



## Treatment Of Pharmaceutical Wastewater By Electrocoagulation & Optimization Using Response Surface Methodology

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**Abstract:** Pharmaceutical wastewater is considered as one of the primary source of environmental pollution. Their occurrence, fate and transformation in the environment are causing significant concerns. The presence of a high COD, dissolved organic nitrogen, total solids, chloride, and complex compounds, etc. makes pharmaceutical wastewater more toxic and unfit. Electrocoagulation (EC) is a process in which electric current is supplied to sacrificial electrodes for the removal of pharmaceutical contaminants from wastewater. The study was experimentally investigated taking into account various factors such as current density (30-100mA/cm<sup>2</sup>), distance between the electrodes (10-15mm), and electrolysis time (60-120 min) which has impact on the % reduction of chemical oxygen demand (COD), using GI galvanized iron electrode. The response surface design based on the central composite design (CCD) has been used to optimize different process parameters for treatment of pharmaceutical wastewater using Electrocoagulation process. The % reduction of COD under different operating conditions was predicted with the help of a quadratic model. The significance and their interaction with independent variables was assessed by analysis of variance (ANOVA). The optimum operating conditions were obtained through mathematical and statistical methods to reach maximum % COD reduction. The result shows that the maximum reduction of COD (94%) was achieved at pH-7, current density-114.24 mA/cm<sup>2</sup>, distance between electrodes-10mm and reaction time 132 minutes. This means that, the process of EC can remove pharmaceutical pollutants from pharmaceutical industrial wastewaters effluent under different operating conditions.

**Index Terms**–Electrocoagulation, Pharmaceutical wastewater, response surface methodology.

### I. INTRODUCTION

#### 1.1 Pharmaceutical industry

Pharmaceutical industry is one of the category among the others that generates strong wastewater of high COD which require effective treatment and disposal effluents. Generally the pharmaceutical industries are provided with conventional ETPs based on activated sludge process. <sup>[58]</sup>The Conventional pharmaceutical industry ETPs consists of

1. Primary treatment (neutralization, flocculation & primary settling),
2. Secondary treatment (biological treatment comprising of surface aeration, secondary settling)
3. Tertiary treatment (dual media filtration: pressure sand & activated carbon filter, MEE, RO).

<sup>[58]</sup>According to their production activity, the pharmaceutical industry can be split into two groups Active Pharmaceutical Ingredient (API) and Formulation. <sup>[58]</sup>Bulk drug means a pharmaceutical, chemical, biological or plant product including its salts, esters, stereo-isomers and derivative which can be used in such form or in the formulation as an ingredient. Three different methods used for manufacturing Bulk Drugs i.e. Biological & Natural Product Extraction, Chemical Synthesis and Fermentation. Active Pharmaceutical Ingredient (API) goes for “Formulation” which is then available as Tablets, Capsules, Liquids and Ointments. Chemicals used in chemical synthesis largely include organic and inorganic reactants and catalysts. Formulation refers to a medicine made from or containing one or more bulk pharmaceuticals, with or without pharmacological aids, for internal or external use in the diagnosis, treatment, mitigation, or prevention of disease in humans or animals.

Environmental impacts of pharmaceuticals effluent are significant. <sup>[10]</sup>The Pharmaceutical residues have been found in aquatic environmental samples such as groundwater, surface water and municipal wastewater. <sup>[13]</sup>Pharmaceutical drugs given to people as well as to domestic animals are excreted and distributed into the environment by flushing toilets as well as by spreading manure and sewage sludge onto the soil <sup>[26]</sup>. These chemicals persist in the environment for longer time, enter the food chain, bio accumulate, bio magnify and cause harmful effects in wildlife and humans.

