

Full Length Research Paper

Decolorization of the dye reactive black 5 using Fenton oxidation

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Study on the effect of Fenton process on decolorization and mineralization of RB-5 in aqueous solution is presented in this work. Batch experiments were carried out to find the optimal operational conditions including pH, temperature, H₂O₂ dosage, FeSO₄ dosage and RB-5 concentration at low cost. Degradation of RB-5 dye as an organic pollutant was monitored based on decolorization and mineralization extents of the model solution determined by UV-vis spectrophotometric and chemical oxygen demand (COD) analysis respectively. Optimum pH and temperature for 250 mg/L RB-5 were observed at 3.0 and 40°C respectively and using 50 mg/L of FeSO₄ and 300 mg/L of H₂O₂ resulted in generation of 58.9% COD and 97% color removal after 10 min reaction by Fenton. It is concluded that Fenton process can provide better result for decolorization compared to COD removal in the treatment of textile effluent.

Key words: Decolorization, black 5, Fenton oxidation, dye.

INTRODUCTION

The textile industry daily consumes large amounts of water in dyeing and finishing processes (Garca-Montano et al., 2006). Effluents products are intensely colored and contaminated with high concentrations of chemical oxygen demand, suspended and dissolved salts and refractory organics (Meric et al., 2004; Garca-Montano et al., 2006). Reactive azo dyes which can be characterized by one or more azo bonds (-N=N-) are the most commonly ones among more than 10000 dyes used in textile processing industries (Meric et al., 2004; Sun et al., 2007; Zhou et al., 2009). The release of azo dyes in natural environment has resulted in hazardous problems because of their toxicity to aquatic life and humans (Meric et al., 2004; Lucas and Peres, 2006). As a result, azo dyes are major threat to surrounding ecosystems even at low concentration (Zhou et al., 2009; Kusic et al., 2007). The traditional treatment techniques such as coagulation/flocculation, membrane separation (ultra

filtration, reverse osmosis) and adsorption usually transfer the waste compounds from one phase to another causing secondary waste (Sun et al., 2007; Zhou et al., 2009; Lucas and Peres, 2006; Lucas et al., 2007).

Hence, it is necessary to find an effective method of wastewater treatment for both water recourses management and nature preservation (Zhou et al., 2009). Advanced oxidation processes (AOPs) have been proposed and employed for the treatment of hazardous materials in wastewater since 1990s (Sun et al., 2007). In principle, AOPs, regarding to generation of highly reactive hydroxyl radicals (OH[•]), is capable of destroying a wide range of organic pollutants in water and wastewater (Sun et al., 2007; Zhou et al., 2009; Lucas and Peres, 2006; Lucas et al., 2007). Fenton's reagent is a homogeneous catalytic oxidation process that uses a mixture of hydrogen peroxide and ferrous ions in an acidic medium to induce a complex redox for generating

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Table 1. Chemical structure, absorption maxima and molecular weight of RB5.

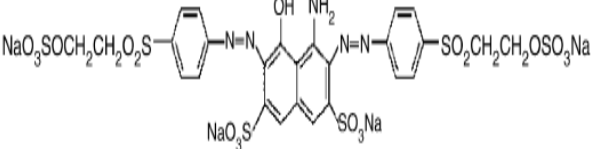
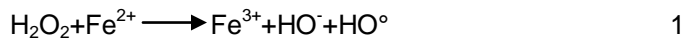
Name	Chemical structure	λ_{max} (nm)	Molecular weight (g/mol)
Reactive Black 5 (diazo dye)		595	991.8

Table 2. First-order rate constant (k), R² values, and half-life (t_{1/2}) of RB5 decolorization.

RB5 decolorization (mg/L)	K (min ⁻¹)	R ²	t _{1/2} (min)
500	0.125	0.944	2
250	0.164	0.952	1
50	0.322	0.961	0.5

of hydroxyl radicals, HO°. The processes are rather complex, but can be shortly represented by following the equation (Eq) (Meric et al., 2004):



Despite the significant oxidizability of Fenton's reagent, producing sludge containing high amount of Fe (III) and high operational costs due to chemical reagent consumption are taken into account as some limitation of its applications (Meric et al., 2004; Garca-Montano et al., 2006; Sun et al., 2007; Zhou et al., 2009).

