

A Comparison Between Aluminum and Iron Electrodes on Removal of Phosphate from Aqueous Solutions by Electrocoagulation Process

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ABSTRACT: This study presents a comparison between aluminum and iron plate electrodes on phosphate removal from aqueous solutions by electrocoagulation process. The effectiveness of main operational parameters such as initial pH, initial phosphate concentration and current density were examined. In addition, treated solution and sludge characteristics, energy consumption and electrode mass depletion were compared for both electrodes. All experiments were done in an electrocoagulation cell with effective volume of 2 liters. It was found that the aluminum electrode has higher removal efficiency compared to the iron electrode in the same operational conditions. Phosphate removal efficiencies of 100% and 84.7% were observed for aluminum and iron electrode in a special case with optimum initial pH, maximum current density and maximum initial phosphate concentration equal to 3, 250A/m², 400mg/l PO₄³⁻, respectively. Furthermore, less treated solution turbidity, higher secondary pollution (remained Al) and less electrode mass depletion was observed for the aluminum electrode.

Key words: Electrochemical Process, Removal efficiency, Operational parameters, Different electrodes, Solution characteristics

INTRODUCTION

Modern technologies have been introduced for removing specific elements from different kinds of waste during recent years (Rajasimman and Karthikeyan, 2009, Nouri *et al.*, 2010, Rahmani *et al.*, 2010, Naim *et al.*, 2010, Tashauoei *et al.*, 2010). Presence of excessive amounts of various anions such as phosphate may cause problems in water resources (Peleka and Delianny, 2009). The main forms of phosphate in water bodies are orthophosphate, polyphosphate and organic phosphate (Caravelli *et al.*, 2010). As a main nutrient, phosphate is very essential for growth of organisms in most ecosystems, but excessive phosphate concentrations release to confined water bodies lead to eutrophication and subsequent deterioration of water quality mainly through oxygen depletion as the algae decay (Zhang *et al.*, 2009; Zhao *et al.*, 2009) Therefore, phosphate content of effluents or streams that are discharged directly to the environment is regulated by the national and international wastewater standard authorities with maximum limits ranging from 0.1 to 2mg/l as P (Kamiyango *et al.*, 2009). For example the US,

India and Iran discharge limits of phosphate are (0.5-1), 5 and 6 mg/l as P respectively (Vasudevan *et al.*, 2009; Iranian environmental protection regulations & standards, 2004).

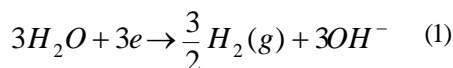
Phosphate removal has received considerable attention since the late 1960s (Peleka and Delianny, 2009; Vasudevan *et al.*, 2009). Various technologies were used for phosphate removal from wastewater that divided into physical, chemical and biological methods (Hosni *et al.*, 2007; Huang *et al.*, 2008; Kamiyango *et al.*, 2009; Peleka and Delianny 2009; Vasudevan *et al.*, 2009; Zhang *et al.*, 2009; Zhao *et al.*, 2009; Caravelli *et al.*, 2010). Physical methods are usually too expensive, as in the case electrodialysis or reverse osmosis (Zhao *et al.*, 2009). In a biological treatment plant, it is necessary to transfer phosphate from liquid to sludge phase, removal efficiency usually doesn't exceed 30%, which means that the remaining phosphate should be removed by other technologies (Hosni *et al.*, 2007). Nowadays, chemical processes are also less attractive due to disadvantages such as further cost for buying and installation of dosing equipments, problems of

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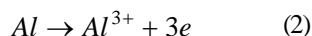
sludge handling and disposal, maintenance costs and chemical used (Vasudevan *et al.*, 2009; Kim *et al.*, 2010).