## 1.2 Electrocoagulation Process

Electrocoagulation is the process in which metal ions are produced in the electrolyte. The basic principle of Electrocoagulation is “Electrolysis” which means to break down the substances apart using electricity<sup>[18]</sup>. These metal ions act as in-situ coagulant which destabilizes and neutralizes the electrical charges of pollutants present in wastewater<sup>[3]</sup>. The mechanism of Electrochemical process in aqueous medium is quite complex. There are 3 possible mechanism taking place in electrochemical process like electrocoagulation (EC), electro floatation (EF), and electro oxidation (EO).<sup>[15]</sup> Electrocoagulation is the process of destabilizing dissolved, emulsified and suspended contaminants present in an aqueous medium by using electricity. The metal hydroxide cations formed in aqueous medium takes part in EC process to remove pollutants by sorption, coagulation, and other process occurring in the space between the electrodes<sup>[8]</sup>. Electro floatation is the process in which hydrogen and oxygen bubbles generated takes pollutants and particles along with them to the surface by buoyant force.<sup>[16]</sup> In Electro oxidation organic materials get decompose by means of oxidation into carbon dioxide and water and other oxides.

Electrocoagulation is a technique that evolved from traditional chemical coagulation. The electrocoagulation process causes colloidal particles in an aqueous media to coagulate when an electric current is applied<sup>[9]</sup>. Contaminants that are suspended, emulsified, or dissolved are destabilized during this procedure. Electric current is applied to electrodes in an electrolytic cell, which produces a coagulating agent as well as gas bubbles. The cathode produces hydrogen, and the bubbles aid in the elimination of contaminants<sup>[5]</sup>. Filtration can easily remove these flocs, which have good stability, low bound water, and a big particle size. The following factors contribute to the interest in electrocoagulation for water treatment: in situ generation of coagulants, which minimizes the expense of chemical transportation and storage in large-scale applications<sup>[10]</sup>. During electrocoagulation, the effluent is not enriched with anions and salts, as it is in chemical coagulation, resulting in compact sludge generation, the following reaction takes places during Electrocoagulation process.

### At the Anode:



### At the Cathode:



Metal anode generates cations as per above reactions.<sup>[21]</sup> Highly charged cations forms polyvalent polyhydroxide complexes which destabilizes the colloidal particles. As these complexes have high adsorption properties they start forming aggregates with pollutants<sup>[12]</sup>. Formation of hydrogen and oxygen gases at cathode aids in mixing and flocculation process. The summarized destabilization mechanism of pollutants, particulate suspension, and breaking of emulsions taking place in a Electrocoagulation reactor is as follows.

1. Compression of diffuse double layer around the charged particles generated due to oxidation of anode metal metal.
2. Charge neutralization of ionic species present in wastewater by ions produced at anode.
3. Floc generated as result of coagulation creates sludge that traps and bridges the colloidal particles to settle which are still in suspension.

<sup>[24]</sup> Electrocoagulation process is much affected by the parameters like type of electrode materials, electrode gap, current density, reaction time, initial pH, conductivity, Total dissolved solids, total suspended solids, electrode arrangement, shape of electrode, stirring time, etc.

## 1.3. Response Surface Methodology

Design of Experiment (DoE) is a set of useful mathematical strategies for statistical modeling and systematic study of a problem by employing variables or factors to optimize intended responses or output measurements<sup>[41, 42]</sup>. Response Surface Methodology (RSM) is a common empirical statistic method used to set mathematical models, optimize multi-factor tests, and explore relationships between the response and explanatory variable<sup>[44]</sup>. RSM is a key consecutive technology for original process, improving the design and formulation of new products and maximizing their performance<sup>[41, 43]</sup>.

According to<sup>[45]</sup>, the Box–Behnken Design (BBD) and the Central Composite Design (CCD) are two most common RSM design types that provide a sufficient amount of information for statistically analyzing lack of fit, which is required for subsequent tests. BBD is used to design all quantitative numerical values varied over three levels, according to<sup>[46]</sup>, and<sup>[47]</sup> while CCD was employed to analyze the lower experiment. The CCD was employed to investigate the effects of factors on responses and in subsequent optimization studies, as this technique is suitable for the installation of the quadratic surface and improves viable parameters with a small number of experiments<sup>[48]</sup>

The majority of previous researches concentrated on the removal of contaminants using synthetic solutions via the EC process, with only a small amount of work using real wastewater. Furthermore, the majority of the research concentrated on wastewater pollutant removal efficiency<sup>[34]</sup>. From an economical point of view, energy consumption is an important component in the EC process. As a result, the current research focuses on calculating the % reduction COD from pharmaceutical industrial wastewater using an EC process<sup>[35]</sup>.

