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ORIGINAL ARTICLE

Removal of pollutants with determination of power consumption from landfill leachate wastewater using an electrocoagulation process: optimization using response surface methodology (RSM)

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Abstract

Treatment of landfill leachate wastewater by electrocoagulation process using an aluminium electrode was investigated in a batch electrochemical cell reactor. Response surface methodology based on central composite design was used to optimize the operating parameters for the removal of % color and % total organic carbon (TOC) together with power consumption from landfill leachate. Effects of three important independent parameters such as current density (X_1), inter-electrode distance (X_2) and solution pH (X_3) of the landfill leachate sample on the % color and % TOC removal with power consumption were investigated. A quadratic model was used to predict the % color and % TOC removal with power consumption in different experimental conditions. The significance of each independent variable was calculated by analysis of variance. In order to achieve the maximum % color and % TOC removal with minimum of power consumption, the optimum conditions were about current density (X_1)—5.25 A/dm^2, inter-electrode distance (X_2)—1 cm and initial solution of effluent pH (X_3)—7.83, with the yield of color removal of 74.57%, and TOC removal of 51.75% with the power consumption of 14.80 kWh/m^3. Electrocoagulation process could be applied to remove pollutants from industrial effluents and wastewater.

Keywords Electrocoagulation · Landfill leachate · Color and TOC removal · Power consumption · Central composite design

Introduction

Increases in world population and new patterns of consumption have resulted in huge production of wastes that are usually discarded in sanitary landfills, since this is relatively simple and inexpensive (Azni 2009). Landfill leachate wastewater can be generated from precipitation, surface run-off, infiltration or intrusion of groundwater percolating though the landfill (Li et al. 2011). Various types of pollutants can be found in sanitary landfill leachate such as organic and inorganic compounds, toxic and heavy metals (Fernandes et al. 2015). Due to its complex and recalcitrant composition, the sanitary landfill leachate represents a significant source of pollutants. Discharge of the landfill leachate into the environment can have a detrimental effect on aquatic life, cause infertility of soil and mutagenic effect on humans as well as affecting the ecological balance. The treatment of landfill leachate wastewater is difficult due to the discharge standards, variable composition and its high pollutant load. Several treatment methods have been used to treat the landfill leachate, such as biological processes (Li et al. 2017; Zhang et al. 2016; Robinson. 2017), membrane processes (Ahn et al. 2002), coagulation and flocculation methods (Wang et al. 2015; Liu et al. 2012), flotation methods (Adlan et al. 2011), adsorption and chemical precipitation (Hur and Kim 2000; Erabee et al. 2017), osmosis (Iskander et al. 2017), chemical oxidation (Derco et al. 2010), Fenton and electrochemical (Vallejo et al. 2012), advanced oxidation techniques Hu et al. 2011; Zhang et al. 2012; Chys et al. 2015) and electro-Fenton (Zhang et al. 2014). However, these methods are found to have certain shortages such

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as operating cost, transfer of one phase to another, lower pollutant removal efficiency and decreasing the process performance. Thus, it is essential to design and develop an economic and effective treatment method for removing pollutants from landfill leachate wastewater.

Electrochemical processes have shown high effectiveness in eliminating persistent pollutants from landfill leachate wastewater (Ricordel and Djelal 2014; Panizza et al. 2010) and have some advantages such as energy efficiency, versatility and cost-effectiveness (Juttner et al. 2000). Among the electrochemical methods, electrocoagulation process appears to be the most effective substitution for the conventional coagulation and flotation process as it can deal with pollutants with a variety of compositions (Wang et al. 2009; Butler et al. 2017).

Electrocoagulation is a simple process in terms of its equipment setup and easy-to-handle methodology, high efficiency with production of less sludge (Kalyani et al. 2009; Sharma and Chopra 2017). It can be operated at ambient temperature and pressure. Electrocoagulation is an electrolytic process involving the dissolution of the sacrificial anodes, made of aluminium (Al), upon application to a current between the two electrodes to supply ions to the wastewater, allowing suspended, emulsified or dissolved contaminants to form agglomerates (Fernandes et al. 2015). The coagulating ions are produced in situ and the successive stages for current theory of electrocoagulation are described as follows: firstly, the formation of coagulants induced by the electrolytic oxidation of the sacrificial anode followed by generation of metal hydroxides; secondly, destabilization of the contaminants and particulate suspension and breaking of emulsions; and lastly, the aggregation or coalescence of the destabilized phases to form larger and separable agglomerates (Moreno-Casillas et al. 2007). Hydrogen (H_2) bubbles that evolve from the cathode surface are adsorbed onto the suspended particles. The separation of the solid matter is achieved either by flotation upon the adsorption of H_2 bubbles, or allowing the solid to settle down due to its higher density which the buoyant force produced by the H_2 bubbles is insufficient to lift the suspended solid (Zodi et al. 2009). The mechanism of electrocoagulation process depends on the chemistry of the aqueous medium, especially the conductivity. The mechanism of ion formation is proposed as in Eqs. (1)–(4) below using aluminium electrode (Fernandes et al. 2015).

