ORIGINAL ARTICLE

Optimization of the Parameters Affecting the Fenton Process for Decolorization of Reactive Red 198 (RR-198) from the Aqueous Phase

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Abstract

Background: Recently, there has been a great concern about the consumption of dyes because of their toxicity, mutagenicity, carcinogenicity, and persistence in the aquatic environment. Reactive dyes are widely used in textile industry. Advanced oxidation processes are one of the cost-effective methods for the removal of these dyes. The main aims of this study were determining the feasibility of using Fenton process in removing Reactive Red 198 (RR-198) dye from aqueous solution and determining the optimal conditions.

Methods: This is a cross-sectional study conducted at a laboratory scale. A total of 69 samples were considered and the effect of pH, Fe (II) concentration, H_2O_2 concentration, initial dye concentration and reaction time were investigated.

Results: According to the results, a maximum removal efficiency of 92% was obtained at pH of 3 and the reaction time of 90 min; also, the concentration of Fe (II), H_2O_2, initial dye concentration were 100 mg/L, 50 mg/L, and 100 mg/L, respectively. The results revealed that by increasing the concentration of Fe (II), H_2O_2 and initial dye, the removal efficiency was increased.

Conclusions: The results showed that Fenton process could be used as a cost-effective method for removing RR-198 dye from textile wastewater efficiently.

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Introduction

Dye production is rapidly increasing in the world.[1] Textile industry is one of the largest consumers of water which produces a significant amount of wastewater.[2] Dye consumption in textile industry is estimated to be more than 10000 tons per year, of this amount approximately 1000 tons of dye is discharged into the wastewater.[3]

Textile wastewater mainly contains several pollutants including acids, bases, dissolved solids, toxic compounds, and organic dyes.[4] Most of the dyes used in textile industries contain organic compounds containing benzene ring.[5] Dyes consist of different chemical structures such as acidic, basic, reactive, dispersive, azo, diazo, anthraquinone-based and metal-complex dyes.[6]

Discharge of industrial wastewater containing dyes into the aquatic environment significantly reduces light transmittance and dissolved oxygen, and increases Chemical Oxygen Demand (COD), which severely disturbs the aquatic life.[7] High concentration of dye significantly decreases the removal efficiency of the biological process in the conventional wastewater treatment plant due to the toxicity of dye on microorganisms.[8] Studies also showed the difficulty of removing dye from textile industrial wastewater because of the wide varieties of complex structure and the presence of a wide range of dyeing additives materials, and detergents.[9]

Different methods have been used to remove the dye including ultrafiltration, reverse osmosis, coagulation and flocculation, ion exchange, biological methods, and adsorption.[10] Advanced oxidation processes (AOPs) is an efficient eco-friendly method in which hydroxyl radicals (OH°) are produced to oxidize dyes.[11] In Fenton process, Fe^{2+} ion and H_2O_2 are used as the reductant and oxidant, respectively, and produce OH° with a very high redox potential. The advantages of Fenton process are low toxicity, high decolorization efficiency, lower cost,[9] and faster oxidation rate.[12] Moreover, studies have showed that these processes have the capability to convert them into CO_2 and H_2O.[13]

The immune system *responds* to RR-198 dyes resulting from allergic *reactions*.[14] Moreover, RR-198 dye is an anionic dye with a very high water solubility. The main environmental issue regarding RR-198 dye is the huge amount of dye wasted during dyeing process.[15] RR-198 dye is considered a potential threat to aquatic life due to its low biodegradability and persistence in the environment.[16]

