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Optimal Fenton Process Using the Modified Taguchi Approach

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Abstract

Unavoidable scatter in repeated test results can be due to influence of unknown process variables (if any) and measurement errors to a certain extent. Modified Taguchi approach recommends few tests as per the orthogonal array and provides the range of estimates for combinations among the levels of process variables. It identifies optimal process variables and demands additional experimentation for confirmation (if necessary). One of the widely applied advanced oxidation processes (namely, Fenton oxidation) utilizes ferrous iron and hydrogen peroxide under acidic conditions to produce hydroxyl radical. This article presents optimal process variables for COD and decolorization efficiency of Fenton oxidation adopting multi-objective optimization. Empirical relations are presented for COD and decolorization efficiency in terms of Fenton oxidation process variables. Comparative study indicates reasonably good agreement between empirical relations and test results.

Keywords: Acid blue dye 113, confirmation of experiments, chemical oxygen demand (COD), decolorization, efficiency, expected range, Fenton treatment, multi-objective optimization.

1. Introduction

In 1894 Fenton discovered a special oxygen transfer properties in many metals. They improve the utilization of hydrogen peroxide. Strong catalytic power in metals gets highly reactive hydroxyl radicals (OH). The iron catalysed hydrogen peroxide (called as the Fenton's reaction) is employed to treat varieties of water pollution (like BTEX, formaldehyde, pesticide, phenols, rubber chemicals, etc.). This process is being applied to contaminated soils, sludges, and wastewater for improving biodegradability, removing BOD/COD as well as odor and colour, destructing organic pollutant as well as resin in radioactive contaminated sludge, and minimizing toxicity (https://www.lenntech.com/fenton-reaction.htm). Andreozzi et al. (1999) studied Fenton oxidation utilizes Fe^{+2} (ferrous iron) and H_2O_2 (hydrogen peroxide) under acidic conditions for advantageous formation of HO^* (hydroxyl radical). Fenton reaction performance Equation (1) is influenced by the process parameters (viz., pH, reaction time, initial concentrations of Dye, H_2O_2 and Fe^{+2} catalysts) given by Ntampegliotis et al. (2006) and Hameed and Lee (2009). Gomathi et al. (2009) tested various conditions of Fenton process. Asghar et al. (2009) examined advanced oxidation processes for in-situ production of hydrogen peroxide/hydroxyl radical for textile wastewater treatment. Albert (2010) focused on treatment of industrial wastewater by Fenton process combine with coagulation. Adopting Taguchi techniques many engineering/industrial optimization problems are solved. Few of interesting works among them are: Srinivasa Rao et al. (2008) observed the effect of drilling induced damage on notched tensile strength and pin-bearing strength of woven GFR-epoxy composites. Stage and satellite separations in space launch vehicle flights by Singaravelu (2009) and (2012). Pillai et al. (2011) studied Taguchi's approach to examine the effect of drilling induced damage on the notched tensile strength of woven GFR-epoxy composite. Rossie (2014) applied Fenton's process to wastewaters treatment: Heterogeneous and homogeneous catalytic operation modes. There is a need for minimizing the consumption of chemicals and cost of H_2O_2.

Asghar et al. (2014) mentioned the reaction in the process is as follows:

$$Fe^{+2} + H_2O_2 \rightarrow Fe^{+3} + HO^* + OH^-. \tag{1}$$

Asghar et al. (2014) have performed Fenton oxidation at atmospheric pressure and room temperature. They have prepared different concentrations of acid blue dye 113 in batch laboratory scale Erlenmeyer flask equipped with a magnetic stirrer. They have presented the efficiency (%) of the Fenton process considering COD removal and decolorization. All experiments are designed as per the response surface methodology (RSM) using central composite design (CCD) and the design expert. Initial concentrations of Dye, $H_2O_2 : Fe^{+2}(wt/wt)$ratio, $Dye : Fe^{+2}(wt/wt)$ratio and pH are the independent Fenton process variables, which are designated by X_1, X_2, X_3 and X_4, respectively. The efficiency (%) of Fenton process for COD removal and decolorization are designated by Y_1 and Y_2.

Optimal manufacturing process parameters are suggested by several authors such as: Bharathi et al. (2016) and Sahiti et al. (2016), Optimum WEDM process parameters of SS304 using Taguchi method. Eaton (2017) measured COD as per the standard method. For this, COD test cells (procured from Merck Sdn., Malaysia) are heated in a thermoreactor (Spectroquant TR 420, Merck Sdn., Selangor, Malaysia) after filling the required amount of sample. Aramyan and Moussavi (2017) reviewed advances in Fenton and Fenton based oxidation processes for industrial effluent contaminants control. Krishnan et al. (2017) compared various advanced oxidation processes used in remediation of industrial wastewater laden with recalcitrant pollutants. Rajeev et al. (2017) Optimized drilling parameters of coir fibre-reinforced polyester composites. Sastry et al. (2017) applied Taguchi approach to seek optimum drilling parameters for woven fabric carbon fibre/epoxy laminates. Roudi et al. (2018) tried to predict and optimize the Fenton process for the treatment of Landfill leachate using an artificial neural network. Buthiyappan et al. (2018) made a study on textile wastewater treatment efficiency by Fenton oxidation with integration of membrane separation system. The COD removal efficiency (%) is evaluated from Asghar et al. (2014) are provided by the Equations (2) and (3).