Another method that has attracted great attention in wastewater treatment is electrocoagulation (EC). EC has been successfully used to remove different kinds of pollutants such as organic compounds (Wang *et al.*, 2010), dyes and textile wastewater (He *et al.*, 2007; Phalakornkule *et al.*, 2010), heavy metals (Zaroual *et al.*, 2009; Zongo *et al.*, 2009; Nouri *et al.*, 2010; Sanjeev Kumar and Goel, 2010; Shafaei *et al.*, 2010), dairy wastewater (Tchamango *et al.*, 2010), biodiesel wastewater (Chavalparit and Ongwandee, 2009), hardness (Malakootian *et al.*, 2010), petroleum refinery wastewater (El-Naas *et al.*, 2009), COD (Wang *et al.*, 2009), important anions such as nitrate (Emamjomeh *et al.*, 2009), fluoride (Ghosh *et al.*, 2008; Zuo *et al.*, 2008) and phosphate (Bektas *et al.*, 2004; Irdemez *et al.*, 2006(a, b, c); Yu *et al.*, 2006; Vasudevan *et al.*, 2009; Kobya *et al.*, 2010). In this process, passing direct current through sacrificial anodes (usually aluminum or iron) causes corrosion of electrodes and production of metal cations. These cations then form polymeric metal hydroxide species in solution similar to coagulant salts like alum in conventional chemical coagulation. These cations and other charged polymeric metal hydroxide species can neutralize negatively charged particles. After neutralization, the particles bind together to form aggregates of flocs, resulting in pollutant removal. Additionally, generation of gas bubbles at both electrodes cause electroflotation, which results in better removal of contaminants (Arroyo *et al.*, 2009; El-Naas *et al.*, 2009; Linares-Hernandez *et al.*, 2009; Wang *et al.*, 2009; Zaroual *et al.*, 2009; Phalakornkule *et al.*, 2010; Sanjeev Kumar and Goel, 2010). The main cathodic and anodic reactions for aluminum and iron are as follows:

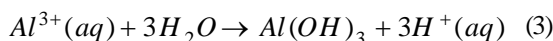
At the cathode:



At the anode:

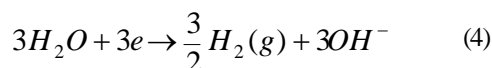


In the solution:

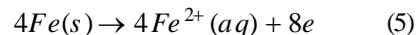


And for the iron electrodes

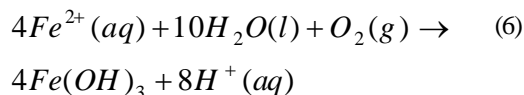
At the cathode:



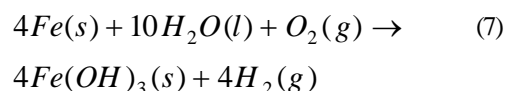
At the anode:



And with dissolve oxygen in solution:



Overall reaction:



EC process like other treatment methods has some advantages and disadvantages. Main advantages of EC are: no requirement for additional chemicals, less sludge production, compact equipments, minimum secondary pollutions and ease of operation (El-Naas *et al.*, 2009; Zaroual *et al.*, 2009; Phalakornkule *et al.*, 2010; Sanjeev Kumar and Goel, 2010). On the other hand, 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 (Zaroual *et al.*, 2009; Sanjeev Kumar and Goel, 2010). The aim of this study is to compare aluminum and iron plate electrodes comprehensively on phosphate removal by EC process. For this purpose, the effects of main parameters like initial pH (pH_i), initial phosphate concentration (C₀) and current density (A/m²) were compared for both electrodes. Additionally, treated solution and sludge characteristics, energy consumption and depletion of anodic metal mass were checked.

MATERIALS & METHODS

The electrocoagulation cell used in this study is schematically shown in Fig. 1. Net volume and effective volume of batch cell is 3.5 and 2 liters, respectively. Four plate electrodes (aluminum or iron) with total effective area of 240 cm² were used. Thicknesses of aluminum and iron plates were 3 mm and 2mm, respectively. The distance between electrodes was kept constant at 3cm. Electrodes were connected to a DC power supply (Micro, PW4053R, 0-5A, 0-40V) in a monopolar mode. The XRF analysis of both electrodes is shown in Table 1.

