Investigation of electrocoagulation process for the removal of phosphate: full-scale process optimization, operation cost and adsorption kinetics

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ABSTRACT

The studies on phosphate removal methods that will respond to needs in terms of both cost and high recover efficiency are ongoing. Electrocoagulation is also one of these treatment technologies that have been successfully applied in the last few years. In this study, the basic mechanism, optimization parameters, cost calculation, kinetics and isotherms of electrocoagulation method in aqueous solutions are investigated. The maximum phosphate removal efficiency was obtained as 100% where electrode combination (Fe-SS), pH (7), current (9 mA/cm^2), electrode distance (2 cm), phosphate concentration (25 ppm) and retention time (20 min). Operating cost of the electrocoagulation process was calculated as 0.94 \$/m^3. The obtained results also proved that second-order kinetic model and the Langmuir isotherm model are correlated with electrocoagulation data.

Keywords: Adsorption kinetics; Cost calculation; Electrocoagulation process; Optimization; Phosphate removal

1. Introduction

Phosphorus (P) is the 17th element that is common in the earth. In nature, phosphates are most commonly found in the form of orthophosphate and organophosphate [1]. It is an essential nutrient for life and the different properties exhibited by the various nonmetal compounds make it possible to use many of the phosphate compounds in the industry [2]. As a result, these industries produce phosphate-containing wastewater. The discharge of phosphate rich wastewater into the aquatic environments causes eutrophication, and this phenomenon leads to hazardous algae bloom, dissolved oxygen depletion and fish death [3,4]. Because of that, discharge limits of *P* in wastewater has been set in as 1 mg/L by many countries and the natural background levels of total phosphorus are generally accepted as less than 0.03 mg/L in freshwater aquatic systems. Also, natural cycle of phosphate has been significantly altered in the past 50 years depending on industrial usage of phosphate containing rocks [5,6]. According to the US Geological Survey's latest report on phosphate rock, global phosphate demand is seen growing by about 14% [7]. However, phosphate rock is the only economic resource of *P* for production of phosphate fertilizers and phosphate chemicals [2,8].

Recent studies have shown that phosphate-containing wastewater can be used as a secondary phosphate source, e.g., as sludge supernatant, municipal and industrial wastewater [9,10]. For the removal and recovery of phosphate from wastewater, several techniques have been studied such as struvite crystallization [11], calcium precipitation [12], amorphous calcium silicate hydrates adsorption [13], marine macro algae biosorption [14] and integrated selectrodialysis/crystallization [15] and reverse osmosis [16]. Besides these methods, electrocoagulation (EC) technology, which is one of the dephosphorization/recovery methods, attracted much interest from researchers because it provides advantages over other technologies [17].

In EC process, multiple reactions take place simultaneously, metal ions from sacrificial anode are driven into the water while water is hydrolyzed into hydrogen gas,

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which helps to remove pollutants by flotation. Meanwhile, electrons flow freely to destabilize surface charges on suspended solids, emulsified oils and other pollutants [18,19]. The reactions at the anode/cathode electrodes and in the EC cell can be summarized by the following equations [20–22].

Anode reactions:

$$M_{(s)} \rightarrow M^{3+}_{(aq)} + 3e^- \tag{1}$$

$$M^{3+}_{(aq)} + 3OH^- \rightarrow M(OH)_3 \quad \text{Alkaline pH range} \tag{2}$$

$$M^{3+}_{(aq)} + 3H_2O \rightarrow M(OH)_3 + 3H^+ \quad \text{Acidic pH range} \tag{3}$$
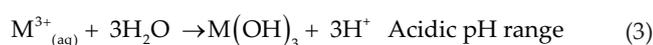
Cathode reactions:

$$3H_2O + 3e^- \leftrightarrow \frac{3}{2}H_2 \uparrow + 3OH^- \tag{4}$$

If phosphate is present, reaction can be as [23,24]:

