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Optimization of electrocoagulation system for municipal wastewater treatment

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ABSTRACT

This study describes the design and construction of a wastewater treatment package using an electrocoagulation system and examines the optimization of the system. For optimization, the factors including number of electrodes, gap between electrodes, current density, time and pH were optimized using the Design-Expert Software. According to response surface tests, the optimum range was selected to maximize the removal efficiency of chemical oxygen demand, total dissolved solids, and biological oxygen demand. This range for current density was between 49.8 and 80 A/m^2, the process time was from 9 to 15 min, and the pH between 6.5 and 8, the number of electrodes was 2 pairs and the gap between the electrodes was 1 cm. It was observed that with increasing the current density, time, number of electrodes and the electrode gap, energy consumption increases and pH changes do not affect energy consumption. The optimum range in which the treatment efficiency is above 80% and the energy consumption is less than 5 kWh/m^3 of the sewage is current density between 49.8 and 63 A/m^2, time between 9 and 11 min, pH 6.5–8, the number of electrodes 1 pair and the gap between the electrodes, 1 cm. In this optimal range, the energy consumption per one cubic meter of wastewater is between 4.5 and 5 kWh.

Keywords: Electrocoagulation; Design–Expert; Purification package; Urban wastewater

1. Introduction

The process of coagulation and flocculation in water and wastewater treatment is currently carried out through chemical means and adding organic coagulants, minerals, etc. to the water and sewage samples [1]. High operating costs and adverse environmental impacts have led to the discontinued use of chemical coagulation in water and wastewater industry [2,3]. In order to find other suitable alternatives to replace the chemical treatment process, several approaches have been taken into consideration in recent years [4,5]. One such process is electrocoagulation, which is a significant innovation in the water and wastewater industry [6,7]. Electrocoagulation involves the production of coagulants in situ using electrolysis of aluminum or iron electrodes. In this process, two or more electrodes, usually made of iron or aluminum, are used. The two electrodes are placed in an electrolyte environment, and on the other side they are connected to the electrical current source. In the anode, which is the victim's electrodes, iron or aluminum metal ion is released and hydrogen bubbles are generated around the cathode [8–10].

The advantages of this system can be fully automated for continuous operation, reducing the number of process units in the treatment plant, resulting in a sharp reduction in the required surface of the treatment plant and reducing the operating costs of the treatment system [11,12]. Due to the lack of chemical addition in this method, the amount of waste is almost equal to the same amount of the deposited materials in the separation section. Therefore, the amount of sludge resulting from this process is much less than other methods. Due to lack of chemicals in the system recycling and reuse of wastewater is possible. The system has the ability to reduce heavy metals such as arsenic, cadmium, lead, nickel and zinc up to 99%–95%, reduce soluble silica, clay particles and other suspended materials up to

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98%. In addition, this process also reduces bacterial counts from 110 million to 2,700 cells per milliliter, and oil waste from various human industries, up to 99%–95%, with considerable decrease in fats, oils, and grease, total dissolved solids (TDS), total suspended solids (TSS) and biological oxygen demand (BOD) [12].

This method was first performed in 1889 for the treatment of sewage by mixing it with seawater before performing electrodialysis at a sewage treatment plant in London. In 1909, in the United States, this method was used to treat sewage by dissolving aluminum and iron metals at the anode [13–15]. Since the early 1970s, this method has been used especially in Europe and Russia for the initial purification of water needed for the plating and completion of metals as well as for water treatment [12]. In North America, waste water treatment plants are more widely used for wood and paper industry, mines and metal processing [13–15]. With the proper design, these systems can be widely used in treating all types of wastewater, groundwater and surface water. Electrocoagulation is a common process for the coagulation and removal of contaminants, organic matter and waste metal ion and elimination of water hardness [8]. Most of the articles related to electrocoagulation are for wastewater treatment [9] and eliminating the hardness of water [10]. In general, the electrocoagulation process is done in three stages [16].

- The victim's electrodes are oxidized to produce coagulant.
- The particle suspension then becomes unstable and the emulsion breakdown occurs.
- The destabilized phase re-aggregates and forms the masses.

The reactions in the anode and cathode occur as follows:

Anode:

- Generation of metal ions

$$Fe \leftrightarrow Fe^{+2} + 2e^{-} \quad (1)$$

$$Fe \leftrightarrow Fe^{+3} + 3e^{-} \quad (2)$$

- Hydrolysis of metal ions and generation of metal hydroxides and polyhydroxides

$$Fe + 6H_2O \rightarrow Fe(H_2O)_4(OH)_{2(aq)} + 2H^{+1} + 2e^{-1} \quad (3)$$

$$Fe + 6H_2O \rightarrow Fe(H_2O)_3(OH)_{3(aq)} + 3H^{+1} + 3e^{-1} \quad (4)$$

- Electrolysis of water:

Anode:

$$2H_2O_{(1)} \rightarrow O_{2(g)} + 4H^{+} + 4e^{-} \quad (5)$$

Cathode:

$$2H^{+} + 2e^{-} \rightarrow H_{2(g)}\uparrow \quad (6)$$

This process is very efficient and can result in the removal of 80%–95% of waste material [16]. The energy consumption in this method is favorable in comparison with centrifuges, which is between 0.3 and 2 kWh/m^3 and improves when processing saline waters. Electrocoagulation is suggested as an attractive method for treating effluents with high TDS and TSS. This method has the advantage of showing that coagulants are not always required [17]. The electrocoagulation on the bench scale has been shown to be an appropriate method and removes 95% of the contaminants present in the suspension with an energy consumption of 0.3 kWh/m^3.

The use of wastewater treatment packages that operate independently of environmental conditions such as temperature can be used for low-population and difficult-to-pass areas, where it is not possible to implement structural wastewater treatment systems; also due to the low temperature, it is not possible to set up systems such as active sludge. Therefore, in this research, a new electrocoagulation package has been designed and manufactured, which in addition to being portable, offers higher efficiency than the previous routine systems due to its novel design. Another innovation in this research is the simultaneous optimization of system performance and energy consumption for municipal wastewater treatment. In this study, municipal wastewater treatment was investigated using an electrocoagulation system and the optimal treatment conditions were determined.

