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Procedia Engineering 33 (2012) 155 – 162

**Procedia
Engineering**

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ISWEE 11

Treatment of Water Loaded With Orthophosphate by Electrocoagulation

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Abstract

In this study, the effective performance of electrocoagulation process in the treatment of water solution that is loaded with orthophosphate was investigated using sacrificial iron electrodes. Various operating parameters (e.g., pH, current intensity and supporting electrolyte concentration) were studied in an attempt to achieve a higher removal capacity. Results obtained with synthetic wastewater revealed that the most effective removal of orthophosphate could be achieved when the pH was kept between 5 and 8. The optimum concentration of supporting electrolyte was found to be 2g/L, which was adjusted using proper amount of NaCl with the orthophosphate concentration of 10 mg/L. In addition, the increase of current intensity, in the range 0.6–2.0 A, enhanced the treatment rate without affecting the energy consumption. The method was found to be highly efficient and relatively fast compared to conventional existing techniques

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Keywords: Electrocoagulation; orthophosphate; Iron electrode; water

1. Introduction

Many causes of pollution, including sewage and fertilizers, refer to nutrients such as phosphates. In excess levels, nutrients over stimulate the growth of aquatic plants and algae. Excessive growth of these types of organisms consequently clogs the water ways, uses up dissolved oxygen as they decompose, and blocks light to reach deeper waters.

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Phosphorus (P) is a macronutrient necessary for crop growth. Theremoval of Phosphorus is a key issue in wastewater treatment due to the risk of eutrophication of the wastewater recipients. This phenomenon, that is responsible for the dramatic growth of algae occurring in internal and coastal waters, is caused by the excess phosphorus concentration in the effluents from municipal or industrial plants discharged in the environment [1].

There are three main sources of anthropic nutrient input. They are runoff, erosion and leaching from fertilized agricultural areas, and sewage from cities and industrial wastewater[2]. Atmospheric deposition of nitrogen (i.e., from animal breeding and combustion gases) can also be important. According to the European Environment Agency[2], most phosphorus pollution comes from households and industry, including phosphorus- based detergents.

The effects of eutrophication on the environment may have deleterious consequences for the health of exposed animal and human populations through various pathways. Specific health risks appear when fresh water that is extracted from eutrophic areas, is used for the production of drinking water. Severe impacts can also occur during animal watering in eutrophic waters.

In its simplest form, electrocoagulation (e.g., used as an electrochemical water and wastewater treatment technology) uses an electrochemical cell where a DC voltage is applied to the electrodes; usually made of iron or aluminum [3].This technique has some advantages when compared to conventional methods in terms of its simple equipment, easy to operate, less retention time, reduction or absence of adding chemicals, rapid sedimentation of the electrogenerated flocks, and less sludge production[4].

Electrocoagulation introduces metal cations in situ rather than via external dosing. Simultaneously electrolytic gases (typically hydrogen) are generated at the cathode. Depending on the reactor operating conditions and the pollutant, these bubbles may float some portion of the coagulated pollutant to the surface [5].

Electrocoagulation with Fe electrodes results in metal dissolution as shown below:

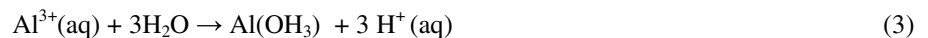
At the cathode:



At the anode:



In the solution:

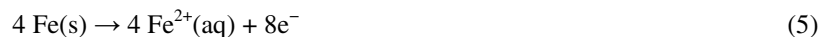


When iron is used as an electrode material, the reactions are the following:

At the cathode:



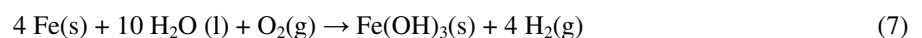
At the anode:



And with dissolved oxygen in solution:



Overall reaction:



Looking at the above reactions, it can be seen that electrocoagulation has occurred in three steps. In first step, coagulant has formed because of oxidation of anode. In second step, pollutants have stabilized. In last step, stabilized matters have united.

EC has been successfully used for decades in order to treat the wastewaters of textile [6, 7], food and protein [8], phosphate [9], tannery wastewater [10], restaurant wastewater [11], and defluoridation [12,13].

Accordingly, the purpose of this paper was to study the feasibility of the removal of orthophosphate from aqueous solution by electrocoagulation method using iron electrodes. The method was tested under various parameters including current density, pH, number of electrodes and concentration of supporting electrolyte. The main target was to determine the optimum operating conditions, with the aim of applying this method to a real wastewater.

