

Optimization of diazo dye disappearance by the UV/H₂O₂ process using the Box–Behnken design

Yasmine Laftani, Baylassane Chatib, Abdelghani Boussaoud, Mohammed El Makhfouk, Mohsine Hachkar and Mohammed Khayar

ABSTRACT

The purpose of this study was to apply the experimental Box–Behnken design (BBD) to evaluate the effect and, therefore, the optimal values of three chosen factors on the efficiency of the UV/H₂O₂ process to decolorize Ponceau S (PS) aqueous solutions. The factors studied at three levels were the irradiated volume of dye solution, the dye solution turbidity and the H₂O₂ dosage. The equations generated, analysis of variance (ANOVA), contour plots and response surface plots were used to analyze the relationship between independent variables and the outcomes of experiments. The fitted model was significant, with an adjusted coefficient of determination ($\text{adj-R}^2 = 0.9835$). The results showed that factors such as H₂O₂ dosage and irradiated volume were the main parameters that affected the decolorization efficiency of the PS aqueous solution, while the turbidity had a slight effect on the response. In addition, significant values were obtained for irradiated volume and H₂O₂ dosage interaction and square terms of all studied factors. Furthermore, the optimal conditions for decolorization of the PS aqueous solution were found to be an irradiated volume of 257.59 mL, a turbidity of 13 NTU and an H₂O₂ dosage of 1.76 mM.

Key words | advanced oxidation processes, Box–Behnken, decolorization, H₂O₂, optimization, Ponceau S

Yasmine Laftani (corresponding author)

Baylassane Chatib
Abdelghani Boussaoud
Mohammed El Makhfouk
Mohsine Hachkar
Mohammed Khayar

Laboratory of Process, Signals, Industrial Systems
and Computer Science, High School of
technology,

Cadi Ayyad University,
Dar Si-Aissa road, P.O. Box 89, Safi,
Morocco

E-mail: laftani90yasmine@gmail.com

INTRODUCTION

Since the discovery of the first synthetic dye in 1856, the progressive industrialization of dye production has done colossal damage to the aquatic environment in the form of toxic dye effluent. Textile industries use a maximum percentage of the synthetic dyes (around 56%) of world total annual production (Das & Mishra 2017). It has been estimated that among the various classes of dye, azo and diazo dyes are used most extensively in industrial dyeing and printing processes.

Moreover, some azo and diazo dyes are used in laboratories as either biological stains or pH indicators. They are characterized by the presence of the nitrogen-nitrogen bond ($-N=N-$) in addition to aromatic and naphthalenic systems with substituted auxochromes (such as OH, NH₂, CO₂H, SO₃ and Cl). The strong electron withdrawing character of the azo group stabilizes the aromatic substances against conversion by oxygenases. This durability makes azo and diazo dyes persistent pollutants, which are not

only harmful for aquatic life but also mutagenic to humans (Akbar Babaei *et al.* 2017; Muslim *et al.* 2013).

Ponceau S diazo dye is considered to be recalcitrant due to the presence of $-N=N-$ bonds and complex aromatic molecular structures, which are not easy to degrade by conventional methods (Vijayaraghavan *et al.* 2013; Quadrado & Fajardo 2017). It is known for its toxicity to aquatic life, along with carcinogenic and mutagenic effects in living organisms (Bharat *et al.* 2011). As a result, it is necessary to treat these colored compounds before discharging them into the environment (Das & Mishra 2017).

Many processes have been employed to eliminate azo and diazo dyes such as oxidative processes, physicochemical treatments (e.g. adsorption, coagulation, precipitation, among others), and synthetic biology (Marathe Sunil & Shrivastava Vinod 2015). Due to some drawbacks of the above mentioned processes, an alternative to conventional methods, advanced oxidation processes (AOPs), that refer

to a set of chemical treatment procedures (UV/H₂O₂, semi-conductors/UV, O₃/UV, Fe²⁺/H₂O₂, etc.) designed to degrade synthetic dyes using ambient temperatures and normal pressure. AOPs present several benefits such as simplicity of operation and lower investment costs. Such similar processes involve the generation of hydroxyl radicals (•OH) which act as a strong oxidizing agent (2.8 V vs. NHE) and react rapidly and non-selectively with most organic chemicals (Lin et al. 2016; Das & Mishra 2017). Moreover, these systems lead, in most cases, to the mineralization of the organic pollutants to H₂O, CO₂ and other non-toxic inorganic compounds, i.e. they do not cause secondary pollution (Velo-Gala et al. 2014; Laftani et al. in press).

Combining UV and hydrogen peroxide (UV/H₂O₂) is one of the AOPs that is useful for organic pollutant treatment: hydrogen peroxide can be photolysed by UV radiations, yielding the homolytic scission of the O-O bond of the H₂O₂ molecule and leading to the formation of HO• radicals as shown in Equation (1) (Ghodbane & Hamdaoui 2010).