Phosphate solutions were prepared synthetically by dissolving proper amounts of KH₂PO₄ (Merck, 99.5%) and NaCl (prolabo, 99.5%) as supporting

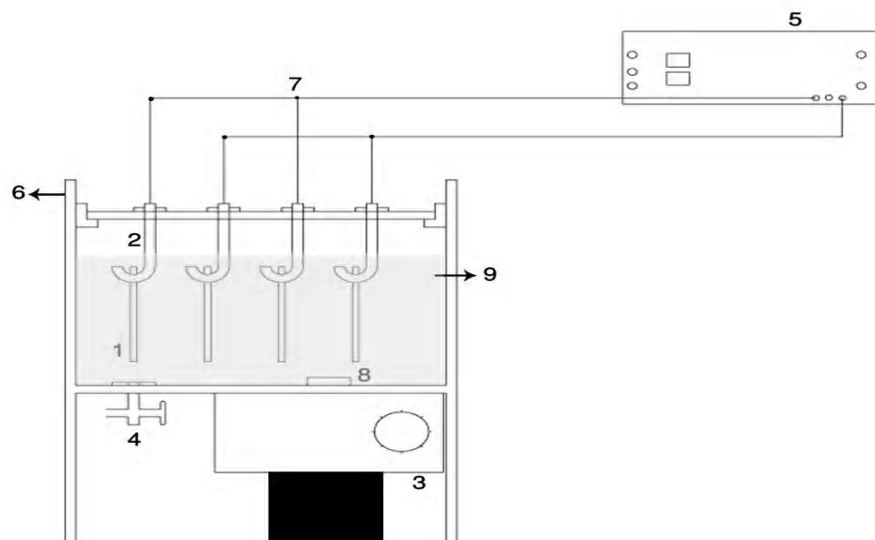


Fig. 1. Schematic diagram of EC cell used in this study 1- aluminum or iron plate electrodes 2- copper hook 3- magnetic stirrer 4- release valve 5- DC power supply 6- plexiglass vessel 7- cables 8- magnet 9- electrolyte

Table 1. XRF analysis of electrodes (wt %)

Elements	Electrode types		Elements (Cont.)	Electrode types	
	Aluminum	Iron		Aluminum	Iron
Al	98.67	-	S	0.029	0.031
Fe	0.36	95.63	Ca	0.014	0.021
Mg	0.024	0.5	Ni	-	0.067
Si	0.88	-	Pb	-	1.23
Cr	-	0.27	Bi	-	1.38
Mn	0.014	0.77	Cu	-	0.016
P	0.0058	0.011	Mo	-	0.026

electrolyte in distilled water. Initial pH of solution was adjusted using HCl and NaOH (1N, 5N) before experiments. For preparing a complete mix solution a magnetic stirrer (VELP, Scientifica, Italy) was used.

pH of solution was measured using pH meter (340i, WTW, Germany). Samples were taken over selected periods of time (5, 10, 20, 40 min) from the reactor and were filtered by cellulose acetate membrane filters with the pore diameter of 0.45µm (Gema medical, Spain) and then analyzed. To prevent passivation of electrodes, cathode and anode were changed every 10 minutes during experiments. All experiments were accomplished at room temperature. The analysis of phosphate was carried out using the amino acid method by a UV-visible spectrophotometer (HACH, DR4000, USA) adopted from the standard methods for examination of water and wastewater (APHA, AWWA, WPCF, 1992). Treated solution turbidity was measured after 1 hour sedimentation by a turbidity meter (HACH, 2100N, USA). Remained aluminum and iron in the treated solution (for a special case) were measured with

aluminum and ferover methods, respectively, using a UV-visible spectrophotometer (HACH, DR4000, USA). For determination of produced sludge constituents, XRF analyses were done using X'Unique II (Philips, Netherlands).

