Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2017.21475

99 (2017) 282–288
December

Mucilage of *Plantago ovata* as natural coagulation–flocculation aid in an electrocoagulation process for phosphate removal from aqueous environments

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Received 1 November 2016; Accepted 22 September 2017

ABSTRACT

The aim of this study was investigating the phosphate removal from aqueous environments by electrocoagulation (EC) in bipolar connection mode. In the study, iron plates were used as electrode with mucilage of *Plantago ovata* as a natural aid-coagulant and aid-flocculent. The several operational parameters such as the concentrations of phosphate, pH, operating time on the removal efficiency of phosphate were determined. The results showed that in the operating removal condition when mucilage was added as aid coagulant, the removal efficiency was improved. In this condition, the formed flocs in EC easily floated on surface of the solution. The process is completely effective in low concentration (100% removal in 5 mg/L phosphate). The sludge formed in EC with mucilage could be used to improve poor soils with low iron, phosphorus and organic matters.

Keywords: Electrocoagulation; Mucilage; *Plantago ovata*; Phosphate; Aid coagulants

1. Introduction

As stewards of our environment, we are responsible for the protection of the environment, for our own sakes and also in favor of next generations. Indeed, the sustainable growth of our civilization as the only reasonable and feasible way requires coexistence with nature on our planet that we protect our fragile environment [1]. Furthermore, phosphorus compounds from wastewater have an important role in managing the environmental and economic concerns such as eutrophication of surface waters and reduction of phosphorus resources. Eutrophication could lead to plentiful development of aquatic plants, dramatic growth of algae and upset the balance of the water bodies by depletion of oxygen level because of high biological oxygen demands, and acidification as well as it causes reductions in biodiversity [2]. Furthermore, severe standard legislations are followed for phosphorus discharge range in water and wastewater such as 6 mgP/L in Iran [3], 0.5 mgP/L in China [4], 0.5–1 mgP/L in US and 5 mgP/L in Indian [5]. Several techniques have been extensively investigated for phosphorus removal from wastewater, including adsorption, chemical precipitation, physical and biological processes [6–8]. Biological process is widely used at the industrial scale as a cost-effective and environmentally method alternative for the chemical precipitation treatment, but its effectiveness could become progressively worse by avoiding fewer fluctuations in operating conditions [9] and inefficiency at low phosphate concentrations [10]. Physical methods are too expensive and inefficient [2,11]. The aluminum sulfate and ferric chloride are the common chemicals used for chemical processes that are widely used for phosphate removal, but chemical treatments are unfavorable due to high cost of preservation, problems of disposal and handling sludge, and neutralization of the effluent [11]. The high cost of activated carbon limits its use as an adsorbent also it needs to chemical consumption and

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temperatures for activation [12]. Some adsorbents are low efficiency or low capacity for adsorbate in practice [12,13]. Additionally, the most important industrial adsorbents are oxygen-containing compounds such as silica gel, activated alumina, zeolites and molecular sieves, clay and pillared clays with hydrophilic properties, and adsorbents with carbon as the major component such as activated carbon, activated coke, carbon molecular sieves, carbon nanotubes and polymeric resins, which are hydrophobic in nature. In fact, selecting an adsorbent from a practical point of view is limited by these criteria: capacity of the sorbent, selectivity, regeneration method, length of unused bed (for fixed bed systems) and price. In addition, chemical resistance and thermal stability are important in application adsorbents [14]. Coagulation process is a highly efficient method in removing phosphate. It is regarded as a vital option in tertiary treatment of wastewater that is proposed for prevention of eutrophication. Electrochemical coagulation is one of the electrochemistry techniques. Metallic ions generated electrochemically from electrodes (usually aluminum or iron plates) could produce a series of activated intermediates near the anode which could upset the stability of the finely dispersed particles present in the water and wastewater. The destabilized particles then aggregate to form flocs that can float [15]. The advantages of electrocoagulation (EC) include a compacted treatment facility, simple operating condition with minimum requirement of chemicals, high particulate removal efficiency, relatively low-cost sludge and ease of operation [11,16].In addition, the EC process is easily managed under high temperature because at higher temperatures, the dissolution of anode is more better thus amount of formation of hydroxide is very high for complex with phosphate ions [11].

