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Sono-photo-Fenton degradation of Reactive Black 5 from aqueous solutions: performance and kinetics

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ABSTRACT

In this study, degradation of Reactive Black 5 (RB5) as a reactive azo dye from aqueous solutions has been performed by an integrated system namely sono–photo-Fenton (H_2O_2/Fe^{2+}/UV/US). The Fenton/UV/US process could show synergistic removal of dye, as compared to Fenton/UV or Fenton process. Batch reactor based on one factorial optimization has been applied for operational parameters: reaction time, pH, H_2O_2 dosage, Fe^{2+} dosage, and initial RB5 concentration. The integrated system significantly removed dye (100%) at the best conditions pH of 3, 50 mg L^{-1} Fe^{2+}, 200 mg L^{-1} H_2O_2, 100 mg L^{-1} RB5 concentration, and reaction time of 30 min. The present study demonstrated that the sono–photo-Fenton could be used as an efficient, reliable method for the removal of RB5 from water and wastewater.

Keywords: Sono–photo-Fenton; Reactive Black 5; Wastewater; Kinetic; Fenton; Decolorization

1. Introduction

Nowadays various pollutants such as dyes [1,2], nitrogen components [3,4], organic carbon [5], heavy metals[6,7] release into receiving waters through industrial wastewater effluent, domestic sewage effluent, landfill leachate, which can cause different environmental and human health problems [8]. Among the pollutants, much attention has recently been focused on dyes removal from water and wastewater. Results of previous research works show that dyes are resistant to biodegradation and they must be removed before discharging into the environment [1,2]. Various physical and chemical methods such as membrane processes [9], coagulation [10], cation exchange membranes [11], and absorption [12] have been used to remove dye from water and wastewater. The aforementioned methods only transfer pollution from the aqueous phase to the solid phase. According to their relatively low efficiency of the above-mentioned methods, they are not considered as acceptable processes [13] and other methods such as advanced oxidation processes (AOPs) are effective and can be performed for pretreatment and final treatment of colored waters. The AOPs work based on producing free radicals of hydroxyl with high oxidation power that can convert many organic chemical compounds to inorganic materials (water and carbon dioxide) [14–16]. Over the last decades, AOPs using Fenton's reagent (H_2O_2/Fe^{2+}), photocatalytic process, photo-Fenton, Ozone, etc. have received great attention as effective methods for water and wastewater treatment [17]. The conventional Fenton's reagent can degrade organic and refractory compounds by producing $^{\bullet}OH$, which is a powerful reactive radical [18]. One drawback of the Fenton-based process is sludge production because of using iron, which needs further separation steps, resulting increase of process overall

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costs [18,19]. To overcome the drawbacks of the conventional Fenton process, an integrated system using UV and ultrasonic irradiation (US) has been studied as a promising way for the degradation of contamination from water and wastewater over the past several years [18,20,21]. Results of previous research works show that accumulation of H_2O_2 as a main product of the sonication can be consumed by combination with other processes such as US/UV and US/Fe^{2+}. The combinations of US/UV and US/Fe^{2+} as homogeneous sono–photo-Fenton is under investigation for performance and synergistic mechanisms [20]. Torres et al. [22] reported that the mineralization of bisphenol A can be achieved through US/UV/Fe^{2+}. Vaishnave et al. [21] studied the sono–photo-Fenton's reaction for the degradation of azure-B and the results displayed that the dye was completely oxidized and degraded into CO_2 and H_2O. Results of a study using the sono–photo-Fenton method for the removal of phenol showed that the integrated process reduced the Fe^{2+} concentration by 30%–50% and the H_2O_2 concentration by 12.5%. This study confirmed that the integrated system (Fenton/UV/US) has a better ability to degrade phenol than individual processes [23].

In this study, an integrated sono–photo-Fenton process has been applied for the degradation of Reactive Black 5 (RB5). To our knowledge, only a few papers have been published on the use of sono–photo-Fenton decolorization. The performance of the integrated system and the effects of variables namely H_2O_2, Fe^{2+}, reaction time, and initial dye concentration on the oxidation process have been investigated.

