



Use of Fenton reagent as advanced oxidative process for removing textile dyes from aqueous solutions

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Abstract

Decolorization of aqueous solutions containing an azo dye (Reactive Yellow 84) was achieved by advanced oxidative process using Fenton reagent. The optimum amounts of Fenton reagent was 25 mg/L of Fe²⁺ and 250 mg/L of H₂O₂ for an initial Reactive Yellow 84 concentration at 60 mg/L. The initial Fe²⁺ concentration in the Fenton reagent affected the degradation efficiency, rate and kinetics. The ratio of Fe²⁺/H₂O₂ is found equal to 0.1 to give the best result for the decolorization efficiency. The Fenton process was effective under pH 3 and the decolorization efficiency of Reactive Yellow 84 attained 85% for 20 min reaction time. Kinetics decolorization of RY84 followed pseudo second-order reaction. The reaction characteristic of oxidative reaction for decolorization efficiency process was evaluated as thermodynamically spontaneous under natural conditions. The value of activation energy is determined and is equal to 16.78 kJ/mol, this low value may show that the oxidative reaction proceeds with low energy barrier.

Keywords: Decolorization, reactive yellow 84, fenton's oxidation, kinetics, thermodynamics

I. Introduction

Many industrial processes use different chemical dyes for various purposes (textile, cosmetics, food, pharmaceutical, paper, pulp manufacturing, dyeing of cloth, leather treatment, printing...). The presence of dyes in water is most undesirable, so a very small amount of these coloring agents are highly visible and may be toxic to the aquatic environment. Dyes absorb and reflect sunlight entering the water and so they can interfere with the growth of bacteria and hinder photosynthesis in aquatic plants [1,2]. Also it is well known that dye effluents from dyestuff manufacturing and textile industries, may exhibit toxic effects on microbial populations and can be toxic and/or carcinogenic to mammalian animals [3]. Therefore, removal of dyes from industrial effluents before discharge is an important aspect of wastewater treatment; it is a major environmental problem. Conventional methods for the removal of dyes in effluents include physical, chemical, and biological processes [4-7]. Currently, we use different chemical and physical processes such as the elimination by adsorption on activated carbon, coagulation by chemical agents, oxidations by ozone or hypochlorite, electrochemical methods [8-11]. Recently, we have tendency to use solid wastes as low-cost adsorbents [12-18]. These methods do not eliminate the colors completely, sometimes they are expensive and usually they cause other waste pollutants as secondary products. Therefore, the purification of colored water discharges remains a problem for many industries and it is necessary to develop new technologies to treat these waters.

Recently, there is a potential interest for so-called of advanced oxidation processes (AOPs) which constitute an attractive alternative to treating wastewater containing toxic and persistent pollutants. Among them, the oxidation using Fenton's reagent has proved to be a promising and attractive treatment method for the effective decolorization and degradation dyes, as well as for the destruction of a large number of hazardous and organic pollutants [19-21]. Fenton's reagent is a homogeneous catalytic oxidation process use a mixture of hydrogen peroxide iron II ions in acidic medium to induce a complex redox for generating of hydroxyl radicals with a oxidation potential of 2.8 V [31-35] are stronger oxidants than ozone 2.07 V and hydrogen peroxide 1.8 V. Hydroxyl radicals react rapidly and indiscriminately with most of the organic compounds, either by addition to a double bond or by abstraction of a hydrogen atom from an aliphatic organic molecule [31]. The resulting

organic radicals then react with oxygen to initiate a series of oxidation reactions ultimately leading to mineralization products of carbon dioxide and water [36,37].

In the current experimental research, we use the Fenton process for decolorization of aqueous solutions by the determination of the optimum conditions of some experimental factors. The aim of this work is to determine the effect of various parameters on the degradation of dye in aqueous solution by this advanced oxidative process. The effects of pH, the concentration of hydrogen peroxide, iron II and RY84, the temperature to determine the optimal experimental conditions for a potential capability of the degradation dye.

