Asian Journal of Chemistry; Vol. 33, No. 1 (2021), 113-119

ASIAN JOURNAL OF CHEMISTRY

https://doi.org/10.14233/ajchem.2021.22958

Optimization of Fenton's Process for the Treatment of Cotton Textile Wastewater using Response Surface Methodology

SANGEETHA PARASHURAM MULGUND*, P.B. KALBURGI and P.G. RAKARADDI

Department of Civil Engineering, Basaveshwar Engineering College, Bagalkot-587102, India

*Corresponding author: E-mail: sangita5679@gmail.com

Received: 20 August 2020; *Accepted*: 30 September 2020; *Published online*: 10 December 2020; AJC-20186

Degradation of cotton textile wastewater using Fenton's process was investigated. The effect of individual and interactive operating parameters on the response was analyzed using central composite design (CCD), a commonly used form of response surface methodology (RSM). The operating parameters selected were pH, dosage of hydrogen peroxide, dosage of iron and the responses (dependent parameters) were chemical oxygen demand (COD) and colour. The model derived correlation coefficients R^2 and R^2_{adj} for COD were 0.982 and 0.966, respectively and the values were almost similar for colour also. The optimum values for various operating parameters namely pH, H_2O_2 and Fe^{2+} dosage were found to be 3.33, 60.57 and 1.56 mM, respectively for electrolysis time of 60 min. By performing the experiments with these values of operating parameters, the COD and colour removal efficiencies were found to be 83.5% and 98.1%, respectively.

Keywords: Cotton textile wastewater, Fenton's process, Response surface methodology, Central composite design, Hydrogen peroxide, Iron.

INTRODUCTION

Textile mills consume large quantity of water (according to USEPA a unit producing 20,000 lb/day of fabric consume 36000 L of water) [1]. Since textile waste consists of different kind of dyes, heavy metals, pigments, recalcitrant dye molecules and organic compounds, direct discharge of the textile effluent into the water body results in intense pollution in terms of colour, chemical oxygen demand (COD), pH, toxicity, suspended solids and low biological oxygen demand (BOD) [2]. Large amount of wastewater is generated in dyeing and finishing operations from textile industry (approximately 21 to 377 m^3 of water per ton of textile produced) [3]. The organic matter present in the waste causes the diminution of dissolved oxygen in river water bodies which has an adverse effect in aquatic ecosystem [4]. The biological degradation processes are usually ineffective in the degradation of complex organic recalcitrant compounds [5]. The other conventional physico-chemical treatment methods that includes coagulation/precipitation, adsorption are not generally effective for the removal of the water soluble dyes from the effluents. These processes also have some major drawbacks in the treatment as well as disposal of the chemical sludge generated due to the phase transfer of pollutants [6]. Hence the textiles industries, due to the stringent pollution control regulations, have been forced to adopt the new and sustainable technologies for the treatment of their waste. In this connection, the advanced oxidation processes (AOPs) like ozonation, photo-Fenton's process, photocatalytic reaction, Fenton's process, which involve the generation of hydroxyl radicals ($^{\bullet}OH$) for faster oxidation process, present effective means for degradation of toxic pollutants [7-10]. Hydroxyl radicals generated in the AOPs are highly reactive and non-selective with almost all the biologically refractory chemical compounds [11]. Further, after the removal of sludge formed during the Fenton's process and followed by biological treatment, discharge can be recycled or reused [12]. Thus, due to their higher efficiency in removing the colour and COD and simple operation, AOPs are gaining more popularities from the industries and researchers [13]. The present work involves studies on the degradation of cotton textile effluent by Fenton oxidation process. In this process iron is employed as a catalyst for carrying out oxidation with hydrogen peroxide in acidic condition [14]. For the generation of hydroxyl radicals, Fenton's process is more economic and also it is easy to operate and maintain as compared to other advanced oxidation process (AOPs).

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In case of the effluents with high content of suspended and total dissolved solids (SS & TDS), Fenton's process is more advantageous over photo oxidation processes as the ability of photon penetration across the depth of the solution is limited [15].

