

Comparison of the Efficiency of Fenton and Photo-Fenton Processes for the Removal of Reactive Red 198 Dye from the Aqueous Solution

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ABSTRACT

The textile industry produces a large amount of polluted effluents discharged into the environment. Therefore, this research was carried out to compare the efficiency of removal of Reactive red 198 (RR-198) dye by fenton and photo-fenton processes and determine the optimal conditions for maximum removal.

This study was conducted on a laboratory scale. The effect of influential parameters, including pH (3–9), Fe(II) concentration (10–200mg/L), H₂O₂ concentration (25–150mg/L), dye concentration (50–200mg/L) and reaction time (15–90min.) on dye removal was investigated and the optimal conditions were determined according to the maximum dye removal efficiency.

The results indicated that the dye removal rate increased as the pH and Fe(II) concentration decreased. The optimal conditions for RR-198 removals from the aqueous solution are pH of 3, Fe (II) concentration of 10mg/L, H₂O₂ concentration of 50mg/L, the initial dye concentration of 50mg/L, and the reaction time of 15min.. The maximum dye removal efficiency under optimal conditions was 98.82%.

The results of this study revealed that the photo-fenton process was superior to the removal of dye compared to fenton process.

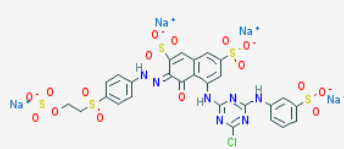
Keywords: Dye removal, Reactive Red 198, Fenton, Photo-Fenton

INTRODUCTION

Dyes are aromatic organic compounds that absorb light at a wavelength of 350–700 nm (visible light region) [1, 2]. According to their chemical structure, dyes can be divided into different categories. One of the highly used dyes in many industries including textiles industries is Azo dyes [3]. Azo dyes contain one or more Azo bond (N = N⁻) and the most important group of synthetic dyes are Azo dyes [4]. Considerable amounts of reactive dyes are detected in wastewater due to their high water solubility and low biodegradability [5]. RR-198 dyes are hydrophilic and also have a low tendency toward uptake by the biomass in biological treatment methods [6]. It has been estimated that 1–15% of dyes during dyeing and finishing processes enter the environment [7, 8]. The presence of chemical dyes pollutes many water resources [9]. The basic characteristics and molecular structure of RR-198 are presented in Table 1.

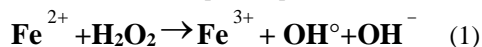
Due to the complexity of the dyes with aromatic molecular structure, the conventional biological and physical-chemical processes are less suitable for their efficient treatment and mineralization [11].

Table 1: The basic characteristics of Reactive Red 198 Dye [10]

Parameter	Characteristic
Chemical name	Reactive Red 198
Molecular formula	C ₂₇ H ₁₈ ClN ₇ Na ₄ O ₁₆ S ₅
C.I. number	18221
Molecular weight	967.5 g/mol
UV absorption	λ _{max} 518 nm
molecular structure	

A review of the literature shows a growing interest in developing advanced oxidation processes (AOPs) for

the treatment of liquid streams containing toxic and xenobiotic compounds, including dyes [10] relying on the production of free radicals OH^\bullet [12]. Recently, photo-fenton and fenton processes have attracted much attention due to high oxidation potential of OH^\bullet to remove the contaminants, especially organic pollutants [13]. In fenton process, ferrous ion and hydrogen peroxide is used as the reductant and oxidant, respectively [14, 15] and peroxide generates hydroxyl radicals (OH^\bullet) at room temperature as described by Walling (1975) in Eq. (1) [16]. The photo-fenton process was conducted using H_2O_2 , Fe^{2+} , and UV radiation [12, 13].



The rate of reaction highly depends on the optimization of the influential parameters, including pH, hydrogen peroxide concentration, Fe (II) concentration, and reaction time [17].

Ghanaian *et al.* showed that the maximum removal rate of Reactive Blue 19 (98.2%) was obtained at the wavelength of 592 nm, the reaction time of 3 hours, pH = 4, and potassium persulfate concentration of 5 mM [18]. Another study showed that the maximum removal of RR-198 was 99%, using electrochemical coagulation process in a voltage of 40 V, distance of 1 cm between the electrodes, and reaction time of 30 minutes [19]. Dehghani *et al.* showed that the maximum removal efficiency at 32 V and 40min. operation time was 98.1% [19]. The maximum removal of acid yellow-powder 36 dyes from aqueous solutions was 97.9%, using fenton's reagent [7].

