



Decolorization of Acid Orange II Dye by Fenton Reagent: Optimization of Parameters and Kinetic Study

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Abstract Decolorization of Acid Orange II in aqueous solution by Fenton reagent in a batch reactor was investigated. The effects of different reaction parameters such as hydrogen peroxide concentration, ferrous ion concentration, pH of solution and initial dye concentration were investigated. In this study the experimental ranges for H₂O₂ dose was varied from 50 mg/l to 500 mg/l and Fe²⁺ ion was 10-40 mg/l. All experiments were carried out at temperature of 38 °C. The optimized dose of H₂O₂ and Fe²⁺ were 250 mg/l and 30 mg/l. The optimum pH condition was found to be 3.07. Under optimum conditions the initial 300 ppm dye solution was completely decolorized within 12 minutes at 38 °C and 87 % chemical oxygen demand (COD) was removed which proves that the dye was not only decolorized but mineralization also took place. A pseudo first-order reaction rate with respect to Acid Orange II concentration was found to fit the data collected adequately.

Keywords Advanced Oxidation Process (AOP), Fenton's process, Degradation of dye, Acid Orange II

1. Introduction

Wastewater generated from dyes and textile industries are highly colored and have a high concentration of chemical oxygen demand. Synthetic dyes are extensively used by different industries like textile, paper production, food technology and plastics. It is estimated that about 10,000 tons of dyes are produced all over the world per year [1]. Moreover azo dyes, characterized by one or more azo bonds (-N=N-), which are considered to be carcinogenic are widely used in textile industries and other industries to color solvents, inks, paints, varnishes, paper, rubber, foods, drugs and cosmetics [2]. The release of wastewater from textile and other industries into the water bodies causes serious environmental pollution by imparting intensive color and toxicity to aquatic environment. The colored wastewater can block sunlight penetration and oxygen dissolution which are essential for aquatic life. Therefore dye wastewater should be treated properly to meet the environmental discharge standards before discharging to the water bodies.

A wide range of methods like physico-chemical and biological processes have been used for removal of dyes from wastewater. Physico-chemical processes i.e. coagulation, precipitation, adsorption, ion exchange, filtration etc do not lead to organics degradation but rather to their transfer from one phase to another phase which should be regenerated and post treated with expensive operations [3]. Biological processes proceed at low rates and are often



inhibited by several substances, particularly those that are toxic to the microorganisms. Classical oxidation processes are also not able to remove dyes cost effectively. For these reasons, research efforts have been focused in developing alternative better cost effective oxidation processes that might be employed as a pre-treatment stage (to increase biodegradability by partial oxidation) or as final treatment, in some cases for simple color/dye removal. Recent progress in this field has led to the development of advanced oxidation processes (AOPs). Advanced Oxidation Process is based on the chemical oxidation technologies that use hydroxyl radical ($\bullet\text{OH}$) generated in situ. This hydroxyl radical is one of the strongest oxidants known after fluorine with oxidation potential of 2.06. (with reference $\text{Cl}_2 = 1.0$). [4]. The radical oxidizes the organic and/or inorganic contaminants to produce environmentally friendly fragments and eventually to CO_2 and H_2O if sufficient reagents and time are allowed. There are various ways of generating hydroxyl radical. Among them, homogeneous Fenton reaction ($\text{Fe}^{+2}/\text{H}_2\text{O}_2$) is one of the most important processes to generate hydroxyl radicals. [5] and Fenton's reagent has proved to be a promising treatment method for the effective decolorization and mineralization of dyes as well as for the destruction of a large number of hazardous and organic pollutants. Moreover this process is cost effective and simple, and takes place at normal room temperature and pressure.

Fenton's reagent consists of hydrogen peroxide (H_2O_2) and ferrous ions (Fe^{2+}). First hydroxyl radical is produced by the reaction of ferrous ion and H_2O_2 as given in equation (1).



According to equation (2), ferric ion is then reduced back to ferrous ion with another H_2O_2 molecule and produces a superoxide radical and a proton.