$$M^{3+}_{(aq)} + PO_4^{3-} \rightarrow MPO_{4(s)} \tag{5}$$

Nowadays, EC process has been used for removal of oil [25], arsenic [26], chromium [27,28], boron [29], organics [30–32], zinc and lead [33], pesticides and drugs [34,35] and fluoride [36] from wastewater. Although, there are many studies on the removal of phosphate from wastewater by the electrocoagulation process [37–39] a full-scale optimization study has to be carried out in these studies, especially in the absence of the electrode optimization parameter which makes a difference in the results, the calculation of the operating cost and the lack of studies on the adsorption kinetics. The aim of this paper is to determine optimum operating conditions for phosphate removal by electrocoagulation process. For this purpose, parameters such as electrode combination (Al-Al, Al-Fe, Al-SS, Fe-Al, Fe-Fe and Fe-SS), pH (3–9), current density (3–15 mA/cm²), inter-electrode distance (1–3 cm), initial concentration (25–200 mg/L) and electrolysis time (10–90 min) were studied with a phosphate synthetic solution. Cost calculation, isotherm models and adsorption kinetics were also determined.

2. Material and methods

2.1. Phosphate-containing aqueous solution and chemical analysis

Phosphate-containing aqueous solution was prepared with appropriate amount of potassium dihydrogénophosphate (KH_2PO_4) per liter of distilled water. Samples of 25, 50, 100, 150 and 200 mg/L were prepared by dilution from the stock solution. The pH was adjusted with 0.1 N –1 N H_2SO_4 and NaOH solutions (Thermo -Orion 3 Star) and conductivity was adjusted with NaCl (Hach-Lange HQ40d). All chemicals used in this study were supplied from Merck Company.

For the phosphate analyses, the standard vandomolyb-dophosphoric acid calorimetric method [40] was applied with a UV spectrophotometer (T90+UV/VIS) at 430 nm after separating the particles with centrifuge (Hettich-ZentrufugenEBA20) at 6,000 rpm for 5 min.

The phosphate removal efficiency (RE) was calculated using the following equation:

$$RE\ (\%)\ = \frac{A_0 - A}{A_0} \times 100 \tag{6}$$

where A_0 and A represent the initial and final phosphate concentrations, respectively.

2.2. Reactor setup

The experimental setup scheme is shown in Fig. 1. The EC experiments were performed in a 1 L glass beaker with an effective reaction volume of 800 mL. The EC system consisted of a magnetic stirrer, a DC power supply unit and a pair of electrodes. All the electrodes were vertically immersed in the liquid for an active surface of 6 x 8.5 cm² (width x height). Distance between electrodes were set as 2 cm at early experiments, it has been changed at inter-electrode distance optimization. The electrodes were connected to a rectifier to supply required electrical current (AATech ADC-3303D).

After each experiment, electrodes were rinsed with 0.1 M HCl and deionized water to remove surface impurities and the electrodes were weighed to calculate electrode consumption for the calculation of operation cost.

2.3. Operation cost

The operating cost of the EC process can be calculated by two major parameters, electrode consumption and energy consumption. The relevant equations are given below [41]:

$$C_{energy} = \frac{U \times i \times t}{v} \tag{7}$$
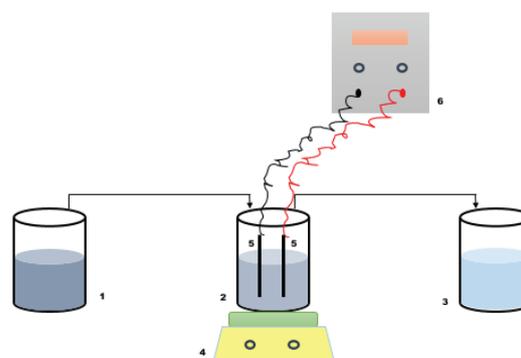
Fig. 1. Schematic illustration of EC system (1. wastewater storage tank, 2. EC reactor, 3. treated water storage tank, 4. magnetic stirrer, 5. anode and cathode, and 6. DC power supply).

where C_{energy} (kWh/m³) is the electrical energy consumption, U is the voltage(V), i is the applied current (A), t is the treatment time (h), v is the active volume (m³).

