

# Phosphate removal from wastewater by electrocoagulation using aluminium electrodes

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# Abstract

The removal of phosphate from wastewater by electrocoagulation using aluminum electrodes was investigated. The main objectives of the experiments were to investigate the effects of the various operating parameters, such as current density, Electrocoagulation(EC)duration, initial phosphate concentration, temperature, pH of the solution, salt (NaCl) concentrations and electrolyte type [NaCl, KCl, NaNO<sub>2</sub>, NaNO<sub>3</sub> ] on the phosphate removal efficiency from synthetic wastewater in batch EC process. The ranges of experimental parameters were between 10 and 150 mg PO<sub>4</sub>/L for initial phosphate concentration, 3-10 for initial pH of the wastewater, 0.5-5 g NaCl/L for supporting electrolyte concentration, 2g/L of other supporting electrolytes KCl, NaNO<sub>3</sub>, NaNO<sub>2</sub> and 1.13-4.54 mA/cm<sup>2</sup> for current density. The results showed that the maximum removal efficiency was achieved at a current density of 4.54mA/cm<sup>2</sup>, at a pH of 7.0 and 4-5 g/L NaCl.

# **Keywords**

Phosphate Removal, Electrocoagulation, Water Treatment, Nutrient Removal, Aluminum Electrode

# 1. Introduction

Wastewater can create severe water pollution problems for aquatic life because of its various contents [1]. Water pollution by nutrients is a historical and ever-growing concern in developed and developing countries alike. On one hand phosphorus (p) is an important nutrient that is critically needed for the normal functioning of ecosystems [2]. Phosphorus is found as phosphate  $(PO_4^{3-})$  in nature and present as orthophosphate, polyphosphate and organic phosphate in water. Phosphorus compounds came from various sources but agriculture and cattle are the main direct and indirect origins of its presence [3]. On the other hand phosphate remains a critical environmental pollutant [4], it is one of the nutrients responsible for eutrophication of the receiving water bodies [5] and subsequent deterioration of water quality [6]. Eutrophication is a common environmental problem that arises in the interface between human activity and surface water [7]. Environmental problems arise as the algae decays, consuming dissolved oxygen required for higher organisms and degrading general water quality.

Because of phosphate later problems, various methods have been used for its removal from wastewater. Phosphorus removal techniques fall into three main categories: physical [8]-[10], chemical [11]-[13], and biological [14],[15]. Physical methods are too expensive. Phosphorous removal by chemical treatments is not used due to disadvantages like high maintenance cost, problems of sludge handling and its disposal, and neutralization of the effluent. In biological treatment removal efficiency usually doesn't exceed 30%, which means that we should use other technologies [16]. In recent years, another method has attracted great attention in wastewater treatment which is electrochemical technique including, electroflotation (EF), electrodecantation and electrocoagulation (EC) [17]-[20]. EC has been successfully used to remove different kinds of pollutants such as organic compounds, heavy metals [21], biodiesel wastewater, dyes, textile wastewater [22],[23], hardness [24], COD [25],[26] and important anions such as nitrate [27], fluoride[28] and phosphate [29].

# 2. Electrocoagulation

Electrocoagulation (EC) is a technology in which the coagulant is generated in situ by oxidation of a metal anode material when applying electrical current [29]. As well known,  $Al^{3+}$  ions dissolve and combine with hydroxyl ions in water [16], when direct current passes through the Al anodes, they form metal hydroxides, which are partly soluble in the water under definite pH values.

Additionally, electrolytic reactions evolve gas (usually as hydrogen bubbles) at the cathode that can enhance the process; this effect is known as electroflotation which results in better removal of contaminants [30]-[32]. Electrocoagulation has occurred in three steps. In first step, coagulant has formed because of oxidation of anode. In second step, pollutants have destabilized. In last step, destabilized matters have united [33]. The most common electrode materials for electrocoagulation are aluminum and iron. They are cheap, readily available, and proven effective [34]. When aluminum is used as electrode material, the reactions are as follows:

At the anode:

$$Al \to Al^{3+} + 3e^{-} \tag{1}$$

At the cathode:

$$2H_2O + 2e^- \rightarrow H_{2(g)} + 2OH^-$$
(2)