Regeneration of Fe^{+2} can also happen as given in equations (3) and (4).



If both $\bullet\text{OH}$ and Fe^{2+} ions are in excess, equation (5) will terminate the chain reaction. [6].



Literature shows that Fenton's reagent has been used for treatment of different types of wastewater like aromatic amines [7] petroleum refinery [8], phenol [9, 10, 11], pharmaceutical waste [12] p-nitro aniline [13] pesticides [14] and different types dye wastewater [15-21].

Present work deals with the degradation of Acid Orange II dye, using Fenton's reagent. Acid Orange II is an azo dye and has the chemical formula 4-[(2-hydroxy-1-naphthalenyl) - azo] - benzene sulphonic acid, monosodium salt; $\text{C}_{16}\text{H}_{11}\text{N}_2\text{O}_4\text{SNa}$. Structure of the dye is shown in figure 1. The dye is soluble in water with reddish yellow color. The dye finds its extensive use in dyeing and printing of wool, nylon, and silk. Other uses are in paper, leather, biological stain and indicator. Heavy metal salts of the dye are used for paper coating, transparent pigments in tin printing, and in molding powders.

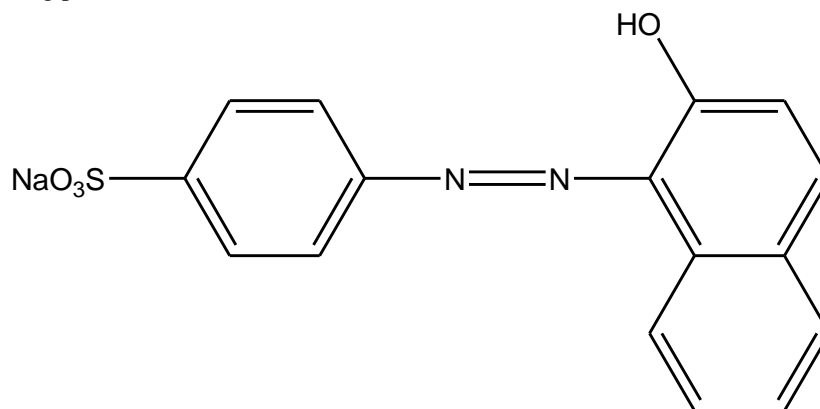


Figure 1: Acid Orange II dye



2. Materials and Methods

Acid Orange II dye was obtained from a dye company (Madhavdas Manilal & Co. “Devarsons House” Ahmadabad). All other chemicals like ferrous sulphate hexahydrate (98%), H_2O_2 (50% w/v) were taken from E Merk India Ltd.

The dye concentration was analyzed by colorimetric method using HACH Spectrophotometer (Model DR/2500). The dye solution was scanned for wavelength and λ_{max} was found to be 484 nm. A calibration curve was plotted for absorbance vs. concentration at 484 nm for known concentration of the dye samples i.e. 2, 4, 6 and 8 ppm (Fig. 2).