2. Materials and methods

2.1. Design and construction of a pilot

The desired pilot design was made according to Fig. 1. As seen in the figure, the inside of the tank is divided into four sections using pairs of separator walls (No. 9). After the effluent entry section (No. 5), there is a separating ditch, which is lower than the edge of the reservoir by about 10 cm and adheres to the bottom of the reservoir. At a distance of 10 cm, there is another wall with 10 cm space from the bottom of the reservoir. This wall prevents turbulence inside the chamber when the effluent enters. The same pattern

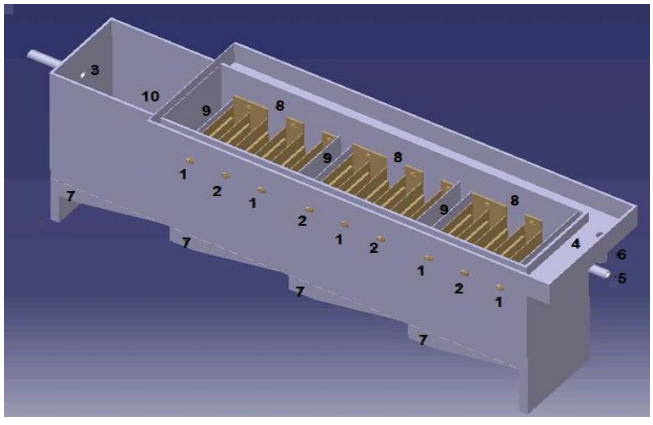

Fig. 1. An overview of the electrocoagulation system. The different sections of this design are as follows: (1) positive device connection screws, (2) the negative pole connecting screws, (3) refined waste effluent, (4) floating material collecting stones, (5) raw wastewater input, (6) output of floating materials, (7) decanter at the end of each section, (8) different parts of electrical purification, (9) insulating plates for separating the electric filtration sections, (10) suspension section.

is repeated to the end of the tank. In the final part, the treated wastewater overflows from the first partition wall and then enters the sedimentation section from the lower part of the second separator wall and after discharge the effluent is removed from the exhaust pipe (No. 3).

2.2. Response surface method

The response surface method is a collection of mathematical and statistical techniques useful for modeling, analyzing many issues and providing a solution as a function of several variables. The purpose of this method is to optimize the response by determining the optimal range of factors that affect the process and analyzing the relationship between these factors, while minimizing the number of experiments and, consequently, spending less cost and time [18]. At this point, the DX10 software was used. In the response surface method, among the various designs that the software has provided, the D-optimal method has usability for optimization 1 to 30 factors, and minimizes the variance of estimated coefficients for the model. This method also provides less testing than other response-surface methods.

2.3. Electrocoagulation test

In order to perform optimization tests, after experimental design, according to Tables 1 and 2, the experiments were performed in the order provided by the software. For this purpose, the initial conditions of the wastewater, such as chemical oxygen demand (COD), BOD, TSS and TDS were determined. Afterwards the initial pH of the effluent was first adjusted according to the value provided in Table 3 for each experiment. For example, for experiment number 1, the initial pH was increased to 9. For this purpose, a normal HCl and NaOH were used. Then 250 cc of pH regulated sewage was poured into the electrocoagulation chamber and according to Table 3 the number and internal distance of the electrodes were adjusted. To test No. 1, a pair of electrodes with an internal distance of 2 cm was used. After placing the electrodes at the intended distance in the chamber and turning on the power supply, the desired current density (Table 3) was created by changing the voltage, which was 43.95 A/m^2 for test number 1. After the specified time, which was determined by the software for each test, the power supply was turned off (e.g., 15 min for test number 1). Later, samples were left for 15 min, to allow clusters of suspended particles created in the liquid due to turbulence generated by the current, to float to the surface. Samples were taken from the transparent liquid below, and the factors mentioned earlier were measured. The efficiency of eliminating the mentioned factors was determined by obtaining the difference. The same conditions were met for the next 52 experiments according to the proposed design.

2.4. Calculation of energy consumption

To calculate the amount of energy consumed, in each test, the voltage is measured on the power supply unit. The duration is specified in minutes for all tests. The amount of current is different for different current densities and the number of electrodes and is summarized in Table 3.

The volume used in the tests is 250 mL. Therefore, the amount of energy consumed in kWh/m^3 of wastewater is obtained from the following equation [19]:

$$\text{Energy Consumption}\left(\frac{\text{kwh}}{\text{m}^3}\right)=\frac{10^6\times\text{time}(\text{min})\times\text{Current}(\text{A})\times\text{Voltage}(\text{V})}{250\times60\times10^3} \quad (7)$$

Table 1
Range of factors influencing the coagulation process

Factor code	Name	–1 Level	+1 Level
A	C.D., A/m^2	7.9	80
B	Time, min	5	15
C	pH	5	9

Fig. 2. A view of the system built at the laboratory and pilot level.

Table 2
Results of optimization tests

Run	*A*: c.d	*B*: time	*C*: pH	*D*: Electrode number	*E*: gap	TDS reduction	COD reduction	BOD reduction	Voltage	Energy consumption
	(A/m^2)	(min)		(pairs)	(cm)	(%)	(%)	(%)	(v)	(kWh/m^3)
1	43.95	15	9	1	2	68.78	65.9	64.867	29.9	9.867
2	7.9	5	7	1	2	10.02	8.2	4.66	8.2	0.66
3	80	5	9	1	1	62.81	64.9	63.069	27.9	3.069
4	7.9	15	5	2	2	31.69	36.9	34.554	6.9	4.554
5	80	15	9	2	1	91.12	90.2	82.572	34.2	22.572
6	80	15	5	1	2	70.49	71.2	63.596	41.2	13.596
7	43.95	10	5	1	1	53.01	49.1	44.202	19.1	4.202
8	43.95	5	5	1	2	43.71	38.1	32.211	28.1	2.211
9	7.9	5	5	2	2	19.13	15.8	13.476	15.8	3.476
10	80	5	7	2	2	86.9	89.3	88.646	39.3	8.646
11	43.95	15	7	2	2	75.91	71.7	73.2	31.7	13.2
12	80	15	5	2	1	86.89	82.4	81.384	32.4	21.384
13	43.95	5	9	2	1	58.38	55.3	53.366	15.3	3.366
14	80	5	9	1	2	51.37	51.3	44.543	41.3	4.543
15	61.975	10	7	1	2	79.46	75.5	77.81	35.5	7.81
16	7.9	15	7	1	1	37.23	33.8	31.65	3.8	1.65
17	43.95	5	5	1	2	48.09	46.5	42.915	26.5	2.915
18	7.9	5	5	1	1	19.67	17.3	14.803	7.3	0.803
19	7.9	15	9	2	1	48.63	44.5	42.97	4.5	2.97
20	80	10	5	2	2	75.41	71.1	68.084	41.1	18.084
21	7.9	5	9	1	1	30.8	34.1	30.451	4.1	0.451
22	7.9	15	9	2	1	45.92	46.1	44.026	6.1	4.026
23	80	5	9	1	2	51.32	48.5	44.565	41.5	4.565
24	7.9	5	9	2	2	14.81	18.3	14.026	18.3	4.026
25	7.9	5	9	2	2	13.23	14.8	10.056	4.8	1.056
26	80	15	9	2	1	91.61	82	81.12	32	21.12
27	80	5	5	1	1	63.72	62.9	62.519	22.9	2.519
28	80	5	9	2	1	70.37	71	69.02	41	9.02
29	7.9	10	5	2	1	40.98	44.4	31.936	4.4	1.936
30	61.975	10	7	1	2	84.66	80	78.8	40	8.8
31	43.95	15	9	1	2	74.86	69	69.57	29	9.57
32	7.9	5	7	1	2	13.26	16	10.66	6	0.66
33	80	5	9	1	1	63.39	58.15	53.0965	28.15	3.0965
34	7.9	15	5	2	2	30.09	25.1	23.366	5.1	3.366
35	80	15	5	1	2	74.34	71.4	73.662	41.4	13.662
36	43.95	10	5	1	1	51.33	50.4	44.488	20.4	4.488
37	7.9	5	5	2	2	19.47	15.4	11.188	5.4	1.188
38	80	5	7	2	2	82.75	75.5	77.81	35.5	7.81
39	43.95	15	7	2	2	78.06	70.15	69.899	30.15	19.899
40	80	15	5	2	1	91.81	84.7	16.302	24.7	16.302
41	43.95	5	9	2	1	58.96	51.1	49.042	41.1	9.042
42	7.9	15	7	1	1	38.71	33.3	31.089	3.3	1.089
43	7.9	5	5	1	1	19.47	16.5	12.915	26.5	2.915
44	80	10	5	2	2	78.76	71.1	68.084	41.1	18.084
45	7.9	5	9	1	1	28.15	24.5	20.495	4.5	0.495
46	80	5	5	1	1	59.29	50.2	53.322	30.2	3.322
47	80	5	9	2	1	67.94	61	59.02	41	9.02
48	7.9	10	5	2	1	36.28	33.2	31.408	3.2	1.408
49	43.95	5	5	1	2	53.19	49.4	43.234	29.4	3.234
50	7.9	15	9	2	1	50.28	43.9	42.574	3.9	2.574
51	80	5	9	1	2	51.42	44	44.51	41	4.51
52	7.9	5	9	2	2	16.2	14.9	11.078	4.9	1.078
53	80	15	9	2	1	91.63	85.1	76.566	25.1	16.566