The study of Ay and colleagues showed that the removal rate increased by increasing the concentration of Fe (II) ion and H_2O_2 using photo-Fenton process. The optimum ratio of H_2O_2/Fe (II)/dyes for the maximum dye removal (100%) was predicted to be equal to 715/71/250 (mg/L).[17] Emami and colleagues indicated that the maximum removal rate of Reactive Red dye 120 (more than 99%) was achieved at optimum pH of 3 by Fenton oxidation method.[9] According to the results of Ghaneyian and colleagues' study, TiO_2 nanoparticles were capable of adsorbing Reactive Red dye 198 considerably in the acidic pH.[18] Bouafia-Chergui and colleagues indicated that 90% of TOC and 100% of the dye were removed using 0.6 mM of Fe^{3+} and 12 mM hydrogen peroxide in photo-Fenton process.[19] Since textile industry is the main consumer of dyes and RR-198 dyes are highly used in many dyeing processes in Iran, there is a concern regarding the effect of dyes in water pollution on people's health and aquatic life. There are various physical, chemical, and biological methods for decolorization, but most convincing is the Fenton process due to its simplicity, low-cost and environmentally-friendly. Therefore, the present paper aimed to (i) investigate the feasibility of using Fenton process to remove RR-198 dye by using Fenton process and (ii) determine the removal efficiency at the optimal conditions.

Methods

Chemicals Characteristics

RR-198 dye is an anionic dye with the molecular weight of 967.5 g/mol ($C_{27}H_{18}C_1N_7Na_4O_{15}S_5$). All the chemicals were purchased from Merck (Germany).

Analytical Method

This cross-sectional study was conducted at a bench-scale using a four-liter glass reactor equipped with a magnetic stirrer that was used for the integration of solution. The study was designed based on one factor at time. Each experiment was performed in triplicates. Finally, 69 samples were used in this study. All data were presented based on the mean value.

Batch Adsorption Experiments

Previous studies showed that the main effective parameters on the oxidation process are pH, Fe (II) concentration, H_2O_2 concentration, initial dye concentration and reaction time [12]. The studied parameters were pH (3, 5, 7, and 9), Fe (II) concentration at five levels (0, 10, 100, 150, and 200 mg/L), H_2O_2 concentration at five levels (0, 25, 50, 75, and 150 mg/L), the initial dye concentration at four levels (50, 100, 150, and 200 mg/L), and reaction time at five levels (15, 30, 45, 60 and 90 min). The maximum dye removal efficiency was determined based on optimal conditions. A four-liter glass reactor equipped with a magnetic stirrer was used for integration of solution. According to the standard method (No 2120), dye concentration was determined at a wavelength of 520 nm using spectrophotometer (DR 5000) [20]. Finally, the data were analyzed using the SPSS software (version 16, SPSS Inc., Chicago, IL, USA).

Dye removal efficiency was calculated by the following equation:

$$R(\%) = \left(\frac{A0 - A}{A0}\right) \times 100$$

Where A_0 and A represented the initial dye concentration before and after Fenton process, respectively. All experiments were done in the Laboratory of the School of Health at Shiraz in 2015.

Results

Effect of pH

According to Figure 1, pH of 3 was optimal for RR-198 dye decolorization with a reduction rate of more than 54%. The removal rate of dye significantly decreased as pH was increased to 7 (11%). Further increase of pH (more than 7) considerably decreased the removal efficiency.

Effect of Ferrous Ion (Fe (II) Concentration)

Figure 2 shows the effects of Fe (II) concentration on decolorization rate of RR-198 dye. The maximum removal efficiency was at Fe (II) concentration of 100 mg/L. The results showed that by increasing the concentration of Fe (II) from 10 to 100 mg/L, the

