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ORIGINAL PAPER

Influence of operating parameters on treatment of egg processing effluent by electrocoagulation process

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Abstract The treatment of egg processing effluent was investigated in a batch electrocoagulation reactor using aluminum as sacrificial electrodes. The influence of operating parameters such as electrode distance, stirring speed, electrolyte concentration, pH, current density and electrolysis time on percentage turbidity, chemical oxygen demand and biochemical oxygen demand removal were analyzed. From the experimental results, 3-cm electrode distance, 150 rpm, 1.5 g/l sodium chloride, pH of 6, 20 mA/cm^2 current density, and 30-min electrolysis time were found to be optimum for maximum removal of turbidity, chemical oxygen demand and biochemical oxygen demand. The removal of turbidity, chemical oxygen demand and biochemical oxygen demand under the optimum condition was found to be 96, 89 and 84 %, respectively. The energy consumption was varied from 7.91 to 27.16 kWh/m^3, and operating cost was varied from 1.36 to 4.25 US \$/m^3 depending on the operating conditions. Response surface methodology has been employed to evaluate the individual and interactive effects of four independent parameters such as electrolyte concentration (0.5–2.5 g/l), initial pH (4–8), current density (10–30 mA/cm^2) and electrolysis time (10–50 min) on turbidity, chemical oxygen demand and biochemical oxygen demand removal. The results have been analyzed using Pareto analysis of variance to predict the responses. Based on the analysis, second-order polynomial mathematical models were developed and found to be good fit with the experimental data.

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Introduction

The egg processing industry is a water-intensive industry, it generates more than 10 billion liters of effluent annually, and it has high organic loads due to presence of egg proteins (Xu et al. 2001, 2002). The volume of effluent generated and its characteristics normally depend on the process used for egg processing. The effluent, which is generated from all the process stages mainly, contains detergent, chemicals, egg proteins and fats. High concentrations of organic components in effluent streams involve a serious environmental problem and must be properly treated before discharge from the environment. The State Pollution Control Board of Tamil Nadu, India, has directed the industries to implement zero discharge facilities.

An extensive literature analysis has been made, and it is found that only a few authors have made an attempt to study the treatment of egg processing effluent. Bough (1975) studied the treatment of wastewater generated from egg breaking section using chitosan as an coagulation and reported that 70–90 % total solids and 55–75 % chemical oxygen demand (COD) were reduced depends on the process conditions. Bulley (1976) used aluminum sulfate as a coagulation agent to treat egg processing wastewater using and reported that the total solids was removed 38–92 % and biochemical oxygen demand (BOD_5) was 80–89 % depend on the quantity of coagulant added. Harris and Moats (1975) investigated to recover egg solids from egg processing wastewater and reported more than 90 % BOD_5 and 97 % COD reduction. Xu et al. (2001) studied the treatment of egg processing wastewater using different coagulants such as lignosulfonate, bentonite, carboxymethylcellulose and ferric chloride for simulated and industrial wastewater and found that chemical oxygen demand, turbidity and total solids were reduced over 90, 97 and 95 %, respectively, for all coagulants. Xu et al. (2002) studied the recovery of protein from simulated and industrial egg processing wastewater using electrocoagulation method and reported that digestibly of protein and fats recovered from settled solids was good. From the analysis of literature, it is found that though some authors made an attempt to recover protein from egg processing effluent, detail extensive work has not been reported for treatment of effluent function of fundamental and operating variable.

Conventionally, the industrial effluents are treated by many different techniques such as adsorption, membrane filtration, coagulation–flocculation and advanced oxidation processes such as ozone, photochemical and Fenton's method, etc. (Akyol et al. 2004; Ersoy et al. 2009; Minhalma et al. 2006; Zhang et al. 2006) to remove COD, BOD_5 and color. These technologies require an extensive setup for treatment of effluent. Moreover, each step takes place in a separate tank, and the entire treatment requires several pH adjustments as well as the addition of chemicals. These conventional processes generate a considerable quantity of secondary pollutants and large volumes of sludge which needs further treatment. The biological treatment processes are more suitable for treatment of high-strength organic effluent when two-stage (anaerobic + aerobic) treatment technique is used. (Kalyuzhnyi et al. 2005). The drawbacks associated with the conventional and biological techniques forced the effective treatment method for complete degradation of pollutants. In recent years, electrocoagulation process has been attracting a great attention for treatments of industrial effluents such as poultry slaughterhouse (Bayramoglu et al. 2006), cork process (Beltran de Heredia et al. 2004), yeast industry (Kobya and Delipinar 2008), dairy industry (Tchamango et al. 2010), olive mill (Un et al. 2006) and distillery industry (Yavuz 2007), etc., because of the flexibility and the environmental compatibility. Electrocoagulation technique has some advantages compared to conventional methods such as easy to operate, less retention time, reduction or absence of adding chemicals, rapid sedimentation of the electro-generated flocs and less sludge production (Holt et al. 2005). In order to develop an electrocoagulation process for egg processing effluent, the fundamental and operating variables, which affect the removal efficiency and operating cost, have to be optimized. Hence in the present study, the effects of the operating parameters such as electrode distance, stirring speed, electrolyte concentration, initial pH, current density and electrolysis time using aluminum electrodes are investigated in detail, and energy consumption and operating costs were calculated. Box–Behnken design was also employed to develop mathematical models for describing the interactive effects of electrolyte concentration, initial pH, current density and electrolysis time on the performance of an electrocoagulation process for the treatment of egg processing effluent. The present research was carried out in the Department of Food Technology at Kongu Engineering College, Perundurai, in April–May 2012.

Materials and methods

Effluent and characteristics

The egg processing effluent used in this study was obtained from an egg processing plant located in Erode, Tamil Nadu, India, and stored in airtight plastic cans at 4 °C to prevent natural degradation until it was used. The characteristics of egg processing effluent were as follows: pH, 7.34–7.95; COD, 3,200–4,300 mg/l; BOD_5, 1,800–2,250 mg/l; total suspended solids, 1,050–1,280 mg/l; turbidity, 950–1,100 NTU; and conductivity, 0.455–0.614 mS/cm.

