

Application of Electrocoagulation Process in Treatment of Tea Factory Effluent: A Case Study of Emrok Tea Factory Nandi County

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Abstract: Large quantities of tea wastes are usually discarded into the environment without any treatment. This discharge of untreated wastewater into the ecosystem has substantial impacts on the environment and human health. The study sought to establish, evaluate and analyse the effect of optimizing electrocoagulation operating variables on removal efficiency. An experimental design using response surface methodology of full factorial design with three factors at two levels was used to optimize and investigate the influence of current intensity, electrode distance and electrolysis time process variables on removal efficiency of colour and reduction of COD in tea effluent. Design Expert 9 statistical software was used to analyse the data and carry out optimization using response surface methodology. Analysis of variance (ANOVA) was used to check the adequacy of the mathematical models. The findings of the study was that at optimal conditions of colour and COD removal were 99.43 % and 98.62 % respectively at Electrolysis time of 18 minutes, Electrode distance of 6 mm and current intensity of 250 mA/cm². In conclusion EC provides the most feasible alternative for treatment of tea effluent with a view of COD reduction and colour removal.

Key Words: Effluent, Electrocoagulation

Date of Submission: 12-04-2018

Date of acceptance: 30-04-2018

I. INTRODUCTION

Globally, tea is the most widely consumed beverage after water (Williams, 2016). With great production and consumption, large quantities of tea wastes are usually released and discharged into the environment without any treatment (Nandal *et al.*, 2014). Like other biomass residues, tea waste is an unused resource and poses increasing disposal problems (Arvanitoyannis & Varzakas, 2008). According to Maghanga *et al.*, (2009); during the cleaning operations, flushing steps in operations and production processes, 18-20 m³ of water is used per cleaning operation releasing large amounts of tea factory effluents. These effluents are characterized by a strong colour and high turbidity due to the presence of large amounts of suspended solids, dissolved organic matter and inorganic wastes which include detergents, grease/waste oil from machine parts (Onchari, 2010). Inorganic pollutants such as phosphates and nitrates from effluents promotes eutrophication leading to depletion of oxygen in water thus a major threat to aquatic life (Price *et al.*, 1998). Colour pigments from the effluents reduce light penetration in aquatic environment significantly affecting photosynthetic activity hence affecting aquatic diversity and occasions adverse aesthetic effect (Kumar and Sahu, 2013). This has occasioned demands for efficient cleaning of industrial and domestic waste water to avoid environmental problems, and especially contamination of pure water resources which are of national and international concern (Khanittha & Wichan, 2009).

Electrocoagulation (EC) has been proposed as an effective method for treatment of many types of industrial effluents (Kumar *et al.*, 2004; Emamjomeh, *et al.*, 2006; Lai & Lin, 2006). EC is one of the effective techniques to remove colour, COD and organic compounds from wastewater (Patel *et al.*, 2010). An EC system may contain either one or multiple anode-cathode pairs and may be connected in either a monopolar or a bipolar mode (Emamjomeh and Sivakumar, 2009). The electrodes are usually made of aluminum, iron, or stainless steel (SS), because these metals are cheap, readily available, non-toxic with proven effectiveness.

EC process has been applied to treat various wastewaters such as electroplating waste water, paper mill bleaching waste water, chemical mechanical polishing waste water, textile waste water and olive oil waste water (Wasewar, 2010). The technology removes metals, colloids particles and soluble organic pollutants which are sources of high colour intensity and COD levels from aqueous media by introducing highly charged polymeric hydroxide species (Patel *et al.*, 2010). The treatment prompts the precipitation of certain metals, reduces the

amount of waste sludge and maximizes effluent through put rates, which need to be disposed (Ahmad, Mostafa, & Sara, 2013). EC as a waste water treatment technique is still an empirically optimized process that requires more fundamental knowledge to realize its full potential (Satish, 2013).

EC technique has been used in different parts of the World; for example in South America and Europe for treatment of industrial waste water (Joffe and knieper; 2000). In India it has been used for treatment of municipal waste water at optimal condition of 25 minutes electrolysis time, 10 V applied potential and 2cm inter electrode distance to remove 80.70% COD and 61.38% Total Solids (Alan and Paul, 2015). Electro coagulation is an evolving technology that is effectively being applied today for waste water treatment (Beagles, 2004), however, there is a paucity of scientific understanding of the complex chemical and physical process involved which is hindering progress (Mollah *et al.*, 2001 cited in Deokate, 2015). This inspired the need for this study in order to assess application of EC process in treatment of tea factory effluent.

1.1 Objective

To examine the optimal electrocoagulation operating variables in removal of colour and reduction of COD.

II. MATERIALS AND METHODS

Tea wastewater was sampled from the Ponds of Emrok tea industry in Nandi County. The sample and the blank were stored at 4°C prior to the experiments. The composition of the tea wastewater and blank were analyzed to determine the colour intensity and COD levels.

2.1 Reagents and materials

All reagents which were used in this study were of analytical grade (AR). Molar solutions were prepared at room temperature using deionised water so that it does not interfere with the specificity, accuracy, and precision of the procedure (National Institute of Health, 2013). They were 500 g platinum-cobalt, 1M Sodium hydroxide (NaOH), 1M Hydrogen chloride (HCl), Acetone and filter papers

2.2 Apparatus and Equipment

Iron sheets 15 cm × 3mm, DC power supply (12 V to 24 V), Digital Multimeter ET 580 and DT 9205A, 600 ohms rheostat R360, water bath MEMMAT W760, colorimeter spec DR 4000, Laboratory shaker MULTIFIX M80, 1000ml borosilicate beaker, COD adapters, cooling racks, Mn III COD vials ,Hanna PH 211, Hanna EC 215, Colorimeter DR/820.

