

Eco-Friendly Electrocoagulation of Crude Oil-Contaminated Water Using Recycled Aluminium Electrodes and Coconut Shell Ash derived Electrolytes

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Abstract. The treatment of crude oil contaminated water using Electrocoagulation (EC) and Coconut Shell Ash (CSA) derived supporting electrolyte was investigated. Aluminium electrodes produced from recycled beverage cans were used in the setup. Nine experiments were designed using the Taguchi's L9 orthogonal array. The operating voltage (10 – 30V), electrolyte concentration (5 - 15 g/l), and treatment time (10 – 30 mins) were chosen as controllable factors, and their impact on Chemical Oxygen Demand (COD), turbidity, Oil & Grease and electrode consumption were studied. The Maximum COD, turbidity, oil and grease removal of 65.47% , 100%, and 93.26% were obtained at 20, 30 and 10V, respectively. Increased electrode consumption was evident in the experiments without the CSA derived electrolyte. This research successfully demonstrated the viability of EC for remediating crude oil-contaminated water.

1 Introduction

Crude oil spills are a persistent environmental concern, causing severe ecological and economic damage [1]. The Niger Delta region of Nigeria is particularly vulnerable, experiencing frequent oil spills that contaminate vital water resources for local communities [1, 2]. Remediation techniques are crucial for restoring the quality of contaminated water and safeguarding public health. Electrocoagulation (EC) has emerged as a promising method for treating wastewater from mining [3-4], dairy processes [5, 6], textile industry [7], crude oil-contaminated water [8-11] and more. An EC system utilizes an electric current to generate metal cations from sacrificial electrodes. These cations destabilize oil emulsions, promoting flocculation and separation from the water.

The core of an EC system lies in the electrochemical cell which is shown in Fig. 1. This cell comprises two electrodes, an anode and a cathode, submerged in a conductive solution. During operation, the anode undergoes a sacrificial oxidation process, releasing metal cations (often iron or aluminum) into the solution [12]. These metal cations play a crucial role in destabilising the targeted pollutants and particulates within the water and in forming flocs.

Essentially EC can be summarized as three-stage process, encompassing anode oxidation, pollutant destabilization, and floc formation, underpins the effectiveness of this method. The equations that show how these reactions proceed is highlighted in Equation 1 - 3.

At the Anode: $Al \rightarrow Al^{3+} + 3e^{-}$ (1)

$Al^{3+} + 3OH^{-} \rightarrow Al(OH)_3$ (2)

At the Cathode; $2H_2O + 2e^{-} \rightarrow H_2 + 2OH^{-}$ (3)

In recent studies supporting electrolytes have been used to improve the outcomes of the electrocoagulation process [14, 15]. Not much has been reported on their use for oil contaminated wastewater. This research investigated the use of recycled aluminium anodes and Coconut Shell Ash derived electrolyte in the electrocoagulation process.

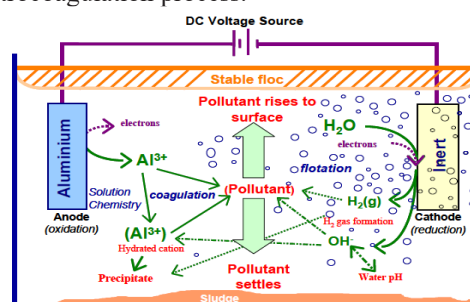


Fig. 1. An Electrochemical Cell

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2 Methodology

2.1 Materials selection

2.1.1 Crude Oil-Contaminated Water Samples

The crude oil-contaminated river water sample was collected from a riverine community in Niger delta area of Nigeria. The Collected Sample was tested for COD, oil & grease, and turbidity using standard testing procedures[13] and the values presented in Table 1.