The Fenton reagent is able to completely decolorize and partially mineralize textile industry dyes in short reaction time (Meric et al., 2004; Feng et al., 2003). Moreover, it can improve organic biodegradability of toxic or non-biodegradable wastewaters (Lucas et al., 2007; Rodrigues et al., 2009). Fenton's reagent is usually used because it is relatively cheap, it can easily be operated and maintained, and there is no need for special equipment (Lucas and Peres, 2006; Kusic et al., 2007; Fongsatitkul et al., 2004). The main objective of this study was to investigate the FO process to remove color and mineralize organic content from synthetic wastewater containing RB-5. The reason of selecting RB-5 is that it is one of the reactive dyes consumed in most cases. Many operational parameters such as pH, Fe²⁺ dosage, H₂O₂ dosage, temperature, and RB-5 concentration influencing on the degradation efficiency are investigated to find the optimum value and make a great comparison with the results obtained from previous research works (Meric et al., 2004; Lucas et al., 2007).

MATERIALS AND METHODS

Azo dye RB-5 (that is color index 20505) was purchased from NimaNasaj Co., Iran. Hydrogen peroxide (30% w/w), ferrous sulfate (Fe₂SO₄·7H₂O), sulfuric acid, and sodium hydroxide were all

obtained from Merck., Germany. All chemicals quantities were of analytical grade and were used without any further purification. Molecular structure of RB-5 has been illustrated (Table 1).

Batch experiments of Fenton's oxidation process were performed in a glass beaker of 2 L at different conditions (that is Fenton dosage in the range of 50 to 400 mg/L H₂O₂, 10 to 100 mg/L FeSO₄, (pH = 2-8), and temperature of 20 to 60°C). The pH of the solution was adjusted using 1 N H₂SO₄ and 1 N NaOH. After optimizing the pH condition, the effects of other parameters were investigated through changing one parameter and keeping the other ones constant (Fongsatitkul et al., 2004).

Experiments were performed using dye solution synthesized (Table 2) containing different concentrations of dye sample from 50 to 500 mg/L (Kusic et al., 2007). A magnetic stirrer was used to mix the solution continuously for 10 min at 100 rpm (Zhou et al., 2009). After the mixing process; pH of the supernatants drawn from each beaker was readjusted up to 8 using 1 N NaOH (Tantak and Chaudhari, 2006; Jozwiak et al., 2004).

The pH of the solution was measured using a digital pH meter. Before the measurement, the pH meter was calibrated with standard buffers of 4, 7, and 10 (at room temperature). The UV-vis spectra of RB-5 solution were recorded from 400 to 700 nm using a Cecil 7100 model spectrophotometer with a 1 cm path length spectrometric quartz cell, showing that the maximum absorbance wavelength of RB-5 was at 595 nm. Prior to the measurement, a calibration curve was obtained using the standard RB-5 solution with the known concentrations (Sun et al., 2007). COD was measured using closed reflex method as outlined in standard methods (APHA, 1998). All results were calculated via averaging the results of triplicate experiments.

RESULTS AND DISCUSSION

Effect of pH

The effect of the initial pH on the decolorization of RB-5 using Fenton process was investigated and the results are demonstrated in Figure 1 (Sun et al., 2007; Lucas and Peres, 2006). The previous studies showed that the pH would influence the amount of OH° generation, and the preferable condition for OH° generation was under acidic conditions (Hsing et al., 2007). Therefore, the

