International Journal of Aquatic Sciences
ISSN: 2008-8019
Vol 12, Issue 03, 2021

Removal of COD and Turbidity from Urban Wastewater by Electrocoagulation Using Aluminium Electrodes: Process Optimization by Response Surface Methodology

S. Raj Kamal[1], M. Kalyan Kumar[2]

[1,2] *Department of Chemical Engineering, JNTUA College of Engineering Ananthapuramu, Ananthapuramu, India, 515002*

Email: [1]kamalstar11@gmail.com, [2]kalyankumarmeda.chemengg@jntua.ac.in

Abstract - Due to extensive industrialization, increase in population and rapid urbanization, management of wastewater has become essential to prevent damage to the receiving water bodies. Urban wastewater includes wastewater from domestic, commercial and industrial areas. There are various conventional treatment methods in use. In this study electrocoagulation technique is adopted to reduce COD and turbidity. Urban wastewater is rich in organic matter and nutrients. It is subjected to the electrocoagulation treatment using aluminium electrodes and the reduction in the COD and turbidity was examined. Conductivity, pH, runtime, voltage and agitation speed are the factors that affect the performance of electrocoagulation. The experimental factors are optimized using response surface methodology (RSM) and a second order polynomial regression model is developed. At pH 6.95, conductivity 3.07 mS/cm, runtime of 48 min, voltage 6 V and agitation speed 215 rpm, the COD and turbidity removal efficiency are 88.42% and 85.16% respectively. The anodic dissolution at optimal conditions is 0.00432 g/cm^2. The energy consumption for the optimal reduction of pollutants is 1.68 KWh/m^3 and the quantity of sludge produced is 0.102 kg/m^3. The treated water is fit to be discharged into natural water bodies.

Keywords: Electrocoagulation, Urban wastewater, Aluminium electrodes, COD, Turbidity, RSM.

I. INTRODUCTION:

Water plays an irreplaceable role in every living creature's life. Life without water is unimaginable. Due to huge rise in human population and rapid urbanization the available sources are not sufficient to meet the demand. The earth's fresh water resources are getting contaminated due to disposal of both industrial and domestic wastewater into them directly without any prior treatments. Domestic wastewater mostly contains organic pollutants [1]. This activity completely degrades the quality of freshwater and also damages the aquatic life. To terminate this damage, wastewater should be treated in wastewater treatment units before disposing into the rivers and oceans so that the pollutants concentrations will get reduced to permissible limits and it doesn't affect the natural ambience and quality of fresh water resources. Characteristics of wastewater are divided into three categories they are physical, chemical and biological characteristics. The physical characteristics include color, odour, turbidity and temperature. Chemical characteristics include pH, conductivity, TOC, nitrogen, phosphorous, chlorides, heavy metals, COD, DO and BOD. Biological characteristics include the population of microorganisms such as bacteria, viruses, algae and protozoa. The aim of the wastewater treatment process or techniques is to degrade or dilute the concentration of impurities before discharging into the natural water bodies or flowing streams. As flowing streams exhibit self purification ability, the diluted ortreated wastewater will not affect the natural quality of the stream. There are many conventional wastewater treatment techniques available in market.

A conventional sewage treatment plant (STP) is the combined unit of all the physical, chemical and biological treatment processes. Physical treatment process includes screening, grit chambers, comminutors, floatation units, skimming, aeration. Chemical treatment process includes coagulation, flocculation, adsorption, ion exchange and chlorination. Biological treatment processes include aerobic and anaerobic methods. Activated sludge process, trickling filters, oxidation ponds, aeration tanks comes under aerobic treatment method. Anaerobic digesition is an anaerobic treatment process. UV treatment, ultra filtration, nano filtration, thermal evaporation, electrocoagulation, are the advanced methods of wastewater treatment. In this study, an advanced wastewater treatment technique is adopted to treat the urban sewage. Electrocoagulation is a reliable and alternative treatment technique to chemical coagulation. Generally, chemical coagulation and electrocoagulation differ in the mode of operation.

A. Electrocoagulation

The process in which metal ions are generated electrochemically, which further act as destabilizing agents in aqueous medium to remove pollutants which are in colloidal state is called “electrocoagulation”. The fundamental principle of electrocoagulation is based on ‘electrolysis’, where two electrodes are placed in an electrolyte and a potential difference is applied between two electrodes which induces electric field between two electrodes. This electric field generates an electromotive force which promotes movement or transfer of ions between electrodes. Oxidation occurs at anode and reduction occurs at cathode. Disassociation of compounds occur at anode so that these compounds are converted to ions and contribute electrons to the anode and cations migrate towards cathode and gain electrons from cathode so that it forms a closed circuit where free flow of charge takes place. Due to electrolysis chemical reaction occurs and coagulants are generated in-situ by metallic dissolution of electrodes. Different species of metal hydroxides are formed which have stronger affinity towards the counter ions and dispersed particles. These hydroxides adsorb and neutralize the charged particles which are in suspension ([2], [3]). Any metal can be used as electrodes in electrolysis. But the most commonly used electrode materials in electrocoagulation are aluminum, iron and steel as they are economical, easily available and show good efficiency in pollutant removal operation ([4], [5], [6], [7], [8]).

1) Mechanism: Aluminum is used as electrode material. External DC source is connected to these electrodes which generates potential difference between the electrodes. The positive terminal acts as anode and negative terminal as cathode. At anode, the aluminum gets dissolved into aqueous medium as metal cations. As the anode is getting dissolved it is also called as ‘sacrificial anode’. Water molecules get dissociated into hydrogen ion and oxgen gas. Oxygen bubbles are liberated at anode [9].

Reactions at anode

$$Al\,(s) \rightarrow Al^{3+}\,(aq) + 3e^- \qquad (1)$$

$$2H_20 \rightarrow 4H^+\,(aq) + O_2\,(g) + 4\,e^- \qquad (2)$$

At cathode, reduction takes place where the metal cations produced at anode move towards cathode, forms stable solid compound by gaining electrons and gets deposited on cathode. Other reactions occur at cathode where water molecules get reduced and results in production of hydrogen gas and hydroxyl ions at cathode.Hydrogen ions also gets reduced at cathode and results in liberation of hydrogen gas at cathode (Fig. 1). This depends on the pH of the solution [9].

Reactions at cathode

$$Al^{3+}(aq) + 3e^- \rightarrow Al\,(s) \qquad (3)$$

$$3H_20 + 3e^- \rightarrow H_2\,(g) + 3OH^-\,(aq) \qquad (4)$$

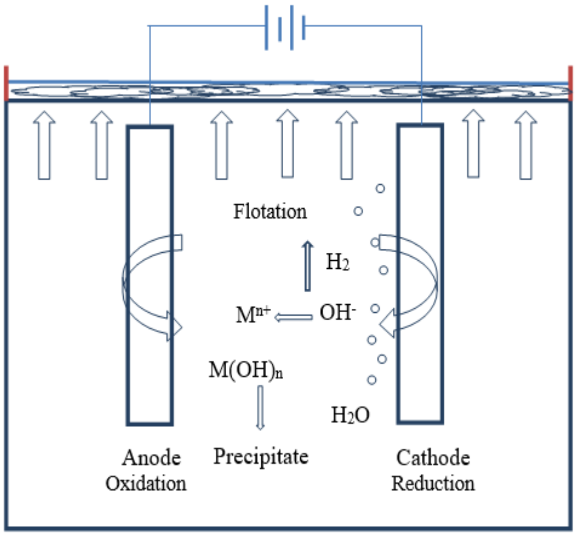

Fig. 1. Mechanism of Electrocoagulation

2. Materials and Methods

A. Urban Wastewater

The urban wastewater sampling location is located in the premises of Ananthapuramu Municipal Corporation. The GPS coordinates of the sampling site are 14.684303° N, 77.587821° E. Wastewater from the residential and commercial areas of the town gets discharged into this main sewer through branched sewers of the town. Grab sampling is done, where the wastewater is collected at a point in the sewer in a 5 L plastic can. Then the can is properly sealed so that there is no exposure of the sample to the atmosphere. The container is labeled with the collection time, date and initial pH. Then sample is transported to the laboratory and stored at 4℃ in the laboratory refrigerator so that the characteristics of collected sample don't change with respect to time as all the reactions in the sample are at inactive state at this temperature.