The hydrogen peroxide gets decomposed by ferrous ions under acidic pH conditions [16]. The reactions involved in the formation of hydroxyl radicals ($^{\bullet}OH$) and ferrous ions Fe^{2+} are as given below [17]:

$$Fe^{2+} + H_2O_2 \longrightarrow Fe^{3+} + OH^- + OH^{\bullet} \quad (1)$$

$$Fe^{3+} + H_2O_2 \longrightarrow Fe^{2+} H^+ + OOH^{\bullet} \quad (2)$$

$$Fe^{3+} + OOH^{\bullet} \longrightarrow Fe^{2+} + H^+ + O_2 \quad (3)$$

$$OH^{\bullet} + H_2O_2 \longrightarrow OOH^{\bullet} + H_2O \quad (4)$$

$$OH^{\bullet} + Fe^{2+} \longrightarrow + OH^- \quad (5)$$

$$OH^{\bullet} + OH^{\bullet} \longrightarrow H_2O_2 \quad (6)$$

$$OH^{\bullet} + OOH^{\bullet} \longrightarrow O_2 + H_2O \quad (7)$$

However, Fenton process is associated with some drawbacks also, for example, (i) the effluent needs to be acidified as waste solution is alkaline in most of the cases and (ii) The sludge generated during the process needs to be treated properly before final disposal [18].

Response surface methodology (RSM) is an effective mathematical and statistical tool for optimization of operating parameters in industrial processes. This technique can be used for the design of experiments and assessment of individual and interactive effects of various operating parameters on process output or response [19]. In RSM the polynomial equation is generated to find the optimum values of independent operating parameters. This technique is faster for generating research results as compared to the time consuming one variable at a time (OVAT) process [20].

In current work, experiments are designed by central composite design (CCD), a commonly used form of RSM. The individual and interactive effect of operating parameters [21] (pH, dosage of H_2O_2 and Fe^{2+}) on the response (removal efficiency of COD and colour) has been assessed. The model predicted parameters were experimentally validated.

EXPERIMENTAL

Cotton textile wastewater: The wastewater of cotton textile was collected from a cotton textile industry plant located in Northern part of Karnataka and was preserved in cold storage at temperature of 4 °C within 3 to 4 h of collection. This textile wastewater majorly consists of reactive red HE3B and blue HEGN dyes used for cotton dyeing. The collected textile wastewater has a pH = 10.12, COD = 1165 mg/L, TOC = 436.1 mg/L, TDS = 3323 mg/L and maximum absorption wavelength in the visible range is 306 nm.

All the chemicals and reagents including hydrogen peroxide (30% w/w) and ferrous sulphate heptahydrate ($FeSO_4 \cdot 7H_2O$) used in the present work were of analytical grade procured from Fisher Scientific, India. Sulfuric acid (98-99%) and sodium hydroxide were used for the adjustment of pH of the solution. COD analysis was carried out using mercuric sulphate, potassium iodide, potassium dichromate and FAS (ferrous ammonium sulfate) solution. The ultra-pure water was used to prepare the solutions.

Fenton's experiments: Fenton's experiments were carried out at room temperature in Erlenmeyer flask. Sample size selected was 500 mL kept with continuous stirring at 100 rpm. As per the design of experiments generated by CCD, 20 experiments were performed for various pH values ranging from 3 to 4, dosage of hydrogen peroxide and Ferrous ions ranging from 55.54 to 77.76 mM and 0.89 to 2.32 mM, respectively. Experiments were performed for a designed reaction time of 60 min with settling time of 30 min. The final pH of the treated sample was adjusted to 7-10 using 4 N NaOH solution to stop the Fenton's reaction. The sample after each designed electrolysis time was then taken into a cylindrical glass and filtered through filter paper of 0.45 µ. Then the final total organic carbon (TOC) and absorption value was analyzed. The interference of H_2O_2 in COD was corrected as per the method discussed in the literature [22] and the final COD removal efficiency was calculated.