Mechanistically, in several steps of oxidizing reaction, the •OH radical attacks the unsaturated dye molecules and the azo bond (N=N) in the chromogen, thereby decolorizing the dye wastewater (Ghodbane & Hamdaoui 2010). The UV/H₂O₂ process has several advantages, such as the practicability of the process under ambient temperature and pressure, considerable solubility and stability of H₂O₂ in water over a wide pH range. Furthermore, the cost of the process is low and there is a lack of sludge formation compared to the Fenton process (Ghodbane & Hamdaoui 2010).

Chemometrics based on design of experiments (DOE), which is a set of variants that are relevant to studying the influence of different variables on the response of a controlled experiment. In recent years, chemometric tools have been frequently applied to the optimization of operating parameters in analytical methods. Their advantages include a reduction in the number of experiments, resulting in lower reagent consumption and considerably less laboratory work, and they give access to a predictive mathematical model in which assessment of relevance is achieved using the statistical significance of the factors aiming to optimize the process (Ferreira et al. 2007).

The Box-Behnken design (BBD) is a specific type of experimental design introduced in 1960 and used in the optimization of process variables with three factors at three levels (maximum, minimum and center point) with 17 runs, including five central points. It does not contain points on the

vertices of the experimental region representing level combinations that are prohibitively expensive or impossible to test because of physical process constraints (Ferreira et al. 2007). As a chemometric methodology, the BBD is often presented as a good design for response surface to optimize several processes and/or analytical techniques such as adsorption process (Kannan et al. 2004), advanced oxidation processes (Kumar et al. 2016), struvite precipitation from wastewater (Daneshgar et al. 2019), photoelectrocatalytic oxidation system (Fu et al. 2007) and electrochemical process (Ruotolo & Gubulin 2005). The BBD permits access, through its mathematical model, to an appropriate statistical significance of factors with their interactions. The BBD is more cost-effective than three-level full factorial designs, and allows the evaluation of the quadratic model parameters as well as the detection of the lack of adjustment of the model (Ferreira et al. 2007).

The present study was aimed at the optimization of selected determinant factors in Ponceau S diazo dye decolorization by the UV/H₂O₂ process using the Box-Behnken design. The factors studied were the irradiated volume, in order to measure the UV dose absorbed by the solution, the turbidity and the H₂O₂ dosage. Each of them was assigned three levels (-1, 0, +1) after assessing their importance in terms of practicability and the influence on outcome of experiments.

MATERIALS AND METHODS

Materials

Ponceau S (abbreviation: PS; color index number: 27195; chemical class: diazo; molecular formula: C₂₂H₁₂N₄Na₄O₁₃S₄; molecular weight: 760.6 g/mol; λ_{max}: 520 nm) was purchased from REACTIFS RAL. The molecular structure of PS is shown in Figure 1. Hydrogen peroxide (H₂O₂, 50%) was obtained from Prochilabo. Otherwise, the kaolin was supplied by SDS.

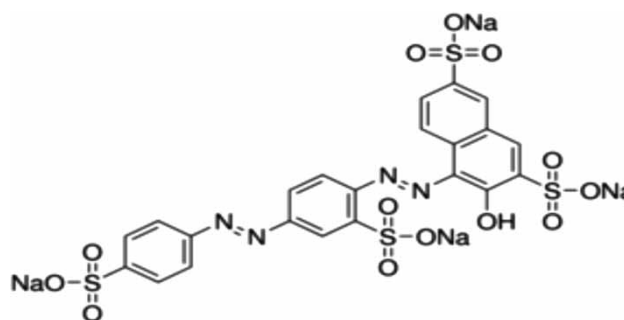


Figure 1 | Molecular structure of Ponceau S (PS).

The ultrapure water with a resistivity of 0.055 μS/cm, which was obtained using a VWR Puranity TU, was used to prepare all stock and working solutions.

All chemicals were used as received without further purification. The UV-vis spectra of PS diazo dye was recorded from 200 to 800 nm using a UV-vis spectrophotometer (Rayleigh UV-1800) with a spectrometric quartz cell (1 cm path length). The maximum absorbance wavelength (λ_{max}) of PS could be found at 520 nm.

Methods

Procedure

Decolorization of the PS aqueous solution was carried out by the UV/H₂O₂ process. A stock of 0.0657 mM of PS dye was prepared by dissolving the required amount in ultrapure water. The working solution of hydrogen peroxide was obtained from the commercial solution (H₂O₂, 50%) by dilution in ultrapure water to the required concentration. It may be mentioned that all volumetric flasks were covered with aluminium foil to protect the contents from sunlight.

Throughout this work, the required amount of kaolin was added to the dye solution in order to achieve the turbidity desired, measured using a turbidimeter (HACH 2100P). A magnetic stirrer was used to get a homogeneous mixture. The reaction mixtures were prepared from dye stock solution and a desired amount of hydrogen peroxide. The reaction time was recorded when the H₂O₂ was added to the reactor.