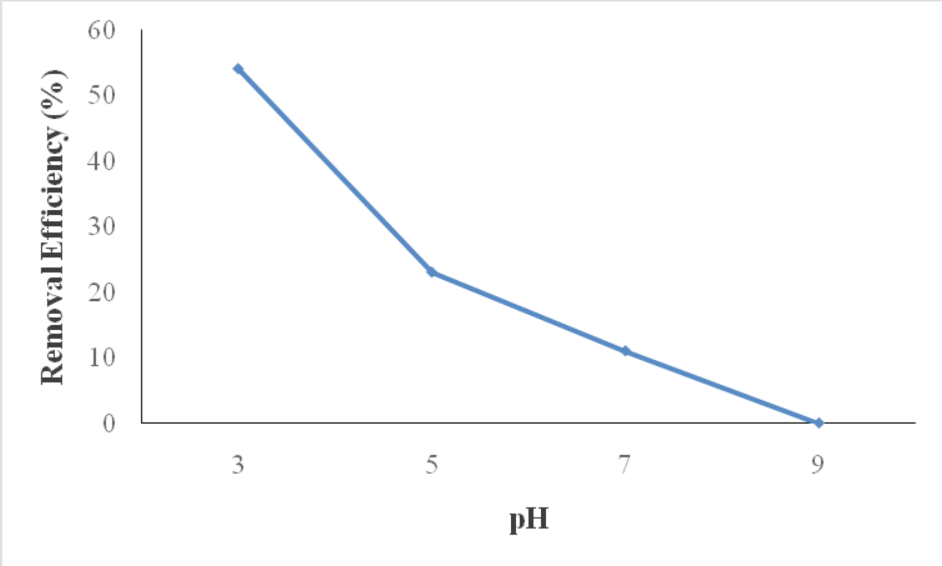

Figure 1: The effect of pH on the removal efficiency of Reactive Red 198 (RR-198) dye by Fenton process at Fe (II) concentration=100 mg/L, H2O2 concentration=50 mg/L, initial dye concentration=50 mg/L and contact time=30 min.

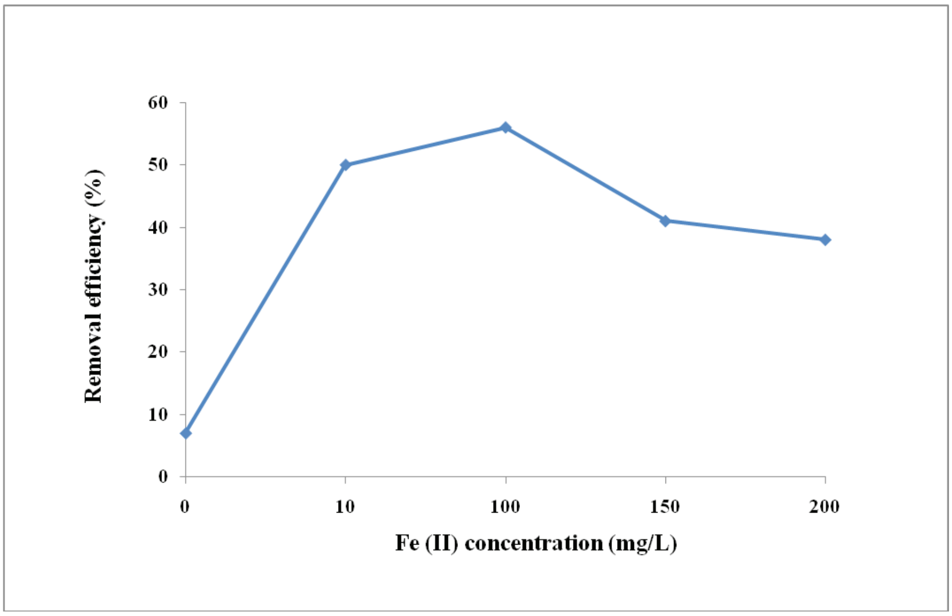

Figure 2: The effect of ferrous ion (Fe (II)) concentration on the removal efficiency of Reactive Red 198 (RR-198) dye by Fenton process at pH=3, H_2O_2 concentration=50 mg/L, initial dye concentration=50 mg/L and contact time=30 min.

removal efficiency also increased to (56%). Increasing the Fe (II) concentration from 100 to 200 mg/L caused a decrease in the removal efficiency (38%).

Effect of Hydrogen Peroxide (H_2O_2) Concentration

Figure 3 shows the effects of H_2O_2 on decolorization rate of RR-198 dye. The result demonstrated that the maximum decolorizing efficiency (68%) was at the H_2O_2 concentration of 50 mg/L. However, by increasing the H_2O_2 concentration from 50 to 150 mg/L, the removal efficiency of RR-198 dye decreased to 47%.