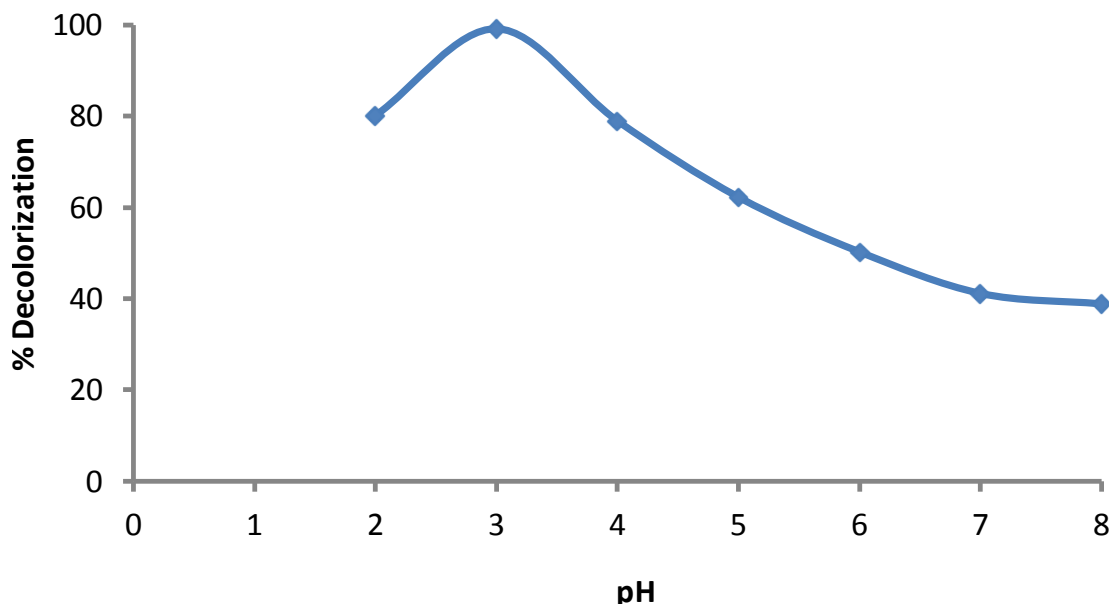


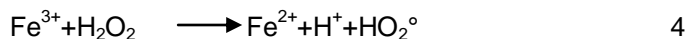
Figure 1. Effect of pH on the decolorization of RB5. Experimental conditions included were: RB5 = 250 mg/L; H₂O₂ = 300 mg/L; FeSO₄ = 50 mg/L; temperature = 40°C; reaction time = 30 min.

experiments were carried out at different pH values ranging from 2 to 8 by adding 1 N H₂SO₄ or NaOH to adjust the pH value. The reaction was carried out for 30 min using 50 mg/L (0.33 mM) FeSO₄ and 300 mg/L (8.82 mM) H₂O₂ under controlled pH conditions. It is apparent from the Figure 1 that the extent of decolorization decreased with increasing the pH and removal efficiency at pH 3 was 99.1%, whereas it reduced to 41.1% at pH 7. These results also are similar to results obtained from previous studies. Sun et al. (2007) observed 98.83% dye decolorization in 30 min. Lucas reported that the optimum pH was 3 for the decolorization of dyes (Lucas and Peres, 2006). Decreasing the oxidation potential of OH° in the high pH condition might be the reason for the decrease in the decolorization rate (Sun et al., 2007).

In addition, under high alkaline conditions (pH > 4), the precipitation of hydroxide ions (Fe(OH)₃) occurs, decreasing the concentration of Fe³⁺ dissolved. Besides, in such conditions, hydrogen peroxide is less stable, therefore, less hydroxyl radicals are formed, reducing the efficiency of process (Rodrigues et al., 2009). On the other hand, for pH condition below 3, the °OH can be consumed via scavenging hydroxyl radicals with H⁺ ions (Equation 2). Subsequently, hydrogen peroxide can capture a proton to form an oxonium ion (H₃O₂⁺) (Equation 3) and H₃O₂⁺ will make hydrogen peroxide to be electrophilic presumably reducing the reactivity of the reaction between hydrogen peroxide and ferrous ion (Peres et al., 2004; Sprink and Woods, 1990; Herney-Ramirez et al., 2008; Oliveira et al., 2006).