Several parameters were chosen to optimize statistically through RSM for the electrocoagulation process. CCD used RSM to improve EC process operating parameters such as current density (A), distance between electrodes (B) and reaction time (C)<sup>[28]</sup>. The key objective of the optimization is to increase % COD reduction from pharmaceutical industrial wastewater at optimum operating conditions<sup>[29]</sup>. The DoE Software was used to optimize the influence of the selected operating variables on wastewater treatment efficiency and to investigate the combined effect of CCD analyzes using statistical analysis of the selected variables<sup>[37]</sup>.

## II. MATERIALS AND METHODS

### 2.1. Wastewater characteristics

The pharmaceutical wastewater obtained from APIs manufacturing industry was sampled directly. The Characteristics of the wastewater used in this study is shown in Table.2.1

Table.2.1 Main characteristics of pharmaceutical wastewater

Parameters	Unit	Amount
pH	-	7
Conductivity	mS/cm	4.54
COD	mg/l	6800
TDS	mg/l	300
Color	Co.pt	469.237

### 2.2. Reagents

The various reagents used in this study, analytically for determining the COD are such as Potassium dichromate K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub>, Sulphuric acid H<sub>2</sub>SO<sub>4</sub>, Silver sulphate AgSO<sub>4</sub> and mercuric sulphate HgSO<sub>4</sub> to produce CO<sub>2</sub> and H<sub>2</sub>O, ferrous ammonium sulphate (FAS) of 0.1N and Ferroin as an indicator, and distilled water.

### 2.3. Experimental setup

The pharmaceutical wastewater sample required for this study was collected from pharmaceutical industry. The experiment setup is shown in Fig.2.1. All the experiments were conducted in a batch mode with dimension 15cmx12cmx10cm to hold a sample of 1.5L and it is made from polypropylene material. All the experiments were conducted at ambient temperature (25±2°C). The Galvanized iron plates of dimension 100mmx120mmx10mm was used as anode and cathode electrode. The distance between the two electrodes anode and cathode was kept 10mm and 15mm. The effective surface area of each electrode was 50cm<sup>2</sup>. The electrode were connected to a DC power supply in a parallel mode. The electrodes used were cleaned manually with 35% hydrochloric acid followed by washing with de-ionized water and dried after each experiment for their reuse. The EC performance was determined by optimizing the operating parameters from varying the ranges; current density (30-100 mA/cm<sup>2</sup>), electrode gap (10mm & 15mm), electrolysis time (60-120 minutes) using RSM. Afterwards the solution was permitted to rest so that flocs and suspended particles gets settled easily. The supernatant water then filtered through Wattmann 42 filter paper, and analyzed for COD reduction.

### 2.4. Design of experiment for optimization

A CCD was executed for three independent variables and DoE is used to minimize the number of runs and needed to combine various independent variables. The parameters selected are current density(A), distance between electrodes (B), and the reaction time (C). The coded values of variables are showed in Table 2.2 and an experimental design matrix resulting from Central Composite Design was revealed in Tables 2.3 and it consists of 26 coded conditions for GI–GI electrode combination.

Variables	Unit	Factors	Levels	
			-1	+1
Electrode gap	Mm	A	10	15
Current density	mA/cm <sup>2</sup>	B	30	100
Run time	Minutes	C	60	120

Table 2.2 Coded values of the variables for the design of experiments for the electrocoagulation process.