Anodic reaction:

$$\mathrm{Al_{(s)}} \rightarrow \mathrm{Al^{3+}_{(aq)}} + 3\mathrm{e^-} \tag{1}$$

Cathodic reaction:

$$2\mathrm{H_2O_{(l)}} + 2\mathrm{e^-} \rightarrow \mathrm{H_{2(g)}} + 2\mathrm{OH^-_{(aq)}} \tag{2}$$

Chemical reaction that takes place in the aqueous medium:

$$\mathrm{Al^{3+}_{(aq)}} + \mathrm{OH^-_{(aq)}} \rightarrow \mathrm{Al(OH)_{3(s)}} \tag{3}$$

Overall reaction is given by:

$$\mathrm{Al^{3+}_{(aq)}} + 3\mathrm{H_2O_{(l)}} \rightarrow \mathrm{Al(OH)_{3(s)}} + 3\mathrm{H^+_{(aq)}} \tag{4}$$

Based on the literature review, many studies on the electrocoagulation process were carried out by varying one factor while the other factors are kept constant (Chopra and Sharma 2013; Sharma and Chopra 2017). However, this approach consumes more time and response surface methodology (RSM) can be an alternative to overcome this problem. Most of the previous studies only focused on the performance of electrocoagulation process such as % COD and % color removal (Saravanan et al. 2010; Janpoor et al. 2011), but did not emphasize on the % TOC removal with power consumption. It was important to determine the power consumption of electrocoagulation process in order to determine its operating cost and feasibility. The objective of this research work is to identify the optimum operating parameters for the removal of pollutants from landfill leachate using central composite design (CCD).

RSM is used to optimize the parameters chosen for the electrocoagulation process. It is a regression analysis used to predict the value of dependent variable based on the controlled values of independent variables. Numerous experiment combinations can be generated within a short period of time, thus allowing researchers to know whether the tested parameter has a significant impact on the research work (Liu et al. 2012; Butler et al. 2017). In many technical fields, it is common that the output variable (Y) exists with a set of predicted variables or the input variables ($X_1, X_2, X_3, \dots X_k$). The output variable is a function of input variable together with the error presence in the model, usually written as $Y = f\left(X_1, X_2, X_3, \dots X_k\right) + \in$, where f is the unknown surface response which is normally described by a first-order or second-order polynomial, while $\in$ is the error in the model. Generally, the first- and second-order models are given as in Eqs. (5) and (6):

$$Y = \beta_o + \sum_{j=1}^{k} \beta_j X_j + \in \tag{5}$$

$$Y = \beta_o + \sum_{j=1}^{k} \beta_j X_j + \sum_{j=1}^{k} \beta_{jj} X_j^2 + \sum_{j=1}^{k-1} \sum_{i=2}^{k} \beta_{ji} X_j X_i + \in_i \tag{6}$$

where X_i and X_j are coded independence variables and β_j, β_{jj}, and β_{ji} ($i = 1, 2, \dots, k$; $j = 1, 2, \dots, k$) are the regression coefficients. A first-order model is used to describe the flat surface, while the curve surface is described by a second-order model, or also known as a quadratic model. A quadratic model is often adequate for RSM in most cases. Besides, the knowledge of statistical fundamentals, regression modeling techniques and optimization methods is required in fitting the response surface model.

The main objective of optimization was to maximize the % color and % TOC removal while minimizing the power consumption by varying operating parameters such as current density (X_1), inter-electrode distance (X_2) and initial pH (X_3). Design of Expert (DoE) Software (11) was used to optimize and study the combined effect of three selected parameters. Each independent variable was coded at three levels between − 1 and + 1, where the variables current density (X_1), inter-electrode distance (X_2) and initial effluent pH (X_3) were set in the range of 1.05–6.25A/dm^2, 1.0–4.0 cm, and pH 5–11, respectively, as indicated in Table 1.

Materials and methods

Materials

Landfill leachate wastewater was collected from Jeram sanitary landfill, Selangor, Malaysia. Various parameters such as pH, chemical oxygen demand (COD), total organic carbon (TOC), color and odor were analyzed and tabulated in Table 2. COD was measured by closed reflux method using potassium dichromate (Spectroquant® TR320); TOC was measured using the TOC analyzer (TOC-LCSH/CPH) and color was determined using UV/Vis spectrophotometer (Spectroquant Pharo®300). Chemicals such as $K_2Cr_2O_7$, NaCl, H_2SO_4, NaOH, etc., were used and supplied from YEW SII SIE lab analytics supplies, Malaysia.

Methods

Experimental setup for the electrocoagulation process is shown in Fig. 1. Experiment was carried out in a batch reactor with a capacity of 500 mL (YEW SII SIE lab analytics supplies, Malaysia). Initial COD concentration of the landfill leachate wastewater was diluted into 1500 ppm. Aluminium (Al) electrodes with dimension of 16 cm × 6 cm were used for both anode and cathode. The effective electrode surface area was 48 cm^2 and the inter-electrode distance between an anode and cathode was varied from 1 to 4 cm. The pH of the landfill leachate was measured by pH meter (Elico; Model LI120) and varied from pH 5 to 11 using H_2SO_4 and NaOH solutions. The electrodes were connected to a direct current (DC) power supply (APLAB Ltd; Model L1606) with aid of crocodile clips for supplying constant current density, varying from 1.05 to 6.25 A/dm^2. 3 g/L NaCl was added in the solution to improve the electrical conductivity of the solution and a magnetic stirrer was used at 500 rpm to increase the probability of particle collision to improve the efficiency of the electrocoagulation process. After the required experimental condition, sample was taken after 1 h of electrolysis time and the filtered using filter paper. Then, the sample was immediately analyzed for color and TOC removal. The removal of the color was determined using the UV/Vis spectrophotometer (Spectroquant® TR320) and TOC was determined using the TOC analyzer (TOC-LCSH/CPH).