Inhabitation of °OH radical formation at pH below 3 seems to be due to decrease of the soluble amount of Fe³⁺ (responsible for the continuity of the oxidation process) that is in equilibrium with other iron species (Fe(OH)² and Fe(OH)²⁺) under such conditions (Lucas and Peres, 2006; Rodrigues et al., 2009).



Effect of H₂O₂ dosage

At the beginning, the effect of H₂O₂ only (without FeSO₄) dosage (from 50 to 400 mg/L, at pH = 3 and Temperature = 40° C) on color removal was investigated. In all cases, colour reduction was negligible (< 10%). Therefore, further experiments were carried out with iron.

The effect of H₂O₂ dose on the decolorization of RB5 through Fenton process was studied and the results are shown in Figure 2. Experiments were carried out at pH 3 with the constant dose of 50 mg/L (0.33 mM) FeSO₄. The H₂O₂ concentration varied in the range from 50 (1.47 mM) to 400 mg/L (11.77 mM).

As shown in Figure 2, the decolorization efficiency that increased from 70.1 to 98.1 is a consequence of increasing H₂O₂ dosage from 50 to 300 mg/L (1.47 to 8.82 mM) after 10 min reaction. The further increase of H₂O₂ from 300 to 400 mg/L (8.82 to 11.77) caused no significant change in decolorization. This is a common behavior in the Fenton's process, which might be due to

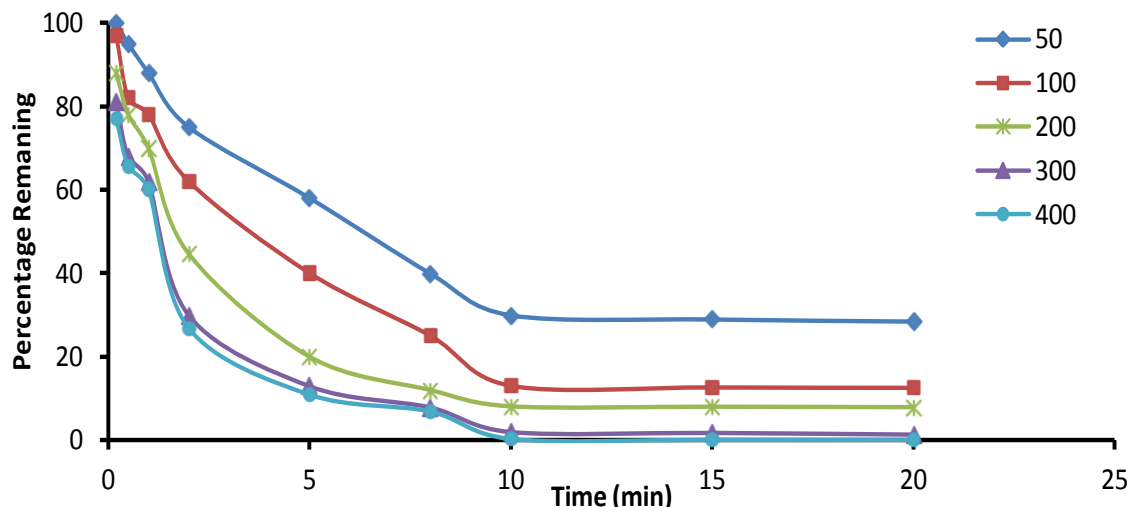
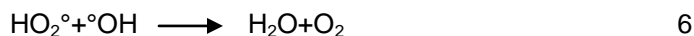
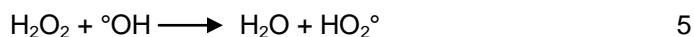


Figure 2. Effect of H₂O₂ concentration on the decolorization of RB5. Experimental conditions: RB5 = 250 mg/L; FeSO₄ = 50 mg/L; Temperature = 40°C; pH = 3.

the hydroxyl radical scavenging effect of H₂O₂ (Equations 5 and 7). According to Lodha and Chaudhari (2007) decolorization rate of dye increases as the H₂O₂ concentration increases until H₂O₂ concentration reaches critical value. However, when the concentration of H₂O₂ was higher than the critical concentration, decolorization of dye was decreased as a result of scavenging effect (Lodha and Chaudhari, 2007; Hsueh et al., 2005; Ramirez and Costa, 2005). The same phenomenon was also observed by Sun et al. (2007) and Lucas and Peres (2006).