Experimental setup and procedure

A laboratory glass beaker of 500 ml was used for the electrocoagulation experiments (Sridhar et al. 2011). The electrodes used in the electrocoagulation system were made of aluminum (Al) having a surface area of 30 cm^2. The area of electrode exposed for the electrolysis was fixed to be 25 cm^2, and the remaining area was prevented from exposure with lacquer. The anode and cathode were positioned vertically and parallel to each other. The spacing between two electrodes in electrocoagulation process was varied in the range of 1–4 cm. Bottom of the electrodes was kept 1 cm above the bottom of the reactor to allow easy stirring. Magnetic stirrer was used to agitate the solution. The current density was maintained constant by means of a precision digital direct current power supply (0–30 V, 0–2 A). All the electrocoagulation runs were conducted at room temperature. The impurities on the surfaces of electrodes were removed by dipping in HCl solution (15 %W/V) for 1–2 min. In each run, 300 ml of egg processing effluent was placed into the electrocoagulation reactor. The current density was adjusted to a desired value, and the run was started. At the end of the run, the solution was filtered and the electrodes were washed thoroughly with distilled water to remove any solid residues on the surfaces, dried and used again. The filtered samples were analyzed for turbidity, COD and BOD_5.

Analytical procedures

The chemical oxygen demand and biochemical oxygen demand analysis were carried out by procedures described in standard methods (Greenberg et al. 1995). The turbidity (NTU) of samples was analyzed using Elico CL52D turbidity meter. The pH was measured using Elico LI120 model pH meter, and the conductivity was determined by Elico CM180 model conductivity meter.

Electrical energy consumption

The electrical energy consumption is very important economical parameter in electrocoagulation process. The electrical energy consumption was calculated using the following equation (Sridhar et al. 2011).

$$E = \frac{VIt}{V_s} \quad (1)$$

where E is the electrical energy (kWh/m^3), V is the cell voltage in volt (V), I is the current in ampere (A), V_s is the volume of solution (l) and t is the time of electrocoagulation process (h).

Results and discussion

The effects of parameters

In the runs, the effects of parameters such as electrode distance, stirring speed, electrolyte concentration, pH, current density and electrolysis time were investigated for percentage turbidity, COD and BOD_5 removal.

Effect of electrode distance

Experiments were carried out by varying the electrode distance between 1 and 4 cm, and the observations are given in Fig. 1a–c. From the figures, it is observed that the percentage removal of turbidity, COD and BOD_5 increases with increasing electrode distance. The results indicate that when the inter-electrode distance is increased from 1 to 3 cm, the removal of turbidity, COD and BOD_5 increased by about 13, 12 and 12 %, respectively, after 30 min of electrolysis time. A further increase in the distance beyond 3 cm decreased the percentage turbidity, COD and BOD_5 removal due to the less interaction of ions with hydroxide polymers (Modirshahla et al. 2007). The removal efficiency is lower for 1–2-cm electrode distance than 3 cm because the gap between anode and cathode is too closed, and solid and fluid transfer was obstructed. The accumulated solid particles and bubbles between the anodes and the cathodes caused a consequent higher electrical resistance. The effect of inter-electrode distance on energy consumption is shown in Fig. 1d. From the figure, it is observed that the energy consumption increases with increasing electrode distance. The energy consumption is varied from 13.41 to 18.91 kWh/m^3 at 30 min of electrolysis for 1–4-cm electrode distance. The results suggest that the 3-cm electrode distance can provide more economical operation.

Effect of stirring speed

The mixing is an important operating factor influencing the performance of electrocoagulation process. To examine its effect on the treatment of egg processing effluent, the stirring speed was varied in the range of 50–200 rpm. As shown in Fig. 2a–c, the percentage turbidity, COD and BOD_5 removal increase by about 18, 16 and 17 %, after 30 min of electrolysis time when the stirring speed was increased from 50 to 150 rpm. This confirms the fact that the removal efficiency is diffusion controlled, and the increase in stirring speed leads to increase in the intensity of turbulence and reduces the diffusion layer thickness at the electrode surface and improves the mixing conditions in the electrolyte bulk (Bouhezila et al. 2011; El-Ashtoukhy et al. 2009). No significant increase in turbidity, COD and BOD_5 removal was observed by increasing stirring speed beyond 150 rpm. The effect of stirring speed (rpm) on energy consumption is shown in Fig. 2d. From the figure, it is found that the energy consumption decreases with increasing stirring speed because mainly it increases the intensity of turbulence and reduces the diffusion layer thickness at the electrode surface. But the energy consumption increases with increasing electrolysis time. The energy consumption was varied from 22.99 to 14.85 kWh/m^3 at 30 min of electrolysis for 50–200 rpm rotational speed. The result suggests that the stirring speed 150 rpm can provide more economical operation because no significant reduction in COD and BOD_5 when mixing speed beyond 150 rpm.

Effect of electrolyte (NaCl) concentration

In general, the electrolytes are used to obtain the conductivity in electrocoagulation process. Solution conductivity affects the current efficiency, cell voltage and consumption of electrical energy in electrolytic cells. In this work, the conductivity of egg processing effluent was adjusted to the desired level by adding an appropriate amount of NaCl (Kobya and Delipinar 2008). The effect of NaCl concentration on turbidity removal efficiency is shown in Fig. 3a. Turbidity removal increased from 73 to 96 % as NaCl concentration increased from 0.5 to 2.5 g/l. The increase in