2. Materials and methods

2.1. Experimental setup and procedure

The electrocoagulation experiments were conducted in a 1L Plexiglas reactor (Fig.1). The anode and cathode, fully immersed in water, with dimensions of 40mm×50 mm×0.5mm made of plate iron were connected to a digital dc power supply (MCP lab.électronics.M10-TP3003L) in bipolar mode. The effective area of the electrode pair was 1600mm². A magnetic stirrer (Linda instrument) was used to maintain the well mixing of the solution. The pH was adjusted by adding 0.5N HCl or 0.5 N NaOH. The conductivity of solutions was raised and adjusted to different values by the addition of NaCl. All experiments were carried out at constant temperature of 25 °C.

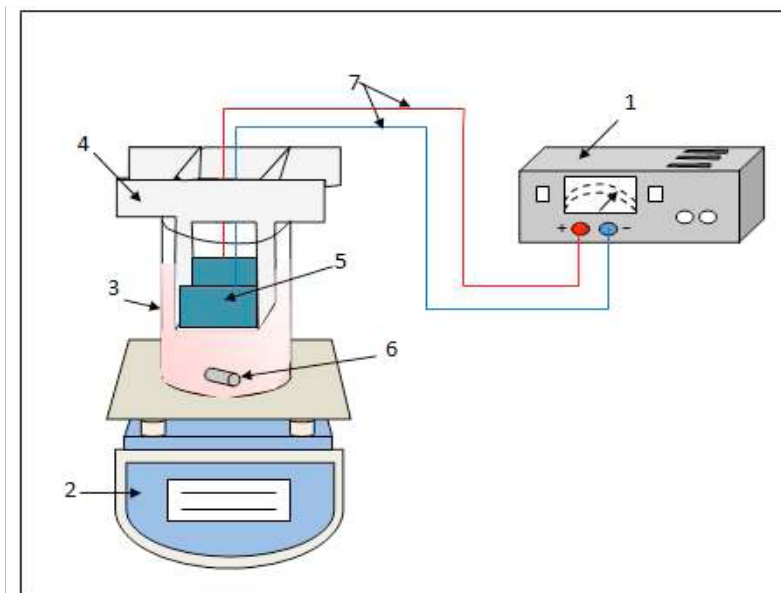


Fig. 1. A schematic diagram of the electrocoagulation reactor, 1: Potentiostat, 2: Magnetic stirrer, 3: Cylindrical column, 4: Plexiglas reactor 5: Electrode, 6: Magnetic stir bar, 7: electrical connections

2.2. Procedure and analysis

Before conducting each run, the iron electrodes were immersed for 5 min in a dilute HCl solution, to remove oxide, and then were rinsed with pure water. They were then polished by sandpaper to further remove oxide. An aqueous solution (1 L) containing KH₂PO₄ was placed into the reactor.

An ultraviolet-visible spectrophotometer (spectrophotometer SHIMADZU U.V mini 1240) was used to measure the maximum wavelength (821 nm) of orthophosphates. Equation (9) was used to calculate the removal efficiency of (PO₄)³⁻ in these experiments.

$$E\% = \frac{(C_i - C_f)}{C_i} \times 100 \quad (8)$$

Where C_i and C_f were the concentrations of (PO₄)³⁻ in the solution at the beginning and the end of the experiment, respectively.

The pH was measured using a Hanna Instrument pH 211 Microprocessor pH Meter. Conductivity was measured using a Hanna Instrument Conductivity Meter EC 214. The turbidity (NTU) of samples was analyzed using a Hanna LP-2000 turbidimeter. At the end of each EC treatment study, a solution with flocks was allowed to settle for 60 min in the container and the resulted water was withdrawn to be filtered (paper filter) before the chemical analysis. The samples for chemical analysis were taken from limpid phase.

2.3. Characteristics of synthetic wastewater

In this study, dried KH_2PO_4 at 105°C that obtained from Sigma–Aldrich Co. were used as samples of wastewater. The properties of these samples are shown in Table 1.

Table1. Physicochemical characteristics of K_2HPO_4

IUPAC Name	Potassium phosphate dibasic anhydrous
Synonyms	Dipotassiumhydrogenphosphate, Dipotassium phosphate, sec.- Potassium phosphate
Purity	99.0%
pH (25 °C, 1 M in H_2O)	8.5- 9.6
Molecularweight	174,1760 g mol^{-1}
Solubility in water	Soluble 1 M at 20 °C, clear, colorless

3. Results and discussion

3.1. Effect of conductivity

The increase of the conductivity, by the addition of supporting electrolyte, is known to reduce the cell voltage (U) at constant current density due to the decrease of the ohmic resistance of the solution [14]. Energy consumption, which is proportional to UI , will therefore be decreased.