Table 1. Wastewater characterization

Parameters	COD (mg/l)	Oil and Grease(g/l)	Turbidity(NTU)	pH
Values	1723.80	1.93	47.73	7.22
Method	APHA 5220-C	APHA 5520-B	Spectro photometric	APHA 4500-H

2.1.2 Electrode Materials

Waste beverage cans (shown in Fig. 2) sourced from a scrap dump were used to produce the aluminum electrodes. Sand casting was used to produce the cylindrical aluminium electrodes after appropriate charge calculations and melting in a furnace. The electrodes produced were cast into a cylindrical shape 2 cm in diameter and 10 cm in height as shown in Fig. 2. Typically these Beverage cans contain 92.5% to 97% aluminium, 5.5% magnesium, 1.6% Manganese, 0.15% chromium and some trace amounts of iron, silicon and copper [12].



Fig. 2. (a) Waste Beverage Cans. (b) Fabricated aluminium anodes

2.1.3 Coconut Shell Ash

Coconut Shells, obtained from Oje market, Ibadan, Nigeria were first subjected to open-air burning followed by calcination at 600°C using a gas-fired furnace. This controlled heating process eliminated any remaining volatile substances, promoting the transformation of the partially burnt shells into coconut shell ash. The major active constituents were determined by XRF Analysis and presented in Table 2.

2.1.4 Electrocoagulation study planned using Taguchi design

Electrocoagulation experiments were performed in a batch-type electrochemical cell of 400 mL, with immersed electrodes and leached by adding the appropriate amount of CSA. The experiments were planned according to Taguchi's L9 orthogonal array

design. A total of 9 experiments were performed to investigate the influence of voltage (10 – 30V), electrolyte concentration (5 - 15 g/l), and treatment time (10 – 30 mins). The distance between the electrodes was kept 1 cm, and the stirring speed was kept at 350 rpm. Turbidity, Oil and Grease, and COD were determined before and after the EC experiments. The amount of electrode dissolution during the treatment was also determined. Fig. 3 gives an overview of the EC setup during the 9 experiments.

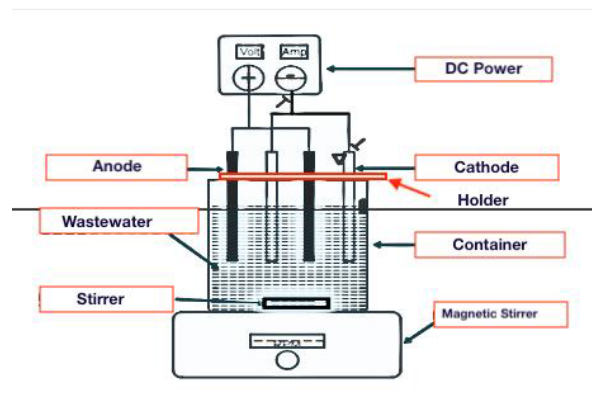


Fig. 3. Electrocoagulation Setup

2.2 Electrode consumption and surface change

Before and after immersion, the electrodes were weighed on an analytical balance, and the electrode consumption was determined.

2.3 Turbidity, COD, Oil & Grease Removal Efficiency

The efficiency of the electrocoagulation process was assessed using Equation (4).

$$\%E = \frac{E_i - E_f}{E_i} \times 100 \quad (4)$$

where E_i and E_f are the initial and final values of the responses for pollutants (Turbidity, COD, and Oil & Grease reduction). A control experiment where no CSA-derived electrolytes was used was also conducted. Table 3 gives an overview of the experimental conditions during the 9 designed experiments and the control experiment.

Taguchi method was used to analyse the result statistically, evaluating the S/N ratios of each factor on response and different graphs like the main effects plot, several factors and response plots.