$$Y_1 = COD(\%) = 100X\left(1 - \frac{COD_f}{COD_o}\right). \tag{2}$$

Here COD_f and COD_o are the chemical oxygen demand of the treated and untreated dye measurements using UV-spectrophotometer. A digital pH meter (Cyberscan pH 300, Eutectic instruments, Wisconsin, USA) is used for pH measurements of the solution. Residual of H_2O_2 is measured in the treated wastewater using peroxide test strips (Merck Sdn., Selangor, Malaysia). The samples (before and after treatment) are scanned using UV-Spectrophotometer (Spectroquant Pharo 300, Merck Sdn., Selangor, Malaysia). Decolorization efficiency (%) of the treated sample is evaluated from Asghar et al. (2014),

$$Y_2 = \text{Decolorization}\left(\%\right) = 100\left(1 - \frac{Abs_f}{Abs_e}\right). \quad (3)$$

Here Abs_f and Abs_e are the UV-absorbance of the treated dye and untreated dye, respectively. To optimize the output responses (Y_1 and Y_2), the design expert suggests 30 test-runs having 16 factorial, 8 axial and 6 center points. The assigned low and high levels of the input process parameters are $X_1 \in [100, 300]$; $X_2 \in [5, 25]$; $X_3 \in [10, 50]$ and $X_4 \in [2, 9]$. Empirical relations developed for Y_1 and Y_2 are presented in the form of Equations (4) and (5) in terms of X_1, X_2, X_3 and X_4 are

$$\begin{aligned} Y_1 = {} & 64.1862 - 0.20098X_1 + 0.70063X_2 + 1.75866X_3 - 0.42339X_4 + 3.48465X10^{-3}X_1X_2 \\ & -1.74043X10^{-3}X_1X_3 + 1.3948X10^{-2}X_1X_4 + 3.1013X10^{-2}X_2X_3 - 0.13479X_3X_4 \\ & +3.88643X10^{-4}X_1^2 - 6.6812X10^{-2}X_2^2 - 2.1456X10^{-2}X_3^2 . \end{aligned} \quad (4)$$

$$\begin{aligned} Y_2 = {} & 85.85276 + 9.2035X10^{-2}X_1 + 1.4344X_2 - 0.70074X_3 - 0.97745X_4 - 1.26375X10^{-3}X_1X_2 \\ & -1.0125X10^{-4}X_1X_3 + 1.6293X10^{-2}X_1X_4 + 1.945X10^{-2}X_2X_3 - 1.6857X10^{-2}X_2X_4 \\ & +3.6054X10^{-2}X_3X_4 - 2.50533X10^{-4}X_1^2 - 3.6003X10^{-2}X_2^2 + 2.85X10^{-3}X_3^2 - 0.50615X_4^2. \end{aligned} \quad (5)$$

They have also performed tests as per the L_9 OA (orthogonal array) of Taguchi. S/N ratio transformation corresponding to the "larger-is-better" is applied and performed ANOVA. The optimized Y_1 and Y_2 from the Taguchi method are comparable with those of CCD, Ross (1989) demonstrates the potentiality of the Taguchi method with few experiments. Many researchers have made investigations on industrial wastewater treatment opting Fenton process.

Dutta and Nageswara (2018) investigated the performance of chevron type plate heat exchangers. Satyanarayana et al. (2018) identified optimum laser beam welding process parameters for E110 zirconium alloy but joint based on Taguchi-CFD simulations. Rajyalakshmi and Nageswararao (2019a) Expected range of the output response for the optimum input parameters utilizing the modified Taguchi approach. Rajyalakshmi and Nageswararao (2019b) Modified Taguchi approach to trace the optimum GMAW process parameters on weld dilution for ST-37 steel plates. Satyanarayana et al. (2019) determined Optimal laser welding process parameters and expected weld bead profile for P92 steel. Dharmendra et al. (2019a, 2019b) suggested a simple and reliable Taguchi approach for multi-objective optimization to identify optimal process parameters in nano-powder-mixed electrical discharge machining of INCONEL800 with copper electrode. Harish et al (2019) obtained optimal process parameters to achieve maximum tensile load bearing capacity of laser weld thin galvanized steel sheets. Prasad et al. (2019) made a study on FSW process parameters to improve the strength of dissimilar AA6061-T6 to Cu Welds with Zn Interlayer. Appreciable scatter in the efficiency (%) of the Fenton oxidation is noticed in few repeated test runs. In fact, S/N ratio transformation in Taguchi method applied by Asghar et al. (2014) on the efficiency (%) is valid only for repeated test runs. Application of such transformation to a single test run data as in Kus (2016), Kumar et al. (2016), Banu et al. (2017), Riberio et al. (2017), Bhirudi and Gawande (2017), Cui et al. (2018), Jaiganesh et al. (2018) and Rajesh et al. (2018) have no added advantages other than transforming test data on the logarithmic scale. Their deterministic approach provides a single value of Y_1 and Y_2, whereas the test data in Asghar et al. (2014) indicate large scatter.

