

Assessing the Efficiency of Electrocoagulation in the Removal of Nitrogen and Phosphorus from Municipal Wastewater

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ABSTRACT

The discharge of nitrogen and phosphorus-rich wastewater into water bodies leads to eutrophication which results in environmental pollution that causes human health-related problems. Therefore, there is a need to regulate anthropogenic activities that produce these nutrients to make the environment safe for both plants and animals. This study aims to evaluate the efficiency of electrocoagulation to remove nitrogen and phosphorus from municipal wastewater. An electrocoagulation reactor was fabricated and a test run was to determine the physiochemical composition of wastewater (pH, temperature, colour, electrical, chloride, total hardness, alkalinity, nitrogen, and phosphorus) before and after treatment. The percentage removal of Nitrogen and phosphorus is 68% and 45% respectively with a treatment time of 60 minutes, current intensity of 3amps, and pH 6.5. There were significant changes in the initial physiochemical composition of wastewater, pH changes from 6.5 to 7.0, electrical conductivity from 1725µS/cm to 577µS/cm, chloride 864ppm to 288ppm, hardness 540mg/l to 320mg/l and alkalinity 75mg/l to 28.4mg/l. From the regression analysis phosphorus removal ratio has a variability of 91.31% while nitrogen has a variability of 85.40%. Current intensity and time have a statistically significant effect on nitrogen removal while pH, time, and current intensity have a statistically significant effect on phosphorus removal.

INTRODUCTION

Electrocoagulation (EC) is a method of water and wastewater treatment based on the electrochemical dissolution of sacrificial metal electrodes, which produces soluble or insoluble species that enhance coagulation, adsorption, or precipitation of soluble or colloidal pollutants, achieving high removal efficiency (AJER,2018). Electrocoagulation is an alternative electrochemical treatment method for nutrient removal that has gained increasing attention in recent years due to its simple operation, high removal efficiency, little sludge production and reduced chemical requirements. The most commonly used methods before these discoveries are chemical precipitation and biological processes. However, other technologies such as electrocoagulation are also being investigated (Edwar, 2020). In recent years, electrocoagulation methods have been successfully used to treat wastewater. The method has several advantages over chemical treatment methods, such as fewer coagulant ions required, no additional chemicals needed, relatively low area demand, low investment cost, and small sludge volume produced. Due to these advantages, electrocoagulation has gained widespread support because of its limited chemical requirements and relative comfort of operation (Magnisali *et al.*, 2021). Pranjal *et al.* (2022) in their study described that the electrocoagulation process is widely

used to destabilize the pollutants present in the form of dissolved or suspended particles in the electrolytic solution through the application of electric current. The electrocoagulation set-up comprises an electrolytic cell and a series of sacrificial metal electrodes (usually Fe or Al) coupled to a controlled DC power source. The efficiency of the electrocoagulation process, on the other hand, depends on several factors, such as electric voltage, ionic concentration, electrode material, reaction time, and temperature. Optimizing these factors plays an important role in the success of the process (Hatice and Elif, 2014).

The use of electrocoagulation is not limited to wastewater. Elkacmi et al. (2023) in their study use the process for the removal of chloroquine from an aqueous solution. World Health Organization (WHO) and U.S. Environmental Protection Agency (USEPA) have set the maximum discharge limit for different species of nutrients as 0.5-1.0 mg/L for phosphate, ≤ 50 mg/l for ammoniacal nitrogen, < 10 mg/L for nitrate, < 5 mg/L for ammonia, < 0.1 mg/L for nitrite, etc. (EPA, 2009; WHO, 2011). Phosphorus is one of the essential elements for many living organisms; however, excess amounts in lakes and other natural water bodies can cause a significant increase in algae (eutrophication), which can harm water quality, food resources, and aquatic life. These algae produce elevated toxins and promote bacterial growth that can affect human health when individuals are exposed to polluted water (U.S.E.P.A, 2022). Nitrogen/nitrate is a risk to human health, especially as a possible cause of infant methemoglobinemia (Johnson, 2019). The removal of nitrogen and phosphorus from wastewater has become an emerging global concern because these compounds cause eutrophication in natural water. Thus, controlling phosphorus discharged from municipal and industrial wastewater treatment plants is a key to preventing eutrophication of surface waters. Although many studies on wastewater properties (i.e., initial pH and initial phosphorus concentration) and operating parameters (i.e., initial density and treatment time) for phosphorus removal by EC process using Fe and Al anode were reported in the literature, however, this report evaluates the efficiency of using electrocoagulation in eliminating nitrogen and phosphorus from municipal wastewater, and to determine how its main operating parameters (current intensity, contact time, pH) influences the process and the thicness of the cathode and anode materials.