Analytical procedure: The chemical oxygen demand (COD) and total organic carbon (TOC) were analyzed as per the standard methods (APHA). The COD was determined by open reflux method and TOC was analyzed using the Shimadzu make TOC analyser. The λ_{max} value of real wastewater at wavelength of 306 nm was measured using UV spectrophotometer (Hach D4000). Orion pH meter was used for measuring the pH values. Hydrogen peroxide remaining in the solution after the treatment was analyzed using iodometric titration.

Experimental design: For the design of the experiments, a statistical and mathematical tool called central composite design is used in the present study. It is a widely used technique for the design of experiments and modelling of the experimental results using RSM [19]. By using this technique, the interactive effects of various operating parameters on the response can be effectively analyzed. The main purpose of using RSM is to optimize several variables of given process to get the optimized output or response [23]. It efficiently predicts a quadratic model equation at predefined level of the independent operating variables to obtain the optimum operating parameters [24]. The optimization of the operating parameters is faster and accurate in RSM than the time consuming OVAT process. The three-level design of CCD has axial or star points which are represented as $+\alpha$ and $-\alpha$. Due to these two values, the number of levels will be increased from three to five. This is incorporated to impart more flexibility and accuracy to the design of experiments and the model output. Thus, a total number 20 experiments were generated for the CCD designed at three factorial and five levels ($-\alpha$, -1, 0, +1, $+\alpha$).

The three independent operating parameters namely, initial pH, H_2O_2 dosage and Fe^{2+} dosage were employed to investigate the colour and COD removal efficiency (dependent parameter or response). The quadratic equation giving functional relationship between the response (Y) and set of independent operating variables (X_i) [25] generated by the model is given in the form of following equation:

$$Y = \beta_0 + \beta_1X_1 + \beta_2X_2 + \beta_3X_3 + \beta_{12}X_1X_2 + \beta_{13}X_1X_3 + \beta_{23}X_2X_3 + \beta_{11}X_1^2 + \beta_{22}X_2^2 + \beta_{33}X_3^2 \quad (8)$$

where Y is the predicted response or output, X_i represents the independent operating parameters and β_i values represents the regression coefficients [5]. Design expert 11.0 software was used for performing the CCD analysis. For process optimization, the desired target of dependent variable (responses) and independent variables (Operating parameters) should be obtained first. In the present study, "maximum" degradation of wastewater in terms of colour and COD removal efficiencies was chosen as response whereas initial pH, H_2O_2 dosage and Fe^{2+} dosage were "within the range" [21].

For obtaining the functional relationship between response and the independent operating variables, analysis of variance (ANOVA) was performed [25]. Best fit for the second order polynomial model was analyzed using the regression coefficient (R^2), probability value (P) and Fisher's test (F test) at its confidence level of 95%. The response surface plot (3D plots) was employed to obtain the most sensitive relations between operating variables and response.

The dosage of hydrogen peroxide was fixed as per the stoichiometric ratio for degradation of COD. The theoretical stoichiometric ratio of hydrogen peroxide to COD is 2.125 [26]. Hence, considering the COD of real wastewater as a reference, hydrogen peroxide dosage was fixed. The five levels of independent operating variables are presented in Table-1.

TABLE-1
FIVE LEVELS OF INDEPENDENT OPERATING VARIABLES

Code	Variables	–α	+1	0	+1	+α
X_1	pH	2.65	3.00	3.50	4.00	4.34
X_2	H_2O_2 (mM)	47.96	55.54	66.65	77.76	85.33
X_3	Fe^{2+} (mM)	0.40	0.89	1.60	2.32	2.80

RESULTS AND DISCUSSION

Response surface methodology design and statistical analysis: Total 20 numbers of experimental runs with different operating variables as generated by CCD are shown in Table-2.