Modified Taguchi approach Rajyalakshmi and Nageswararao (2019a, 2019b) provides the range of estimated responses for the levels of input process variables considering few experimental data as per the OA (orthogonal array) of Taguchi. It recommends the Chavunet's criterion to consider statistically acceptable repeated test data for reporting average value. By doing this, there is no need to apply S/N ratio transformation on repeated test data The current trend is towards the range of estimated responses. This article utilizes a simple multi-objective optimization approach to identify a set of optimal Fenton oxidation process variables for achieving the optimal efficiency (%) of COD removal and decolorization. Empirical relations for the responses are developed in terms of Fenton process variables. Their adequacy is demonstrated compared with test data.

2. Test Data Acquisition

Asghar et al(2014) have performed Fenton oxidation considering initial concentrations of Dye, $H_2O_2 : Fe^{+2}$ (wt/wt) ratio, $Dye : Fe^{+2}$ (wt/wt) ratios and pH as independent process parameters. Their design of experiments involves the RSM using CCD. The efficiency of process is analyzed from the output responses, viz., COD removal efficiency (%) and decolorization efficiency (%). The Fenton process variables are designated by X_1, X_2, X_3 and X_4, respectively, whereas Y_1 and Y_2 are the efficiency (%) of COD removal and decolorization respectively. Empirical relations in Equations (4) and (5) are developed from the design expert. To examine the effect of Fenton process variables on Y_1 and Y_2, L_{25} OA of Taguchi as in Rajyalakshmi and Nageswararao (2019b) is considered assigning five levels to each process parameter (see Table 1). For the number of experiments, $N_{Taguchi}$=25 and 5 levels, there is a possibility of accommodating six process variables, whereas in Aramyan and Moussavi (2017) only four process variables are considered. Hence, two fictitious factors (viz., X_5 and X_6) are introduced in Table 1 as in Singaravelu et al. (2019). Table 2 gives L_{25} OA of Taguchi and the levels of Fenton process variables for each test run. Specifying the values of Fenton process variables corresponding to the levels in each test run, the output responses are generated from Equations (4) and (5) and presented in Table 2.

3. ANOVA

Analysis of variance (ANOVA) on Y_1 and Y_2 is performed and presented the results in Table 3. Initial concentrations of Dye, H_2O_2 : Fe^{+2} (wt/wt) ratios, $Dye : Fe^{+2}$ (wt/wt) ratios and pH are designated by X_1, X_2, X_3 and X_4, respectively. The output responses are Y_1 (COD removal efficiency (%)) and Y_2 (Decolorization efficiency (%)). The independent process parameters X_1, X_2, X_3 and X_4 have 6.64, 25.76, 18.55 and 16.86% contributions on the response Y_1. They have 15.03, 16.86, 9.32 and 55.31% contributions on the response Y_2.

The results in ANOVA (Table 3) indicate the increasing tendency of decolorization efficiency (%), Y_2 with increase in process parameters, X_1 (dye concentration) and X_2 ($H_2O_2 : Fe^{+2}$). Also, Y_2 decreases with increasing X_3 ($Dye : Fe^{+2}$) and X_4 (pH). Kang and Hwang (2000) have noticed unavailability of HO^* radical in the reaction at high pH due to decomposition of H_2O_2 to oxygen and conversion of ferrous ions to ferric hydroxy complex. This confirms active Fenton's reagents for decolorization. Mean values of the COD removable efficiency (%), Y_1 are above its grand mean for the process parameters: $X_1 \in [250,300]$; $X_2 \in [10,20]$; $X_3 \in [10,30]$ and $X_4 \in [2,3.75]$. Maximum COD removal efficiency (%) can be achieved within that range of process variables. Meric et al. (2004) and Argun and Karatas (2011) provided the efficiency of can be improved with chemical consumption and COD removal efficiency.

