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Modeling and optimizing of sonochemical degradation of Basic Blue 41 via response surface methodology

Research Article

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Abstract: The nanocatalyst-assisted sonodegradation of Basic Blue 41 (BB41) dye in aqueous medium was modeled and optimized using response surface method (RSM) based on Box-Behnken design. The studied variables included pH, initial dye concentration, H_2O_2 concentration and sonolysis time while each factor varied at three levels: Low level (-1), Medium level (0) and High level (+1). The ultrasound -assisted degradation was well described by developing quadratic model with correlation value squared (R^2) of 0.9114. Factor effects along with interaction effects were evaluated. The graphical optimization step was conducted to achieve the best experimental condition in dye removal. pH, H_2O_2 concentration and initial dye concentration of the reaction were investigated. It was recognized that at lower pH values the dye removal rate decreased. However, dye removal rate increased (82.5%) by increasing the concentration of H_2O_2 and by lowering the initial dye concentration.

Keywords: *Sonocatalytic • Optimization • Nanocatalysis • Box-Behnken • Response Surface Methodology*

1. Introduction

Rapidly growing world population and rise in side effects of human activity on the environment result in a need for its protection and in a requirement for sustainable development, These are the drivers of future technological progress. The strategies for environmental protection in industry usually include processes for waste treatment or new products development with less harmful effects on the environment [1]. Large amounts (~10 megatons) of different chemical dyes are produced annually world-wide. They are used extensively in the dye and printing industries, and 5–10% of the dyestuffs are lost in the industrial effluents [2]. Textile industry involves the use of a multitude of chemical products in different sub-sectors, leading to a wastewater with varying degrees of pollutant contents [3].

Azo dyes are the most important group of synthetic colorants (60–70%). They are characterized by the azo group (-N=N-), in association with aromatic systems and auxochromes (e.g. -OH, $-SO_3$). These groups can be combined with variety of dye molecules in order to make their bio-elimination from effluents a complex matter.

Colorant wastewaters are usually treated by conventional methods such as biological oxidation [4] or adsorption [5,6]. Recently, there has been a considerable interest in utilizing advanced wastewater treatment techniques such as photochemical oxidation [7,8], electrochemical technology [9,10] and Sonolysis [11-13].

Ultrasonic wave spans the frequencies of roughly 18 kHz – 10 MHz with associated acoustic wavelengths of 10 to 0.01 cm. The chemical effects of high intensity ultrasound arise from acoustic cavitation of liquids. This rapid formation, growth and implosive collapse of gas vacuoles generate short-lived (nanoseconds) localized “hot spots” whose peak temperatures and pressures reach 5000 K and 1000 atm [14].

High temperatures and pressures exist inside the collapsing gas bubbles, such as in the core. Under these conditions, the solvent vapor inside the bubble undergoes pyrolysis reactions at the interface between

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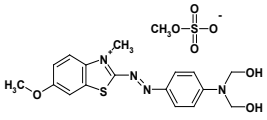
Table 1. Characteristics of Basic Blue 41

Name and color index (C.I)	Structure	λ max (nm)	Molecular weight
Basic Blue 41; 11105		617 nm	482.57 g mol^{-1}

the collapsing bubble and the bulk solvent. In aqueous solutions, the relative efficiency of non-volatile solutes to decompose thermally or to scavenge radicals formed in the hot spot depends on the solutes' ability to accumulate at the gas/solution interface of the growing micro bubble and in bulk solution at ambient temperature. Free radicals formed in the hot regions may diffuse to the bulk solution and react to yield products similar to those found in aqueous radiation chemistry. Substrates such as bisphenol [15], chlorinated hydrocarbons [16], various aromatic compounds [17,18] and dyes [19,20] are transformed into short-chain organic acids, inorganic ions, CO, CO_2, and H_2O as the final products.

Two reaction mechanisms have been previously proposed for sonodegradation. The first mechanism, pyrolysis, which occurs in cavitations bubbles, is expected to be the major reaction path for the degradation of polar compounds. The second mechanism is the generation of hydroxyl radicals in the cavitations which subsequently oxidize the polar organic compounds [14].