Although there are several reports that are related with usage of EC for removal of pollutants (i.e., chromium(VI) [17], boron [18], cadmium [19], treatment of textile wastewater [20] and phosphate [15,19,21–23]), they are focused on optimizing of process parameters for improving the removal efficiency by change in parameter values. The coupling of EC with adsorption was reported earlier in the removal of methylene blue using banana peel [16] but the main aim of the present work was to investigate the efficiency of coupling the EC with natural coagulant.

Lee et al. [24] has classified flocculants into three categories including chemical flocculants (metals salts, polyacrylamide, and polyacrylic acid), natural bio-flocculants (chitosan, cellulose, tannin, etc.), and grafted flocculants. Chemical flocculants are commonly used due to its low cost and ease of use but their applications are limited due to low flocculating efficiency and the presence of residue metal ions in the treated water. The organic polymeric flocculants are significant ability to flocculate efficiently with low dosage. However, it is related to lack of biodegradability and dispersion of monomers residue in water that may affect health. Therefore, natural bio-flocculants have been attracting wide interest of researchers due to their advantages of biodegradability and being environmentally friendly. The use of natural coagulants can not only reduce the requirements of conventional coagulants but could also reduce the cost of overall treatment process by decreasing the amount of chemical coagulant [25].

The mucilage is a safe alternative for conventional polymers in wastewater treatment because of its production process and applications which are environmentally friendly and beneficial to human and ecology. Moreover, it is derived from plant species include *Abelmoschus esculentus* [26], *Plantago ovata* [25], *Ocimum basilicum* [27,28] and cactus [29]. Although these plant-based flocculants are generally obtained through aqueous extraction, precipitation with alcohol and drying [24], mucilage of *P. ovata* can be extracted by grinding of seeds surface. The usage of flocculants enhances the formation of larger flocs from fine colloidal particles formed during EC. These larger flocs settle more rapidly and are easily floated and removed [30].

In this study, seed mucilage of *P. ovata* as a plant-based natural coagulant was used for improvement of coagulation and floating during the EC process. The *P. ovata* are small gray-colored seeds with ≈30% weight husk. Psyllium husk contains about 30% mucilage, mainly xylose, arabinose, and galacturonic acid [31]. The galacturonic acid is generally predominant active coagulation agent that enhances turbidity removal [32]. *P. ovata* seed for pollutant adsorption was used in few studies but it was not implemented for improvement of EC process. In this study, the effect of mucilage on phosphate removal was investigated by EC.

2. Materials and methods

2.1. Mucilage extraction of seed

The seeds of the *P. ovata* plant were collected from Iranshahr (Iran). Pods and impurities were removed from the seeds by sieving and winnowing. Domestic mill was used to separate the psyllium husk from seeds. The mill blades scratch the shell so that the husk could be separated and then sieved by a fine mesh.

2.2. Electrocoagulation reactor

A reactor (Fig. 1) with a 1.5 L glass container below the water surface with four iron plates (4 × 7 cm and total area of 56 cm^2) was used. Sheets of the same size were placed with 2 cm gap of electrodes in the reactor. The solution was stirred on the magnetic stirrer. A regulated direct current (DC) was employed from a precise DC power supply PS-303 model