2. Materials and Methods

The azo dye RY84 (CAS 61951-85-7) was selected for this study since it has been commercially important and commonly used in textile industries to dye the natural fibers/fabrics. As dyeing operators want to fix all of dye to fabric, textile wastewater usually contains lower dye concentrations. However, some previous studies have stated that textile industry wastewaters contain different type of dyes and their concentrations vary from 10 to 200 mg/L [38]. Therefore, RY84 concentration of aqueous solution was selected as 60 mg/L to represent actual textile wastewater. The chemical structure of the dye is shown in Figure 1.

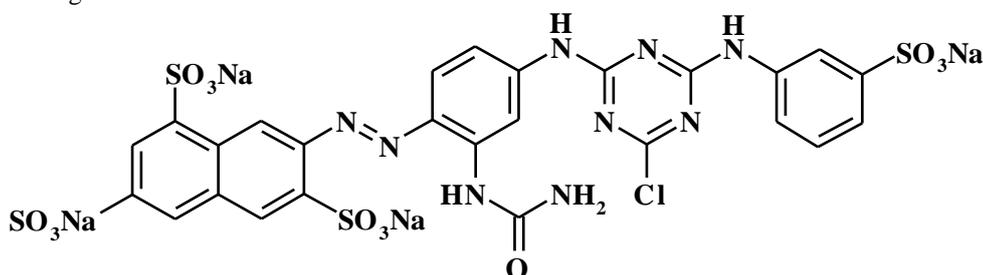


Figure 1: Chemical structure of Reactive Yellow 84

All tests were conducted in 250 mL capacity glass beakers and reaction solution stirred during 30 min. Temperature was controlled through a thermostat. The required amounts of RY84 from stock solution and ferrous sulfate were introduced for reaction and then diluted with deionized water to 200 mL. The pH of each solution was adjusted to the appropriate value by adding NaOH 10^{-1} mol/L or H₂SO₄ 10^{-1} mol/L. The reaction was initiated by adding different amounts of hydrogen peroxide to the reactor after the pH was adjusted. The start time was recorded in order to measure the reaction period when the solution of H₂O₂ is added. Samples were periodically taken out from the reactor using a pipette and then absorbance measurements of the reaction solutions were immediately performed.

Color measurement of RY84 was recorded from 200 to 800 nm wavelengths using a UV/Vis spectrophotometer (BioMate 6, England). The samples were centrifuged at 500 rpm for 5 min and the absorbance values of supernatants were measured. Therefore, the concentration of the RY84 in the reaction mixture at different reaction times was determined by measuring the absorption intensity at $\lambda_{\max} = 618$ nm and from a calibration curve. Prior to the measurement, a calibration curve was obtained by using the standard RY84 solution with known concentrations. Because the reaction continued after sampling, the measurement of absorbance of reaction solution was done within 1 min. The degradation efficiency of RY84 was defined as follows:

$$\text{Decolorization..Efficiency} = \left(1 - \frac{C_t}{C_0}\right) \times 100 \quad (1)$$

Where C_0 (mg/L) is the initial concentration of RY84, and C_t (mg/L) is the concentration of RY84 at reaction time t (min).

3. Results and discussion

3.1. Effect of H₂O₂ and Fe²⁺ concentrations on the removal of RY84

Concentrations of H₂O₂ and Fe²⁺ are important parameters to optimize for the determination of the Fe²⁺/H₂O₂ ratio to achieve the maximum removal efficiency of RY84 from aqueous solutions and this ratio has been used in the following experiments. Blank experiments showed that neither decolorizing nor mineralization of RY84 occurs in the presence of Fe²⁺ ions alone. Color removal was also negligible in the presence of only H₂O₂. The first step consists to take H₂O₂ concentration constant at 250 mg/L and studying the effect of the variation of Fe²⁺ concentrations (10, 25, 50 and 100 mg/L) on the decolorization efficiency (%) at initial pH solution and at ambient temperature. Figure 2 depicts the effects of H₂O₂ concentration and oxidative reaction time on the decolorization efficiency. In the light of these results, it appears that for all H₂O₂ concentrations the reaction time don't affect the decolorization efficiency. Equilibrium is reached in a short time much less than 20 min.