B. Analytical Procedures

Analysis of the wastewater sample collected is done before and after the treatment process. Sample pH, conductivity, solid analysis, COD and turbidity are determined. Sample pH and conductivity are some of the guiding parameters of the electrocoagulation process. COD and turbidity define the magnitude or intensity of pollutants in wastewater. In this study COD and turbidity are the pollutants of concern so they are determined before and after the treatment process. Characterization is done by following the standard procedures suggested by IS 3025: 2006.

Sample's pH is measured by using Systronics 361 digital pH meter. The conductivity of the sample is measured by using Systronics 306 digital conductivity meter. COD of the sample is determined by closed reflux method. Turbidity of the sample is determined using Systronics digital nephelo- turbidity meter 132.

Calculation of COD ($\frac{mg}{L}$) and turbidity (NTU) removal efficiencies after the electrocoagulation of the wastewater sample is done using the data of initial and final values of COD and turbidity. It is calculated using the following formula:

$$\text{Removal efficiency (\%)} = \frac{C_i - C_f}{C_i} \text{ x } 100 \qquad (6)$$

C_i - Initial concentration of COD or turbidity

C_f - Final concentration of COD or turbidity

C. Experimental Setup

The setup consists of an electrolytic cell, a glass beaker of 2 liter volume. This beaker is placed on a magnetic stirrer. In this study aluminum plates of size 12 cm x 4.5 cm x 0.1 cm are used. The effective electrode surface are is 45 cm^2. Distance between the electrodes is 10 mm, which is maintained throughout the experiment. These electrodes are connected to an external DC source by using alligator clips to perform electrolysis. A battery eliminator is used as an external DC source (3 – 12 V) which has different voltage levels in it. A magnetic strring bar is introduced into the cell for proper agitation of the solution during electrolysis. Experimental runs are performed at the room temperature.

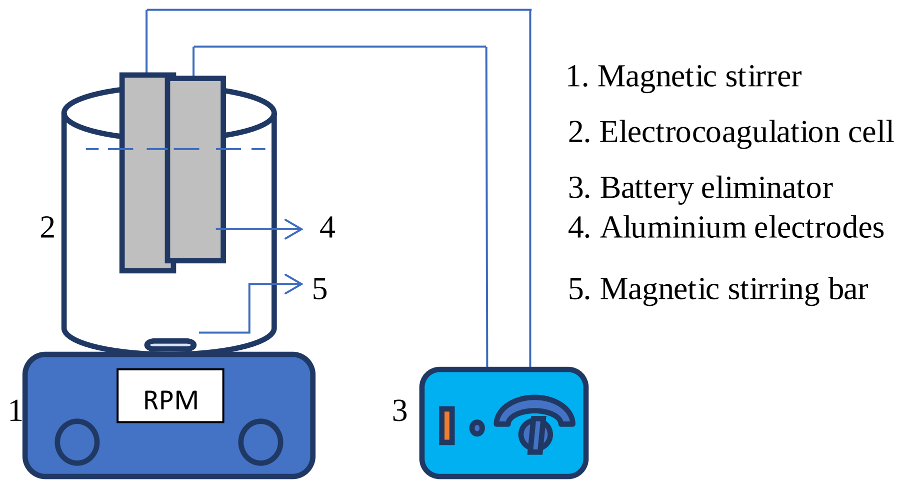

Fig. 2. Schematic diagram of experimental setup

C. Preliminary Studies

Preliminary studies are conducted to identify the operational ranges of the operative parameters which provide input values for the central composite design (CCD) to design the experiment. The interactive effects of operational parameters on pollutant removal efficiency are studied and process optimization is done using response surface methodology. One factor experiments are conducted and the corresponding pollutant removal efficiencies are noted.

D. Design of Experiments

As there are five operational parameters, it is difficult and time consuming to conduct the single factor varying experiments. One factor at a time experiments leads to large number of experimental runs, also interactions between the factors and their combined effect on the responses cannot be determined. Design of experiments (DoE) is a statistical tool which provides solution to all the drawbacks that arise in the conventional one factor at a time approach. Design of experiments is a well planned and structured statistical approach for the smooth conduct of the experiments. It is very useful in development and improvement of the process. It reduces the number of the experimental runs. Interactions between the factors can be evaluated perfectly. Finally we get the optimum settings where the response can be maximized or minimized. High precision can be obtained by replications and clear conlusions are made based on the hypothesis statements.

'Design-Expert 8.0.7.1' developed by 'Stat Ease' is used to structure the experiments and evaluate the optimum settings based on the results obtained through RSM. Here the inputs or the factors are pH, conductivity, voltage, run time, agitation speed and each factor has five levels. 'COD removal efficiency' and 'turbidity removal efficiency' are the responses in the experiment. Half fraction 'central composite design' is adopted to get the planned experimental design. This design suggested 30 experimental runs with different combinations of the factors at different levels. Each factor is varied over five levels, of which two axial points (+α, -α), two factorial points (+1, -1) and one center point. Four replicate runs are established. Experiments are conducted according to the data or input settings provided by the CCD and the responses are collected after each experimental run. Based on the data of response values, results are evaluated and regression analysis is performed. The analysis of varience (ANOVA) is executed and the important factors which are showing greater impact on responses are determined. Finally, using response surface methodology, a mathematical model is built [10]. Regression analysis is carried to fit the data to a non-linear second order polynomial equation so that a relation is plotted between responses and input factors.

$$y = f(x_1, x_2, x_3, x_4, x_5, \ldots\ldots, x_n) + \varepsilon \qquad (7)$$

In the above equation, f defines the response function of an unknown correlation, ε denotes the residual error i.e the differentiation between the observed values and predicted values. Analysis of variance, coefficient of determination (R^2) and response surface plots are used to analyse the results. The experimental data is fitted to a second-order polynomial equation and the order of the model was identified by analyzing the experimenal data by the software. All the terms such as linear, interaction terms and square terms are considered and they were expressed as a quadratic response model.

$$Y = \beta_0 + \sum \beta_i x_i + \sum \beta_{ii} x_{ii}^2 + \sum \beta_{ij} x_i x_j + \varepsilon \qquad (8)$$

where, Y is the reponse function, β_0 is the coefficient of the constant, β_i is the coefficient of the individual or linear effect, β_{ii} is the quadratic coefficient and β_{ij} is the coefficient of interaction factors.

E. Electrode Consumption

During the electrocoagulation process, as the current is allowed to pass through the electrodes, the electrode material gets dissolved in the solution. This dissolution rate depends on the applied current density. The current density is the ratio of applied current to the effective electrode surface area. The relation between the amount of anode material that gets dissolved into solution and the applied current desnity is given by Faraday's law.

$$W = \frac{itM}{ZF} \qquad (9)$$

'W' is anode dissolution in g/cm^2, 'i' is current density in A/cm^2, 't' is time in seconds, 'M' molar mass of anode material (for Al, M = 26.98), 'Z' is number of electrons during oxidation (3), 'F' is Faraday's constant (96,487).