Effect of Initial Concentration of Reactive Red 198 (RR-198)

Figure 4 illustrates the effect of initial concentration of RR-198 dye on the removal efficiency. According to Figure 4, the highest removal efficiency occurred at an initial dye concentration of 100 mg/L. The results showed that by increasing the dye concentration from 50 to 100 mg /L, the removal efficiency increased up to 87%. After that, by increasing the concentration of dye concentration to 200 mg/L, the removal efficiency was decreased to 76%.

Effect of Reaction Time

Figure 5 shows the effects of the reaction time (15, 30, 60 and 90min) on the removal efficiency of RR-198 dye in the aqueous phase. According to Figure 5, the maximum removal efficiency was at the reaction time of 90 min (92%). Data showed that by

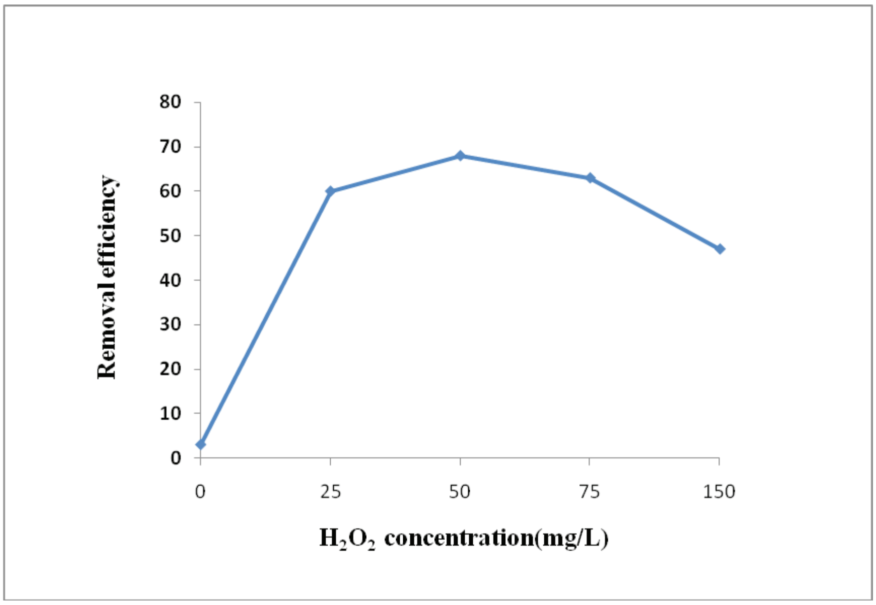

Figure 3: The effect of hydrogen peroxide (H_2O_2) concentration on the removal efficiency of Reactive Red 198 (RR-198) dye by Fenton process atpH=3, Fe (II) concentration=100 mg/L, initial dye concentration=50 mg/L and contact time=30 min.