Table 3
Amount of electrical current (A)

Electrodes No.	Current densities (A/m^2)			
	80	61.98	43.95	7.9
1	0.61	0.47	0.33	0.06
2	1.25	0.94	0.67	0.12

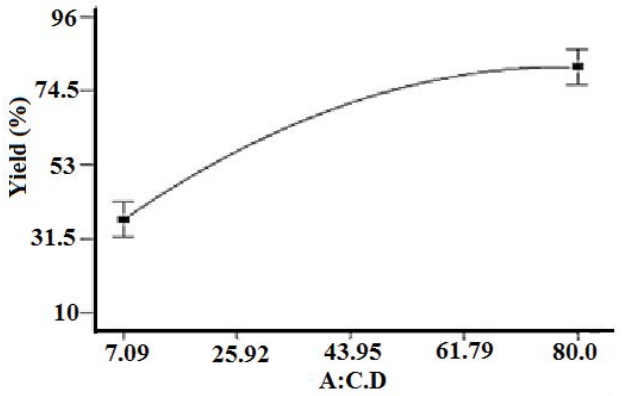

Fig. 3. Effect of current density on the efficiency of sewage treatment.

2.5. Cost of power consumption

Power consumption is a function of the electric current, voltage and time. In the electrocoagulation process, for an optimal current density, the voltage is controlled by the solute resistance. Given Eqs. (1) and (2), the amount of cost needed to treat 1 m^3 of waste water can be obtained from the following equation [19].

$$\frac{\$}{\mathrm{m}^3}=\left(\frac{\$}{\mathrm{kWh}}\right)\times\left(\frac{\mathrm{Voltage(V)}\times\mathrm{Current(A)}\times\mathrm{time(h)}}{1{,}000}\right)\times\left(\frac{1}{\mathrm{m}^3}\right) \tag{8}$$

2.6. Cost of corrosion of the electrode

The metal content is separated from the electrode, at the certain time and the current density can be calculated by Faraday's law, according to the following equation. This equation can be expressed in simpler form, according to the following equation [19].

$$m = c\times I\times t \tag{9}$$

In this regard, m is the mass of the abandoned metal with the Gram unit, c is the electrochemical component of the metal, which is a constant value for each metal and its unit is grams per ampere hour, I is the electrical current with amperes unit and t is the time (h).

The following equation is correct, assuming that all current is spent on releasing the metal in solution. While some amount of the current is also spent on hydrogen production. The above equation is corrected as follows [20]:

$$m = c\times I\times t\times \eta \tag{10}$$

η is the current efficiency and is expressed as a percentage. The current efficiency (η) is obtained from the following equation [19].

$$\eta=\frac{\Delta M\ \text{experimental}}{\Delta M\ \text{theoritical}}\times 100 \tag{11}$$

This equation is based on the comparison of the weight of the electrode that was reduced in the test and the weight of the electrode, which is reducing by Faraday's law. The theory of ΔM is obtained from the same equation. In this study, due to the ease of doing the calculations, η is considered equal to one [19].

$$\frac{\$}{\mathrm{m(kg)}}=\left(\frac{\$}{\mathrm{kg}}\right)\times\left(\frac{c\times I\times t}{1{,}000}\right) \tag{12}$$

2.7. Wastewater tests

2.7.1. Biological oxygen demand

The BOD of water was determined by titration method. Samples in duplicate were taken and one set was fixed immediately with azide reagent and the initial DO is measured titrimetrically. The other set of samples was kept in BOD incubator at 20°C for 5 d and then analyzed for final DO.

2.7.2. Chemical oxygen demand

COD was determined by titration method. 50 mL of water sample was taken in three 100 mL flask (in triplicates). Triplicates of blank were also prepared. 5.0 mL of $K_2Cr_2O_7$ solution was added to each of the six flasks. The flasks were kept at 100°C in the water bath for 1 h. The samples were allowed to cool for 10 min and then 5.0 mL of KI was added. 10 mL of H_2SO_4 was added in each flask, contents of each flask were titrated with 0.1 M $Na_2S_2O_3$ till the appearance of pale yellow colour. 1.0 mL of starch solution was then added due to which the solution turns pale yellow to blue colour. The sample was titrated again until the blue colour disappeared completely.

Calculation:

$$\text{COD of the sample (mg/L)} = 8\times C\times (B-A)/S \tag{13}$$

where C = concentration of titrant (mL/L), A = volume of titrant used for blank (mL), B = volume of titrant used for sample (mL), S = volume of water sample taken.

2.7.3. Total dissolved solids

100 mL of filtered sample was taken in the previously heated, cooled and weighed evaporating dish. Residue was heated at 103°C–105°C in hot air oven till all the water evaporated. Final weight of dishes was noted, the final weight was taken after cooling in desiccators. Total dissolved solids were calculated by following formula:

Total dissolved solids mg/L = $(A - B \times 1{,}000 \times 1{,}000)/V$ (14)

where A = final weight of the dish in g, B = initial weight of the dish in g, V = volume of sample taken in mL.