F. Energy Consumption

Electrical energy consumption by electrocoagulation process is helpful in estimation of operational cost of EC process. The following formula is used to calculate the electrical energy consumed per cubic meter of treated wastewater.

$$E = \frac{itu}{1000V} \qquad (10)$$

Where, u = applied voltage in volts.
i = applied current in amperes
t = runtime in hours
V = volume of wastewater treated in m^3

G. Sludge Volume Calculation

During the electrocoagulation process, hydrogen bubbles are produced at the anode. These bubbles cause the floatation of pollutants to the surface of the treated water. The pollutants accumulated at the surface of the treated water are separated and collected. This sludge is oven dried for 10 hours at 110 °C so that the moisture present in it is evaporated. This oven dried sludge is weighed [11].

$$\text{Volume of sludge} = (W_1 - W_2)/\text{volume of wastewater} \qquad (11)$$

W_1 - Initial weight of the sludge,

W_2 - Final weight of sludge after oven drying

H. Cost Analysis

Electrodes and electricity are the major factors which contribute to the operational cost of the Electrocoagulation treatment of wastewater. Electrode consumption and the energy consumption are obtained from the experimental data. Unit price of the electricity and the unit price of the electrode material are obtained from the local market.

$$\text{Operating cost}_{(EC)} = i\,C_{electrode} + j\,C_{energy} \quad ([4], [12]) \qquad (12)$$

where, $C_{electrode}$ – Electrode consumption per m^3 of urban wastewater treated

C_{energy} - Energy consumption per m^3 of urban wastewater treated

i – Unit price of electrode material per kg

j – Unit price of electricity

The major cost-effective factors of chemical coagulation treatment of wastewater are coagulant dosage and electricity consumption for the mixing of the coagulant. The energy consumption for the mixing of the coagulant using a impeller in conventional chemical coagulation is calculated by using the formula mentioned below.

$$P = N_p \rho N^3 D^5 \quad (13)$$

Where, N_p is dimensionless power number (depends on the Reynolds number of the fluid), ρ is density of fluid in kg/m^3, N is agitation or mixing speed in rpm, D is impeller diameter in m.

$$\text{Operating cost}_{(CC)} = i\, C_{coagulant} + j\, C_{energy} \quad [13] \quad (14)$$

$C_{coagulant}$ – Consumption quantity of coagulant
C_{energy} – Consumption of energy for mixing of coagulant.
i – Unit price of coagulant.
j – Unit price of electricity.

3. Results and Discussion

A. Characterization of Urban Wastewater

The physicochemical characteristics of the urban wastewater sample collected are determined. Following table shows the characteristics of the urban wastewater sample collected (Table I). The COD and turbidity values of the wastewater sample are beyond the tolerance Limits for discharge of effluents into inland surface water by IS:2296-1982.

TABLE I
PHYSICOCHEMICAL CHARACTERISTICS OF URBAN WASTEWATER

Parameter	Value
pH	7.28
Conductivity (mS/cm)	2.37
Dissolved Solids (ppm)	596
Suspended Solids (ppm)	600
COD (mg/L)	641
Turbidity (NTU)	87

B. Preliminary Studies

1) Effect of initial pH on COD and turbidity removal efficiency: Sample pH influences the COD and turbidity removal efficiency. The pH of the sample is varied using 0.1M H_2SO_4 and 0.1M NaOH, maintaining the other factors constant. COD removal has increased till pH 6.5, where maximum COD removal of 87.31% observed. Further increase in pH, the COD removal has decreased. Turbidity removal has increased with increase in pH. Maximum turbidity removal of 86.68% is observed at pH 8.0 (Fig. 3). The formation of different aluminum hydroxyl species is highly dependent on pH of the solution. The rate of interaction mechanism between pollutants and hydroxyl products is dominated by the pH of the influent. The flocculation at lower pH ranges i.e., pH 4 - 7 is called as 'precipitation' [4]. Here the cation monomeric metal species such as Al^{3+}, $Al(OH)_2^+$ are predominant. At higher pH ranges i.e. pH 7 - 10, the flocculation mechanism is called 'adsorption' where both monomeric and polymeric species were formed such as $Al(OH)_2^+$, $Al(OH)_2^{2+}$, $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_{13}(OH)_{34}^{5+}$. These species are finally transformed into insoluble amorphous $Al(OH)_3(s)$. This amorphous $Al(OH)_3(s)$ is also called as "sweep floc" which has larger surface area [14]. If the pH of the wastewater is below 7, it tends to increase after EC. If the initial pH is above 8 the pH gets reduced after EC. This confirms that the EC process exhibits pH buffering nature [11].

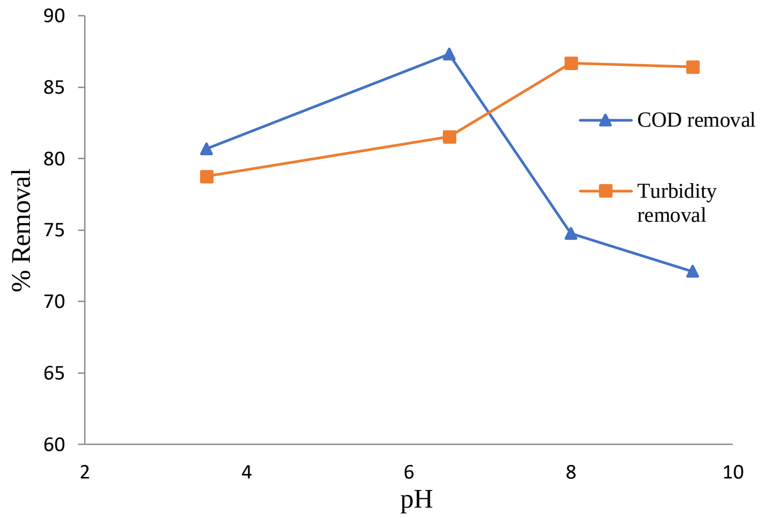

Fig. 3. Effect of initial pH on COD and turbidity removal, conductivity 2.24 mS/cm, voltage 6 V, runtime 20 min and agitation speed 200 rpm.

2) Effect of conductivity on COD and turbidity removal efficiency: Conductivity is an important parameter which affects COD and turbidity removal. Conductivity of the sample is varied using NaCl and deionized water. Conductivity is increased by adding NaCl and deionized water is used to reduce conductivity. Other parameters are kept constant to study the variation of COD and turbidity removal with conductivity. Both COD and turbidity removal shown increasing trend till a conductivity of 3.0 mS/cm, where corresponding COD and turbidity removal efficiencies are 85.19% and 84.15% respectively (Fig. 4). Rise in conductivity reduces the power consumption. Conductivity defines the ionic strength of the solution [14]. Higher conductivity reduces the ohmic resistance and increases the current density at the constant cell voltage and vice-versa.

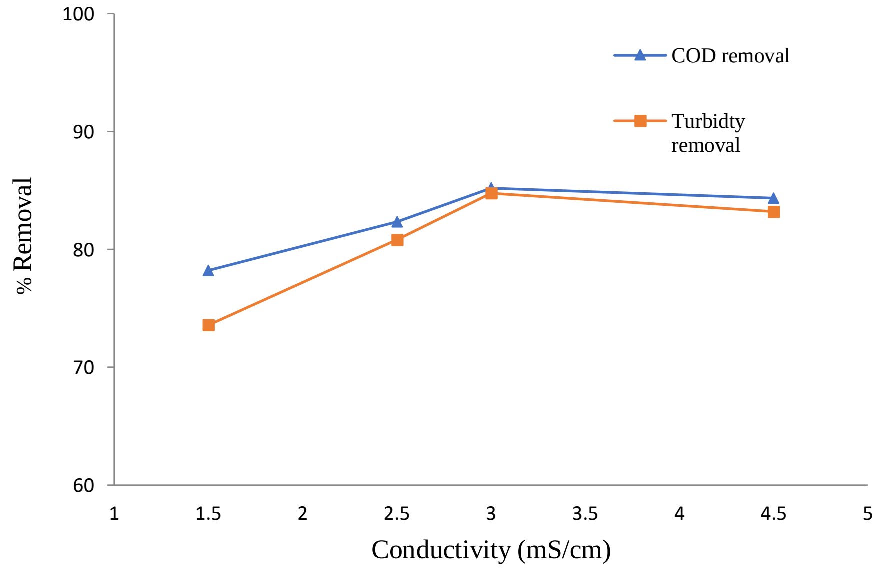

Fig. 4. Effect of conductivity on COD and turbidity removal at pH 6.89, voltage 6 V, runtime 20 min and agitation speed 200 rpm