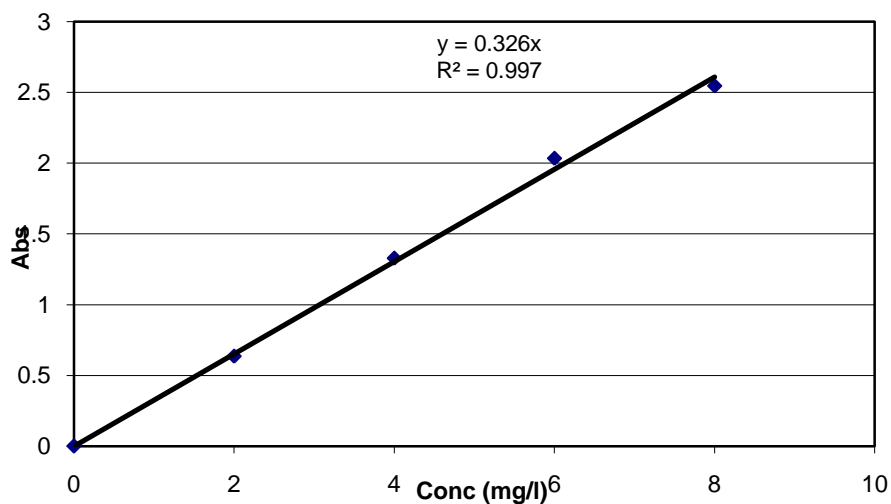


Figure 2: Calibration Curve for Acid Orange II dye

The strength of H_2O_2 used was around 50 % w/v. The strength was checked regularly using the permanganate titration method. [22] COD determination was done by oxidation with potassium dichromate in sulfuric acid and heating for 2 hrs to 150 °C [23]. Commercially available test kits from HACH (USA) Company was used for determination of COD.

Synthetic dye solution was prepared in distilled water. The ambient pH of the solution was 7.06. Experiments were carried out in a batch process. The initial dye concentration of all experiments were 300 ppm. For the study, 250 ml dye solution was taken in a bottle after adjusting the pH of the solution by acid or alkali. The required dose of Fe^{+2} and hydrogen peroxide was added. Samples were taken at regular time intervals for analysis.

3. Results and Discussions

Initial experiments were carried out with H_2O_2 alone without adding of Fe^{+2} catalyst. But it was found that rate of decolorization was very slow. For a H_2O_2 dose of 200 mg/l, it was found that after one hour dye decolorization took place only 5%. So it was necessary to add Fe^{+2} catalyst.

3.1. Effect of H_2O_2 dose on decolorization of Acid Orange II

The main chemical consumption of Fenton's process is hydrogen peroxide. To make the Fenton process cost effective compared to other processes, it is necessary to optimize the hydrogen peroxide dose. The effect of H_2O_2 concentration on Fenton's treatment was investigated in a H_2O_2 concentration range between 50 to 500 mg/l, while keeping the ferrous ion dose, pH, and temperature constant at 20 mg/l, 4.2, and 38°C respectively. The results are shown in Fig. 3. The results indicated that the decolorization of dye was significantly influenced by the H_2O_2 dose of the solution, and the best decolorization efficiency was obtained at H_2O_2 dose of 250 mg/l

It is seen that with increasing hydrogen peroxide concentration from 50 mg/l to 250 mg/l, the dye decolorization rate increases but further increase of H_2O_2 concentration, the rate decreases. The basic reaction mechanism of Fenton's process is first formation of hydroxyl radicals and this hydroxyl radical is responsible for further oxidation of organic molecules. So initially with increasing hydrogen peroxide concentration, more hydroxyl radicals are



produced by reaction (1) and the efficiency of decolorization process increases. But after reaching a certain value of H_2O_2 concentration the decolorization of dye decreases because H_2O_2 in excess acts as hydroxyl radical scavenger effect. The hydroxyl radicals could be consumed by H_2O_2 and result in the generation of less reactive HO_2° radical which again can react with hydroxyl radical and forms water and oxygen, as per equation (6) and (7) [24].



Moreover some competitive reaction could take place where two hydroxyl radicals can recombine as given in equation (8) and (9) [25].



The optimum concentration of hydrogen peroxide has to be determined experimentally for every wastewater before designing any large scale application.