$$C_{\text{electrode}} = \frac{i \times t \times M}{z \times F \times v} \tag{8}$$

where $C_{\text{electrode}}$ is the electrode consumption (kg/m³) in theoretically, i is the applied current (A), t is the treatment time (h), M is the molecular weight of anode (Fe) (55.85 g/mol), z is the number of electron involved in the reaction ($z_{\text{Fe}} = 3$), F is the Faraday's constant (96,485 C/mol) and v is the active volume (m³).

$$OC = \alpha \times C_{\text{energy}} + \beta \times C_{\text{electrode}} \tag{9}$$

According to the Turkish electricity market in August 2018, price for electrical energy was 0.05 US $/kWh ($\alpha$) [42], and price for Fe electrode materials was 0.58 US $/kg ($\beta$) [43].

2.4. Modeling methods

The adsorption process for the electrochemical removal of the pollutant takes place in two steps; (i) metal flocs generation at the anode, (ii) adsorption of the cations on floc surface [44]. The removal mechanism of conventional adsorption and electrocoagulation is the same except the formation of coagulant [45].

In order to determine the mechanisms of the adsorption process, various kinetic models: first order, second order, pseudo-first-order and pseudo-second-order models were applied to describe the adsorption kinetics onto iron hydroxides. The relevant kinetic models are given below [46,47];

First order,

$$C_t = C_0 \times e^{-K_1 t} \tag{10}$$

where C_t (mg/L) is the concentration at EC time t (min), K_1 (min⁻¹) is the constant rate of adsorption and C_0 is the initial concentration.

Second order,

$$t/q = 1 / \left(k_2 \times q^2_e\right) + t / q_e \tag{11}$$

where k_2 is the rate constant of the second-order adsorption.

The pseudo-first order,

$$d_q/d_t = K_1 (q_e - q_t) \tag{12}$$

where K_1 (min⁻¹) is the constant rate of adsorption, q_t and q_e are the adsorbed amounts at a given time t and at equilibrium (mg/g) respectively.

The pseudo-second order,

$$1 / q_t = 1 / (k_2 \times q^2_e) + t / q_e \tag{13}$$

Moreover, isotherm models, Langmuir and Frendluich, were established the mechanism of adsorption process. These two models can be generally expressed as [45];

The Langmuir isoterm,

$$C_e/q_e = 1/b + (q_0/b) \times C_e) \tag{14}$$

where b (L/mg) is the binding constant and q_0 (mg/g) refers the maximum adsorption capacity, evaluated by potting the C_e/q_e against C_e.

The Freundlich isoterm,

$$\ln q_e = \ln K_f + 1/n \ln C_e \tag{15}$$

where K_f and n are the constants which give adsorption capacity and intensity respectively.

Coefficient of determination (R^2) was used to choose the most accurate kinetic & isotherm model.

3. Results and discussion

3.1. Effect of electrode combination

The chemical reactions that occur in EC process are given in Eqs. (1)–(5). In this study, electrode combinations as Fe-Al, Fe-Fe, Fe-SS, Al-Al, Al-Fe and Al-SS were studied to determine optimum electrode pairs. The results are shown in Fig. 2. As can be seen from the removal efficiency results, there is a high effect of the electrode choice on the removal of phosphate from aqueous solutions. Even after a change in cathode choice with using the same anode, the difference between the phosphate removal efficiencies is about 30% (Fe-Fe, Fe-SS).

The phosphate removal efficiencies were 86.5%, 97.7%, 84.2%, 74.2%, 74.8% and 95.3% with electrode pairs of Al-SS, Fe-SS, Al-Fe, Fe-Al, Fe-Fe, Al-Al, respectively. The sacrificed Fe-SS combination supplied the highest phosphate removal efficiency. This result is reasonable because iron has higher oxidation potential compared with the other electrodes and can produce more bubbles than aluminum [48]. Therefore,
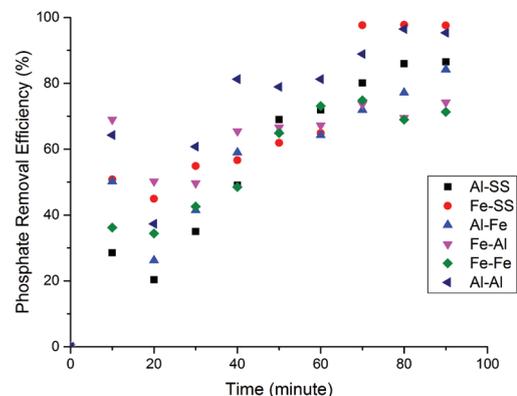
Fig. 2. Effect of electrode combination on phosphate removal via time where initial concentration, 100 mg/L; pH, 7; electrode distance, 20 mm; and current density, 10 mA/cm².