3) Effect of runtime on COD and turbidity removal efficiency: Runtime or operating time is one of the governing factors of electrocoagulation process. The impact of run time on pollutant removal efficiency is studied by varying the run time, while the other factors are kept constant. Both COD and turbidity removal efficiency increased with increase in runtime. At runtime of 50 min, the COD and turbidity removal efficiency are 89.47% and 87.39% respectively which are maximum values (Fig. 5). As runtime increases the anodic dissolution increases and the formation of metal hydroxyl species also increases. This results in increase of flocs so that the flocs available for pollutant removal are more in number. Eventually as more flocs are available the pollutant removal efficiency also increases [15]. Beyond optimum runtime, even though the flocs formation continues, the pollutant removal efficiency remains constant as the sufficient amount offlocs are available for the pollutants to be removed [16]. The increase in the runtime beyond the optimum value will increase the energy consumption. This leads to rise in the operational cost. Anode dissolution also increases due to increase in runtime which leads to inefficient consumption of anode material and decreases the life of electrodes and needs regular replacement.

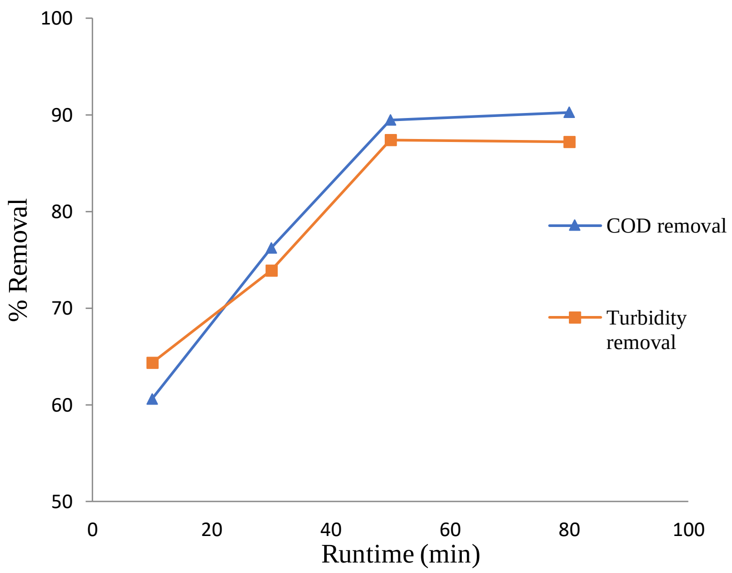

Fig. 5. Effect of voltage on COD and turbidity removal efficiencies; pH 6.89, conductivity 2.24 mS/cm, runtime 20 min and agitation speed 200 rpm.

4) Effect of applied voltage on COD and turbidity removal efficiency: Voltage is an important operating parameter of electrocoagulation process. Voltage for each run is varied and the remaining parameters are kept constant. Both COD and turbidity removal efficiency increased with increase in voltage. Maximum COD and turbidity removal efficiency of 88.58% and 91.33% are observed at 12 V (Fig. 6). As the cell voltage increases, ohmic resistance decreases and current density increases [17]. The coagulant dosage depends on current density. The anodic dissolution rate is directly proportional to the current density. Increase in cell voltage leads to increase in anodic dissolution and finally the formation of metal hydroxides increases. The cell voltage is regulated using a battery eliminator. The bubble formation rate is highly dependent on cell voltage [3]. These bubbles promote the flotation of the pollutants to the surface of the solution. The voltage rise beyond the optimum value will cause increase in operational cost as the electrical energy consumption increases. The current density is directly proportional to voltage. Rise in voltage simultaneously increases the anode dissolution. The current density can be increased by reducing ohmic resistance between the electrodes at constant voltage.

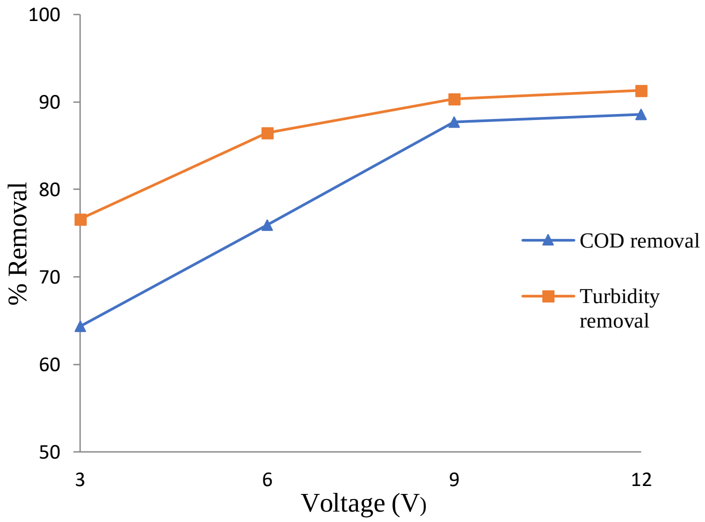

Fig.6. Effect of voltage on COD and turbidity removal efficiencies; pH 6.89, conductivity 2.24 mS/cm, runtime 20 min and agitation speed 200 rpm.

5) Effect of agitation speed on COD and turbidity removal efficiency: Agitation speed or stirring speed is an important operational parameter of electrocoagulation process. The effect of agitation speed on the COD and turbidity removal is studied by running EC process at different agitation speeds, while the other operational parameters under consideration are kept unchanged. COD and turbidity removal efficiency increased with increase in agitation speed till 200 rpm, where the maximum COD and turbidity removal efficiency observed was 88.26% and 90.47% respectively. But a huge drop is observed in COD and turbidity removal efficiency at agitation speed of 300 rpm (Fig. 7).

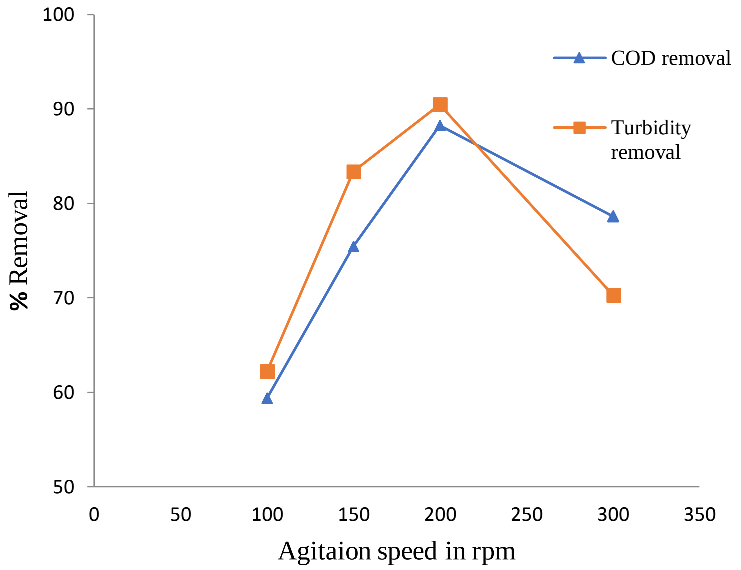

Fig. 7. Effect of agitation speed on COD and turbidity removal; pH 6.89, conductivity 2.24 mS/cm, voltage 6.0 V and runtime 20 min.

