NCNE-2020 Special Issue

J. Indian Chem. Soc.,
Vol. 97, July 2020, pp. 1066-1071

Significance of hybrid central composite design for remediation of paper mill wastewater through electrocoagulation

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Manuscript received online 20 April 2020, accepted 12 June 2020

In the present study, hybrid central composite design (HCCD) was chosen to optimize four variables based remediation of paper mill wastewater by using electrocoagulation (EC). Response surface methodology incorporates with HCCD designs are rarely used by researchers for optimization of paper mill wastewater remediation. pH, electrode distance, conductivity and current density are operating variables which were selected for optimization over response parameters like COD, color and TDS. EC experiments were performed in 1.5 L capacity reactor with four aluminum electrodes connected to DC power supply. 35 min of electrolysis time for remediation of paper mill wastewater was determined by performing EC experiments with respect to time. Quadratic model was suggested by HCCD design for 20 sets of experimental runs. The signification of this design was measured in terms of regression analysis and t-test. For all response variables regression coefficient and *p*-value are > 0.80 and < 0.05 respectively. Desirable solution for optimization is obtained at pH = 7.11, electrode distance = 1.98 cm, conductivity = 7.07 mS/cm and current density = 13.76 mA/cm^2. Reduction in COD, color and TDS were 80.89%, 93.37%, and 87.54% respectively.

Keywords: Hybrid central composite design, paper mill wastewater, response surface methodology, optimization, aluminum electrode.

Introduction

Wastewater generation in the paper industry has been problematic due to the massive amount of water consumed in the production process[1]. The wastewater generated in paper industries contains different types of chemical, which is toxic in nature. Recycled fiber industry produces white wastewater with high Chemical Oxygen Demand (COD), Total Dissolved Solid (TDS) and color[2]. Treatment of this wastewater is necessary to reuse and dispose of safely in the environment. Paper mill effluent characteristics vary based on raw material used for production[3]. Various researchers had reported different treatment methods for paper mill wastewater such as biological treatment methods, adsorption, phytoremediation, electrocoagulation, oxidation ozone treatment and coagulation[4–8]. Electrocoagulation is one of a simple method to treat wastewater efficiently. EC process does not require any additive of chemicals, can be easily automated, cost-efficient, energy-efficient, safe, versatile and rapid achievement of results is profitable[9]. Various variables influence the EC process amongst which main considerations include conductivity, current density and distance between the electrodes[10]. Optimization of these factors is important to decrease the treatment cost. Most of the researchers have been used Response surface methodology (RSM) for multivariate optimization[11]. Central composite design and box behnken design are mostly used designs for optimization from available RSM designs, while researchers has been less attentive toward hybrid central composite design (HCCD)[11,12]. HCCD design was developed by Roquemore (1976), to accomplish a similar level of orthogonality as central composite or regular polyhedral designs, to be close rotatable, to be close least point in size and to have some simplicity in coding[13]. In this study paper mill wastewater remediation by electrocoagulation using aluminum electrodes take place. Optimization of remediation process was evaluated by HCCD design for four variable parameters such as conductivity, current density, electrode distance and pH over response parameters removal efficiency like Chemical Oxygen Demand (COD), Total Dissolved Solid (TDS) and Color.

Material and methods

Wastewater sample utilized in this work was obtained from reuse fiber based paper plant situated close Champa town and kept in cold storage at 4°C. Characterizations of paper mill wastewater were performed by standard method given by APHA[13]. Table 1 presents the characteristics of wastewater.

Table 1. Physicochemical characteristics of paper mill wastewater

Sr. No.	Name of parameters	Before	After
1.	Chemical Oxygen Demand (mg/L)	2700–3200	200–400
2.	Total Organic Carbon (mg/L)	1500–1700	140–200
3.	Color (CU)	2500–2800	200–275
4.	Total Dissolved Solid (mg/L)	2650–3000	400–600
5.	Total Suspended Solid (mg/L)	100–150	40–60
6.	Total Solid (mg/L)	2750–3150	440–660
7.	pH	7.18–7.55	7.67–8.00

EC arrangement utilized is introduced in Fig. 1. Reactor with measurements of 130×130×150 mm with 2 L volume capacity was used for execution of experiments. Four aluminum electrodes were used with measurements of 85×90×2 mm having working surface of 144 cm^2. Electrodes were assosiated in monopolar manner with DC power supply (0–30 V, 0–5 A). Magnetic stirrer was used for proper mixing with 250 rpm maintained throughout EC experiment. Percentage COD, color and TDS removal were obtained to validated the performance of EC.