MATERIALS AND METHODS

Fabrication

A batch-type acrylic material(non-reactive) plastic was used to fabricate the electrocoagulation reactor of size 40cm wide, 50cm long, and 40cm high to treat 80 litres of wastewater. A sealant (special glue) was used to join and hold the material together, and a water outlet with a tap was fixed at a side, 20mm from the bottom, and 20mm away from the point of attachment. Four Aluminium plates were used as electrodes at the anode, 20cm wide, 20cm long, and 0.05cm thick. Four iron plates were used as electrodes at the cathode, 20cm wide, 20cm long, and 0.1cm thick. The anode and cathode plates were connected by conductive clips and connected to a variable power supply following the procedures described by Edwar (2020).

Experimental Tests

Samples of wastewater were collected from an open drain channel at a local market, Ogbomoso North, Oyo State Nigeria. The reactor was set up, and 20 litres of the sample with an initial pH of 6.5 were poured into it. The power supply was switched on and pre-heated for 10 minutes to set the experiment in motion. The power supply was set to

1 amp for the first stage of the experiment, and effluents were taken for 1 hour at 20mins intervals. The reactor and plates were cleaned after the first hour. The process was repeated the second time with the power supply set at 3 amps. NaOH (Sodium hydroxide) and HCL (Hydrochloric acid) were used to adjust the pH to 7.5 respectively in a dropwise manner and checked using a pH meter for the second stage, the procedure was repeated twice for 1 amp and 3 amps, and effluents were taken at 20mins interval for 1hour. For the third stage, the pH was adjusted to 9 and the process was repeated for 1 amp and 3 amps current intensities. A total of 18 wastewater samples were tested for physicochemical analyses. Nitrogen and phosphorus percentages removal were calculated using equation 1.

$$\%Y = \left(\frac{Ci - Cf}{Ci}\right) \tag{1}$$

Where: %Y; nitrogen and phosphorus removal percentage

ci; Initial nitrogen and phosphorus concentration

cf; Final nitrogen and phosphorus concentration

The raw wastewater samples and the treated effluent samples were taken to the laboratory for temperature, electrical conductivity, colour, chloride, total hardness, alkalinity, turbidity, pH, nitrogen, and phosphorus analyses using APHA standard methods of examination. Regression analysis was used as a statistical tool for estimating relationships between the dependent variable and the three independent variables.

RESULTS AND DISCUSSION

The experimental setup is shown in Figure 1a, the reactor contains the raw wastewater sample while Figure 1b shows the treated effluent samples obtained at intervals from the electrocoagulation process.

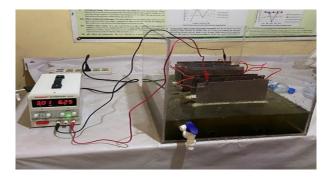


Figure 1(a): Experimental set up in operation



Figure 1(b): Effluents removed at interval

Tables 1 and 2 give the average value of the physiochemical analysis of raw wastewater samples collected and effluent samples obtained. Figures 2a and b reveal the treatment capacity of the reactor as all the parameters considered drastically improved in quality.

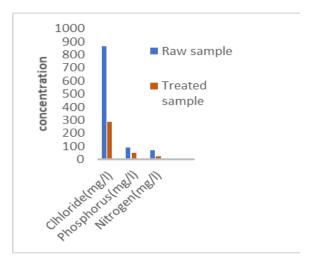
Table 1: Result of Physiochemical Analysis of Raw Wastewater Sample

Parameters	Result		
Temperature	27.9°C		
Electrical Conductivity	1725 (μ S/cm)		
Colour	Deep Green		
Chloride	864ppm		
Total Hardness	540mg/l		
Alkalinity	75mg/l		
Turbidity	1.43NTU		
pH	6.5		
Nitrogen	69.47mg/l		
Phosphorus	92.13mg/l		

Table 3 gives the experimental design with the response of variable removal (%) of nitrogen and phosphorus. The pollutant removal efficiency increases along with the electrolysis time, but beyond the optimal electrolysis time of 60 minutes this removal efficiency become constant as indicated in Figure 3 to Figure 8. At 60 minutes high removal values were reached for Nitrogen and phosphorus at pH 6.5 and 7.5 respectively and correspond with the report of Bharath and Manoj, 2018. However, the removal of total phosphorus was lower as compared with Nitrogen, the highest efficiencies were reached after 60 minutes of retention, with a value of 45% and 68% respectively.