Agitation speed influences the performance of the electrocoagulation. Agitation is required to maintain the homogeneity of the mixture in the reactor [18]. It ensures uniform distribution of coagulant produced in the cell. It also enhances the mixing speed of coagulant by imparting velocity to the coagulant and pollutant particles so that the rate of interaction increases. Finally, it results in enhancement of removal efficiency of pollutants. If the agitation speed exceeds the optimum range it results in shearing of the flocs which ultimately reduces the pollutant removal efficiency ([15], [19]). If the agitation speed is less than the optimum range then the coagulant produced is not evenly distributed and offers less pollutant removal efficiency. So, it is necessary to maintain the optimum agitation speed for efficient removal of pollutants.

C. Central Composite Design (CCD)

Circumscribed central composite design is preferred where each factor is studied at five levels and some of the levels are beyond the domain values (Table II).

TABLE II
ORIGINAL AND CODED FACTORS WITH LEVELS IN CCD

Factor	Coded factor	Low	High	-α	+α
Ph	A	6	8	5	9
Conductivity (mS/cm)	B	2.5	3.5	2	4
Runtime (min)	C	30	50	20	60
Voltage (V)	D	4.5	7.5	3	9
Agitation speed (rpm)	E	150	250	100	300

The half fraction central composite design is generated with the different combinations of operating parameters, where 30 experimental runs are obtained. These experimental runs are conducted and the corresponding removal efficiencies are calculated (Table III).

The maximum COD removal efficiency is 91.75 % (Std. Run No. 28). The minimum COD removal efficiency value is 61.32 % (Std. Run No. 2). The maximum turbidity removal efficiency value is 96.76 % (Std. Run No. 18). The minimum turbidity removal efficiency value is 54.64 % (Std. Run No. 20).

TABLE III
CENTRAL-COMPOSITE DESIGN FOR THE GIVEN FACTORS AND THEIR CORRESPONDING REMOVAL EFFICIENCIES

Std. Run	pH	Cond. (mS/cm)	Run time (Min.)	Voltage (V)	Agitation speed (RPM)	COD removal (%)	Turbidity removal (%)
1	6	2.5	30	4.5	250	76.32	61.38
2	8	2.5	30	4.5	150	61.32	80.15
3	6	3.5	30	4.5	150	76.42	64.37
4	8	3.5	30	4.5	250	67.33	83.11
5	6	2.5	50	4.5	150	82.61	69.45
6	8	2.5	50	4.5	250	70.79	82.11
7	6	3.5	50	4.5	250	84.23	72.62
8	8	3.5	50	4.5	150	65.58	78.42
9	6	2.5	30	7.5	150	79.44	73.17
10	8	2.5	30	7.5	250	65.71	85.29
11	6	3.5	30	7.5	250	79.49	75.38
12	8	3.5	30	7.5	150	65.16	80.17
13	6	2.5	50	7.5	250	90.22	75.37
14	8	2.5	50	7.5	150	70.21	83.93
15	6	3.5	50	7.5	150	87.17	71.53
16	8	3.5	50	7.5	250	66.83	90.45
17	5	3.0	40	6	200	84.33	60.48
18	9	3.0	40	6	200	62.48	91.42
19	7	2.0	40	6	200	78.94	81.11
20	7	4.0	40	6	200	86.56	85.32
21	7	3.0	20	6	200	70.72	76.34
22	7	3.0	60	6	200	88.28	88.74
23	7	3.0	40	3	200	65.22	74.97
24	7	3.0	40	9	200	89.54	90.47
25	7	3.0	40	6	100	75.25	75.22
26	7	3.0	40	6	300	84.81	76.83
27	7	3.0	40	6	200	84.76	83.58
28	7	3.0	40	6	200	91.75	88.54
29	7	3.0	40	6	200	88.73	87.95
30	7	3.0	40	6	200	86.44	85.52

D. RSM Optimization

1) Statistical analysis of COD removal efficiency: The CCD data is analyzed utilizing ANOVA to determine the model suitability and model significance (Table IV). The terms or factors whose 'p - values' are $p < 0.05$ and $p < 0.01$ are considered as significant and highly significant terms respectively, where similar studies are carried out by Nawel et.al [5].

TABLE IV
ANOVA TABLE FOR COD REMOVAL EFFICIENCY

Parameter	Sum of Squares	Df	F Value	P value	Estimated coefficients	
Model	2435.4690	20	4.4426	0.0132	88.45	Significant
A	1157.4537	1	42.227	0.0001	-6.94	
B	4.8870	1	0.1782	0.6828	0.45	
C	277.2360	1	10.1144	0.0112	3.40	
D	194.1997	1	7.0850	0.0260	2.84	
E	43.0140	1	1.5692	0.2419	1.34	
AB	0.2139	1	0.0078	0.9315	-0.12	
AC	21.7855	1	0.7948	0.3959	-1.17	
AD	11.9889	1	0.4373	0.5250	-0.87	
AE	0.8883	1	0.0324	0.8611	0.24	
BC	15.2685	1	0.5570	0.4745	-0.98	
BD	5.5814	1	0.2036	0.6625	-0.59	
BE	2.1830	1	0.0796	0.7842	-0.37	
CD	0.4935	1	0.0180	0.8962	0.18	
CE	6.25E-06	1	2.28E-07	0.9996	-6.250E-004	
DE	9.7188	1	0.3545	0.5662	-0.78	
A^2	432.1508	1	15.7662	0.0033	-4.03	
B^2	76.1063	1	2.7766	0.1300	-1.69	
C^2	166.9167	1	6.0896	0.0357	-2.50	
D^2	245.1270	1	8.9430	0.0152	-3.30	
E^2	149.7050	1	5.4617	0.0442	-2.37	
Residual	246.6890	9				
Lack of Fit	219.1880	6	3.9850	0.1419		Not Significant
Pure Error	27.501	3				

From the above table it is evident that the linear terms A, C and D are significant as their p - value is less than 0.05. The quadratic parameter of C, D and E are significant as their p-value is less than 0.05. Quadratic parameter of A is highly significant as its p-value is less than 0.01. The model F-value of 4.44 implies the model is significant. There is only a 1.32 % chance that a "model F-value" this large could occur due to noise. The coefficient of determination (R^2) is 0.9080 denotes that the sample variations of 90.8% efficiency were assigned to independent variables only 9.2% of the variation could not be explained by the model. The 'adjusted R^2 value is 0.7836 which is having a reasonable agreement with R^2indicating a good statistical model [20]. The "Lack of fit F-value" for the model is 3.98 and it is not significant relative to pure error which confirms the validity of the model. The lack of fit p-value is 0.1419 which means lack of fit is not significant. The "adequate precision" ratio of the model is 7.639 which indicates that the model is reliable and reproducible. These results conclude that the model suits best for the COD removal by electrocoagulation from the urban wastewater. The second order quadratic polynomial equation correlating the COD removal efficiency and electrocoagulation treatment parameters is given below which includes both significant and non-significant parameters in it.

$$\begin{aligned} y = \; & 88.45 - 6.94A + 0.45B + 3.40C + 2.84D + 1.34E - 0.12AB - 1.17AC \\ & - 0.87AD + \quad 0.24AE - 0.98BC - 0.59BD - 0.37BE + 0.18CD \\ & - (6.205E + 004)CE - 0.78DE - \quad 4.03A^2 - 1.69B^2 - 2.50C^2 \\ & - 3.30D^2 - 2.37E^2 \end{aligned}$$

Here, y - COD removal efficiency, A - pH, B - conductivity, C-runtime, D- voltage and E- RPM

2) Statistical analysis of turbidity removal efficiency: The central composite design data is analyzed utilizing ANOVA to determine the model suitability and model significance (Table V). The terms or factors whose 'p - values' are $p < 0.05$ and $p < 0.01$ are considered as significant and highly significant terms respectively.