Table 1 Coded and actual values of the variables of the design of experiments for the electrocoagulation process

Variable	Unit	Factor	Levels		
			− 1	0	1
Current density	A/dm^2	X_1	1.05	3.65	6.25
Inter-electrode distance	cm	X_2	1.0	2.50	4.0
Initial effluent pH	–	X_3	5	8	11

Table 2 Characterization of landfill leachate wastewater

Parameter	Value
COD (mg/L)	7225
TOC (mg/L)	4000
Absorbance (Au)	4.534
pH	8.1
Color	Dark brown
Smell	Pungent ammonia smell
Temperature (°C)	33
Turbidity (NTU)	230

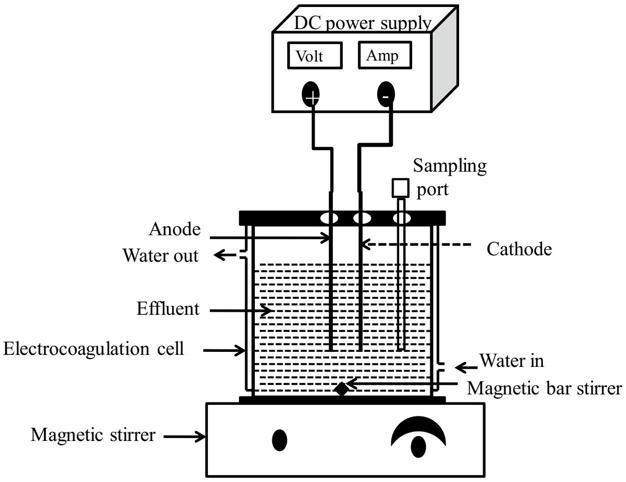

Fig. 1 Experimental setup for the electrochemical process

Analysis

Color and TOC removal (%)

The % color and % TOC removal were calculated using Eqs. (7) and (8):

$$\text{Color removal efficiency (\%)} = \frac{([\text{Abs}_i]) - ([\text{Abs}_t])}{\text{Abs}_i} \times 100 \quad (7)$$

where Abs_i and Abs_t are absorbance of samples at initial and reaction time t for a corresponding wavelength $\lambda_{\max}$.

$$\text{TOC removal efficiency (\%)} = \frac{([\text{TOC}_i]) - ([\text{TOC}_t])}{\text{TOC}_i} \times 100 \quad (8)$$

where TOC_i is the initial of TOC and TOC_t is the TOC at any reaction time, t (mg/L).

Power consumption

The power consumption for the removal of % color and % TOC from landfill leachate using the electrocoagulation process was calculated using Eq. (9):

$$P = \frac{\text{VIt}}{V_{\text{R}}}, \left(\frac{\text{kWhr}}{\text{m}^3}\right) \quad (9)$$

where V is the cell voltage (V), I is the applied current (A), t is the electrolysis time (h) and V_{R} is the volume of wastewater used (m^3).

Results and discussion

Central composite design

A 3-factor and 3-level CCD was used to optimize the operating parameters of an electrocoagulation process on the responses such as the % color and % TOC removal efficiency as well as the power consumption. The total number of experiment combinations was 20, with 6 replications at the design central to determine the pure error. The total number of runs, experimental conditions, response of % color removal, % TOC removal and power consumption together with the predicted values are shown in Table 3.

Evaluation of experimental results with design of experiments

The % color removal (Y_1), % TOC removal (Y_2) and power consumption (Y_3) are the function of operating parameters such as current density (X_1), inter-electrode distance (X_2) and initial pH (X_3) at constant electrolysis time of 1 h. The

Table 3 Experimental design matrix and response based on the experimental runs and predicted values on the color removal (%), TOC removal (%) and power consumption proposed by the CCD

Run	X_1	X_2	X_3	Color removal (%)		TOC removal (%)		Power consumption (kWhr/m^3)	
	A/dm^2	cm	–	Actual	Predicted	Actual	Predicted	Actual	Predicted
1	1.05	1	5	45.25	46.67	33.5	33.36	5.12	4.81
2	6.25	1	5	70.05	70.63	48.25	48.34	15.50	14.54
3	1.05	4	5	41.5	39.97	21.35	21.46	15.75	14.16
4	6.25	4	5	58.75	58.13	35.75	35.64	36	38.18
5	1.05	1	11	40.5	41.57	27	27.02	3.21	3.18
6	6.25	1	11	60.15	62.13	38.5	38.30	10	10.58
7	1.05	4	11	34.4	34.27	18.25	18.06	9.5	9.46
8	6.25	4	11	50	49.03	28.5	28.54	35	34.30
9	1.05	2.5	8	50.5	49.68	39	39.20	4.5	5.0
10	6.25	2.5	8	70	69.04	51.75	51.93	28	26.91
11	3.65	1	8	73.5	68.46	46	46.23	16.15	20.02
12	3.65	4	8	55.3	58.56	35.25	35.40	36.4	36.56
13	3.65	2.5	5	60	60.16	42	42.05	28	28.68
14	3.65	2.5	11	55	53.06	35	35.33	21	24.35
15	3.65	2.5	8	64	64.59	47	46.87	30	28.66
16	3.65	2.5	8	64	64.59	47	46.87	30	28.66
17	3.65	2.5	8	64	64.59	47	46.87	30	28.66
18	3.65	2.5	8	64	64.59	47	46.87	30	28.66
19	3.65	2.5	8	64	64.59	47	46.87	30	28.66
20	3.65	2.5	8	64	64.59	47	46.87	30	28.66

quadratic model regression equations were obtained from Design Expert Software as shown in Eqs. (10), (11) and (12):