The second order polynomial equation (quadratic equation) as generated by Design Expert 11.0 software for the percentage removal of COD and colour is given by eqn. 9 and 10.

$$\%\ \text{COD removal}\ (Y_1) = +83.88 - 0.6384*\text{pH} - 2.15*H_2O_2 + 0.0160*Fe^{2+} + 0.6725*\text{pH}*H_2O_2 + 3.11*\text{pH}*Fe^{2+} - 0.9200*H_2O_2*Fe^{2+} - 1.78*(\text{pH})^2 - 2.12*(H_2O_2)^2 - 4.60*(Fe^{2+})^2 \quad (9)$$

$$\%\ \text{Colour removal}\ (Y_2) = +97.98 - 0.6842*\text{pH} - 3.09*H_2O_2 + 0.8226*Fe^{2+} + 0.7475*\text{pH}*H_2O_2 + 2.54*\text{pH}*Fe^{2+} - 0.9200*H_2O_2*Fe^{2+} - 2.72*(\text{pH})^2 - 1.90*(H_2O_2)^2 - 5.86*(Fe^{2+})^2 \quad (10)$$

The values 83.88 and 97.98 shown in eqns. 9 and 10, respectively are the intercept values for COD (Y_1) and colour (Y_2). They imply a positive impact on model. The values of coefficients for the variables such as X_3, X_1*X_2 and X_1*X_3 has positive effect on percent removal of COD and colour, respectively.

The results of ANOVA found for COD and colour removal by Fenton's process are presented in Table-3. The mean square values were obtained by dividing the sum of squares of each source by their respective degrees of freedom (DF). The significance of the statistics was analyzed at a confidence level of 95 % (α = 0.05) *i.e.*, the P-value less than 0.05 represents a significant model.

The goodness of fit of the model was measured using the regression coefficient (R^2) and the adjusted regression coeffi-

TABLE-2
EXPERIMENTAL DESIGN AND RESULTS WITH PREDICTED RESPONSES OF COD AND COLOUR PERCENT REMOVAL AS PER CCD DESIGN

Run	Independent variable			Response			
	X_1	X_2	X_3	Experimental		Predicted	
	pH	H_2O_2 (mM)	Fe^{2+} (mM)	COD	Colour	COD	Colour
1	3.50	66.65	2.81	72.03	83.45	70.90	82.78
2	4.00	55.54	2.32	79.12	91.57	80.27	91.66
3	4.34	66.65	1.61	77.86	88.56	77.77	89.14
4	3.50	66.65	1.61	83.65	98.58	83.88	97.98
5	3.50	66.65	1.61	84.88	96.89	83.88	97.98
6	4.00	77.76	2.32	75.56	88.54	75.47	88.68
7	4.00	55.54	0.89	72.86	88.12	72.17	86.65
8	3.50	47.97	1.61	81.91	96.55	81.49	97.80
9	3.50	66.65	1.61	84.12	98.50	83.88	97.98
10	3.00	77.76	0.89	78.55	86.15	77.21	85.20
11	3.50	85.33	1.61	73.58	87.45	74.27	87.40
12	3.00	77.76	2.32	68.69	82.86	69.18	83.48
13	3.50	66.65	1.61	83.55	98.33	83.88	97.98
14	3.00	55.54	0.89	81.12	95.58	81.02	94.59
15	2.66	66.65	1.61	79.56	90.81	79.92	91.44
16	4.00	77.76	0.89	71.44	80.68	71.06	80.26
17	3.50	66.65	1.61	83.45	98.65	83.88	97.98
18	3.50	66.65	1.61	83.65	97.11	83.88	97.98
19	3.00	55.54	2.32	76.48	89.88	76.67	89.45
20	3.50	66.65	0.40	69.45	78.15	70.85	80.02

TABLE-3
ANOVA RESULTS FOR PERCENT OF COD AND COLOR REMOVAL BY FENTON'S PROCESS

	Source	Sum of squares	Degree of freedom	Mean square	F-value	P-value
COD removal	Model	521.25	9	57.92		
	Residual	9.48	10	0.95		
	Lack of Fit	8.02	5	1.6	61.09	0.001
	Pure Error	1.46	5	0.29		
	R^2	0.982	R^2_{adj}	0.966		
Colour removal	Model	787.86	9	87.54		
	Residual	14.19	10	1.42		
	Lack of Fit	11.05	5	2.21	61.70	0.001
	Pure Error	3.14	5	0.63		
	R^2	0.982	R^2_{adj}	0.966		

cient (R^2_{adj}). The R^2 value of the model is 0.982 whereas the R^2_{adj} value is 0.966 for percent removal of COD and the values obtained for colour were almost in the same range. These values indicates a good fit of the model.