When the anode potential is sufficiently high, secondary reactions may occur, especially oxygen evolution (Eq.3):

$$2H_2O \rightarrow O_2 + 4H^+ + 4e^-$$
 (3)

Aluminum ions  $(Al^{3^+})$  produced by electrolytic dissolution of the anode (Eq.1) immediately undergoes spontaneous hydrolysis reactions which generate various monomeric species according to the following sequence [35]:

$$Al^{3+} + H_2O \rightarrow Al(OH)_2^+ + H^+$$
(4)

$$Al(OH)_{2}^{+} + H_{2}O \rightarrow Al(OH)_{2}^{+} + H^{+}$$
(5)

$$Al(OH)_{2}^{+} + H_{2}O \rightarrow Al(OH)_{3} + H^{+}$$
(6)

The overall reaction in the solution:

$$Al_{(aq)}^{3+} + 3H_2O \rightarrow Al(OH)_3 + 3H_{(aq)}^{+}$$
(7)

EC process like other treatment methods has some advantages and disadvantages. Main disadvantages of this method are lack of a systematic approach to EC reactor design and operation, replacement of electrodes at regular intervals, high cost of electricity and anode passivation [36].

On the other hand, main advantages of EC are: the simplicity of the equipment [37], no need of additional chemical matter after or before treatment, relatively low area demand, also sludge from this process is intensive and has low water [33].

For these reasons the presented work is focused on the EC processes using a sacrificial aluminum anode to remove phosphorous from waste water by studying variables like

current density, initial phosphorous concentration, time, temperature, pH, electrolyte concentration (NaCl), and different types of electrolytes.

## **3. Experimental Part**

#### 3.1. Materials

All chemicals used were analytically graded and used without further treatment. Distilled water was used in all experiments. Stock phosphate solutions were prepared from A.R. potassium dihydrogen phosphate. The initial pH was adjusted to a desired value using sodium hydroxide or hydrochloric acid solution.

#### **3.2. Analytical Method**

The analysis of phosphate was carried out using yellow vanodomolybdo-phosphoric acid method by a double beam spectrophotometer (UNIC0, Spectrophoto-meter, 1200 series, united products & instruments, Inc, USA) according to the standard methods of examination of water and wastewater [38].

#### **3.3. Experimental Setup**

A two litres plexi glass rectangular tank, with dimensions of  $10 \times 10$  cm square base and a height of 20 Cm with four plate electrodes (2 anodes and 2 cathodes) with an area of 10  $\times$  22 cm<sup>2</sup> was used as shown in Fig(1). The plates were placed vertically in the tank and separated by a distance of 2 Cm. Perforated stainless steel sheets were used as a cathode while aluminum plates were used as anode. Alternating electrodes were connected to a digital DC power supply (containing amperometer and voltmeter was used to measure the current intensity passing through the circuit and the applied potential difference respectively), characterized by 10 A for current intensity and a range of 0-40 V for voltage in monopolar mode. Synthetic phosphate solutions (with concentrations varing from 10 to 150 mg PO<sub>4</sub>/L) were used. At the beginning of each run the phosphate solution of the desired concentration was fed into the cell and solution of NaCl (as electrolyte) was added to it to increase the solution conductivity. Each run was timed starting with the DC power supply switching on.

#### **3.4. Electrocoagulation Procedure**

For each run a 1.5 L of synthetic waste solution was mixed with the appropriate amount of sodium chloride which was used as a conductor. The solutions were placed into the electrolytic rectangular cell. The pH was adjusted by the addition of NaOH or HCl solutions. Direct current from the D.C power supply was passed through the solution via the four electrodes during the 180 minutes of electrolysis run. 10 mL of the solution was drawn at every 5 minutes interval for the first half hour and every 30 minutes interval for the remaining time of the run. The location of the drawn samples was kept constant for each run. Samples were filtered, and then taken for absorbance measurements at wave length 470nm. The measured absorbance was then converted to the residual concentration using a calibration curve. The electrodes were washed with HCl solution (15% w/v) before each run in order to remove any adhering scales or oxides. Following each run, the electrodes were washed with distilled water, dried until they are used again. The efficiency of phosphorus compounds removal, was calculated as

Removal efficiency, 
$$\% = \frac{\text{Ci-Cf}}{\text{Ci}} * 10$$
 (8)

Where; Ci is the initial phosphate concentration (mg/L) and Cf is the final phosphate concentration (mg/L).