RESULTS & DISCUSSIONS

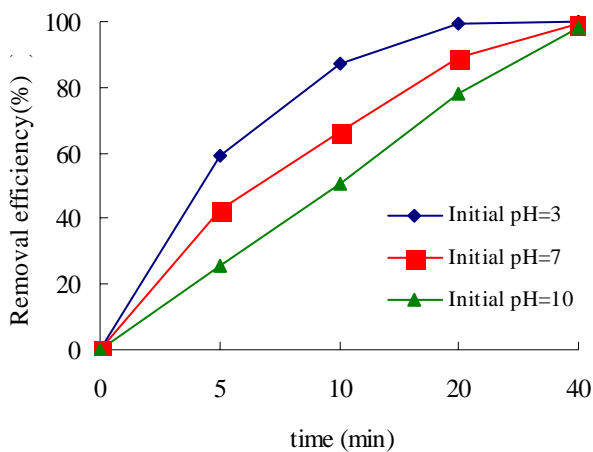
Electrocoagulation is a complex process that may be affected by different parameters such as electrode material, electrode connection mode, initial pH, initial pollutant concentration, current density, applied potential, supporting electrolyte, coexisting ions. In this study, aluminum and iron plate electrodes were used at the same operational conditions (initial pH, current density, initial phosphate concentration (C₀), supporting electrolyte and time intervals) to compare their performance on EC process for phosphate removal. For this purpose, phosphate removal efficiency, turbidity of treated solution and electrode weight depletion were investigated. Overall results for aluminum and iron plate electrodes are illustrated in Tables 2 and 3.

Table 2. Overall results for aluminum plate electrode

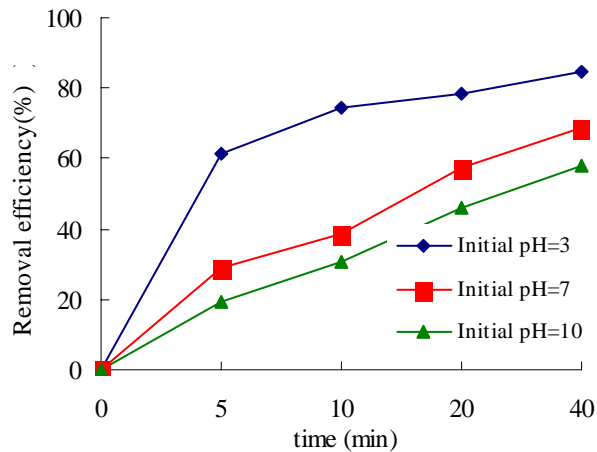
pH _i	Current density (A/m ²)	C ₀ (mg/l)	Phosphate Removal Efficiency (%)				pH of treated solution	Turbidity of treated solution (NTU)	Electrode mass depletion (gr)
			time (min)						
			5	10	20	40			
3	83.33	25	97.6	99.6	100	100	7.98	4.73	0.644
		100	98.8	100	100	100	8.01	0.52	0.622
		400	27.7	50.05	84.05	98.2	9.03	1.81	0.632
	166.66	25	100	100	100	100	8.37	3.3	1.201
		100	100	100	100	100	8.65	0.42	1.161
		400	35.05	60.05	87	98.15	9.17	0.435	0.876
		25	100	100	100	100	8.47	5.15	1.711
		100	99.8	100	100	100	8.73	1.92	1.897
		400	59.35	87.15	99.15	100	9.03	0.78	1.503
7	250	25	94	98.8	100	100	8.55	0.44	0.625
		100	56.2	84.8	100	100	9.06	1.18	0.674
		400	23.55	41	63.25	87.6	9.51	1.33	0.669
	166.66	25	100	100	100	100	8.93	0.407	1.153
		100	68.2	97	100	100	9.17	2.18	1.136
		400	27.2	41.9	65.9	92.35	9.67	1.7	0.955
		25	100	100	100	100	8.81	0.53	1.825
		100	87.8	100	100	100	9	0.95	1.753
		400	42.95	66.2	89.25	99.5	9.5	0.53	1.709
10	83.33	25	82	96	99.6	100	8.91	0.96	0.67
		100	39	64.4	95.8	100	9.25	0.22	0.662
		400	16.1	23.35	41.75	64.7	9.7	26.1	0.56
	166.66	25	100	100	100	100	8.99	1.63	1.212
		100	54.2	84.6	100	100	9.25	0.28	1.135
		400	18.4	32.25	58.25	89.5	9.71	1.54	1.083
		25	100	100	100	100	8.66	0.88	1.758
		100	82	100	100	100	9	0.78	1.796
		400	25.55	50.6	78.3	98.05	9.6	1.07	1.621

Initial pH is an important parameter which affects electrocoagulation performance (Irdemez *et al.*, 2006a). To investigate the influence of initial pH on phosphate removal, the initial pH of solutions was adjusted to 3

(acidic condition), 7 (neutral condition) and 10 (alkaline condition). Phosphate removal efficiency versus time and initial pH is shown in Fig. 2 (a,b) for aluminum and iron plate electrodes.