Response surface plots for Fenton's process: The three-dimensional (3D) response surface plots obtained from deign expert 11.0 software were used to analyse the interactive effect of process variables.

Interactive effect of H_2O_2 dosage and pH on removal of COD: Fig. 1 displays 3D response surface plots which represents the interactive effects of H_2O_2 dosage and initial pH for the reduction of COD. From the figure it can be observed that with the increase in pH, COD removal also increases. At a pH of 3.5 a maximum of 84 % COD removal occurs with an oxidizing dosage of 66.65 mM in the presence of 1.61 mM catalyst dosage with a reaction time of 60 min. With the increase in the pH value above 3.5-3.6, a decrease in the COD reduction was observed which may be attributed to the breakdown of H_2O_2 to O_2 and H_2O. From the previous studies, the maximum reduction was observed at a pH value of 3.0-3.5. On the other hand, the same Fig. 1 indicates the removal of COD slightly decreased with the increase in the dosage of H_2O_2. This negative effect of excess dosage of H_2O_2 may be attributed to the scavenging effect by free radical, the formation which is shown in the following reaction [27]:

$$H_2O_2 + OH^{\bullet} \longrightarrow H_2O + HO_2^{\bullet} \quad k = 2.7 \times 10^7 \ M^{-1} \ s^{-1} \quad (11)$$

Interactive effect of H_2O_2 dosage and pH on colour removal: The interactive effect H_2O_2 dosage and pH on colour removal for catalyst dosage of 1.605 mM and reaction time of 60 min is shown in Fig. 2. High colour removal efficiency (more than 80%) was observed at all the working conditions. Maximum removal of colour of 99% was observed at pH 3.5 and H_2O_2 dosage of 66.65 mM. Hence it can be said that the optimum range of H_2O_2 dosage for the maximum reduction of colour is 60 to 63 mM.

Interactive effect of H_2O_2 and Fe^{2+} dosage on removal of COD: In Fenton's process Fe^{2+} acts as a catalyst and it is a key parameter for the COD reduction. As shown in Fig. 3 for the interactive effect of H_2O_2 and Fe^{2+} dosages at pH 3.5 and reaction time of 60 min on removal of COD, the increase in the iron (Fe^{2+}) dosage increases the removal of COD. Even though the dosage of H_2O_2 plays an important role in the Fenton's process, it is to a lower extent as compared to Fe^{2+}. It

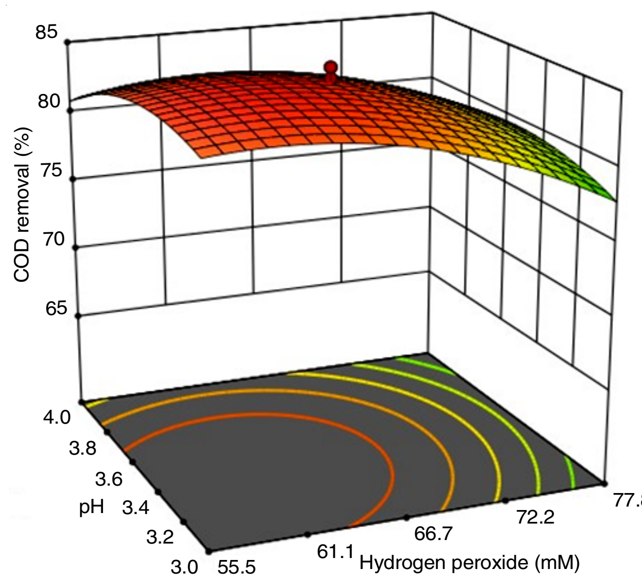

Fig. 1. Response surface plots percent COD removal from the wastewater, as a function of pH and hydrogen peroxide dosage (Fe^{2+} dosage = 1.605 mM and reaction time of 60 min)