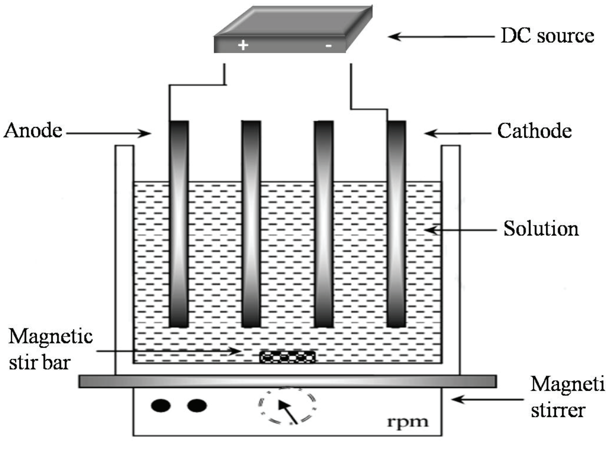

Fig. 1. Laboratory scale reactor for electrocoagulation phosphate solution.

(3.5 A, 0–30 V). Traditionally, the DC source was employed to generate electric field and ions transportation between the sacrificial electrodes immersed in the reactor.

The potassium dihydrogen phosphate (KH_2PO_4) was dissolved in distilled water as the stock solution (1,000 mg/L). Experimental solutions were obtained by diluting the stock solution and 1 L of the solution was used for each experiment. Before the experiments, the pH of the solution was adjusted with 1 M HCl and 1 M NaOH solutions. The solution conductivity was adjusted by adding 1 g of NaCl in 1 L of synthetic solutions. The used chemicals and reagents were of analytical grade (Merck). The phosphate removal efficiency was calculated according to the following equation:

$$\%\ \text{Removal} = \frac{C_0 - C_f}{C_0} \times 100 \qquad (1)$$

where C_0 is the concentration of phosphate before EC (mg/L), and C_f is the concentration of phosphate after a few minutes of EC (mg/L).

Current density j (A/m^2) can be calculated from the current in the EC reactor per effective anode area below the water surface A_{eff} (m^2) [33]:

$$j = \frac{I}{A_{\text{eff}}} \qquad (2)$$

Cost of electrical energy (kWh/m^3) is calculated as Eq. 3, where U is the voltage (V), I is the current (A), t is the time of electrolysis (h) and V is the volume (m^3) of solution [19]:

$$C_{\text{Energy}}\ (\text{kWh/m}^3) = \frac{U \times I \times t}{V} \qquad (3)$$

2.3. Analytical method

The analysis of phosphate was carried out using the ascorbic acid method (4500-P.E) by a spectrophotometer according to the Standard Methods for Examination of Water and Wastewater [34].

3. Results and discussion

3.1. Effect of pH

To investigate the effect of pH, a series of experiments were performed using 100 mg P/L phosphate solutions with an initial pH varying in the range of 3–10. As illustrated in Fig. 2, the removal efficiency of phosphate was decreased with increase of the pH and the maximum removal efficiency was obtained in pH = 3 more than 85% and in range 3–6 approximately was stable (85%). However, an increase was observed in pH = 6 which then decreased. The minimum removal efficiency of phosphate was 55% at pH = 10.

The minimal pH increase (pH increase of less than 0.5) led to a localized solution chemistry at the anode surface favorable to green rust (GR) crystallization [35]. Moreover, it causes the GR in a higher pH to carry less positive surface charge in combination with a more important negative charge for phosphate anions; furthermore, phosphate exists in solution in different forms $H_3PO_4^-$, $H_2PO_4^-$, HPO_4^{2-} and PO_4^{3-} and at different ratios depending on pH. From pH 3 to 6 $H_2PO_4^-$ is dominant form in solution [36]. Two other reasons for this decline could be expressed as follows: (1) increasing the pH value can destroy the passive of iron oxide, which is formed at lower pH. (2) Turbulence generated by the rising gas bubbles can set off the rate of mass transfer at anode and cathode with a finally decrease in the concentration of polarization [37].