Wastewater treatments, like many other industrial processes, are optimized by using "one at a time" variation of treatment variable. This method assumes that various treatment parameters do not affect each other and that the response is only function of a single factor. An effective tool for optimizing the process in which several independent variables affect desired responses is the response surface methodology (RSM).

Response surface methods have been designed to include factors with more than three levels (Low level (-1), Medium level (0) and High level (+1)) in which quadratic models can be established. The main objective is to find a desirable location in the design space. This could be a maximum, a minimum or an area where the response is stable over a range of conditions. After establishing the goal, methods to measure responses need to be developed. Achieving a quantifiable response is one of the most important steps in a successful design of experiments (DOE). The most popular response surface methodologies are Central Composite, Box-Behnken and Doehlert designs. Box-Behnken designs require only 3 levels, coded as -1, 0, and +1 and are available for 3 to 10 factors. They are formed by combining two-level factorial designs with incomplete block designs. This procedure creates designs with desirable statistical properties and it involves only a fraction of the experiments required for a three-level factorial. Since there are only three levels, the quadratic model is the most suitable one. The number of required experiments for Box-Behnken design can be calculated according to the formula $N=k^2+k+c_p$, where k is the factor number and c_p is the replicate number of the central point. If viewed as cube, the quadratic model consists of a central point and the middle points of the edges [21-23].

The main goal of this work is to develop a proper mathematical model which describes the sonodegradation of Basic Blue 41 dye as a function of pH, initial dye concentration, H_2O_2 concentration and sonolysis time. With the model, one should also be able to optimize and compare the effect of various factors on the process yield. We explained degradation of Basic Blue 41 by sonochemical method in our previous work [12].

2. Experimental Procedure

2.1. Chemicals and equipment

Basic Blue 41 [1-Amino-6-methoxybenzotiazole] was purchased from Fluka chemical company and used without further purification (Table 1). All other chemicals were of analytical grade and purchased from Merck chemical company. The nanocatalyst used in this experiment was titanium dioxide (TiO_2) with initial average particle size ~100 nm, 97% purity and 80:20 anatase to rutile ratio. A fixed amount of nanocatalyst was applied (0.2 grams for 1 L of solution) in all experiments.

The ultrasound irradiation was done using an

Table 2. Experimental factors with their actual and coded levels

Numerical factors	Low level (-1)	Medium level (0)	High level (+1)
pH / A	4	6.25	8.5
Initial dye conc.(mg L^{-1}) / B	15	30	45
H_2O_2 conc.(mg L^{-1}) / C	0	250	500
Sonolysis time (min) / D	0	90	180