NaCl is generally used to increase the solution conductivity by electrocoagulation treatment. It also allows decreasing the passivity of the electrodes by removing the passivating oxide layer formed on electrode surface [15] due to its catalytic action. Chloride ions could significantly reduce the adverse effects of other anions (due to their oxidation), and increases the availability of iron hydroxide in the solution. Thus, this parameter should have a significant impact on removal efficiencies of pollutants.

The effect of the concentration of the supporting electrolyte on orthophosphate removal was examined at 0.5, 0.75, 1.0 and 2.0 g/NaCl . Two iron electrodes were used, current intensity of 1.5A, distance between electrodes of 1.8 cm and pH of the initial solution were kept constant in the experiments.

For lower concentrations, there was a decrease of that rate, probably because there were not enough ions to conduct the current. Thus, it may be concluded that increasing the electrical resistance of the solution would decrease the efficiency of the process. The effect of NaCl concentration on the removal efficiency as a function of treatment time is shown in Fig.2. It can be seen that the orthophosphate ions removal efficiency increased from 85 to 92% as the NaCl concentration increased from 1 to 2 g/L , after 70 min reaction time. On the other hand, at lower NaCl concentrations of 0.50 and 0.75 g/L , the orthophosphate removal was seen to be less significant compared to that of the higher electrolyte concentration. These results can be explained by the fact that when the NaCl concentration increased, the conductivity of the electrolyte was enhanced correspondingly. This was probably, because the Cl^- anions can destroy the passivation layer and increase the anodic dissolution rate of metal, either by the incorporation of Cl^- into the oxide film or by the participation of Cl^- in the metal dissolution reaction [16].

Similar kind of effect of increase in removal efficiency with increase in conductivity was also reported by Kobya et al [17], and Mollah et al [18].

According to the results of this study, best orthophosphate removal efficiency with low electrocoagulation time can be obtained with NaCl of 2 g /L. In this respect, 2.0 g /L NaCl was used in the rest of the experiments of this work.

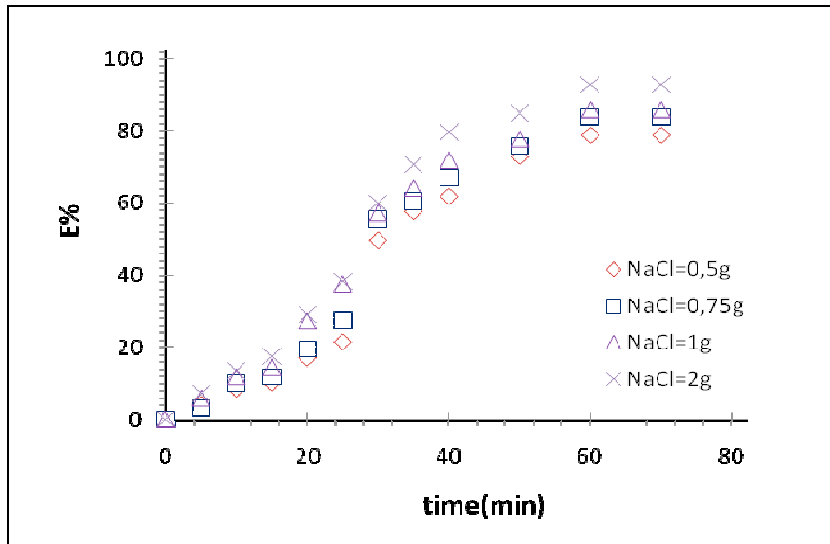


Fig. 2. Effect of NaCl on orthophosphate removal (operating time = 70 min, $C_i=10\text{mg}\cdot\text{L}^{-1}$, $N=2$, $D=1.8\text{cm}$, $I=1.5\text{A}$)

3.2. Effect of current intensity

Operating current density is critical in batch electrocoagulation as it is the only operational parameter that can be controlled directly [19]. The supply of current to the electrocoagulation system determines the amount of Fe^{2+} ions released from the anode and adjusts the rate and size of the bubble production. Accordingly, this will affect the growth of flocs which in turn influence the treatment efficiency of the electrocoagulation [20, 21]. Moreover, it will strongly influence the solution mixing and the mass transfer at the electrodes. Thus, this parameter is expected to have a significant impact on orthophosphate removal efficiencies.