Table 3. Experimental conditions of experiments planned using Taguchi L9 Orthogonal array

Experiment No.	Voltage (V)	Electrolyte Concentration (g/l)	Treatment time (Mins)
1	10	5	15
2	10	10	20
3	10	15	30
4	20	10	15
5	20	15	20
6	20	5	30
7	30	15	15

8	30	5	20
9	30	10	30
Control	30	0	30

The experimental factors and levels are outlined in Table 4. The commonly utilized Signal-to-Noise (S/N) ratio functions include "smaller is better," "larger is better," and "nominal is better." Typically, a stronger signal is achieved when the noise is minimal, hence a higher S/N ratio leads to improved final results for the study. "Larger is better" is used when the process outcome is to be maximized while "Smaller is better" minimizes the process outcome. The criteria for selection was based on the goals of optimization which is the removal of COD, oil and grease, and turbidity. Higher removal efficiencies of COD, oil and grease, and turbidity are the desired performances for the treatment process therefore, "Larger is better" was used to analyse and optimize the study.

Larger is better:

$$S/N = -10 \log \frac{\sum_{i=1}^n \frac{t}{y_i^2}}{n} \quad (5)$$

Where n is the number of runs for an experimental combination and Y_i is the performance value of the i th experiment.

Table 4. Factors and levels in coded and uncoded matrix

Levels	Voltage (V)	Treatment Time (Mins)	Electrolyte Concentration (g/l)
1	10	15	5
2	20	20	10
3	30	30	15

3 Results and Discussion

The results of laboratory and statistical analysis of the treated crude oil-contaminated water sample are presented and discussed below. The results of the nine experiments using Taguchi Design of Experiments are presented in Table. 5 For the nine experiments between 10 and 30V, the range of COD, turbidity, oil & grease were (595.24 - 1011.24) g/l, (0 - 3.56) NTU and (0.13 - 0.79) g/l respectively. Maximum COD removal of 65.47% was obtained and corresponds to experiment 6 (20V, 5g/l and 30 mins).

Maximum turbidity removal of 100% was achieved for experiment 7 (30V, 15g/l, 15 mins) and control (no ash). Maximum oil & grease removal of 93.26% was obtained at experiment 2 (10V, 10g/l, 20 mins). The addition of CSA electrolytes provided higher removal percentages compared to the control EC process.

3.1 Analysis of Response tables for Signal-to-Noise ratios obtained from Taguchi

Statistical analysis of the results was performed used Taguchi DoE and they are presented in Table 6, 7 and 8 and visualized in Fig. 4, 5 and 6. From the tables, the

larger value is considered better. The levels are presented in coded matrix 1, 2 and 3 and the corresponding S/N ratios are provided for each level. The delta values in the tables show the difference in S/N ratios between adjacent levels of the factors (Voltage, Electrolyte Concentration, and Treatment time).

3.2 COD Removal Efficiency

The response for S/N ratios provides insights into the effect of different levels of voltage, treatment time and Electrolyte concentration on COD (Chemical Oxygen Demand) removal efficiency. Larger S/N ratios indicate better COD removal efficiency.

From Table 6, we note that Voltage was ranked 1, indicating that it had the most significant effect on COD removal efficiency. The S/N ratio at level 2 of voltage was 35.21, which was the highest among the three voltage levels. Treatment Time was ranked 2, with a lower S/N ratio compared to voltage. The S/N ratio at level 2 of Treatment time was 35.36. Electrolyte Concentration was ranked 3, with the lowest S/N ratio among the three factors. The S/N ratio at level 1 of treatment time was 35.19. The delta values provide a measure of the difference between the S/N ratios of each level and the best level. A larger delta value indicates a more significant effect on the response. In this case, the delta value for voltage was 1.25, suggesting that it had the most substantial impact on COD removal efficiency. The delta value for Treatment time was 1.20, indicating a lower but still significant effect. Electrolyte concentration had a delta value of 0.9, implying that it had a smaller influence on COD removal efficiency compared to Voltage and Treatment time.