rest of the experiments was carried out with using Fe-SS electrodes.

3.2. Effect of pH

Many researchers have reported that EC efficiency is remarkably sensitive to the initial pH value [49,50]. Therefore, the effect of pH on the phosphate removal efficiency has been investigated with different initial pH values (3, 5, 7 and 9). Fig. 3 shows the effect of pH on phosphate removal efficiency. It can be said that neutral medium provides highest removal efficiency that exceeds 97%.

The lowest removal efficiency was obtained with pH 9. This can be easily explained with magnetic attraction and repulsion. It is more likely to produce positive charged coagulants in neutral or slightly acidic pH [21,51]. Other studies also reported the lower efficiencies in basic pH in EC process [52].

3.3. Effect of current density

Rate of bubble properties and coagulant dosage are both depend on current density [53,54]. Therefore, it is one of the most effective parameter in EC process optimization. Several electrocoagulation experiments were carried out in the current density range of 3.0–15.0 mA/cm². It can be seen from Fig. 4. phosphate removal efficiency varies from 60% for 3 mA/cm² to 97% for 9 mA/cm². However, removal efficiency was decreased when the applied current increased from 9 to 15 mA/cm². These variable results were also obtained in different studies which were carried out with iron electrodes [29,55]. On the other hand, determining endpoint of the highest current density with the highest removal efficiency is good for cost calculation. It is a known fact that power consumption increases as the current density increases.

3.4. Effect of inter-electrode distance

All of the studies conducted so far have proven that inter-electrode distance determines the increase of coagulants and energy consumption [2,56,57]. Therefore, optimizations of inter-electrode distance experiments were carried out for the distance of 10, 20 and 30 mm. Obtained results were given in Fig. 5. After 70 mins of treatment, the phosphate removal efficiency increased from 77% to 97% when the distance decreased from 30 to 20 mm.

3.5. Effect of initial phosphate concentration

The removal efficiencies of different initial phosphate concentrations (25, 50, 100, 200 and 250 mg/L) from aqueous solutions were investigated under optimum removal conditions determined in previous experiments.

After 20 mins of treatment, over 99% phosphate removal efficiency has been achieved in 25 and 50 ppm concentrations (Fig. 6.). However, the phosphate removal efficiency was decreased when initial phosphate concentration was increased. This could be because of that, the generated flocs are insufficient to remove all the phosphate ions at higher concentrations (>50 ppm). It can be said that, it requires more reaction time to treat higher initial concentrations. This statement is supported by literature studies [58].
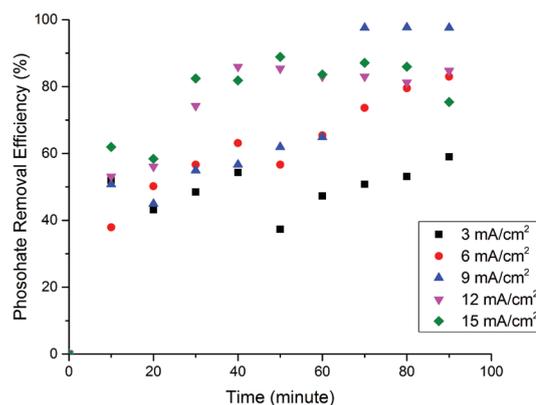
Fig. 4. Effect of current density on phosphate removal via time where initial concentration, 100 mg/L; electrode pair, Fe-SS; pH, 7; and electrode distance, 20 mm.