2. Materials and methods

2.1. Chemicals

All the chemicals applied in this research were of analytical standard. RB5 (chemical formula: $C_{26}H_{21}N_5Na_4O_{19}S_6$; molecular weight: 991.82 g mol^{-1}), $FeSO_4{\cdot}7H_2O$ (P 99.0%), H_2O_2 (P 35%), sulfuric acid (P 97%) and sodium hydroxide were purchased from Merck Company (Germany). Thiosulfate sodium was used for stopping the reaction at the end of the processing time. The structure of the RB5 has been given in Fig. 1.

2.2. Reactor set up and operation

All experiments carried out in a bench-scale reactor to evaluate the efficiency of the Fenton process individually and in combination with ultrasonic and UV processes to remove RB5 from aqueous solutions (Fig. 2). At the first step of experiments, the optimization of Fenton process carried out for H_2O_2 at different reaction time (2, 4, 6, 8, 10, 15, 20, 25, and 30 min) when other factors were constant (pH 4, 25 mg L^{-1} Fe^{+2}, initial dye concentration of 100 mg L^{-1}, agitation of 150 rpm, and temperature 20°C). RB5 degradation was performed by adding H_2O_2 at the desired concentration (10, 25, 50, 100, 200, 300, and 400 mg L^{-1}) to the RB5 solution and then the pH was adjusted to 4 value using H_2SO_4 (0.1 mol L^{-1}) or NaOH (0.1 mol L^{-1}). After pH adjustment, Fe^{2+} was added to the solution. At the end of each run neutralization of radicals carried out by adding equilibrium concentration of sodium thiosulfate. The effects of Fe^{+2} concentration (10–100 mg L^{-1}) and pH values (2, 2.5, 3, 3.5, 4, 5, 6, 7 and 8) on the process investigated as stepwise for each factor by keeping other factors. At the next step, the effect of ultrasonic and UV on the dye degradation was studied separately and in combination with Fenton as an integrated system, after determining the optimal conditions for the H_2O_2 concentration and pH value. The ultrasonic bath (DSA100-SK2-4.0, China) with the capacity of 4 L (Internal dimensions of 235 mm × 135 mm × 150 mm) was used to sonicate RB5 at a frequency of 40 kHz and 100 W and the photo-related tests were conducted using a 25 W lamp as UV-C at 254 nm that located above the ultrasonic bath.

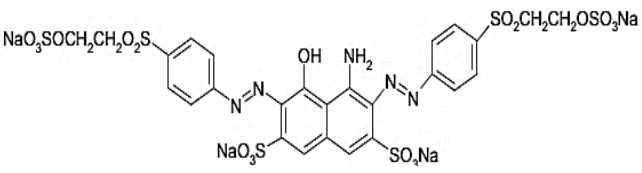

Fig. 1. Structure of Reactive Black 5.

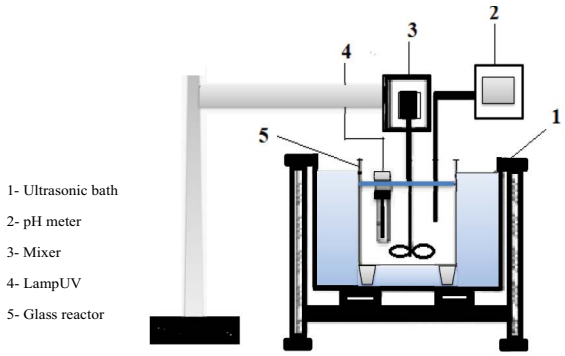

Fig. 2. Schematic of the experimental setup (H_2O_2/Fe^{2+}/UV/US).

2.3. Analytical methods

The concentration of RB5 was determined by measuring the absorption intensity of the solution at 599 nm using a UV/Vis spectrophotometer (Jenway 6305, UK). The dye removal efficiency was defined as follows:

$$\text{RB5 removal efficiency} = \frac{C_0 - C_t}{C_0} \times 100 \quad (1)$$

where C_0 is the RB5 initial concentration (mg L^{-1}) and C_t is the RB5 concentration (mg L^{-1}) at reaction time t (min).