TABLE V
ANOVA TABLE FOR TURBIDITY REMOVAL EFFICIENCY

Parameter	Sum of Squares	Df	F value	P value	Estimated Coefficients	
Model	1966.3768	20	11.7563	0.0003	86.89	Significant
A	1096.7424	1	131.1416	0.00009	6.76	
B	7.7293	1	0.9242	0.3615	0.57	
C	86.8681	1	10.3871	0.0104	1.90	
D	232.3792	1	27.7864	0.0005	3.11	
E	32.06281	1	3.8338	0.0819	1.16	
AB	0.9312	1	0.1113	0.7463	-0.24	
AC	4.4944	1	0.5374	0.4822	-0.53	
AD	8.3810	1	1.0021	0.3429	-0.72	
AE	9.0902	1	1.0869	0.3243	0.75	
BC	0.0484	1	0.0058	0.9410	-0.055	
BD	2.0022	1	0.2394	0.6363	-0.35	
BE	54.8340	1	6.5567	0.0307	1.85	
CD	2.4964	1	0.2985	0.5981	-0.39	
CE	6.1504	1	0.7354	0.4134	0.62	
DE	7.3712	1	0.8814	0.3723	0.68	
A^2	236.5326	1	28.283	0.0005	-2.98	
B^2	36.0065	1	4.3054	0.0678	-1.16	
C^2	47.2238	1	5.6467	0.0415	-1.33	
D^2	44.0840	1	5.2713	0.0473	-1.29	
E^2	233.5637	1	27.9280	0.0005	-2.96	
Residual	75.2673	9				
Lackof Fit	59.5584	6	1.8957	0.3205		Not Significant
Pure Error	15.7088	3				

The linear terms A, C and D are significant as their p-value is less than 0.05. The quadratic parameter of A, C, D and E are significant as their p - value is less than 0.05, quadratic parameters of A, E are highly significant as its p-value is less than 0.01. The interaction parameter BE is significant as its p-value is less than 0.05. The model F-value of 11.7 implies that the model is significant. There is only a 0.3% chance that a "model F-value" this large could occur due to noise. The coefficient of determination (R^2) 0.963 denotes that the sample variations of 96.3% efficiency are assigned to independent variables and 7.28% of the variation could not be explained by the model. The 'adjusted R^2 value 0.881 is in reasonable agreement with R^2 which implies the model obtained is a good statistical model. The "lack of fit" F-value for the model is 1.89 and it is not significant relative to pure error which confirms the validity of the model. The "lack of fit" p-value is 0.32 which means lack of fit is not significant. The "adequate precision" ratio of the model is 13.43 which indicates the model is reliable and reproducible. These results conclude that the model suits best for the turbidity removal from urban wastewater by electrocoagulation. The second order quadratic polynomial equation correlating the turbidity removal efficiency and electrocoagulation treatment parameters is given below which includes both significant and non-significant parameters in it.

$$y = 86.89 + 6.76A - 0.57B + 1.90C + 3.11D + 1.16E - 0.24AB - 0.53AC - 0.72AD + 0.75AE - 0.55BC - 0.35BD + 1.85BE - 0.39CD + 0.62CE + 0.68DE - 2.98A^2 - 1.16B^2 - 1.33C^2 - 1.29D^2 - 2.96E^2 \quad (9)$$

Here, y- Turbidity removal efficiency, A- pH, B- Conductivity, C- Runtime, D- Voltage, E- RPM

3) Response surface plots of COD removal efficiency: The COD removal efficiency is plotted along the z-axis against the independent variables and the remaining independent variables are kept stationary or at zero level (Fig. 8 (a-j)).

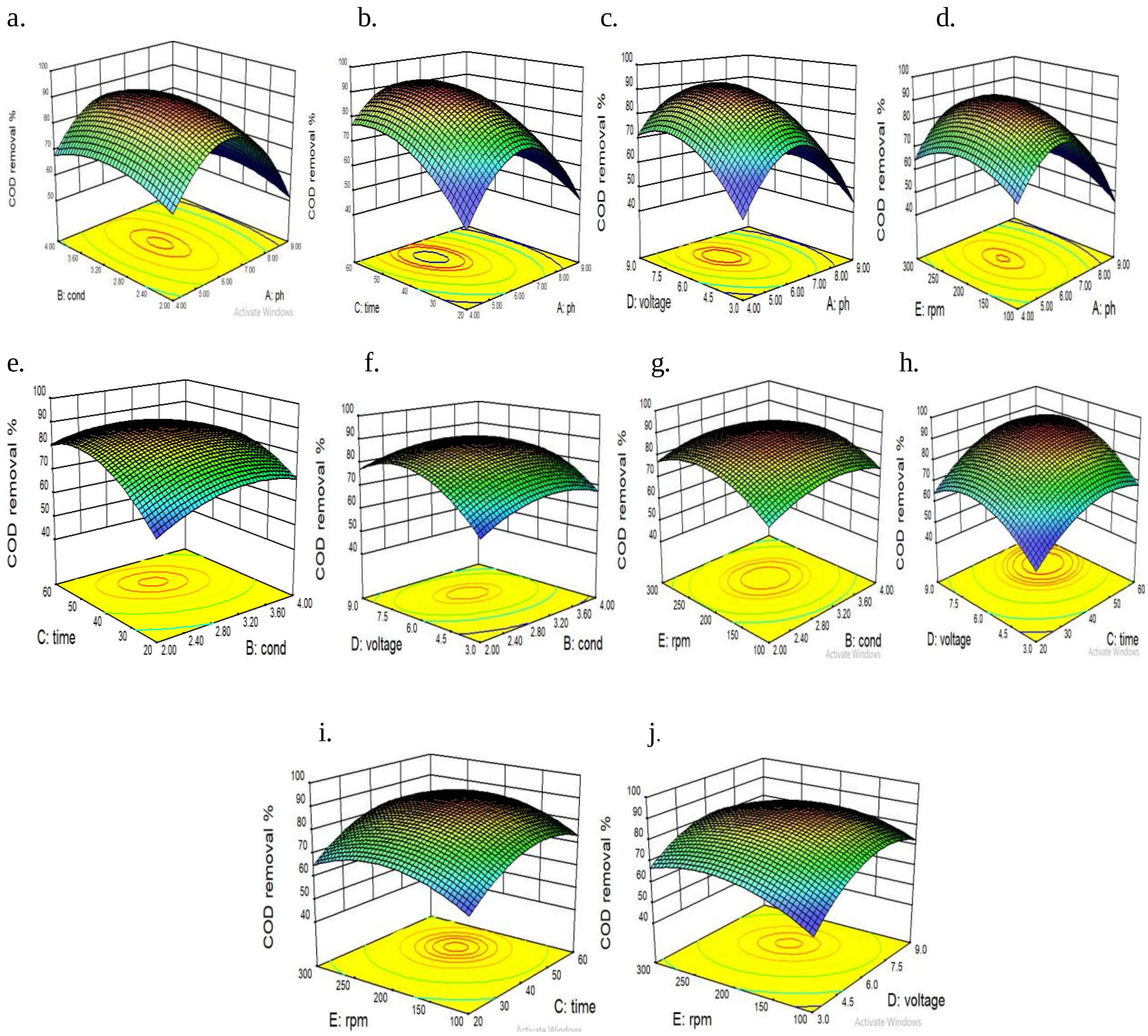

Fig. 8. Response surface plots; interactive effects of pH, conductivity, runtime, voltage and agitation speed (a-d); conductivity, runtime, voltage and agitation speed (e-g); voltage, runtime, agitation speed (h-i); voltage and agitation speed on COD removal efficiency.

4) Response surface plots of turbidity removal efficiency: The turbidity removal efficiency is plotted along the z-axis against the independent variables and the remaining independent variables are kept stationary or at zero level (Fig. 9 (a-j)).

