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Design of Experiment As a Tool for Optimizing the Batch Condition of Methylene Blue Degradation Using Palladium Based Catalyst

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Abstract: ***Present work deals with the application of Design of Experiment to study the effect of operational parameter on the degradation of methylene blue dye using palladium based catalyst and H_2O_2. Design of Experiment is used to optimize the degradation of methylene blue. Four factors and two level is used which give 16 experimental set (2^4). Fit values were found to be good as the experimental values (R=94.1% and Adj R=91.9). Graphical responses were used to locate the optimum points.***

Keywords: Immobilised Pd-CoO, Design of Experiment, Factorial Design, methylene Blue Degradation, Waste-Water Treatment

1. Introduction

Due to the Continuing processes of industrialization and urbanization population growth, deforestation, and pollution are exerting pressure on the depleting freshwater resources in many parts of the world. Clean and non-polluted water is one of the basic requirements for all living organisms including human beings. Colour is one of the most important hazards for the water bodies [1-3].Advanced oxidation processes (AOPs) is one of the most efficient, cheap, and eco-friendly in the degradation of any kind of toxic pollutants. AOPs generate hydroxyl radical, a strong oxidant, which can completely degrade or mineralize the pollutants no selectively into harmless products. For the batch process of degradation, the main parameter to be considered is time, pH, H_2O_2, and Concentration of dye (ppm). So we need to investigate interaction between the parameters and their effects [4-7].

2. Experimental

2.1 Synthesis of Pd-CoO

1.0gm of cobalt sulphate is mixed in 70ml dist. Water. Then in another beaker add 0.1gm of palladium acetate is mixed 30ml ethanol[8]. Then both the solutions are mixed together and then 1gm pvp is add to the solution mixture then it is sonicated for 30 mins maintaining sonication parameter as

1. Pulse = 05on/02off
2. Probe temperature = 46c
3. Set point temperature = 70c
4. Amplitude = 30%
5. Timer = 30 mins.

After 30 mins of sonication the solution obtained is dried at 100c in hot air oven to get dry powder. Then further it is immobilized over the surface and then is used for carrying out degradation.

2.2. Degradation of methylene blue

A certain volume of Methylene Blue is taken in beaker and the slab of immobilised catalyst Pd-CoO is dipped in that and certain amount H2O2 is add and kept it for few hours. Definite volumes of resulting solution at fixed intervals of time are taken out and the samples are analysed for absorbance using UV-spectrophotometer.Percentage Degradation is then calculated according to the following formula

% deg = (Ai - Af) x 100 /Ai

Where,

% deg = Percentage Degradation
Ai = Initial Absorbance
Af = Final Absorbance

2.3. Statistical design of Experiments

2.3.1 Full Factorial Design

Table 1 and table 2 given below explain about the various factor and their level. 16 no. of experiment with different factor and different level which is obtained using the Minitab software 16.

Factors: 4 Base Designs: 4, 16
Runs: 16 Replicates 1
Blocks: 1 Center pts (total): 0

Table 1: Full 2^4 factorial designs

Factor	*Levels*	
	+1	-1
Time (hour)	8	4
Concentration (PPM)	100	50
PH	7.5	4.5
H_2O_2 Dosage (ml)	40	10

Table 2: Optimization of Methylene blue using Pd-CoO

C1	C2	C3	C4	C5	C6	C7	C8	C9
StdOrder	RunOrder	CenterPt	Blocks	TIME	CONCENTRATION	pH	H2O2 DOSAGE	%DEGRATATION
7	1	1	1	-1	1	1	-1	49.740
15	2	1	1	-1	1	1	1	67.048
12	3	1	1	1	1	-1	1	100.000
5	4	1	1	-1	-1	1	-1	62.660
4	5	1	1	1	1	-1	-1	72.467
8	6	1	1	1	1	1	-1	61.730
16	7	1	1	1	1	1	1	86.000
14	8	1	1	1	-1	1	1	100.000
11	9	1	1	-1	1	-1	1	74.000
2	10	1	1	1	-1	-1	-1	90.493
6	11	1	1	1	-1	1	-1	68.020
9	12	1	1	-1	-1	-1	1	86.020
10	13	1	1	1	-1	-1	1	100.000
13	14	1	1	-1	-1	1	1	81.500
1	15	1	1	-1	-1	-1	-1	62.595
3	16	1	1	-1	1	-1	-1	51.180