Fig. 3. Effect of pH on phosphate removal via time where initial concentration, 100 mg/L; electrode pair, Fe-SS; electrode distance, 20 mm; and current density, 10 mA/cm².
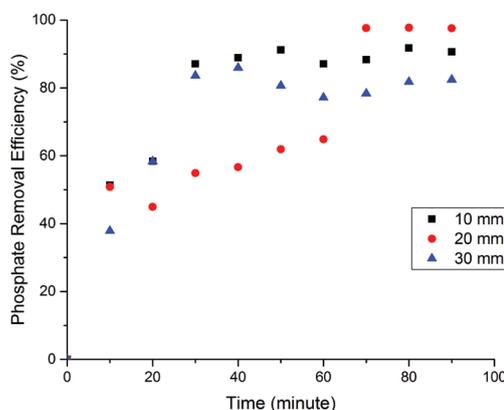
Fig. 5. Effect of inter-electrode distance on phosphate removal via time where initial concentration, 100 mg/L; electrode pair, Fe-SS; pH, 7; and current density, 10 mA/cm².

3.6. Operating cost of the electrocoagulation process

In the end of the optimization study parameters for optimum phosphate removal were determined as 10 min reaction time; pH 7, electrode combination of Fe-SS;
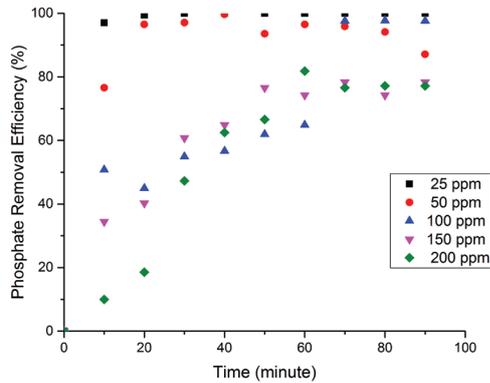
Fig. 6. Effect of initial phosphate concentration on phosphate removal efficiency versus time. (experimental conditions: electrodes, Fe-SS; pH, 7; electrode distance, 20 mm; and current density, 10 mA/cm²).

initial phosphate concentration of 25 ppm; current density of 10 mA/cm², inter electrode distance of 1 cm. The cost analysis was made by using these obtained results and Eqs. (7)–(9) which were given above. Energy and electrode consumptions were determined as 0.96 and 1.54 kg/m³, respectively.

$$\begin{aligned} OC &= 0.96 \text{ kWh/m}^3 \times 0.05 \text{ \$/kWh} \\ &\quad + 1.54 \text{ kg/m}^3 \times 0.58 \text{ \$/kg} \\ &= 0.94 \text{ \$/m}^3 \end{aligned} \tag{16}$$

It was examined that the cost of EC process is quite low when compared with similar studies for phosphate removal technologies from aqueous solutions. Huang and others [11] were studied to recover phosphate ions with chemical coagulation. They obtained that higher efficiency was achieved with iron ions and the removal cost of phosphate was calculated as 1.99 \$/m³.