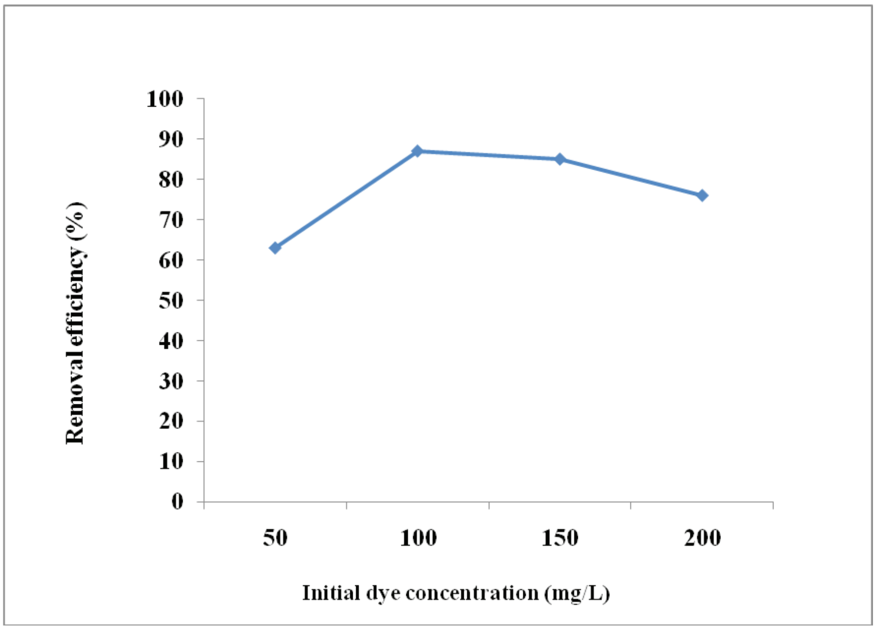

Figure 4: The effect of initial dye concentration of Reactive Red 198 (RR-198) dye by Fenton process at pH=3, Fe (II) concentration=100 mg/L, H_2O_2 concentration=50 mg/L and contact time=30 min.

increasing the reaction time from 15 to 90 min, the removal efficiency increased by 30%.

Discussion

Generally, pH has an important role in the characteristics of the textile industrial wastewater and OH° production. Textile industrial wastewater usually has a wide range of pH.[21] Based on the reports of many researchers, the appropriate pH for the Fenton reaction is in the acidic range, around pH of 3 to 4.[22] Based on the results of other researchers at higher pH (more than pH of 3), the solubility of Fe (II) ion reduced and the formation of ferric hydroxide decreased the formation of hydroxyl radicals. In other words, the dye removal efficiency is reduced due to the instability of the Fe (II) ions at higher pH. In addition, Fe $(OH)^{+2}$ was maintained in the completely soluble form at pH=3.[2,23,24] Moreover, studies also showed that at pH lower than 3, the formed ferrous hydroxide reacted with OH° which in turn reduced the removal efficiency.[2] Ghaneyian and colleagues also demonstrated that the removal of Reactive Blue 19 was obtained in the acidic pH range of 4, using photochemical oxidation process with potassium persulfate and sunlight.[18] Moreover, Ahrampush and colleagues declared that maximum removal of RR-198 dye was obtained at pH=4, using the photo-catalytic TiO_2/ UV-C.[25] Shirmardi and colleagues also had the

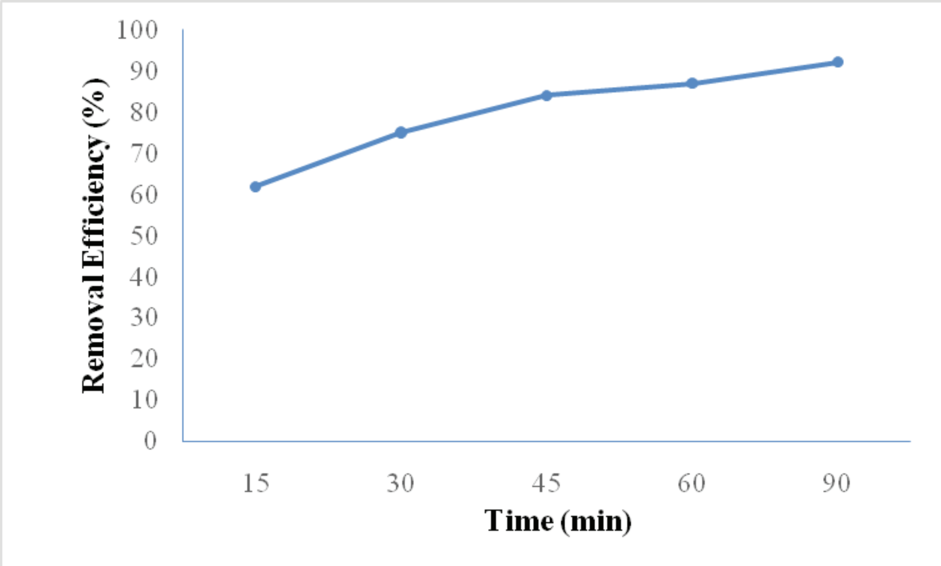

Figure 5: Effect of reaction time on the removal efficiency of Reactive Red 198 (RR-198) dye by Fenton process (pH=3, Fe (II) concentration=100 mg/L, H_2O_2 concentration=50 mg/L, initial dye concentration=100 mg/L)

same results for the highest removal of Acid Red 18 at acidic conditions.[26] This study demonstrated the same results in acidic condition. According to our findings, dye removal efficiency decreased with increasing pH.

