



TREATMENT OF MUNICIPAL WASTEWATER USING ELECTROCOAGULATION PROCESS

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Abstract

Treatment of Municipal wastewater by Electrocoagulation (EC) process using punched aluminium and zinc electrodes was studied in a batch EC cell reactor. Response surface methodology (RSM) based on Central Composite Design (CCD) was utilized to optimize the operating parameters for the removal of % Total Suspended Solids (TSS) and % Chemical Oxygen Demand (COD) from Municipal Sewage. Effect of operating parameters such as Electrode Distance (x_1), Electrolysis Time (x_2) and Voltage (x_3) has been optimized for the removal of TSS and COD. The prediction of removal percentage of TSS and COD in various Operational circumstances is done by using Quadratic model. The significance of each operating parameter was computed by Analysis of variance (ANOVA). To achieve the maximum removal of % TSS and % COD, the optimum conditions were Electrode distance(x_1)—3 cm, Electrolysis Time (x_2)—70.299 minute and Voltage (x_3)—6.5V. It was observed that the performance of electrocoagulation process increased up to 61.45% for COD removal, and 73.73% for TSS removal using punched electrode compared to plane electrodes.

Keywords: electrocoagulation (EC), municipal sewage, central composite design (CCD), response surface methodology (RSM), total suspended solids (TSS)

1. INTRODUCTION

Water pollution is of great concern, since water is the prime necessity of life and extremely essential for the survival of all living organisms. Moreover, water pollution is considered to be an environmental problem worldwide, and among the various water pollutants generated is due to the usage of water for

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municipal, commercial and industrial purposes. This result in huge quantity of wastewater usually disposed in nearby water bodies like lakes, rivers, and ocean etc. The municipal sewage can be generated from domestic places, hotels, Institutions, public parks, road cleaning and also some contribution from rainwater. The municipal sewage contains many hazard causing contaminants and causes many adverse effects on human health as well as on the environment. Due to adverse effect on human health and environment, the municipal sewage represents a major pollution source. The discharge of municipal sewage into the water bodies have a negative effect on water quality, on aquatic life due to reduction of Dissolved Oxygen (DO) and on human's health as well as affecting the environmental stability. To prevent the pollution and control the adverse effects on the environment as well as on human health the treatment of Municipal sewage is very necessary. Several methods have been used to treat Municipal sewage which includes Electro-Fenton [7], combined coagulation & flocculation [8], membrane bioreactor [9], submerged membrane reactor [16], and some biological methods such as sequential batch reactor and activated sludge [4], etc. Previous research shows that a lot of work has been done in the area of Municipal sewage treatment. Various developing and under-developing countries using septic tanks to collect and treat wastewater, where only 40% of Biological Oxygen Demand (BOD) is removed and the majority of pathogenic bacteria and organic pollutants remain in the wastewater [7, 13]. In the major cases, biological treatment methods are utilized for the treatment of municipal wastewater, which is inexpensive but not very effective. Therefore, it is very important to develop an efficient and economical method for removing contaminants from Municipal wastewater.

Electrocoagulation has the potential to eradicate the disadvantages of traditional treatment methods and also to provide economical and sustainable wastewater treatment. This method does not require additional chemical additives, and reduces the volume of sludge that produced [2]. The process itself derives that the treatment of wastewater is achieved by using electricity. The principle of EC process based on reactions of aqueous pollutants to electrically persuade the reduction and oxidation reactions by powerful electric fields. The EC process employs the direct current to remove unnecessary pollutants by precipitation technique and chemical reactions. The Aluminum, Zinc, cadmium, Copper, and Iron are the various types of electrodes utilized in this method [1]. The working of EC process based on the liquid chemistry means, specially the electric conductivity. The mechanism in the formation of ion is expressed as in below Eqs. (1.1) & (1.2).

Anode reactions:



Cathode reactions:



When Zinc and aluminum electrodes were used, the formation of, $\text{Zn}_{(aq)}^{2+}$ and $\text{Al}_{(aq)}^{3+}$ ions immediately enter into spontaneous reactions with the formation of polyhydroxides and hydroxides, respectively. These polyhydroxides and hydroxide are precipitated and settled at the bottom [1].