Fig (1). Laboratory scale cell assembly: 1-cathode, 2-anode, 3-plexi glass tank, 4solution level, 5-D.C power supply.

# 4. Results and Discussion

Electrocoagulation is a complex process that may be affected by different parameters such as electrolysis time, initial pH, initial pollutant concentration, current density, supporting electrolyte and temperature.



#### 4.1. Effect of Current Density

**Fig (2).** Effect of current density on the percentage of removal of phosphate (Co = 20 mg/L, NaCl = 1 g/L, pH = 5.2, Temperature = 25°C and time=180 min)

Current density is described as the amount of current applied per surface area of the anode. As seen in Fig (2) at constant initial phosphate concentration, pH, temperature and NaCl concentration it is found that the efficiency of phosphate removal has increased from 85% to 95% by increasing current density from 1.13 to 4.54 ( $mA/cm^2$ ). When direct current passes through the Al anodes, Al<sup>3+</sup> ions dissolve and combine with OH- in the water to form aluminum hydroxide which is responsible for electrocoagulation efficiency. At high current density the quantity of Al<sup>3+</sup> from anode dissolution increases according to Faraday's low. As a result, the amount of aluminum hydroxide increases, which adsorbs a large amount of phosphate on its surface. In other words by increasing the current of the cell the amount of hydrogen bubbles at the cathode increases, resulting in a greater upwards flux and a faster removal of the pollutant and sludge flotation. An increase in current density above the optimum current density does not result in an increase in the pollutant removal efficiency as sufficient number of metal hydroxide flocs is available for the sedimentation of the pollutant. It can easily be concluded that the increase in current density results in an increase in the removal efficiency in accordance with earlier studies [39], [40].

## 4.2. Effect of Electrolysis Time

Time of electrocoagulation is the time taken by the process to generate metal hydroxides and to complete coagulation of the impurities. To explore the effect of time, current density is kept constant.



**Fig (3).** Effect of electrolysis time on the percentage of removal of phosphate ( $Co = 20 \text{ mg/L}, \text{ } \text{C.D} = 3.41 \text{ mA/cm}^2, \text{ } \text{NaCl} = 1 \text{ g/L}, \text{ } \text{pH} = 5.2, \text{ } \text{Temperature} = 20^{\circ}\text{C}$ )

As seen in Fig (3) most of phosphate removal is reached under some optimal values of electrocoagulation duration (90 mins). Further increase of the electrocoagulation time have a small influence on the degree of phosphate removal, and from the fig, after 90 mins of electrocoagulation results become very close to each other.

Generally, for a fixed current density, the generated metal hydroxide increases with an increase in the electrolysis time. For a longer electrolysis time, there is an increase in the generation of flocs resulting in an increase in the pollutant removal efficiency. For an electrolysis time beyond the optimum electrolysis time of 90 mins, the pollutant removal efficiency does not increase as sufficient numbers of flocs are available for the removal of the pollutant. These results are similar to the results of the previous workers [41],[42].

#### 4.3. Effect of Temperature

The effect of varying temperature from 20 to 50°C has been studied as shown in Fig (4). It can be observed that increase in temperature causes an increase in removal efficiency. This may be attributed to the increase in the mass transferred of  $Al^{3+}$  from the anode surface to the solution bulk and the increase of the rate of  $Al^{3+}$  hydrolised to  $Al(OH)_3$ . This agree with previous results [42],[43]. Increasing the temperature above 50°C was found to be uneconomic as there was a slight change in the rate of the reaction and the percentage of removal.

#### 4.4. Effect of Initial pH

The pH is one of the important factors affecting the performance of electrochemical process. The pH value increased as the operating time of EC process was increased due to the OH<sup>-</sup> ion accumulation in aqueous solution during the process. The effect of change of initial pH values on the removal efficiency is illustrated in Fig (5).