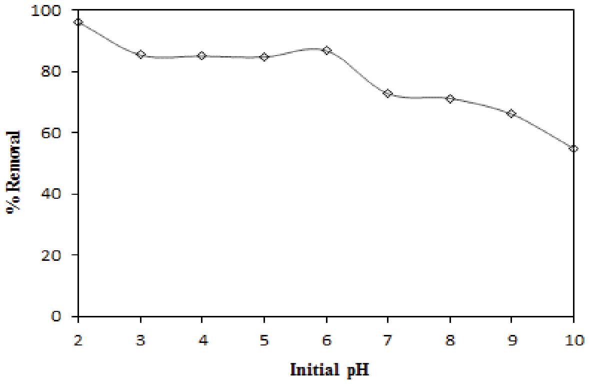

Fig. 2. The effect of pH on phosphate removal (100 mgP/L, 11 mA/cm^2, 30 min).

This decreased trend in line result was observed on phosphate removal in hybrid (Al/Fe) [33] and iron electrodes [37]. The decline in removal efficiency at more acidic and alkaline pH was observed with aluminum alloy and stainless steel as anode and cathode, respectively. In this the experimental set, maximum removal efficiency was obtained at pH 7.0 and the minimum at pH 10 for phosphate [11].

The final pH in all the experiments was augmented after phosphate removal. The rise in pH during EC was mainly attributed to the increase of hydroxide ion concentration in solution resulting from water reduction at cathode (Eq. 4) [38].

$$2H_2O_{(l)} + 2e^- \rightarrow H_{2(g)} + 2OH^- \qquad (4)$$

An initial pH value of 6 was chosen for following experiments; this is close to the usual pH level of wastewater in practice.

3.2. Effect of initial concentration

The effectiveness of variant phosphate concentrations in wastewater was evaluated (5–200 mg/L) on EC process. During these assays, the current density was held at 11 mA/cm^2 over 30 min with pH = 6. The results obtained are illustrated in Fig. 3.

As seen from Fig. 2, the removal rate of phosphate is declined with an increase in phosphate concentration and maximum removal is obtained in low concentration. Percentage of phosphate removal can be similar to result of Tran et al. [38] with an efficiency of 100% in low concentration. In fact, phosphate was removed due to co-precipitation with Fe^{2+} or Fe^{3+} generated by anodic dissolution during electrolysis.

Table 1 shows the changes of parameters on electrical conductivity and pH after phosphate removal in solution. Increased trend in electrical conductivity and pH could be associated with augmentation of Fe concentration during the electrolysis which leads to increase of the percentage of phosphate removal but it is not effective in high rate of concentration sufficiently which may be due to increasing pH in solution. Phosphorus forms can be less adsorbed in pH > 10. Another reason expressed for this behavior is due to the lack of flocs for adsorption of excess phosphate at high concentrations and also due to the lower rates of iron corrosion and increasing iron surface passivation at higher concentrations. The same results were observed for hexavalent chromium that could be explained by Faraday's law, in the constant current density. A constant amount of Fe^{2+} is released into the solution. As a result, the Fe^{2+} ions produced at high initial concentrations are insufficient to reduce all the pollutant ions [17]. Furthermore, in higher initial concentration, the adsorption sites as $Fe(OH)_2$ and/or $Fe(OH)_3$ would block, and its ability is decreased to absorb phosphate from solution [37].

3.3. Influence of the operation time

Operating time is a significant factor in EC process. It is necessary to provide sufficient current to the electrodes where the metal ions are generated by the dissolution to form hydroxyl and metal ions [39,40]. To investigate the effects of operating time on the EC, a solution was used with pH 6 and initial concentration of 100 mgP/L. The impact of the operating time on the phosphate removal is presented in Fig. 4.

As seen from Fig. 3, the phosphate removal increases until a certain operating time value (30 min) and then it remains steady. An increase in the electrolysis duration leads to more generation of the ions resulting in the higher phosphate removal efficiency of the wastewater [40]. Sometimes the removal efficiency does not augmented as sufficient numbers of flocs are available for the removal of the pollutant [20]. Furthermore, removal efficiency does not increase (on the contrary, it decreases) due to the anodic passivation and cathodic polarization which can slow down the performance of EC [39]. The operating time of 30 min was selected for the experiments.