In previous research (17), they studied on the removal efficiency of BOD and TDS from sewage by EC using punched aluminum electrodes, and observed about 70-80% of BOD and TSS removal efficiency. Hence, in this research, the removal efficiency of COD and TDS from municipal sewage by EC using punched Zinc and aluminum electrodes is studied. Based on several researches on the EC (electrocoagulation) method were conducted by varying one factors and the further factors held constant [6, 15]. However, this method took maximum time; hence Response surface methodology (RSM) could be a possible choice to overcome this issue. The objective of this study is to identify the optimum

conditions of operating parameters (Independent variables) for the contaminants removal from municipal sewage using Central Composite Design (CCD).

The use of RSM helps to optimize the operating parameters preferred for EC process. It gives the predicted value of dependent variables (output or responses) depend on the controlled values of operating parameters using regression analysis. Several test combinations can be prepared within a small period, hence it helping the analyzer to understand the impact of the operating parameter on the research work [5,11]. In several technical platforms, it is familiar that the response (y) available with a set of operating parameters ($x_1, x_2, x_3, \dots, x_k$). The response is a function of operating parameter simultaneously with the error presence in the model, generally expressed as $y = f(x_1, x_2, x_3, \dots, x_k) + \epsilon$, where f stands for unknown surface response which is generally described by a second or first order polynomial, and ϵ stands for error in the model. In general, the second and first order models are expressed as in below Equations (1.5) and (1.6):

$$Y = \beta_0 + \sum_{j=1}^k \beta_j X_j + \epsilon \quad (1.5)$$

$$Y = \beta_0 + \sum_{j=1}^k \beta_j X_j + \sum_{j=1}^k \beta_{jj} X_j^2 + \sum_{j=1}^{k-1} \sum_{j=2}^k \beta_{jj} X_j X_i + \epsilon_i \quad (1.6)$$

Here, both X_i and X_j are referred as coded operating parameters and β_j, β_{ji} and β_{jj} ($i = 1, 2, \dots, k$; and $j = 1, 2, \dots, k$) are the regression coefficient. A first-order model is utilized to describe the plane surface, while the second-order model is utilized to describe curvature surface, or also called as a quadratic model. In various cases of RSM, the quadratic model is frequently sufficient for analysis. Moreover, the understanding of regression modeling methods, statistical fundamentals, and optimization techniques are necessary in fitting the response surface model [3]. The objective of optimization was to increase the TSS and COD removal by varying independent variables such as Electrode distance (x_1), Electrolysis time (x_2) and Voltage (x_3). Minitab-19 software was utilized to optimize and determine the combined effect of three preferred operating parameters.

The main aim of this study is to analyze treatment efficiency of Electrocoagulation process using punched aluminum and zinc electrodes by different independent variables (Electrode distance, Electrolysis time, Voltage) for the maximum removal of responses (COD and TSS) based on Central Composite Design and to study the optimum condition of the independent variables for maximum removal efficiency of responses by utilizing Response Surface Methodology based on Central Composite Design.

2. MATERIALS AND METHODOLOGY

2.1. Material

Borosilicate beaker of 2liter capacity, the punched aluminum and zinc plate electrodes of dimension 125mm×25mm×4mm with the 4 punches on its body with the diameter of 0.5mm with 1cm c/c, DC supply of 0-30V capacity, Magnetic stirrer with hot plate up to 700rpm capacity were important materials used for the electrocoagulation process.

2.2. Methods

2.2.1 Initial Characteristics of sewage

The Municipal wastewater for this research is collected from the Sewage Treatment Plant (STP) in Davangere. Several initial characteristics such as Total Dissolved solids (TDS), Turbidity, Total

Suspended Solids (TSS), Color, Chemical Oxygen Demand (COD), pH, and Biological Oxygen Demand (BOD) of the wastewater sample are analyzed as per IS 3025.