3. Results and discussion

3.1. Results of process optimization

To optimize the process more precisely, response surface and D-optimal design were used. The results of response surface tests are given in Table 3. As shown in Table 4, the proposed software model is quadratic and meaningful. Factors whose p-value is less than 0.05 are affected factors [18]. The current density, time, pH, electrodes number, the distance between the electrodes, the interaction between the current density and the distance between the electrodes, the interaction of current density and pH, the time and pH interaction, the interaction between the number of electrodes and pH are important factors.

In optimization experiments, p-value is used to determine the significant effect of parameters on the process. This value, which is specified for each parameter in the software, indicates the effect of the parameter on the system response, so if the value of this scale is less than 0.05, it indicates the significant effect of this factor on the system response. In this study, all factors except pH had a significant effect and the effect of pH is only insignificant with very little deviation (Table 4). Usually, factors with a p-value greater than 0.1 are removed from the model.

As can be seen in the table above, the p-value for the model is less than 0.0001, which indicates the importance of the model. On the other hand, adequate precision (comparison between the predicted range using the model and the average prediction error of more than 4 is desirable) is equal to 27.041 and more than 4, which is also a desirable factor for the model. The adjusted-R^2 and predicted-R^2 values are 0.9601 and 0.9739, respectively, which are in agreement. In addition, the p-value for the lack of fit equal to 0.9687 indicates that this test is insignificant for the model, which is desirable.

To ensure that there are no significant errors in the laboratory data and the proposed model, a number of statistical tests were performed. The first test is to check the normal probability function of the residuals. The results of this test show that the laboratory points are located around the line, so it can be ensured that there is no abnormal term in the system error. If the pattern of points has a nonlinear state, it indicates a non-normality error. The next test looks at the quality of the model. In this test, the predicted

Table 4
Analysis of variance for sewage treatment efficiency in response surface experiments

ANOVA for response surface quadratic model						
Source	Sum of squares	df	Mean square	F-value	p-value	Prob. > F
Model	30,832.957	18	1,712.9421	70.56183	<0.0001	Significant
A-c.d	16,415.863	1	16,415.863	676.2245	<0.0001	
B-time	1,605.827	1	1,605.827	66.1494	<0.0001	
C-pH	94.10106	1	94.10106	3.876338	0.0572	
D-number	1,039.3567	1	1,039.3567	42.81459	<0.0001	
E-gap	414.28705	1	414.28705	17.06587	0.0002	
AB	70.946977	1	70.946977	2.922544	0.0965	
AC	103.53408	1	103.53408	4.264916	0.0466	
AD	0.0362662	1	0.0362662	0.001494	0.9694	
AE	155.6481	1	155.6481	6.411667	0.0161	
BC	449.1016	1	449.1016	18.5	0.0001	
BD	2.0971558	1	2.0971558	0.086389	0.7706	
BE	64.964935	1	64.964935	2.676124	0.1111	
CD	227.14783	1	227.14783	9.356981	0.0043	
CE	47.878932	1	47.878932	1.972294	0.1693	
DE	22.952784	1	22.952784	0.945502	0.3377	
A^2	777.61409	1	777.61409	32.03253	<0.0001	
B^2	1.632381	1	1.632381	0.067243	0.7970	
C^2	503.84616	1	503.84616	20.75511	<0.0001	
Residual	825.37583	34	24.27576			
Lack of fit	14.18		2.84	0.16	0.9687	Not significant
TStd. dev.	4.927044	R-Squared	0.9739			
Mean	54.03321	Adj. R-Squared	0.9601			
C.V. %	9.118548	Pred. R-Squared	0.9362			
PRESS	2,019.575	Adeq.	27.041			

points are plotted using the model according to the laboratory data. In a suitable model, the resulting points are placed around the 45-degree line. Since the data are located around the 45-degree line, it can be said that the model predicts the data well to the desired extent. Another test is the error chart based on the predicted values. In this test, the dots must follow a random scattering pattern. If the pattern of the points is funnel-shaped and the amount of deviation increases, it indicates that the points must be transferred to another form. On the other hand, in a suitable model, the amount of deviation is in the range of ±3 of the standard deviation. The model obtained in this study also has a pattern of scattered behavioral points and none of the predicted points are outside the range.

Three models of response surface method (RSM) including the first-order (FO) response surface model, the two-way interactions (TWI) model and the full second-order (FSO) model are usually selected to fit of data, and finally the best model is selected using analysis of variance (ANOVA). The model with higher R^2 and insignificant lack of fit will be selected as appropriate model [21–23].

FSO model with a higher R^2 and also an insignificant lack of fit (0.96) indicated superiority than the rest. Therefore, self-organizing map (SOM) model was selected as proper model for prediction of sewage treatment. Multiple R-squared for FO, TWI, FSO was 0.83, 0.88 and 0.97, respectively. Lack of fit for FO and TWI model were significant (<0.0001 and 0.005, respectively) while the lack of fit of FSO with the value of 0.967 was insignificant.

Finally, four equations regarding different states; the number of electrodes and the distance between the electrodes were presented for non-coded value:

- Number of electrodes = 1 pair, distance = 1 cm

$$\text{Yield} = -110.03557 + 1.49850 \times \text{c.d} - 1.82920 \times \text{time} + 36.38503 \times \text{pH} + 0.010259 \times \text{c.d} \times \text{time} - 0.027667 \times \text{c.d} \times \text{pH} + 0.43727 \times \text{time} \times \text{pH} - 9.02052\text{ E-}003 \times \text{c.d}^2 - 0.024870 \times \text{time}^2 - 2.61242 \times \text{pH}^2 \quad (15)$$

- Number of electrodes = 2 pair, distance = 1 cm

$$\text{Yield} = -75.52856 + 1.49619 \times \text{c.d} - 1.92969 \times \text{time} + 33.43398 \times \text{pH} + 0.010259 \times \text{c.d} \times \text{time} - 0.027667 \times \text{c.d} \times \text{pH} + 0.43727 \times \text{time} \times \text{pH} - 9.02052\text{ E-}003 \times \text{c.d}^2 - 0.024870 \times \text{time}^2 - 2.61242 \times \text{pH}^2 \quad (16)$$

- Number of electrodes = 1 pair, distance = 2 cm

$$\text{Yield} = -120.29635 + 1.63269 \times \text{c.d} - 1.21013 \times \text{time} + 35.20897 \times \text{pH} + 0.010259 \times \text{c.d} \times \text{time} - 0.027667 \times \text{c.d} \times \text{pH} + 0.43727 \times \text{time} \times \text{pH} - 9.02052\text{ E-}003 \times \text{c.d}^2 - 0.024870 \times \text{time}^2 - 2.61242 \times \text{pH}^2 \quad (17)$$

- Number of electrodes = 2 pair, distance = 2 cm

$$\text{Yield} = -89.12960 + 1.63038 \times \text{c.d} - 1.31061 \times \text{time} + 32.25792 \times \text{pH} + 0.010259 \times \text{c.d} \times \text{time} - 0.027667 \times \text{c.d} \times \text{pH} + 0.43727 \times \text{time} \times \text{pH} - 9.02052\text{ E-}003 \times \text{c.d}^2 - 0.024870 \times \text{time}^2 - 2.61242 \times \text{pH}^2 \quad (18)$$

Due to the coefficients of the factors in the above equation as well as the *p*-value rate of each factor, the importance of each factor on the purification rate can be understood. Given the above equation, it can be concluded that the single effects of all parameters on the purification rate except time are positive. This means that by increasing all parameters except time, the purification rate increases. According to the coefficients, the most important factors are current intensity, time, number and the distance between the electrodes and pH, respectively. In addition, according to the equation, it was found that all factors have a curvature effect on the purification rate.