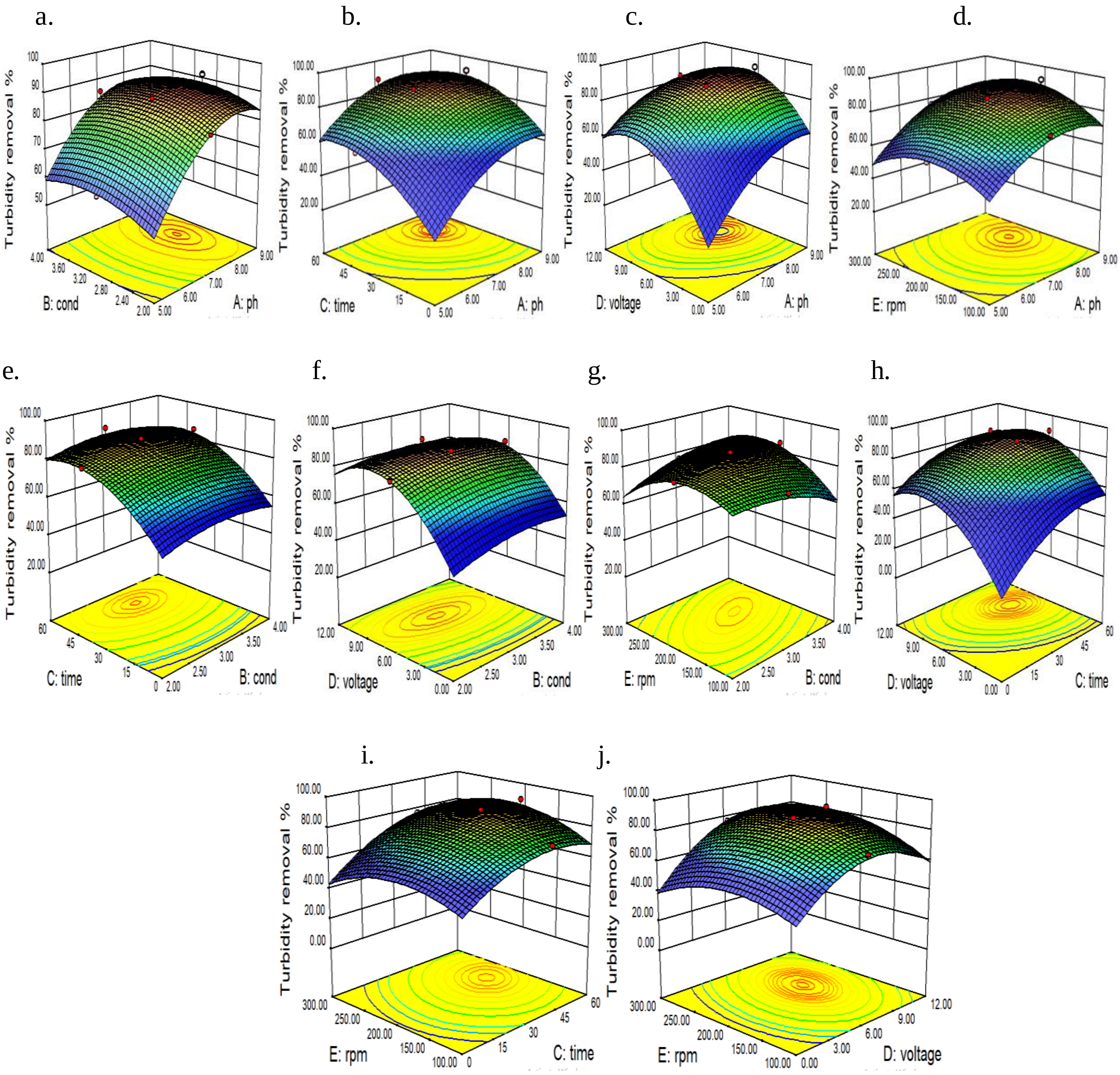

Fig. 9. Response surface plots; interactive effects of pH, conductivity, runtime, voltage and agitation speed (a-d); conductivity, runtime, voltage and agitation speed (e-g); voltage, runtime, agitation speed (h-i); voltage and agitation speed on turbidity removal efficiency.

3) Validation and verification of both COD and turbidity models: The reliability of the model equation is to be validated and verified. The model is optimized and the optimal independent variable values are developed. Target responses are entered and predicted optimal response values are generated. Response values are to be set to maximum efficiency. The model predicted COD and turbidity removal efficiencies are verified and validated by conducting the experimental runs with the operational settings generated by the model. The predicted response values (P) for the given operational parameters are found to be in reasonable agreement with the actual or experimental response values (A). These results confirmed the adequacy of the derived regression model in reflecting the expected optimization (Table VI) ([5],[17]).

TABLE VI
PREDICTED AND ACTUAL EXPERIMENTAL VALUES OF COD AND TURBIDITY REMOVAL EFFICIENCY AT THE OPTIMIZED OPERATING CONDITIONS

pH	Conductivity (mS/cm)	Runtime (min)	Voltage (V)	Agitation Speed (RPM)	COD removal % (P)	Turbidity removal % (P)	Desirability	COD removal % (A)	Turbidity removal % (A)
6.95	3.07	48	6.0	215	90.10	87.56	0.910	88.42	85.16
6.00	3.35	45	7.5	221	91.30	80.70	1.000	90.46	79.85
6.67	2.89	48	9.0	210	86.24	86.40	0.850	87.62	85.33
6.74	2.93	47	8.5	210	88.70	87.53	0.890	86.72	85.92
8.00	3.50	50	7.5	250	71.40	92.27	1.000	70.39	91.58

E. Electrode Consumption

The anodic dissolution during the electrocoagulation process is dependent on the current density and time. The optimal pollutant removal efficiencies are obtained at the current density 0.0161 A/cm^2, beyond this value the removal efficiency doesn't show any increment (Table VII). Further increase in runtime and voltage impacts operational cost. The electrode consumption corresponding to the optimal current density is 0.00432 g/cm^2, which is equivalent to 0.129 kg Al per m^3 of urban wastewater. The aluminium consumed by EC process for the efficient pollutant removal is low in comparison with chemical coagulation, where 78% of COD removal was obtained at the aluminium dosage of 0.32 kg/m^3 [4].

TABLE VII
ANODE DISSOLUTION VALUES AT OPTIMIZED OPERATING CONDITIONS.

Voltage (V)	Current density (A/cm^2)	Run time (Min)	COD removal (%)	Turbidity removal (%)	W (g/cm^2)
6.0	0.0161	48	88.42	85.16	0.00432
7.5	0.0172	45	90.46	79.85	0.00430
7.5	0.0179	50	70.39	91.58	0.00500
8.0	0.0178	47	86.72	85.92	0.00467
9.0	0.0189	48	87.62	85.33	0.00507

F. Energy Consumption

Energy consumption is highly dependent on voltage and runtime. The maximum and minimum energy consumption values for the optimized experimental conditions are 1.68 KWh/m^3 and 3.24 KWh/m^3 respectively (Table VIII). The operational cost will be economical if the energy consumption is low.

TABLE VIII
ELECTRICAL ENERGY CONSUMPTION IN KWh/m^3 AT THE OPTIMIZED EXPERIMENTAL CONDITIONS.

Voltage (V)	Current (A)	Runtime (min)	Treated volume (m^3)	Energy consumption (KWh/m^3)
6.0	0.70	48	0.002	1.68
7.5	0.85	45	0.002	2.39
7.5	0.85	50	0.002	2.65
8.0	0.89	47	0.002	2.78
9.0	0.90	48	0.002	3.24

G. Amount of Sludge Produced

Sludge is accumulated on the surface of the wastewater during electrocoagulation. It is observed that at the optimal operating condition, when the initial COD and turbidity values are 653 mg/l and 105 NTU respectively, the sludge obtained is 0.102 kg/m^3. The sludge produced by chemical coagulation of wastewater ranges 0.24 – 0.4 kg/m^3 [21]. The quantity of sludge produced is lower by electrocoagulation.