2.2.2 Design of Experiments

The Electrocoagulation process is conducted based on the design of experiments for operating parameters such as Electrode distance (x_1), Electrolysis time (x_2) and Voltage (x_3) was done by Central Composite Design using Minitab-19 software. Each operating parameters were coded at three levels between -1 and $+1$, where the parameters Electrode distance (x_1), Electrolysis time (x_2) and Voltage (x_3) were set in the range of 3-5cm, 30-60minute, and 5-8V, respectively, as specified in Table 1.

Table 1. The experimental range and levels of independent variables assessed

Independent variables	Unit	Levels		
		-1	0	+1
Electrode distance (x_1)	cm	3	4	5
Electrolysis time (x_2)	minute	30	45	60
Voltage (x_3)	V	5	6.5	8

2.2.3 Experimental Procedure

The electrocoagulation equipment setup as shown in Fig. 1 comprises of a cylindrical borosilicate beaker with two parallel plates of punched aluminum and zinc electrodes. The experimentation will be conducted in batch mode. The punched Aluminum and zinc electrodes of dimension (125mm*25mm*4mm) with the 4 punches on its body with the diameter of 0.5mm with 1cm c/c were used as cathode and anode with the surface area of the electrodes was 12.2cm² were immersed into the beaker containing Municipal sewage at the depth of 50mm. In this research, the Zinc was used as anode and aluminum is used as Cathode. The electrode distance between cathode and anode was varied between 3-5cm. The Voltage varying from 5-8V is supplied to the test sample of sewage through both anode and cathode connected to DC supply with the help of clips until the electrolysis time varied from 30-60 minutes. This test sample is stirred at 400 rpm at the temperature of 20–22°C using magnetic stirrer. After the completion of experimentation in various operational circumstances, the test sample is immediately taken from beaker and conducted the standard laboratory tests to determine the final concentration of TSS and COD and the removal percentage (actual results) is calculated. The analysis of actual results with the design of experiments and also the prediction of TSS and COD in various operational circumstances are done by using quadratic model. The Analysis of variance (Analysis of Variance) was used to analyze the Significance of the quadratic model and the comparison of actual and predicted results. The combined effect of operating parameters for the percentage of COD removal and percentage of TSS removal was studied using surface plots. The optimization of independent variables of the EC process for the maximum removal of dependent variables such as TSS and COD is done by regression equation of quadratic model in RSM.

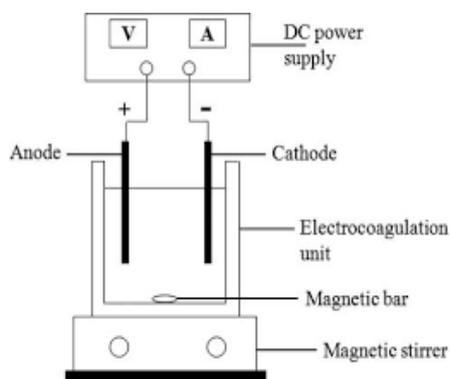


Fig. 1. Typical experimental setup of EC process

3. RESULTS AND DISCUSSIONS

3.1 Initial Characteristics of Municipal Sewage

Several initial characteristics of sewage sample are analyzed using standard methods and tabulated in Table 2.

Table 2. The Initial Characteristics of Municipal sewage wastewater

Sl.no.	Parameters	Laboratory Results
1	pH	7.1
2	Color	Blackish grey
3	Turbidity(NTU)	215
4	Total Dissolved Solids	890mg/L
5	Total Suspended Solids	120mg/L
6	Conductivity	35mS/cm
7	BOD	164mg/l
8	COD	262mg/L

During the analysis of initial characteristics, it is observed from the Table 2. that above parameters is exceeds the permissible limit as recommended under Schedule -VI of Environmental (protection) rules, 1986.

3.2 Central Composite Design (CCD)

In this study, the design of experimentation was performed by CCD using Minitab-19 software for three independent variables such as Electrode distance (x_1), Electrolysis time (x_2) and Voltage (x_3). There was 20 number of test combinations in the design to find out the optimum condition of independent variables for maximum TSS and COD removal efficiency. The set of various experimental runs and test conditions of Independent variables, the actual results of TSS and COD removal along with the predicted results are represented in Table 3 [13].