3.1.1. Effect of current density

With increasing the current density, efficiency has increased (Fig. 3). This can be explained by the fact that the amount of aluminum released from the anode increases with increasing current density, according to Faraday's law [24]. Faraday's first law states that the mass separated from the electrodes is directly proportional to the amount of electricity passing through the electrodes. When aluminum ions increase in the environment, the surface of the coagulation contact and the number of active sites increases, which improves the accumulation of particles and formation of the clot [25–27]. Also, by increasing the current density, the bubble increases and the size of the bubbles decreases, which makes the clusters float faster on the liquid surface.

3.1.2. Effect of time

As shown in Fig. 4, efficiency increases as time increases. As the time increases, more metal ions are released in the sewage medium, and particles also have more chance of contact with metal ions, resulting in more clusters and increased COD, TDS and BOD reduction efficiency [28].

3.1.3. Effect of pH

As shown in Fig. 5, with increasing pH to a value between 6 and 8, the efficiency increases and then decreases. In surface-response experiments, pH was considered at three levels and it was observed that there should be an optimal point between pH 5 and 9. This can be due to the fact that the pH is suitable for producing more aluminum hydroxide in a liquid medium at a point between 6 and 8. In some studies, a pH of about 6.5 is said to be optimum [25,29]. In other electrochemical process such as E-Fenton treatment process, also the initial pH is an important factor, which can influence the overall removal efficiencies of turbidity and COD. In this regard researchers indicated that the percentage removal of turbidity and COD were increased with increasing pH up to 7 [30,31].

3.1.4. Effect of the number of electrodes and the distance between them

Increasing the number of electrodes and reducing the distance between them increases the efficiency (Fig. 6). With increasing number of electrodes, more metal ions are released in the sewage system, thus increasing the contact

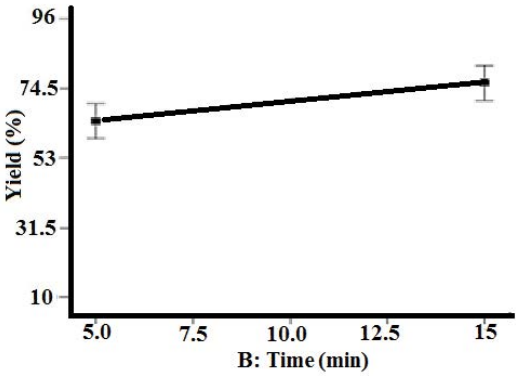

Fig. 4. Effect of time on the efficiency of sewage treatment.

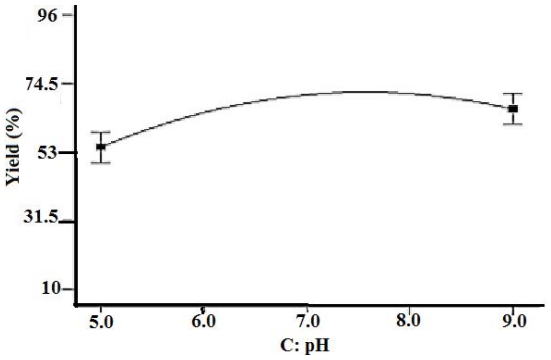

Fig. 5. Effect of pH on the efficiency of sewage treatment.

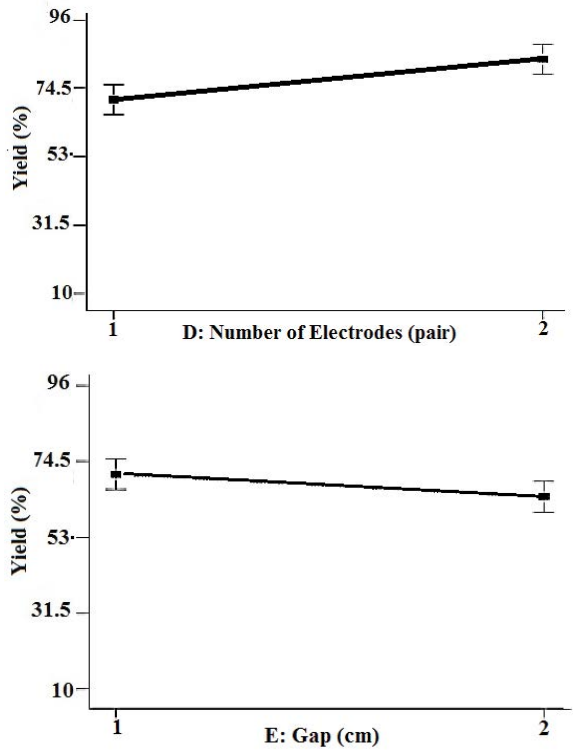

Fig. 6. Effect of the number of electrodes and the distance between them on the efficiency of sewage treatment.

surface of the materials with increasing material removal efficiency [25,26]. Of course, this increases the cost of the electrical coagulation process. Thirugnanasambandham and Shine [27] also reported that one of the most important parameters affecting the variance of industrial wastewater treatment is the electrode's surface. They have indicated that the hydrogen gas yield increases in a linear fashion with increasing electrode surface area [27]. By increasing the distance between the electrodes, the efficiency decreases. This can be due to the low ion displacement, during the coagulation process, and the less collision of particles with ions. However, with the increase in the distance between the electrodes, the electrical coagulation process can be done on a larger volume, but the need for higher voltages will increase the amount of energy and thus the cost [25,26].

3.1.5. Interaction effect of current density and pH

As shown in Fig. 7, pH in the range of 5 and 9 has a greater effect on yield. For outputs above 80%, the current density range is 49 to 80 and the pH is 6.5 to 2.8.

3.1.6. Interaction of time and pH

According to Fig. 8, for a yield above 8%, the pH range is between 6.5 and 8.8 and the time range is about 9–15 min. With regard to these three Contour charts, it can be said that for high efficiency, an appropriate range of current density should be between 49.8 and 80 A/m^2, the time range is from 9 to 15 min and the pH range is from 6.5 to 8.