The reactor used in all experiments was an aerated cylindrical vessel in which we immersed a double walled quartz sleeve containing a high-pressure mercury lamp (250 W, Ingelec). Continuous circulation of water in the sleeve was used to keep the temperature constant in the treated solution. Constant agitation was assured by means of a magnetic stirrer placed at the reactor base.

After 40 min of decolorization of the PS aqueous solution by the UV/H₂O₂ process, an aliquot of 5 mL was taken from the reaction mixture and centrifuged at 2,300 rpm (for 3 min) with a centrifuge (FiRLaBO FC 40). The supernatant solution of centrifuged samples of the PS aqueous solution were filtered and then analyzed using the UV-visible spectrophotometer. Decolorization of the PS dye was monitored by measuring the absorbance of the cell free supernatant at 520 nm.

The decolorization efficiency was determined by Equation (2).

$$\% \text{ decolorization efficiency} = \left(\frac{A_t - A_0}{A_0} \right) \times 100 \quad (2)$$

where A_0 is the initial absorption of PS, and A_t is the absorption of PS aqueous solution at reaction time $t = 40$ min.

Experimental design

The BBD using the response surface methodology (RSM) was used as an empirical optimization technique for evaluating the relationship between responses and three chosen factors at three levels with 17 runs, including five central points.

The BBD is an independent quadratic design in that it does not contain an embedded factorial or fractional factorial design. These designs are rotatable (or near rotatable) and require three levels of each factor (Ferreira et al. 2007; Masomboon et al. 2010; Anuar et al. 2013).

The BBD helps to accumulate a second-order polynomial equation and also assists in getting an optimal formulation with the execution of the least number of trial runs in order to avoid experiments performed under extreme conditions (Sathyamoorthy et al. 2017; Ghosal et al. 2018).

The software product, Design-Expert 7 trial version, was used for determining the optimum conditions for decolorization of the PS aqueous solution by UV/H₂O₂ process using the BBD. The irradiated volume, the turbidity and H₂O₂ dosage was selected as independent factors A , B , and C respectively. The concentration of PS was fixed as 0.0657 mM for all experiments. These variables are studied at three different levels, i.e. low (−1), medium (0) and high (+1). The conditions of the BBD are summarized in Table 1.

A non-linear quadratic model Equation (3) was utilized to evaluate the experimental response (Y) as follows:

$$Y = b_0 + b_1A + b_2B + b_3C + b_1b_2AB + b_1b_3AC + b_2b_3BC + b_{11}A^2 + b_{22}B^2 + b_{33}C^2 \quad (3)$$

Y is the measured response, b_0 is the intercept, $b_1 - b_{33}$ are the regression coefficients, A , B , C are the independent

Table 1 | Chosen variables and their levels in Box-Behnken design

Variables	Symbol	Variable level		
		Low (−1)	Center (0)	High (+1)
Irradiated volume (mL)	A	250	375	500
Turbidity (NTU)	B	10.5	38.9	64
H ₂ O ₂ (mM)	C	0.46	1.23	2

factors, AB , AC , BC are the interaction effects and A^2 , B^2 , C^2 are the quadratic expressions of chosen factors. The model given in order to predict and control the response under different chosen variables was checked in terms of the values of R-squared and the adjusted R-squared (adj-R²).

RESULTS AND DISCUSSION

Effect of formulation variables on the decolorization efficiency of PS aqueous solution

The 17 various experimental runs were conducted for each set of process factors by the BBD to estimate the effect of different variables on the responses. The data in Table 2 give all formulae prepared according to the BBD for each combination of the three selected factors and corresponding responses.

The BBD with three factors including the irradiated volume, the turbidity and the H₂O₂ dosage each at three levels, was built with a view to describing and evaluating the influence of the above parameters on the decolorization of the PS dye by the UV/H₂O₂ process.

Table 2 | Ponceau S decolorization by the UV/H₂O₂ process designed by the Box–Behnken design

Run number	Irradiated volume (mL)	Turbidity (NTU)	H ₂ O ₂ (mM)	Decolorization efficiency (%) at t = 40 min
1	250 (−1)	10.5 (−1)	1.23 (0)	91.53
2	500 (+1)	10.5 (−1)	1.23 (0)	99.29
3	250 (−1)	64 (+1)	1.23 (0)	90.64
4	500 (+1)	64 (+1)	1.23 (0)	98.73
5	250 (−1)	38.9 (0)	0.46 (−1)	59.07
6	500 (+1)	38.9 (0)	0.46 (−1)	78.62
7	250 (−1)	38.9 (0)	2 (+1)	98.50
8	500 (+1)	38.9 (0)	2 (+1)	99.48
9	375 (0)	10.5 (−1)	0.46 (−1)	66.83
10	375 (0)	64 (+1)	0.46 (−1)	59.77
11	375 (0)	10.5 (−1)	2 (+1)	98.59
12	375 (0)	64 (+1)	2 (+1)	96.11
13	375 (0)	38.9 (0)	1.23(0)	79.37
14	375(0)	38.9 (0)	1.23(0)	77.97
15	375 (0)	38.9 (0)	1.23(0)	80.26
16	375 (0)	38.9 (0)	1.23(0)	78.53
17	375 (0)	38.9 (0)	1.23(0)	77.73

Table 2 lists the values of variables, the experimental data according to the BBD (−1, 0, +1) and the measured responses. The maximum decolorization of the PS aqueous solution was 99.48% and the minimum was 59.07%.