In recent years, RR-198 is largely used in textile and dye industries. In addition, there is a concern regarding the contamination of water resources and its side effect on people's health, as well. Since the Reactive dyes are persistent in the environment and conventional wastewater treatment was unable to remove the dye efficiently, the necessity of a suitable treatment to reduce the dyes is highly required. Therefore, the present study aimed to (i) evaluate the possibility of using fenton and photo-fenton processes for removing RR-198 in aqueous solution, (ii) compare the removal efficiency using these processes, and (iii) assess the optimum operating conditions for removing the dye effectively.

MATERIALS AND METHODS

Chemicals and Analytical Method

RR-198 ($\text{C}_{27}\text{H}_{18}\text{ClN}_7\text{Na}_4\text{O}_{15}\text{S}_5$) with a molecular weight of 967.5 g/mol was supplied by Hoechst Company, Germany. The rest of the chemicals were purchased from Merck (Germany). UV lamp (F8T5) with a length of 25 cm, 8 W and 356 nm wavelength,

(Hitachi, Japan) was used as the radiation source for photo-fenton processes.

Dye concentration was determined at a wavelength of 520 nm, using spectrophotometer (DR 5000) according to the standard method (No 2120) [20].

Photocatalytic Experiments

The experiments were carried out in triplicates in the batch mode. The studied parameters were reaction time (15, 30, 45, 60 and 90min.), H_2O_2 concentration (25, 50, 75, and 150mg/L), Fe (II) concentration (10, 100, 150, and 200mg/L), and initial dye concentration (50, 100, 150, and 200mg/L) at different pH levels (3, 5, 7, and 9). The optimal conditions were determined according to the maximum dye removal efficiency. In photo-fenton process, the studied parameters were exactly as above, except for using a UV lamp. The data were analyzed using SPSS for windows (version 16, SPSS Inc., Chicago, IL, USA) by Pearson's correlation coefficient to analyze the relationship between these parameters.

Reactor specifications

The experiment was performed in a 4-litre volume reactor. The test was performed in a closed glass reactor with adjustable mixer. The source of radiation was a UV lamp which was protected by a Quartz tube with a height of 30 cm and inside diameter of 5 cm. The UV radiation source was immersed in the solution for better radiation. The whole system was wrapped in an aluminium foil in order to prevent reflection. The specification of the photochemical reactor is shown in Fig. 1.



Fig. 1: The specification of the photochemical reactor

RESULTS

Effect of pH

The variations of pH on the removal rate of RR-198 by fenton and photo-fenton processes are shown in Fig. 2. The data on the effect of pH showed that as pH increased from 3.0 to 9.0, the rate of dye reduction

decreased (Fig. 2). Based on our data obtained in the present study, pH of 3 is optimal for dye degradation for both of the processes. According to Figure 3, the maximum removal efficiencies for fenton and photo-fenton processes were 34.46% and 36.03%, respectively.

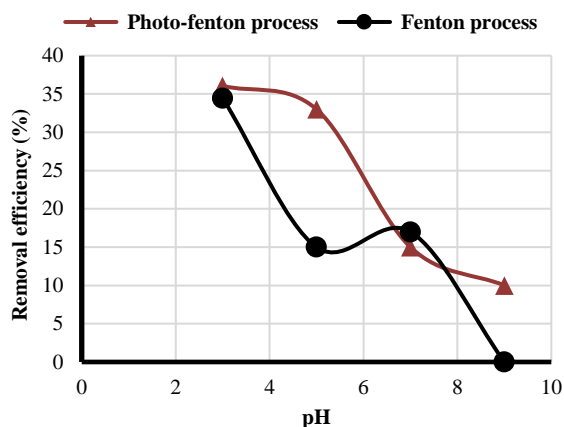


Fig. 2: Effect of pH on the removal of RR-198 by fenton and photo-fenton processes (Fe (II) = 100mg/L, H₂O₂ = 50mg/L, initial dye concentration = 50mg/L, reaction time = 30min.)

Effect of ferrous ion (Fe(II))

According to Fig. 3, photo-fenton process was more efficient in the removal of RR-198 at Fe(II) concentration of 10 mg/L. As Fe(II) concentration increased from 10 to 200 mg/L, the removal efficiency of RR-198 decreased from 81.98 to 43.42% for photo-fenton process, respectively. However, in fenton process, an increase in the reduction rate was observed as Fe(II) concentration was increased from 10 to 100 mg/L. Therefore in fenton process, the dye reduction rate increased with Fe(II) concentration up to a specific level (10–100mg/L) and then began to decrease (100–200mg/L).