Our observations indicated that the decolorization of dye at 300 mg/L (8.82 mM) and 400 mg/L (11.77 mM) of H₂O₂ did not vary significantly as shown in Figure 2. The decolorization efficiency at 300 and 400 mg/l of H₂O₂ dose were 98.1 and 99.7, respectively. Hence, optimum concentration of H₂O₂ was chosen as 300 mg/L (8.82 mM).

Effect of FeSO₄

The effect of adding FeSO₄ ion on the decolorization of RB5 has been studied at optimum pH (pH 3) and lower optimum H₂O₂ dose through varying the concentration of FeSO₄ ions in the range of 10 (0.066 mM) to 100 mg/L (0.66 mM). The results are shown in Figure 3. It was observed that the amount of ferrous ion is one of the main parameters influencing the Fenton process.

The decolorization was increased from 60.1 to 96.6 as the concentration of ferrous ion was increased from 10 to 100 mg/L (0.066 to 0.66 mM) at 10 min. The decrease in the decolorization can be explained by the redox reaction since HO[•] radicals are scavenged by the reaction or with another Fe²⁺ molecule (Equations 8 and 9) (Hsueh et al., 2005).



The Fe³⁺ formed can react with H₂O₂ as well as with hydroperoxy radicals that regenerate Fe²⁺ in the solution resulting in decrease in dye removal (Bentize et al., 2001). It was seen during the studies that maximum dye removal were reached at ferrous ion concentration of 50 mg/L.

Considering the weak distinction in the decolorization efficiency at ferrous ion concentration from 50 to 100 mg/L (0.33 to 0.66 mM), as seen in Figure 3, 50 mg/l (0.33 mM) of FeSO₄ dosage was selected as an optimum dosage for decolorization of RB-5 by Fenton (Sun et al., 2007).

Hence, it was found that 50 mg/l (0.33 mM) of FeSO₄ ion and 300 mg/l (8.82 mM) of H₂O₂ dose at pH 3 is required for > 95% of decolorization of RB-5 dye. The dye: H₂O₂:FeSO₄ mass ratio (w/w/w) of 1:1.2:0.16 was required for 97% RB5. Lodha and Chaudhari (2007) studied decolorization of RB5, RB13, and AO7 and found that the dye: H₂O₂:Fe²⁺ mass ratio (w/w/w) was 1:1:0.3 for 97, 98 and 97% decolorization respectively. Thus, at a lower optimization of FeSO₄ ion and H₂O₂ dosage, operating cost of the treatment can be saved and the amount of sludge generation can be minimized.