(a) Aluminum plate electrode



(b) Iron plate electrode

Fig. 2. Phosphate removal efficiency versus time and initial pH (C₀ = 400mg/l, Current density = 250 A/m²)

Table 3. Overall results for iron plate electrode

pH _i	Current density (A/m ²)	C ₀ (mg/l)	Phosphate Removal Efficiency (%)				pH of treated solution	Turbidity of treated solution (NTU)	Electrode mass depletion (gr)
			time (min)						
			5	10	20	40			
3	83.33	25	100	100	100	100	7.2	95.8	0.75
		100	56.2	95.8	100	100	8.78	26.8	0.768
		400	14.3	34.4	60.05	79	10.48	28.7	0.77
	166.66	25	100	99.2	99.2	98.8	8.39	117	1.537
		100	94	100	100	100	9.3	29.2	1.516
		400	39.35	67.2	77.05	80.75	10.93	22.5	1.526
	250	25	100	100	100	100	9.6	80.3	2.305
		100	99	93.6	89	84.6	10.26	10.3	2.394
		400	61.3	74.25	78.4	84.7	10.7	2.33	2.062
7	83.33	25	65.6	71.2	99.6	100	10.5	21.1	0.718
		100	23.8	42.6	76	87.8	10.87	25.5	0.727
		400	15.85	32.05	35.95	54.55	11.13	7.3	0.712
	166.66	25	78.8	91.6	94.8	96	10.57	36.8	1.5
		100	48.2	84.2	92	94	10.96	21.2	1.413
		400	30.9	34.4	47.35	58.55	11.2	2.55	1.447
	250	25	91.6	98	98.4	96.4	10.6	20.4	2.142
		100	55	78.2	75.2	74	10.81	3.58	2.234
		400	28.8	38.8	57.45	68.95	11.2	11.4	2.198
10	83.33	25	48.4	73.6	99.2	100	10.75	27.1	0.81
		100	21.4	27.8	66.4	86.8	10.67	24.8	0.743
		400	13.2	17.15	22.25	41	11.27	15.7	0.71
	166.66	25	66	90.8	95.2	96	10.66	25	1.503
		100	40	71.8	95.4	99.8	11.05	24.2	1.479
		400	16.55	24.9	39.5	47.6	11.2	8.05	1.383
	250	25	80.4	91.2	94.8	91.2	10.68	7.6	2.132
		100	45.6	65.4	65.4	66.4	10.9	3.91	2.263
		400	19.55	30.4	46.2	57.95	11.23	1	2.271

As it is shown in Fig. 2 (a,b), phosphate removal is completely depends on the initial pH value and at the lowest initial pH value, phosphate removal efficiencies were higher for both kinds of electrodes. Maximum removal efficiencies (100%, 84.7%) obtained at pH_i 3 for aluminum and iron electrodes, respectively. The effect of pH on the process performance was explained as follows: At acidic pH, the oxide surfaces exhibited a net positive charge, and adsorption of anionic phosphate was enhanced by columbic attraction. At higher pH, the oxide surface had a net negative charge and would tend to repulse the anionic phosphate in the solution (Kobyta *et al.*, 2010). Irdemez study groups also demonstrated that optimum initial pH value is 3 when pH has not been controlled (Irdemez *et al.*, 2006a, c) whereas Vasudevan study group and Bektas study group have reported that phosphate removal reached to its maximum values at initial pH 7 and pH 6 respectively (Bektas *et al.*, 2004; Vasudevan *et al.*, 2009).