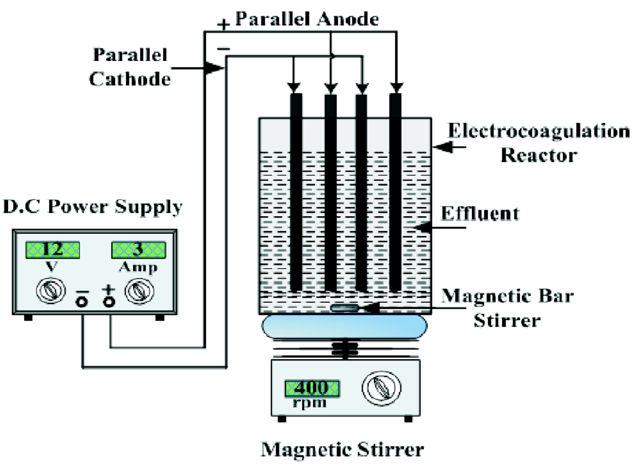

Fig. 1. Experimental setup used for electrocoagulation (EC) process.

An experimental design was employed for optimizing electrocoagulation processes. For response surface study, HCCD design is chosen with subtype randomized design for quadratic model, four factors were varied through high and low value over a three response parameter i.e. Chemical Oxygen Demand (COD), color and Total Dissolved Solid (TDS). Initial pH, conductivity, electrode distance (ED) and current density (CD) is selected as factors for the EC experiments with coded value A, B, C and D respectively. The levels of coded factors using HCCD design shown in Table 2.

Table 2. Ranges of experimental factors and their coded levels

Factor	Name	Units	Lowest	Highest	Low (–1)	High (+1)
A	pH		3.00	8.00	4.50	7.50
B	Conductivity	mS/cm	3.50	9.00	5.46	8.48
C	ED	Cm	1.00	2.50	1.38	2.13
D	CD	mA/cm^2	5.00	20.00	8.75	16.25

Results and discussion

The set of 20 experiments designed by HCCD for EC treatment of reuse fiber based paper mill wastewater with four variable factors over the three responses are shown in Table 3. To perform response surface regression investigation, full quadratic model by giving regression coefficients chosen to fit experimental set of data[14]. The full quadratic model utilized in the response results were reported with actual factors of A, B, C and D were coded given as follows:

$$\text{COD}(\%) = 79.04 + 7.57\,A + 0.1218\,B - 1.92\,C + 6.91\,D - 0.2525\,AB + 2.25\,AC - 0.8463\,AD - 0.3787\,BC + 0.9456\,BD - 1.10\,CD - 8.94\,A^2 + 0.1342\,B^2 - 0.7303\,C^2 - 2.83\,D^2$$

$$\text{Color}(\%) = 91.08 + 9.37\,A + 1.07\,B - 1.21\,C + 9.92\,D + 0.5050\,AB + 4.12\,AC - 2.44\,AD - 0.1894\,BC - 0.5414\,BD + 0.2705\,CD - 14.37\,A^2 - 2.00\,B^2 + 0.211\,C^2 - 7.36\,D^2$$

$$\text{TDS}(\%) = 86.12 + 10.00\,A + 0.0216\,B - 1.76\,C + 9.92\,D$$

Table 3. Design of experiment by HCCD design with response, observed and predicted values