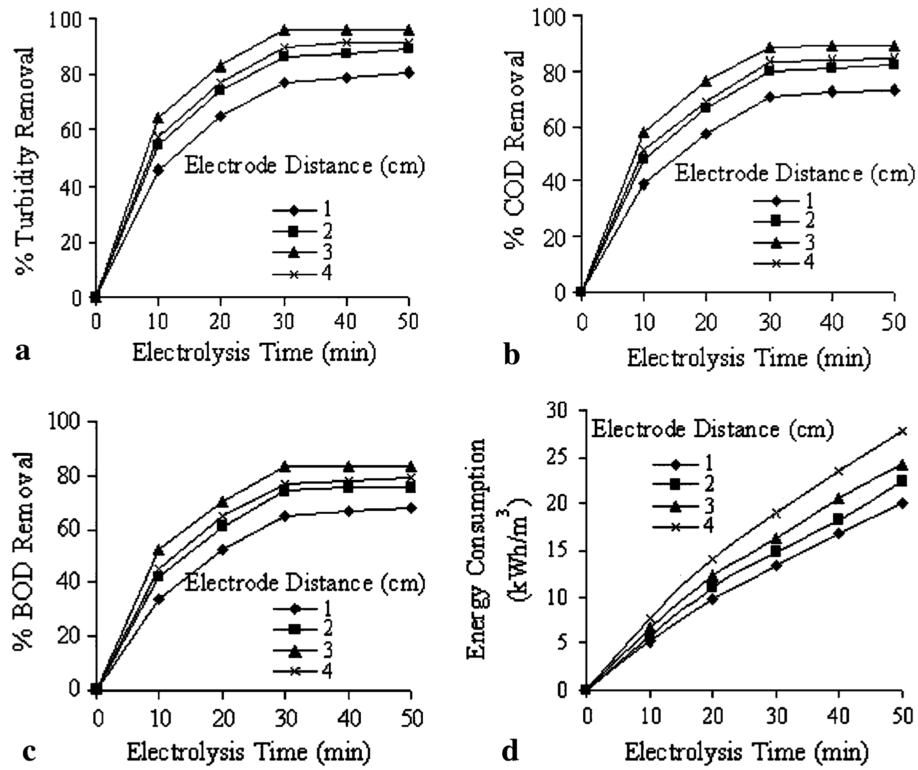


Fig. 1 Effect of electrode distance on, **a** percentage turbidity removal, **b** percentage COD removal, **c** percentage BOD_5 removal and **d** energy consumption (pH = 6, current density = 20 mA/cm^2, rpm = 150 and NaCl = 1.5 gm/l)

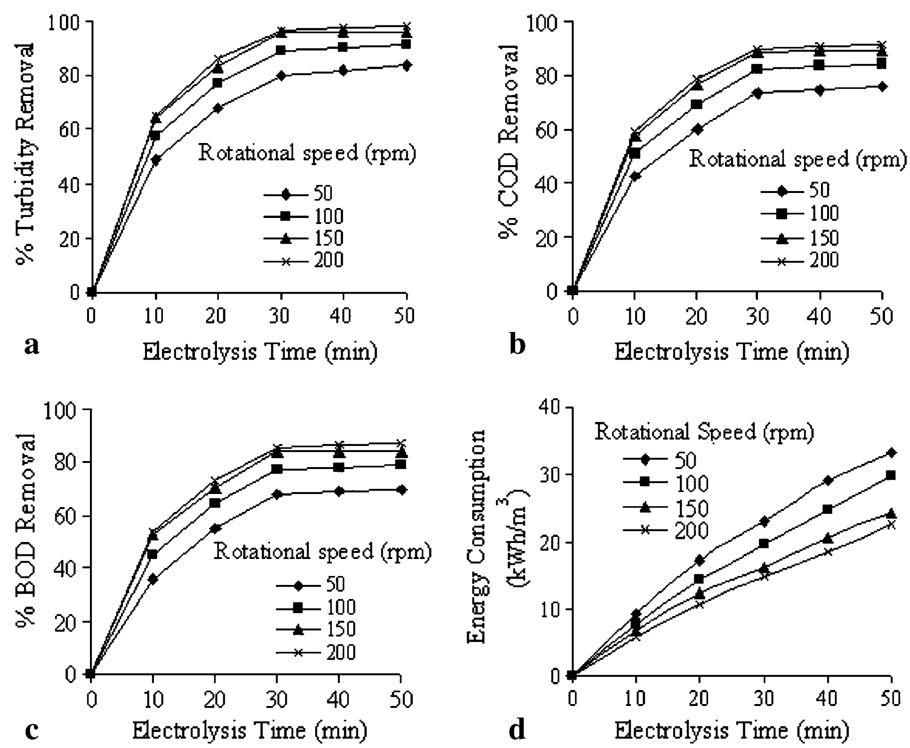


Fig. 2 Effect of rotational speed (rpm) on **a** percentage turbidity removal, **b** percentage COD removal, **c** percentage BOD_5 removal and **d** energy consumption (pH = 6, current density = 20 mA/cm^2, NaCl = 1.5 g/l and electrode distance = 3 cm)

the removal efficiency may be attributed to a change in the ionic strength due to the changing conductivity of aqueous medium. The higher ionic strength will generally cause an increase in current density in the same cell voltage or, equivalently, the cell voltage decreases with increasing effluent conductivity at constant current density. Also, with the addition of NaCl to the medium, following reactions take place in the effluent (Chen 2004).

$$2Cl^- \rightarrow Cl_2 + 2e^-$$

$$Cl_2 + H_2O \rightarrow HOCl + Cl^- + H^+$$

$$HOCl \rightarrow OCl^- + H^+$$

As shown in above reactions, when sodium chlorides are added into the solutions, the products discharged from anode are Cl_2 and OCl^-. The OCl^- itself is a strong oxidant, which capable of oxidizing organic molecules present in effluent and hence turbidity removal increases. Therefore, addition of NaCl not only increases the conductivity but also contributes strong oxidizing agents (Sridhar et al. 2011). The presence of NaCl has a considerable effect on the percentage COD and BOD_5 removal up to 1.5 g/l, beyond that there is no significant reduction in COD and BOD_5 (Fig. 3b, c). The effect of electrolyte concentration on energy consumption for Al electrode is shown in Fig. 3d. The energy consumption decreased with increasing concentration of supporting electrolyte because the potential decreased under constant current density. The energy consumption was varied from 21.91 to 14.74 kWh/m^3 at 30 min of electrolysis for 0.5–2.5 g/l NaCl. According to the results, high removal percentage with low cell voltages and low energy consumption was obtained with NaCl concentration of around 1.5 g/l.

Effect of pH

The influent effluent pH is one of the important factors which affect the performance of electrocoagulation process. To evaluate this effect, a series of experiments were performed. The effect of pH on the turbidity, COD and BOD_5 removal was examined at 4, 5, 6, 7 and 8 pH. From the Fig. 4a–c, it is observed that the percentage of turbidity, COD and BOD_5 removal increases with increasing pH up to 6 and then decreased. The extent of hydrolysis depends upon total pollutant concentration and pH, as well as the amount of other species present in solution. At lower pH less than 6, the protons in the solution are reduced to H_2 at the cathode, and the same proportion of hydroxide ions could not be produced, which leads to the formation of $Al(OH)^{2+}$ and $Al(OH)_2^+$, and hence, COD and BOD_5 reduction was found to be less (Mollah et al. 2001). When is pH 6–6.5, there is a formation of Al(III) species in the form of $Al(OH)_{3(s)}$ which increases the BOD_5 and COD removal efficiency. $Al(OH)_4^-$ forms at the higher pH, which is dissolving nature in the effluent and does not form flocks, and hence, there is no reduction in turbidity, COD and BOD_5 (Zaied and Bellakhal 2009). The relationship between the energy consumption and pH is shown in Fig. 4d. The energy consumption increased with increasing the pH values. The minimum and maximum energy consumption was observed at pH 4 and 8, respectively. The