2.3 Experimental Setup and Procedure

Measurements of pH, Conductivity, chemical oxygen demand (COD) and Colour (CR) were done in accordance with the standard methods adopted from American Society For Testing and Materials, (1995) and platinum-cobalt standard method (Clesceri & Eaton, 2005) respectively.

The EC experiment was performed according to Thirugnanasambandham *et al.* , (2014) with modification on working volume (1000 ml), type of electrode (Iron), electrode size and distance between the electrodes (3 mm to 10 mm). The EC setup that was used is as shown in fig 2.1.

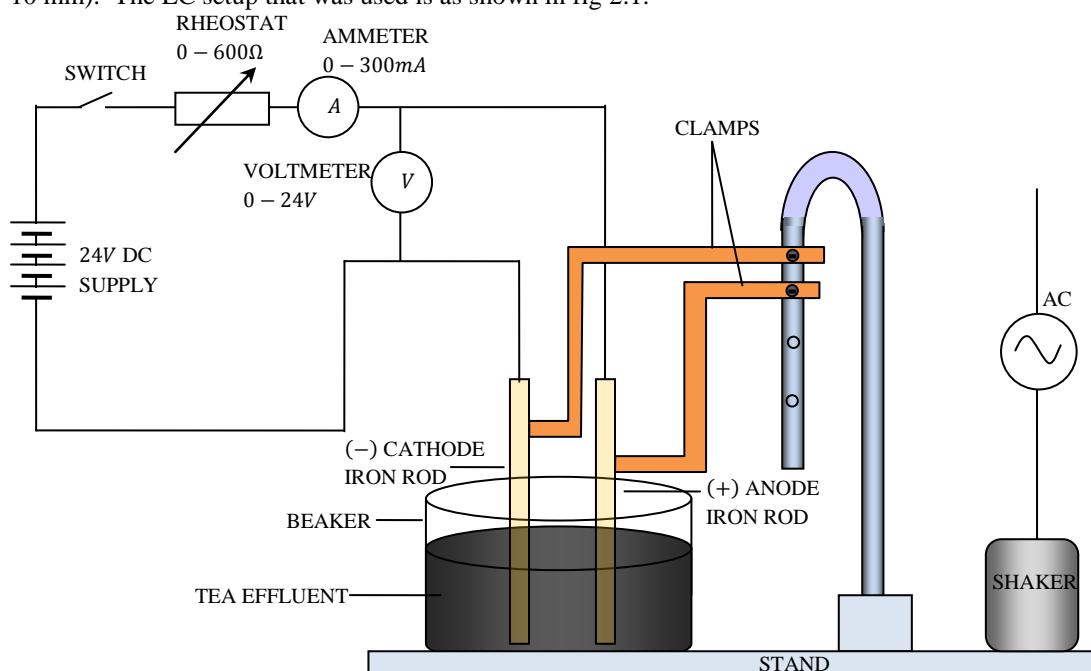


Fig 2.1: Schematic diagram of electrocoagulation (EC) process unit

2.4 Mathematical Model

The influences of all experimental factors and interactions effects on the responses were investigated. The general mathematical models for the full factorial design with three factors at two levels in coded values were as follows according to (Zivorad, 2004):

$$Y_1 = \beta_0 + \beta_1X_1 + \beta_2X_2 + \beta_3X_3 + \beta_{12}X_1X_2 + \beta_{13}X_1X_3 + \beta_{23}X_2X_3 + \beta_{123}X_1X_2X_3 + \epsilon \dots \dots \dots (2.4.1)$$

$$Y_2 = \beta_0 + \beta_1X_1 + \beta_2X_2 + \beta_3X_3 + \beta_{12}X_1X_2 + \beta_{13}X_1X_3 + \beta_{23}X_2X_3 + \beta_{123}X_1X_2X_3 + \epsilon \dots \dots \dots (2.4.2)$$

2.5 Removal efficiency of COD and colour.

According to Kumar, *et al.*, (2009) the colour and COD parameters removal efficiency was determined as follows:

$$\text{Removal Efficiency (\%)} = \frac{C_0 - C}{C_0} \times 100 \dots \dots \dots (2.5.1)$$

Where; C_0 : Initial concentration (mg/L)

C: Concentration at t (mg/L)

III. FINDINGS

3.1 Examination of optimal electrocoagulation operating variables in removal of colour and reduction of COD.

3.1.1 Predictive analysis

Before multiple regression was conducted the assumptions were tested .in this study the assumption of normality was tested. Normal probability plots indicate whether the residuals follow a normal distribution, in which case the points will follow a straight line (Antony, 2003) . The normal probability plot of the residuals of the COD and Colour removal is shown in Figure 3.1.1. The data points fairly close to the straight line indicating that the experiments came from a normally distributed population. The normal percentage probability plot of the residuals is an important diagnostic tool to detect and explain the systematic departures from the assumptions that errors are normally distributed and are independent of each other and that the error variances are homogeneous (Hsuan-Liang, *et al.*, 2004).The normal probability plot of the residuals indicates no violation of the assumptions underlying the analyses of the COD and Colour removal.