It was found that increasing H₂O₂ concentration increased the decolorization efficiency of the PS aqueous solution. When 0.46 mM of hydrogen peroxide was added to the volume of 250 mL, the decolorization efficiency was only 59.07% (run number 5). However, by increasing the H₂O₂ concentration from 0.46 to 2 mM, the decolorization efficiency increased to 98.50% (run number 7). This proves that the effect of increasing hydrogen peroxide concentration is positive for efficient decolorization of PS dye, and can be attributed to generation of more hydroxyl radicals.

The results also show that increasing the irradiated volume can improve the decolorization efficiency of the PS aqueous solution. Maximum decolorization of 59.07% (run number 5) was observed when 0.46 mM of hydrogen peroxide was added to 250 mL of turbid solution at 38.9 NTU. Increasing the irradiated volume from 250 mL to 500 mL increased the decolorization efficiency to 78.62% (run number 6). By keeping the reactor position at the same level, and increasing the irradiated volume of the dye solution, the amount of UV radiation absorbed by H₂O₂ increased, leading to the formation of plentiful hydroxyl radicals in the treated solution.

Increasing the turbidity of the treated solution decreased the decolorization efficiency of the PS aqueous solution. When 2 mM of hydrogen peroxide was added to 375 mL of turbid solution at 10.5 NTU, the decolorization efficiency was 98.59% (run number 11). Increasing the turbidity of the treated solution from 10.5 to 64 NTU reduced the decolorization efficiency to 96.11% (run number 12).

Box–Behnken analysis

The sequential model sum of squares and model summary statistic were outputted to examine the adequacy of the models generated from the input data. The results are shown in Table 3. Sequential model sum of squares selects the highest order polynomial where the additional terms are significant and the model is not aliased. The results showed that, for quadratic versus 2FI (two-factor interaction), the *p*-value was less than 0.0001 which shows high significance. It is clear that the cubic versus quadratic model is not adequate for the experimental data.

The model summary statistic focussed on the model maximizing the adjusted R-squared and the predicted

Table 3 | Adequacy of model tested**Sequential model sum of squares**

Source	Sum of squares	Df	Mean square	F-value	p-value	Remarks
Mean versus total	1.205×10^5	1	1.205×10^5			
Linear versus mean	2,241.03	3	747.01	12.25	0.0004	
2FI versus linear	91.48	3	30.49	0.43	0.7328	
Quadratic versus 2FI	678.36	3	226.12	69.11	<0.0001	Suggested
Cubic versus quadra	18.54	3	6.18	5.67	0.0635	Aliased
Residual	4.36	4	1.09			
Total	1.235×10^5	17	7,264.33			
Model summary statistic						
Source	Std Dev.	R ²	Adjusted R ²	Predicted R ²	PRESS	Remarks
Linear	7.81	0.7387	0.6784	0.5008	1,514.32	
2FI	8.37	0.7688	0.6302	0.0112	2,999.75	
Quadratic	1.81	0.9925	0.9827	0.9000	303.50	Suggested
Cubic	1.04	0.9986	0.9943		+	Aliased

+Case(s) with leverage of 1.0000: PRESS statistic not defined.

R-squared (pred-R²). The results showed that the quadratic model has the highest values for R² and pred-R² compared to the other models, which means that it presents the highest significance, so it was suggested as a model to fit the experimental data. The cubic model was disregarded as it is aliased.

Model fitting and statistical analysis of data

Model equation

The Box–Behnken design was performed with a view to obtaining a formula for the optimum decolorization efficiency of the PS aqueous solution. The relationship between the response and the three chosen factors is shown in Equation (4):

$$\begin{aligned} \% \text{ decolorization} = & +78.77 + 4.55A - 1.37B + 16.05C \\ & + 0.083 AB - 4.64 AC + 1.14 BC + 9.93A^2 + 6.34 B^2 \\ & - 4.79 C^2 \end{aligned} \quad (4)$$

The above equation described the individual main effect, interaction effect and quadratic effect of the selected independent variables. The sign of each coefficient shows how the factor influences the response. If the coefficient is negative the response decreases, and if it is positive, the response increases (Sadoun *et al.* 2018). It should be noted

that the precision of the model represented by Equation (4) measures the signal to noise ratio; any value greater than 4 is acceptable. This model has a ratio of 30.424, indicating an adequate signal. As a result, this model can be used to navigate the design space.