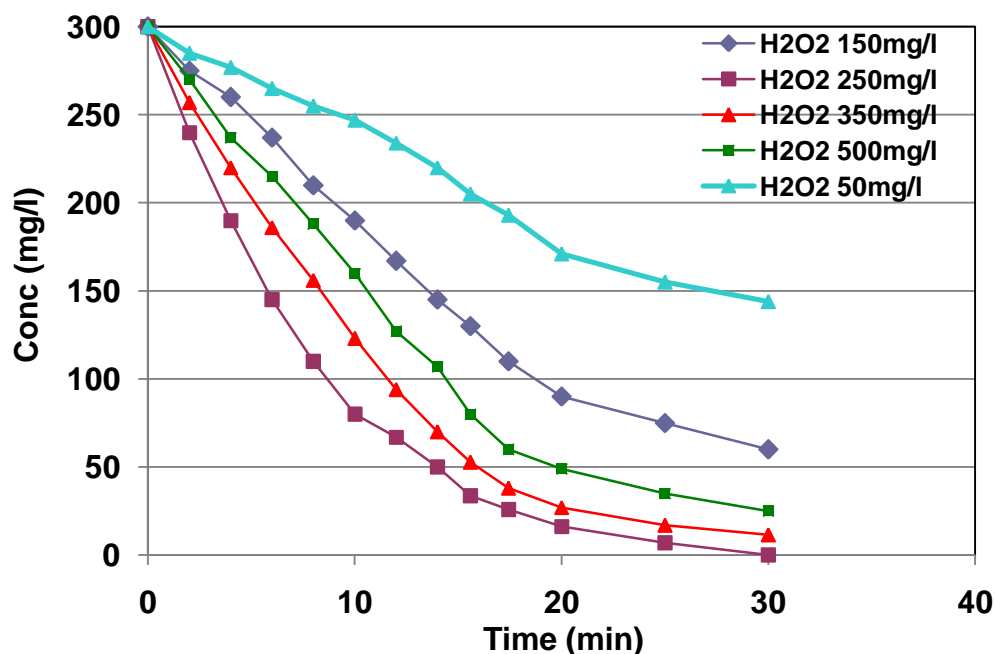


Figure 3: Effect of H_2O_2 conc on decoloration of Acid Orange II by Fenton reagent (Fe^{2+} conc.=20 mg/l; $\text{pH}=4.2$)

3.2. Effect of Fe^{2+} concentration on decolorization of dye

The amount of ferrous ion is also a very important parameter in Fenton oxidation process because the ferrous ion is responsible for the generation of hydroxyl radicals with reaction of H_2O_2 . The effect of Fe^{2+} ion concentration on the decolorization of dye has been studied in the Fe^{2+} concentration range of 10-40 mg/l while keeping other parameters constant (H_2O_2 dose = 250 mg/l, $\text{pH} = 4.2$). The results are shown in Fig. 4. The results indicate that the extent of decolorization increased with increase in initial Fe^{2+} concentration from 10 mg/l to 30 mg/l. The concentrations higher than 30 mg/l resulted in decrease in decolorization efficiency of dye under study. Initially with increasing ferrous ion concentration decolorization increases due to more hydroxyl radical produced by reaction of ferrous ion with hydrogen peroxide. But after a certain concentration of ferrous ion, the efficiency decreases because hydroxyl radicals may be scavenged by the reaction with the hydrogen peroxide or with another Fe^{2+} molecule as given by equation (10) and (11) [26,27].



In the present study the optimum concentration of ferrous ion concentration was found to be 30 mg/l and the H_2O_2 and Fe^{+2} ratio (w/w) was = 0.12. Chang et al (2008) [28] have shown Fe^{+2} and H_2O_2 ratio equals to 0.147 seems to be the most effective when the hydroxyl radicals generation is considered. Pegah et al (2013) [29] have shown that the dye, H_2O_2 and FeSO_4 mass ratio (w/w/w) of 1.2:0.16 was required for 97% degradation of reactive black 5 dye. Similarly Lodha and Chaudhari (2007) [30] have studied decolorization of RB5, RB13, and AO7 and found that the dye: H_2O_2 : Fe^{2+} mass ratio (w/w/w) was 1:1:0.3 for 97, 98 and 97% decolorization respectively.