**Fig (5).** Effect of the pH Value of the solution on the percentage of phosphate ( $C_o = 20 \text{ mg/L}$ ,  $C.D = 3.41 \text{ mA/cm}^2$ , NaCl = 1 g/L, Temperature = 20°C and time= 150 min)



Fig (6). Effect of pH on percentage of removal (Time= 1hr; C.D= 3.41, Temperature=  $20^{\circ}$ C)

As shown in Fig (5), very low range of pH is not desirable. Highest removal efficiency can be achieved near neutral pH. The lower percentage of removal at higher acidic and alkaline pH can be explained by amphoteric behavior of Al(OH)<sub>3</sub> which does not precipitate at very low pH. Fig (6) show the effluent pH after EC treatment would increase for acidic influent but decrease for alkaline influent. The increase of pH at acidic condition was attributed to H<sub>2</sub> evolution at cathodes and formation of Al(OH)<sub>3</sub> near at the anode would relase H<sup>+</sup> leading to decrease of pH. In addition, there is also O<sub>2</sub> evolution reaction leading to pH decrease. As for the pH decrease at alkaline conditions, it can be the result of formation of Al(OH)<sup>-</sup><sub>4</sub> which is soluble and useless for adsorption of phosphorus.

$$Al(OH)_3 + OH^- \rightarrow Al(OH)_4^- \tag{9}$$

These results are in accordance with previously published works [34],[44],[45].

#### 4.5. Effect of Initial Phosphate Concentration



**Fig** (7). Effect of initial concentration on the percentage of removal of phosphate (pH = 5.2, C.D = 3.31 mA/cm2, NaCl = 1 g/L, Temperature =  $20^{\circ}C$  and time = 180 min)

Effect of phosphate concentration from 10 to 150 mg/L on the removal efficiency has been illustrated in Fig (7). It can be observed that increase in concentration at constant EC time and current density result in decrease in removal efficiency.

At a constant current density, the same amount of aluminum ions passes through the solution at different phosphate concentration. Consequently, the formed amount of aluminum hydroxides was insufficient to coagulate the greater number of phosphate molecules at higher phosphate concentration, previous works support this phenomena [37],[46].

#### 4.6. Effect of NaCl Concentration

Different doses of NaCl were added as supportive electrolyte to enhance the conductivity of the solution. Fig (8) presented that the increase in the amount of NaCl results in increasing removal efficiency. The amount of aluminum generated increased rapidly as the NaCl dose varies from 0.5 to 5 g/L. This may be explained by the fact that the higher chloride ion concentration, the higher the ability of Cl<sup>-</sup> to destroy any passive oxide film which tends to be formed on the anode and limit anode dissolution, hence it increases the availability of Al(OH)<sub>3</sub> in solution and improve the efficiency of phosphorus removal. Previous works shows similar results [47], [48]. Varieties of electrolytes like NaCl, KCl, NaNO<sub>3</sub>, NaNO<sub>2</sub>, NaSO<sub>4</sub>, etc., are available, But due to low cost and easy availability, NaCl has been selected as the best electrolyte [49], [50]. The addition of NaCl would also lead to the decrease in power consumption due to the increase in conductivity.



Fig (8). Effect of NaCl concentration on the percentage of removal of phosphate (Co = 20 mg/L, pH = 5.2, C.D. = 3.41 mA/cm<sup>2</sup>, Temperature =  $20^{\circ}C$  and time = 150 min)

#### 4.7. Effect of Nature of Electrolyte

Most electrocoagulation studies have used chloride as anion to enhance the conductivity of the solution and some of them have utilized nitrate and sulphate as the electrolyte [45],[51]. A passive oxide film on the electrodes surface might increase IR drop across the electrode. These problems were overcome by adding electrolyte into the system.

In this study, different types of supporting electrolytes sush as NaCl, KCl, NaNO<sub>2</sub>, and NaNO<sub>3</sub> were studied. And it is known that conductivity of  $NaCl > KCl > NaNO_3 > NaNO_2$ . When the conductivity increases, anodic dissolution increases resulting in enhanced coagulation percentage. From Fig (9), 88% and 84% of phosphorus removal were obtained while using NaCl and KCl respectively as electrolyte. Whereas 74% and 70% were observed in case of NaNO3 and NaNO<sub>2</sub> respectively. This may be explained by the presence of chloride ions that not only increase the solution conductivity, but also decrease the passivity of the electrode surface due to catalytic action, unlike nitrate and nitrite ions. These results are in accordance with previously published works [52], [53].