4. ANOVA

The additive law Ross (1989) estimates the output responses from the results of ANOVA analysis. For simplicity, ζ is assumed here as any one of the output responses (Y_1 and Y_2). Let $\zeta(X_i, j)$ is the mean value of ζ to the process parameter (X_i) and level '*j*' and $\overline{\zeta}$ is the grand mean of ζ for the 25 test runs in L_{25} orthogonal array. Using the results of ANOVA (Table 3), estimates of ζ given in Equation (6) (i.e., $\zeta_{estimate}$) for each test run are made from Ross (1989)

$$\zeta_{estimate} = \sum_{i=1}^{n_p} \zeta(X_i, j) - (n_p - 1)\overline{\zeta}. \tag{6}$$

Here, n_p is the number of process variables. The grand mean of the output responses are $\overline{Y}_1 = 67.51$ and $\overline{Y}_2 = 90.93$. The process variables identified to achieve maximum output response, $Y_{1\max}$ are $X_1^{(5)}, X_2^{(4)}, X_3^{(2)}, X_4^{(1)}$ in which superscript denotes the level of the process parameter. Similarly, $X_1^{(5)}, X_2^{(5)}, X_3^{(1)}, X_4^{(1)}$ are for $Y_{2\max}$. Using Equation (6), one can find $Y_{1\max} = 91.95$ and $Y_{2\max} = 108.71 > 100$. As in Pillai et al. (2011), the least and highest corrections with inclusion of fictitious parameters for the output response Y_1 are found to be -10.8 and 11.95 respectively. For the output response Y_2, the respective corrections arrived are –3.3 and 3.17. Applying these corrections, one can find the expected range of the output responses as $Y_{1\max} \in [81.85, 103.90]$, which implies that $Y_{1\max} \geq 81.85$; and $Y_{2\max} \in [105.41, 111.88]$; which implies that $Y_{2\max} \geq 100$. Tables 4 and 5 give estimates of Y_1 and Y_2 for different sets of process parameters. Test data in Asghar et al. (2014) are within/close to the estimated range.

5. Development of Empirical Relations

Using the results of ANOVA (Table 3), empirical relations presented in the form of Equations (7) and (8) for Y_1 and Y_2 are developed in terms of X_1, X_2, X_3 and X_4 in the form

$$Y_1 = Y_{1X1} + Y_{1X2} + Y_{1X3} + Y_{1X4} - 3\overline{Y}_1, \tag{7}$$

$$Y_2 = Y_{2X1} + Y_{2X2} + Y_{2X3} + Y_{2X4} - 3\overline{Y}_2. \tag{8}$$

Using Equations (6)-(8), the following equations obtained are as follows used to estimate the optimum parameters

$$Y_{1X1} = 2.27\zeta_1^2 + 3.93\zeta_1 + 66.4 \tag{9}$$

$$Y_{1X2} = -1.21\zeta_2^2 + 2.88\zeta_2 + 73.6 \tag{10}$$

$$Y_{1X3} = -7.89\zeta_3^2 - 5.15\zeta_3 + 71.5 \tag{11}$$

$$Y_{1X4} = -4.15\zeta_4^2 - 5.87\zeta_4 + 65.4 \tag{12}$$

$$Y_{2X1} = 0.57\zeta_1^2 + 4.49\zeta_1 + 89.5 \tag{13}$$

$$Y_{2X2} = -2.21\zeta_2^2 + 4.55\zeta_2 + 90.9 \tag{14}$$

$$Y_{2X3} = 4.54\zeta_3^2 - 2.31\zeta_3 + 87.5 \tag{15}$$

$$Y_{2X4} = -4.34\zeta_4^2 - 8.24\zeta_4 + 91.9 \tag{16}$$

$$\zeta_1 = 0.01X_1 - 2 \tag{17}$$

$$\zeta_2 = 0.1X_2 - 1.5 \tag{18}$$

$$\zeta_3 = 0.05X_3 - 1.5 \tag{19}$$

$$\zeta_4 = \frac{1}{7}(2X_4 - 11). \tag{20}$$

Figures 1 and 2 show a good comparison of Y_1 and Y_2 for the specific Fenton process variables in Tables 4 and 5 using the additive law Equation (6) and those obtained from empirical relations (7) and (8). Applying the least and highest corrections (-10.8 and 11.95) to empirical Equation (7), minimum and maximum estimates of Y_1 are obtained for the specific Fenton process variables. Similarly, the least and highest corrections (–3.3 and 3.17) are applied to empirical relation of Equation (8) to find the minimum and maximum estimates of Y_2 for the specific Fenton process variables. Table 6 presents estimates of Y_1 and Y_2 for the specific Fenton process parameters of the repeated tests. The test results of Y_1 are within/close to the estimated range. The discrepancy in the estimated range of Y_2 is due to large scatter in test data. Chauvenet's criterion is applied as in Rajyalakshmi and Nageswara Rao (2019a) and found no outliers in the test data.