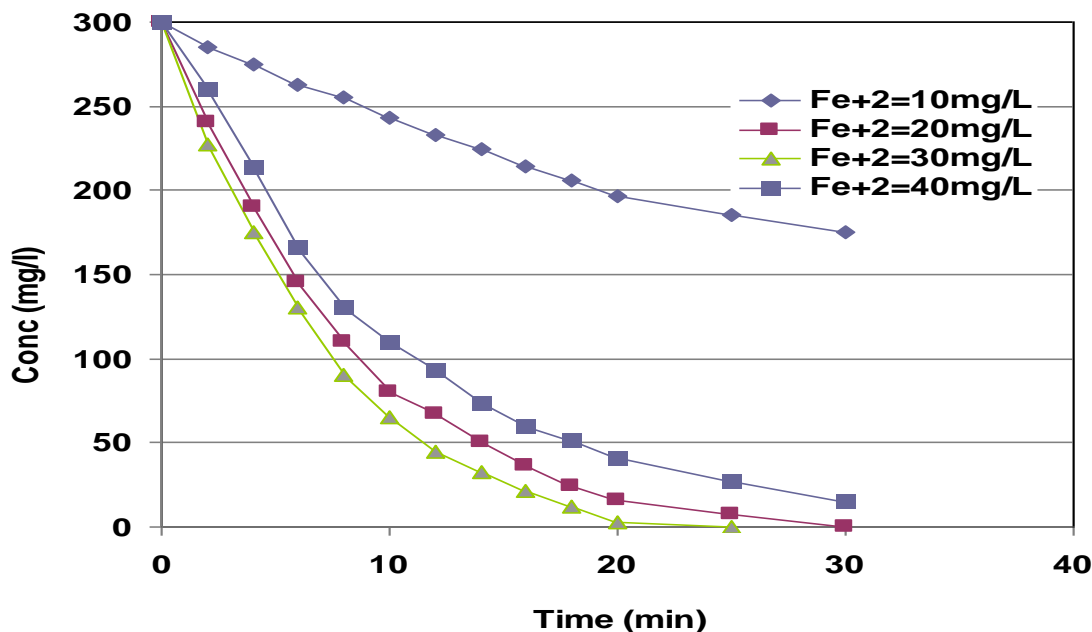


Fig.4: Effect of Fe^{+2} concentration on decolorization of Acid Orange II by Fenton Reagent. ($\text{H}_2\text{O}_2=250$ mg/l, $\text{pH}=4.2$)

Figure 4: Effect of Fe^{+2} concentration on decolorization of Acid Orange II by Fenton Reagent (H_2O_2 250 mg/ml, $\text{pH}=4.2$)

3.3. Effect of initial pH of the solution on decolorization of Acid Orange II by Fenton's reagent

The effect of initial pH of the solution on dye decolorization by the Fenton oxidation process was studied in the pH range of 2-7.5. The initial dye concentration was 300 ppm and the dose of H_2O_2 and Fe^{2+} ion were 250 mg/l and 25 mg/l respectively. The results are shown in Fig. 5. The results show that pH has a very important role on decolorization of the dye solution. The dye decolorization rate is better in acidic medium than basic medium and the best decolorization efficiency was observed at pH of 3.07. If the pH is raised beyond 3.07 or lowered than it, the efficiency of dye decolorization decreases.

The lower efficiency at high pH values may be due to the precipitation of $\text{Fe}(\text{OH})_3$. At high pH, ferrous ions are unstable and it will easily form ferric ions which have a tendency to produce a colloidal ferric hydroxo complex. The generation of $\text{HO}\cdot$ gets slower because of the formation of this species. In the latter form, the iron catalytically decomposes the H_2O_2 into oxygen and water, without forming hydroxyl radicals [31]. Another reason for lower efficiency at higher pH is due to self decomposition of H_2O_2 . The base catalyzed decomposition involves the HO_2^- anion, the conjugated base of H_2O_2 reacts with a non dissociated molecule of H_2O_2 according to equation (12) [32].



On the other hand when pH condition below 3, the $\cdot\text{OH}$ can be consumed via scavenging hydroxyl radicals with H^+ ions as can be seen in equation (13) [27, 33].



According to equation (14), hydrogen peroxide can capture a proton to form an oxonium ion (H_3O_2^+) and H_3O_2^+ can make hydrogen peroxide to be electrophilic and thus reduce the reactivity of the reaction between hydrogen peroxide and ferrous ion [34, 35].