$$Y_1 = 64.59 + 9.68X_1 - 4.95X_2 - 3.55X_3 - 1.45X_1X_2 - 0.85X_1X_3 - 0.15X_2X_3 - 5.23X_1^2 - 1.08X_2^2 - 7.98X_3^2 \tag{10}$$

$$Y_2 = 46.87 + 6.36X_1 - 5.42X_2 - 3.36X_3 - 0.2X_1X_2 - 0.93X_1X_3 + 0.74X_2X_3 - 1.30X_1^2 - 6.05X_2^2 - 8.18X_3^2 \tag{11}$$

$$Y_3 = 28.66 + 8.64X_1 + 8.27X_2 - 2.17X_3 + 3.57X_1X_2 + 0.21X_1X_3 + 0.02X_2X_3 - 10.39X_1^2 - 0.37X_2^2 - 2.14X_3^2 \tag{12}$$

Experimental data were analyzed by sequential model sum of squares and model summary statistics to obtain the most suitable models among various models such as linear, interactive, quadratic and cubic. The results are tabulated in Tables 4, 5 and 6 for the % color removal, % TOC removal and power consumption, respectively. From Tables 4, 5 and 6, it can be seen that quadratic model

Table 4 Sequential model sum of squares and model summary statistics for percentage color removal (%)

Sequential model sum of squares

Source	Sum of square	*df*	Mean square	*F* value	*P* value	Prob > *F*
Mean vs total	65,998.56	1	65,998.56			
Linear vs mean	1308.07	3	436.02	7.97	0.0018	
2FI vs linear	22.78	3	7.59	0.1158	0.9492	
Quadratic vs 2FI	797.58	3	265.86	48.69	< 0.0001	Suggested
Cubic vs quadratic	47.36	4	11.84	9.80	0.0084	Aliased
Residual	7.25	6	1.21			
Total	68,181.60	20	3409.08			

Model summary statistics

Source	Std. dev.	R^2	Adjusted R^2	Predicted R^2	PRESS	
Linear	7.39	0.5992	0.5240	0.2672	1599.81	
2FI	8.10	0.6096	0.4295	− 1.9223	6379.45	
Quadratic	2.34	0.9750	0.9525	0.8068	421.76	Suggested
Cubic	1.10	0.9967	0.9895	− 3.0784	8903.33	Aliased

df degree of freedom

Table 5 Sequential model sum of squares and model summary statistics for percentage TOC removal (%)

Sequential model sum of squares

Source	Sum of square	*df*	Mean square	*F* value	*P* value	Prob > *F*
Mean vs total	30,584.02	1	30,584.02			
Linear vs mean	811.25	3	270.42	4.57	0.0170	
2FI vs linear	11.52	3	3.84	0.0534	0.9830	
Quadratic vs 2FI	934.10	3	311.37	6249.77	< 0.0001	Suggested
Cubic vs quadratic	0.1545	4	0.0386	0.6743	0.6338	Aliased
Residual	0.3437	6	0.0573			
Total	32,341.39	20	1617.07			

Model summary statistics

Source	Std. dev.	R^2	Adjusted R^2	Predicted R^2	PRESS	
Linear	7.69	0.4616	0.3607	0.0406	1685.96	
2FI	8.48	0.4682	0.2227	− 2.8880	6832.70	
Quadratic	0.2232	0.9997	0.9995	0.9976	4.22	Suggested
Cubic	0.2393	0.9998	0.9994	0.7597	422.31	Aliased

df degree of freedom

Table 6 Sequential model sum of squares and model summary statistics for power consumption, kWhr/m^3

Sequential model sum of squares

Source	Sum of square	*df*	Mean square	*F* value	*P* value	Prob > *F*
Mean vs total	9862.57	1	9862.57			
Linear vs mean	1477.19	3	492.40	8.64	0.0012	
2FI vs linear	102.45	3	34.15	0.5488	0.6577	
Quadratic vs 2FI	724.69	3	241.56	28.69	< 0.0001	Suggested
Cubic vs quadratic	47.00	4	11.75	1.89	0.2308	Aliased
Residual	37.22	6	6.20			
Total	12,251.11	20	612.56			

Model summary statistics

Source	Std. Dev.	R^2	Adjusted R^2	Predicted R^2	PRESS	
Linear	7.55	0.6184	0.5469	0.3484	1556.40	
2FI	7.89	0.6613	0.5050	− 0.9141	4571.90	
Quadratic	2.90	0.9647	0.9330	0.8584	672.72	Suggested
Cubic	2.49	0.9844	0.9507	− 18.1442	45,726.80	Aliased

df degree of freedom

gives the highest R^2, adjusted R^2 and predicted R^2 values when compared to the other models after excluding the cubic model. The cubic model cannot be used for further modeling of experimental data because it was found to be aliased. An aliased model was a result of insufficient experiments run to independently estimate all the terms of the model. Thus, not all parameters can be estimated and it is unwise for further studying an aliased model. The highest order polynomial from the sequential model sum of squares, quadratic model, was selected for modeling the treatment of landfill leachate using electrocoagulation process where the additional terms are significant and the model is not aliased.