The normal probability plot (a) revealed that the residuals generally align with the reference line, suggesting approximate normality with slight deviations at both tails. The residual versus fitted values plot (b) showed a random scatter around zero with no clear pattern, but the spread is relatively wide, with some large positive and negative values. This trend is further confirmed by the histogram (c), which shows a roughly symmetric but dispersed residual distribution that is centred around zero.

3.3 Turbidity Removal Efficiency

Table 7 provides insights into the effect of treatment time on turbidity removal efficiency for different levels of voltage, Electrolyte concentration. In this table, larger S/N ratios indicate better turbidity removal. From the results Electrolyte concentration was ranked 1, indicating that it had the most significant effect on turbidity The residual plots in Fig.7 collectively indicate that the Taguchi model for turbidity removal is reasonably adequate with minor deviations. The normal plot (a) showed that the residuals lie approximately along the straight reference line, indicating close normal distribution of the error terms. The residuals versus fitted values plot (b) showed a random scatter of points around zero without any discernible pattern or funnel shape, indicating that there is no bias or misspecification in the model. This is supported by the histogram (c), which showed that the residuals were roughly

symmetrically distributed about zero despite the small sample size.

Treatment time, with the lowest S/N ratio among the three factors, was ranked 3. The delta values provide a measure of the difference between the S/N ratios of each level and the best level. A larger delta value indicates a more significant effect on the response. In this case, the delta value for Electrolyte concentration was 0.31, suggesting that it had the most substantial impact on turbidity removal efficiency. The delta value for Voltage was 0.17, indicating a lower but still significant effect. Treatment time had a delta value of 0.17, implying a similar influence on turbidity removal efficiency compared to voltage. The S/N ratio at level 3 of Electrolyte concentration was 39.83, which was the highest among the three Electrolyte concentration levels. Voltage was ranked 2, with a lower S/N ratio compared to Electrolyte concentration. The S/N ratio at level 1 of voltage was 39.75.

3.4 Oil and Grease removal Efficiency

Table 8 provides insights into the effect of different levels of voltage, Electrolyte concentration, and treatment time on Oil and Grease removal efficiency. In this table, larger S/N ratios indicate better Oil and Grease removal.

It was observed that Electrolyte concentration was ranked 1, indicating that it had the most significant effect on turbidity removal efficiency. The S/N ratio at level 3 of Electrolyte concentration was 38.38, which was the highest among the three Electrolyte concentration levels. Voltage was ranked 2, with a lower S/N ratio compared to Electrolyte concentration. The S/N ratio at level 1 of voltage was 38.49. Treatment time, with the lowest S/N ratio among the three factors, was ranked 3. The S/N ratio at level 1 of treatment time was 37.65.

The delta value for Electrolyte concentration was 2.12, suggesting that it had the most substantial impact on Oil and Grease removal efficiency. The delta value for Voltage was 1.69, indicating a lower but still significant effect. Treatment time had a delta value of 0.14, indicating a relatively smaller influence on Oil and Grease removal efficiency than voltage.

The residual diagnostics for oil and grease removal are presented in Fig. 9. It shows that the Taguchi model is generally acceptable. The normal probability plot (a) indicates that most of the residuals follow the straight reference line with a few deviated points at the tails. The residuals versus fitted values plot (b) indicates a largely random dispersion around zero, which supports the assumption of constant variance, while the histogram (c) presents a roughly symmetric distribution about zero, further confirming the assumption of normality. The residuals versus order plot

Level	Voltage	Treatment Time	Electrolyte Concentration
1	34.71	34.13	35.19
2	35.21	35.36	34.29
3	33.99	34.16	34.45
Delta	1.25	1.20	0.90
Rank	1	2	3

Table 7. Response Table for Signal-to-Noise Ratios for Turbidity removal efficiency

Level	Voltage	Treatment Time	Electrolyte Concentration
1	39.75	39.75	39.53
2	39.58	39.58	39.67
3	39.70	39.71	39.83
Delta	0.17	0.17	0.31
Rank	2	3	1