Table 3. The Experimental design and the actual and predicted results of COD and TSS removal (%) performed by the CCD

Run	Electrode Distance: x_1	Time: x_2	Voltage: x_3		Efficiency %		
	Centi meter	Minutes	Volts	COD		TSS	
				Actual	Predict	Actual	Predict
1	4.00000	30.0000	5.00000	56.64	55.2851	68.92	58.0304
2	4.00000	45.0000	6.50000	53.68	55.2851	65.96	58.0304
3	5.00000	45.0000	6.50000	58.08	58.4253	70.36	58.7631
4	4.00000	19.7731	6.50000	54.10	55.2851	66.38	58.0304
5	4.00000	45.0000	6.50000	61.45	63.0133	73.73	64.2925
6	3.00000	45.0000	3.97731	43.82	42.6025	56.16	48.2231
7	2.31821	45.0000	6.50000	57.00	57.3892	69.28	59.9930
8	4.00000	60.0000	8.00000	55.45	55.2851	68.73	58.0304
9	4.00000	60.0000	5.00000	56.85	57.0909	66.13	58.7879
10	5.68179	45.0000	6.50000	54.16	55.5749	66.44	55.9473
11	4.00000	60.0000	5.00000	25.70	28.9733	37.98	35.0809
12	4.00000	60.0000	8.00000	55.50	55.2851	67.30	58.0304
13	5.00000	30.0000	8.00000	55.90	55.8417	67.18	57.9612
14	5.00000	30.0000	5.00000	52.68	50.4753	64.96	51.9905
15	4.00000	45.0000	9.02269	56.65	55.2851	68.93	58.0304
16	3.00000	30.0000	5.00000	53.85	52.2290	66.13	55.5711
17	3.00000	45.0000	6.50000	53.48	53.7005	65.76	66.1054
18	3.00000	45.0000	6.50000	57.90	58.8290	70.18	70.6503
19	5.00000	19.7731	6.50000	39.80	38.3038	52.08	50.1250
20	4.00000	45.0000	6.50000	48.60	47.1308	60.88	59.5163

3.3 Analysis of actual (experimental) results with design of experiments

The % COD (y_1) and % TSS (y_2) removal are the functions of Independent variables such as electrode distance(x_1), Electrolysis time(x_2), and Voltage(x_3). The regression equations of quadratic model are obtained from Minitab 19 software as shown in below equations (3.1) and (3.2);

$$y_1 = -87.1 - 10.05 x_1 + 0.339 x_2 + 43.58 x_3 + 0.423 x_1^2 + 0.00749 x_2^2 - 2.708 x_3^2 + 0.0424 x_1 \times x_2 + 0.649 x_1 \times x_3 - 0.1639 x_2 \times x_3 \quad (3.1)$$

$$y_2 = -82.2 - 9.73 x_1 + 0.613 x_2 + 43.75 x_3 + 0.481 x_1^2 + 0.00539 x_2^2 - 2.682 x_3^2 + 0.0486 x_1 \times x_2 + 0.571 x_1 \times x_3 - 0.1692 x_2 \times x_3 \quad (3.2)$$

The experimental results of COD and TSS removal were analyzed by quadratic model and the values of R^2 (coefficient of determination), $R^2 (adj)$ values from model summary are represented in below Table 4. From Table 4. it shows that the highest values of R^2 and $R^2 (adj)$ about 98.76% and 98.61% for COD and 97.99% and 97.75% for TSS respectively, which shows that the excellence of the relationship between actual and predicted results.