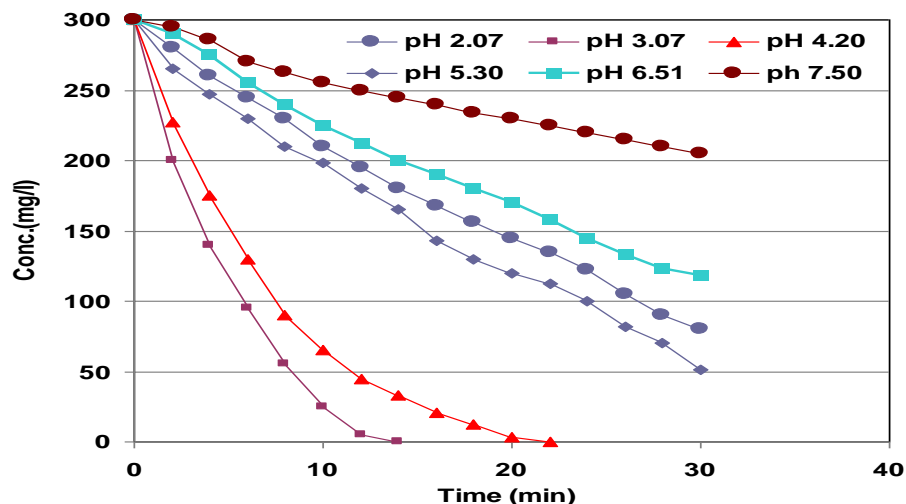


Figure 5: Effect of pH on discoloration of Acid Orange II by Fenton's reagent (Fe^{+2} conc.= 30 mg/l, H_2O_2 conc= 250mg/l)

3.4. Effect of initial concentration on degradation of Acid Orange II

The effect of initial dye concentration on decolorization was studied at pH of 3.07 with initial H_2O_2 dose of 250 mg/l and Fe^{2+} dose of 30 mg/l and the results are shown in Fig. 6. The results show that with increasing initial dye concentration from 50 to 300 ppm the time for decolorization increases. This is because when the number of dye molecule increases, more hydroxyl radicals are required to decolorize it. But the hydroxyl radical concentration remains same for the particular reaction medium and thus the decolorization time increases. This result is well agreement with literature [36, 37].

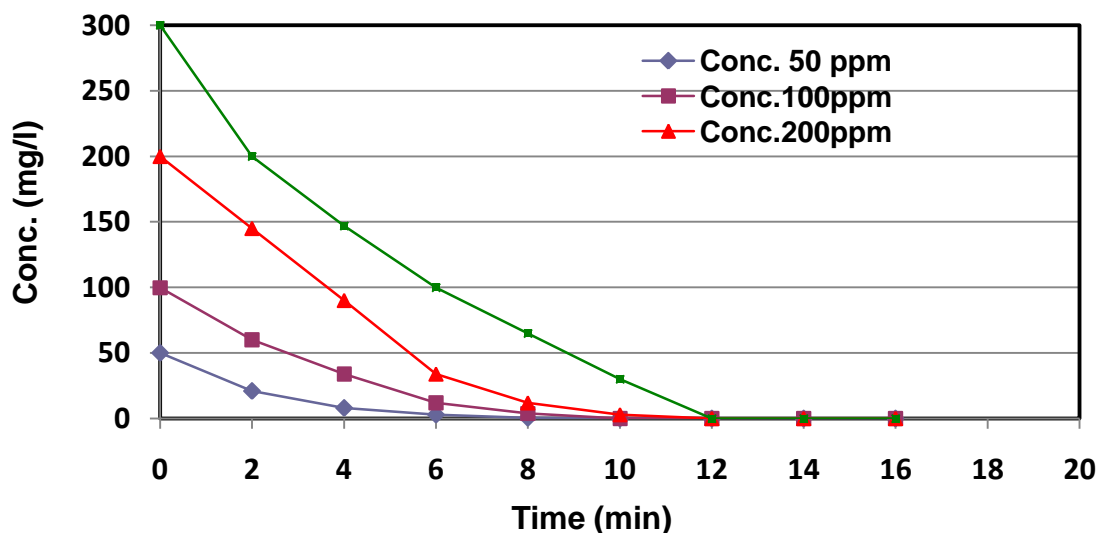


Figure 6: Effect of initial dye concentration on discoloration of Acid Orange II by Fenton's reagent (Fe^{+2} conc.= 25 mg/l, H_2O_2 conc= 250mg/l, pH=3.07)