Based on the model analysis, the coefficients of main effects for the H₂O₂ dosage and the irradiated volume are highly significant, which means that these factors have a desirable effect for PS decolorization by the UV/H₂O₂ process. In other words, the decolorization efficiency increases with increasing hydrogen peroxide concentration and irradiated volume. The H₂O₂ dosage has the high coefficient of 16.05.

For the interaction effects only, the interaction between the irradiated volume and H₂O₂ dosage is significant, for the reason that the reaction zone between the solution and the UV source increases with increasing irradiated volume. In addition, more hydroxyl radicals are generated with increasing hydrogen peroxide concentration. Thus, the coefficients for the quadratic terms are shown to be significant.

The shape of the contour plot shows the nature and extent of the interactions between factors. It should be noted that a negligible effect appears as a circular contour plot when an elliptical contour plot indicates a prominent interaction (Qiu *et al.* 2014).

Whereas the response surface plot helps to visualize the tendency of each factor on the response, the observed surfaces were obtained by plotting the measured values of the

response against two factors, keeping the third one at its middle level. Figure 2 shows the three-dimensional surface and contour plots of the effect of hydrogen peroxide concentration and irradiated volume on PS decolorization when using the turbidity of 38.9 NTU.

Figure 2(a) shows that the decolorization efficiency increases considerably with both H₂O₂ dosage and irradiated volume. In other words, increasing both H₂O₂ dosage and irradiated volume from -1 to $+1$ has a positive effect for efficient decolorization. These results are supported by the contour plot given in Figure 2(b): as can be seen using 0.46 mM of H₂O₂ and 250 mL of irradiated volume, the decolorization efficiency was 63.18%. Increasing the H₂O₂ dosage and the irradiated volume to 2 mM and 500 mL respectively increases the decolorization efficiency to 92.68%.

Validation of the model

Analysis of variance (ANOVA) is an analytical technique that is used to identify the significance and the fit of the model and its parameters, using Fisher's F-test and Student's t-test. The ANOVA shows that the model is significant through both the p and F values. In general, larger F-values and smaller p -values indicate more significant coefficient terms (Qiu et al. 2014; Ismail & Khattab 2018). A summary of ANOVA test for the acquired model is shown in Table 4.

The coefficient of determination (R^2) is defined as the ratio of the sum of squares due to regression to the total sum of squares, while the adjusted R^2 value corrects the R^2 value for the sample size and for the number of terms in the model (Anuar et al. 2013).

It was found that the model evaluated in this study has a pred- R^2 of 0.9000 that is in reasonable agreement with the adj- R^2 of 0.9827 at a confidence level of 95%, which indicated a good fit for the model. In other words, the high values imply that more than 95% of experimental data can be explained by the model.

The model F-value of 102.25 implies the model is significant. There is only a 0.01% chance that a model F-value this large could occur due to noise. A p -value less than 0.05 indicates that the model is statistically significant and a value greater than 0.1000 indicates that the model is not significant (Qiu et al. 2014).

This model presents a very low probability value $p < 0.0001$, hence it is significant. Lack of fit is the variation due to the model inadequacy. The lack of fit was not significant, therefore there is no evidence to indicate that the model does not adequately explain the variation in the response.

The significance of each coefficient was determined by the F-value and the p -value. The data in Table 4 show that only the irradiated volume and the H₂O₂ dosage are the significant linear components ($p < 0.05$), with H₂O₂ dosage having the highest effect on the decolorization of