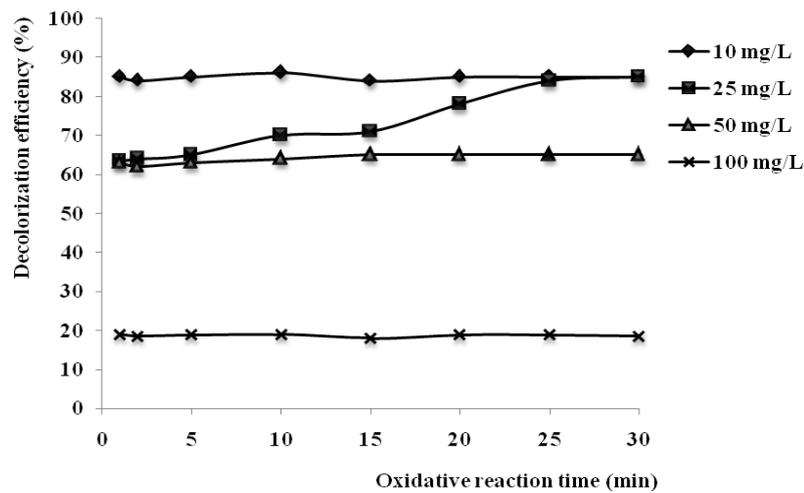


Figure 2: Effect of Fe²⁺ concentration and oxidative reaction time on the decolorization efficiency (%) with [H₂O₂] = 250 mg/L, initial pH and ambient temperature

Also, the decolorization efficiency decreases when H₂O₂ concentration increases. After 20 min of reaction time, it varied from 19% to 85%. Moreover, removal RY84 dye by oxidative process was also studied at constant Fe²⁺ concentration and with variable H₂O₂ concentration (50, 100, 250 and 500 mg/L). The required data are summarized in Figure 3. H₂O₂ concentration causes an important effect to the decolorization efficiency and with the same manner after 20 min of reaction time, when H₂O₂ concentration increases the decolorization efficiency increases and is varied from 64% to 85%. Accordingly with previous work [39,40], the excessive Fe²⁺ and H₂O₂ reagents reduced the RY84 removal of oxidation efficiency.

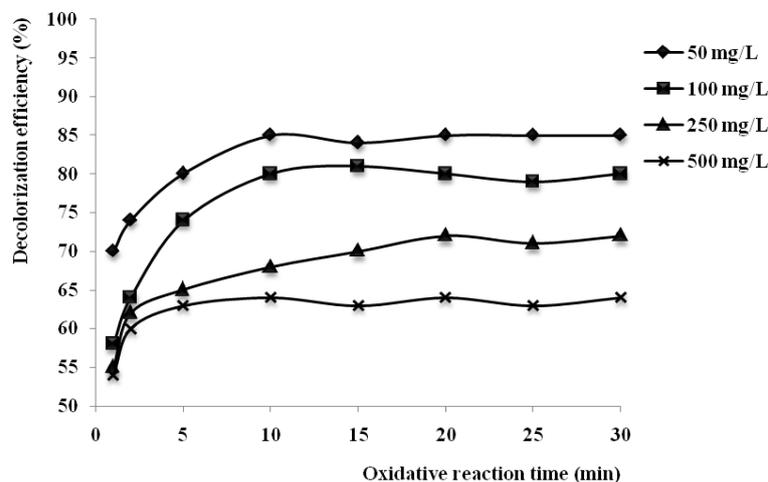


Figure 3: Effect of H₂O₂ concentration and oxidative reaction time on the decolorization efficiency (%) with [Fe²⁺] = 25 mg/L, initial pH and ambient temperature

