide radicals, with small amount of hydroxyl and sulfate radicals, are the main radicals generated in the alkaline activated persulfate system. The base-catalyzed hydrolysis of persulfate to a hydroperoxide anion and sulfate is the main approach of base activated persulfate. Furthermore, the HRs can be produced through the reaction of sulfate radicals with hydroxyl ions.<sup>[42]</sup> In addition, increasing COD removal at the alkaline pH can be related to the generation of HRs via the above-mentioned approach. Zhao *et al.* reported that elevated pH enhances the removal of PAHs in soil. They also showed that superoxide radicals were the main radicals

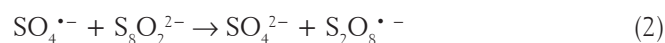
**Table 2: Comparison the color and chemical oxygen demand removal percentages using different methods**

Method	Color	COD removal (%)	Color removal (%)	References
Catalytic ozonation with MgO nanocrystals	RR198	61	100	[16]
Sono-Fenton	Reactive yellow 145	51	95	[39]
UV/H <sub>2</sub> O <sub>2</sub>	Basic violet 16	45	97	[41]
O <sub>3</sub>	Direct blue 71	-	97.4	[38]
O <sub>3</sub>	RR135	66.66	99.9	[44]
Ultrasonic degradation with CCl <sub>4</sub> enhancement	Brilliant red K-2BP	60	100	[37]
O <sub>3</sub>	RR198	57	90	Present study
O <sub>3</sub> /H <sub>2</sub> O <sub>2</sub>	RR198	55	100	Present study
Persulfate oxidation	RR198	64	98	Present study

COD: Chemical oxygen demand, UV: Ultraviolet, RR: Reactive red

generated in the alkaline activated persulfate system.<sup>[42]</sup> In contrast, Xiang *et al.* (2010) showed that the degradation rate of orange G dye by persulfate/Fe<sub>2</sub> process was decreased with increases in solution pH. They also illustrated that the performance of persulfate/Fe<sub>2</sub> was more efficient at the acidic pH than of neutral and alkaline pHs.<sup>[43]</sup> Increase of COD and color removal with increasing temperature confirm that the sulfate free radicals (SO<sub>4</sub><sup>•-</sup>) have been generated by activation of persulfate through heat energy.<sup>[22]</sup>

The higher removal efficiency of color and COD at higher persulfate dosage may be due to the more production of SO<sub>4</sub><sup>•-</sup> in the reaction because persulfate acts as the main source of SO<sub>4</sub><sup>•-</sup> radicals. Similar results have been reported in previous studies.<sup>[20,25,43]</sup> Although the generation of SO<sub>4</sub><sup>•-</sup> radicals is increased by increasing persulfate dosage in the process, at the same time, the reaction of SO<sub>4</sub><sup>•-</sup> radicals with each other or with various scavengers instead of target pollutants reactions is enhanced.<sup>[23]</sup> Therefore, insignificant increase in COD removal percentage can be occurred at higher persulfate dosage. Possible chemical scavenging reactions that compete with the SO<sub>4</sub><sup>•-</sup>/RR198 interaction can be presented by the following reactions.<sup>[23]</sup>



The comparison of the present study with the other methods in the removal of color and COD is summarized in Table 2. As seen, persulfate oxidation is considered as an efficient process, especially in the removal of COD due to its higher efficiency.

Further, destruction of azo bands and aromatic rings of RR198 by the oxidants compounds have been led to enhancement of the dye biodegradability. The same results were reported by others.<sup>[16,45]</sup>

## CONCLUSION

In this study, degradation of RR198 using several AOPs including O<sub>3</sub>, O<sub>3</sub>/H<sub>2</sub>O<sub>2</sub>, and persulfate was studied. The

effects of various parameters such as ozone, hydrogen peroxide, and persulfate doses, initial dye concentration, and solution pH at reaction time of 40 min were studied on the degradation of RR198. The degradation of dye was influenced by the initial solution pH, initial dye concentration, H<sub>2</sub>O<sub>2</sub> concentration, and persulfate dosage. Increasing initial solution pH, H<sub>2</sub>O<sub>2</sub> concentration, and persulfate dosage led to enhance the dye degradation while increasing initial dye concentration led to a decrease the dye degradation. It was observed that, in comparison with other processes, persulfate as the newest oxidant agent caused higher COD removal (67%) of RR198 at reaction time of 40 min. The greatest biodegradability of the dye-containing wastewater was also achieved by oxidation with persulfate.

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## Conflicts of interest

There are no conflicts of interest.

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