Table 8. Response Table for Signal-to-Noise Ratios for Oil and Grease removal efficiency

Level	Voltage	Treatment Time	Electrolyte Concentration
1	38.49	37.65	38.10
2	36.80	37.51	36.27
3	37.44	37.54	38.38
Delta	1.69	0.14	2.12
Rank	2	3	1

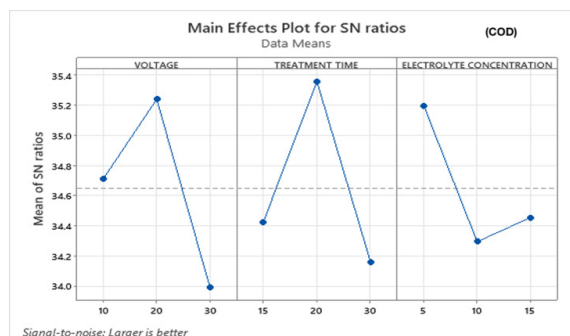


Fig. 4. Main effects plot for Signal-to-Noise ratios for COD

Table 6. Response Table for Signal-to-Noise Ratios for COD removal efficiency

Table 2. Composition of Coconut Shell Ash

Element	CaO	SiO ₂	Fe ₂ O ₃	Al ₂ O ₃	LOI	MgO	Na ₂ O	SO ₃	TiO ₂	P ₂ O ₃	K ₂ O	MnO
%	60.5	25.48	7.15	5.2	0.65	0.42	0.3	0.11	0.1	0.08	0.05	0.04

Table 5. EC results

Experiment No	Voltage (V)	Electrolytes Concentration (g/l)	Time (mins)	Initial COD (mg/l)	Final COD (mg/l)	COD Removal (%)	Initial Turbidity (NTU)	Final Turbidity (NTU)	Turbidity Removal (%)	Initial Oil & Grease (g/l)	Final Oil & Grease (g/l)	Oil & Grease Removal (%)
1	10	5	15	1723.81	838.1±41.9	51.38±2.6	47.73	1.53±0.08	96.79±4.84	1.93	0.42±0.02	78.24±3.91
2	10	10	20	1723.81	624.79±31.2	63.76±3.2	47.73	1.49±0.07	96.88±4.84	1.93	0.13±0.01	93.26±4.66
3	10	15	30	1723.81	876.19±43.8	49.17±2.5	47.73	1.04±0.05	97.82±4.89	1.93	0.36±0.018	81.35±4.07
4	20	10	15	1723.81	812.34±40.6	52.88±2.6	47.73	2.55±0.13	94.66±4.73	1.93	0.51±0.03	73.58±3.68
5	20	15	20	1723.81	761.9±38.1	55.8±2.8	47.73	1.66±0.08	96.52±4.83	1.93	0.46±0.023	76.17±3.81
6	20	5	30	1723.81	595.24±29.8	65.47±3.3	47.73	2.49±0.12	94.78±4.74	1.93	0.79±0.039	59.07±2.95
7	30	15	15	1723.81	798.56±39.5	53.67±2.7	47.73	0	100±4.88	1.93	0.44±0.022	77.2±3.86
8	30	5	20	1723.81	749.09±37.5	56.54±2.8	47.73	3.56±0.19	92.54±4.63	1.93	0.78±0.039	59.59±2.98
9	30	10	30	1723.81	1011.24±50.6	41.34±2.1	47.73	1.23±0.06	97.42±4.87	1.93	0.32±0.016	83.42±4.17
Control Experiment (No Ash)	30	0	30	1723.81	800±40	53.59	47.73	0	100±5.1	1.93	0.38±0.019	80.31±4.02