Table 3. Box-Behnken design with actual and coded factor levels

Run	pH	Initial dye conc. (mg L^{-1})	H_2O_2 conc. (mg L^{-1})	Contact time (min)	Percentage BB41 removal (%)
1	6.25 (0)	30 (0)	250(0)	90 (0)	38.1
2	4 (-1)	30 (0)	250 (0)	180 (+1)	35.8
3	8.5 (+1)	0 (0)	250 (0)	0 (-1)	0
4	6.25 (0)	15 (-1)	0 (-1)	90 (0)	9
5	4 (-1)	30 (0)	500 (+1)	90 (0)	42
6	4 (-1)	30 (0)	0 (-1)	90 (0)	4.2
7	6.25(0)	30 (0)	250(0)	90 (0)	38.3
8	6.25 (0)	15 (-1)	250(0)	180 (+1)	63
9	6.25 (0)	45 (+1)	250(0)	0 (-1)	0
10	4 (-1)	30 (0)	250(0)	0 (-1)	0
11	6.25 (0)	45 (+1)	250(0)	180 (+1)	33.5
12	8.5 (+1)	45 (+1)	250(0)	90 (0)	73.3
13	6.25 (0)	15 (-1)	250(0)	0 (-1)	0
14	6.25 (0)	30(0)	0 (-1)	180 (+1)	18
15	4 (-1)	45 (+1)	250(0)	90 (0)	32
16	8.5	15 (-1)	250(0)	90 (0)	79.8
17	6.25 (0)	15 (-1)	500 (+1)	90 (0)	71.1
18	8.5 (+1)	30 (0)	500 (+1)	90 (0)	81.4
19	8.5 (+1)	30 (0)	0 (-1)	90 (0)	16
20	6.25 (0)	30(0)	250(0)	90 (0)	38.5
21	6.25 (0)	30 (0)	250(0)	90 (0)	38.1
22	6.25 (0)	45 (+1)	0 (-1)	90 (0)	6.5
23	4 (-1)	15 (-1)	250(0)	90 (0)	51.6
24	6.25 (0)	30 (0)	500 (+1)	180 (+1)	42
25	6.25 (0)	30 (0)	500 (+1)	0 (-1)	0
26	6.25 (0)	30 (0)	0 (-1)	0 (-1)	0
27	8.5 (+1)	30(0)	250(0)	180 (+1)	82.1
28	6.25 (0)	45 (+1)	500 (+1)	90 (0)	36
29	6.25 (0)	30 (0)	250(0)	90 (0)	25

ultrasonic bath (Bundelin Electronic RK 255H, Germany) at 35 KHz and 160 W. The 150 mL cylindrical reactor was double-surrounded. The temperature was controlled with a thermostatic bath and water circulation system (LAUDA, RE 104, Germany) and aqueous temperature of 25+1˚C for 180 min. [12]. Spectrophotometric measurements were conducted by a PDA-Multispect Shimadzu 1501 spectrophotometer. All pH measurements were performed using a digital pH meter (Hack).

2.2. Procedure

The four variables studied were pH, initial dye concentration, H_2O_2 concentration and contact time (Table 2). Experimental matrix based on Box-Behnken design containing 29 trials was planned by Design-Expert-v.7 (STATE-EASE, Corp., Minnesota) and it is described in Table 3. The percentage of dye removal was considered a quantifiable response. The average of three replicates was used for each datum. The definitions of statistical terms are explained in index. Dye decomposition was performed using 100 mL aqueous BB41 solution mixed at appropriate concentrations under constant stirring. The acidity levels were adjusted using dilute HCl and NaOH solutions.

3. Results and Discussion

3.1. Box-Behnken design method

A Box-Behnken design containing 29 experiments was studied. The designs have fewer runs compared to 3-level a factorial design which makes the Box-Behnken method more economic, convenient and time-efficient.

Four factors under investigation were designated as A, B, C and D. The experimental conditions were selected based on our previous work in which “one factor at a time” method was utilized to optimize the BB41 removal [12]. The design levels in terms of coded and actual forms and relevant response values are shown in Table 3. Percentage of BB41 removal was being recorded and analyzed.

Results revealed that the best dye removal was achieved at alkaline medium (pH 8.5) using 30 mg L^{-1} initial dye concentration, 250 mg L^{-1} H_2O_2 concentration and contact time of 180 min. Box-Behnken design methodology provides the evaluation of major and interaction effects. The major effect refers to the effect caused by varied factor, while the interaction effect is related to the case in which the effect of one factor is dependent on the value of another factor [24]. The significant factors in the regression model can be estimated via analysis of variance.

Table 4. ANOVA for response surface quadratic model

Source	Sum of squares	DF	Mean square	F value	Prob>F
Model	18934.15	1	1352.44	10.28	<0.0001
A	2324.08	1	2324.08	17.67	0.0009
B	723.85	1	723.85	5.50	0.0342
C	3989.45	1	3989.45	30.34	<0.0001
D	6274.61	1	6274.61	47.71	<0.0001
A^2	947.73	1	947.73	7.21	0.0178
B^2	298.83	1	298.83	2.27	0.1539
C^2	547.67	1	547.67	4.37	0.0553
D^2	1637.27	1	1637.27	12.45	0.0033
AB	42.90	1	42.90	0.33	0.5769
AC	190.44	1	190.44	1.45	0.2488
AD	535.92	1	535.92	4.08	0.0631
BC	265.69	1	265.69	2.02	0.1771
BD	217.56	1	217.56	1.65	0.2192
CD	144.00	1	144.00	1.09	0.3131
Residual	1841.16	14	131.51		0.0711
Lack of fit	1700.60	10	170.06	4.84	
Pure error	140.56	4	35.14		
Cor total	20775.31	28			