3.5 Kinetic Study

The degradation of dye can be expressed by equation (15).

$$(-r_{\text{dye}}) = k C_{\text{OH}} C_{\text{dye}} \quad (\text{Eq. 15})$$

Where k is the rate constant, C_{OH} is the concentration of hydroxyl radical and C_{dye} being the concentration of the dye. By assuming that the concentration of hydroxyl radicals are constant, the kinetics of dye degradation will follow pseudo first order reaction mechanism and the above equation can be written as given in equation (16).

$$(-r_{\text{dye}}) = k' C_{\text{dye}} \quad (\text{Eq. 16})$$

where k' is the overall rate constant. Integrating and rearranging equation (16), yields the typical pseudo first order equation as given in equation (17).

$$-\ln(C_{\text{dye } t} / C_{\text{dye } 0}) = k' t \quad (\text{Eq. 17})$$

A plot of $-\ln(C_{\text{dye } t} / C_{\text{dye } 0})$ against time shown in fig 7 shows a linear relationship. Good correlation coefficients ($r^2 = 0.98$) were obtained in our systems. The value of the rate constants k' (min^{-1}) were obtained from the slope of the equation and presented in Table 1. From Table 1 it is seen that highest rate constant value (0.921 min^{-1}) was obtained at pH 3.07.

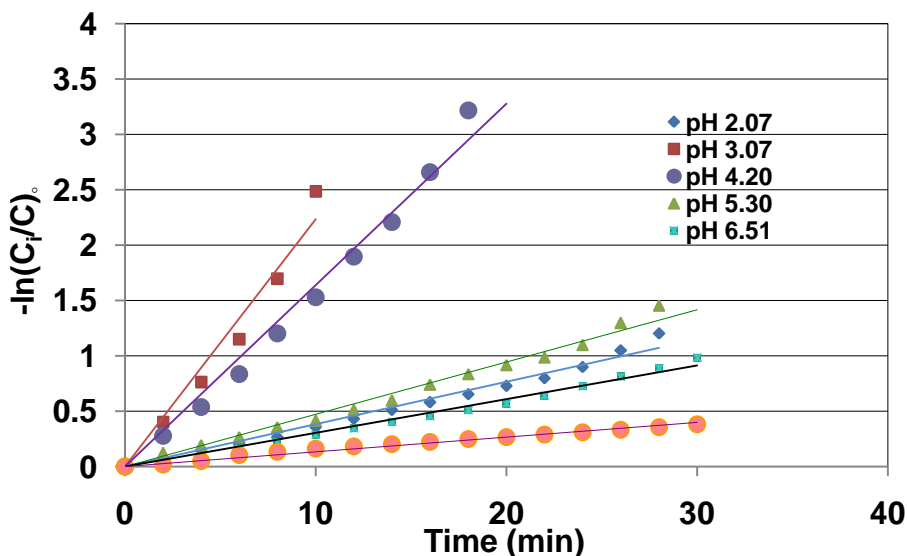


Figure 7: Plot of $-\ln(C_t/C_0)$ with time for decoloration of Acid Orange II at different pH

Table 1: Pseudo first order rate constant values at different pH

pH	$K'(\text{min}^{-1})$	R^2
2.07	0.038	0.983
3.07	0.223	0.969
4.20	0.163	0.985
5.30	0.047	0.985
6.51	0.030	0.988
7.50	0.013	0.980

Similar pseudo first order kinetic behavior of dye degradation was also observed by other investigators [38-43].