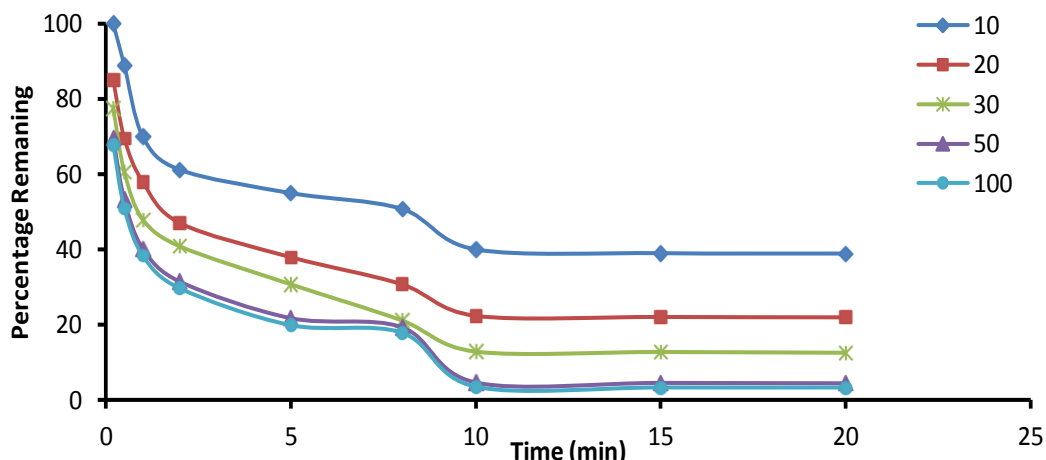


Figure 3. Effect of FeSO_4 concentration on the decolorization of RB5. Experimental conditions: $[\text{RB5}] = 250 \text{ mg/L}$; $[\text{H}_2\text{O}_2] = 300 \text{ mg/L}$; Temperature = 40°C ; $\text{pH} = 3$.

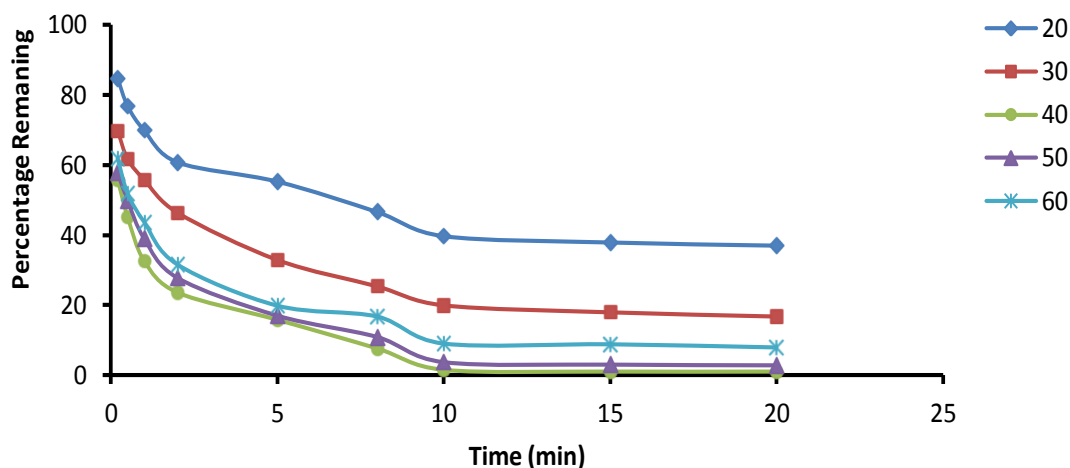


Figure 4. Effect of temperature on the decolorization of RB5. Experimental conditions: $\text{RB5} = 250 \text{ mg/L}$; $\text{H}_2\text{O}_2 = 300 \text{ mg/L}$; $\text{FeSO}_4 = 50 \text{ mg/L}$; $\text{pH} = 3$.

Effect of temperature

As it can be seen in Figure 4, the decolorization of RB-5 increased with the increase of temperature. The decolorization efficiency increased from 60.3 to 98.5% as a consequence of increasing the temperature from 20 to 40°C after 10 min. The rate of the redox reaction can be accelerated by increasing the temperature. However, it does not mean that it will be better at too high temperature. Some studies have indicated that increasing the temperature had negative effect on the dye removal due to the destabilization of flocks resulting in increasing the ferrous ions, which scavenged OH radicals during Fenton's reaction. The removal results have shown that optimum temperature was 40°C at which 98.5% dye removal was obtained after 10 min reaction (Merici et al., 2004; Sun et al., 2007).

Effect of dye concentration

The effect of initial dye concentration of aqueous solution on the decolorization of RB-5 by Fenton process was investigated and the results obtained are shown in Figure 5. The results show that increasing initial concentration from 50 to 500 mg/L decreased the decolorization from 98.2 to 78.9 mg/L in 10 min. This is because when the number of dye molecules increases, the concentration of OH radicals does not increase correspondingly and hence the removal rate decreases (Sun et al., 2007; Lucas and Peres, 2006).