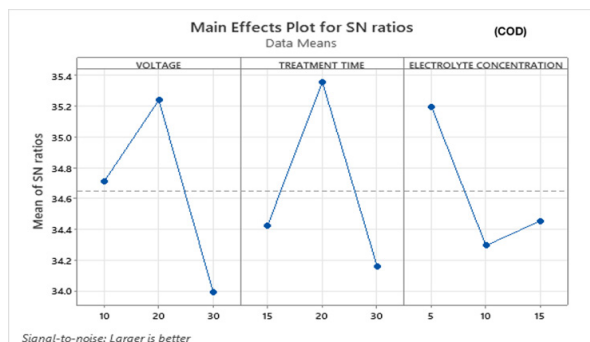


Fig. 6. Main effects plot for Signal-to-Noise ratios for Turbidity

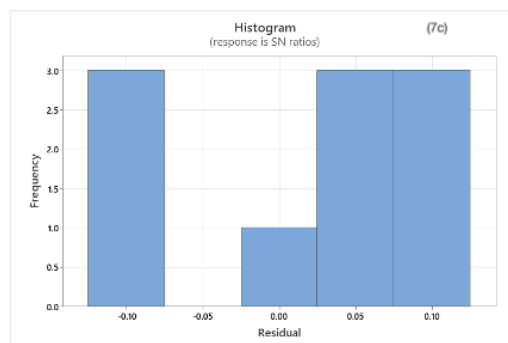
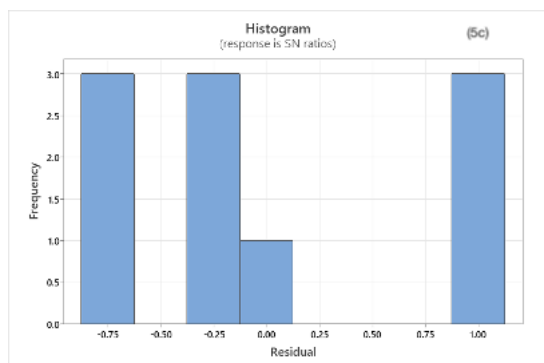
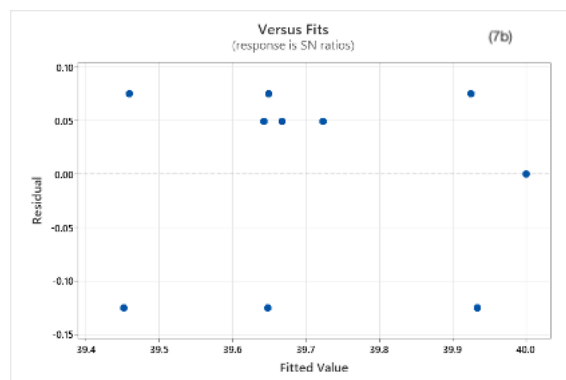
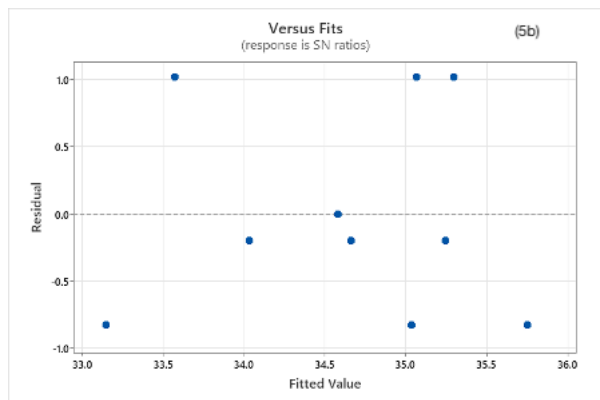
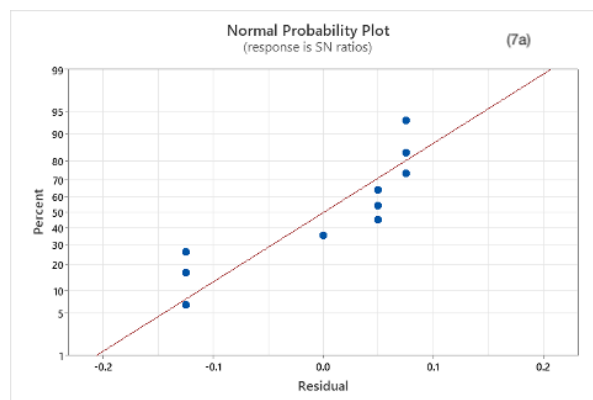
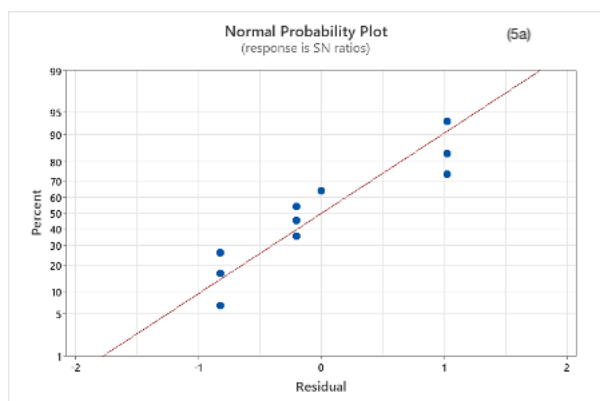