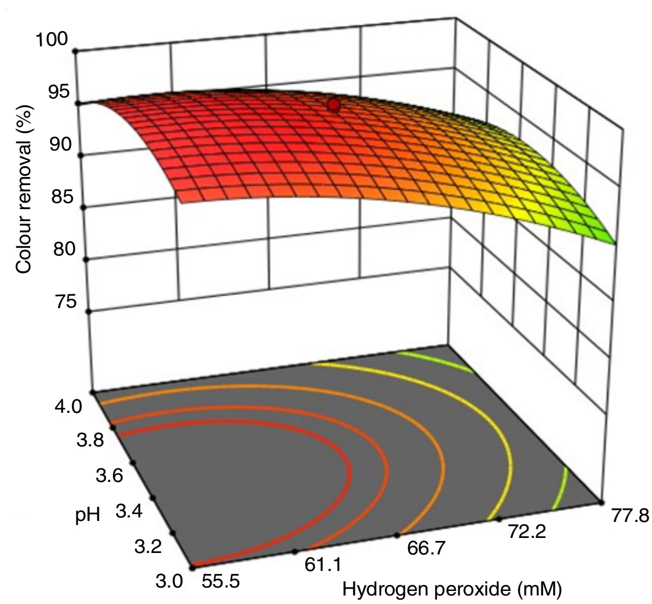

Fig. 2. Response surface plots for percent colour removal as a function of pH and hydrogen peroxide dosage (Fe^{2+} dosage = 1.605 mM and reaction time of 60 min)

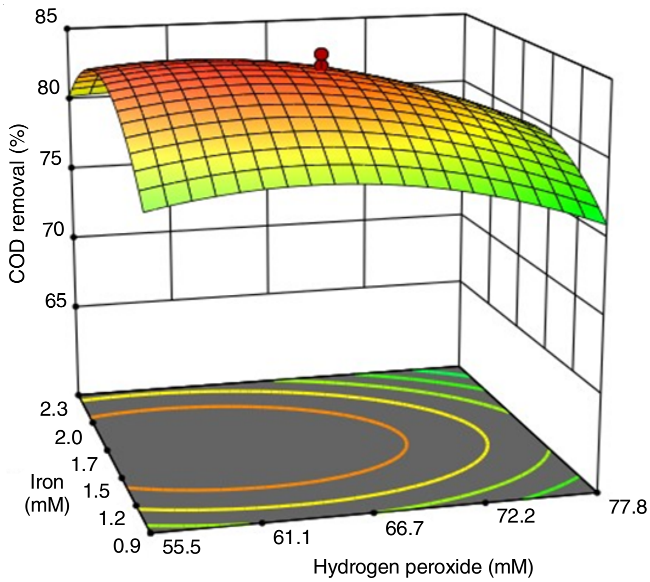

Fig. 3. Response surface plots percent COD removal from the wastewater, as a function of hydrogen peroxide dosage and Fe^{2+} dosage (pH = 3.5 and reaction time of 60 min)

can be seen that the increase in the dosage of Fe^{2+} produces not only more hydroxyl radicals ($^{\bullet}OH$) but also provides rapid oxidation of wastewater. With further increase in iron (Fe^{2+}) dosage, the decrease in the reduction of COD can be observed which imply that the H_2O_2 dosage becomes the limiting factor for further production of hydroxyl radical ($^{\bullet}OH$). An increase in the H_2O_2 dosage decreases the removal of COD which is due to the generation of another radical ($HO_2^{\bullet}$) which has electro potential lesser than hydroxyl radicals ($^{\bullet}OH$) [28]. From these results, it can be stated that a significant dosage of H_2O_2 and Fe^{2+} is required for the maximum reduction of COD. As can be seen from surface plots, the highest removal of COD (84.88%) is observed at a catalyst dosage of 1.6 mM, oxidising agent dosage at 66.7 mM, a electrolysis time of 60 min and 3.5 pH. Lowest removal of COD was observed at a lower dosage of a catalyst of 0.9 mM.