Table 4. Summary of Quadratic Model

Response	S	R^2	$R^2 (adj)$
COD	4.87771	98.76%	98.61%
TSS	2.13221	97.99%	97.75%

3.4 Significance of the model tested for Responses removal

The Analysis of variance (ANOVA) was used to analyze the adequacy and Significance of the model. The ANOVA results of the quadratic model for COD and TSS removal are shown in Table 5a & b. The

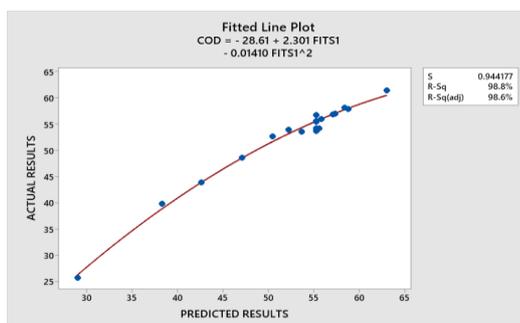
F-test of model provides a small *P*-Value (less than 0.05), which shows that the all models are statistically significant can be used for the predict the responses for the EC process. In Table 4, it shows that for COD removal, the linear co-efficient of the Electrode distance (x_1), Electrolysis time (x_2) and Voltage (x_3), the quadratic coefficient of Electrode distance (x_1^2), Electrolysis time (x_2^2) and Voltage (x_3^2) are statistically significant with the *P*-values <0.05. For TSS removal, it was observed that the linear co-efficient of the Electrode distance (x_1), Electrolysis time (x_2) and Voltage (x_3), the quadratic coefficient of Electrode distance (x_1^2), Electrolysis time (x_2^2) and Voltage (x_3^2) and the interaction coefficient of Electrode distance (x_1) with Electrolysis time (x_2), Electrode distance (x_1) with Voltage (x_3) and Electrolysis time (x_2) Voltage (x_3) are statistically significant.

Table 5(a). ANOVA (analysis of variance) of the quadratic model % COD removal

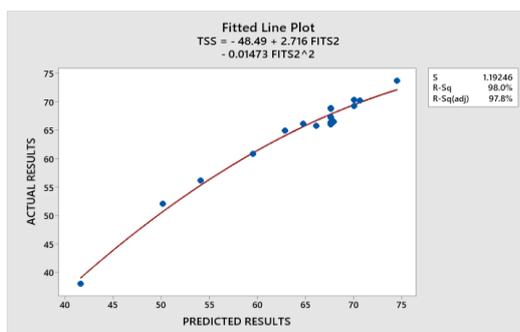
Source	df	Sum of Squares	Mean Square	F-Value	P-Value	Prob > F
Model	9	1185.19	131.688	35.09	0.000	Significant
Linear	3	444.29	148.096	39.47	0.000	Significant
X ₁	1	3.97	3.973	1.06	0.328	
X ₂	1	42.34	42.339	11.28	0.007	Significant
X ₃	1	397.98	397.975	106.05	0.000	Significant
Square	3	621.23	207.075	55.18	0.000	Significant
X ₁ * X ₁	1	2.58	2.581	0.69	0.426	Significant
X ₂ * X ₂	1	40.94	40.936	10.91	0.008	Significant
X ₃ * X ₃	1	534.98	534.976	142.56	0.000	Significant
2 – Way Interaction	3	119.68	39.893	10.63	0.002	
X ₁ * X ₂	1	3.24	3.239	0.86	0.375	
X ₁ * X ₃	1	7.59	7.586	2.02	0.186	
X ₂ * X ₃	1	108.86	108.855	29.01	0.000	Significant
Error	10	37.53	3.753			
Lack of Fit	5	29.79	5.958	3.85	0.083	
Pure Error	5	7.74	1.547			
Total		1222.72				

Table 5(b). ANOVA (analysis of variance) of the quadratic model for % TSS removal