6. Optimal Process Parameters

Asghar et al. (2014) have used the RSM and predicted $Y_1 = 81.64$ and $Y_2 = 99.4$ (whereas $Y_1 = 79.1$ and $Y_2 = 98.4$ are achieved) for the optimal Fenton process variables have determined using Equation from 9 to 16 for the given $X_1 = 300; X_2 = 19.15, X_3 = 25.92$ and $X_4 = 3$. The estimated range: $Y_1 \in [75.91, 98.66]$ and $Y_2 \in [98.12, 104.59]$ $\Rightarrow Y_2 \geq 98.12$. They have also adopted the Taguchi's L_9 OA for conducting experiments. They have performed ANOVA analysis after applying the S/N ratio transformation corresponding to the "larger-is-better" while evaluating S/N value for each response. They have predicted $Y_1 = 79.06$ for optimal parameters: $X_1 = 300; X_2 = 25; X_3 = 10$ and $X_4 = 3$. The achieved value is 81.2, whereas the estimated range for $Y_1 \in [64.12, 86.87]$. They have predicted $Y_2 = 99.31$ for optimal parameters: $X_1 = 300; X_2 = 25, X_3 = 15$ and $X_4 = 3$. The achieved value is 99.04, whereas the estimated range, $Y_2 \in [102.6, 109.1] > 100$.

7. Multi-Objective Optimization

To specify a set of process variables for optimum Y_1 and Y_2, the problem of multi-objective is converted for a single objective optimization. Both Y_1 and Y_2 are normalized with their respective maximum values determined from the Equations (17)-(20): $Y_{1\max} = 103.9$ and $Y_{2\max} = 111.87$ Defining $\zeta_1\left(\equiv \frac{Y_1}{Y_{1Max}}\right)$ and $\zeta_2\left(\equiv \frac{Y_2}{Y_{2Max}}\right)$ and introducing weighing factors ω_1 and ω_2 satisfying $\omega_1 + \omega_2 = 1$ as in Rajyalakshmi and Nageswararao (2019a, 2019b) single objective function (ζ) is of the form

$$\zeta = w_1\zeta_1 + w_2\zeta_2. \tag{21}$$

Mean values of Y_1 and Y_2 in ANOVA (Table 3) are normalized by $Y_{1\max} = 103.9$ and $Y_{2\max} = 111.87$, respectively. The normalized mean values of Y_1 and Y_2 are nothing but the mean values of ζ_1 and ζ_2, respectively. Multiplying the mean values of ζ_1 and ζ_2 with the weighing factors ω_1 and ω_2, and using Equation (21), the mean values of the multi-objective function (ζ), are obtained as in ANOVA (Table 3). The level of process variables corresponding to the maximum mean values of ζ represent the set of optimal processing variables for achieving maximum Y_1 and Y_2.

For the specified $\omega_1 = 1 (\Rightarrow \omega_2 = 0)$, maximization of (ζ) yields only the maximum Y_1 and the optimal process parameters are $X_1^{(5)}, X_2^{(4)}, X_3^{(2)}, X_4^{(1)}$. For the specified $\omega_2 = 1 (\Rightarrow \omega_1 = 0)$, maximization of ζ yields only the maximum Y_2 and the optimal process parameters are $X_1^{(5)}, X_2^{(4)}, X_3^{(2)}, X_4^{(1)}$. Equal weighing factors ω_1 and ω_2 are specified to achieve common optimal process conditions $X_1^{(5)}, X_2^{(5)}, X_3^{(1)}, X_4^{(1)}$. Hence, the optimized operating parameters for maximum Y_1 and Y_2 are: $X_1 = 300; X_2 = 20, X_3 = 10$ and $X_4 = 2$. Figure 3 shows variation of Y_1 and Y_2 with X_2 for the specific optimal process parameters $(X_1 = 300; X_2 = 10, X_4 = 2)$. The estimated range for the identified optimal process parameters are: $Y_1 \in [75.46, 98.21]$ and $Y_2 \in [104.8, 111.2]$.

The lower bound output responses Y_1 and Y_2 from the expected range should be preferable to have conservative estimates for optimal process variables X_1, X_2, X_3 and X_4. Due to large scatter in the output responses, $Y_2 > 100$, which indicates active Fenton's reagents for decolorization. This does not impact on the identified optimal process variables.

8. Concluding Remarks

A simple multi-objective optimization procedure is adopted in this article to select a set of optimal Fenton process variables for achieving maximum COD and decolorization efficiency. Modified Taguchi approach provides an estimated range of responses for the optimal process variables. Test data on Fenton oxidation is considered for validating the estimates. S/N ratio transformation in the Taguchi approach of single value output response will involve additional calculations and no improvements while seeking optimal solution. Empirical relations are presented for the output responses. Test results are within/close to the estimated range. Repetition of experiments with application of Chauvenet's criterion, is more appropriate to consider the statistically valid output responses in complex optimization problems. Active Fenton's reagents for decolorization are noticed for the optimal process parameters. Maximum COD removal efficiency (>75%) is assured for the identified set of optimal process variables. Taguchi approach is thus helpful in minimizing the consumption of chemicals and cost of H_2O_2 and providing the optimal solution with few experiments.