The results revealed that by increasing the concentration of Fe (II) up to 100 mg/l, nitrate removal efficiency increased and, then its further increase led to a decrease in the removal efficacy. The same results were also reported by other studies.[27] However, Bahmani and colleagues showed that increasing the concentration of Fe (II) ions had no significant effect on the decolorization rate.[2] When Fe (II) concentration was increased to the optimal concentration (100 mg/L), the OH° production and decolorization rate increased. Increasing Fe (II) concentration more that the optimum level had an inhibitory effect on the production of OH° and acted as the scavenger for hydroxyl radicals, and also reduced the decolorization rate.[28-31]

The findings demonstrated that by increasing the hydrogen peroxide concentration, the rate of removal efficiency was increased. This is due to the formation of more hydroxyl radicals. From the economical point of view, it is substantial to use an appropriate concentration of hydrogen peroxide because excessive hydrogen peroxide increases the final cost of the process. The maximum dye removal efficiency was achieved at optimal H_2O_2=50 mg/L. The optimum molar ratio of H_2O_2 to Fe (II) is equal to 0.5. The excessive addition of H_2O_2 may promote OOH^- formation and act as the hydroxyl radical's scavengers which reduce the oxidation efficiency.[9,29] Daneshvar and colleagues also showed the same results, indicating that addition of hydrogen peroxide up to a certain amount increased the removal rate.[32]

By increasing the initial concentration of the dye to 100 mg/L, the removal efficiency increased to some extent. This result has also been confirmed by other researches.[16,33] However, extra addition of initial dye concentration decreased the removal rate. This can be described by the fact that by increasing the dye concentration, a high competition might occur between the intermediate compounds formed during the degradation process and the former dye molecule. As a matter of fact, the production of intermediate compounds increased with increasing the initial dye concentration.[12] Similar results have also been reported by many studies.[34-39]

Reaction time is an important parameter in many advanced oxidation process. The dye removal efficiency was increased by increasing the contact time. Based on the results, the maximum decolorization rate (92%) occurred at 90 min. According to the results, at the first 30 minutes, the rate of decolorization was very fast and a large amount of dye was removed. In fact, the removal rate improved rapidly in the first 30minutes and then it became slower. Other studies also declared that dye removal efficiency increased significantly by increasing the reaction time.[40-42] Bazrafshan and colleagues showed that dye removal efficiency was 56% at 5 minutes and by increasing the reaction time to 60 minutes, the removal rate was increased to more than 95%. Further increase in the contact time had no significant effect on the removal efficiency.[40] Although the maximum removal efficiency was at 90 min, from the economical point of view, the reaction time of 30 min was optimal for the removal of RR-198 dye using Fenton method.

Conclusion

In conclusion, Fenton process enhances the removal rate of RR-198 dye in the aqueous phase. The optimal conditions for decolorization of RR-198 dye using Fenton method (Fe (II)/H_2O_2) are as follows: the initial RR-198 dye concentration of 100 mg/L, Fe (II) concentration of 50 mg/L, H_2O_2 concentration of 100 mg/L, pH=3, and the reaction time of 90 min. The results indicated that the Fenton process is able to remove 92% RR-198 dye in 90 min. The results showed that Fenton process could be used as an appropriate method for the removal of RR-198 dye from polluted water because of cost effectiveness, lower toxicity, higher removal efficiency, and higher oxidation potential. Therefore, it can be concluded that the Fenton oxidation can be applied successfully for the treatment of industrial dyeing wastewater due to high removal efficiency and cost-effectiveness.

Authors' Contributions

The overall implementation of this study including design, experiments and data analysis, and

manuscript preparation were the results of the corresponding author's efforts. All authors have made extensive contribution to the review and finalization of this manuscript. All authors read and approved the final manuscript.

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Conflict of Interest: None declared.

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