3.1.7. Interaction of qualitative factors

As mentioned earlier, because of the limitations of the electrocoagulation chamber and the impossibility of changing the levels of the number of electrodes and the distance between them, these two factors were considered as qualitative factors in response surface experiments. In the ANOVA, it was stated that only the interaction between the current density and the distance between the electrodes,

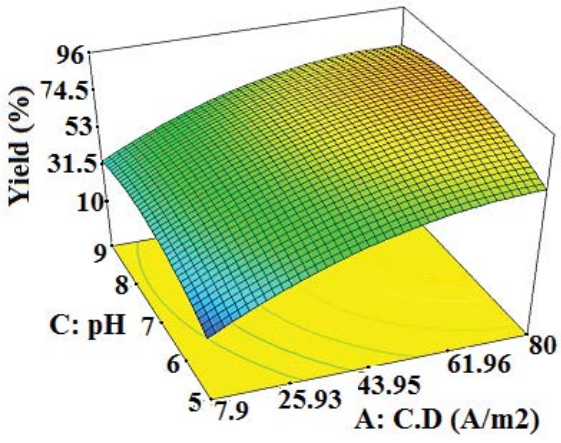

Fig. 7. Effect of current density and pH on the efficiency of sewage treatment, where the time is 10 min and the number of electrodes 1 pair and their distance 1 cm.

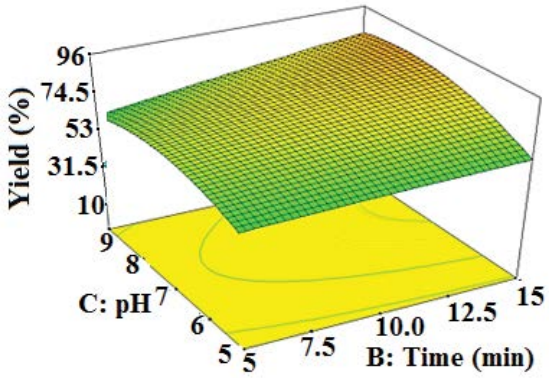

Fig. 8. Effect of time and pH on the efficiency of sewage treatment in the case of a current density of 49.8 and the number of electrodes is 1 pair and their distance is 1 cm.

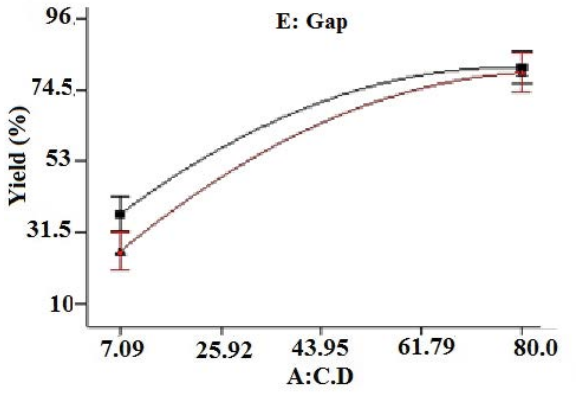

Fig. 9. Interaction of the current density and the distance between the electrodes on the efficiency of sewage treatment. ■: 1 cm gap, ▲: 2 cm gap.

and the interaction of the number of electrodes and pH have significant effects [32,33].

As shown in Fig. 9, when the distance between the electrodes is 2 cm (red line), with increasing the current density, the efficiency increases with a slope greater than the distance of 1 cm (black line). When the distance between the electrodes is high, the displacement of the ions in the environment is low. At low current densities, the amount of metal ion released in the environment is also low, so the efficiency is very low, but with increasing current density, more ions and bubbles are released, which is resulting in higher efficiency [32,33].

On the graph of Fig. 10, when the pH is 5 (black line), with increasing the number of electrodes, the efficiency increases with a slope higher than that of the pH of 9 (red line). When the number of electrodes is higher, more hydroxide ions are released from the cathode. This increases the pH of the solution. When the initial pH of the sewage is 5, this increase in pH causes the solution to be closer to the optimal pH of the aluminum hydroxide, thus increasing the removal efficiency of the process with a sharp slope. But when the initial pH of the sewage is 9, this increase in pH has a negative effect, and on the other

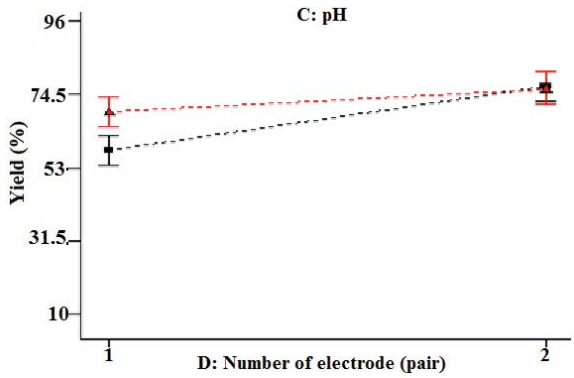

Fig. 10. Effect of electrode number and pH on the efficiency of sewage treatment. ■ pH: 5, ▲ pH: 9.

hand, increasing the number of electrodes has a more positive effect on the process, resulting in a slight increase in gradient efficiency [34].

3.2. Find the stable conditions of the process

The terms of a stable condition are situations in which the response does not change much with factors variation. For this purpose, the error propagation parameter was used [18]. This parameter derives the final response from the factors affecting the process. Therefore, less value of this parameter shows that the changes in the final response are less in response to the changes in the factors; therefore, the conditions are more stable.

According to the software analysis, it can be said that there is a stable condition in a current density between 95.94 and 80 A/m^2 and a time of 5–15 min (Fig. S1). Also there is a stable condition in the system at pH 6 to 8.3 and between 5 and 15 min. According to these results, it can be said that the stable conditions are in the current density between 43.95 and 80 A/m^2 and the pH ranges from 6 to 8.8 and a time of 5–15 min (Fig. S2 and Table S1).

3.3. Results of response surface tests for energy consumption

The amount of energy consumed was calculated for each experiment. The voltage value of each test and its energy consumption are given in Table 3. Table 5 shows the analysis of the variance of this response.

As can be seen in Table 5, the current density, the time, the number of electrodes, the distance between the electrodes, the interaction of current density and time, the interaction of current density and the number of electrodes, the interaction of time and distance between the electrodes are the important factors.