Fig (9). Effect of nature electrolyte on the percentage of removal of phosphate ( $C_o = 20 \text{ mg/L}$ , pH = 5.2,  $C.D. = 3.41 \text{ mA/cm}^2$ , Temperature = 25°C, electrolytes concentration = 2g/L and Time = 150 min)

#### 4.8. Electrical Energy Consumption and Electrode Consumption



Fig (10). Effect of current density on the energy consumption and Al consumption of phosphate (C\_o = 40 mg/L, pH =5.2, NaCl = 1 g/L, *Temperature* =  $25^{\circ}C$ )

In order to assist in assessing the economic feasibility of electrocoagulation in comparison with other techniques, the energy consumption and Al metal consumption were calculated as follows[55]:

(i) Energy consumption (kWh/g phosphate removed) = 
$$\frac{EIEC}{(Co-Ct)V}$$
 (10)  
Where:

E is the cell voltage (Volt), I is the current (A),  $t_{EC}$  is the electrocoagulation time (h), Co is the initial compound concentration (mg/L),  $C_t$  is the concentration at time t (mg/L), V is the treated volume (L).

(ii) The amount of Al metal consumed in electrocoagulation was calculated using Faraday's law:

Al consumption (g Al/g phosphate removed) = 
$$\frac{\text{ItM}}{\text{ZFV(Co-Ct)}}$$
 (11)

Where:

M is the molecular weight of aluminum (27 g/mol), Z is the number of electron transfer ( $Z_{AI} = 3$ ), F is Faraday's constant (96,500 Columb).

The variation of electrical energy consumption and electrode consumption with current density, initial electrolyte concentration and initial phosphate concentration were presented in Figures (10 to 12).



**Fig (11).** Effect of NaCl Concentration on the energy Consumption and Al consumption of phosphate, ( $C_o = 40 \text{ mg/L}$ , pH = 5.2,  $C.D. = 3.41 \text{ mA/cm}^2$ , Temperature = 25°C)



**Fig (12).** Effect of the initial concentration on the energy consumption and Al consumption of phosphate, (NaCl = 1 g/L, pH = 5.2, C.D. =  $3.41 \text{ mA/cm}^2$ , Temperature =  $25^{\circ}$ C)

As seen from previous figures the experimental energy consumption values ranged from 0.31 to 2.24 kWh/g phosphate removed, 1.96 to 1.27 kWh/g phosphate removed and from 5.47 to 0.44 kWh/g phosphate removed. The variation of aluminum consumption ranged from 0.004 to 0.017 g aluminum/g phosphate removed for initial phosphate concentration, 0.016 to 0.011 g aluminum/g phosphate removed for initial phosphate removed for initial phosphate concentration and from 0.035 to 0.0041 g aluminum/g phosphate removed for initial phosphate removed for initial phosphate concentration. It is clear from these figures that energy consumption increases with increasing the current

density and decreases with increasing sodium chloride concentration, and initial concentration of phosphate removed. Aluminum consumption increases with increasing the current density, and decreases with increasing initial concentration of phosphate removed and sodium chloride concentration [55],[56].

#### 4.9. Kinetics



**Fig (13).** Effect of current density on the kinetics of the reaction, (Co = 40 mg/L, pH = 5.2, NaCl = 1 g/L, Temperature = 25°C, Time= 180min)



**Fig (14).** Efect of NaCl concentration on the kinetics of the reaction, (Co = 40 mg/L, pH = 5.2, C.D. =  $3.41\text{mA/cm}^2$ , Temperature =  $25^{\circ}$ C, Time= 150 min)



Fig (15). Effect of pH values on the kinetics of the reaction, Co = 40 mg/L, C.D. =  $3.41\text{mA/cm}^2$ , NaCl = 1 g/L, Temperature =  $25^{\circ}$ C, Time = 150min)

The kinetic study is an important issue in order to examine the order of the reaction. In an attempt to throw some light on the kinetics of the present process which is difficult to quantify owing to its complexity and it is being controlled by many interacting variables. It was assumed that the process takes place according to first order mechanism [57], which can be expressed by:

$$\ln\left(\frac{Co}{Ct}\right) = Kt \tag{12}$$

Where;

 $C_o$  is the initial concentration (mg/L),  $C_t$  is the concentration at time t (mg/L), K is the rate constant (min.<sup>-1</sup>) and t is the time (min.).