3. Results and Discussion

3.1. Screening of factors for the degradation of methylene blue

Dye degradation by the use of catalyst depends upon several factors such as time, concentration of dye, acidic and basic pH, dosage of H_2O_2. Optimization of these variable are very tedious and all the factors cannot be optimized and we can't get interaction of different variables, so in order to overcome this problem the statistical design of experiment can be used to optimize the system. In this work we are screening the factor like time, concentration, pH and dosage of H_2O_2. Table 2 shows the percentage degradation of methylene blue. The Minitab Software 16 has been used to obtain the effect and optimize the experiment.

Table 3: Factorial Fit: %DEGRATATION versus TIME, PPM, pH, H2O2 DOSAGE
Estimated Effects and Coefficients for % DEGRATATION (Coded Units)

Term	*Effect*	*Coef*
Constant		75.841
time	17.996	-5.570
PPM	-11.140	-3.754
pH	-7.507	10.980
H2o2 Dosage	21.960	1.750
Time*PPM	1.561	0.781
Time*pH	-4.295	-2.148
Time*h2o2 Dosage	1.362	0.681
PPM*pH	-0.775	-.388
PPM*h2o2 Dosage	1.022	.511
pH*h2o2 Dosage	1.139	.570
Time*PPM*pH	.209	.105
Time*PPM*h2o2 Dosage	1.557	.778
Time*pH*h2o2 Dosage	3.663	1.832

Table 4: Analysis of Variance for %degradation (coded units)

Source	*DF*	*Seq SS*	*Adj SS*	*Adj MS*
Main Effects	4	3946.3	3946.3	986.57
TIME	1	1295.41	1295.41	1295.41
PPM	1	496.43	496.43	496.43
pH	1	225.43	225.43	225.43
H2O2 DOSAGE	1	1929.03	1929.03	1929.03
2-Way Interactions	6	102.75	102.75	17.12
TIME*PPM	1	9.75	9.75	9.75
TIME*pH	1	73.8	73.8	73.8
TIME*H2O2 DOSAGE	1	7.42	7.42	7.42
PPM*pH	1	2.4	2.4	2.4
PPM*H2O2 DOSAGE	1	4.18	4.18	4.18
pH*H2O2 DOSAGE	1	5.19	5.19	5.19
3-Way Interactions	4	107.98	107.98	27
TIME*PPM*pH	1	0.17	0.17	0.17
TIME*PPM*H2O2 DOSAGE	1	9.69	9.69	9.69
TIME*pH*H2O2 DOSAGE	1	53.68	53.68	53.68
PPM*pH*H2O2 DOSAGE	1	44.43	44.43	44.43
4-Way Interactions	1	38.47	38.47	38.47
TIME*PPM*pH*H2O2 DOSAGE	1	38.47	38.47	38.47
Residual Error	0			
Total	15	4195.5		

Estimated effects and coefficient for %degradation in coded units.