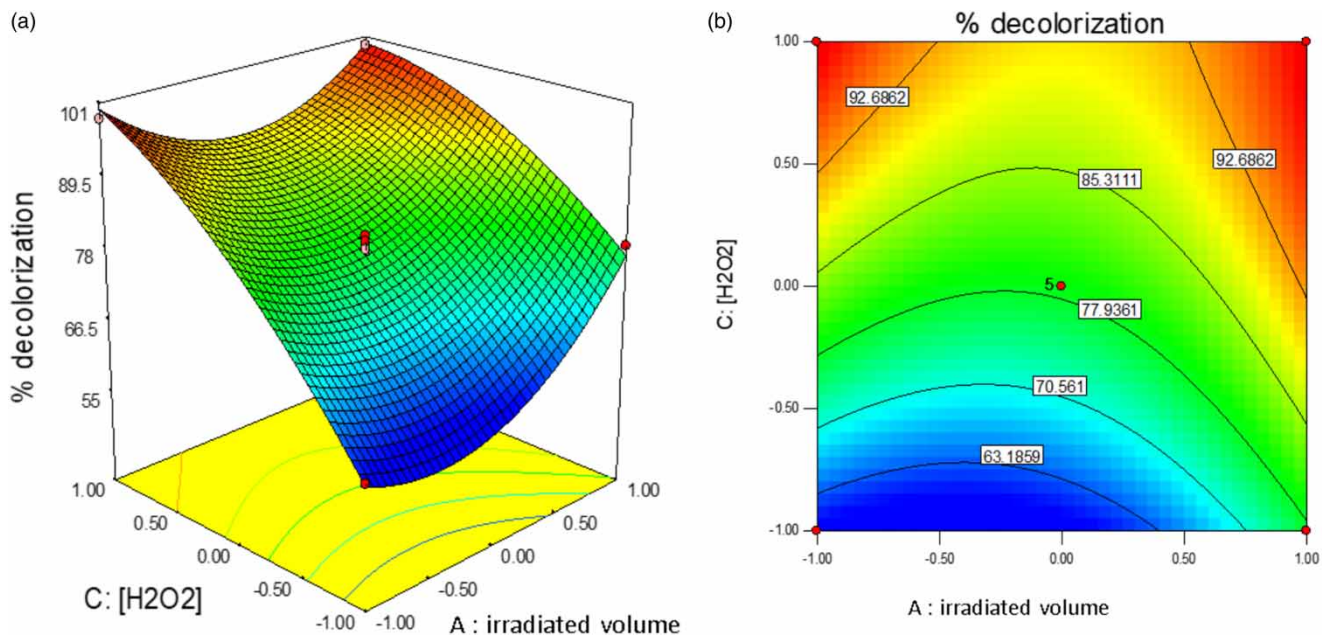


Figure 2 | Three-dimensional surface (a) and contour plots (b) of the effect of H₂O₂ dosage and irradiated volume on the efficient decolorization of PS dye.

Table 4 | ANOVA for the acquired model

Source	Sum of squares	Df	Mean square	F-value	p-value	Judgment
Model	3,010.88	9	334.54	102.25	<0.0001	Significant
A-irradiated volume	165.44	1	165.44	50.56	0.0002	Significant
B-turbidity	15.10	1	15.10	4.61	0.0688	
C-[H ₂ O ₂]	2,060.50	1	2,060.50	629.78	<0.0001	Significant
AB	0.027	1	0.027	8.321×10^{-5}	0.9299	
AC	86.21	1	86.21	26.35	0.0013	Significant
BC	5.24	1	5.24	1.60	0.2460	
A ²	415.51	1	415.51	127.00	<0.0001	Significant
B ²	169.32	1	169.32	51.75	0.0002	Significant
C ²	96.55	1	96.55	29.51	0.0010	Significant
Residual	22.90	7	3.27			
Lack of fit	18.54	3	6.18	5.67	0.0635	
Pure error	4.36	4	1.09			
Cor total	3,033.78	16				

PS aqueous solution followed by the irradiated volume. It should be noted that the turbidity has a *p*-value that exceeds 0.05, which confirms that is a non-significant factor.

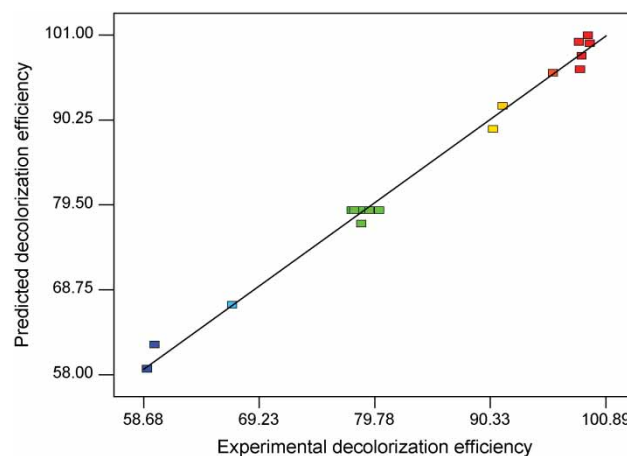
Model modification and optimized conditions

The final model can be improved by keeping the terms that are significant and deleting the others that are not very significant. The model obtained is shown in Equation (5):

$$\begin{aligned} \% \text{ decolorization} = & +78.77 + 4.55A - 1.37B + 16.05C \\ & - 4.64 AC + 9.93A^2 + 6.34 B^2 \\ & - 4.79 C^2 \end{aligned} \quad (5)$$

The pred-R² has increased from 0.9000 to 0.9459 and the adj-R² has changed from 0.9827 to 0.9835 which indicates a good fit of a regression model. Moreover, the *p*-value is still less than 0.0001, which means that the model is more significant. It is necessary to check the model accuracy by comparing the experimental and the predicted responses; it is clear from Figure 3 that the predicted and experimental responses have a linear relationship. The normal probability plot of residuals was generated to check the normality of the residuals. It represents the relationship between the normal probability (%) and the internally studentized residuals and it is shown in Figure 4.

It must be noted that the internally studentized residuals measures the standard deviations separating the experimental and predicted values while the residuals

**Figure 3** | Comparison of predicted and experimental PS dye decolorization.

determine how well the model satisfies the assumptions of ANOVA (Qiu et al. 2014). The residuals followed normal distribution. Moreover, the data points followed the fitted line fairly closely, with no outliers.