3. Results and discussion

3.1. Effect of H_2O_2 concentration

The effect of H_2O_2 concentration on the Fenton degradation of RB5 was examined while the other parameters were constant (Fig. 3). The initial concentration of H_2O_2 as a powerful oxidizing agent is an important parameter for producing active radicals and also in the performance of dye removal in this process. It was observed that the efficiency of dye removal increases on increasing the

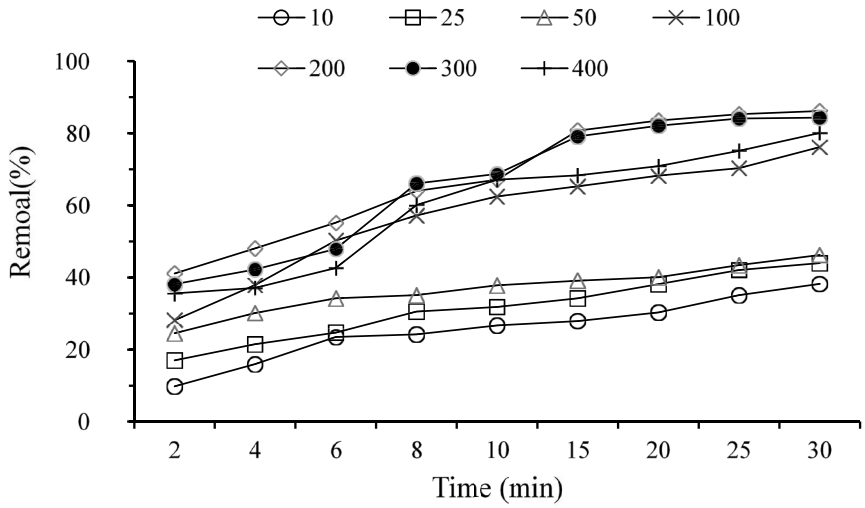

Fig. 3. Effect of H_2O_2 dosage on the RB5 degradation by Fenton process (25 mg L^{-1} Fe^{+2}, 100 mg L^{-1} dye, pH 4).

concentration of H_2O_2, attaining a maximum removal at H_2O_2 of 200 mg L^{-1}. As the concentration of H_2O_2 increased to 400 mg L^{-1} results showed a slight decrease in the removal efficiency of RB5 from the solution because of consuming $^{\bullet}OH$ radicals by extra hydrogen peroxide at high concentration instead of reacting with RB5 (Eqs. (2)–(4)) [24]. The same results have been attained in previous studies. The study of Bai et al. [25] on removing Rhodamine B dye showed that increasing the concentration of H_2O_2 to Fe ions partly increases the removal efficiency and then decreases the removal efficiency.

$$^{\bullet}OH + H_2O_2 \rightarrow {}^{\bullet}HO_2 + H_2O \quad (2)$$

$$^{\bullet}HO_2 + {}^{\bullet}OH \rightarrow H_2O + O_2 \quad (3)$$

$$^{\bullet}OH + {}^{\bullet}OH \rightarrow H_2O_2 \quad (4)$$

3.2. Effect of Fe^{+2} concentration

At the constant value of H_2O_2 (200 mg L^{-1}) and pH 4, the effect of different concentrations of Fe ions has been investigated. It is obvious from Fig. 4 that the removal efficiency of dye increases on increasing the concentration of Fe^{2+} ions from 20 to 50 mg L^{-1} about 87.1% to 92.2%, respectively. These results are because of enhancement in the hydroxyl radicals production that can improve biodegradation of dye [26]. However, on increasing the concentration of Fe^{2+} ions to 75 mg L^{-1}, the RB5 removal efficiency decreased in the Fenton process (Fig. 4). This is because further increases of Fe^{2+} due to the combination of ferrous ion with hydroxyl radical and also because hydroxyl radicals are out of reach, the efficiency of dye removal by hydroxyl radicals decreased [27]. Katsumata et al. [28] studied the photo-Fenton and ultrasonic processes to degrade herbicide linuron. Results showed that the removal efficiency of process increases by increasing the