To investigate the effect of current intensity on the orthophosphate removal, a set of experiments were conducted by solutions containing constant solution concentration with current intensity being varied from 0.6 to 2.0 A. The pH of solution, the distance between electrodes, and the supporting electrolyte concentration were kept constant in the experiments. It was found that the dissolving rate of iron electrode increased with the increasing current density. Consequently, the orthophosphate removal was found to be increased with the increasing current density as more Fe^{2+} will pass to the solution at higher current density and hence formation rate of $\text{Fe}(\text{OH})_2$ will be increased.

Fig. 3 illustrates the effect of current density on the orthophosphate removal efficiencies. The $(\text{PO}_4)^{3-}$ removal efficiency was increased to 92% at 2 A from 82% at 0.6 A after 70 min reaction. At a high current intensity, the extent of anodic dissolution of iron increases, resulting in a greater amount of precipitate and removal of orthophosphate.

It should be noted that an increase in current intensity from 1.5 to 2 A resulted in an increase in the efficiency of $(\text{PO}_4)^{3-}$ removal from 92 to 89%, which was not a significant change. As a result of increasing the current intensity, the applied potential increased. Thus, it is advisable to limit the current intensity in order to prevent excessive oxygen evolution as well as to eliminate other adverse effects, like heat generation and also to avoid high electrical energy consumption [18]. Therefore, the current 1.5 A was a reasonable current intensity in our experiments.

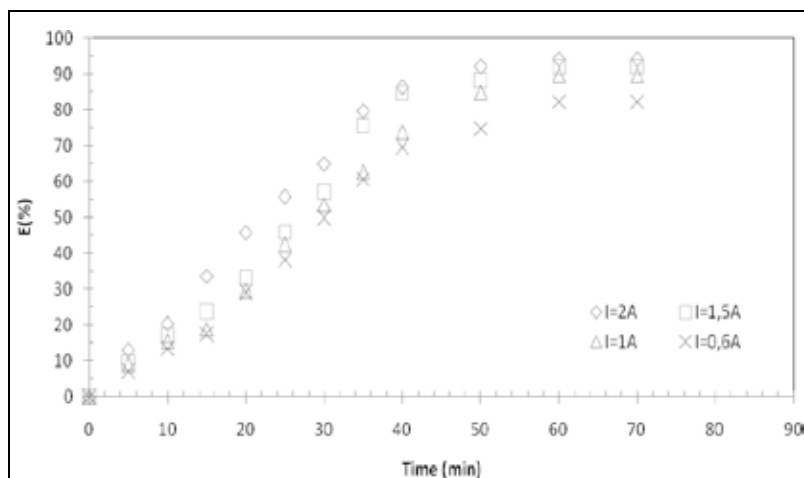


Fig. 3. Influence of the current intensity on the removal rates of orthophosphate ($C_i=10\text{mg}\cdot\text{L}^{-1}$, $N=2$, $D=1.8\text{cm}$, $N_{ac}=2\text{g/l}$)

3.3. Effect of pH

The pH is known to play a key role on the performance of EC [22]. The initial pH is one of the important factors in affecting the performance of electrochemical process. It has been established that the pH has a considerable influence on the performance of electrocoagulation process. To investigate this effect in this work, a series of experiments were performed using synthetic solution containing orthophosphate concentration of 10 mg/L. Experiments were carried out at various values of pH (3.0, 5.0, 6.0, 8.0 and 9.0) under which the current intensity was kept at 1.5 A. The distance between electrodes and concentration and supporting electrolyte concentration were kept constant during these experiments.

Variation of orthophosphate removal efficiency versus time at different initial pH is shown in Fig. 4. The highest efficiency of orthophosphate removal was obtained when the initial pH was 6. Efficiencies of orthophosphate removal using iron electrodes by electrocoagulation were 89% for pH 5, 98% for pH 6, 95% for pH 8; at the end of 60 min.

It can be also seen that the initial pH of 3 had no significant effect on the removal efficiency of the orthophosphate; indeed a dramatic decrease of the removal efficiency of $(\text{PO}_4)^{3-}$ was observed (57%) after a same time of electrocoagulation. This behavior is in accordance with the character of iron that does not precipitate at acidic medium.

The results of this study agree very well with that presented in the open literature indicating that the maximum amount of phosphate removal had occurred at pH 6.0. Therefore, it can be concluded that at this pH, the majority of iron complexes (coagulants) are formed and it is the optimum pH for carrying out the electrocoagulation. According to Pourbaix [23], it was deduced that the major complexes formed at this pH were $\text{Fe}(\text{OH})^+$, $\text{Fe}(\text{OH})_2$, $\text{Fe}(\text{OH})_4^{2-}$ and $\text{Fe}(\text{OH})_3$.