### 2.5. Removal analysis

The reduction % of COD [45, 49, 50, 51] were determined according to the formula given in Equation.(4).

$$\text{COD reduction \%} = \frac{(\text{COD}_0 - \text{COD}_t)}{\text{COD}_0} \times 100$$

Where, COD<sub>0</sub> and COD<sub>t</sub> are the chemical oxygen demand at time = 0 (initial) and at t (reaction time, t) respectively

**III. RESULTS AND DISCUSSIONS**

## 3.1 Statistical analysis

Minitab software was used for determination of results of response COD reduction for the treatment of Pharmaceutical wastewater during EC process. The efficiency of response were greatly affected by current density, electrolysis time, electrode gap, pH, conductivity, TDS. For the investigation of the combined influence of components, a statistically designed experiment was utilized, and physical characteristics were used to determine the performance of trials in various combinations. A central composite design (CCD) is a widely used design with RSM, which include runs with factors at their extreme limits.

Table 2.3 Experimental conditions and responses

Sr. No.	Run Time	Current Density	Electrode Gap	% Reduction
1	47.57	65	15	41.18
2	120	100	10	88.24
3	90	65	10	58.82
4	90	65	10	58.82
5	47.57	65	10	47.06
6	120	100	15	82.35
7	60	100	15	47.06
8	90	65	15	52.94
9	90	65	15	52.94
10	90	15.5	15	41.18
11	90	65	15	52.94
12	90	65	15	52.94
13	90	65	10	58.82
14	132.4	65	15	64.71
15	60	30	15	23.53
16	60	100	10	52.94
17	120	30	15	52.94
18	90	65	15	52.94
19	90	65	10	58.82
20	90	15.5	10	47.06
21	120	30	10	64.71
22	90	114.49	15	82.35
23	60	30	10	52.94
24	90	114.49	10	88.24
25	90	65	10	58.82
26	132.4	65	10	76.47

Using Minitab software trial version. Analysis of variance (ANOVA) was utilized to evaluate the data. The total of 26 experiments were performed. In the present study, the regression model analysis was done for the response: % reduction of COD with GI electrode by using the terms of encoded factors and the results of analysis are summarized.

Based on the experimental results of EC and regression analysis for CCD, the full quadratic model is given by  
Regression equation uncoded units for electrode gap =10mm

$$10 \text{ COD Reduction\%} = 41.1 + 0.141 \text{ Run Time} - 0.413 \text{ Current Density} \\ + 0.00000 \text{ Run Time*Run Time} \\ + 0.00300 \text{ Current Density*Current Density} \\ + 0.00350 \text{ Run Time*Current Density}$$

Regression equation uncoded units for electrode gap =15mm

$$15 \text{ COD Reduction\%} = 22.2 + 0.180 \text{ Run Time} - 0.308 \text{ Current Density} \\ + 0.00000 \text{ Run Time*Run Time} \\ + 0.00300 \text{ Current Density*Current Density} \\ + 0.00350 \text{ Run Time*Current Density}$$

The test run values along with their optimal operating conditions for GI electrode are given in Table.3.1

Table 3.1 Optimum Operating for COD reduction

Electrode	pH	Conductivity	Current density	time	%COD Reduction
GI electrode	7	4.54	114mA/cm2	132 mins	94%

The P-Value is 0.001 which is lower than 0.05, this confirm that the regression model is statistically significant. The Analysis of variance is shown in Table.3.2

Table 3.2 Analysis of Variance (ANOVA)

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Model	8	5342.91	667.86	26.79	0.000
Linear	3	4983.90	1661.30	66.65	0.000
Run Time	1	2177.12	2177.12	87.34	0.000
Current Density	1	2326.39	2326.39	93.33	0.000
Electrode Gap	1	480.40	480.40	19.27	0.000
Square	2	191.46	95.73	3.84	0.042
Run Time*Run Time	1	0.00	0.00	0.00	0.999
Current Density*Current Density	1	188.22	188.22	7.55	0.014
2-Way Interaction	3	167.55	55.85	2.24	0.121
Run Time*Current Density	1	108.12	108.12	4.34	0.053
Run Time*Electrode Gap	1	5.42	5.42	0.22	0.647
Current Density*Electrode Gap	1	54.01	54.01	2.17	0.159
Error	17	423.75	24.93		
Lack-of-Fit	9	423.75	47.08	*	*
Pure Error	8	0.00	0.00		
Total	25	5766.65			