Interactive effect of H_2O_2 dosage and Fe^{2+} dosage on removal of colour: For catalyst dosage of 1.605 mM, oxidising dosage of 66.7 mM and reaction time of 60 min at pH 3.5, the interactive effect of Fe^{2+} and H_2O_2 dosages on colour removal is shown in Fig. 4. High colour removal efficiency (more than 80%) was observed at all the experimental conditions. The maximum removal of the colour of 99% was observed at Fe^{2+} dosage of 1.605 mM.

Interactive effect of Fe^{2+} and pH on COD removal: The interactive effect of catalyst dosage and pH on the COD removal is shown in Fig. 5 acidic pH value above 3.3 leads to the formation of active iron species of Fe^{2+} ions due to which the generation of hydroxyl radicals ($^{\bullet}OH$) increases [28]. The maximum reduction of COD was observed at pH 3.5 for Iron dosage of 1.605 mM, H_2O_2 dosage of 66.65 mM at a reaction time of 60 min.

Interactive effect of Fe^{2+} and pH on colour removal: Fig. 6 displays the interactive effect of Fe^{2+} dosage and pH value on colour removal for catalyst dosage of 1.605 mM and

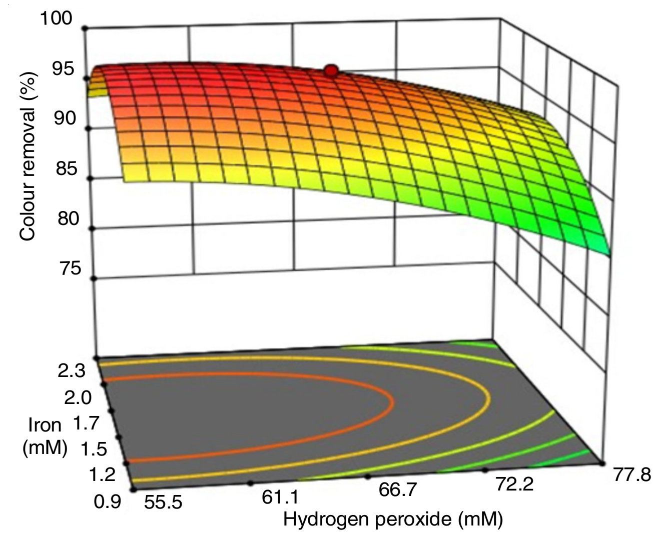

Fig. 4. Response surface plots percent colour removal from the wastewater, as a function of hydrogen peroxide dosage and Fe^{2+} dosage (pH = 3.5 and reaction time of 60 min)

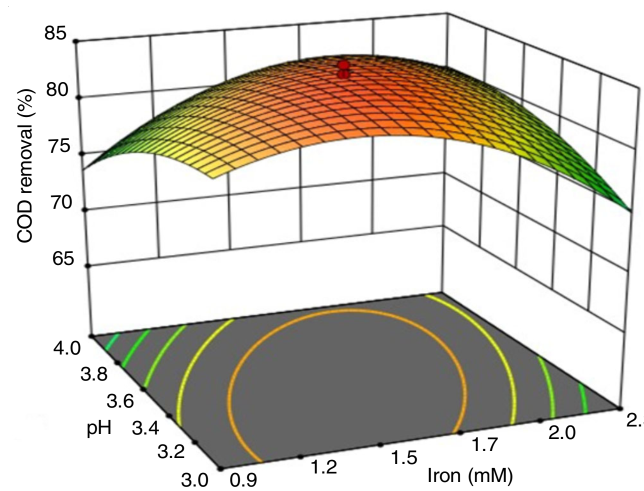

Fig. 5. Response surface plots percent COD removal from the wastewater, as a function of Fe^{2+} dosage and pH (hydrogen peroxide dosage = 66.65 mM and reaction time of 60 min)