Variation of phosphate removal efficiency versus time at different initial phosphate concentrations (25,100,400 mg/l PO₄³⁻) using aluminum and iron plate electrodes were also compared. Phosphate removal efficiencies versus time are compared in Fig. 3(a, b) at three initial concentration for constant initial pH(3) and current density (250 A/m²).

As seen from Fig. 3 (a, b), at the highest initial concentration (400mg/l), phosphate removal efficiencies were 61.3%, 59.35 % for iron and aluminum electrodes, respectively for the reaction time of 5 min. At this initial concentration of phosphate, additional time is necessary to obtain higher removal efficiencies, while at lower initial concentrations (25, 100 mg/l), at the first time interval (5 minute), the removal efficiencies reached to their maximum values and after that (except for iron plate electrode at C₀ 100 mg/l) did not change significantly. The reason of this phenomenon is limited adsorption capacity of metallic hydroxide flocs at higher phosphate concentrations.

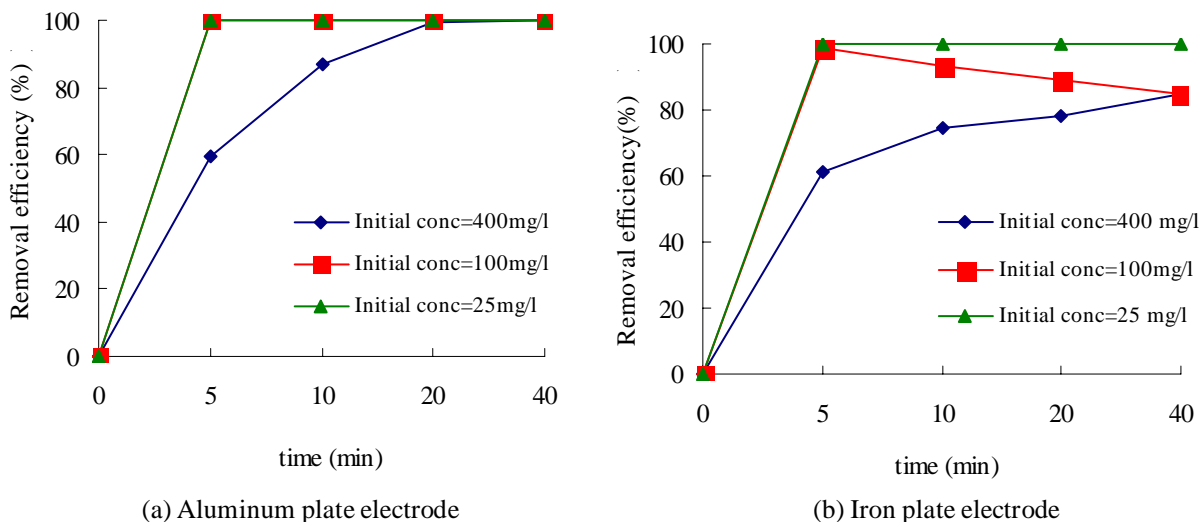


Fig. 3. Phosphate removal efficiency versus time and initial concentration (initial pH=3, current density=250 A/m²)

Trend of removal efficiencies versus time at different initial phosphate concentrations in this study is almost similar to the obtained results of Bektas study group (Bektas *et al.*, 2004).

Another parameter that affects electrocoagulation process is current density because it has direct relationship to the amount of aluminum or iron released to the solution according to the Faradays law. To evaluate the effect of current density on phosphate removal efficiency, three different currents (1, 2, 3A) that are producing current densities of 83.33A/m², 166.66A/m², 250A/m² were examined. Phosphate removal efficiency versus time and current density for

a selected operational condition (initial pH 3, initial phosphate concentration of 400mg/l PO₄³⁻) is shown in Fig. 4 (a, b).

Maximum removal efficiencies of 98.2%, 98.15%, 100% by aluminum electrode and 79%, 80.75%, 84.7% by iron electrode obtained at current densities of 83.33A/m², 166.66A/m², 250A/m², respectively. It is clear that the increase of current densities enhance the phosphate removal efficiencies for aluminum and iron plate electrodes.