Adequacy of the model tested for % color removal, % TOC removal and power consumption

The significance and adequacy of the model was analyzed by the analysis of variance (ANOVA) and the results for % color removal, % TOC removal and power consumption are given in Tables 7, 8 and 9, respectively. The *F* test of the quadratic models gives a small *P* value (< 0.05), which indicates that all the models were significant and could be used to predict the outcome for the electrocoagulation process. From Table 7, it can be seen that for the % of color removal, the linear coefficient of the current density (X_1), inter-electrode distance (X_2) and initial pH (X_3)

Table 7 ANOVA of the second-order polynomial equation for percentage color removal, (%)

Source	Sum of squares	*df*	Mean square	*F* value	*P* value	Prob > *F*
Model	2128.44	9	236.49	43.31	< 0.0001	Highly significant
X_1	937.02	1	937.02	171.61	< 0.0001	Highly significant
X_2	245.03	1	245.03	44.87	< 0.0001	Highly significant
X_3	126.03	1	126.03	23.08	0.0007	Significant
X_1X_2	16.82	1	16.82	3.08	0.1098	
X_1X_3	5.78	1	5.78	1.06	0.3278	
X_2X_3	0.1800	1	0.1800	0.0330	0.8596	
X_1^2	75.27	1	75.27	13.79	0.0040	Significant
X_2^2	3.22	1	3.22	0.5894	0.4604	
X_3^2	175.20	1	175.20	32.09	0.0002	Significant
Residual	54.60	10	5.46			
Lack of fit	54.60	5	10.92			
Pure error	0.0000	5	0.0000			
Cor total	2183.04	19				

df degree of freedom

Table 8 ANOVA of the second-order polynomial equation for percentage TOC removal (%)

Source	Sum of squares	*df*	Mean square	*F* value	*P* value	Prob > *F*
Model	1756.87	9	195.21	3918.22	< 0.0001	Highly significant
X_1	405.13	1	405.13	8131.85	< 0.0001	Highly significant
X_2	293.22	1	293.22	5885.58	< 0.0001	Highly significant
X_3	112.90	1	112.90	2266.06	< 0.0001	Highly significant
X_1X_2	0.3200	1	0.3200	6.42	0.0296	Significant
X_1X_3	6.84	1	6.84	137.39	< 0.0001	Highly significant
X_2X_3	4.35	1	4.35	87.34	< 0.0001	Highly significant
X_1^2	4.66	1	4.66	93.61	< 0.0001	Highly significant
X_2^2	100.73	1	100.73	2021.91	< 0.0001	Highly significant
X_3^2	183.89	1	183.89	3690.98	< 0.0001	Highly significant
Residual	0.4982	10	0.0498			
Lack of fit	0.4982	5	0.0996			
Pure error	0.0000	5	0.0000			
Cor total	1757.36	19				

df degree of freedom

Table 9 ANOVA of the second-order polynomial equation for power consumption, (kWhr/m^3)

Source	Sum of squares	*df*	Mean square	*F* value	*P* value	Prob > *F*
Model	2304.33	9	256.04	30.40	< 0.0001	Highly significant
X_1	746.84	1	746.84	88.69	< 0.0001	Highly significant
X_2	683.43	1	683.43	81.16	< 0.0001	Highly significant
X_3	46.92	1	46.92	5.57	0.0399	Significant
X_1X_2	102.10	1	102.10	12.12	0.0059	Significant
X_1X_3	0.3445	1	0.3445	0.0409	0.8438	
X_2X_3	0.0032	1	0.0032	0.0004	0.9848	
X_1^2	296.97	1	296.97	35.26	0.0001	Significant
X_2^2	0.3700	1	0.3700	0.0439	0.8382	
X_3^2	12.62	1	12.62	1.50	0.2490	
Residual	84.21	10	8.42			
Lack of fit	84.21	5	16.84			
Pure error	0.0000	5	0.0000			
Cor total	2388.54	19				

df degree of freedom

and the quadratic coefficient of current density (X_1^2) and initial pH (X_3^2) were significant, with p value less than 0.05. For the % TOC removal, it can be observed from Table 8 that the linear coefficient of current density (X_1), inter-electrode distance (X_2), initial pH (X_3), interaction effect of current density (X_1) with inter-electrode distance (X_2), current density (X_1) with initial pH (X_3) and inter-electrode distance (X_2) with initial pH (X_3) and quadratic coefficient of current density (X_1), inter-electrode distance (X_2^2 and initial pH (X_3^2) were significant variables. For the power consumption from Table 9, the linear effect of current density (X_1), inter-electrode distance (X_2) and initial pH (X_3), interaction effect of current density (X_1) with inter-electrode distance (X_2) and the quadratic effect of the current density (X_1^2) were found to be significant. "Adeq Precision" measures the signal-to-noise ratio; it was desirable to obtain a value greater than 4. The signal-to-noise ratio was 22.01, 214.61 and 18.60 which is greater than 4 for the % color removal, % TOC removal and power consumption, respectively. Thus, the second-order model can be used to navigate the design space. Adequacy check is crucial to make sure the approximation model can give adequate approximation to prevent poor and misleading result.