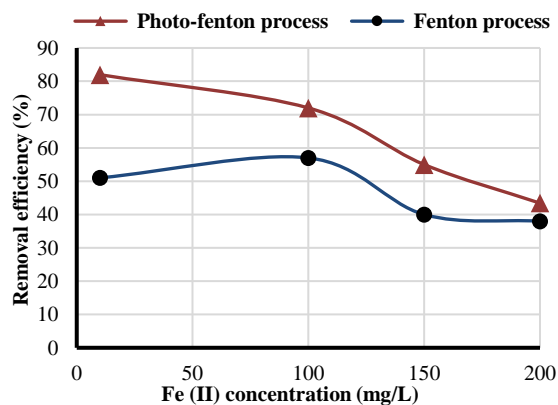


Fig. 3: Effect of ferrous ion (Fe (II)) on the removal of RR-198 by fenton and photo-fenton processes (pH = 3, H₂O₂ =

50mg/L, initial dye concentration = 50 mg/L, reaction time = 30min.)

Effect of H₂O₂ concentration

The effect of H₂O₂ concentration on the removal of RR-198 is shown in Fig. 4. In photo-fenton process, an increase in the concentration of H₂O₂ from 25 to 50 mg/L can lead to a significant increase in the removal efficiency (from 43.22% to 85.22%). Further increase in the peroxide concentration (from 50 to 150 mg/L) had no effect on the removal rate. Based on the data obtained in the fenton process, H₂O₂ concentration of 50 mg/L was optimal and the removal efficiency was 67.5%. Further increase in the concentration of hydrogen peroxide leads to a significant decrease in the removal rate.

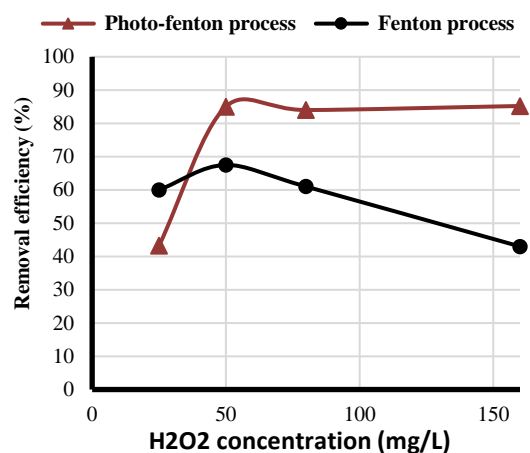


Fig. 4: Effect of H₂O₂ on the removal of RR-198 by fenton and photo-fenton processes (pH = 3, initial dye concentration = 50mg/L, reaction time = 30min, Fe (II) = 10 and 100mg/L for photo-fenton and fenton, respectively)

Effect of RR-198's initial concentration

The initial concentrations of RR-198 at four levels (50, 100, 150 and 200mg/L) had a considerable effect on the reduction rate of the dye in the aqueous phase. The effect of initial dye concentration on its removal efficiency is shown in Fig. 5. As the initial dye concentration increased from 50 to 200mg/L, the removal efficiency was decreased in the photo-fenton process. Therefore, the maximum removal rate in the photo-fenton process (74.98%) was observed at the initial concentration of 50mg/L. However, in the fenton process, an increase in the concentration of the dye from 50 to 100mg/L had a significant increase in its removal rate. In the fenton process, the maximum removal rate (87%) was observed at the RR-198 concentration of 100mg/L. After that, the removal efficiency of the dye decreased when the initial dye concentration was more than 100mg/L.

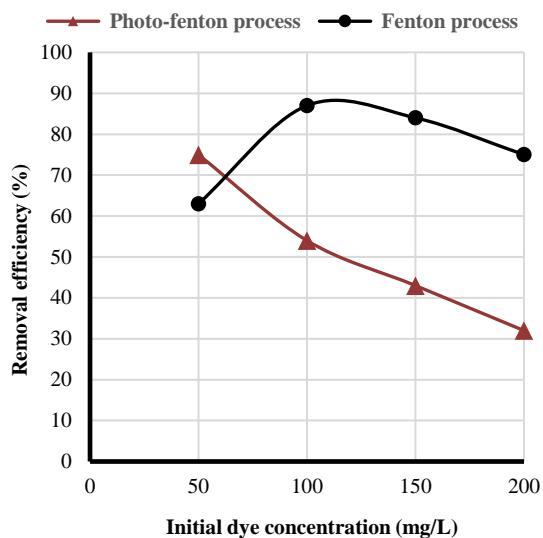


Fig 5: Effect of initial dye concentration on the removal of RR-198 by fenton and photo-fenton processes (pH = 3, H₂O₂ = 50mg/L, reaction time = 30min, Fe (II) = 10 and 100mg/L for photo-fenton and fenton, respectively).