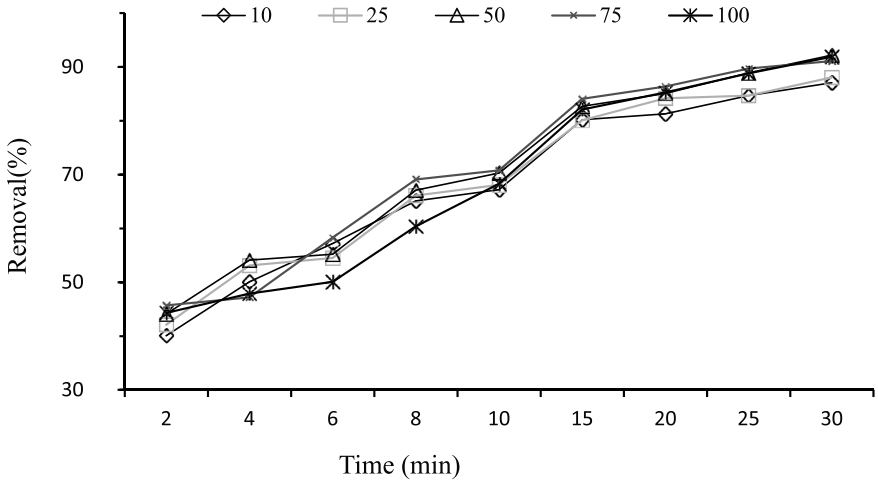

Fig. 4. Effect of Fe^{2+} dosage on the RB5 degradation by Fenton process (100 mg L^{-1} dye, pH 4, 200 mg L^{-1} H_2O_2).

concentration of ferrous ions from 0.05 to 0.1 mM L^{-1} and then the removal efficiency decreases.

3.3. Effect of pH

The effect of different pH values has been investigated to evaluate the effect of pH on the efficiency of the Fenton process in removing RB5 (Fig. 5). The results indicated that at the pH value 2.5 and 3.5, the dye removal efficiency was 91.6% and 93.1%, respectively. Nevertheless, at higher and lower values of pH, the Fenton oxidation efficiency was decreased significantly. At the acidic condition (pH 3), Fenton process has more ability to produce hydroxyl radical, which resulting high level of oxidation [29]. However, at the lower pH, the hydroxyl radicals are used by H^+ ions and the removal rate decreases. Furthermore, H_2O_2 is electro escape and can take a proton in low pH and turn to $H_3O_2^+$, that is a reason to decrease the decolorization (Eqs. (5) and (6)) [30].

$$OH + H^+ + e^- \rightarrow H_2O \quad (5)$$

$$H_2O_2 + H^+ \rightarrow H_3O_2^+ \quad (6)$$

The hydroxyl radicals production can be reduced at the high value of pH and therefore the rate of degradation decreases [31]. Previous studies confirm the achieved results of the present study; Zheng et al. [32] reported that at acidic conditions (pH, 2.5 to 3.5) the removal of Eosin Y by photo – Fenton was in the high level of efficiency. Wang et al. [33] studied the effect of pH on the Brilliant Red KBP. Results confirmed that the reaction rate of degradation at the pH value (2–3) is more than the range of 5 to 8. The same results have been reported by Lu et al. [34] and Özdemir et al. [27].

2.4. Effect of integrated processes

The sono–photo-Fenton degradation of RB5 was carried out at the constant values of H_2O_2, ferrous ions and pH that have been determined in the first phase of this study. In order to investigate the properties of different process; US/H_2O_2, US/UV/H_2O_2, H_2O_2/Fe^{2+}/US/UV, H_2O_2/Fe^{2+}/US, US/UV/H_2O_2, UV/H_2O_2 and H_2O_2/Fe^{2+}/UV in the removal of RB5, a batch reactor have been performed for comparison; the results showing the dye removal efficiency for all methods in Fig. 6. In this research, the individual and different integrated AOPs of Fenton reaction, photo-oxidation, and sonication were investigated in the presence of RB5. It can be seen from the results that the applied integrated process (H_2O_2/Fe^{2+}/US/UV) has a sufficient efficiency of 100% to remove dye than other processes or integrated systems. It is because sonication can produce more hydroxyl radicals by breaking down of H_2O_2 (Eq. (7)), and also react with Fe^{2+} (Eq. (8)) [21,35,36].