Sr. No.	pH	Conductivity (mS/cm)	ED (cm)	CD (mA/cm^2)	COD (%)		Color (%)		TDS (%)	
					O^a	P^b	O^a	P^b	O^a	P^b
1.	6.0	7.0	1.5	12.5	80.0	80.0	92.0	92.0	87.0	87.0
2.	6.0	7.0	1.5	20.0	84.0	84.0	82.0	82.0	80.0	80.0
3.	6.0	9.0	1.5	8.5	68.0	68.0	72.0	72.0	65.0	65.0
4.	4.5	5.5	2.0	15.0	60.0	62.2	64.0	67.0	64.0	66.3
5.	4.5	7.0	1.5	12.5	65.0	65.0	71.0	71.0	66.0	66.0
6.	7.5	8.5	1.0	15.0	82.0	79.7	85.0	82.0	84.0	81.6
7.	4.5	8.5	1.0	15.0	73.0	75.2	79.0	82.0	75.0	77.3
8.	6.0	7.0	1.5	12.5	80.0	80.0	92.0	92.0	87.0	87.0
9.	6.0	4.5	1.5	8.5	70.0	70.0	65.0	65.0	65.0	65.0
10.	6.0	7.0	1.0	8.5	67.0	67.0	76.0	76.0	68.0	68.0
11.	6.0	7.0	1.5	12.5	80.0	80.0	92.0	92.0	87.0	87.0
12.	6.0	7.0	1.5	12.5	80.0	80.0	92.0	92.0	87.0	87.0
13.	7.5	8.5	2.0	15.0	78.0	80.2	86.0	89.0	80.0	82.3
14.	4.5	5.5	1.0	15.0	74.0	71.7	84.0	81.0	75.0	72.6
15.	7.5	5.5	2.0	15.0	82.0	79.7	90.0	87.0	84.0	81.6
16.	6.0	7.0	2.5	8.5	64.0	64.0	70.0	70.0	62.0	62.0
17.	4.5	8.5	2.0	15	66.0	63.7	70.0	67.0	68.0	65.6
18.	7.5	5.5	1.0	15	75.0	77.2	76.0	79.0	73.0	75.3
19.	6.0	7.0	1.5	12.5	80.0	80.0	92.0	92.0	87.0	87.0
20.	8.0	7.0	1.5	8.5	62.0	62.0	60.0	60.0	65.0	65.0

^{a}O = observed. ^{b}P = predicted.

$+ 0.3788\ AB + 2.34\ AC - 5.35\ AD - 1.04\ BC + 1.10\ BD - 0.2616\ CD - 12.56\ A^2 - 1.59\ B^2 - 0.691\ C^2 - 6.81\ D^2$

The effect of operating variables on paper mill wastewater by electrocoagulation was investigated by selecting quadratic model. Also, significance of quadratic model was determined by analysis of variance (ANOVA)[14]. Fit of the model supported by higher value of regression coefficients and adjusted regression coefficients as presented in Table 4. ANOVA results for fitted quadratic model were shown in Table 5. Replicated runs at centre point was performed to obtained the pure error sum of squares. The lack of fit sum of squares calculated based on affects that were seen little on the normal probability. The lack of fit test had small *F* ratios, so the model was correctly specified. The *F*-value less than 1 for lack of fit imply that lack of fit is not significant relative to the pure error. Optimization of parameters for electrocoagulation treatment of paper mill wastewater was completed by Design expert response optimizer. Variation in predicted response with the variables represented by optimization plots. The optimization plot obtained by HCCD is given in Fig. 2. Most desirable solution determined over 100 design point shown by these graphs. The desirable solution for optimization was obtained at pH = 7.11, electrode distance = 1.98 cm, conductivity = 7.07 mS/cm and current density = 13.76 mA/cm^2. Reduction in COD, color and TDS were 80.89%, 93.37%, and 87.54% respectively. The Global desirability was equal to 1 for HCCD design as shown in Fig. 3.