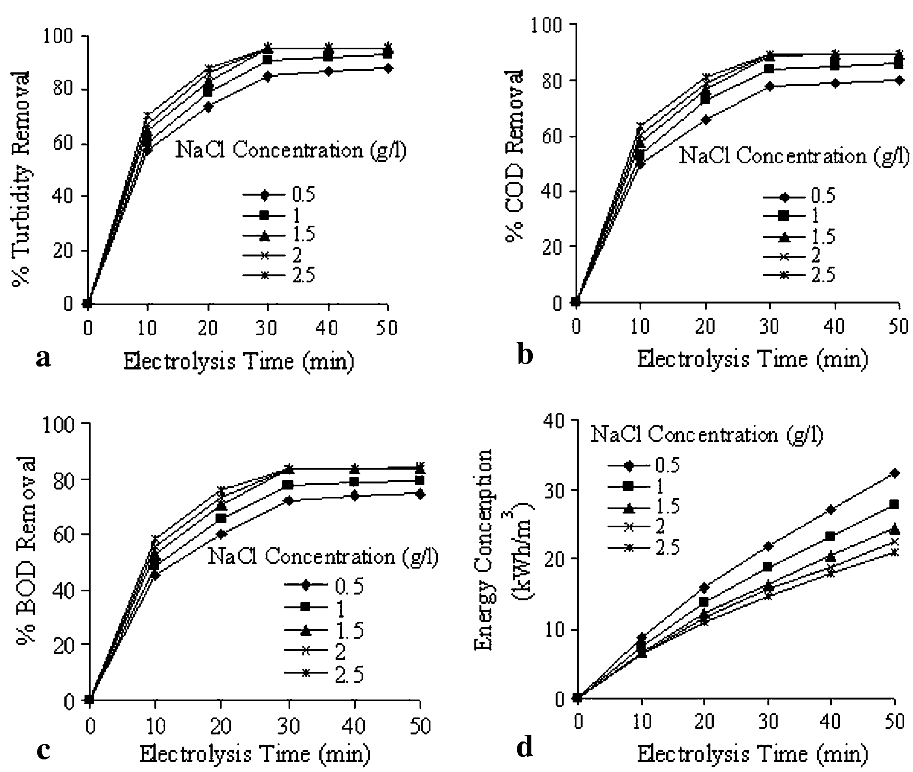


Fig. 3 Effect of NaCl concentration on **a** percentage turbidity removal, **b** percentage COD removal, **c** percentage BOD_5 removal and **d** energy consumption (pH = 6, current density = 20 mA/cm^2, rpm = 150 and electrode distance = 3 cm)

energy consumption is varied from 14.41 to 19.24 kWh/m^3 at 30 min of electrolysis for the pH range of 4–8.

Effect of electrolysis time and current density

From the Fig. 5a–c, it is observed that the turbidity, COD and BOD_5 removal efficiency increases with increasing electrolysis time. After 30 min of electrolysis turbidity, COD and BOD_5 removal efficiency reached a maximum at all current density except for 10 mA/cm^2.

The current density is the most important parameter in all electrocoagulation process. The current density determines not only the coagulant dosage rate but also the bubble production rate, size and the flocs growth, and they have strong influence on treatment efficiency of the electrocoagulation process. Fig. 5a–c illustrate that the turbidity, COD and BOD_5 removal efficiency increased from 84 to 96 % by increasing the current density from 10 to 30 mA/cm^2 for 30 min electrolysis time. This is mainly due to direct and indirect oxidation. When sufficient voltage is developed across the electrodes, direct oxidation takes place near the anode, due to the release of electrons by the organic compounds in order to maintain the flow of current, whereas indirect oxidation occurs due to the strong oxidants that form during the reaction. Here, the Cl^- can be the principal charged species which carry the current in the solution. If Cl^- carries the current, then Cl_2 gas is produced at the anodes which rapidly hydrolyze to form hypochlorous acid, which is a strong oxidant, and has the ability to oxidize the organic compounds effectively (Kobya and Delipinar 2008; Adhoum et al. 2004). The removal of turbidity, COD and BOD_5 was increased with increasing current density up to 30 min. After 30 min, increasing current density beyond 20 mA/cm^2 did not show any significant improvement on the percentage of turbidity, COD and BOD_5 removal. The effect of current density on energy consumption is shown in Fig. 5d. The energy consumption increased with increasing current density due to increase in ion production on the anode and cathode. The energy consumption is varied from 7.91 to 27.16 kWh/m^3 in the current density range of 10–30 mA/cm^2.

Effect of parameters on operating cost

The operating cost is very important economical parameter in electrocoagulation process. The operating cost involves costs of chemicals, electrodes and energy consumptions as well as labor, maintenance, sludge disposal and fixed costs. The energy, electrode and chemical costs were taken into account as major cost items in the calculation of operating cost (Bayramoglu et al. 2004; Sridhar et al. 2011).

$$\text{Operating cost}\ \left(\text{US}\ \$/\text{m}^3\right) = aC_{\text{energy}} + bC_{\text{electrode}} + cC_{\text{chemicals}} \quad (2)$$

where C_{energy} is the electrode and magnetic stirrer energy consumption (kWh/m^3), $C_{electrode}$ is the electrode consumption (kg/m^3) and $C_{chemicals}$ is the chemical