Fig. 5. Residual plots for COD removal (a) Normal probability plot (b) Versus fits (c) Histogram

Fig. 7. Residual plots for turbidity removal (a) Normal probability plot (b) Versus fits (c) Histogram order

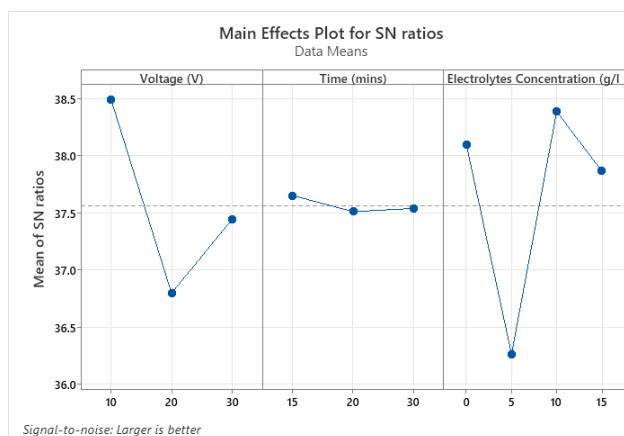


Fig. 8. Main effects plot for Signal-to-Noise ratios for Oil & Grease

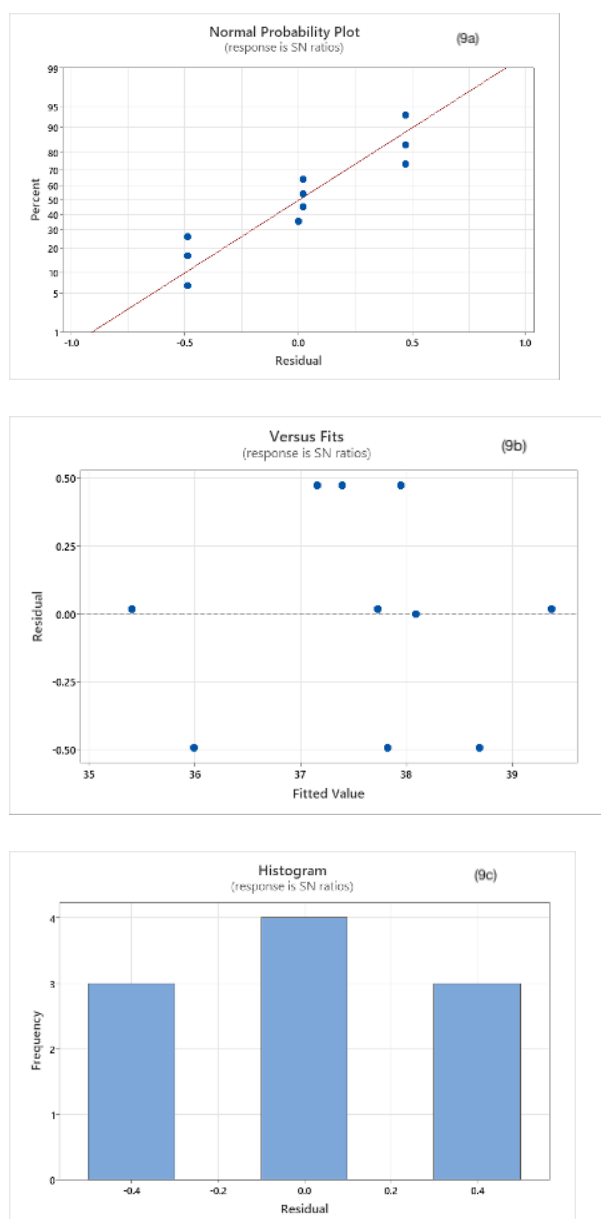


Fig. 9. Residual plots for oil and grease removal (a) Normal Prob. Plot (b) Versus fit (c) Histogram

3.5 Electrode Consumption Analysis

The anodes and cathodes were weighed before and after each experiment and mass consumption was compared

in Fig. 10. It can be observed that the highest electrode mass loss occurred with experiment 10, which is the only experiment carried out without application of Coconut Shell Ash as supporting electrolyte. This suggests that the use of CSA as supporting electrolyte reduced electrode consumption because it increases the conductivity of the solution and affects the metal dissolution rate [14].

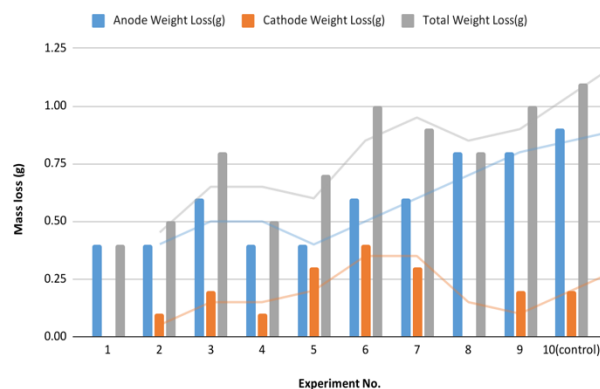


Fig. 10. Electrode mass consumption.

3.6 Crude Oil-Contaminated Water Colour Change

The Crude Oil-contaminated water had a characteristic black-brown color with emulsified oil and other visible impurities. The color of the wastewater was observed to change drastically within the first 5 - 10 mins of treatment during which the water had a turbid appearance. During the process, bubbles generated could be instantly seen carrying oil flocs and impurities to the top and sludge were observed at the top and bottom of the beaker. Fig. 11 show the wastewater before, during and after treatment respectively.



Fig. 11. Change in Colour of Crude Oil-Contaminated water. (a) Untreated water (b) Water during EC treatment (c) Water filtered after EC treatment

4 Conclusions

Crude Oil-Contaminated water was treated by Electrocoagulation using aluminum electrodes from

waste beverage cans and CSA based supporting electrolyte. From the results obtained, the following conclusions can be drawn:

- Maximum COD removal of 65.47% was obtained and corresponds to experiment 6 (20V, 5g/l and 30 mins).
- Maximum turbidity removal of 100% was achieved for experiment 7 (30V, 15g/l, 15 mins) and control (no ash).
- Maximum oil & grease removal of 93.26% was obtained at experiment 2 (10V, 10g/l, 20 mins).

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Data will be made available on request.

Nosa Idusuyi and Michael Femi John designed the experiments, wrote the initial drafts. Thomas Orhadahwe performed data analysis. All authors proofread the article and made necessary corrections.

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