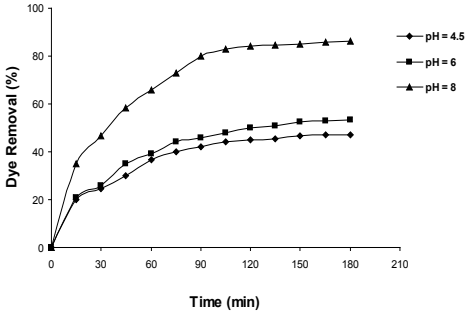


Figure 1. Effect of pH: initial dye concentration: 15 mg L^{-1}, US: 35 kHz, H_2O_2 concentration: 250 mg L^{-1} and Nano TiO_2:0.2 g L^{-1}.

Analysis of variance (ANOVA) for individual factors and their interactions is displayed in Table 4. ANOVA showed the best fit for the data with a quadratic model (values of Prob>F less than 0.0001). The model F-value of 10.28 implied that the model was significant. There is only a 0.01% chance that a large value for "model F-value" could be due to noise. The lack of a fit F-value of 4.84 showed that the lack of fit was not significant relative to the error.

Values of "Prob>F" less than 0.0500 indicated that the model terms were significant. In this case of A, B, C and D, the second-order main effects (A^2 and D^2) were significant model terms. Values greater than 0.1000 indicated that the model terms were not significant. F-values suggested that sonolysis time were the most determinant factors in the sonodegradation process, H_2O_2 concentration was the second important factor, whereas pH and initial dye concentration were rated last. The correlation coefficient for obtained model was found to be 0.9114, which demonstrated that the quadratic model fit the data well.

"Adeq Precision" measured the signal to noise ratio with a desirable ratio to be greater than 4. The ratio of 12.726 indicated an adequate signal, thus our model could be used to navigate the design space.

The final regression equation expressing the dependence of percentage BB41 removal (R) on practical variables in terms of coded factors was obtained as:

$$R = +35.60 + 13.92A - 7.77B + 18.23C + 22.87D + 12.09A^2 + 6.79B^2 - 9.41C^2 - 15.89D^2 + 3.27AB + 6.90AC + 11.57AD - 8.15BC - 7.37BD + 6.00CD \quad (1)$$

where A is pH, B is initial dye concentration, C is hydrogen peroxide concentration and D is the sonolysis time. Term coefficients in Eq. 1 demonstrate that in the ANOVA results the effects of each variable can be directly attributed to their coefficient values. Moreover, more effective factors possess higher mathematical coefficients. Since the equation is represented in terms of coded factors, the relative effect of each variable can be evaluated by comparing the absolute value of its coefficient and its algebric sign [25]. Comparison of the factors showed that the term D, which is indicative of the contact time, had larger algebric coefficient, as could be understood from ANOVA results. In contrast, factor B (dye concentration) had the lowest influence on the percentage dye removal.

As shown in Eq. 1, the effect of all single factors resulted in a positive algebric sign, with the exception of the factor B, which indicated that increasing the initial dye concentration would result in decrease of response (BB41 removal). Other variables had a direct relation with the dye removal amount. Second order main interactions (C^2 and D^2) along with BC and BD interaction terms had a negative algebric sign which indicated a negative effect on dye removal.