Table1 Levels of Fenton oxidation process variables

Process parameter	Designation	Level 1	Level 2	Level 3	Level 4	Level 5
Dye (mg/L)	X_1	100	150	200	250	300
H_2O_2: Fe^{+2} (wt/wt) ratio	X_2	5	10	15	20	25
Dye: Fe^{+2} (wt/wt) ratio	X_3	10	20	30	40	50
pH	X_4	2	3.75	5.5	7.25	9
Fictitious	X_5	x_{51}	x_{52}	x_{53}	x_{54}	x_{55}
Fictitious	X_6	x_{61}	x_{62}	x_{63}	x_{64}	x_{65}

Table 2 Efficiency (%) of COD removal and decolorization for the levels of Fenton process variables in test runs of Taguchi's L_{25} orthogonal array

Test run	Levels of Fenton process variables				Fictitious parameter levels		Efficiency of Fenton process (%)	
	X_1	X_2	X_3	X_4	X_5	X_6	Y_1 (COD)	Y_2 (decolorization)
1	1	1	1	1	1	1	66.05	92.17
2	1	2	2	2	2	2	74.63	90.24
3	1	3	3	3	3	3	73.97	86.89
4	1	4	4	4	4	4	64.05	82.13
5	1	5	5	5	5	5	44.89	75.94
6	2	1	2	3	4	5	66.05	84.37
7	2	2	3	4	5	1	66.04	80.44
8	2	3	4	5	1	2	56.78	75.09
9	2	4	5	1	2	3	82.64	99.13
10	2	5	1	2	3	4	53.40	98.78
11	3	1	3	5	2	4	57.42	71.61
12	3	2	4	1	3	5	75.28	91.04
13	3	3	5	2	4	1	69.22	96.77
14	3	4	1	3	5	2	64.53	97.8
15	3	5	2	4	1	3	65.46	89.83
16	4	1	4	2	5	3	60.51	86.89
17	4	2	5	3	1	4	55.10	92.04
18	4	3	1	4	2	5	74.96	94.44
19	4	4	2	5	3	1	76.55	85.89
20	4	5	3	1	4	2	77.46	99.52
21	5	1	5	4	3	2	40.28	84.93
22	5	2	1	5	4	3	84.69	88.71
23	5	3	2	1	5	4	77.61	97.73
24	5	4	3	2	1	5	82.40	100.3
25	5	5	4	3	2	1	77.94	101.4

Table 3 ANOVA on the efficiency (%) of COD removal and decolorization

Process parameter	1-Mean	2-Mean	3-Mean	4-Mean	5-Mean	% Contribution
COD removal efficiency (%), Y_1						
X_1	64.72	64.98	66.38	68.91	72.58	6.64
X_2	58.06	71.15	70.5	74.03	63.83	25.76
X_3	68.72	72.06	71.46	66.91	58.42	18.55
X_4	75.81	68.03	67.52	62.16	64.06	16.86
X_5	65.16	73.52	63.89	72.29	62.71	15.46
X_6	71.16	62.73	73.45	61.51	68.71	16.73
Decolorization efficiency (%), Y_2						
X_1	85.47	87.56	89.41	91.76	94.60	15.03
X_2	83.99	88.49	90.18	93.04	93.09	16.86
X_3	94.38	89.61	87.74	87.30	89.76	9.32
X_4	95.92	94.59	92.49	86.35	79.45	55.31
X_5	89.88	91.36	89.51	90.30	87.76	2.05
X_6	91.33	89.52	90.29	88.46	89.21	1.42

Table 4 Estimates of COD removal efficiency (%), Y_1

Levels of process parameters				COD removal efficiency (%), Y_1				
X_1	X_2	X_3	X_4	Test [3]	Eq.(4)	Eq.(6)	Expected range	
							Lower	Upper
3	3	3	3	75.3	73.20	73.34	62.54	85.29
3	3	3	3	76.4	73.20	73.34	62.54	85.29
5	5	5	5	62.8	60.91	56.36	45.56	68.31
3	5	3	3	73.4	69.76	66.66	55.86	78.61
5	1	5	5	34.8	35.06	50.59	39.79	62.54
3	3	3	3	73.4	73.20	73.34	62.54	85.29
1	1	1	5	66.5	63.41	53.03	42.23	64.98
1	5	5	1	87.7	85.27	60.25	49.45	72.20
3	3	3	5	65.8	67.33	69.88	59.08	81.83
5	3	3	3	78.3	80.21	79.54	68.74	91.49
1	3	3	3	77.1	73.96	71.68	60.88	83.63
1	1	1	1	67.0	66.05	64.78	53.98	76.73
1	1	5	5	30.2	32.98	42.73	31.93	54.68
3	3	3	3	74.5	73.20	73.34	62.54	85.29
5	1	1	5	77.9	79.42	60.89	50.09	72.84
3	1	3	3	60.8	63.29	60.89	50.09	72.84
1	5	1	5	48.0	50.51	58.80	48.00	70.75
3	3	3	3	70.3	73.20	73.34	62.54	85.29
5	5	1	1	67.4	63.56	78.41	67.61	90.36

Table 4 (Continued)