3.4. Effect of electrodes number

There is a direct relation between removal efficiency and the total number of electrodes. So when the area of electrodes increases, the distribution of the coagulation agents density will be more effective. This can produce the related coagulation and improve the removal of orthophosphate. Fig. 5 depicts the orthophosphate removal efficiency at different number of electrodes (1, 2, and 4). As the number of electrodes was increased, the efficiency of orthophosphate removal increased (92%, 93%, and 96% respectively, after 70 min reaction time). The increase in the number of electrodes will decrease the electrostatic effects. Hence, the movement of the produced ions would be slower and they will have more opportunity to aggregate and produce coagulants flocs. Consequently, these flocs were able to absorb more orthophosphate.

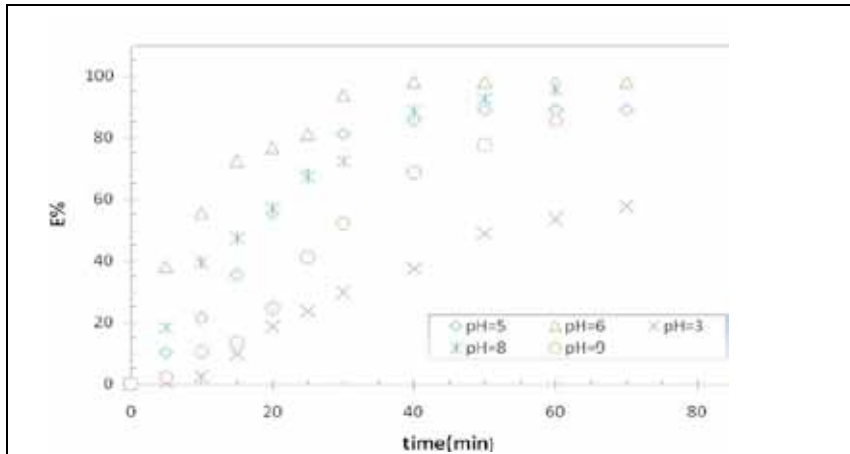


Fig. 4. Influence of initial pH on the elimination of orthophosphate ($C_i=10\text{mg.L}^{-1}$, $N=2$, $D=1.8\text{cm}$, $\text{NaCl}=2\text{g/l}$, $I=1.5\text{A}$)

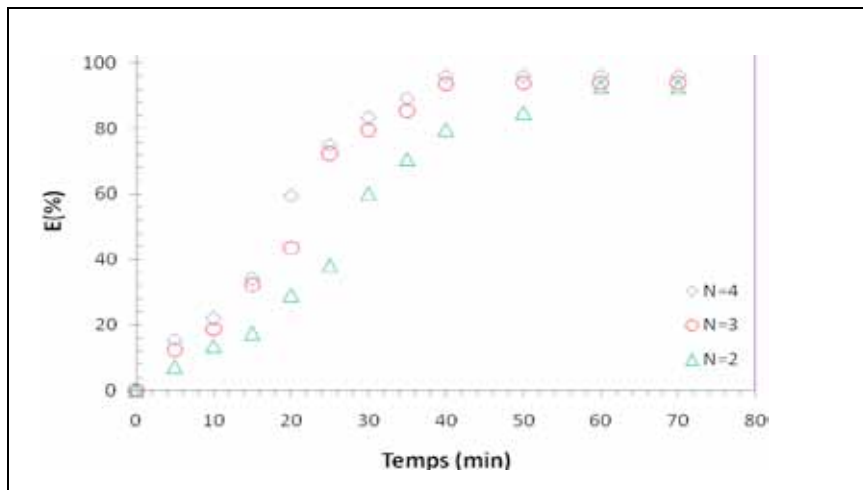


Fig. 5. Influence of the electrodes number on the removal of orthophosphates ($C_i=10\text{mg.L}^{-1}$, $D=1.8\text{cm}$, $\text{NaCl}=2\text{g/l}$, $I=1.5\text{A}$, $\text{pH}=6$)

4. Conclusion

The current study demonstrated the efficiency of electrocoagulation method on the orthophosphate removal. The influence of different parameters on the removal of orthophosphate was determined. These parameters include the pH, the current intensity, the supporting electrolyte concentration, and the number of electrodes

The achieved results indicated that the orthophosphate removal efficiency was over 90% when iron was used as a sacrificial anode under the following conditions: initial pH of 6.0, current intensity of 1.5 A, supporting electrolyte concentration of 2g/L and the number of electrodes of 4. It is recommended that further study is required to investigate the detailed mechanisms of orthophosphate removal by electrocoagulation.

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