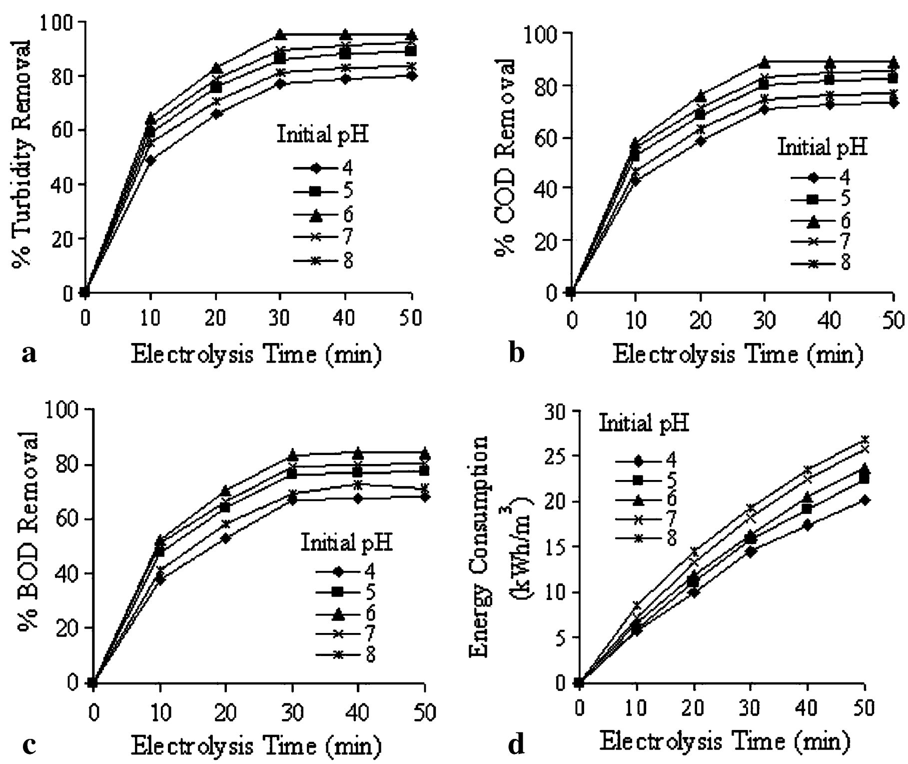
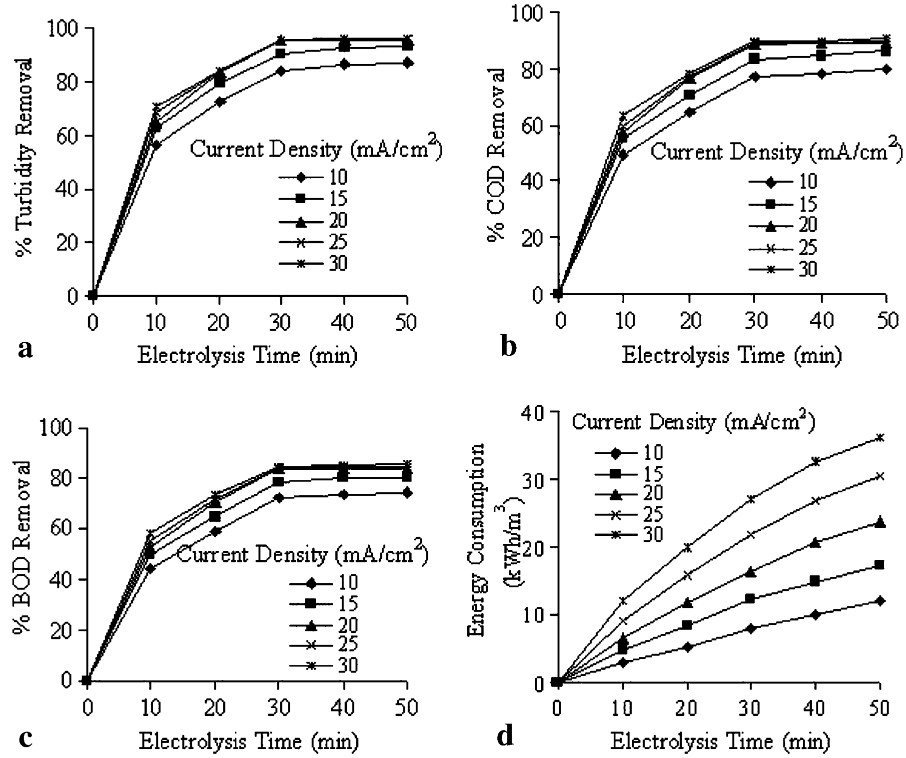


Fig. 4 Effect of pH on percentage **a** percentage turbidity removal **b** percentage COD removal and **c** BOD_5 removal and **d** energy consumption (rpm = 150, current density = 20 mA/cm^2, NaCl = 1.5 g/l and electrode distance = 3 cm)

Fig. 5 Effect of current density on **a** percentage turbidity removal, **b** percentage COD removal, **c** percentage BOD_5 removal and **d** energy consumption (rpm = 150, pH = 6, NaCl = 1.5 g/l and electrode distance = 3 cm)



consumption (kg/m^3). Unit prices a, b and c given for the price at first quarter of 2012, India, are as follows: (a) electrical energy price 0.085 US $/kWh, (b) electrode material (Al) price 2.23 US $/kg and (c) electrolyte (NaCl) price 0.043 US $/kg.

The operating cost increased with increasing electrode distance, pH and current density and decreased with increasing stirrer speed and electrolyte concentration. The operating costs were determined as 2.46–2.92, 2.54–2.95, 1.36–4.25, 3.27–2.58 and 3.14–2.61 $/m^3 at electrode distance of 1–4 cm, pH of 4–8, current density of 10–30 mA/cm^2, stirrer speed of 50–200 rpm and electrolyte concentration of 0.5–2.5 g/l, at 30 min of electrolysis, respectively. Under optimal operating condition such as 20 mA/cm^2 current density, pH of 6, 1.5 g/l NaCl, 150 rpm, 3-cm electrode distance and 30 min of electrolysis, the operating cost is found to be 2.7 US $/m^3.

Development of model

Experimental design and procedure

In this study, the Box–Behnken design with four factors at three levels was applied using Stat Ease Design-Expert 8.0.4 with the limits of the independent variables. Each independent variable was coded at three levels between −1 and +1 in the ranges determined by the preliminary experiments. Table 1 gives the parameters and the operating ranges covered. A total of 29 experiments, including five center points, were employed to evaluate the individual and interactive effects of the four main independent parameters on the turbidity, COD, BOD_5 removal efficiency and energy consumption. Percentage turbidity, COD and BOD_5 removal and energy consumption have been taken as a response (Y) of the system, while four process parameters, namely current density (j): 10–30 mA/cm^2; pH (pH_0): 4–8; electrolyte concentration (c): 0.5–2.5 g/l; and electrolysis time (t): 10–50 min, have been taken as input parameters. Experimental conditions and corresponding results (responses) are present in Table 2. The performance of the process was evaluated by analyzing the responses. A non-linear regression method was used to fit the second-order polynomial equation to the experimental data (Prakash Maran et al. 2013a; Sridhar et al. 2011). The statistical significance of the models was justified through analysis of variance (ANOVA) for polynomial model. The quality of the fit polynomial model was also expressed with the coefficient of determination R^2.