Levels of process parameters				COD removal efficiency (%), Y_1				
X_1	X_2	X_3	X_4	Test [3]	Eq.(4)	Eq.(6)	Expected range	
							Lower	Upper
5	1	1	1	61.4	62.53	72.64	61.84	84.59
1	5	1	1	50.3	53.15	70.55	59.75	82.50
3	3	1	3	67.7	67.68	70.6	59.8	82.55
5	5	1	5	80.4	80.45	66.66	55.86	78.61
5	5	5	1	75.5	81.76	68.11	57.31	80.06
3	3	3	1	74.1	79.07	81.63	70.83	93.58
1	5	5	5	44.7	44.89	55.18	44.38	67.13
3	3	5	3	62.7	61.56	60.30	49.50	72.25
3	3	3	3	73.4	73.20	73.34	62.54	85.29
1	1	5	1	72.1	73.36	54.48	43.68	66.43
5	1	5	1	61.2	55.91	62.34	51.54	74.29

Table 5 Estimates of decolorization efficiency (%), Y_2

Levels of process parameters				Decolorization efficiency (%), Y_2				
X_1	X_2	X_3	X_4	Test [3]	Eq.(5)	Eq.(6)	Expected range	
							Lower	Upper
3	3	3	3	95.4	95.34	90.55	87.25	93.72
3	3	3	3	95.4	95.34	90.55	87.25	93.72
5	5	5	5	96.8	96.30	87.62	84.32	90.79
3	5	3	3	98.1	97.67	93.45	90.15	96.62
5	1	5	5	80.0	80.38	78.52	75.22	81.69
3	3	3	3	95.5	95.34	90.55	87.25	93.72
1	1	1	5	55.8	59.69	74.01	70.71	77.18
1	5	5	1	99.2	100.2	94.96	91.66	98.13
3	3	3	5	78.9	80.54	77.50	74.20	80.67
5	3	3	3	94.5	98.78	95.74	92.44	98.91
1	3	3	3	90.1	86.89	86.61	83.31	89.78
1	1	1	1	93.4	92.17	90.48	87.18	93.65
1	1	5	5	56.1	54.97	69.40	66.10	72.57
3	3	3	3	95.4	95.34	90.55	87.25	93.72
5	1	1	5	89.1	85.92	83.14	79.84	86.31
3	1	3	3	84.3	85.82	84.36	81.06	87.53
1	5	1	5	67.9	65.11	83.11	79.81	86.28
3	3	3	3	94.6	95.34	90.55	87.25	93.72
5	5	1	1	98.9	98.31	108.70	105.40	111.87
5	1	1	1	95.2	95.59	99.61	96.31	102.78
1	5	1	1	97.6	99.95	99.58	96.28	102.75
3	3	1	3	98.2	97.68	97.19	93.89	100.36
5	5	1	5	84.4	86.28	92.23	88.93	95.40
5	5	5	1	99.4	98.24	104.09	100.79	107.26
3	3	3	1	98.3	97.75	93.97	90.67	97.14

Table 5 (Continued)

Levels of process parameters				Decolorization efficiency (%), Y_2				
X_1	X_2	X_3	X_4	Test [3]	Eq.(5)	Eq.(6)	Expected range	
							Lower	Upper
3	3	3	1	98.3	97.75	93.97	90.67	97.14
1	5	5	5	73.6	75.94	78.49	75.19	81.66
3	3	5	3	93.5	95.28	92.57	89.27	95.74
3	3	3	3	96.6	95.34	90.55	87.25	93.72
1	1	5	1	76.5	77.35	85.87	82.57	89.04
5	1	5	1	78.9	79.96	94.99	91.69	98.16

Table 6 Efficiency (%) of COD removal and decolorization from repeated tests for the specific Fenton process parameters (viz., Dye, H_2O_2: Fe^{+2} (wt/wt) ratios, Dye: Fe^{+2} (wt/wt) ratios, and pH which are designated by X_1, X_2, X_3 and X_4, respectively)

Fenton process parameters	COD removal efficiency (%), Y_1		Decolorization (%), Y_2	
	Test [3]	Estimated range	Test [3]	Estimated range
$X_1 = 200$; $X_2 = 15$; $X_3 = 30$; $X_4 = 5.5$	75.3, 76.4, 73.4, 74.5, 70.3, 73.4	63.57–86.32	95.4, 95.4, 95.5, 95.4, 94.6, 96.6	87.25–93.72
$X_1 = 100$; $X_2 = 15$; $X_3 = 30$; $X_4 = 5.5$	77.1, 53.07	54.21–76.96	90.1, 97.99	83.31 - 89.78
$X_1 = 100$; $X_2 = 5$; $X_3 = 10$; $X_4 = 2$	67.0, 74.86	61.91–84.66	93.4, 98.95	87.18–93.65
$X_1 = 100$; $X_2 = 25$; $X_3 = 50$; $X_4 = 9$	44.7, 36.31	37.93–60.68	73.6, 89.56	75.19–81.66