$$H_2O_2 \xrightarrow{US} 2OH^{\bullet} \quad (7)$$

$$H_2O_2 + Fe^{2+} \rightarrow OH^{\bullet} + OH^- + Fe^{3+} \quad (8)$$

3.5. Effect of dye concentrations

The most important parameter affecting the efficiency of the process is the concentration of pollutants. So that increasing the concentration of pollutants can reduces the efficiency of pollutants [37]. The removal efficiency of dye using the sono–photo-Fenton process has been investigated by varying the concentration of RB5 (50–400 mg L^{-1}) at constant 200 mg L^{-1} H_2O_2, 50 mg L^{-1} Fe, pH of 3, and reaction time of 30 min. It can be observed from Fig. 7 that the initial dye concentration at the higher value of 400 mg L^{-1} attained lower decolorization for RB5 (37.1%) by the sono–photo-Fenton process. It is because of constant OH radicals in solution when the concentration of dye molecule increases and therefore more radical consumption takes place during the oxidation process. Furthermore, OH radical generation can be reduced at the higher number of dye molecule because of decrease in the penetration of photons entering into the solution [18]. Özdemir et al. [27] investigated the degradation of Reactive Yellow 145 by Fenton-ultrasonic process. Results showed that the efficiency of process decreased from 95%,

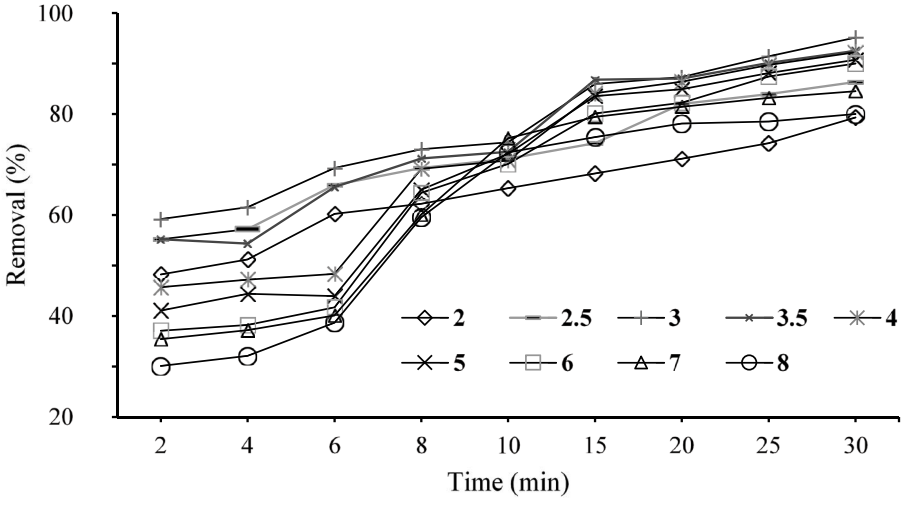

Fig. 5. Effect of pH on the RB5 degradation by Fenton process (50 mg L^{-1} Fe^{+2}, 100 mg L^{-1} dye, 200 mg L^{-1} H_2O_2).