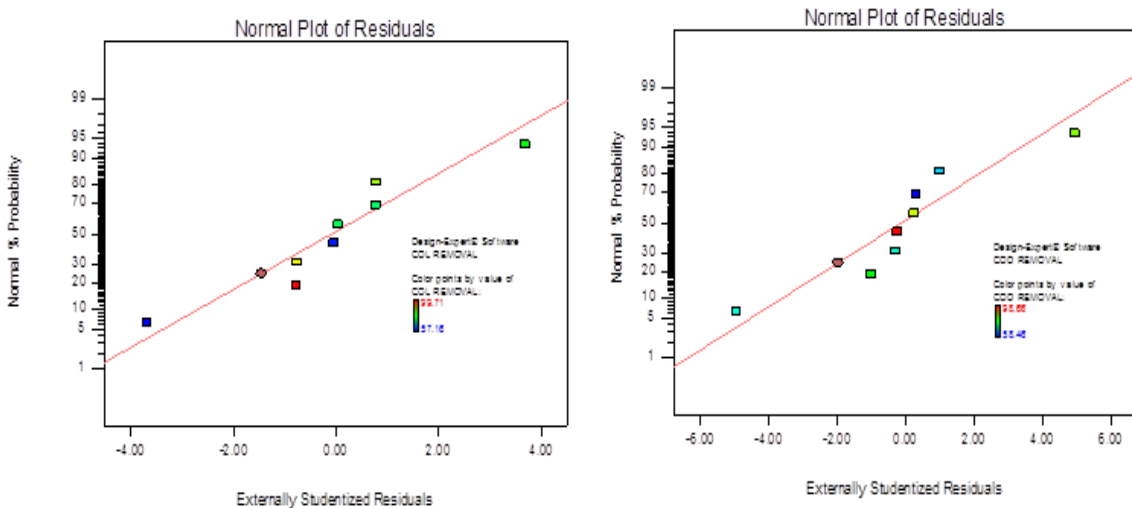


Fig 3. 1.1: Normal probability plot for colour and COD

For colour and COD removal experimental points are reasonably aligned suggesting a normal distribution

3.1.2 Experimental design matrix for colour and COD removal

In this study, three factors with two levels were used to evaluate the effect and optimize the process variables on the responses. A total number of 8 batch experiments were carried out in duplicate using statistically designed experiments. The percentage removal of colour and COD (Y) at different levels of electrolysis time, electrode distance and current intensity was calculated using equation 2.5.1 and presented in table 3.1.

Table 3.1: Experimental design matrix for colour and COD removal

RUN	ELECTROLYSIS TIME (MIN)	ELECTRODE DISTANCE (MM)	CURRENT INTENSITY mA/cm ²	RESPONSES COD REMOVAL (%)	RESPONSES COLOUR REMOVAL (%)
CODE	A	B	C	Y ₁	Y ₂
1	5	10	100	70.45	74.85
2	25	10	300	86.61	85.39
3	25	3	100	66.82	74.16
4	5	3	300	83.92	77.88
5	5	3	100	78.13	88.49
6	5	10	300	98.66	99.78
7	25	10	100	58.46	58.23
8	25	3	300	69.79	57.16

The highest response for Colour and COD removal was 99.78% and 98.66% respectively.

3.1.3 Optimal electrocoagulation operating variables in removal of colour and reduction of COD

Multiple regression was used to answer the research questions. The individual runs of experimental design were conducted and the responses were measured shown in Table 3.1. A linear regression model was fitted for the experimental data. The model coefficients and effects of the factors and interactions are shown in Table 3.2. The model summary was analyzed to establish the strength of conceptualized optimized variables in predicting COD and Colour removal. Results represented on table 3.2 reveal that the variables electrolysis time, electrode distance and current intensity contributes 99.5% (Adjusted R square = 0.9955) and 98 % (Adjusted R squared = 0.9785) to COD and Colour removal respectively. The "Pred R-Squared" of 0.9862 and 0.9343 for COD and colour removal were in reasonable agreement with the "Adj R-Squared" of 0.9955 and 0.9785; i.e. the difference is less than 0.2. "Adeq Precision" measures the signal to noise ratio. R² values are greater than 80%, suggesting the desirability of the models (Olmez, 2009). This indicates that less 20% of the total variation could not be explained by the empirical model. These values confirm that the equations of the models are reliable. This also indicates that the model terms are significant. The models are also reproducible (values of reproducibility close to 1). A ratio greater than 4 was desirable and therefore the ratio of 59.439 and 25.214 indicates an adequate signal.

3.1.3.1 Model summary

Table 3.2 : Model summary statistics tested for the responses

SD	MEAN	C.V.%	PRESS	R ²	Adjusted R ²	Pred R ²	Adeq Precision
Chemical Oxygen Demand							
0.86	76.61	1.13	15.89	0.9981	0.9955	0.9862	59.439
COLOUR							
2.13	76.98	2.77	97.01	0.9908	0.9785	0.9343	25.214

3.1.3.2 Analysis of variance (ANOVA) of COD and Colour

Analysis of variance (ANOVA) was also used to check the adequacy of the models in tables 3.2. The Model F-values were 385.89 and 80.48 for COD and colour removal respectively. Values of "Prob > F" less than 0.0500 indicate model terms in the model have a significant effect on response. In this case A, B, C, BC were significant model terms. These results indicate a good adequate agreement between experimental data and the data predicted by the developed models. This argument is corroborated by Thirugnanasambandham et al., (2013) who opines that adequately developed models shows low Coefficient of Variation, acceptable mean square, F-value and p-value for individual and interactive effects.