Table 2: Result of Physiochemical Analysis of the Treated Effluent

Parameters	Result
Temperature	29.3°C
Electrical Conductivity	577 (μ S/cm)
Colour	Colourless
Chloride	288ppm
Total Hardness	320mg/l
Alkalinity	28.4mg/l
Turbidity	1.0NTU
pH	7.0
Nitrogen	22.23mg/l
Phosphorus	50.67mg/l



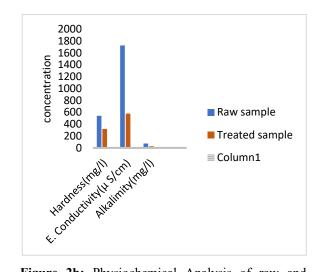


Figure 2a: Physiochemical Analysis of raw and treated

Figure 2b: Physiochemical Analysis of raw and treated wastewater

Table 3: Experimental Design with the Response Variable Removal (%) of Nitrogen and Phosphorus

EXP	FACTORS			REMO	OVAL (%)	CONTENT	
	Current intensity(A)	Time(min)	pH X ₃	Nitrogen Y ₁	Phosphorus Y ₂	Nitrogen mg/l	Phosphorus mg/l
1	1	20	6.5	13	8	60.79	81.43
2	1	40	6.5	26	27	51.75	69.43
3	1	60	6.5	64	37	25.55	57.35
4	1	20	7.5	24	13	52.44	80.40
5	1	40	7.5	25	31	52.40	63.68
6	1	60	7.5	68	38	22.48	57.43
7	1	20	9.0	27	14	50.99	79.23
8	1	40	9.0	29	28	49.75	66.42
9	1	60	9.0	60	44	28.27	52.49
10	3	20	6.5	39	27	42.77	67.44
11	3	40	6.5	45	35	38.73	60.50
12	3	60	6.5	68	45	22.37	54.48
13	3	20	7.5	32	25	47.75	69.43
14	3	40	7.5	42	33	40.30	62.49
15	3	60	7.5	65	41	24.75	55.22
16	3	20	9.0	35	27	45.43	67.48
17	3	40	9.0	54	39	32.09	56.46
18	3	60	9.0	66	44	23.75	52.13

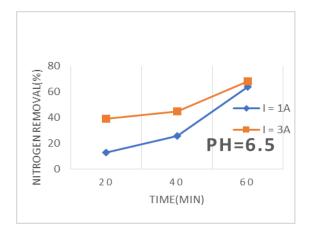


Figure 3: Nitrogen removal (%) versus time (Nitrogen =6.947mg/100ml, pH = 7.5)

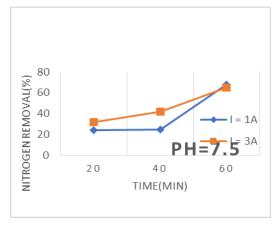


Figure 4: Nitrogen removal (%) versus time (Nitrogen =6.947mg/100ml, pH = 6.5)

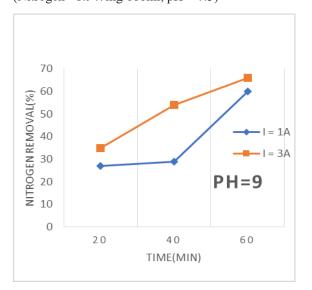


Figure 5: Nitrogen removal (%) versus time. (Nitrogen =6.947mg/100ml, pH = 9)

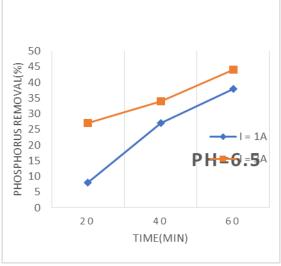


Figure 6: Phosphorus removal (%) versus time (Phosphorus = 9.213mg/100ml, pH =6.5)

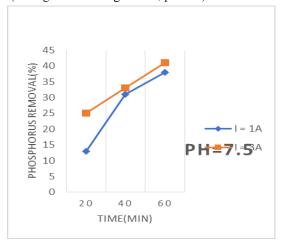


Figure 7: Phosphorus removal (%) versus time. (Phosphorus = 9.213mg/100ml, pH = 7.5)

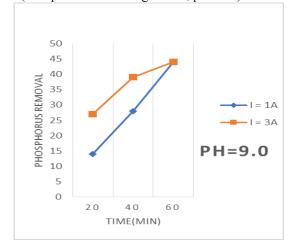


Figure 8: Phosphorus removal (%) versus time (Phosphorus = 9.213mg/100ml, pH =9)

Current intensity has an important effect on the electrocoagulation process. In the case of nitrogen, it was observed in Figure 3 to Figure 8 that there is a greater influence of current intensity than for phosphorus. The highest efficiency of 68% removal was obtained at 3 amps after 60 minutes of treatment at a pH of 6.5 and 1 amps after 60 minutes of treatment at a pH of 7.5, which indicates that there is a correlation between current intensity and pH value. The range of applied current density may differ depending on the types of effluents as recorded by Pranjal *et al.* (2022).