3.7. Determination of kinetics of phosphate adsorption

Kinetic studies were carried out using the results of the optimization experiments. First order, second order, pseudo first-order and pseudo second-order kinetics were formed. The acceptability of a kinetic model depends on the formation of a linear line, accordingly, it can be seen that the effect of the diffusion of the boundary layer is so remarkable (Figs. 7(a)–(b)).
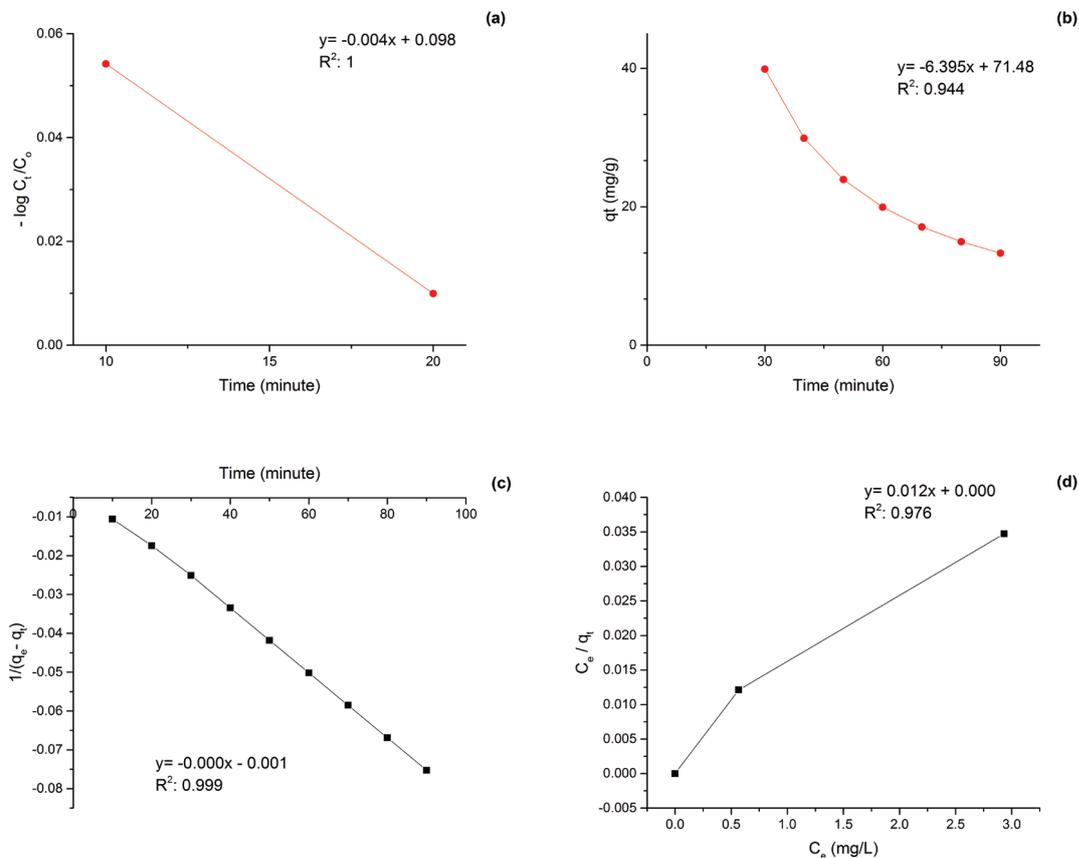
Fig. 7. (a–b) effect of the diffusion of the boundary layer, (c) phosphate adsorption in the second steps corresponds to the pseudo-second-order model and (d) Langmuir isotherm model.

It was also found that, second order and pseudo-second order kinetic were found to be appropriate, but the experimental results showed that the adsorption process did not follow the first order and pseudo first order models. When the obtained results were set increasing order: second order (R^2 : 0.99) > pseudo second order (R^2 : 0.95), it can be said that second order model is better fitted (Fig 7(c)). The Freudlich and Langmuir isotherm models were considered to analyze adsorption experiments. As seen from the high correlation value, the reactions are consisted with Langmuir isotherm model; however negative values for the Freudlich isotherm show the inadequacy of the isotherm models in explaining the adsorption process (Fig 7(d)).

4. Conclusion

In this paper, removal efficiency of phosphate ions was investigated from aqueous solutions by EC process. Electrode combination (Al-Fe-SS), pH (3–9), current (0.15–0.75 A), electrode distance (1–3 cm), initial phosphate concentration (25–200 ppm) and retention time (10–90) were studied as optimization parameters. Removal efficiency was achieved as 100% at pH 7.0, current density of 9mA/cm², electrode combination of Fe-SS, concentration of 20 ppm and reaction time of 20 min. It can be seen that each parameter can change the results between 20% and 40%. For this reason, such a detailed optimization study is important to decide applicability of EC process for phosphate removal. The results also showed that EC process is more efficient in phosphate removal at neutral pH levels. Cost analyze of the electrocoagulation process at optimum conditions was calculated as 0.94 \$/m³. Therefore, it can be said that EC is an economically sustainable treatment process for the removal of phosphate from aqueous solutions. The obtained results also proved that second-order kinetic model and the Langmuir isotherm model are correlated with electrocoagulation data.

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