Table 3.2 : ANOVA Table for COD and Colour

SOURCE	Chemical Oxygen Demand				Colour			
	Df	Mean Square Value	F Values	P values	df	Mean Square Value	F Values	P values
Model	4	287.36	385.89	0.0002	4	365.98	80.48	0.0022
A-ELC	1	306.03	410.97	0.0003	1	544.34	119.70	0.0016
TIME								
B-ELEC	1	30.11	40.43	0.0079	1	52.48	11.54	0.0425

DIST								
C-CUR	1	530.08	711.83	0.0001	1	74.48	16.38	0.0272
INSTY								
BC	1	283.22	380.33	0.0003	1	792.62	174.30	0.0009
Residual	3	0.74			3	4.55		
Cor Total	7				7			

3.1.3.3 Regression coefficients for electrocoagulation variables

The results obtained from regression coefficients presented in tables 3.3 shows the estimates of β (coefficients) value and gives an individual contribution of each predictor to the model. β value shows the relationship between optimized variables and COD and colour removal with each optimization variable being a predictor of colour and COD removal. The coefficients for COD removal (Y_1) were electrolysis time -6.19, electrode Distance 1.94, Current intensity 8.14 and BC 5.95. The model can be specified as

$$Y_1 = 76.61 - 6.19A + 1.94B + 8.14C + 5.95BC + \epsilon \tag{3.1.1}$$

The coefficients for Colour removal (Y_2) were electrolysis time -8.25, electrode Distance 2.56, Current intensity 3.05 and BC 9.95. The positive values indicate that the direction of the relationship is positive while the negative value indicates that the direction of the relationship is negative. From the results in table 3.3.1 the model can be specified as

$$Y_2 = 76.98 - 8.25A + 2.56B + 3.05C + 9.95BC + \epsilon \tag{3.1.2}$$

The coefficients for each variable indicate the amount of change one could expect in removal of COD and Colour given a one unit change in the optimization variables. Therefore the model is chosen to describe the effects of operating variables on the EC process to treat waste water (Thirugnanasambandham, Sivakumar, & Prakash, 2013). In this study the VIF for all the optimization variables are within the acceptable range which is 1 hence there is no orthogonality of the design. 95% CI High and Low represents the range that the true coefficient should be found in 95% of the time if the range spans zero then the coefficient of zero could be true indicating that the factor has no effect. A Coefficient with a plus sign (+) means that the factor has a synergistic effect. By varying the factor the response increases and a minus sign (-) shows an antagonistic effect factor; this suggests that by varying the factor the response decreases (Kermet-Said & Moulai-Mostefa, 2015).

Table 3.3: COD coefficients

Factor	Coefficient		Standard Error	95% CI		VIF
	Estimate	df		Low	High	
Intercept	76.61	1	0.31	75.63	77.58	
A-ELC TIME	-6.19	1	0.31	-7.16	-5.21	1.00
B-ELEC DIST	1.94	1	0.31	0.97	2.91	1.00
C-CUR INSTY	8.14	1	0.31	7.17	9.11	1.00
BC	5.95	1	0.31	4.98	6.92	1.00

Table 3.3. 1: Colour coefficients

Factor	Coefficient		Standard Error	95% CI		VIF
	Estimate	df		Low	High	
Intercept	76.98	1	0.75	74.58	79.38	
A-ELC TIME	-8.25	1	0.75	-10.65	-5.85	1.00
B-ELEC DIST	2.56	1	0.75	0.16	4.96	1.00
C-CUR INSTY	3.05	1	0.75	0.65	5.45	1.00
BC	9.95	1	0.75	7.55	12.35	1.00

3.1.4 COD and Colour removal Pareto Plots

To determine whether calculated effects were significant, Student’s *t*-test was used. It was observed that for a 95% confidence level, the *t*-value was equal to 3.18245 for COD and Colour removal. Those evaluations are illustrated by means of Pareto charts in figure 3.2. The horizontal line indicates minimum statistically significant effect for a 95% confidence level. Effects above *t*-value 3.18245 limit are significant terms in the model. The main factors of Electrolysis time (A), Electrode distance (B), Current intensity (C) and interactions such as BC significantly influenced the COD and colour removal. ABC and AB, AC interactions were found to be of no importance in the COD and colour removal. Any factor or interaction of factors above Bonferroni limit 6.57968 had a more effect on COD and Colour removal. These included Current intensity (C), Electrolysis time (A), and interaction of Electrode distance and Current intensity (BC). These included Current intensity (C), Electrolysis time (A), and interaction of Electrode distance and Current intensity (BC).