Statistical analysis

Experiments were performed to study the effect of j, pH_0, c and t on the turbidity removal, COD removal, BOD_5 removal and energy consumption. The results of the Y (response) of turbidity removal, COD removal, BOD_5 removal and energy consumption were measured according to design matrix, and the measured responses are listed in Table 2. In order to quantify the curvature effects, the data

Table 1 Process parameters and their levels for electrocoagulation treatment

Variable, unit	Factors	Level		
	X	−1	0	+1
Current density, j (mA/cm^2)	X_1	10	20	30
Initial pH, pH$_o$	X_2	4	6	8
Electrolyte concentration, c (g/l)	X_3	0.5	1.5	2.5
Electrolysis time, t (min)	X_4	10	30	50

from the experimental results were fitted to four higher degree polynomial equations viz., linear, two factor interaction (2F1), quadratic and cubic models. Two different tests, namely the sequential model sum of squares and model summary statistics were employed to decide about the adequacy of various models to represent turbidity removal, COD removal, BOD_5 removal and energy consumption, and the results of these tests are given in Table 3. Cubic model was found to be aliased and cannot be used for further modeling of experimental data. Though the p values were in the acceptable range of both linear and two factor interaction (2F1) models, the adjusted R^2 and predicted R^2 values were found to be low (refer Table 3); however, and hence, these two models were eliminated. On the other hand, the quadratic model exhibited low p values (<0.0001) and high adjusted R^2 and predicted R^2 values and was chosen for further analyses.

Development of regression model equation and validation of the model

The adequacy of the model was evaluated through ANOVA (analysis of variance) (Sridhar et al. 2011). The ANOVA

Table 2 Box–Behnken design with observed and predicted responses

Run order	j (X_1)	pH (X_2)	c (X_3)	t (X_4)	% Turbidity removal		% COD removal		% BOD_5 removal		Energy consumption (kWh/m^3)	
					Y_{exp}	Y_{pre}	Y_{exp}	Y_{pre}	Y_{exp}	Y_{pre}	Y_{exp}	Y_{pre}
1	20	8	1.5	50	84.00	83.34	76.50	76.10	71.00	70.48	26.79	27.77
2	20	8	1.5	10	55.00	55.65	46.50	47.40	41.00	41.77	8.47	8.37
3	30	6	2.5	30	96.13	97.31	89.14	90.61	84.12	85.71	23.91	23.45
4	20	6	0.5	50	87.75	87.83	79.88	80.16	74.66	75.05	32.34	31.79
5	20	6	2.5	10	70.71	70.57	63.03	62.88	58.44	58.28	7.70	7.81
6	20	4	0.5	30	68.44	68.98	61.17	61.82	56.03	56.70	20.07	19.77
7	10	6	1.5	50	87.32	87.44	79.94	79.66	74.38	74.17	11.06	10.84
8	20	6	1.5	30	95.56	95.56	88.65	88.65	83.66	83.66	16.24	16.24
9	30	8	1.5	30	86.56	85.97	78.16	77.40	72.33	71.73	32.16	31.79
10	10	4	1.5	30	68.94	69.49	60.72	61.63	55.52	56.35	7.67	7.78
11	20	4	1.5	10	49.10	50.03	42.60	43.39	37.77	38.64	5.20	5.19
12	10	6	2.5	30	89.00	89.05	80.90	80.84	75.42	75.34	11.12	11.27
13	30	6	1.5	10	70.95	70.61	63.00	62.73	58.09	57.71	12.05	12.27
14	20	4	2.5	30	81.13	80.16	74.57	73.35	69.17	67.89	17.41	16.66
15	10	6	0.5	30	75.00	74.08	67.64	66.55	62.55	61.30	11.12	11.43
16	20	6	1.5	30	95.56	95.56	88.65	88.65	83.66	83.66	16.24	16.24
17	20	6	2.5	50	96.02	96.65	89.08	90.23	84.14	85.30	20.95	20.60
18	20	4	1.5	50	80.04	79.65	73.48	72.97	68.00	67.57	20.12	20.36
19	20	6	0.5	10	57.27	56.60	50.23	49.23	45.36	44.43	9.80	10.01
20	10	8	1.5	30	71.54	72.16	63.66	64.44	57.11	57.99	10.24	10.66
21	20	6	1.5	30	95.56	95.56	88.65	88.65	83.66	83.66	16.24	16.24
22	20	8	2.5	30	85.79	85.03	78.45	77.25	73.00	71.76	19.07	18.37
23	10	6	1.5	10	56.66	56.23	49.20	48.92	44.30	44.10	1.00	0.99
24	30	6	1.5	50	96.50	96.71	90.52	90.26	85.66	85.28	36.09	36.99
25	20	6	1.5	30	95.56	95.56	88.65	88.65	83.66	83.66	16.24	16.24
26	20	8	0.5	30	72.68	73.42	64.39	65.06	58.15	58.86	28.91	28.65
27	20	6	1.5	30	95.56	95.56	88.65	88.65	83.66	83.66	16.24	16.24
28	30	4	1.5	30	80.00	79.33	73.71	73.08	68.00	67.34	23.66	24.08
29	30	6	0.5	30	89.27	89.48	80.74	81.19	75.24	75.65	37.41	36.69

Table 3 Adequacy of the models tested for turbidity, COD, BOD_5 removal and energy consumption

Source	Sum of squares	Degree of freedom	Mean square	F value	Prob > F	Remark
Sequential model sum of squares for turbidity removal						
Mean	1.878E + 005	1	1.878E + 005			
Linear	3,337.60	4	834.40	8.92	0.0001	
2F1	30.85	6	5.14	0.042	0.9996	
Quadratic	2,205.80	4	551.45	837.72	<0.0001	Suggested
Cube	8.98	8	1.12	28.75	0.0003	Aliased
Residual	0.23	6	0.039			
Sequential model sum of squares for COD removal						
Mean	1.55E + 005	1	1.55E + 005			
Linear	3,453.83	4	863.46	9.10	0.0001	
2F1	12.56	6	2.09	0.017	1.0000	
Quadratic	2,250.96	4	562.74	548.11	<0.0001	Suggested
Cube	13.45	8	1.68	10.86	0.0046	Aliased
Residual	0.93	6	0.15			
Sequential model sum of squares for BOD_5 removal						
Mean	1.335E + 005	1	1.335E + 005			
Linear	3,412.68	4	853.17	8.74	0.0002	
2F1	11.40	6	1.90	0.015	1.0000	
Quadratic	2,317.25	4	579.31	532.83	<0.0001	Suggested
Cube	14.22	8	1.78	10.66	0.0049	Aliased
Residual	1.00	6	0.17			
Sequential model sum of squares for energy consumption						
Mean	9,138.61	1	9,138.61			
Linear	2,165.66	4	541.42	44.46	0.0001	
2F1	141.39	6	23.57	2.81	0.0413	
Quadratic	134.31	4	33.58	28.42	<0.0001	Suggested
Cube	13.04	8	1.63	2.79	0.1141	Aliased
Residual	3.51	6	0.58			