3.2. Effect of contact time

According to the regression equation, dye removal amount increased with an increase in contact time and hydrogen peroxide concentration. More contact time allowed reacting dye molecules to be decomposed. The sonolysis time should be considered carefully as the excess of sonication is believed to reduce the yield due to the production of hydroxyl radicals from aqueous solvent. In this research, optimum contact time was found to be 180 min. After this period, no increase in dye removal was observed, owing it to the established equilibrium conditions.

3.3. Effect of pH

ANOVA data revealed that more dye removal could be achieved in alkaline media compared to acidic conditions [12]. In the presence of various hydrogen peroxide concentrations, maximum BB41 removal was estimated at 51.6% at pH 4, while maximum amounts of 71.1% and 82.1% were attained at pH values of 6.25 and 8.5, respectively. The effect of pH in the presence of TiO_2 for degradation of dyes using advanced oxidation processes has been evaluated by others. The point of zero charge occured at pH 6.8 for TiO_2 particles. As pH of the system increased, the number of negatively charged surface sites on the TiO_2 favored the adsorption of dye cations due to the electrostatic attraction, which resulted in the increase of the dye degradation. This could have been casued by the presence of hydroxyl ions at pH 8 which would cause more hydroxyl radical formation, and hence increase in radical attack to the dye molecules. The effect of varied initial pH values on the decolorization of BB41 during the sonocatalysis is presented in Fig. 1.

3.4. Effect of H_2O_2 concentration

Presence of hydrogen peroxide plays a major role in the dye degradation process in advanced oxidation process technique and it depends on H_2O_2 concentration and the type of reductants [12]. Formation of hydroxyl radicals by hydrogen peroxide occurs via two mechanisms. Firstly, the reduction of H_2O_2 at the conduction band

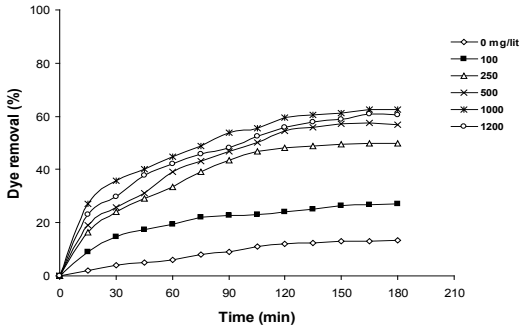


Figure 2. Effect of H_2O_2 concentration: pH: 4.5, initial dye concentration: 15 mg L^{-1}, US: 35 kHz and Nano TiO_2 : 0.2g L^{-1}.

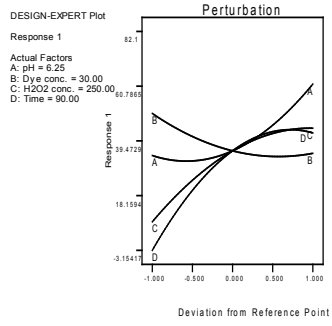


Figure 3. The perturbation plot of percentage BB41 removal against four variables.

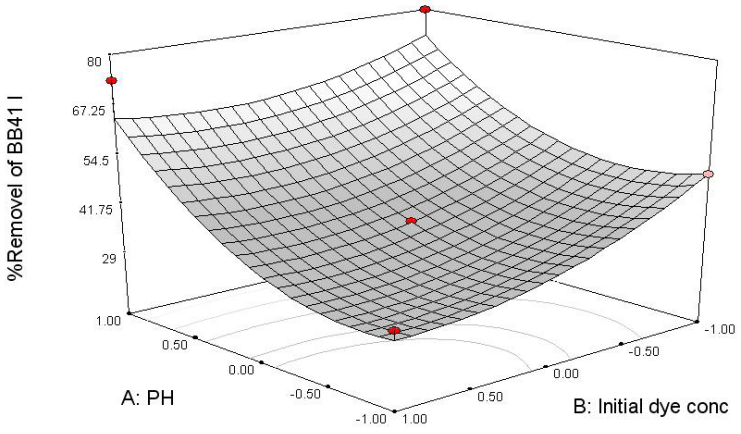
would produce hydroxyl radicals. Secondly, the self-decomposition as a result of ultrasound irradiation would also produce hydroxyl radicals. The degradation rate of the BB41 increases with increasing of H_2O_2 concentration at optimal conditions. Plots of percentage dye removal *versus* time implying the H_2O_2 effect are shown in Fig. 2. It should be noted that the effect of ultrasonic irradiation combined with H_2O_2 is observed only when free radical attack is the controlling mechanism.