One of the most important factors that affect the performance of the electrocoagulation process and directly influences the efficiency of the removal of contaminants, is the initial pH. The effect of pH is directly associated with the formation of coagulants in solution as stated by Pranjal *et al.* (2022). The pH of water directly affects the solubility of metal hydroxides and consequently, the formation of colloidal particles on the anode surface. The results indicate that the difference between the phosphorus removal efficiencies at a pH of 6.5, 7.5, and 9 is minimal. In the beginning, it was observed that the variation of this parameter accelerated the process, but after 60 minutes of treatment, they became practically the same and reached values between 41% and 45%, 38% and 44% for 1 and 3amps current intensity respectively as shown in Figures 3, 4, 5,6, 7,8 and table 3. This is similar to what was found by Attour, 2014 in their studies on synthetic water, which showed that there is no need to realize pH modifications for an efficient removal of phosphates and nitrogen.

Statistical analysis

This analysis was done at $\alpha = 0.05$ for NITROGEN and the result is in Table 4, it shows the p-values for both current intensity and time are less than the level of significance (α), this means that they have a statistically significant effect on nitrogen removal while pH has no statistically significant effect on Nitrogen since it's p-value is greater than α .

The coefficient of determination $(R^2) = 85.40\%$, which means that the resulting regression model explains 85.40% of the variability in the Nitrogen removal ratio.

The proposed regression model is given below;

Nitrogen = -22.1 + 6.74 Current intensity (A) + 0.928 Time(min) + 1.81 pH

Table 5 shows the p-values for all three explanatory variables; current intensity, time and pH, are less than the level of significance (α), which means that they all have a statistically significant effect on phosphorus removal. The coefficient of determination (R^2) is 91.31%, which means that the resulting regression model explains 91.31% of the variability in the phosphorus removal ratio. The proposed regression model is given below; Phosphorus = -11.54 + 4.219 Current intensity(A) + 0.5780 Time(min) + 1.347 Ph

CONCLUSION

The variable parameters (i.e. pH, current intensity and time) influence the removal efficiency of phosphorus and the highest value reached after 60 minutes was 45%. Time and current intensity were the most influential variables in the process for the removal of nitrogen while the effect of pH was not significant and the highest removal value was 68%. This study validates the ability of the electrocoagulation process to remove phosphorus and nitrogen from wastewater. Time and current intensity were the most influential variables in the process, while the effect of pH was not significant for nitrogen. All the parameters (i.e. pH, current intensity and time) affected the removal efficiency of phosphorus.

Table 4: Regression Analysis: Nitrogen versus Current intensity (A), Time (min), pH

Variation source	DF	Adj SS	Adj MS	F-Value	P-value	
Regression 3		5775.95	1925.32	31.20	0.000	
Current (A)	1	887.81	887.81	14.39	0.002	
Time(min)	1	4430.61	4430.61	71.80	0.000	
Ph	1	69.27	69.27	1.12	0.305	
Error	16	987.29	61.71			
Lack-of-fit	14	987.29	70.52			
Pure error	2	0.00	0.00			
Total	19	6763.25				

S = 7.85531, $R^2 = 85.40\%$, Adj $R^2 = 82.66\%$

Table 5: Regression Analysis: Phosphorus versus Current intensity(A), Time(min), pH

Variation source	DF	Adj SS	Adj MS	F-Value	P-value
Regression	3	2265.92	755.31	56.05	0.000
Current (A)	1	347.85	347.85	25.81	0.000
Time(min)	1	1719.07	1719.07	127.56	0.000
рН	1	38.39	38.39	2.85	0.111
Error	16	215.62	13.48		
Lack-of-fit	14	215.62	15.40		
Pure error	2	0.00	0.00		
Total	19	2481.54			

S = 3.67099, $R^2 = 91.31\%$, $Adj R^2 = 89.68\%$

Electrocoagulation is capable of treating pollutants in wastewater: decolourization, chlorine reduction, pH adjustments, reduction of electrical conductivity, removal of turbidity, hardness reduction as well as alkalinity. However, more studies need to be conducted to justify the operating parameters used for scaling up the process, especially when these parameters vary based on the type of effluents.

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