3.3.1. Analysis of the graphs for the response of energy consumption

As shown in Fig. 11, energy consumption increases with increasing current density and time, which is due to increased electric current consumption. According to Fig. 11c, pH changes have no effect on the amount of energy consumed. In Fig. 11d, energy consumption increases with

Table 5
Analysis of variance for the response of energy consumption

Response	Energy consumption					
Source	Sum of squares	df	Mean square	*F*-value	*p*-value	Prob. > *F*
Model	1,974.771	15	131.6514	41.68352	<0.0001	Significant
A-c.d	740.61992	1	740.61992	234.4954	<0.0001	
B-time	328.80072	1	328.80072	104.105	<0.0001	
C-pH	5.2770477	1	5.2770477	1.670821	0.2042	
D-number	179.01918	1	179.01918	56.68113	<0.0001	
E-gap	37.502045	1	37.502045	11.87391	0.0014	
AB	145.23755	1	145.23755	45.98517	<0.0001	
AC	0.2982838	1	0.2982838	0.094443	0.7603	
AD	57.527361	1	57.527361	18.21434	0.0001	
AE	3.8914929	1	3.8914929	1.232126	0.2742	
BC	4.6066834	1	4.6066834	1.45857	0.2348	
BD	10.115783	1	10.115783	3.202864	0.0817	
BE	17.757309	1	17.757309	5.622327	0.0231	
CD	0.0752278	1	0.0752278	0.023819	0.8782	
CE	0.6449209	1	0.6449209	0.204195	0.6540	
DE	1.5734204	1	1.5734204	0.498177	0.4847	
Residual	116.85917	37	3.1583559			
Std. dev.	1.777176		R-squared	0.944		
Mean	6.801425		Adj. R-squared	0.921		
C.V. %	26.12947		Pred. R-squared	0.887		
PRESS	234.7336		Adeq. precision	20.510		

increasing number of electrodes. When the number of electrodes increases, the surface of the electrode's contact with the liquid also increases. Consequently, for constant current density, there is a need for more electrical current, and this increases the amount of energy consumed [28]. In section (E), it can be seen that increasing the distance between the electrodes also increases energy consumption. The distance between the electrodes is directly related to the voltage [19]. As a result, with increasing distance, there is a need for more voltage to reach a given current density, which increases energy consumption [35,36].

For all current densities, it is possible to find a time period in which the energy consumption is less than 3.5 kWh/m^3. For a current density of 80 A/m^2, the time from 5 to 7.5 min for a current density of 61.98 A/m^2, a time of 5–9 min and for current densities less than 43.95, an optimum amount of energy is obtained at a time interval of 5–15 (Table S2 and Fig. S3).

3.4. Optimum range for the process

To find the range for which the efficiency of sewage treatment is the highest, and the energy consumption is the lowest, the common area between these two optimizations is considered. For this current density, using two pairs of electrodes, the energy consumption is greater than 5 kWh/m^3. So when the number of electrodes is 1 pair, process is more optimal.

For a time between 9 and 15 min, a distance of 1 cm is more appropriate. On the other hand, for a current density of 49.50 A/m^2, the energy consumption is less than 5 kWh/m^3 in the period from 5 to 11 min. The current density of 80 A/m^2, in the range of 5–7.5 min, has low energy consumption, but this time does not eliminate the optimal efficiency conditions (Table 6). The highest current density, which the appropriate time is in both areas, is determined to be 63 A/m^2, and in order to have an energy consumption of less than 5 kWh/m^3, the range of time should be between 5 and 9 min. Therefore, it can be said that the optimum current density range is between 49.8 and 63 A/m^2 and the optimum time range is 9–11 min. These conditions show the optimal range of the process with a yield of more than 80% and an energy consumption of less than 5 kWh/m^3.

3.5. Sludge analysis

The sludge collected from the sewage treatment system was analyzed for its COD and BOD, the results of which are given in Table 6.

As we can see, 42% of the initial sewage's BOD is found in the sludge, which can be concluded that about 58% of the BOD of the sewage has been lost during the electro-oxidation process. For COD, the amount of residue in sludge is 31.5%, which indicates the removal of 68.5% of the sewage COD during the electro-oxidation process.

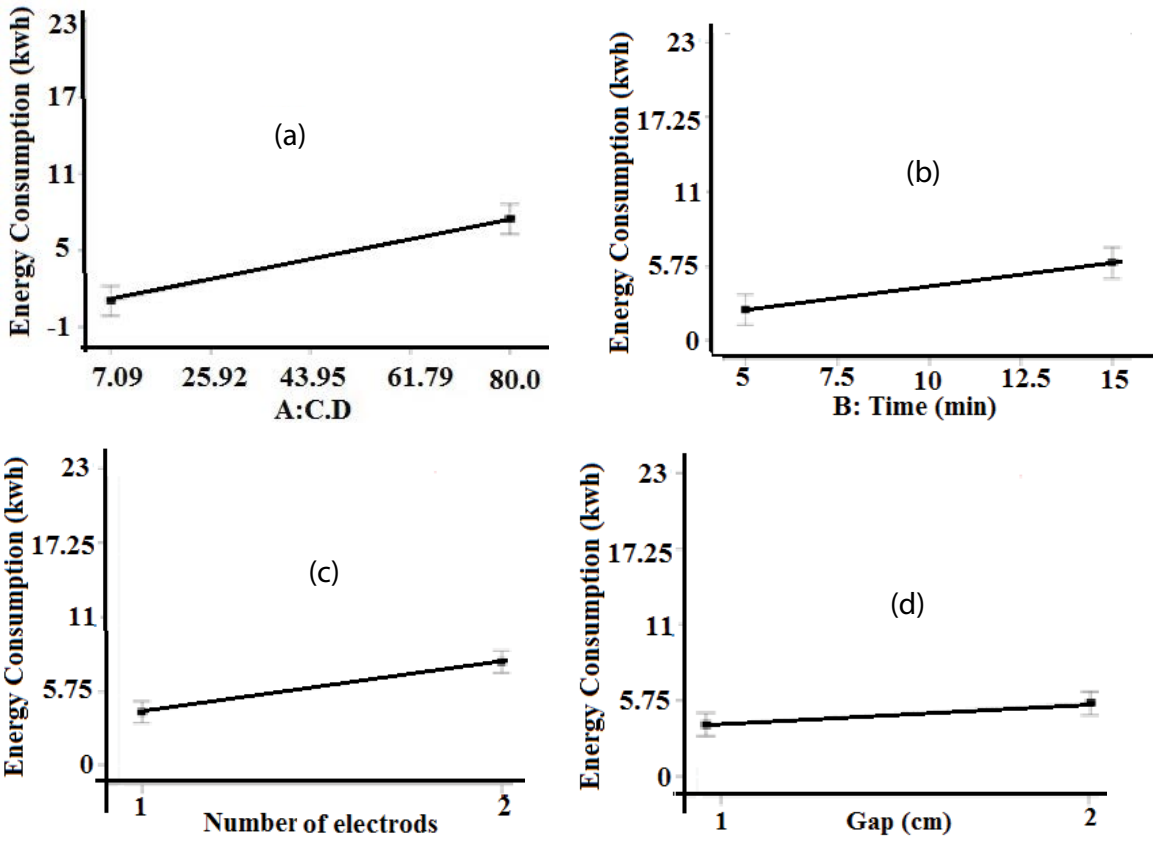

Fig. 11. Effect of (a) current density, (b) time, (c) electrode number, and (d) distance between electrodes on energy consumption.