H. Cost Analysis

Electrode dissolution and the energy consumption are the major cost affecting factors. The electrode consumption at the optimized experimental conditions is 0.129 kg/m^3. The electricity consumption at the optimized pollutant removal is 1.68 KWh/m^3. Unit price of electricity according to tariff order of FY 2021-22 – APSPDCL is Rs.7.25 per unit. Cost of aluminium in Indian market is Rs. 200 per kg.

$$\text{Operating cost in INR} = i\, C_{electrode} + j\, C_{electricity}$$
$$= 200 \times 0.129 + 7.25 \times 1.68$$
$$= 25.8 + 12.18$$
$$= 37.98 \text{ INR}$$

The operating cost for the treatment of urban wastewater by electrocoagulation is Rs. 37.98 per Cu.m. The aluminium dosage of 0.32 kg/m^3 is required for 78% of COD removal through chemical coagulation. Power consumption for rapid mixing of coagulant is 0.42 KWh/m^3. Cost of alum powder (aluminium sulfate) in Indian market is Rs. 300 per kg.

$$\text{Operating cost in INR} = i\, C_{coagulant} + j\, C_{mixing}$$
$$= 0.32 \times 300 + 7.25 \times 0.42$$
$$= 99.04 \text{ INR.}$$

The operating cost for chemical coagulation is Rs. 99.04 per Cu.m. A saving of Rs. 61 per Cu.m. is achieved. Cost reduction of 62 % is achieved through electrocoagulation with higher pollutant removal efficiencies compared to conventional chemical coagulation.

4. CONCLUSIONS

Electrocoagulation with aluminum electrodes has reduced COD and turbidity of urban wastewater sample by 88.42% and 85.16% respectively at the optimum operating conditions provided by the response surface methodology. The optimum conditions for the experimentation are initial pH 6.95, conductivity 3.07 mS/cm, voltage 6.0 V, current density 0.0161 mA/cm^2, runtime 48 min, agitation speed 215 rpm. Among all the operational parameters pH, runtime and applied voltage exhibited significant effect on the pollutant removal. The anodic dissolution at optimal conditions is 0.00432 g/cm^2. Aluminium consumption reduced by almost 58% and the operating cost is reduced by 62% compared to chemical coagulation. The energy consumed for the efficient pollutant removal is 1.68 KWh/m^3 and the quantity of sludge produced is 0.102 kg/m^3. The treated water is fit to be discharged into natural water bodies and can be used for agricultural activity reducing load on irrigational reservoirs.

5. REFERENCES

[1] P. D. Rimeh Daghrir, "Coupled electrocoagulaion-electro-fenton for efficient domestic wastewater treatment," *Environ Chem Lett. 11,* pp. 151-156, 2013.

[2] C. p. E.-B. S. S. K. Tak-Hyun Kim, "Decolorization of disperse and reactive dyes by continuous electrocoagulation process," *Desalination 150* , pp. 165-175, 2002.

[3] B. M. P. C. O. Sahu, "Treatment of wastewater by electrocoagulation: a review," *Environ. Sci. Pollut. Res. - Int. 21 (4),* pp. 2397-2413, https://doi.org/10.1007/s11356-013-2208-6, 2014.

[4] M. K. E. M. B. O.T. Can, "Treatment of textile wastewater by combined Electrocoagulation," *Chemosphere 62,* pp. 181-187, 2006.

[5] D. F. M. B. L. J.-P. M. K. Adjeroud Nawel, "Improvement of electrocoagulation-electroflotation treatment of effluent by addition of Opuntia ficus indica pad juice," *Separation and purification technology 144,* pp. 168-176, 2015.

[6] K. H. M. E. M. Elazzouzi, "Electrocoagulation flocculation as a low-cost process for pollutants removal from urban wastewater," *Chemical engineering research and design volume 117,* pp. 614-626, https://doi.org/10.1016/j.cherd.2016.11.011, 2017.

[7] L. S. Z. M. S. N. S. K. N. W. A. Z. M. Nasrullah, "Treatment of palm oil mill effluent by electrocoagulation with presence of hydrogen peroxide as oxidizing agent and polialuminum chloride as coagulant-aid," *Water Resour. Ind. 17,* pp. 7-10, https://doi/. org/10.1016/j.wri.2016.11.001., 2017.

[8] C. t. I. c. M. A. R. Laura Zaleschi, "A Comparative Study of Electrocoagulation and Chemical coagulation processes applied for wastewater treatment," *Environmental Engineering and Management journal, Vol.11, No. 8,* pp. 1517-1525, 2012.

[9] S. Didar-Ul-Islam, "Electrocoagulaion (EC) technology for wastewater treatment and pollutants removal," *Sustain. Water Resour. Manag,* pp. doi 10.1007/s40899-017-0152-1., 2017.

[10] L. T. S. Carpinteyro-urban, "Use of response surface methodology in the optimization of coagulation-floccuclation of wastewaters employing biopolymers,," *Int. j. Environ. Res.7,* 2013.

[11] G. C. P. Y. X. Chen, "Separation of pollutants from restaurant wastewater by electrocoagulation," *Sep. Purif. Technol. 19* , pp. 65-76, 2000.

[12] O. Sahu, "Suitability of aluminum material on sugar industry wastewater with chemical and electrochemical treatment process," *International journal of Industrial chemistry10,* pp. 335-347,https://doi.org/10.1007/s40090-019-00196-8, 2019.

[13] J. W. Chia Miang Khor, "Performance, Energy and Cost of Produced water treatment by Chemical and Electrochemical coagulation," *Water,* pp. 12,3426; doi: 10.3390/12123426, 2020.

[14] O. C. M. B. M Kobya, "Treatment of textile wastewater by electrocoagulation using iron and aluminum electrodes," *Journal of Hazardous Materials B 100,* pp. 163-178, https://doi.org/10.1016/S0304-3894(03)00102-X, 2003.

[15] S. C. Z. Z. M. A. P. A. A.S. Naje, "A review of electrocoagulation technology for the treatment of textile wastewater," *Rev. Chem. Eng. 33 (3),* pp. 263-292, 2017.

[16] L. M. A. A.-M. a. A. H. M. Edris Bazrafshan, "Heavy metals removal from aqueous environments by electrocoagulation process- a systematic review," *J Environ Health Sci Engineer 13, 74,* pp. https://doi.org/10.1186/s40201-015-0233-8, 2015.

[17] M. O. Orathai Chavalparit, "Optimizing electrocoagulation process for the treatment of biodiesel wastewater using response surface methodology," *Journal of environmental sciences 21,* pp. 1491-1496, 2009.

[18] M. S. J. F. A. Amina Tahreen, "Role of electrocoagulationin wastewater treatment: A developmental review," *journal of water process engineering 37,* p. https://doi.org/10.1016/j.jwpe.2020.101440, 2020.

[19] A. S. V. Khandegar, "Electrocoagulation for the treatment of textile industry effluent - a review," *J. Environ. Manage. 128* , pp. 949-963, https://doi.org/10, 2013.

[20] M. H. L. H. P. F. L. W. J. Z. G. Zhang, "Optimization of a hydrocolloid extract fromwaste peanut shells and evaluation of its antioxidant and antibacterial activities in vitro.," *Food Bioprod. Process 91,* pp. 158-168, 2013.

[21] F. L. B. George Tchobanoglous, Wastewater Engineering: Treatment and resuse/Metcalf & Eddy, Inc, 4th edition, McGraw Hill Education (India) pvt. ltd., 2003.