3.5. Effect of initial BB41 concentration

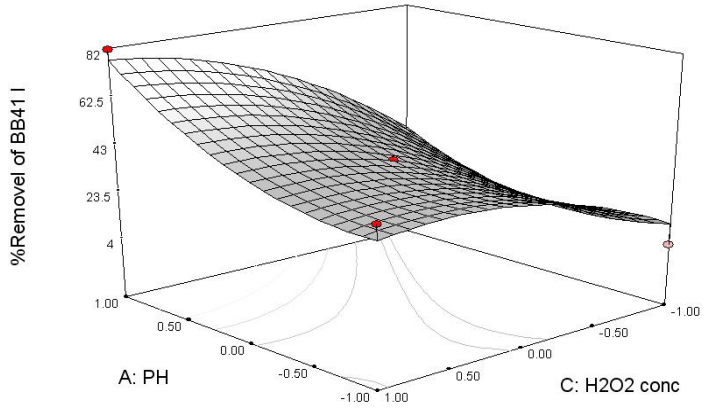
Decolorization efficiency decreased with higher BB41 concentrations. This phenomenon may be related to the interferences caused by dye molecules which inhibited degradation process. This effect could be increased during the formation of more intermediate species.

3.6. Perturbation curve

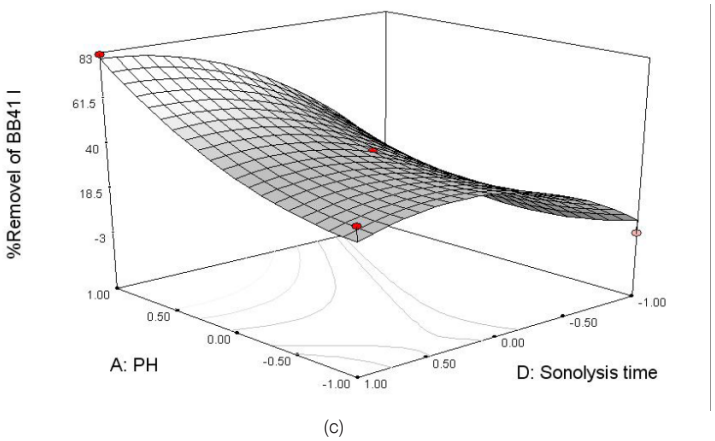
The results from plot of perturbation response (percentage dye removal) against studied factors (Fig. 3) provided supporting evidence of the importance of contact time (factor D) and H_2O_2 concentration (factor C) in the dye removal process. Fig. 4 shows dye removal amount as each variable moves away from the selected reference, with the remaining factors held constant in



(a)

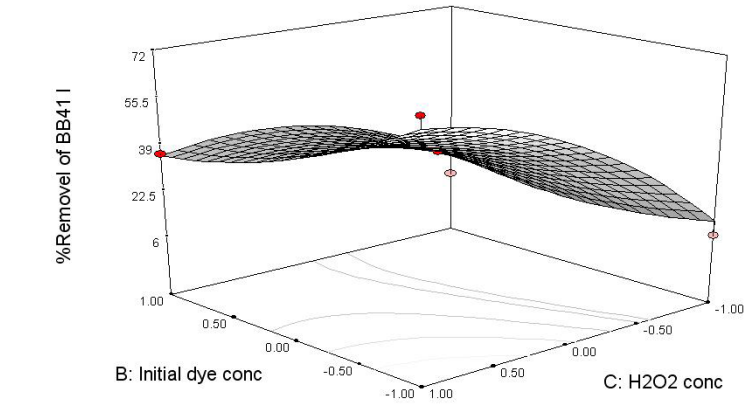


(b)