Optimization of PS decolorization by the UV/H₂O₂ process was performed using numerical optimization. To find the optimum conditions, it was necessary to fix the desired goals for each variable and response (i.e., none, maximum, minimum, target, or in range). The importance of goals (1–5) was chosen for the three variables as 3, while for the response it was set at 5.

In this study the decolorization efficiency of the PS aqueous solution was taken as ‘maximize’. The irradiated volume (250–500 mL), the turbidity (10.5–NTU–64 NTU)

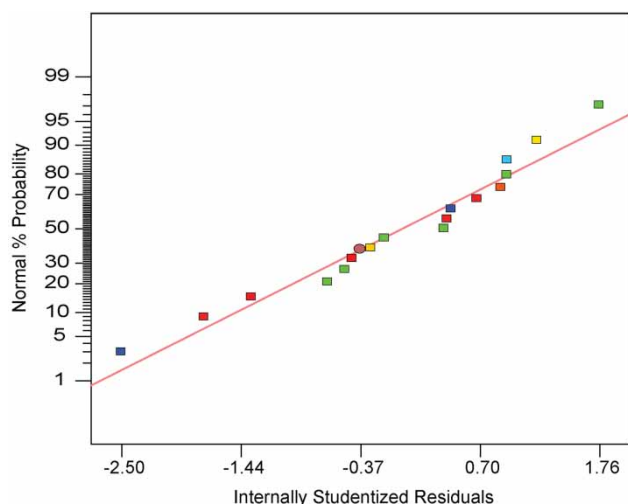


Figure 4 | Normal probability plots of residuals for PS dye decolorization.

and H₂O₂ dosage (0.46–2 mM) were chosen as ‘within the range’. These fixed goals were combined into one desirability function for the optimization process.

By applying the desirability function, the maximum decolorization of the PS aqueous solution was found under the optimum conditions of the irradiated volume of 257.59 mL, the turbidity of 13 NTU and 1.76 mM of H₂O₂.

CONCLUSION

Since the UV/H₂O₂ process can be efficiently used for decolorization of Ponceau S aqueous solution, there is an enhanced need to identify and optimize this process. As a result, the Box–Behnken design using response surface methodology was successfully applied to optimize some parameters that affect the process, i.e. irradiated volume, turbidity and H₂O₂ dosage.

A quadratic model was proposed, studied and evaluated to describe the relationship between the efficient decolorization versus the irradiated volume, the H₂O₂ dosage and the turbidity. The fitted model was significant with a coefficient of determination R² of 0.9459 and an adj-R² of 0.9835. Furthermore, the model has a *p*-value less than 0.0001, which means that the model was very significant.

In this study, among the parameters studied, H₂O₂ dosage was found to be the most prominent factor affecting the decolorization efficiency. The optimal conditions for decolorization of the PS aqueous solution by the UV/H₂O₂ process were an irradiated volume of 257.59, a turbidity of 13 NTU and an H₂O₂ dosage of 1.76 mM.