Table 4. Summary of model fit statistics tested for the responses

Sr. No.	Response parameters	R^2	Adj. R^2	*p*-values	Remarks
1.	COD	0.9634	0.8610	0.0109	Significant
2.	Color	0.9666	0.8730	0.0088	Significant
3.	TDS	0.9735	0.8992	0.0051	Significant

Table 5. ANOVA statistics tested for the responses

Source	COD	Color	TDS	COD	Color	TDS	COD	Color	TDS	COD	Color	TDS	Remark
	Sum of squares			Mean square			*F*-value			*p*-value			
Model	1066.5	2083.0	1655.8	76.1	148.7	118.2	9.4	10.3	13.1	0.010	0.0080	0.0050	Significant
A	204.20	312.65	356.2	204.2	312.6	356.2	25.2	21.71	39.47	0.004	0.0050	0.0010	
B	0.12	9.9	0.004	0.1	9.94	0.004	0.01	0.69	0.0004	0.904	0.4440	0.9840	
C	53.2	21.1	44.75	53.2	21.11	44.75	6.58	1.47	4.96	0.050	0.2800	0.0760	
D	190.7	392.4	392.9	190.7	392.42	392.91	23.55	27.25	43.54	0.004	0.0030	0.0012	
AB	0.5	2.0	1.1	0.5	2.00	1.13	0.06	0.13	0.124	0.813	0.7240	0.7384	
AC	72.0	242.0	78.1	72.0	242.0	78.13	8.89	16.81	8.66	0.030	0.0090	0.0322	
AD	1.1	9.2	44.4	1.1	9.2	44.44	0.13	0.64	4.92	0.726	0.4590	0.0772	
BC	2.0	0.5	15.1	2.0	0.5	15.12	0.24	0.03	1.68	0.640	0.8590	0.2520	
BD	7.1	2.3	9.7	7.1	2.34	9.71	0.88	0.16	1.08	0.391	0.7030	0.3472	
CD	11.2	0.6	0.6	11.2	0.67	0.6345	1.39	0.04	0.07	0.291	0.8360	0.8015	
A^2	166.3	430.3	328.5	166.3	430.3	328.51	20.53	29.89	36.40	0.006	0.0020	0.0018	
B^2	0.1	29.2	18.3	0.1	29.2	18.39	0.01	2.03	2.04	0.903	0.2130	0.2127	
C^2	10.7	0.8	9.6	10.7	0.89	9.61	1.32	0.06	1.07	0.302	0.8130	0.3493	
D^2	72.8	491.3	420.2	72.8	491.3	420.26	8.99	34.12	46.57	0.030	0.0020	0.0010	
Residual	40.5	72.0	45.1	8.1	14.4	9.02							
Lack of fit	40.5	72.0	45.1	40.5	72.0	45.12	0.19	0.32	0.56	0.982	0.873	0.8640	Not significant
Pure error	0.05	1.2	0.6	0.03	0.14	0.08							
Cor total	1107	2155	1700										