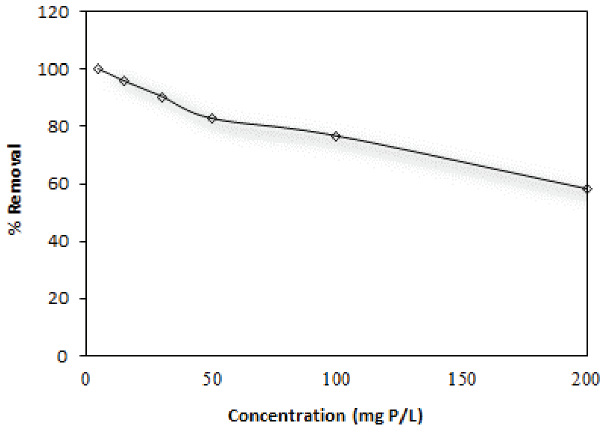

Fig. 3. Effect of initial concentration on phosphate removal (pH = 6, 30 min, 11 mA/cm²).

Table 1
Electrical conductivity (EC) and pH changes after EC with different concentrations of phosphate

Initial concentration	% Removal	Initial pH	Final pH	EC (ms)
5	100.00	6 ± 0.1	8.99	2.45
15	95.93	6 ± 0.1	10.57	2.30
30	90.49	6 ± 0.1	10.7	2.29
50	82.72	6 ± 0.1	10.74	2.43
100	76.58	6 ± 0.1	10.23	2.57
200	58.03	6 ± 0.1	10.81	2.91

3.4. Effect of voltage and mucilage

Current density influences coagulant dosage, bubble formation rate, their size and the flocs growth. It is also an important parameter that impacts on the performance and the economic efficiency of the EC process [16,17]. In order to investigate the effect of current on removal efficiencies, experiments were conducted in two voltages 30 (0.52 A) and 15 (0.27 A).

A decline in current density results in an decrease in the phosphate removal efficiency as the insufficient number of metal hydroxide flocs are available for the decline of the pollutant [20], but energy consumption which is lower than high-current density should be taken into account an economic aspect.

The mucilage of *P. ovata* seeds can be separated easily by scratch unlike seeds of *O. basilicum*. These seeds can be used for pollutants adsorption [43,44]. However, mucilage of *P. ovata* is an anionic polysaccharide [45]; therefore, it cannot remove anionic pollutants such as phosphate (Table 2).

In the EC after completion of the process for a specific electrolysis time, the solution is kept for fixed time (retention time) to allow settlement of the coagulated species. The removal efficiency of pollutant is augmented with increasing

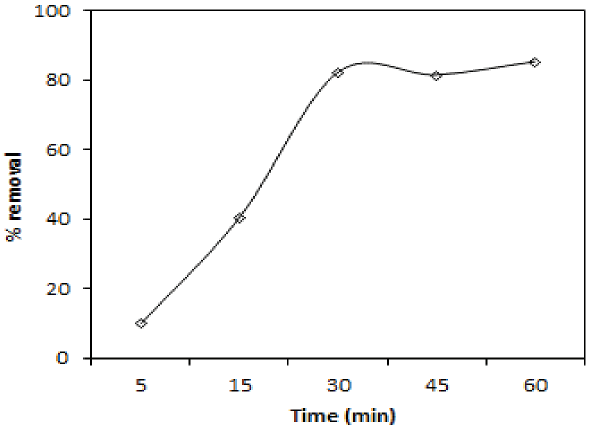

Fig. 4. Influence of the operating time on the removal of phosphate (pH = 6, 100 mgP/L).

the retention time. This is due to the matter that all coagulated species settle down easily to give clear supernatant liquid and the sludge. However, on condition that a retention time more than the optimum retention time results in the reduction of efficiency in the removal of pollutant doe to the adsorbed pollutant desorbs back into the solution [20]. The mucilage of *P. ovata* seed is a natural coagulant that can be suitable for this purpose. In addition, the mucilage is better for floating of flocs. In addition, coagulant aid can contribute to better settling of the coagulated species (Fig. 5). Hence, mucilage of *P. ovata* is a coagulation–flocculation aid.