Treated solutions turbidity was measured after 1 hour sedimentation of flocs which are reported in Tables 2 and 3. Treated solutions turbidity resulted

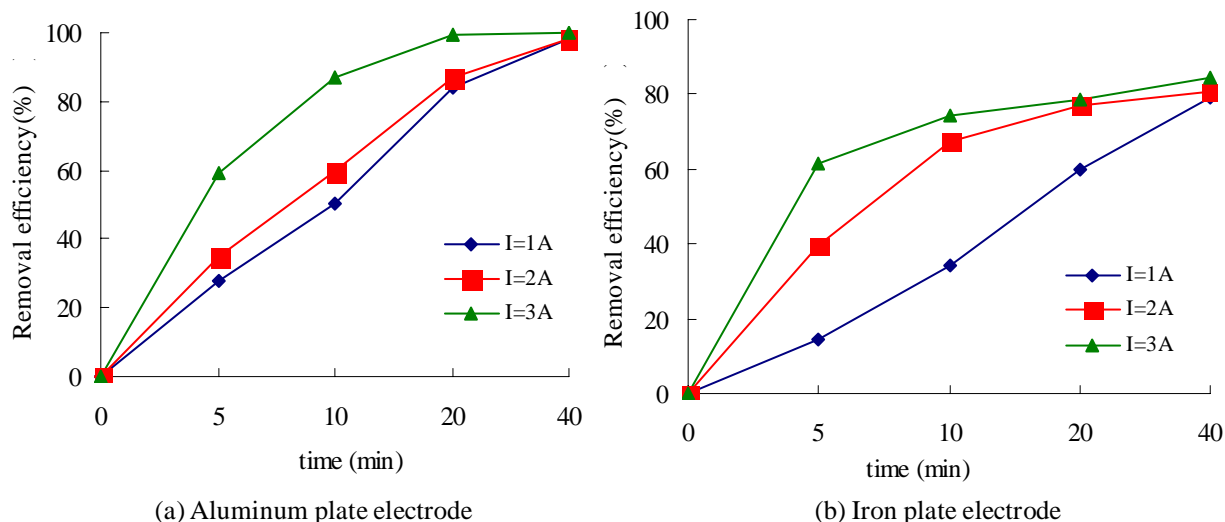


Fig. 4. Phosphate removal efficiency versus time and current values (initial pH=3, initial phosphate concentration=400mg/l PO₄³⁻)

from iron electrode was higher than aluminum electrode. Treated solution turbidity of all cases for aluminum electrodes meet national (Iranian) effluent turbidity standard (50 NTU) (Iranian environmental protection regulations & standards, 2004), while for iron electrodes, 3 cases (out of 27) had higher turbidity than the above-mentioned standard.

Prevention of adding other pollutants to the solution is important during wastewater treatment processes. The amounts of aluminum and iron remained in the treated solution were checked for both electrodes in a special case (initial pH 3, current density of 250A/m² and initial phosphate concentration of 400 mg/l). The remained Fe in treated solution for iron electrode was 2mg/l which is lower than the national (Iranian) effluent discharge standard limit (3 mg/l), whereas the remained Al (for aluminum electrodes) was 14.5 mg/l that is much higher than its allowable amount in the above-mentioned standard (5 mg/l) (Iranian environmental protection regulations & standards, 2004).

Identification of sludge constituents that is produced in each treatment process is an important part of each method evaluation. Therefore the XRF analysis of sludge produced in a special operational condition of (initial pH 3, Initial phosphate concentration of 400mg/l PO₄³⁻ and current density of 250A/m²) is demonstrated for both types of electrodes in Table 4.

As illustrated in Table 4, higher amounts of phosphate found in the sludge of electrocoagulation process using aluminum electrode that confirms its higher removal efficiency.

The energy consumption in electrocoagulation process at a constant voltage and current is calculated by the following equation:

$$E = V \times I \times \int_0^t dt = VIt \quad (8)$$

The amounts of electricity consumption are compared in Table 5 for both electrodes at 40 min reaction time with the current density of 166.66A/m². Total energy at each experiment is calculated by addition of the amount of energy consumed at each time interval. As illustrated in Table 5, the difference between energy consumption for aluminum and iron electrodes is negligible.