**Fig (16).** Effect of initial concentration of phosphate on the kinetics of the reaction,  $(pH = 5.2, C.D. = 3.41 \text{ mA/cm}^2, \text{ NaCl} = 1 \text{ g/L}, \text{ Temperature} = 25^{\circ}\text{C}, \text{ Time} = 150 \text{min})$ 

Parameter	Value	<b>K</b> <sub>1</sub>	$R_{1}^{2}$	$K_2$	$R_{2}^{2}$
Current density, mA/cm <sup>2</sup>	1.13	0.0261	0.9955	0.0064	0.9832
	2.27	0.0307	0.9461	0.0076	0.9942
	3.41	0.0373	0.8973	0.0075	0.9898
	4.54	0.0467	0.8421	0.0084	0.9983
Initial concentration, mg/L	10	0.0491	0.8339	0.009	0.9901
	20	0.0411	0.9092	0.0084	0.9978
	40	0.0371	0.8983	0.0081	0.9968
	60	0.0327	0.8881	0.0084	0.9972
	80	0.029	0.9094	0.0079	0.9947
	100	0.0249	0.9542	0.0068	0.9945
	150	0.0213	0.984	0.0063	0.985
рН	3	0.0282	0.868	0.0077	0.9716
	5.8	0.0371	0.8983	0.0081	0.9968
	6	0.0413	0.8598	0.0087	0.9943
	7	0.0593	0.7846	0.0074	0.965
	10	0.0156	0.9647	0.0088	0.989
NaCl Concentration, g/L	0.5	0.0306	0.965	0.0042	0.899
	1	0.0373	0.8973	0.0081	0.9968
	2	0.0493	0.92	0.0072	0.906
	3	0.0615	0.9273	0.0072	0.8378
	4	0.0974	0.9619	0.0056	0.9525
	5	0.1187	0.8722	0.0048	0.7998

Table 1. Rate Constants for the First- Order Reaction

Figures 13 to 16 show the effect of current density, NaCl concentration, pH value and initial concentration of phosphate on the kinetics of the reaction. The present data fit (Eq.12), however, two slopes are distinguished indicating the presence of two rates of removal. In first stage which occurs

during few minutes, the rate is higher than the rate in second stage. The fast rate of removal in the first few minutes is ascribed to the flocs are still free so the adsorption rate is very fast. The low rate of removal in second stage is due to the fact that the flocs becomes exhausted (the number of free flocs is few) so the adsorption rate is very slow. Table (1) shows the calculated rate constants of the first order reaction.

# 5. Conclusions

- The EC method does not use any chemical reagents and makes the process of phosphate-containing wastewater treatment easy for regulation and automation.
- The efficiency of the electrocoagulation process depends on many operational parameters.
- In this study, the effects of main operational parameters like initial pH, initial phosphate concentration, NaCl Concentration, temperature, nature of electrolyte and current density were investigated for aluminum plate electrodes.
- The obtained results showed that phosphate removal completely depends on:
- The initial pH and at high acidic or alkaline conditions the removal efficiencies were lower.
- The pollutant removal efficiency increases with an increase in the electrolysis time. But beyond the optimum electrolysis time, the pollutant removal efficiency becomes constant and does not increase with an increase in the electrolysis time.
- At the highest initial concentration (150mg/L), the phosphate removal efficiency was decreased.
- At this initial concentration of phosphate, additional time is necessary to obtain higher removal efficiencies. While at lower initial concentrations (10, 40 mg/L) the removal efficiencies reached to their maximum values.
- The increase of current density improves the efficiency of phosphate removal.
- Energy and Aluminum consumption decreases with increasing initial concentration of phosphorus, NaCl Concentration and increases with increasing the current density.

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- 97
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