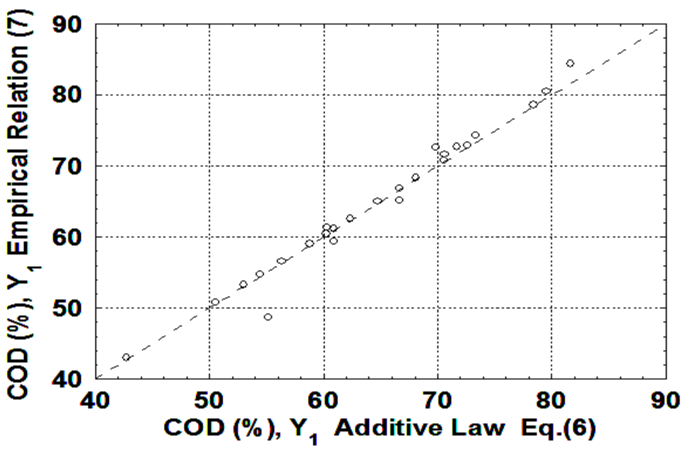

Figure 1 Comparison of COD removal efficiency (%), Y_1 for the levels of Fenton process variables in Table 4

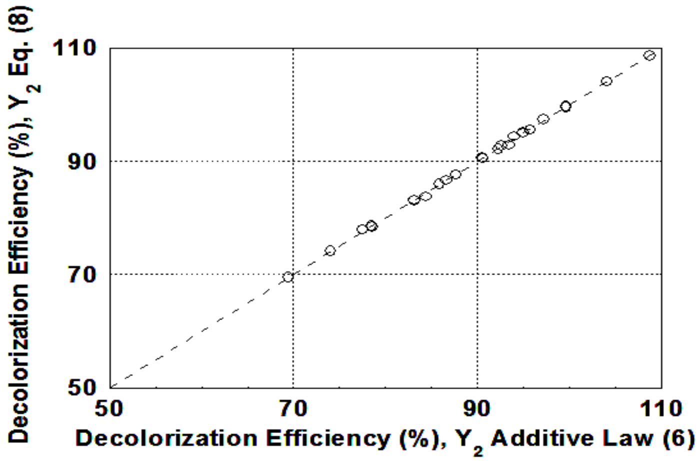

Figure2 Comparison of decolorization efficiency (%), Y_2 for the levels of Fenton process variables in Table 5

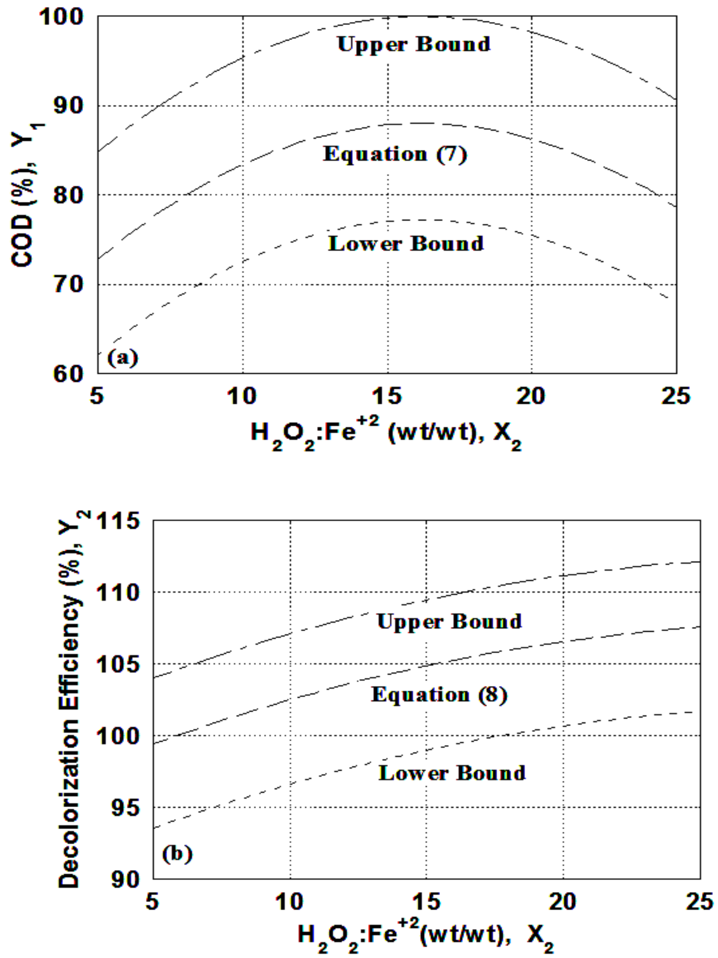

Figure 3 Variation of COD removal efficiency (%), Y_1 and decolorization efficiency (%), Y_2 with X_2 for the optimal Fenton process variables ($X_1 = 300; X_3 = 10;$ and $X_4 = 2$)

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