In most cases, the publications deal with the effect of operating parameters on the removal efficiency of pollutants in water and wastewater (Table 3). The initial pH, mean operating time, nature and type of electrodes, surface area of electrode, initial concentration, supporting electrolyte type, current density, and conductivity are the main parameters that affect the removal efficiency [22,46]. The distance of electrodes is the main parameter. As the distance is augmented, the resistivity of the solution is soared; therefore, the amount of metal dissolved into the solution is also declined [21]. In this study, distance of electrodes is more than the ones stated in other studies but the efficiency is better for which two reasons can be expressed: (1) mucilage of *P. ovata* improved the efficiency of removal in EC; (2) the bipolar mode is more efficient compared with monopolar connections systems for phosphate removal [46–48].

It has been indicated in performed preliminary investigations that aluminum electrodes have the better results compared with iron electrodes [22] but removal

Table 2
The influence of mucilage and voltage on phosphate removal by EC

Electrocoagulation reactor	% Removal	$J = I/A$ (A/m^2)	C_{Energy} (KW/m^3)
With mucilage and electrode	86.99		
Without mucilage with electrode	89.70		
Without electrode with mucilage	−16.86		
Voltage 15	52.07	48.21	2.02
Voltage 30	86.99	92.86	7.80


Fig. 5. EC Reactor with mucilage of *P. ovata.*

Table 3
Review of the studies on phosphate removal by electrocoagulation

References	Type of electrodes	Current density (mA/cm^2)	Gap of electrodes (mm)	Total Area (cm^2)	Operating time	Initial concentration	Volume (mL)	pH	Phosphate removal
44	Six pair of Al	0.5	5	1,500	8	25–150 P–PO4	<850	3	100–42
22	Six pair of Fe	0.75	5	1,500	20	100	<850	3	86
								6	71
								9	64
45	Eight pair of Al	10	3	–	5	10–50 PO_4	500	6.2	81–90
						100–200			50–35
10	Stainless steel as cathode and anode was variable**	20	5	600 × 2^a	30	100	900	7	99 Al alloy
									85 Al
									87 mild steel
20	One pair of Al	2–18	5	–	30	100 $P–PO_4$	–	7	25–80
					140				65–100
This study	Two pair of Fe	11	20	224 × 2	30	5	1,000	6	100
						100			76.5
						200			58

efficiency in this method is acceptable despite the high volume of solution and long distance of electrodes as well as using of iron electrodes. However, these results are very promising for improvement of the efficiency of EC process by natural flocculent–coagulant aid of *P. ovata*.

4. Conclusion

In the current study, the efficiency of EC was investigated for the removal of phosphate from aqueous solution with Fe electrodes. The obtained results showed that EC can be successfully applied for the phosphate removal with and without mucilage. However, mucilage could help improve coagulation, floating of flocs, and reduction in retention time without increase in concentration of phosphate at retention time more than the optimum time. Several parameters were examined to determine the optimal conditions among which EC performs best in phosphate removal. Such parameters which involved in initial phosphate concentrations were applied to current and initial pH of the solution and time. The best result was obtained for iron electrodes placed at 2 cm distance with a current density of 11 mA/cm^2 and initial pH 6. After 30 min of treatment, the total removal reached 100%–83% for initial concentration of 5–50 mg/L. The sludge load with iron and phosphorus on mucilage as natural organic material could be used for improvement of poor soil in agriculture and regeneration of soil in polluted sites by phytoremediation.

Acknowledgments

The research is extracted from an educational project of Soghra Yaghoobi Rahni, in Velayat University, Iranshahr, Iran. The authors wish to thank M. Ramezani (laboratory operator of Gonabad medical university) for supporting this work.

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