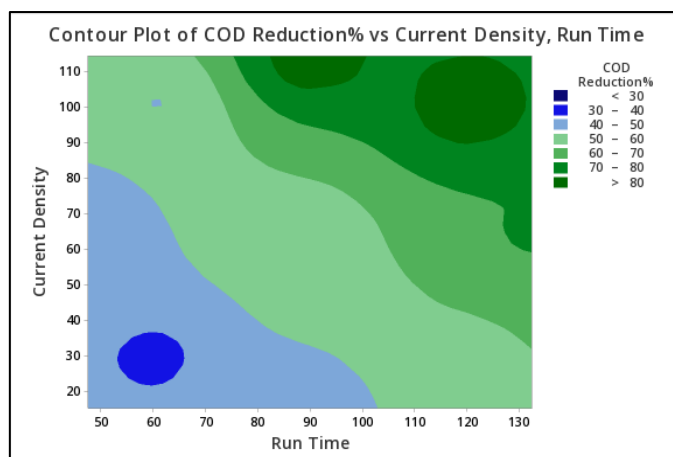
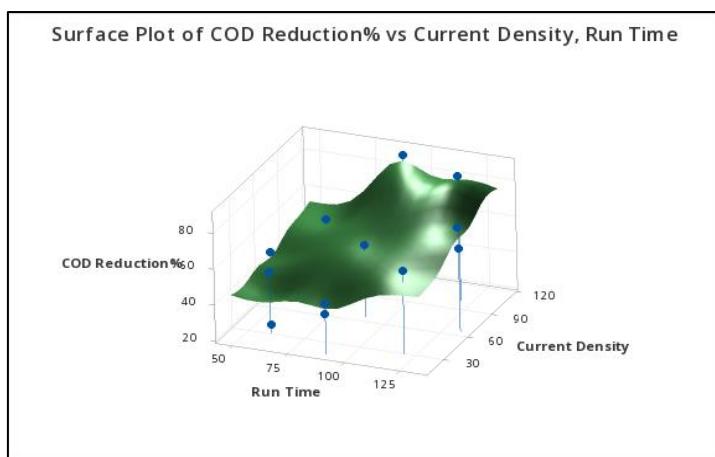
These models take into account quadratic effects, linear effects and two-way interactions between the studied factors. The empirical correlation represented in Equations. Involved factors that produces the actual values of these factors, adequacy of model and the quality of the model are evaluated by the correlation coefficient value  $R^2=86.43\%$ ,  $R^2$  adjusted= $84.58\%$ ,  $R^2$  predicted= $79.17\%$ . The  $R^2$  value always lies between 0 to 1. If the value is near to 1 it is considered as better fit

3.2 3D Surface Plot

The 3D-surface plot shown in Fig.3.1 gives the relationship between the Run time and current density settings used to determine the COD reductions. Current density at the shorter time intervals results in less COD reduction. However, current density at the longest intervals combined with the highest run time results in maximum COD reduction<sup>[59]</sup>. The peak on the plot corresponds with the highest COD Reduction % and occurs at approximately Time = 130 and current density =110mA/cm2