However, further increase of Fe³⁺ concentration, decreases the mineralization efficiency, this may be due to the increase of a brown turbidity that hinders the absorption of light required for the fenton process and to scavenging effects of hydroxyl radicals by Fe³⁺ and Fe²⁺ since these parasitic reactions become competitive at higher Fe²⁺ or Fe³⁺ concentrations. The decrease in the decolorization can be explained by the redox reaction since hydroxyl radicals HO[•] are scavenged by the reaction with another fenton reagent Fe²⁺ by the following reactions:



From these Figures 2 and 3, we can take 250 mg/L as optimum H₂O₂ concentration and 25 mg/L as optimum Fe²⁺ concentration. Consequently, we can say that the optimum Fe²⁺/H₂O₂ molar ratio is 0.1.

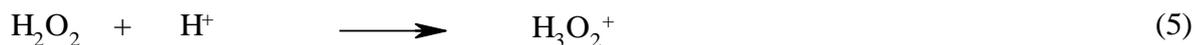
3.2. Effect of pH on RY84 removal from aqueous media

The pH of the textile water discharge is a very important parameter in the Fenton's process. It affects directly the mechanism of oxidation dye, because a change in pH of the solution, involves a variation of the concentration of Fe^{2+} , and therefore the rate of production of HO° radicals responsible for oxidation dyes, will be restricted.

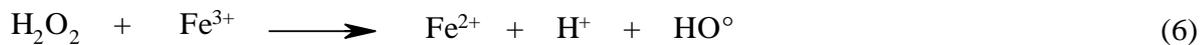
The effect of the initial pH on the decolorization efficiency of RY84 from aqueous solutions using fenton process was investigated and the results have been depicted in Figure 4.

The previous works have shown that the pH would influence on the amount of HO° generation and the preferable condition for HO° generation was under acidic conditions. Therefore, the experiments were carried out at different pH values ranging from 2 to 10. The reaction was carried out for 20 min using 25 mg/L of Fe^{2+} ions and 250 mg/L of H_2O_2 under controlled pH conditions. It is apparent from Figure 4 that extent of decolorization decreases with increasing the pH and the removal efficiency at pH 3 is 85%, whereas it is reduced to 45% at the pH 7. The decreasing of the decolorization rate is attributed to the decreasing of oxidation potential of hydroxyl radicals in the high pH. In addition, under alkaline conditions (pH > 4), the precipitation of hydroxide ions $Fe(OH)_3$ occurs, decreasing the Fe^{3+} concentration dissolved. Besides, in such conditions, hydrogen peroxide is less stable; therefore less hydroxyl radicals are formed reducing the process efficiency. In the other hand, for pH condition

below 3, the HO° can be consumed via scanning hydroxyl radicals with H^+ ions. Subsequently, hydrogen peroxide can capture a proton to form an oxonium ion $H_3O_2^+$ and will make hydrogen peroxide to be electrophilic presumably reducing the reactivity of the reaction between hydrogen peroxide and iron ion. The occurred reactions are described as follows:



Inhibition of HO° formation at pH below 3 seems to be due to decrease of the soluble amount of Fe^{3+} 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 as follows:



The results were in accordance with those obtained in previous studies [41-43].

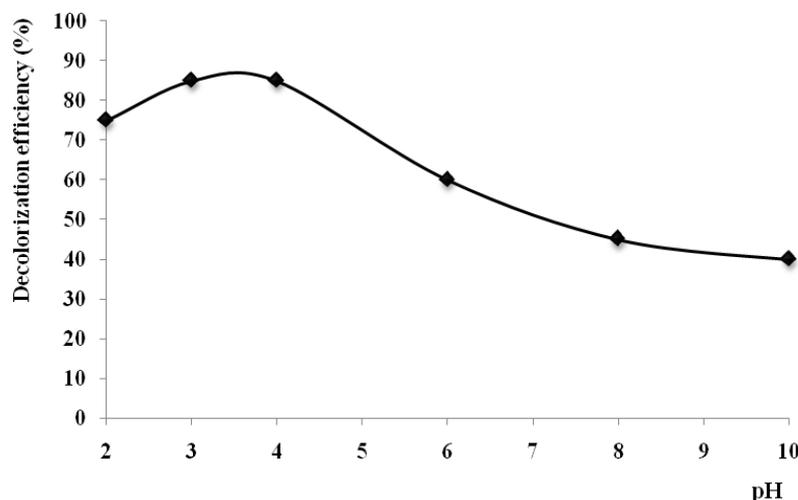


Figure 4: Effect of pH on the decolorization efficiency (%) with Fe^{2+}/H_2O_2 molar ratio is 0.1, ambient temperature