Table 5: Regression Analysis: %DEGRATATION versus TIME, PPM, pH, H2O2 DOSAGE

Predictor	*Coef*	*SE Coef*	*T*	*P*
Constant	75.841	1.19	63.74	0
TIME	8.998	1.19	7.56	0
PPM	***5.57***	1.19	4.68	0.1
pH	*3.754*	1.19	***3.15***	0.9
H2O2 DOSAGE	10.98	1.19	9.23	0

S = 4.75965
R-Sq = 94.1%
R-Sq (adj) = 91.9%
PRESS = 527.228
R-Sq (pred) = 87.43%

Table 6: Analysis of Variance

Source	DF	SS	MS	F	P
Regression	4	3946.3	986.57	43.6	0
Residual Error	11249.2	22.65			
Total	15	4195.5			

3.2. Optimization for degradation of methylene blue %degradation

After doing the screening of all the factor for the %degradation of methylene blue it has been found that there are certain factor which are playing crucial role in degradation as shown in the fig 1 shows that there are certain factor which are significant and some are not significant and fig 2 show how different factors are interacting among themselves. And fig 3 and 4 shows how the different factors are working under the given level. The %degradation of methylene blue using Pd-CoO catalyst follows equation which is given below

The regression equation is
%DEGRATATION = 75.8 + 9.00 TIME - 5.57 PPM - 3.75
pH + 11.0 H2O2 DOSAGE

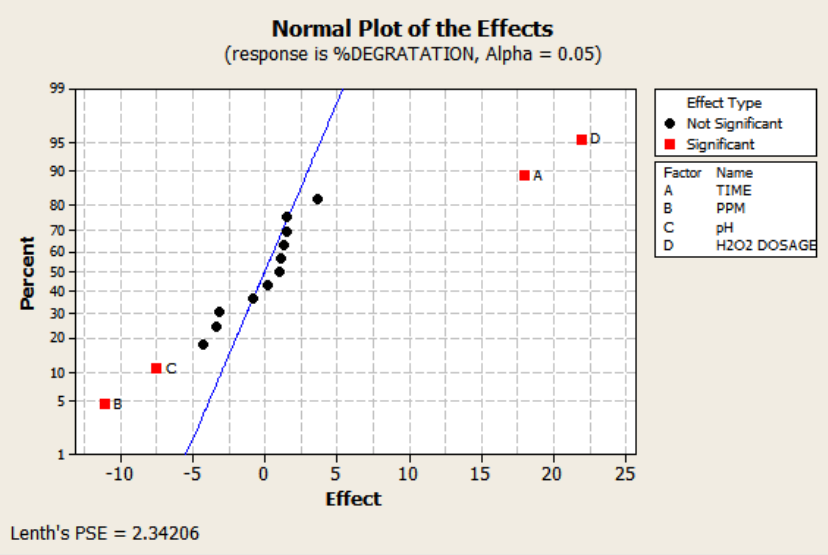

Figure 1: shows the normal plot of effect on %degradation.

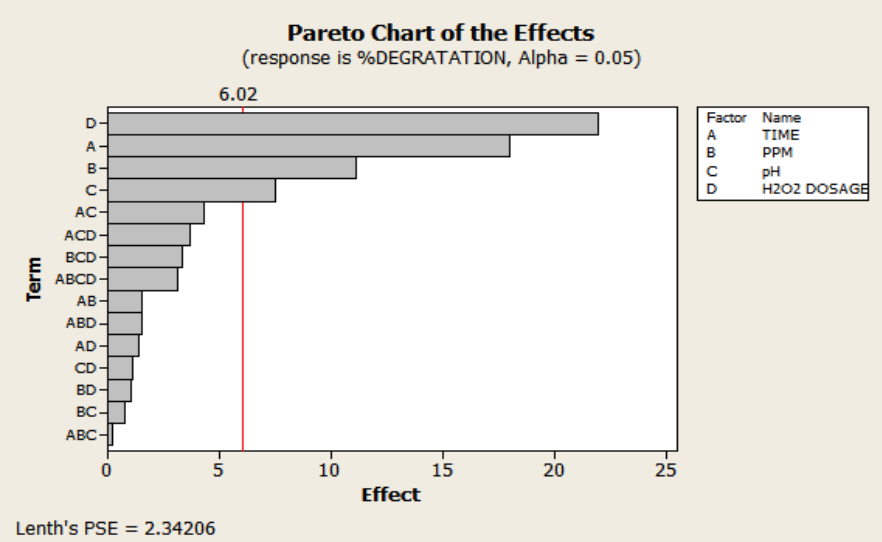

Figure 2: shows the pareto chart of the effect on %degradation.