Figure 3.1 Surface plot of COD reduction % V/s current density, run time

Figure 3.2 Contour plot of COD reduction % V/s current density, run time



### 3.3 Contour Plot

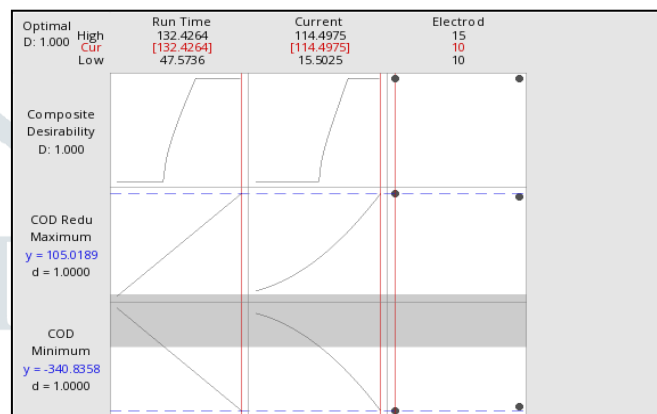
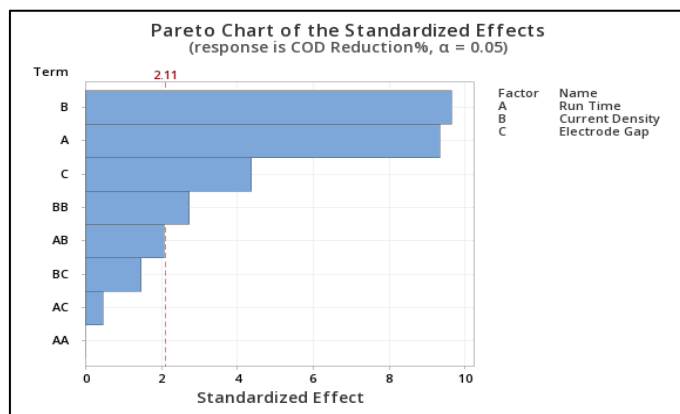
From Contour plot as shown in Fig.3.2, COD reduction can be easily interpreted that at low current density and with smaller run time, reduction of COD is very low. It can be clearly studied from the contour plot that more than 80% COD reduction is obtained somewhere at current density 100-110mA/cm<sup>2</sup> and 120-130 minutes.

### 3.4 Pareto Chart

From the largest to the smallest effect, the Pareto chart shown in Fig.3.3 depicts the absolute values of the standardized effects. A reference line is also plotted on the graph to show which effects are statistically significant<sup>[59]</sup>. Determine the magnitude and importance of the effects using the Pareto chart. Bars that cross the reference line on the Pareto chart are statistically significant. The bars that represent factors A, C, B, and BB cross the reference line that is at 2.11.

Figure 3.3 Pareto chart of standardized effects (response is COD Reduction)

Figure 3.4 Multipurpose optimization plot



### 3.5 Multi-response optimization plot for COD reduction

The multi-response optimization plot is shown in Fig.3.4. For the COD reduction data, the composite desirability is 1.000. The first column of the graph shows the response values at each level of Electrode gap, which is a categorical variable<sup>[59]</sup>. The current variable settings are Run time = 132.42, current density = 114.49. The goal was to Maximize COD reduction. Its predicted value is 105.0189, and its individual desirability is 1.00.

## IV. CONCLUSIONS

The performance of electrocoagulation process for the treatment of pharmaceutical wastewater to reduce COD was investigated. The electrocoagulation process was found to reduce chemical oxygen demand from the pharmaceutical wastewater. It was also found that the reduction efficiency of COD largely depends on the initial pH, conductivity, BOD, TDS, electrolysis time, current density, type of electrode material, electrode arrangement, interaction between electrode and effluent. The result shows that the above parameters are directly or indirectly related to COD reduction. In present study, the maximum 94% COD reduction was observed with Galvanized Iron electrode at CD=114.24 mA/cm<sup>2</sup>, pH=7.00, Run time=132 mins, electrode gap=10mm and conductivity=4.54 mS/cm. Treatment of Pharmaceutical wastewater was done with higher amount of metal cations species at higher current density, which contribute to higher reduction of COD. Thus fixing the metal ions chemically at in-situ and less operating cost compared to conventional treatment process confirmed that EC technique is inexpensive and viable tool for the treatment of pharmaceutical wastewater.

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