3.3. Kinetics studies for the removal of RY84

The decolorization kinetics of RY84 by Fenton process was studied for various contact times varying from 5 to 30 min at different temperatures 293, 303, 313 and 323 K. Already, kinetics models are established to perform such studies and then occurred by the first and second order kinetics as described by the equations 7 and 8 [44-45]:

$$\text{Log}(C_t) = \text{Log}(C_0) - k_1 t \quad (7)$$

$$\frac{1}{C_t} - \frac{1}{C_0} = k_2 t \tag{8}$$

Where C_0 is initial concentration of the RY84, C_t is concentration of the RY84 at time t , k_1 (min^{-1}) and k_2 ($\text{mg} \cdot \text{L}^{-1} \cdot \text{min}^{-1}$) are the rate constants of the first order and second order kinetic equations.

Values of k_1 and k_2 were determined respectively by the plot of the curves of $\text{Log}(C_t)$ versus time and $1/C_t$ versus time at different temperatures (293, 303, 313 and 323 K). All data found were presented in Table 1. The kinetic study was discussed in the first time by using the first and second orders models. In the light of these results and in order to fit the best experimental data, we compare the correlation coefficient of the both models. The calculated of the half times of the both models are very higher. Thus, the temperature affect on both of the rate constants and the half times of the two models. In this fact, with increasing temperature from 293 to 323 K, the rate constants decrease from 0.056 to 0.023 min^{-1} for the first order kinetic model and from 0.223 to 0.173 $\text{L} \cdot \text{min}^{-1} \cdot \text{mg}^{-1}$ for the second order kinetic model. In the same manner, increasing temperature causes an increasing of half time, it varied from 20.56 to 24.79 min for first order model and from 9.78 to 11.98 min for second order model. However, in order to found better results, we tested another time the data given by the pseudo first ad second order models.

Table 1: Data of the first and second orders kinetics

T (K)	First order model			Second order model		
	k_1 (min^{-1})	$t_{1/2}$ (min)	R^2	k_2 ($\text{L} \cdot \text{min}^{-1} \cdot \text{mg}^{-1}$)	$t_{1/2}$ (min)	R^2
293	0.056	20.56	0.656	0.223	9.78	0.798
303	0.045	22.76	0.537	0.229	10.13	0.687
313	0.034	23.87	0.426	0.178	11.78	0.675
323	0.023	24.79	0.349	0.173	11.98	0.547

The following equations 9 and 10 describe respectively the pseudo first and second orders models [44-46].

$$\text{Log}(C_t - C_e) = \text{Log}(C_0 - C_e) - \left(\frac{k_1}{2,303} \right) t \tag{9}$$

$$\frac{t}{C_t} = \frac{1}{k_2 C_e^2} + \left(\frac{1}{C_e} \right) t \tag{10}$$

where C_e is equilibrium concentration of the RY84.

Values of k_1 and k_2 were determined respectively by the plot of the curves of $\text{Log}(C_t - C_e)$ versus time and $1/C_t$ versus time at different temperatures (293, 303, 313 and 323 K). All data found were presented in Table 2.

Figure 5 depicts the plot of the curve of pseudo second order model. Finally, we can deduce that the kinetic study occur favorably by the pseudo second order model.