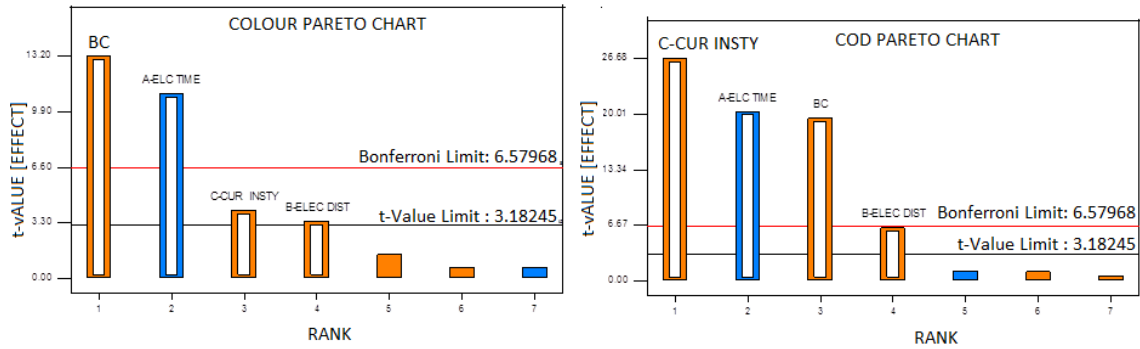


Fig 3. 2: COD and Colour removal Pareto Plots

3.1.5 Main effects

3.1.5.1 Electrolysis Time

Figure 3.3 a and b shows that there is an inverse relationship between electrolysis time on colour and COD removal. The electrolysis time effected colour and COD removal significantly therefore colour and COD removal are a function of electrolysis time. The decline in colour and COD removal with increase in time depends directly on the concentration of ions produced by the electrodes. As electrolysis time increases, an increase occurs in the amount of metal hydroxide flocs ($M(OH)_3$) which promotes the removal of COD and colour via a sweep coagulation followed by precipitation mechanism, thus removal efficiency of colour and COD increases but after 5 minutes concentration of iron ions decreases because of degradation of iron electrodes (Thirugnanasambandham et al, 2014). Hence efficiency of colour and COD removal decreases with electrolysis time (Patel et al., 2010).

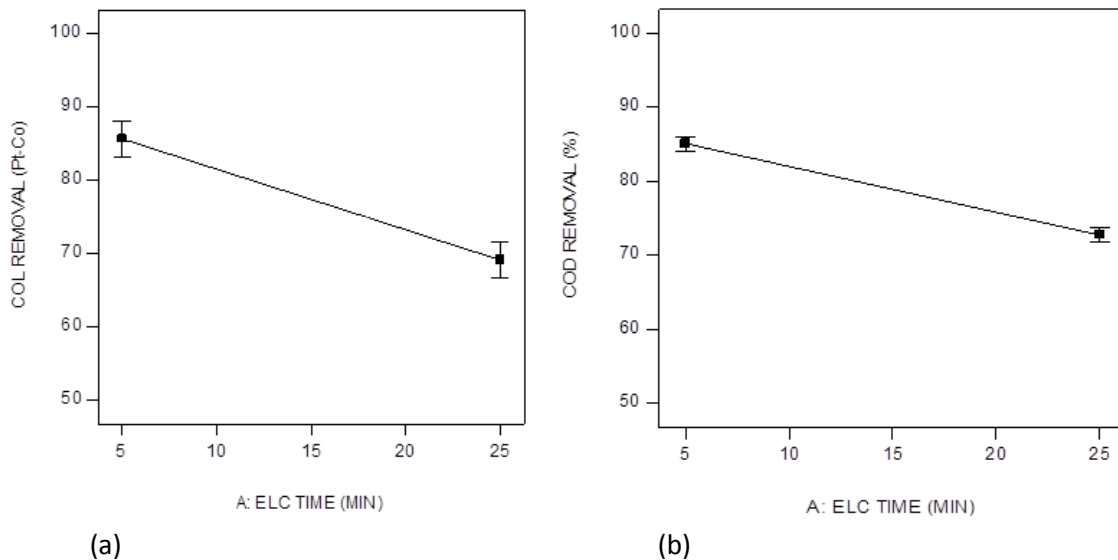


Fig 3. 3: (a), (b) Effect of electrolysis time on colour and COD removal

3.1.5.2 Electrode distance

Figure 3.4 a and b below shows that there is a direct positive relationship between electrode distance on colour and COD removal. It is observed that, removal efficiency of COD and colour increased with the increasing electrode distance. This is in line with the findings of Thirugnanasambandham et al., (2013) who found that the removal efficiency of COD and colour increased with increasing electrode distance up to 5 cm. The resistance for current flow in the reactor is lower at low inters electrode distance which facilitates the EC process for enhanced removal of colour and COD.