Source	df	Sum of Squares	Mean Square	F-Value	P-Value	Prob > F
Model	9	1158.29	128.698	28.86	0.000	Significant
Linear	3	449.35	149.783	33.58	0.000	Significant
X ₁	1	5.20	5.199	1.17	0.306	
X ₂	1	57.54	57.537	12.90	0.005	Significant
X ₃	1	386.61	386.613	86.68	0.000	Significant
Square	3	585.02	195.006	43.72	0.000	Significant
X ₁ * X ₁	1	3.33	3.331	0.75	0.408	
X ₂ * X ₂	1	21.19	21.192	4.75	0.054	Significant
X ₃ * X ₃	1	524.91	524.912	117.69	0.000	Significant
2 – Way Interaction	3	123.92	41.306	9.26	0.003	Significant
X ₁ * X ₂	1	2.15	2.153	0.48	0.503	
X ₁ * X ₃	1	5.87	5.865	1.32	0.278	
X ₂ * X ₃	1	115.90	115.900	25.99	0.000	Significant
Error	10	44.60	4.460			
Lack of Fit	5	35.61	7.122	3.96	0.079	
Pure Error	5	8.99	1.798			
Total	19	1202.89				



(a)



(b)

Fig. 2. Comparison plot of the predicted results with actual results of (a) COD removal and (b) TSS removal

3.5 Comparison of Predicted results with Actual Results

The Comparison between predicted and actual values are represented in Table 3. and Fig. 2.(a) & (b), it shows that the actual results are matched with predicted values that all actual (points) values are very nearer to the predicted value (diagonal line). Analysis by ANOVA shows that overall 3-quadratic models are statistically significant by getting P -value < 0.05 and could be utilized to predict the TSS and COD removal. The excellence of predicted values was studied by the value of R^2 are 98.8% and 98.0% for removal of COD and TSS respectively.

3.5 Combined Effect of Independent Variables on Percentage COD and Percentage TSS Removal

The Combined effect of independent variables such as Electrode distance (x_1), Electrolysis time (x_2), and Voltage (x_3) on the TSS and COD removal efficiency are debated as below with the help of surface plots using Minitab-19 software

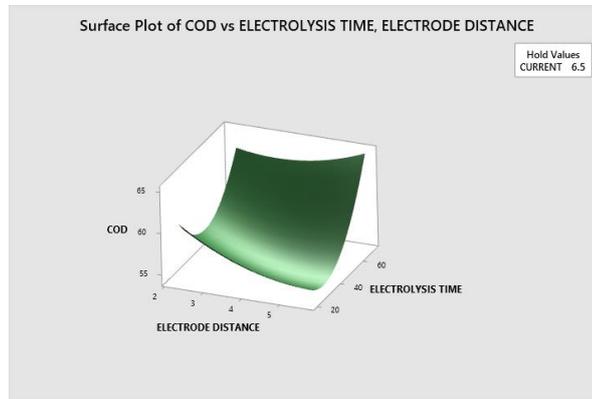
3.5.1 Combined Effect of Electrode distance (x_1) and Voltage (x_3)

The Combined effect of Electrode distance (x_1) and Voltage (x_3) on TSS and COD removal was analyzed by varying x_1 from 3-5cm and x_3 from 5-8V and the output are presented in Table 3 and also surface plot is plotted as shown in Fig. 3a-c for COD and 4a-c for TSS. As seen in these plots, the COD and TSS removal efficiency is increased with increasing in Voltage up to the optimum level and started to gradually decrease beyond the optimum level of Voltage. The increment in Voltage resulted in huge amount of Zn^{2+} ions by anodic metal dissolution, and more H_2 bubbles was formed at cathode, which

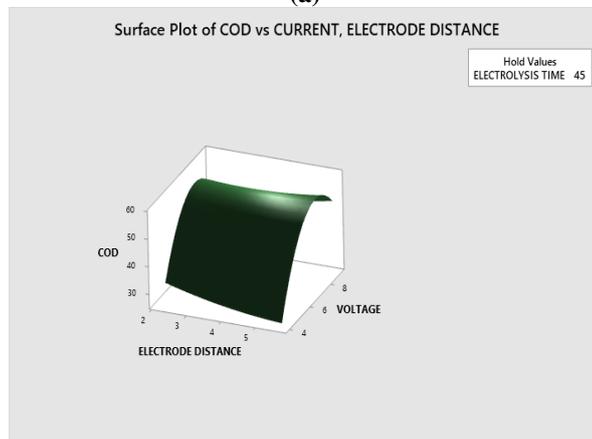
are profitable for the separation or flotation process (Ozyonar 2015). And, the COD and TSS removal efficiency is decreased with increasing in electrode distance at the Voltage range of 5-8V. This is due to the fact that the rising in ohmic voltage drop as increasing electrode distance(Khandegar and Saroha 2013).In addition, Faraday's Law states that the quantity of oxidized metal decreases with increasing distance between the electrodes. [13].