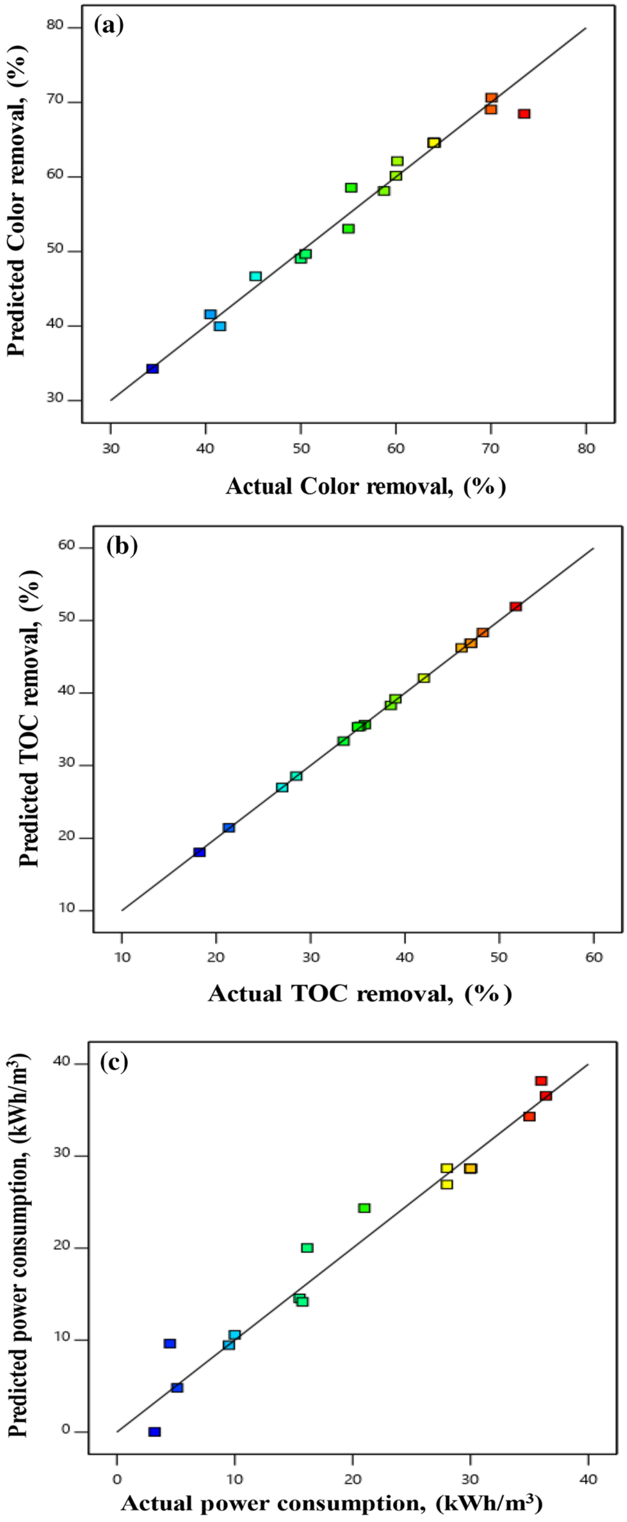

Fig. 2 Plot for relationship between experimental and predicted value for **a** % color removal, **b** % TOC removal and **c** power consumption

Experimental versus predicted

The comparison between experimental and predicted value is shown in Table 3 and Fig. 2a–c. From Fig. 2, it can be seen that the model-predicted values matched the experimental data in which all the points are closed to the diagonal line. The ANOVA analysis showed that all the three quadratic models were significant ($p < 0.05$) and can be used to predict the % of color removal, % TOC removal, and also power consumption. The quality of predicted points was verified by the R^2 value, where the R^2 values were 0.97, 0.99 and 0.96 for % of color removal, % TOC removal and power consumption, respectively.

Combined effect of operating parameters for % color removal, % TOC removal and power consumption

The effect of operating parameters in estimating the maximum % color removal, % TOC removal and minimum of power consumption with respect to each variable and the impact of each operating parameter on the output are discussed as following. The electrolysis time for the electrocoagulation process was 1 h and the initial COD concentration of the leachate is diluted to 1500 ppm to visualize a better and clearer result.

Combined effect of current density (X_1) and inter-electrode distance (X_2)

The combined effect of current density (X_1) and inter-electrode distance (X_2) on % color and % TOC removal with power consumption was tested by varying X_1 from 1.05 to 6.25 A/dm^2 and X_2 from 1 to 4 cm and the results are tabulated in Table 3 and plotted in Fig. 3a–c. From Fig. 3a, b, it can be observed that the % color removal and % TOC removal were increased as the current density increased, but after the optimum value, further increase in current density does not help in improving the removal of % color and TOC (Kalyani et al. 2009). The increase in current density resulted in the production of large amount of Al^{3+} ions via anodic metal dissolution, more H_2 bubbles was formed at the cathode, which are profitable for the separation or flotation process (Ozyonar and Karagozoglu 2015). From Fig. 3c, it can be seen that the increase in current density caused an increase in the power consumption. This is because, an increase in current density caused an increase in cell voltage, which had a direct impact on the power consumption of the electrochemical process. Since a proportional relationship was established between the current density and power consumption, it is necessary to identify the optimum value of current density to reduce the power consumption and operating cost (Heidmann and Calmano 2008).