Source	SD	R^2	Adjusted R^2	Predicted R^2	PRESS	Remark
Model summary statistics for turbidity removal						
Linear	9.67	0.5978	0.5307	0.4705	2,956.33	
2F1	11.09	0.6033	0.3829	0.1414	4,794.02	
Quadratic	0.81	0.9983	0.9967	0.9905	53.08	Suggested
Cubic	0.20	1.0000	0.9998	0.9940	33.74	Aliased
Model summary statistics for COD removal						
Linear	9.74	0.6026	0.5363	0.4813	2,973.19	
2F1	11.22	0.6048	0.3852	0.1609	4,809.66	
Quadratic	1.01	0.9975	0.9950	0.9856	82.79	Suggested
Cubic	0.39	0.9998	0.9992	0.9767	133.71	Aliased
Model summary statistics for BOD_5 removal						
Linear	9.88	0.5928	0.5250	0.4693	3,055.22	
2F1	11.38	0.5948	0.3697	0.1426	4,935.42	
Quadratic	1.04	0.9974	0.9947	0.9848	87.68	Suggested
Cubic	0.41	0.9998	0.9992	0.9750	144.08	Aliased
Model summary statistics for energy consumption						
Linear	3.49	0.8811	0.8613	0.8160	452.29	
2F1	2.89	0.9386	0.9045	0.8067	475.04	
Quadratic	1.09	0.9933	0.9865	0.9612	95.28	Suggested
Cubic	0.76	0.9986	0.9933	0.7946	504.84	Aliased

Table 4 ANOVA results of the quadratic models for turbidity, COD, BOD_5 removal and energy consumption

Factor	% Turbidity removal			% COD removal			% BOD_5 removal			Energy consumption (kWh/m^3)		
	Coeff. of the model	*F* Value	*P* Value	Coeff. of the model	*F* Value	*P* Value	Coeff. of the model	*F* Value	*P* Value	Coeff. of the model	*F* Value	*P* Value
Model	95.56	604.85	<0.0001	88.65	397.76	<0.0001	83.66	377.19	<0.0001	16.24	147.59	<0.0001
X_1	5.91	637.44	<0.0001	6.10	435.15	<0.0001	6.18	421.65	<0.0001	9.36	889.12	<0.0001
X_2	2.33	98.75	<0.0001	1.78	37.21	<0.0001	1.51	25.08	0.0002	2.65	71.22	<0.0001
X_3	5.70	591.93	<0.0001	5.93	410.78	<0.0001	6.02	400.54	<0.0001	−3.35	113.95	<0.0001
X_4	14.33	3,742.08	<0.0001	14.57	2,480.90	<0.0001	14.41	2,291.56	<0.0001	8.64	758.63	<0.0001
X_1^2	−4.12	167.33	<0.0001	−4.54	130.32	<0.0001	−4.80	137.58	<0.0001	1.09	6.58	0.0224
X_2^2	−14.70	2,130.62	<0.0001	−14.97	1,415.91	<0.0001	−15.50	1,434.18	<0.0001	1.25	8.55	0.0111
X_3^2	−3.96	154.39	<0.0001	−4.31	117.32	<0.0001	−4.35	113.13	<0.0001	3.38	62.58	<0.0001
X_4^2	−13.69	1,846.63	<0.0001	−13.72	1,188.46	<0.0001	−13.54	1,094.09	<0.0001	−2.06	23.33	0.0003
$X_1 \times X_2$	0.99	5.99	0.0282	0.38	0.56	0.4685	0.68	1.73	0.2100	1.21	4.94	0.0432
$X_1 \times X_3$	−1.79	19.36	0.0006	−1.22	5.75	0.0310	−1.00	3.64	0.0770	−3.27	36.22	<0.0001
$X_1 \times X_4$	−1.28	9.92	0.0071	−0.80	2.51	0.1355	−0.63	1.45	0.2487	3.72	46.73	<0.0001
$X_2 \times X_3$	0.11	0.067	0.7995	0.17	0.11	0.7495	0.43	0.67	0.4260	−1.79	10.87	0.0053
$X_2 \times X_4$	−0.49	1.43	0.2517	−0.22	0.19	0.6707	−0.06	0.011	0.9175	1.06	3.77	0.0725
$X_3 \times X_4$	−1.29	10.11	0.0067	−0.90	3.12	0.0991	−0.90	2.98	0.1063	−2.25	17.14	0.0010

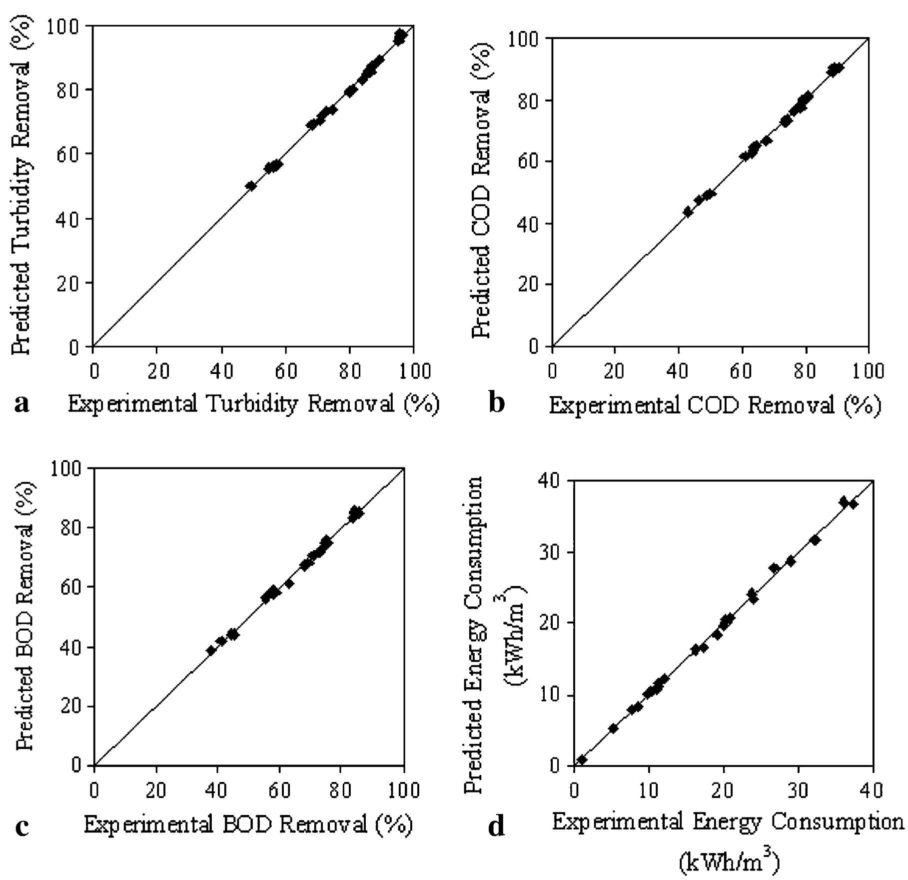


Fig. 6 Comparison of experimental and predicted **a** percentage turbidity removal, **b** percentage COD removal, **c** percentage BOD_5 removal and **d** energy consumption by RSM