3.5. Effect of reaction time

The data on the effect of reaction time showed that as the reaction time increased, the dye removal efficiency increased in both fenton and photo-fenton processes. According to Fig. 6, the maximum removal efficiency (98.82%) was observed at 90 minutes in the photo-fenton process.

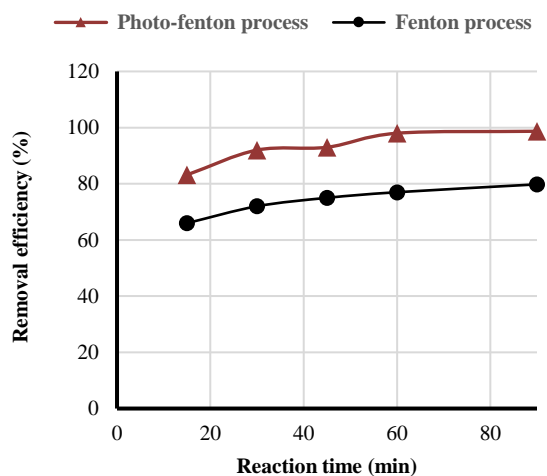


Fig.6: Effect of reaction time of the initial dye concentration on the removal of RR-198 by fenton and photo-fenton processes (pH = 3, H₂O₂ = 50mg/L, initial dye concentration = 50 and 100mg/L for photo-fenton and fenton, respectively, Fe (II) = 10 and 100mg/L for photo-fenton and fenton, respectively).

DISCUSSION

Effect of pH

pH plays an important role in advanced oxidation processes and has a significant effect on the production of hydroxyl radicals. Several studies have shown that hydroxyl radicals production is much higher in acidic media [21]. In addition, these radicals are more stable under acidic conditions and this condition causes higher degradation of contaminants [21, 22]. As pH increased, the removal rate of RR-198 decreased due to the reduction of hydroxyl radical [23]. In fact, Fe (II) is oxidized to Fe (III), and the OH[•] with very high redox potential can be produced [13, 16].



Furthermore, high concentrations of H⁺ ions may increase the formation of H[•] radicals and then by using oxygen they change to HO₂[•] radicals and are eventually converted to OH[•] radicals. The oxidation of pollutants in these processes mainly depends on the formation of OH[•] potential [24, 25]. Many other studies also demonstrated a better removal of the dye at a lower pH [26]. Moreover, a higher dye removal at pH=3 was observed for fenton and photo-fenton process which has also been concluded by other researches [27, 28]. Therefore, it can be concluded that at pH more than 3, the Fe (II) ion solubility reduced and the formation of OH[•] decreased due to the formation of ferric hydroxide [14], and at a pH lower than 3, the generated ferric hydroxide reacted with H₂O₂, which resulted in a decrease in the OH[•], eventually decreasing the removal efficiency [29].

Effect of ferrous ion (Fe (II))

According to our results, the removal efficiency generally decreased with an increase in Fe (II) concentration in fenton and photo-fenton processes. The results revealed that a maximum dye removal efficiency was observed at Fe (II) concentration of 10mg/L for the photo-fenton process. This can be attributed to the fact that extra Fe (II) concentrations more than 10mg/L acted as hydroxyl radicals scavenger and eventually decreased the removal efficiency [26]. Other studies showed that Fe (II) concentration had no effect [27,30,31]. Biglari *et al.* [27] concluded that by increasing Fe (II) concentrations more than the optimum level, the removal efficiency of humic acid dramatically decreased [27].

Data showed that the removal efficiency increased by increasing Fe (II) concentration from 10 to 100mg/L in the fenton process, but further increase in the concentration from 100 to 200mg/L decreased the removal efficiency of the dye. The maximum removal was observed at Fe (II) concentration of 100mg/L. Therefore, the optimal concentration of Fe (II) was 100mg/L in the fenton process. It can be assumed that

increasing the concentration of Fe (II) ions up to this amount led to an increase in the production of hydroxyl ions and the removal rate [32]. The decrease in the removal rate at the concentration of more than 100mg/L can be justified by the fact that high concentrations of iron can increase the production of ferrous hydroxide which increases the turbidity and decreases the efficiency [33], and acted as the scavenger for hydroxyl radicals [34]. Other researches reached the same results [35, 36].