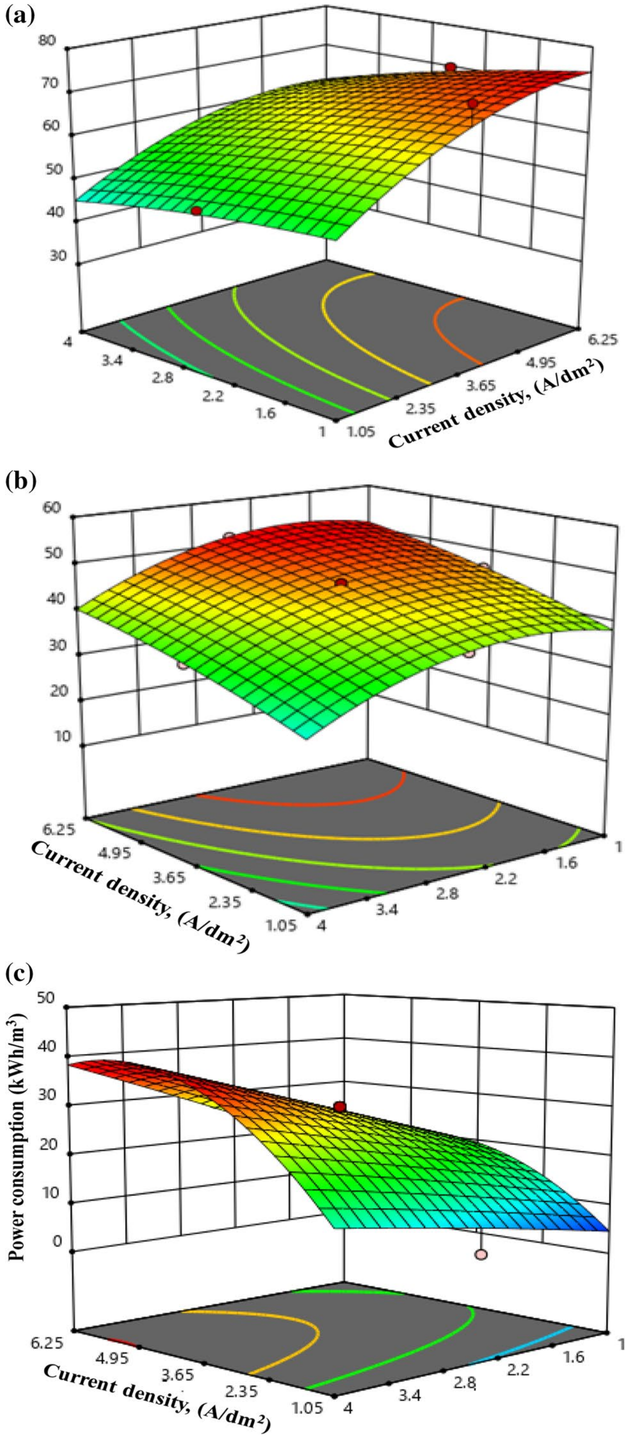

Fig. 3 Combined effect of current density (X_1) and inter-electrode distance (X_2) on **a** % color removal, **b** % TOC removal and **c** power consumption

Inter-electrode distance (X_2) was varied from 1 to 4 cm in order to study its effect on the % color removal, % TOC removal and power consumption. From Fig. 3a, b, it was seen that the % color removal and % TOC removal was decreased as the inter-electrode distance increased from 1 to 4 cm at any value of current density in the range of 1.05–6.25 A/dm^2. This is because there is an increased in ohmic voltage drop as the distance between the anode and cathode was increased (Khandegar and Saroha 2013). Besides, Faraday's law also stated that the amount of oxidized metal decreased as the gap between the electrodes was increased. However, Fig. 3c shows that the power consumption increased as the inter-electrode distance increased. This was due to the fact that there is more resistance offered when the electrodes gap increase and power consumption is directly proportional to the cell voltage (Ricordel and Djelal 2014).

Combined effect of initial pH (X_3) and current density (X_1)

Initial pH of the landfill leachate (X_3) was adjusted in the range of pH 5–11 to investigate the impact of the pH on the % color removal, % TOC removal and power consumption. The result is given in Table 3 and plotted in Fig. 4a–c. From Fig. 4a, b, it can be seen that the % color and % TOC removal were increased at effluent pH from 5 to 7.5; however, further increase in pH from 7.5 to 11 decreased the removal efficiency. This can be explained by the formation of aluminium species formed in the reaction. For the Al electrodes in acidic medium, monomeric hydroxometallic cation $Al(OH)_3$ is formed. At neutral medium, both polymeric hydroxometallic cations and metal hydroxides precipitates coexist while at higher pH or alkali medium, the net charge on the surface of the amorphous metal hydroxide precipitate changes from positive to negative and the polymeric cations will only remain in the solution. More $^{\bullet}OH$ can be formed in neutral condition compared to acidic and alkaline mediums in the electrocoagulation process (Modirshahla et al. 2007; Kobya et al. 2003). However, from Fig. 4c, it can be seen that the initial pH of the leachate had no impact on the power consumption for the electrocoagulation process. This is because the conductivity of the landfill leachate did not change as a result of pH adjustment; thus, 3 g/L of NaCl or mediator has been added in before starting the experiment.

Optimization

The main objective of this study is to determine the optimal operating parameters for the maximum % color and % TOC removal with the minimum of power consumption from landfill leachate wastewater using the electrocoagulation process. The results were optimized using the regression equation of RSM based on CCD. While optimizing, all the input variables such as current density (X_1), inter-electrode distance (X_2) and initial pH (X_3) were selected as within the range while the output variables such as % color removal and % TOC removal were maximized with power consumption