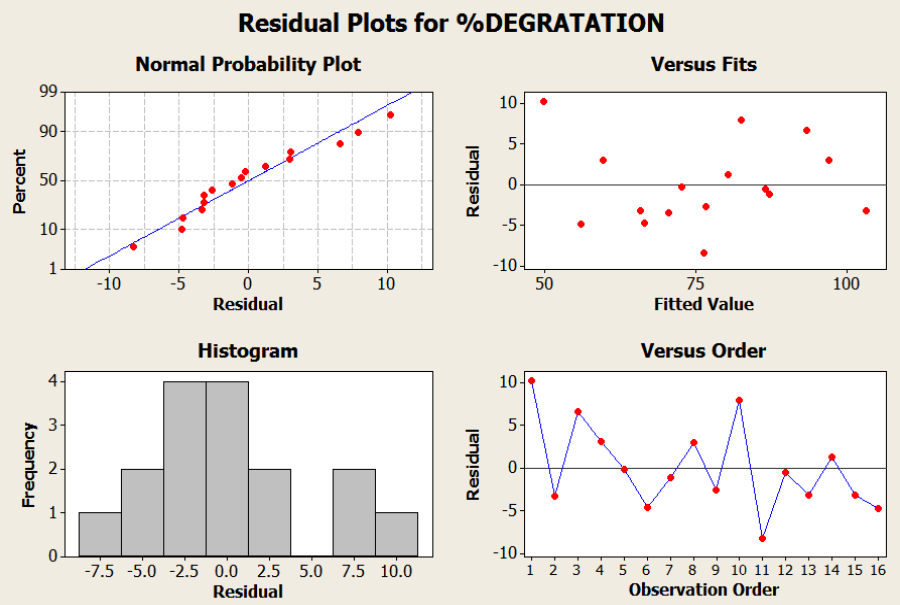

Figure 3: shows the Residual plot on the effect on %degradation

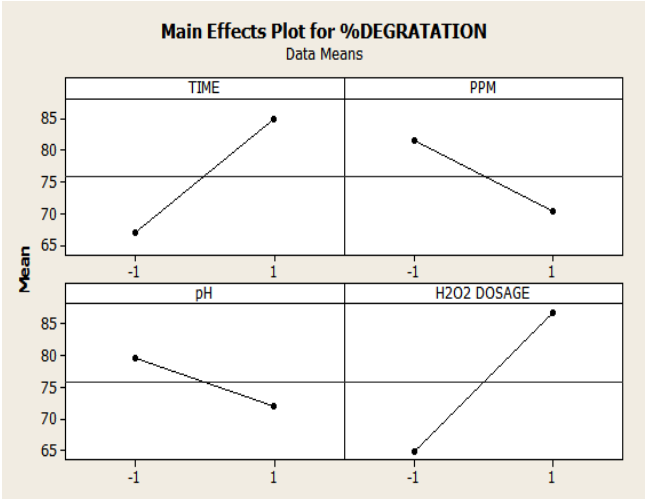

Figure 4: shows the main effect of different factor on %degradation

4. Conclusions

Full 2^4 factorial designs were applied for the screening of the factor which would influence the overall optimization of methylene blue degradation with the Pd-CoO catalyst. The optimum condition for the different factor is obtained by the regression equation is %DEGRADATATION = 75.8 + 9.00 TIME - 5.57 PPM - 3.75 pH + 11.0 H2O2 DOSAGE. This equation can be validated by substituting the different values of the time, ppm, pH andH_2O_2 dosage. So by this equation we can find the best optimum condition. And hence we can also enhance the rate of %degradation by improving the factors and level range so that we can get best optimum result.

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