Mineralization of dye

Extent of mineralization of the dye by Fenton process can

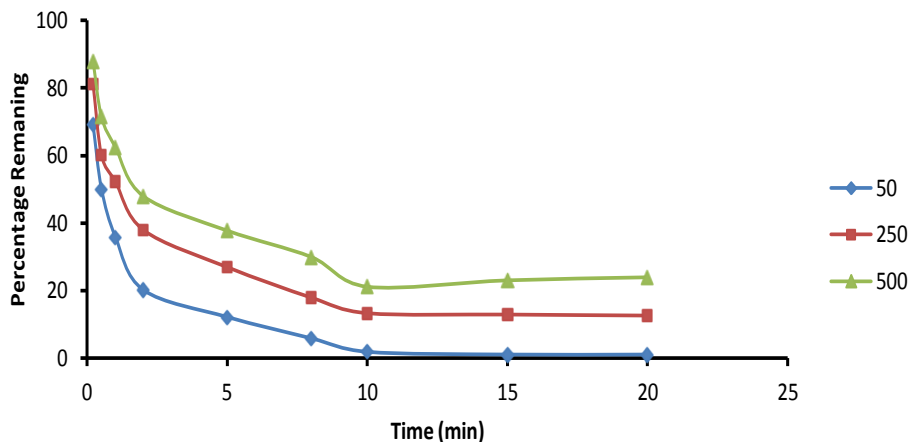


Figure 5. Effect of dye concentration on the decolorization of RB5. Experimental conditions: RB5 = 250 mg/L; H₂O₂ = 300 mg/L; FeSO₄ = 50, mg/L; temperature = 40°C; pH = 3.

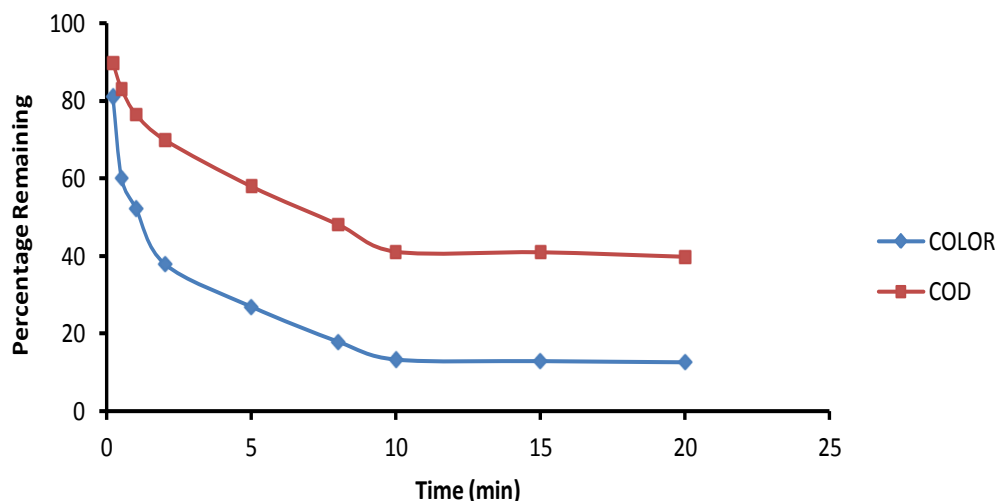


Figure 6. Comparison between the decolorization and COD removal of dye RB5. Experimental conditions: RB5 = 250 mg/L; H₂O₂ = 300 mg/L; FeSO₄ = 50, mg/L; temperature = 40°C; pH = 3.

be evaluated using chemical oxygen demand (COD) measurement. The mineralization of RB5 was evaluated for COD reduction of F.O treated sample. From the results presented in Figure 6, it can be seen that the value of mineralization obtained are significantly low compared to decolorization (Kusic et al., 2007). To characterize the change of RB-5 dye in the COD of reaction medium, initial COD (pure dye solution) and the COD of a sample at different intervals during the reaction were measured and COD reduction was determined as follow:

$$\text{COD removal} = \frac{(\text{COD}_i - \text{COD}_f)}{\text{COD}_i} \times 100 \quad 10$$

Where, COD_i and COD_e are the initial and final COD (mg/L) values respectively. A significant COD reduction

was achieved at a H₂O₂:Fe²⁺ mass ratio of 6:1 for 250 mg/L of RB5 in 10 min, indicating the partial mineralization of dye (Tantak and Chaudhari, 2006; Lodha and Chaudhari, 2007; Kusic et al., 2007; Meric et al., 2004). As could be seen, dye decolorization was much higher than the COD removal. Kuo (1992) reported approximately 90% COD removal in 30 min. Tantak and Chaudhari (2006) observed that about 55.5% COD removal can be achieved in 6 h.