In addition, the amounts of electrodes mass depletion for iron and aluminum electrodes were measured and reported in Tables 2 and 3. This mass depletion was calculated by subtracting the weight of the electrodes taken at the end of experiment from the weight before the experiments of the same electrodes. As it is shown, the amounts of electrodes mass depletion for iron electrode were higher than that of aluminum electrode in the same operational conditions.

CONCLUSION

In this study, the effects of main operational parameters like initial pH, initial phosphate concentration and current density were compared for iron and aluminum plate electrodes. The obtained results showed that phosphate removal completely depends on the initial pH and at the lowest initial pH value (3), removal efficiencies were higher for both kinds of electrodes. At the highest initial concentration (400mg/l), the phosphate removal efficiency was 61.3%, 59.35% for iron and aluminum electrodes, respectively for the reaction time of 5 min, initial pH 3 and current density of 250A/m². At this initial concentration of phosphate, additional time is necessary to obtain higher removal efficiencies, while at lower initial concentrations (25, 100 mg/l), at the first time interval (5 minute), the removal efficiencies reached to their maximum values. The increase of current density improves the efficiency of phosphate removal. Maximum removal efficiencies (100%, 84.7%) obtained at initial pH 3, initial phosphate concentration of 400mg/l PO₄³⁻, and current density of 250A/m² for aluminum and iron electrodes, respectively.

Table 4. XRF analysis of produced sludge in electrocoagulation using aluminum and iron electrodes (%)

Constituents	Electrode types		Constituents (Cont.)	Electrode types	
	Aluminum	Iron		Aluminum	Iron
Al	28.1	-	Mn	0.035	0.57
Fe	0.38	67.33	Mg	0.23	0.72
PO ₄	13.8	9.1	Ca	0.15	0.16
Na	13.82	7.75	K	2.01	0.39
Cl	39	12.8	Cu	2.22	0.82

Table 5. Energy consumption in electrocoagulation using aluminum and iron electrodes (current density 166.66A/m²)

Electrode type	pH _i	C ₀ (mg/l)	Voltage(v)				W _T (kwh)
			time (min)				
			5	10	20	40	
Aluminum	3	25	6.3	6	6	5.8	7.92*10 ⁻³
		100	6.5	6.4	6.3	6	8.25*10 ⁻³
		400	6.5	6.4	6.3	6.1	8.32*10 ⁻³
	7	25	6.5	6.3	6.2	6	8.2*10 ⁻³
		100	6.2	6.2	6.2	6	8.13*10 ⁻³
		400	6.5	6.4	6.1	5.9	8.12*10 ⁻³
Iron	10	25	6.4	6.3	6.1	5.9	8.08*10 ⁻³
		100	6.1	5.9	5.9	5.8	7.83*10 ⁻³
		400	6.1	5.9	5.9	5.8	7.83*10 ⁻³
	3	25	6.2	6.2	6.1	5.9	8.03*10 ⁻³
		100	6.4	6.3	6.1	6	8.15*10 ⁻³
		400	6.5	6.4	6.3	6.1	8.32*10 ⁻³
Iron	7	25	6.5	6.3	6.2	6	8.2*10 ⁻³
		100	6.4	6.3	6.1	5.9	8.12*10 ⁻³
		400	6.5	6.4	6.2	6	8.22*10 ⁻³
	10	25	6.5	6.3	6.2	6	8.2*10 ⁻³
		100	6.4	6.2	6.1	5.9	8.07*10 ⁻³
		400	6.5	6.4	6.2	6	8.22*10 ⁻³

In addition, treated solution turbidity, secondary pollution and sludge characteristics of both electrodes were compared. The results showed that the treated solution turbidity for iron electrode was higher than that of aluminum electrode. The amounts of aluminum and iron remained in the treated solution were 2 and 14.5 mg/l for aluminum and iron electrodes respectively, in a special case (initial pH 3, current density of 250A/m² and initial phosphate concentration of 400 mg/l). Higher amount of phosphate found in the sludge of electrocoagulation process for aluminum electrode that confirms its higher removal efficiency.

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