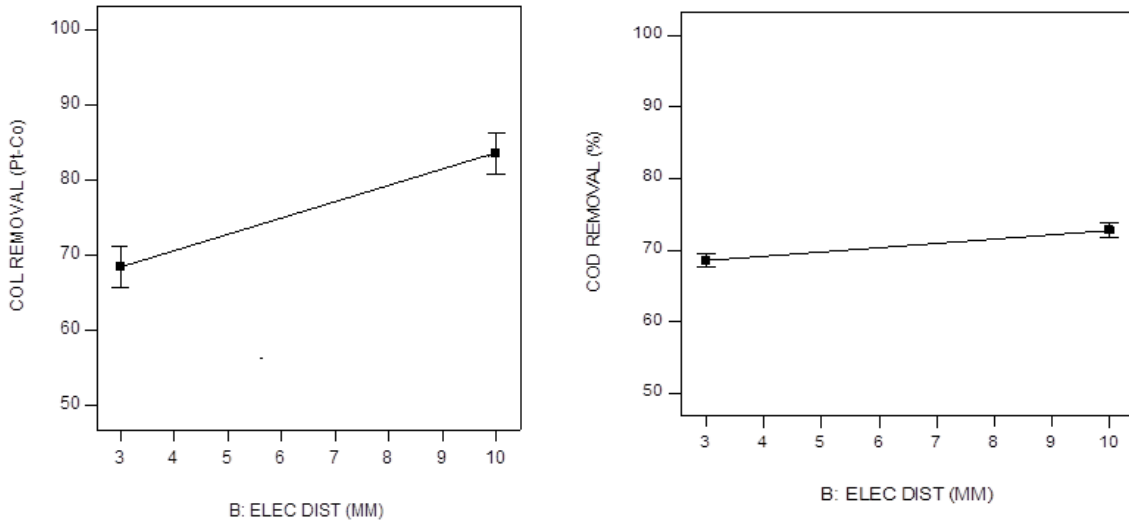


Fig 3. 4: Effect of Electrode distance on colour and COD removal

3.1.5.2 Current intensity

Figure 3.5 a and b below shows that there is a direct positive relationship between current intensity on colour and COD removal. This explained by the fact that the amount of current intensity determines the coagulant production rate, and adjusts the rate and size of the bubble production, and hence affects the growth of flocs which promotes removal of colour and COD. Raising current intensity causes a corresponding increase in the oxidized ion production from electrodes hence an increase in current intensity is favorable for Iron electrodes in removal of colour and COD (Patel et al., 2010).

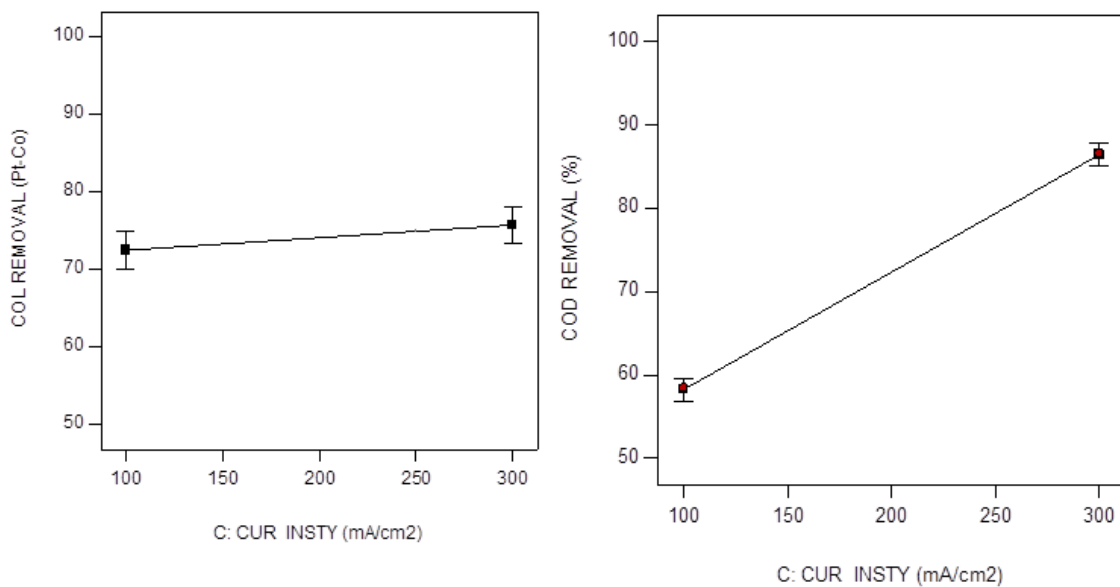


Fig 3. 5: Effect of current intensity on colour and COD removal

3.1.5.3 Interaction of effects

Figure 3.6 a,b,c,d shows the interactions of current intensity and electrode distance. This interaction influenced the removal of colour and COD significantly can be observed from the Pareto chart figure 3.2. From the Table 3.3 and 3.3.1 the interaction of Electrode distance and Current intensity (BC) is seen to be significant in the removal of Colour and COD. The curved nature of contour plots as shown figure 3.6 confirms this interaction.