Kinetic analysis

Kinetic studies for decolorization of dye were carried out under the following experimental conditions. Initial dye concentrations were 50, 250 and 500 mg/l, for Fenton's reagent (0.33 mM FeSO₄ and 8.82 mM H₂O₂) at pH 3.

The decolorization of the dye using Fenton reagent can be described as a first order reaction kinetic (Table 2) (Ashraf et al., 2006). Hence, the kinetic data of first the 5 min were fitted into the following equation:

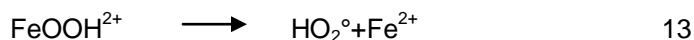
$$\ln\left[\frac{C_t}{C_0}\right] = kt \quad 11$$

Initial decolorization rate constant (k) was determined from the slope of $-\ln\left(\frac{C_t}{C_0}\right)$ versus time plot where C_0 and C_t are dye concentration at zero and t time respectively. Moreover, it is presented half-life time ($t_{1/2}$), that is the time necessary to reduce 50% of initial RB-5 concentration for each process. The half-life time is determined via interpolation of experimental data (Lucas and Peres, 2006; Lucas et al., 2007; Fongsatitkul et al., 2004).

Comparison of k values and $t_{1/2}$ are presented in Table 1. It was observed that the rate constant (k) for a RB-5 concentration of 50 mg/L was more than the concentration of 250 and 500 mg/L. The half-life time ($t_{1/2}$) was four times higher for a concentration of 500 mg/L than that of 50 mg/L (Lucas et al., 2007).

The entire decolorization can be divided into two stages: decolorization was much faster in the first 5 min, and then it proceeded at a slower reaction rate. The first stage followed first order kinetic model while the second one followed the second order kinetic reaction. In the entire experiments, in the first 5 min, more than 60% of color removal occurred and > 75% reduction was observed after 10 min. It is worthwhile to mention that most of the H_2O_2 dosage was consumed in the early stage of the Fenton reaction. Since ferrous ion catalyzed H_2O_2 to form hydroxyl radical in the first stage of reaction, more decolorization occurred in the early stage of reaction.

The probable reason for this well known behavior is that in the first stage, ferrous ions react very quickly with hydrogen peroxide to produce a large amount of hydroxyl radical which can then react rapidly with the dye. Further ferric ions produced in the first stage can react with hydrogen peroxide to produce hydroperoxyl radicals (HO_2°) and ferrous ions through the following reaction scheme:



Thus, hydroxyl radical and hydroperoxyl radicals are formed in the first and second stage respectively. Oxidation capability of hydroxyl radical is much more than that of the hydroxyl radicals. In addition, it can be claimed that the reaction rate decreased on the second stage of the Fenton oxidation is basically due to the fact that ferrous ions are consumed quickly, but reproduced slowly.

Conclusion

In this study, the removal of color and COD of RB-5 were studied using the influence of operating parameters such as pH condition, temperature, $FeSO_4$ and H_2O_2 dosage and RB-5 concentration. The overall process efficiency was investigated and determined based on the UV-vis and COD measurement. Optimal operating conditions included: 300 mg/L of H_2O_2 , 50 mg/L of $FeSO_4$ and temperature and pH of 40°C and 3 for 250 mg/L of RB-5 at 10 min respectively. At these conditions, pseudo-first order degradation rate constants were obtained from batch experimental data. The results obtained indicate that the Fenton's oxidation process leads to decolorization more than 95% of RB-5 dye under these conditions. Also the results have shown that the Fenton process is not only able to decolorize the dye solution but also can partially mineralize the azo dye RB-5.

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