3.5.2 Combined Effect of Electrolysis time (x_2) and Voltage (x_3)

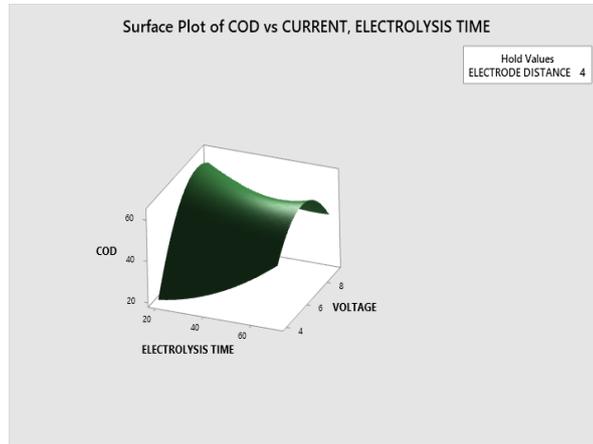
The Interacted effect of Electrolysis time (x_2) and Voltage (x_3) on TSS and COD removal was analyzed by varying x_2 from 30-60min and x_3 from 5-8V and output are presented in Table 3 and also surface plots are plotted as shown in Fig. 3(a) (b) & (c) for COD and Fig. 4(a) (b) & (c) for TSS. As seen in these plots, the COD and TSS removal efficiency is increased with increase in electrolysis time. This because, when the electrolysis time is minimum, there is no sufficient period for both releasing of Zn^{2+} ion by metal dissolution in anode and H_2 bubble release in cathode in the solution. By increasing the Electrolysis time, the increase in the metal dissolution and H_2 bubble production in both anode and cathode respectively resulting in increasing in coagulation process in solution, Hence the removal efficiency also increased.



(a)

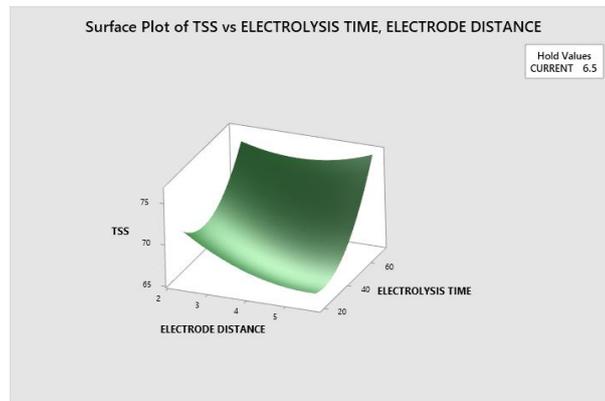


(b)

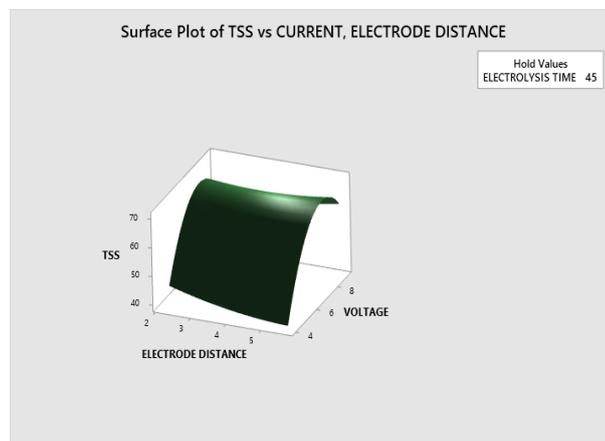


(c)