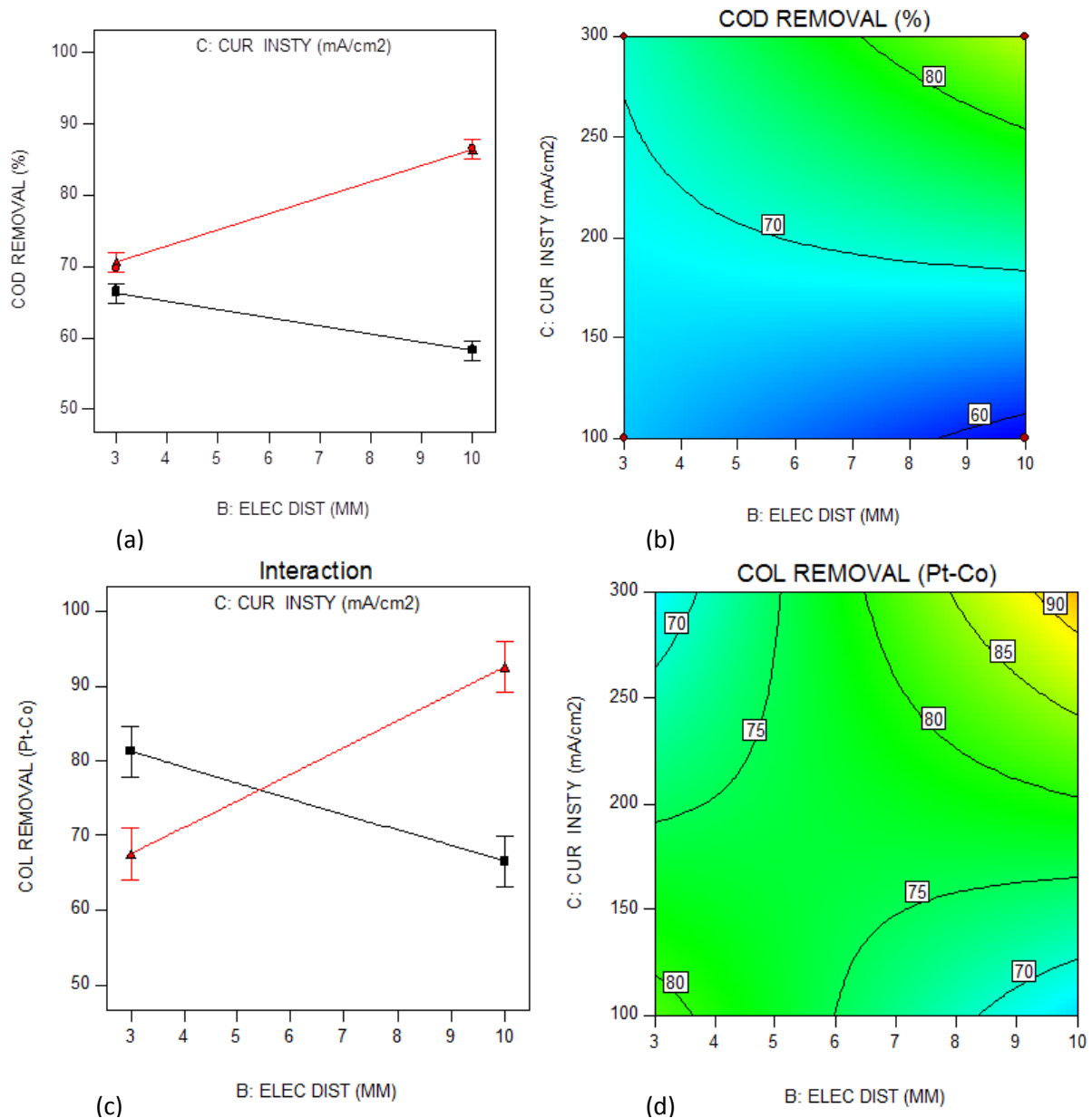


Fig 3. 6: (a), (b), (c), (d) Interaction of BC variable in COD and Colour removal

3.1.6 Optimization of COD and Colour removal

Cube plots display the average response values at all combinations of design parameter settings (Antony, 2003). Figures 3.8a and b illustrates the cube plot for COD and Colour removal average response values at all combinations for optimization study with three parameters; Electrolysis time (A), Electrode distance (B) and Current intensity (C). The optimization of the COD and Colour removal was performed by using a multiple response method called desirability (D) function. The main factors of Electrolysis time (A), Electrode distance (B) and Current intensity (C) were optimized by targeting maximum removal (D=1) of COD and Colour. The Current intensity was set to be minimised, Electrolysis time and Electrode distance were set to be within the studied range, Electrolysis time and Electrode distance were targeted for the optimum within the range. This was done in consideration of cost in EC process and given that the holding time for tea effluent during treatment is long enough to give the allowance of the studied range. The 3D surface plot of COD and Colour removal is shown in Figure 3.7 a and b. The optimisation process desirability, D=0.864 for COD and D=0.999 for Colour removal respectively which was close to the targeted value of 1. The models predicted (98.62) % COD and (99.43) % Colour removal at Electrolysis time of (18) Electrode distance of (6) and Current intensity of (250).