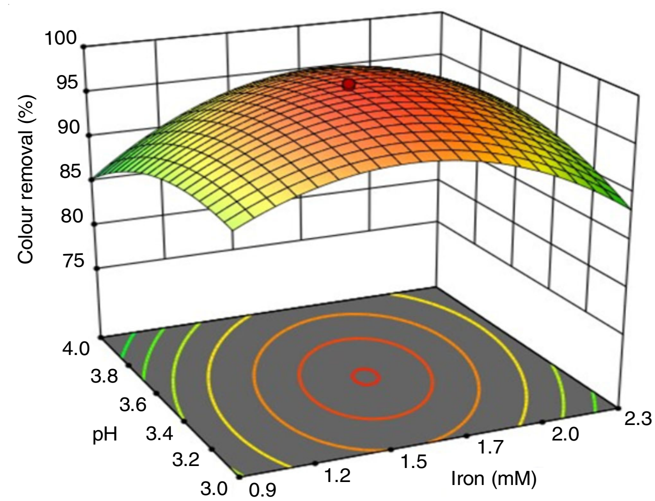

Fig. 6. Response surface contour plots percent colour removal from the wastewater, as a function of Fe^{2+} dosage and pH (hydrogen peroxide dosage = 66.65 mM and reaction time of 60 min

TABLE-4
COMPARISON OF PREDICTED AND EXPERIMENTAL RESULTS FOR OPTIMUM WORKING CONDITIONS FOR TREATMENT OF WASTEWATER USING FENTON's PROCESS

Parameters	Optimum value of the parameters	COD (%)		Colour (%)	
		Predicted	Experimental	Predicted	Experimental
pH	3.33	84.56	83.50	99.17	98.10
H_2O_2	60.57				
Fe^{2+}	1.56				

pH value 3.5 and reaction time of 60 min at H_2O_2 dosage of 66.65 mM. High colour removal efficiency (more than 80%) was observed at all the experimental conditions. Maximum removal of colour of 99% was observed at Fe^{2+} dosage of 1.605 mM.

Process optimization: Contour overlay plots were drawn graphically to analyze the optimum working condition for the process. The optimum region can be found by keeping independent variables in the desired range [19]. In present work, the independent variables namely, pH, H_2O_2 and Fe^{2+} dosage were kept in the desired range for 60 min of electrolysis time. The response was kept maximum for COD and colour removal. The model validation study was conducted at optimum working conditions for electrolysis time of 60 min. The outcomes of this study are shown in Table-4. These results indicate that the model predictions are 84.56 and 99.17% removal for COD and colour under the optimum working conditions of H_2O_2 dosage (60.57 mM), Fe^{2+} dosage (1.56 mM) and pH (3.33). The experimental results under optimum working conditions are 83.50 and 98.1% of COD removal and colour, respectively. Hence it can be said that the model prediction is in best agreement with the experimental result.

Conclusion

The parameters affecting the degradation and colour removal of cotton textile wastewater by Fenton's process at varying operating conditions were investigated using CCD. The second order polynomial equation (mathematical model) generated by CCD from the experimental results represents an empirical relationship between independent operating parameters and dependent parameters (responses). The derived model represents a significant R^2 (0.982) and R^2_{adj} (0.966) values which are almost similar for COD and colour removal. These values represent a best fit correlation with each other. The optimized values for the operating parameters were 3.33, 60.57 and 1.56 mM for pH, H_2O_2 dosage and Fe^{2+} dosage, respectively at reaction time of 60 min. Under these operating conditions, the maximum decolourization was 98.1% and the COD removal efficiency was found to be 83.5%. From the observation of these results it can be said that the model derived results best fit with the experimental results. The results show that the Fenton's process can be efficiently used for the degradation and colour removal of cotton textile wastewater.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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