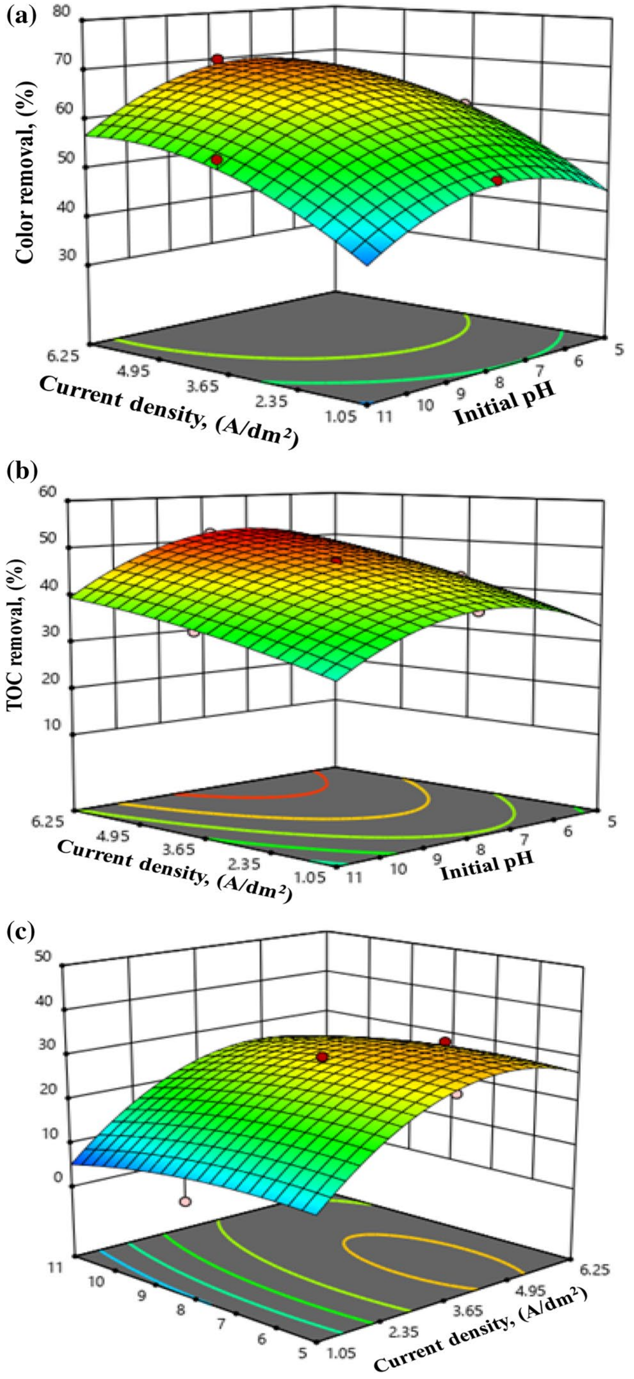

Fig. 4 Combined effect of initial pH (X_3) and current density (X_1) on **a** % color removal, **b** % TOC removal and **c** power consumption

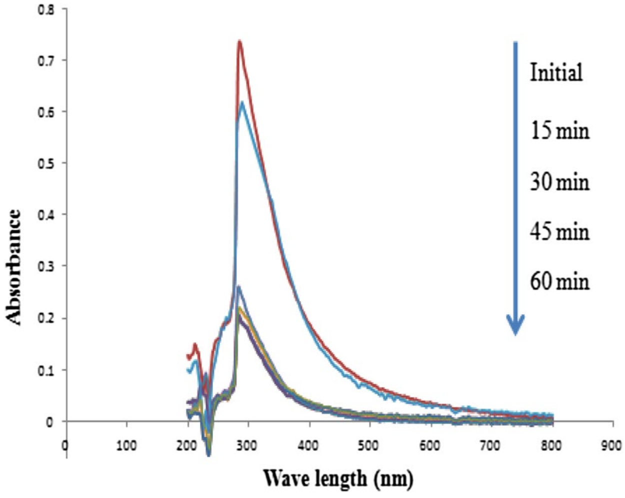

Fig. 5 Spectra of landfill leachate wastewater, recorded before and after the electrocoagulation process at different electrolysis times

minimized. The optimized operating parameters are as following: current density (X_1)—5.25 A/dm^2, inter-electrode distance (X_2)—1 cm and initial pH (X_3)—7.83 with expected result of color removal to be 74.57%, TOC removal of 51.74% and 14.80 kWh/m^3 for power consumption. A mean value of 75.20% for color removal, 50.90% for TOC removal and 13.75 kWh/m^3 for power consumption was obtained experimentally, which is closed to the predicted result. From the expected and actual result, it can be said that there was good correlation between them which indicates that the central composite design could be effectively used to optimize the electrocoagulation process parameters.

Instrumental analysis

The absorption spectra of before and after treatment of electrocoagulation process were analyzed using the UV/Vis spectrophotometer (Spectroquant Pharo®300) to study the color removal rate from landfill leachate wastewater. The absorbance spectrum for the landfill leachate effluent had an absorbance peak at 284 nm which belongs to the coloring agent. From Fig. 5, it can be seen that there was reduction in absorbance of peak with increasing electrolysis time. It might be attributed that, the color of the landfill leachate wastewater was continuously reduced with increasing electrochemical reaction time.

Conclusion

This study investigated the removal of % color and % TOC using electrocoagulation process in a real landfill leachate wastewater. An empirical relationship between the output and independent variables was obtained based on the experimental data and it was expressed by the quadratic model using RSM. The results showed that, the maximum % color removal, % TOC removal and minimum of power consumption were 74.57, 51.75 and 14.80 kWh/m^3, respectively, obtained at the optimum conditions of current density (X_1) of 5.25 A/dm^2, inter-electrode distance (X_2) of 1 cm and initial effluent pH of 7.83. Based on the experimental results, an empirical relationship between the response and independent variables was obtained and expressed by the second-order polynomial equation. The ANOVA analysis showed a high coefficient of determination value, thus ensuring a satisfactory adjustment of the second-order regression model with the experimental data. This technology could be used effectively for the removal of pollutants from industrial effluents and wastewater.

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