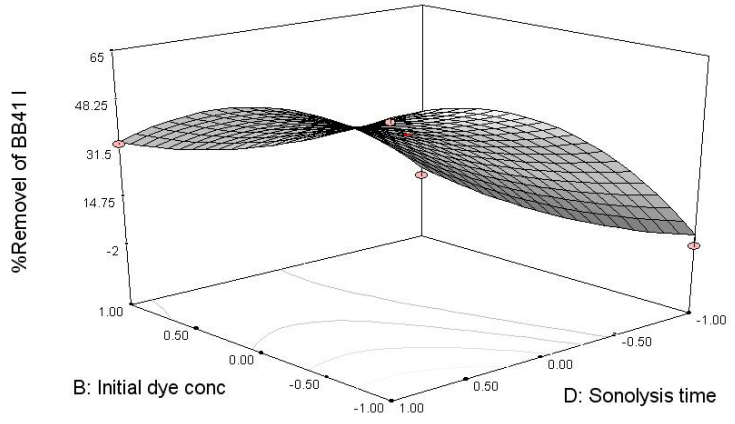


(c)

Figure 4. Second order response surface plot in the sonochemical removal of Basic Blue 41 dye. Dependence of response on various factor A interactions (AB, AC and AD). The other two factors were set at their middle-level values.



(a)



(b)

Figure 5. Second order response surface plot in the sonochemical removal of Basic Blue 41 dye. Dependence of the response on various factor B interactions (BC and BD). The other two factors were set at their middle-level values.

the middle of the design space (the coded zero level). Initial dye concentration had the lowest effect while the effect of pH became more pronounced by moving from middle towards higher levels (more alkaline media). The reciprocal slope of perturbation curve for factor B (dye concentration) confirmed the decrease in dye removal through increasing the dye concentration up to the 45 mg L^{-1}. The reciprocal slope of perturbation curve for factors of A and C (pH and H_2O_2 concentration) confirmed the that the decrease of the dye removal is caused by increasing the pH and H_2O_2 concentration up to the 8.5 and 500 mg L^{-1} respectively.

3.7. Response surface three-dimensional plots

The use of three-dimensional plots of the regression model was used as an appropriate tool for the graphical interpretation of the interactions. Three-dimensional response surface was generated to study the interaction among four variables and to visualize the combined effects of these variables on the response of dye removal. Factors giving quadratic and interaction terms in the fitted model were selected for the axes of the response surface plots to account for curvature of the surfaces, while keeping other factors constant at their medium levels.

It was observed that the maximum dye removal could be achieved using 15 mg L^{-1} BB41 aqueous solution and pH 8.5, as shown in Fig. 4a. Decreasing the related factors would lead to the less dye removal. In the case of pH, the variation of response was more significant. Fig. 4b showed the maximum dye removal at pH 8.5 and maximum amount of hydrogen peroxide concentration. Fig. 4c demonstrated that the most dye removal could be attained in 180 min. at pH 8.5. Decreasing the factor levels would cause less dye removal. According

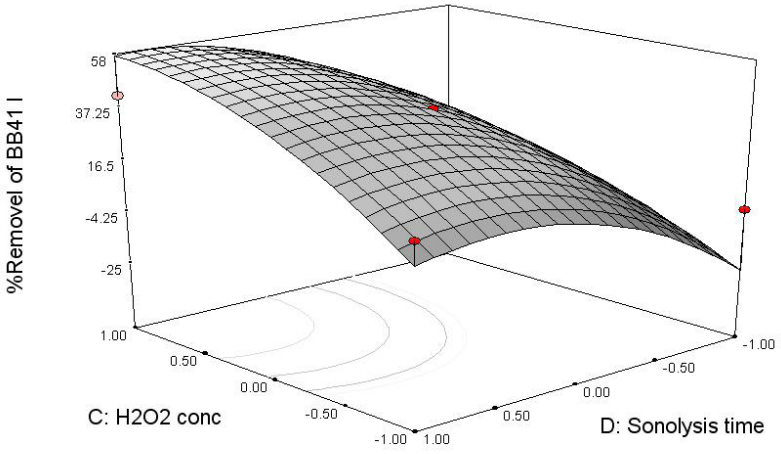


Figure 6. Second order response surface plot in the sonochemical removal of Basic Blue 41 dye. Dependence of response on the CD interaction. The other two factors were set at their middle-level values.