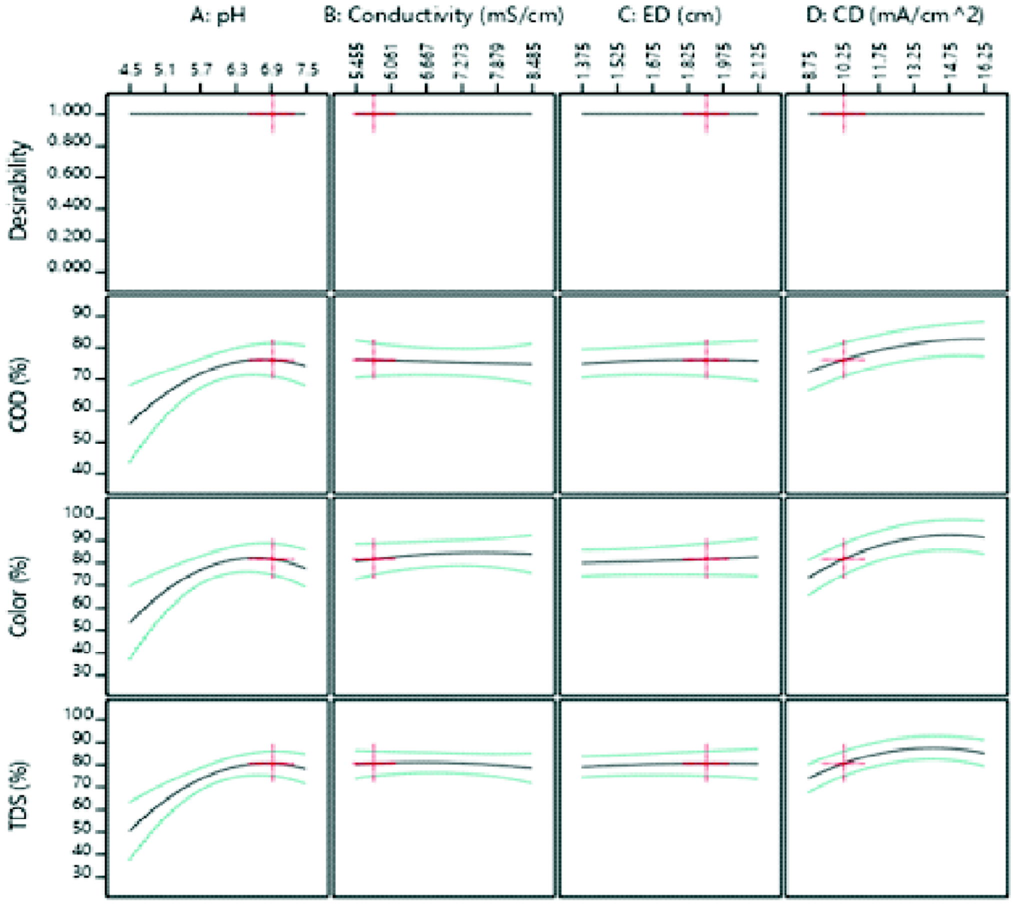

Fig. 2. All variable factors and response parameters plots for a desirable solution.

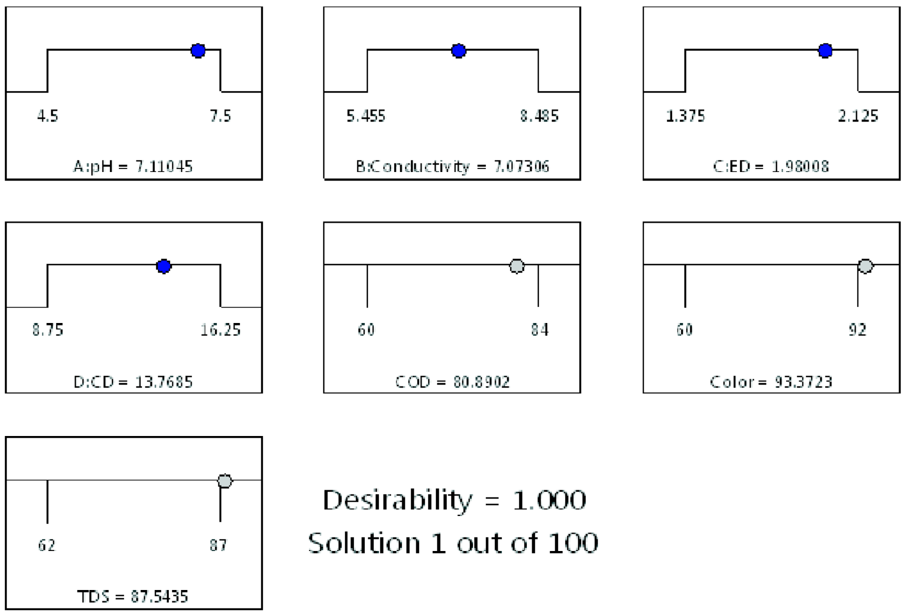

Fig. 3. Desirability analysis solution out of hundred design points for paper mill wastewater treatment by electrocoagulation.

Conclusions

The results obtained in this study, provides an opportunity for the application of treatability through EC treatment utilizing aluminum electrodes for recycled fiber based paper mill wastewater. RSM coupled with HCCD design model has been effectively applied. The impacts of variable factor like initial pH, current density, electrode distance and conductivity on responses COD, color and TDS removal were investigated. HCCD design model used in this study demonstrated that correlation between the experimental data and the values predicted using model design is very high $R^2 > 0.80$. Desirable solution for optimization is obtained at pH = 7.11, electrode distance = 1.98 cm, conductivity = 7.07 mS/cm and current density = 13.76 mA/cm^2. Reduction in COD, color and TDS were 80.89%, 93.37%, and 87.54% respectively.

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