results for turbidity removal, COD removal, BOD_5 removal and energy consumption are shown in Table 4. The large value of F (604.85 for percentage turbidity removal, 397.76 for percentage COD removal, 377.19 for percentage BOD_5 removal and 147.59 for energy consumption) indicates that most of the variation in the response can be explained by the regression equations. The associated p value is used to estimate whether F is large enough to indicate statistical significance. The values of Prob > $F < 0.05$ indicated that model terms are significant. Prob > F values are <0.0001 for turbidity removal, COD removal, BOD_5 removal and energy consumption, which indicate that terms are significant at 95 % probability level (Prakash Maran et al. 2013b). Coefficients of determination (R^2) were found as 0.9983, 0.9975, 0.9974 and 0.9933, whereas adjusted R^2 were determined as 0.9967, 0.9950, 0.9947 and 0.9865 for percentage turbidity, COD, BOD_5 removal and energy consumption, respectively, indicating a good fit for both dependent variables. For all equations, adequate precision signal to noise ratio is greater than 4, which is desirable for sound models (Sridhar et al. 2012). The ANOVA results show that j, pH_0, c and t are the significant factors that affect the turbidity, COD, BOD_5 removal and energy consumption by electrocoagulation. The model intercept coefficient, which does not depend on any factor, shows that the average percentage turbidity, COD, BOD_5 removal and energy consumption are 95.56 %, 88.65 %, 83.66 % and 16.21 kWh/m^3, respectively, and these values are independent of the factors set in the experiment. The ANOVA shows that the model chosen to explain the relationship between the factors and the response is suitable. Table 2 shows the relationship between the actual and the predicted values of Y, and it can be inferred that the residuals for the prediction of each response are minimum, supporting that the results of ANOVA are correct. The second-order polynomial equations for turbidity removal efficiency (Y_1), COD removal efficiency (Y_2), BOD_5 removal efficiency (Y_3) and energy consumption (Y_4) in terms of uncoded factors are given by Eqs. 3, 4, 5 and 6, respectively.

$$\begin{aligned} Y_1 = {} & -149.66531 + 2.40129X_1 + 44.57000X_2 + 22.76333X_3 \\ & + 3.06706X_4 - 0.041208X_1^2 - 3.67615X_2^2 - 3.95833X_3^2 \\ & - 0.034224X_4^2 + 0.049625X_1X_2 - 0.17850X_1X_3 \\ & - 0.0063875X_1X_4 + 0.0525X_2X_3 \\ & - 0.012125X_2X_4 - 0.0645X_3X_4 \end{aligned} \quad (3)$$

$$\begin{aligned} Y_2 = {} & -159.15656 + 2.61621X_1 + 45.46708X_2 \\ & + 22.13333X_3 + 2.96615X_4 - 0.045417X_1^2 - 3.74260X_2^2 \\ & - 4.30917X_3^2 - 0.034289X_4^2 + 0.018875X_1X_2 \\ & - 0.12150X_1X_3 - 0.0040125X_1X_4 + 0.082500X_2X_3 \\ & - 0.0055X_2X_4 - 0.04475X_3X_4 \end{aligned} \quad (4)$$

$$\begin{aligned} Y_3 = {} & -163.99750 + 2.57679X_1 + 46.30313X_2 + 21.14542X_3 \\ & + 2.89027X_4 - 0.048021X_1^2 - 3.87615X_2^2 - 4.35458X_3^2 \\ & - 0.033855X_4^2 + 0.034250X_1X_2 - 0.0995X_1X_3 \\ & - 0.0031375X_1X_4 + 0.21375X_2X_3 \\ & - 0.001375X_2X_4 - 0.045X_3X_4 \end{aligned} \quad (5)$$

$$\begin{aligned} Y_4 = {} & +0.41667 + 0.068519X_1 - 3.07523X_2 + 1.81366X_3 \\ & + 0.38022X_4 + 0.010949X_1^2 + 0.31192X_2^2 \\ & + 3.37616X_3^2 - 0.00515336X_4^2 + 0.060417X_1X_2 \\ & - 0.32708X_1X_3 + 0.018576X_1X_4 - 0.89583X_2X_3 \\ & + 0.026389X_2X_4 - 0.11250X_3X_4 \end{aligned} \quad (6)$$

The models predictions are compared with the experimental observations are shown in Fig. 6a–d. From the figures, it is observed that the developed models are adequate for the prediction of each response because the data points lie close to the diagonal line.

Conclusion

The effects of various operational parameters such as electrode distance, stirring speed, NaCl concentration, pH and current density on electrocoagulation using aluminum electrodes have been examined. The percentage turbidity, COD and BOD_5 removal were found to increases with increasing electrolyte concentration, current density and stirring speed. The results showed that optimal operating conditions were found to be an initial pH of 6, current density of 20 mA/cm^2, stirring speed of 150 rpm, NaCl concentration of 1.5 g/l and electrolysis time of 30 min. This experimental study clearly showed that under the optimal conditions, about 96 % turbidity, 89 % COD and 84 % BOD_5 were successfully removed. Energy consumption and operating cost were found to decrease with increasing stirring speed and NaCl concentration and increased with an increase in current density, electrode distance, pH and electrolysis time. Under the optimal operating conditions, the energy consumption and operating cost were found to be 16.24 kWh/m^3 and 2.7 US \$/m^3, respectively. A Box–Behnken design was successfully employed for experimental design and analysis of results for maximizing the turbidity, COD and BOD_5 removal and for minimizing energy consumption. Analysis of variance showed a high coefficient of determination value $R^2 = 0.9983$ for turbidity, $R^2 = 0.9975$ for COD, $R^2 = 0.9974$ for BOD_5 and $R^2 = 0.9933$ for energy consumption, thus ensuring a satisfactory fit of the second-order regression model with that of the experimental data. The results of this study showed that electrocoagulation could be effectively used for the treatment of effluent generated from the egg processing plants.

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