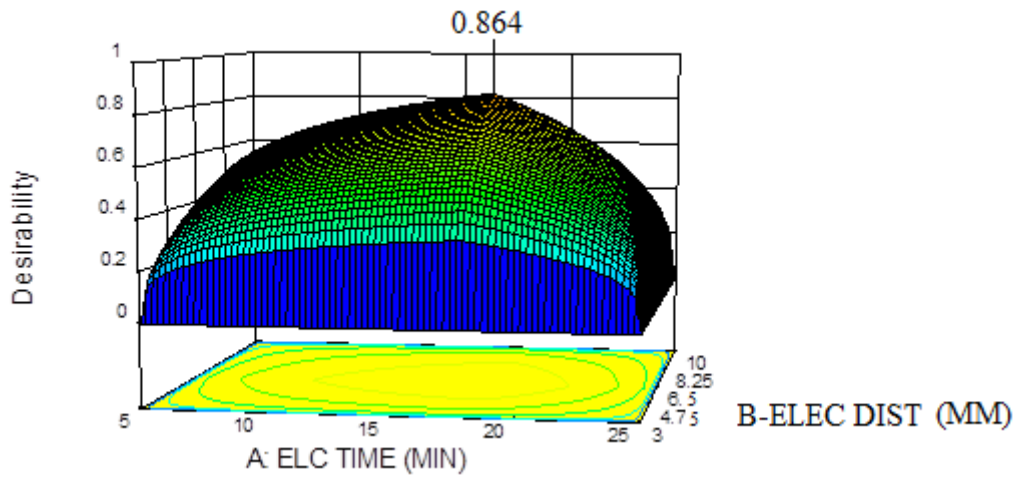


Fig 3.7 a: 3D- Surface plot for COD removal

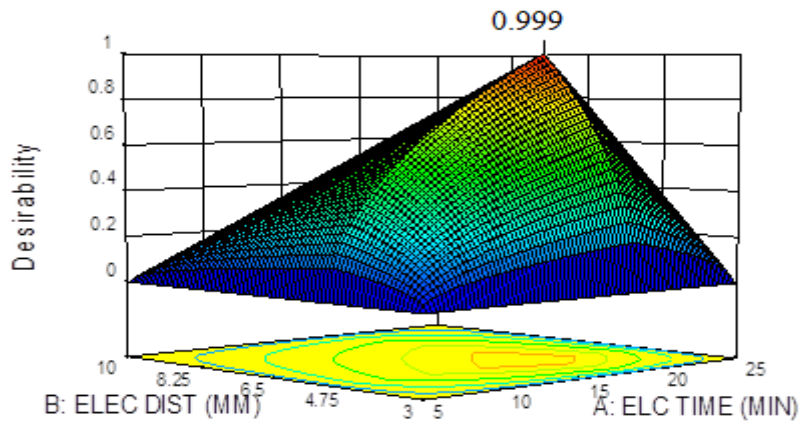


Fig 3.7 b: 3D- Surface plot for colour removal

Design-Expert® Software
 Factor Coding: Actual
 COD REMOVAL (mg/L)
 X1 = A: ELC TIME
 X2 = B: ELEC DIST
 X3 = C: CUR INSTY

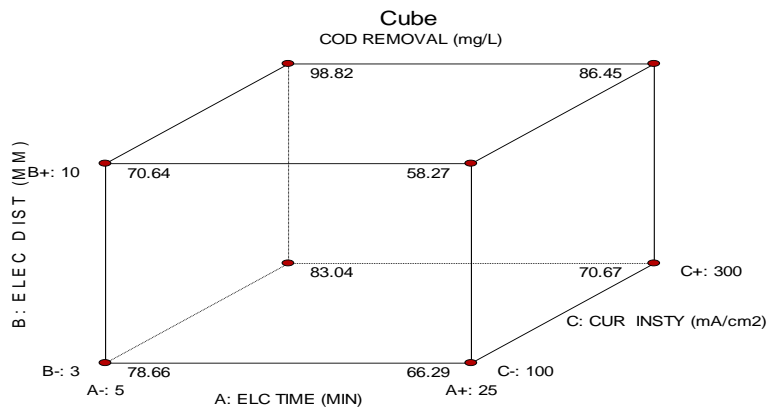


Fig 3.8 a COD Box-Coxes Plot For Power Transformations

Design-Expert® Software
 Factor Coding: Actual
 COL REMOVAL (Pt-Co)
 X1 = B: ELEC DIST
 X2 = C: CUR INSTY
 X3 = A: ELC TIME

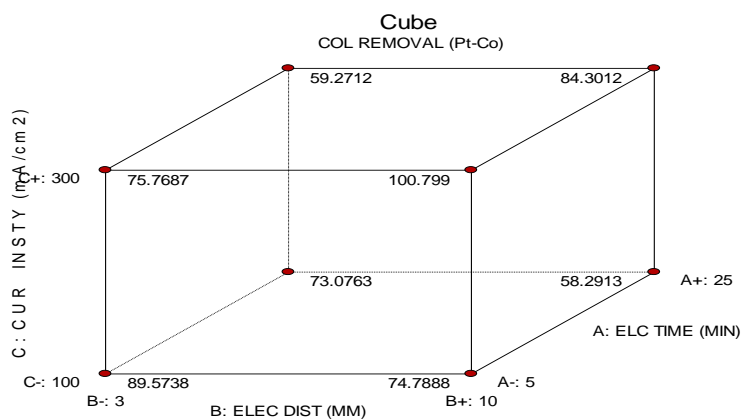


Fig 3.8 b Colour Box-Coxes Plot For Power Transformations

IV. CONCLUSION

Response surface methodology of full factorial designs, it was found from the surface plots that, the optimal conditions of colour and COD removal were 99.43 % and 98.62 % respectively at electrolysis time of 18 minutes, electrode distance of 6 mm and Current intensity of 250 mA/cm². The experimental values obtained at these conditions were 99.87 % colour removal and 98.39 % COD removal thus incongruent with the predicted model at 95% confidence level. This percentage removal reduced the colour intensity and COD to below the National Environment Management Authority maximum allowable values. Therefore RSM was a suitable method to optimize the operating conditions and maximize COD reduction and color removal.

From the findings this study has contributed to the gap in EC literature and the optimal application of EC in tea waste treatment process which has suffered a deficiency in research. The study also contributes to continued research in other electrode systems by recommending further studies on their use in treating tea effluent in an industrial scale. Further studies should be conducted on optimization of EC integrated with ponds in tea effluent treatment strategies for efficient removal of colour and reduction of COD.

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Makau Francis Kalulu "Application of Electrocoagulation Process in Treatment of Tea Factory Effluent: A Case Study of Emrok Tea Factory Nandi County"." *IOSR Journal of Engineering (IOSRJEN)*, vol. 08, no. 4, 2018, pp. 28-38.