REFERENCES

- Akbar Babaei, A., Kakavandi, B., Raffee, M., Kalantarhormizi, F., Purkaram, I., Ahmadi, E. & Esmaeili, S. 2017 Comparative treatment of textile wastewater by adsorption, Fenton, UV-Fenton and US-Fenton using magnetic nanoparticles-functionalized carbon (MNPs@C). *J. Ind. Eng. Chem.* **56**, 163–174.
- Anuar, N., Faris Mohd Adnan, A., Saat, N., Aziz, N. & Mat Taha, R. 2013 Optimization of extraction parameters by using response surface methodology, purification, and identification of anthocyanin pigments in melastoma malabathricum fruit. *Sci. World. J.* **2013**, 10.
- Bharat, P. N., Naik, D. B. & Shrivastava, V. S. 2011 Photocatalytic degradation of hazardous Ponceau-S dye from industrial wastewater using nanosized niobium pentoxide with carbon. *Desalination* **269**, 276–283.
- Daneshgar, S., Vanrolleghem, P. A., Vaneekhaute, C., Buttafava, A. & Capodaglio, A. G. 2019 Optimization of P compounds recovery from aerobic sludge by chemical modeling and response surface methodology combination. *Sci. Total Environ.* **668**, 668–677.
- Das, A. & Mishra, S. 2017 Removal of textile dye reactive green-19 using bacterial consortium: process optimization using response surface methodology and kinetics study. *JECE* **5**, 612–627.
- Ferreira, S. L. C., Bruns, R. E., Ferreira, H. S., Matos, G. D., David, J. M., Brandão, G. C., da Silva, E. G. P., Portugal, L. A., dos Reis, P. S., Souza, A. S. & dos Santos, W. N. L. 2007 Box-Behnken design: an alternative for the optimization of analytical methods. *Anal. Chim. Acta* **597**, 179–186.
- Fu, J., Zhao, Y. & Wu, Q. 2007 Optimising photoelectrocatalytic oxidation of fulvic acid using response surface methodology. *J. Hazard. Mater.* **144**, 499–505.
- Ghodbane, H. & Hamdaoui, O. 2010 Decolorization of anthraquinonic dye, C.I. Acid Blue 25, in aqueous solution by direct UV irradiation, UV/H₂O₂ and UV/Fe(II) processes. *Chem. Eng. J.* **160**, 226–231.
- Ghosal, K., Ghosh, D. & Kumar Das, S. 2018 Preparation and evaluation of naringin-loaded polycaprolactone microspheres based oral suspension using Box-Behnken design. *J. Mol. Liq.* **256**, 49–57.
- Ismail, S. & Khattab, A. 2018 Optimization of proniosomal itraconazole formulation using Box Behnken design to enhance oral bioavailability. *J. Drug. Deliv. Sci. Technol.* **45**, 142–150.
- Kannan, N., Rajakumar, A. & Rengasamy, G. 2004 Optimisation of process parameters for adsorption of metal ions on straw carbon by using response surface methodology. *Environ. Technol.* **25**, 513–522.
- Kumar, S. S., Malyan, S. K., Kumar, A. & Bishnoi, N. R. 2016 Optimization of Fenton’s oxidation by Box-Behnken design of response surface methodology for landfill leachate. *J. Mater. Environ. Sci.* **12**, 4456–4466.
- Laftani, Y., Boussaoud, A., Chatib, B., Hachkar, M., Makhfouk, M. E. & Khayar, M. Comparison of advanced oxidation processes for degrading Ponceau S dye. Application of photo-Fenton process. *MJCCE* (in press).

- Lin, C.-C., Lin, H.-Y. & Hsu, L.-J. 2016 Degradation of ofloxacin using UV/H₂O₂ process in a large photoreactor. *Sep. Purif. Technol.* **168**, 57–61.
- Marathe Sunil, D. & Shrivastava Vinod, S. 2015 Photocatalytic removal of hazardous Ponceau S dye using Nanostructured Ni-doped TiO₂ thin film prepared by chemical method. *Appl. Nanosci.* **5**, 229–234.
- Masomboon, N., Chen, C.-W., Anotai, J. & Lu, M.-C. 2010 A statistical experimental design to determine o-toluidine degradation by the photo-Fenton process. *Chem. Eng. J.* **159**, 116–122.
- Muslim, M., Ahsan Habib, M., Selima Akhter Islam, T., Mohmmad Ibrahim Ismail, I. & Jafar Mahmood, A. 2013 Decolorization of diazo dye ponceau S by Fenton process. *Pak. J. Anal. Environ. Chem.* **14**, 44–50.
- Qiu, P., Cui, M., Kang, K., Park, B., Son, Y., Khim, E., Jang, M. & Khim, J. 2014 Application of Box–Behnken design with response surface methodology for modeling and optimizing ultrasonic oxidation of arsenite with H₂O₂. *Cen. Eur. J. Chem.* **12**, 164–172.
- Quadrado, R. F. N. & Fajardo, A. R. 2017 Fast decolorization of azo methyl orange via heterogeneous Fenton and Fenton-like reactions using alginate-Fe²⁺/Fe³⁺ films as catalysts. *Carbohydr. Polym.* **177**, 443–450.
- Ruotolo, L. A. M. & Gubulin, J. C. 2005 A factorial-design study of the variables affecting the electrochemical reduction of Cr(VI) at polyaniline-modified electrodes. *Chem. Eng. Trans.* **110**, 113–121.
- Sadoun, O., Rezgui, F. & G'Sell, C. 2018 Optimization of valsartan encapsulation in biodegradables polyesters using Box–Behnken design. *Mater. Sci. Eng. C* **90**, 189–197.
- Sathyamoorthy, N., Magharla, D., Chintamaneni, P. & Vankayalu, S. 2017 Optimization of paclitaxel loaded poly(ϵ -caprolactone) nanoparticles using Box Behnken design BENI-SEUF UNIV. *J. Appl. Sci.* **6**, 362–373.
- Velo-Gala, J. I., Sánchez-Polo, M. & Rivera-Utrilla, J. 2014 Comparative study of oxidative degradation of sodium diatrizoate in aqueous solution by H₂O₂/Fe²⁺, H₂O₂/Fe³⁺, Fe(VI) and UV, H₂O₂/UV, K₂S₂O₈/UV. *Chem. Eng. J.* **241**, 504–512.
- Vijayaraghavan, J., Sardhar Basha, S. J. & Jegan, J. 2013 A review on efficacious methods to decolorize reactive azo dye. *JUEE* **7**, 30–47.

First received 17 October 2019; accepted in revised form 11 December 2019. Available online 24 December 2019