Effect of H₂O₂ concentration

H₂O₂ concentration was another factor influencing the removal rate. From the economic aspect, it is substantial to use an appropriate concentration of H₂O₂ because excessive concentration increases the final cost of the process [14].

According to the results, at first, the removal rate of the dye increases significantly as H₂O₂ concentration increased from 25 to 50mg/L. After that, its rate becomes slower (H₂O₂=150mg/L) until reaching a plateau. Therefore, adding proper H₂O₂ concentration (50mg/L) may stimulate the production of OH[•] and increase the capacity to remove the dye efficiently. Extra addition of H₂O₂ concentration forms HO₂[•] with a lower oxidative potential and acts as the scavenger for OH[•] [31, 33]. Many investigations showed the positive effect of H₂O₂ on the removal of the dye by fenton and photo-fenton process [35, 36]. According to Figure 4, photo-fenton process is superior to the fenton process in the removal of the dye since the UV irradiation enhances the production of OH[•].

Effect of RR-198's initial concentration

Based on Figure 5, increasing the concentration of the dye had a negative effect on photo-fenton processes. The removal efficiency of the pollutant was decreased by increasing the dye concentration because of lower penetration of UV photons into the solution causing lower production of hydroxyl radicals [33]. Therefore, some researches revealed that increasing the dye concentration decreased the penetration of the UV light, and lower UV irradiation resulted in the reduction of the hydroxyl radical production, eventually leading to a reduction in the removal efficiency [37]. Other studies also came to similar conclusions [38].

On the other hand in the fenton process, the removal efficiency increased with increasing the initial dye concentrations up to 100mg/L and decreased afterwards.

However, extra addition of the initial dye concentration decreased the removal rate, but this phenomenon can be attributed to the fact that extra addition of the dye (more than 100mg/L) may result in a high competition between the intermediates produced during the degradation of the dye and the original dye molecules. Therefore, increasing the dye

intermediates acts as a disturbing factor in the degradation of the dye [39]. Similar results have also been reported by many studies. 34-39 [40].

Effect of reaction time

Reaction time is an important parameter in many advanced oxidation processes. Based on the results, the removal rate increased when the time increased. Accordingly, the highest colour removal occurred in the first 15min reaction time (66% and 83.19% for fenton and photo-fenton process, respectively) and then the process was slower. Also, the maximum removal rate (92%) occurred at 90 minutes. According to the results, at the first 15 minutes, the rate of removal was very fast and a large amount of dye was removed. In fact, the removal rate improved rapidly in the first 15 minutes and then it became slower. Based on the data obtained in the present study, 90min. reaction time is optimal for the degradation of the dye. It can be concluded that the maximum production of OH[•] occurred at 15min.; then, the production rate was at a slower rate. One of the most important factors for any chemical reactions is the optimization of the reaction time. The optimal reaction time of 15 minutes was selected for this study due to economic reasons. Other researchers also found the same results [41]. Bazrafshan *et al.* showed that the dye removal efficiency was 56% at 5 minutes and the removal rate was increased to more than 95% by increasing the reaction time to 60 minutes. Although the maximum removal efficiency was at 90min., from the economic point of view, the reaction time of 15min was optimal for the removal of RR-198 dye, using the Fenton method.

CONCLUSION

In conclusion, the obtained results suggest that the photo-fenton process had significantly removed the RR-198 in liquid phase compared to the fenton process. The rate of removal showed an initial sharp increase, followed by a relatively slower rate. The removal of the dye was decreased with increasing pH and increased with the reaction time. The optimal conditions for the removal of the dye in the aqueous phase using photo-fenton process (UV/H₂O₂/Fe (II)) are as follows: the initial dye concentration equals to 50mg/L, concentration 10mg/L, H₂O₂ concentration equals to 50mg/L, pH 3, and the reaction time of 15min., respectively. According to the results, the removal efficiency of the dye from aqueous solutions was more than 83% at optimal conditions. The economic aspects were considered for selecting the optimal conditions. Therefore, UV/H₂O₂/Fe (II) process is an efficient, reliable, flexible and cost-effective method to remove RR-198 from industrial wastewater.

ETHICAL ISSUES

Ethical issues such as plagiarism have been observed by authors.

CONFLICT OF INTEREST

There was no conflict of interest.

AUTHORS' CONTRIBUTIONS

The overall implementation of this study including the design, experiments, data analysis, and manuscript preparation were the results of the corresponding author's efforts. All authors have made an extensive contribution to the review and finalization of this manuscript. All authors read and approved the final manuscript.

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