to Fig. 5a, use of 15 mg L^{-1} dye solution along with 500 mg L^{-1} of H_2O_2 concentration provided the maximum response, and variation of hydrogen peroxide reduced the dye removal amount from 67.125% to 7.94%. Fig. 5b shows that the maximum dye removal of 64.54% could be attained again using 15 mg L^{-1} dye solution and 180 min reaction time. However, the lowest reached response in two term interactions was achieved in conditions depicted in Fig. 6 in which 57.532% of BB41 was removed from solution using maximum amount of H_2O_2 and 180 min contact time.

The above plots were obtained in the conditions where other factors were held constant at their medium levels and variation of these factor levels could result in possibly higher responses.

4. Conclusions

The effect of variables such as pH, dye concentration, H_2O_2 concentration and sonolysis time on nanocatalytic sonodegradation of Basic Blue 41 can be well described by Quadratic model using response surface methodology based on Box-Behnken design. Obrained high correlation coefficient (R^2=0.9114) proved the fitness of the selected model to analyze the experimental data. The degradation process was strongly influenced by sonication time and H_2O_2 concentration, as well aso by pH, while initial dye concentration had lower effect (sonication time > H_2O_2 concentration > pH > dye concentration). Significant interaction terms were found to be A^2 and D^2. Classical optimization studies use the "one factor at a time" approach, in which only one factor is variable at a time while all others are kept constant. This approach is time-consuming and expensive. In addition, possible interaction effects between variables can not be evaluated and misleading conclusions may be drawn. The response surface method (RSM) overcomes these difficulties, as it allows accounting for possible interaction effects among variables. If adequately used, this powerful tool can provide the optimal conditions that improve the process. ANOVA optimization reveals that the optimum dye removal condition can be achieved at pH 8.5 during 180 min contact time, using maximum amount (up to 500 mg L^{-1}) of hydrogen peroxide and 30 mg L^{-1} of Basic Blue 41. The results from three-dimensional surface plots and perturbation curves confirm the ANOVA data.

Appendix

Statistical term	Definition
Sum of squares	The sum of the squared distances from the mean due to the variation in average response when a factor shifts from low to high level
Model sum of squares	Total of the sum of squares for the model terms
Residual sum of squares	Total of the sum of squares of all the terms not included in the model
Lack of fit sum of squares	Residual sum of squares after subtracting the pure error sum of squares
Pure error sum of squares	Some of squares from replicated points

Degrees of freedom (DF)	The number of independent comparisons available to estimate a parameter. Usually the number of model parameters minus 1
Model DF	Number of model terms including minus 1
Residual DF	Adjusted total DF minus the model DF
Lack of fit DF	Amount of information available after accounting for blocking model terms and pure error
Pure error DF	Amount of information available from replicated points
Lack of fit	The result of experimentation should be a model which will adequately predict the response within the design space. The variation between the model prediction and the design points is defined as lack of fit.
Pure error	The normal variation in the response which appears when an experiment is repeated
Mean square	The sum of squares divided by the degrees of freedom analogous to variance
F value	The ratio of model mean square to the appropriate error mean square
Model F value	A test for comparing model variance with residual variance
Lack of fit F value	Test for comparing lack of fit variance with pure error variance
Prob>F (probability of a larger F value)	If the F value (the ratio of variances) lies near the tail of the <F> distribution then the probality of a large F is small and the variance ratio is supposed to be significant
Core. Total (corrected total)	The total sum of squares corrected for the mean. It is the sum of squared differences between the individual observations and overall average

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