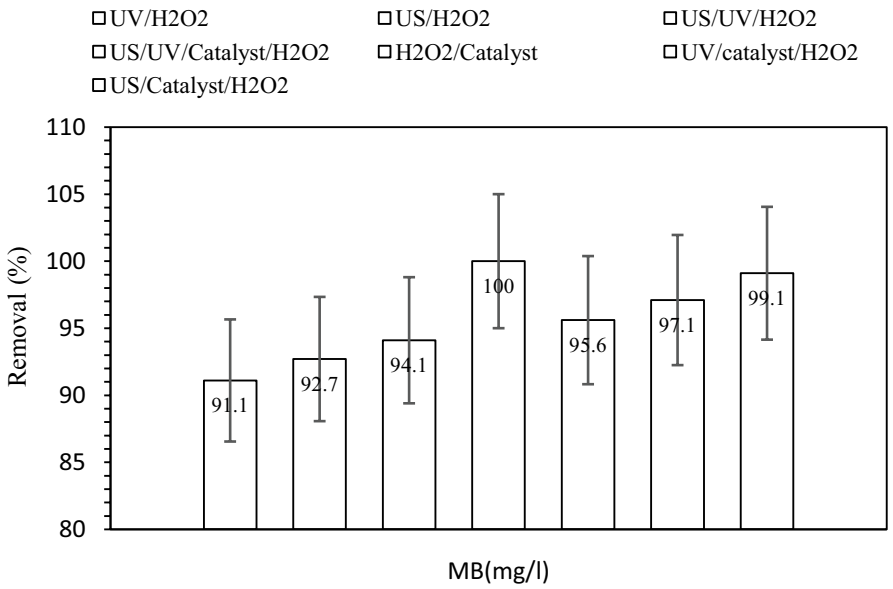

Fig. 6. RB5 degradation performance under different oxidation processes (50 mg L^{-1} Fe^{+2}, 100 mg L^{-1} dye, 200 mg L^{-1} H_2O_2, pH = 3, Time = 30 min).

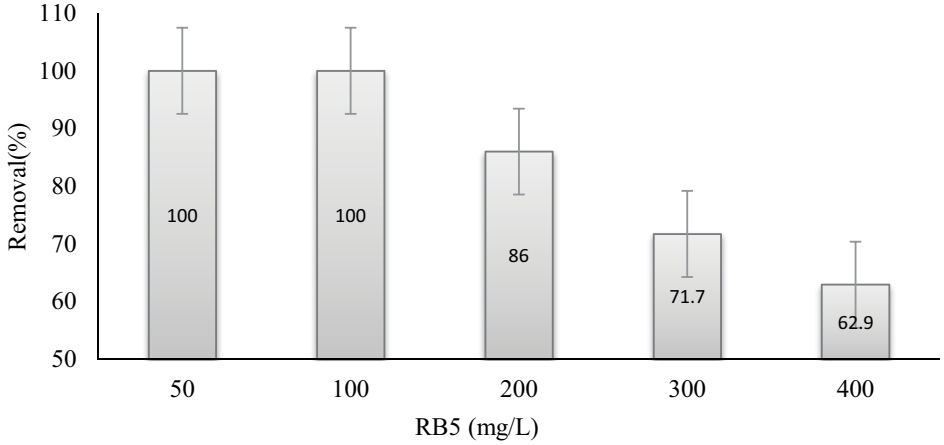

Fig. 7. Effect of initial dye concentration on the RB5 degradation by sono-photo-Fenton (50 mg L^{-1} Fe^{+2}, 100 mg L^{-1} dye, 200 mg L^{-1} H_2O_2, pH 3, and 30 min reaction time).

to 71% on increasing of the dye concentration from 50 to 250 mg L^{-1}, respectively.

3.6. Effect of contact time

In this study, the effect of reaction time on the efficiency of removing RB5 during the sono–Photo-Fenton process was studied. The results indicated that the most dye removal occurred in the first few minutes and the maximum removal of 100% take place at the 30 min (Fig. 8). Lin et al. [38] confirmed that the increase of contact time causes increasing decolorization for Acid Red 14 by the Fenton process.

3.7. Kinetic study

In the present study, several kinetic models were tested to investigate the degradation of RB5 by the integrated system (H_2O_2/Fe^{2+}/UV/US), which regression coefficients (R^2) obtained by graphical representation according to Fig. 9 fits best with pseudo-first-order. From the slope, a first-order rate constant $k_1 = 0.1505$ min^{-1} was calculated for sono–photo-Fenton. The minimum reaction rate constant of 0.0567 min^{-1} is dedicated to the H_2O_2/UV process (Fig. 9).

4. Conclusion

In this study, sono–photo-Fenton processes under different experimental conditions investigated the degradation of RB5 in aqueous solution. The decolorization using the Fenton process was affected by main variables namely pH, initial dye concentration, H_2O_2 dosage, Fe^{+2} dosage, and reaction time. The efficient RB5 degradation carried out at 200 mg L^{-1} H_2O_2, Fe^{+2} of 50 mg L^{-1}, 100 mg L^{-1} dye by Fenton oxidation.