Mineralization of dye

Chemical Oxygen demand gives an average measure of the oxidation state of the organic byproducts generated during the degradation of the dye. Extent of mineralization of the acid orange II dye by Fenton process was evaluated by measuring the COD at different time interval with initial dye concentration of 300 ppm using optimized conditions like H_2O_2 dose = 250 mg/l, Fe^{+2} = 30 mg/l, pH and temperature were kept at 3.07 and 38°C . The COD removal efficiency was determined by equation (18).

$$\text{COD removal efficiency} = (\text{COD}_i - \text{COD}_t) / (\text{COD}_i) \quad (\text{Eq. 18})$$

Where, COD_0 and COD_t are the COD at time 0 and t respectively.

From the results presented in Figure 8, it can be seen that initially COD removal rate was low. Because initially when the solution is coloured, dye molecules are first decomposed to lower molecular weight compounds by hydroxyl radicals and the resulting intermediates still contribute to the COD of solution. With increasing time, the COD decreased sharply and after 30 minutes around 87% COD is removed. So it is seen that after decolorization some more retention time is required for complete mineralization of the solution.

It has been reported by Kusic (2007) that initially mineralization is slow than decolorization process [45]. Kuo (1992) reported that approximately 90% COD removal in 30 min [45].

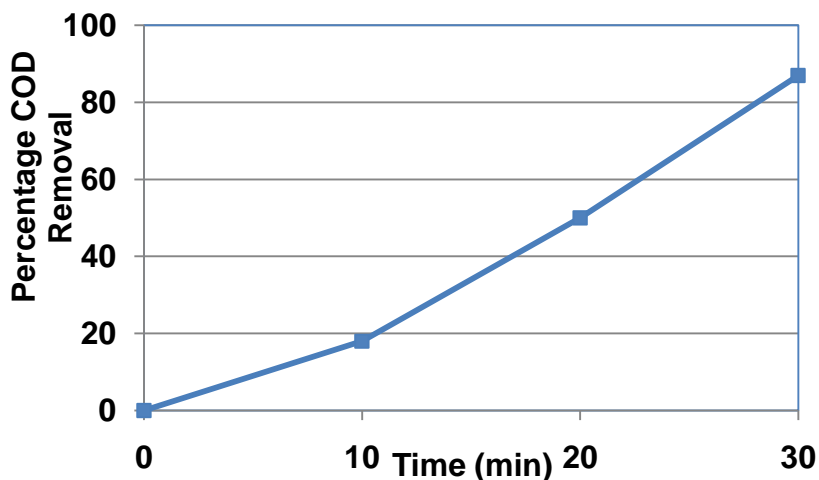


Figure 8: Percentage COD removal with time for degradation of Acid Orange II by Fenton reagent

Malik and Saha (2003) observed that at the optimal ratio of $[Fe^{2+}]$: $[H_2O_2]$: [dye] (initial concentration ratio) and at 30°C with pH 3, 70% COD removal can be achieved in 60 min [46].

4. Conclusion

The degradation of acid orange II was carried out using Fenton's reagent. The effect of various parameters like hydrogen peroxide dose, ferrous ion concentration, pH of the solution and the initial dye concentration were investigated. All experiments were carried out at room temperature of about 38°C with initial dye concentration of 300 ppm. The optimized operating conditions for decolorization of Acid Orange II included: 250 mg/l of H_2O_2 , 50 mg/l of ferrous ion concentration, pH of the solution 3.07 for a dye concentration of 300 ppm. At these conditions, pseudo first order degradation rate constants were obtained from batch experimental data. The results obtained shows that the Fenton's process leads to complete decolorization optimized conditions with initial dye concentration of 300 ppm at 12 minutes. Also the results shows that Fenton's process not only decolorize the dye solution but it can also oxidize the by-products leading to mineralization and 87 % COD reduction was achieved within 30 minutes.

Funding Acknowledgement

We would like to acknowledge the Centre for Fire, Explosive & Environment Safety, Delhi, India for providing funds and equipment to complete this work.

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