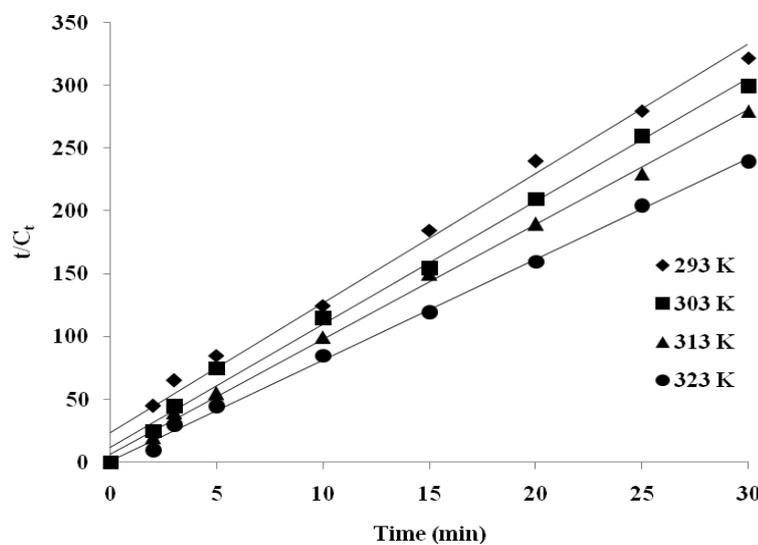


Figure 5: Plot of pseudo second order kinetic model

Table 2: Data of the pseudo first and second orders kinetics

T (K)	Pseudo first order model			Pseudo second order model		
	k_1 (min ⁻¹)	$t_{1/2}$ (min)	R ²	k_2 (L.min ⁻¹ mg ⁻¹)	$t_{1/2}$ (min)	R ²
293	0.061	21.34	0.876	6.56	2.67	0.998
303	0.042	23.17	0.757	9.45	2.13	0.997
313	0.031	24.67	0.754	10.76	1.96	0.998
323	0.019	25.23	0.697	12.75	1.94	0.998

As shown in Table 2, correlation coefficients of kinetic study of pseudo second order are very close to unit and are higher than those of pseudo first order.

3.4. Thermodynamics studies for the removal of RY84

In order to study the effect of reaction temperature on the RY84 decolorization, data were collected at four different temperatures (293, 303, 313 and 323 K). The temperature dependence of the kinetic parameters of Fenton treatment was calculated by Equation (11) [44]. The Gibbs free energy, ΔG , for the Fenton process was also obtained using Equation (12).

$$\ln k = \ln A - \left(\frac{E_a}{2.303R} \right) \left(\frac{1}{T} \right) \quad (11)$$

$$\Delta G = -RT \ln(k) \quad (12)$$

Where k is the rate constant which controls process (pseudo second order reaction rate constant), A is the Arrhenius constant, T is the solution temperature in K, E_a is activation energy (kJ/mol) and R is the ideal gas constant (0.0083 kJ/mol. K).

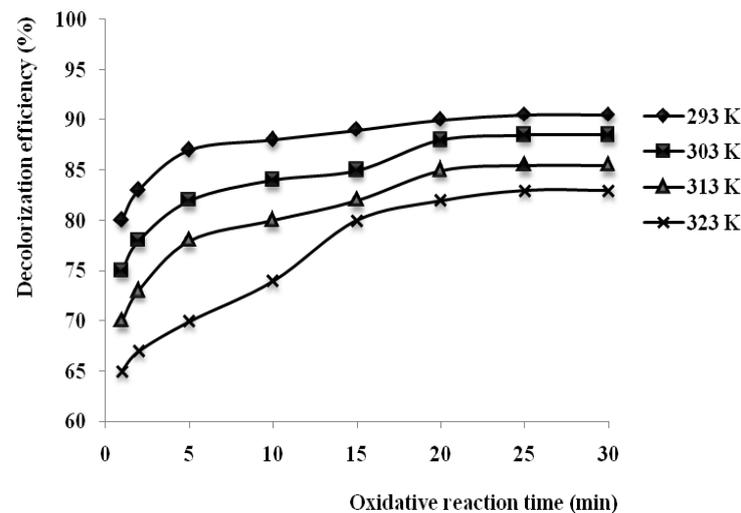


Figure 6: Effect of temperature on the decolorization efficiency of RY84. Concentration Fe^{2+} 25 mg/L, concentration H_2O_2 250 mg/L, pH 3.