3.6. Coliforms elimination rate

Analysis of the fecal coliforms, total coliforms and heavy metals content in sewage was done before and after treatment; the results are presented in Table 7. For the analysis of fecal coliforms and total coliforms, standard methods (9222 B for total coliforms and 9222 D for fecal coliforms) [37] were used and the amount of heavy metals was measured by atomic absorption analysis [38]. As can be seen, the removal rate is 99% for fecal coliforms and 98% for total coliforms content. Also, the amount of heavy metal removal is 99.999%.

4. Conclusion

Electrocoagulation (EC) technology is an electrochemical technique with many applications and high pollutants removal efficiency. It allows the wastewater to electrochemically oxidize or reduce the organic contaminants to non-hazardous inorganic substances. The technology has potential for treating municipal, industrial wastewater, and surface and groundwater, which are normally contaminated, by simple equipment, convenient operation, less operating time and not using any chemicals. In this study, electrocoagulation was approved as an effective method for the reduction of COD, BOD, TDS, TSS, heavy metals, fecal and total coliforms in sewage water. In this treatment, the efficiency of electrocoagulation point out to be dependent on the amount of ion release by electrode, which depended on the amount of current density, number and gap between the electrodes. Meaning that, as the higher current density and more electrodes at shorter distances been given, the higher amount of metal ion been generated, leading to higher treatment efficiency. The influence of various operational variables such as current density, number and inter electrode distance, electrocoagulation time and pH on treatment of sewage wastewater was investigated. The optimal value obtained was the current density of 49.8–80 A/m^2, the process time of 9–15 min, the pH 6.5–8, number of electrodes 2 pairs with inter electrode distance of 1 cm. It was observed that with increased current density, time, and number of electrodes and the distance between them energy consumption increases and pH changes does not affect the amount of expenditure. The optimum range in which the output efficiency is more than 80% and the energy consumption is less than 5 kWh/m^3 is the current density between 49.8 and 63 A/m^2, the time between 9 and 11 min, the pH of 6.5–8, the number of electrodes 1 pair and the distance between the electrodes 1 cm. In this optimal range, the energy consumption per one cubic meter of sewage is between 4.5 and 5 kWh.

Acknowledgment

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Table 7
Amount of COD and BOD of sludge collected and compared with the initial wastewater

	Amount in sewage	Amount in sewage after treatment	Amount in sludge	Percentage remaining in sludge
BOD (ppm)	233	47	97.86	42%
COD (ppm)	320	45	101	31.5%

Table 8
Depletion rate of fecal coliforms, total coliforms and heavy metals from sewage

	Before treatment	After treatment
Fecal coliforms, MPN (100 mL)$^{-1}$	7.3×10^3	8
Total coliforms, MPN (100 mL)$^{-1}$	4.6×10^7	50,643
Fe, ppm	0.012	0.000
Mn, ppm	0.020	0.000
Cu, ppm	0.018	0.000
Zn, ppm	0.040	0.000
Cd, ppm	0.004	0.000
Pb, ppm	0.04	0.000
Ni, ppm	1	0.002
TSS, ppm	312	10
TDS, ppm	298	25

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Supporting information

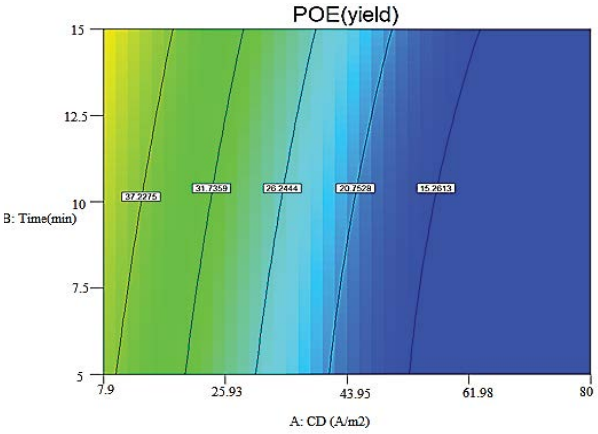

Fig. S1. Stability of the efficiency of sewage treatment compared with current density and time variations.

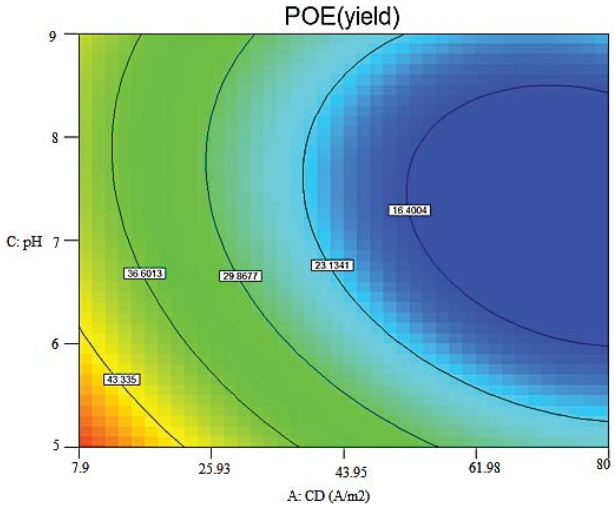

Fig. S2. Sustainability rate of the efficiency of sewage treatment from time and pH changes.

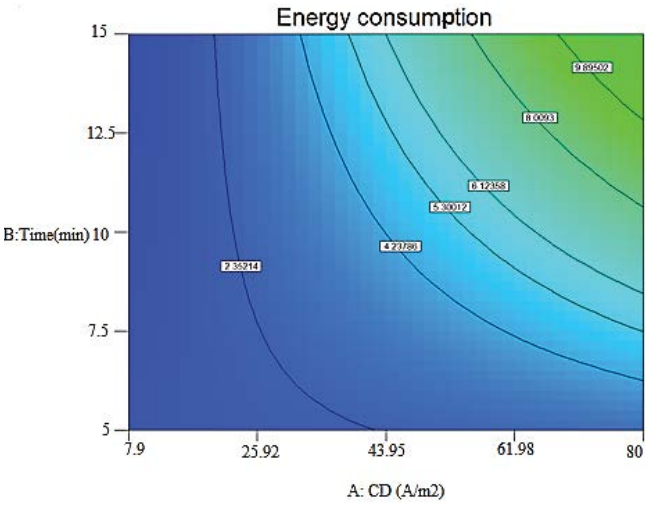

Fig. S3. Effect of current density and time on energy consumption.

Table S1
Optimal and stable range of the process

Factors	Stable condition	Optimal condition
Current density (A/m^2)	43.95–80	49.8–80
Time (min)	5–15	9–15
pH	6–8.3	6.5–8

Table S2
Energy consumption and electrode corrosion

Corrosion (kg)	Energy consumption (kWh/m^3)	Time (min)	Electrical current (A)	Current densities (A/m^2)
1.91×10^{-5}	4.3	9	0.38	49.8
2.41×10^{-5}	5	9	0.48	63
2.2×10^{-5}	5.2	11	0.38	49.8
3.01×10^{-5}	6.11	11	0.48	63