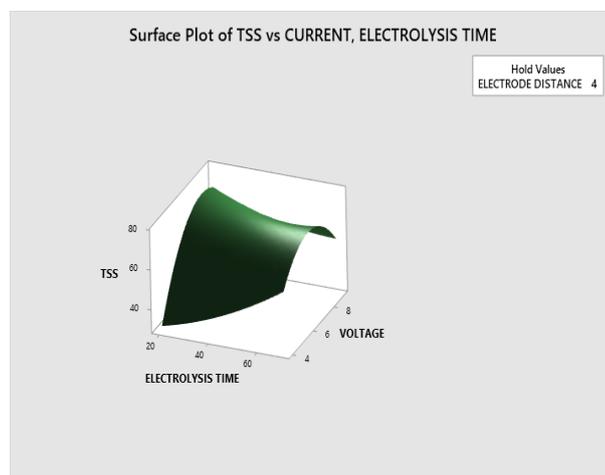
Fig.3. Combined effect of Electrode distance (x_1) Electrolysis time (x_2), and Voltage (x_3) on COD removal



(a)



(b)



(c)

Fig. 4. Combined effect of Electrode distance (x_1) Electrolysis time (x_2), and Voltage (x_3) on TSS removal

3.6 The Optimization

The major aim of this research is to find out the optimal Independent variables for the maximum TSS and COD removal from Municipal sewage using EC process. The regression equation of RSM based on CCD was used to optimize the results. During optimizing, all the Independent variables such as Electrode distance (x_1), Electrolysis time (x_2) and Voltage (x_3) are preferred as within the range while the responses such as TSS and COD removal were increased: Electrode distance (x_1)—4cm, Electrolysis time (x_2) —70.22 min and Voltage (x_3) —6.5V with predicted result of COD removal to be 63.013%, TSS removal of 72.540%. An actual experimental result of 61.45% for COD removal, 73.73% for TSS removal was attained, which is very nearer to the predicted values. From the actual and predicted result, it can be expressed that there was excellent relationship between actual and predicted results which shows that CCD can be successfully utilized to optimize the EC process variables.

4. CONCLUSIONS

This research examined the removal efficiency of TSS and COD from municipal sewage by EC process using punched Zinc and aluminum electrodes. During the analysis of initial characteristics, it was observed that all the parameters were exceeded the standard discharge limits. In that, the Initial concentration of COD and TSS were observed about 262mg/L and 120mg/L respectively, this shows an objectionable for discharge. Hence the electrocoagulation process was conducted with batch mode using three independent variables such as Electrode distance (x_1), Electrolysis time (x_2) and Voltage (x_3) varied between 3-5cm, 30-60minute, and 5-8V, respectively for the removal of TSS and COD. The optimum condition of each independent variables for the maximum removal percentage of COD and TSS was done by design of experiments using Central Composite Design based Response Surface Methodology. The prediction of removal percentage of TSS and COD in various operational circumstances and the analysis of actual results with the design of experiments is done by using quadratic model. The quadratic model shows the excellence of the relationship between predicted and actual results by getting maximum R^2 value for COD and TSS of 98.76% and 97.99% respectively. The Analysis of variance shows that the quadratic model was significant for the electrocoagulation process by getting P-value < 0.05. Actual results indicated that, the maximum COD removal, TSS removal were 61.45%, and 73.73%,

respectively, secured at the optimum conditions of Electrode distance (x_1)—4cm, Electrolysis time (x_2)—70.22 min and Voltage (x_3)—6.5V. Hence, the electrocoagulation process using punched Zinc and aluminum electrodes could be utilized successfully for the removal of contaminants from municipal sewage.

There is always a scope for improving research in this world. Few works can be carried forward in this area are by changing the electrode combinations of two different types of electrodes in the EC process, COD and other contaminants removal efficiency can be studied. By varying the different ranges of electrode distance, electrolysis time, Voltage, the efficiency of EC treatment can be studied. Other than these Independent variables, the pH, agitation speed, can be used as independent variables for studying the treatment efficiency of EC process.

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