Figure 6 depicts the effect of temperature on the removal of RY84 efficiency. It can be seen that the degradation efficiency of RY84 increased from 67.59 to 88.12% as a consequence of increasing the temperature from 293 to 323 K within the first 10 min of oxidation process.

This is because higher temperature increased the reaction rate between hydrogen peroxide and any form of ferrous/ferric iron, thus increasing the rate of generation of oxidizing species such as HO° radical or high-valence iron species [46]. After 20 min of oxidative reaction time, the increase of the decolorization efficiency of RY84 is not affected by the increasing of the temperature process. Therefore, the optimal temperature was chosen as 293 K to remove of RY84 by Fenton under our experimental conditions. To work at high working temperature has also some disadvantage that both of the investment and operational costs would be high to increase the wastewater's temperature [47]. To describe the thermodynamic behavior of the decolorization efficiency of RY84 using Fenton reagent, the thermodynamic parameters ΔG and E_a were calculated using equations (11) and (12). The calculated constants are illustrated in Table 3.

Table 3: Thermodynamic data for RY84 decolorization by Fenton reagent

T (K)	Log k_2	ΔG (kJ/mol)	E_a (kJ/mol)
293	1.881	- 4.574	16.78
303	2.246	- 5.648	
313	2.376	- 6.173	
323	2.546	- 6.826	

The oxidative reaction using Fenton reagent for the decolorization efficiency of RY84 was spontaneous in nature and the degree of spontaneity of the reaction increased by increasing temperature. All these suggestions on the basis of the obtention of a negative value for the Gibbs free energy. This result also supported the suggestion that the resulting reaction mechanism was energetically stable and that the rate of Fenton reaction primarily increased by increasing temperature as shown in Figure 7.

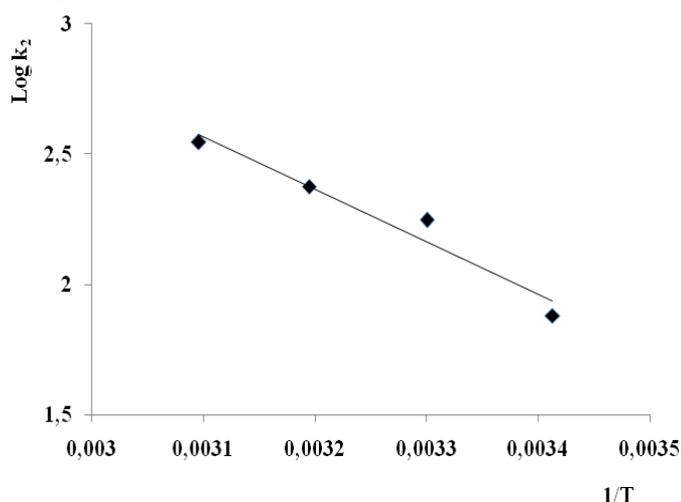


Figure 7: Plot of Log k_2 versus $1/T$

The calculation of activation energy value is determined by the slope of graph in Figure 7. This energy is equal to 16.78 kJ/mol, this low value may show that the oxidative reaction proceeds with low energy barrier.

4. Conclusion

This paper presented the results of a detailed study about kinetics and thermodynamics of the RY84 decolorization process by Fenton reagent. Optimal operation parameters for the Fe^{2+} and H_2O_2 concentration, reaction time, pH and temperature were 25 mg/L, 250 mg/L, 20 min, pH 3 and 293 K, respectively. Under these conditions, 85% decolorization efficiency of RY84 was achieved.

The kinetics of RY84 decolorization could be described by a model of pseudo-second-order. The reaction characteristic of oxidation process was evaluated as thermodynamically spontaneous under natural conditions.

5. References

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