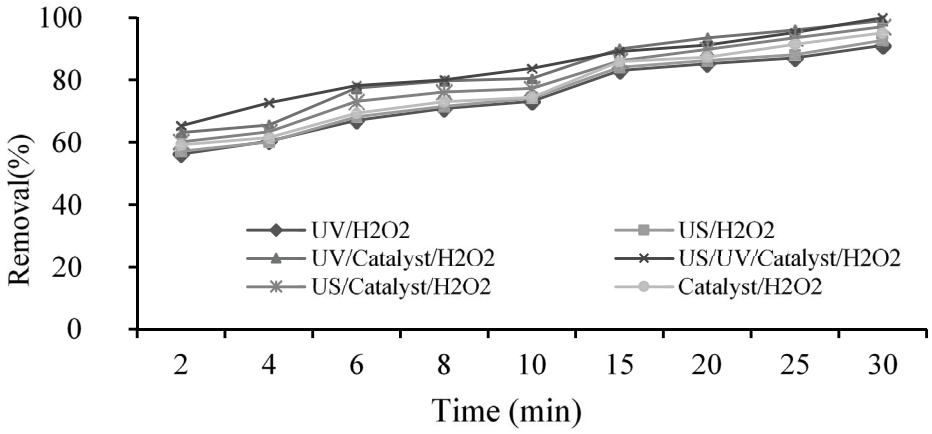

Fig. 8. RB5 degradation at different reaction time using different systems (50 mg L^{-1} Fe^{+2}, 100 mg L^{-1} dye, 200 mg L^{-1} H_2O_2, and pH 3).

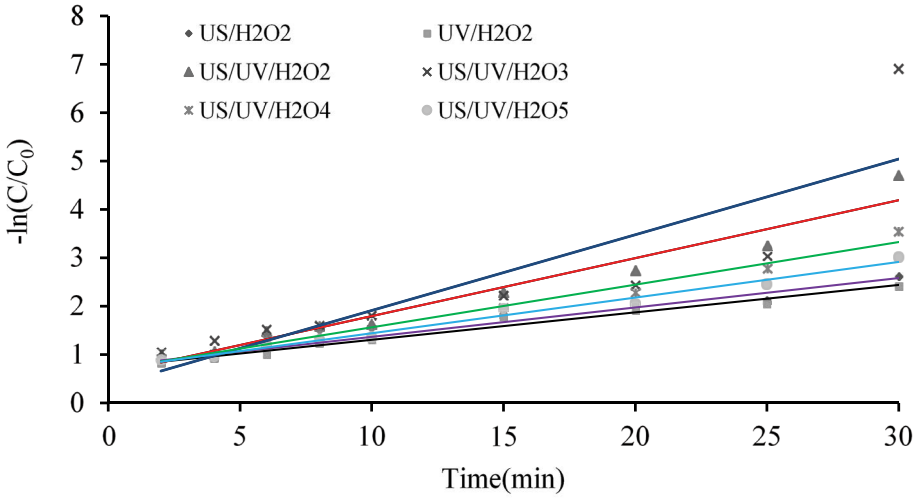

Fig. 9. Kinetic rate constants for the decolorization of RB5 under different oxidation processes (50 mg L^{-1} Fe^{+2}, 100 mg L^{-1} dye, 200 mg L^{-1} H_2O_2, and pH 3).

The removal efficiency of the sono–photo-Fenton process can be affected by alteration in the concentration of H_2O_2 and Fe^{2+}. Furthermore, higher removal of 86.2% and 92.2% achieved when 50 and 200 mg L^{-1} H_2O_2 have been used, respectively. The increase in the initial RB5 concentration harmed the degradation, with the effects of the experimental parameters on the decolorization resulting in comparable reaction rate constants. The rate constant decreased with increasing concentration of dye. The results showed that sono–photo-Fenton is an appropriate integrated process with high removal efficiency of RB5 from aqueous solutions. This integrated process can be considered as a promising and effective method for the removal of dye from water and wastewater.

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