



# The effect of supporting electrolyte type and concentration on the phosphate removal from water by electrocoagulation method using iron electrodes

## Elektrokoagülasyon yöntemi ile demir elektrotlar kullanılarak sulardan fosfat giderimi üzerine destek elektrolit türü ve konsantrasyonunun etkisi

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### Abstract

In this study, the effect of the type and concentration of the supporting electrolyte given to the water to increase the conductivity in the electrocoagulation process on the phosphate removal from synthetically prepared waters was investigated. In addition, experiments were carried out without electrolyte and the results were compared. The effect of support electrolyte type and concentration on phosphate removal efficiency, energy consumption and reaction rate was analyzed. Experiments have shown that electrolyte use has no significant effect on phosphate removal efficiency. However, it has been found that the addition of electrolyte reduces energy consumption and increases the removal rate. From the results obtained, it was determined that NaCl as the most suitable electrolyte type and 5 mM electrolyte was sufficient as the optimum dose. Under these conditions, for 100 mg L<sup>-1</sup> PO<sub>4</sub>-P at 0.5 mA cm<sup>-2</sup> current density, the phosphate removal efficiency was 96.98%, the energy consumption was 1.13 kWh m<sup>-3</sup> and the first order reaction rate constant was 0.0593 min<sup>-1</sup>.

**Keywords:** Electrocoagulation, Phosphate removal, Supporting electrolyte, Iron electrode

### 1 Introduction

One of the parameters used in monitoring environmental water resources in industrialized countries is amount of phosphate in the natural waters. One of the main causes of eutrophication, which is one of the environmental problems occurring in aquatic environments, is phosphorus released into the environment without treatment [1]. Eutrophication, which means excessive growth of algae in aquatic environments, is caused by the concentration of nitrogen and phosphorus in wastewater released from various industries to the receiving environment [2-5]. The main source of phosphorus in wastewater is agricultural, industrial and domestic uses [6].

Phosphorus removal techniques include adsorption [7], chemical precipitation [8], ion exchange [9], electro dialysis [10], membrane filtration [11], electrocoagulation [12] and

### Özet

Bu çalışmada elektrokoagülasyon prosesi kullanılarak atıksuyun iletkenliğini artırmak için suya verilen destek elektrolitin türü ve konsantrasyonunun sulardan fosfat giderimine etkisi araştırılmıştır. Ayrıca elektrolitsiz deneyler yapılmış ve sonuçlar karşılaştırılmıştır. Destek elektrolit tipi ve konsantrasyonunun fosfat giderme verimliliği, enerji tüketimi ve reaksiyon hızı üzerindeki etkisi analiz edilmiştir. Deneyler, elektrolit kullanımının fosfat giderme verimliliği üzerinde önemli bir etkisi olmadığını göstermiştir. Ancak elektrolit ilavesinin enerji tüketimini azalttığı ve uzaklaştırma oranını artırdığı bulunmuştur. Elde edilen sonuçlardan en uygun elektrolit türü olarak NaCl ve optimum doz olarak 5 mM elektrolitin yeterli olduğu belirlenmiştir. Bu koşullar altında, 0.5 mA cm<sup>-2</sup> akım yoğunluğunda 100 mg L<sup>-1</sup> PO<sub>4</sub>-P için fosfat giderme verimi %96.98, enerji tüketimi 1.13 kWh m<sup>-3</sup> ve birinci dereceden reaksiyon hızı sabiti 0.0593 dk<sup>-1</sup> olarak elde edilmiştir.

**Anahtar Kelimeler:** Elektrokoagülasyon, Fosfat giderme, Destek elektrolit, Demir elektrot

biological phosphate removal [13, 14]. The most important process used in the phosphate removal from wastewaters is to convert the soluble phosphate compounds into an insoluble solid phase and separate them from water by solid-liquid separation methods such as precipitation and filtration. The most widely used chemical method in wastewater treatment and phosphate removal is precipitation of phosphate in the form of Al<sup>3+</sup>, Ca<sup>2+</sup> and Fe<sup>3+</sup> compounds.

Electrochemical treatment methods and especially electrocoagulation process have begun to be widely used in domestic and industrial wastewaters treatment today. Electrocoagulation is a process in which some soluble metals such as iron and aluminum are used as anodes and this anode material dissolves into water by electric current. The dissolved anode material performs the treatment by forming metal hydroxides that are insoluble in water. The most important difference that distinguishes electrocoagulation

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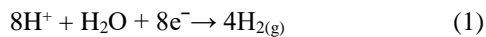
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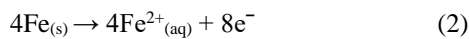
from chemical coagulation is the way the coagulant is given to water [15]. In electrocoagulation, flocs occur not by adding chemicals to the system, but by dissolving aluminum and iron electrodes in the reactor [16]. Electrocoagulation is based on the principle that undissolved suspended solids and emulsions in water are unstable by changing their electrical charges. In this way, the surface charge of the particles is neutralized and colloids come together to form flocs that can collapse [17]. Electrode selection is one of the most important parameters of the electrocoagulation process. Therefore, choosing the right material is very important [18]. The main reason why aluminum and iron are widely used for electrocoagulation is that they are cheap and easily available. They are also preferred because hydroxides of aluminum and iron are poorly soluble in water. In the case of using iron as anode, two different mechanisms occur in the formation of Fe(OH)<sub>n</sub>, depending on the environment pH, with the condition that n = 2 or 3 [12].

First mechanism;

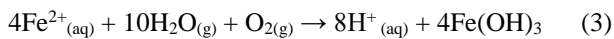
The cathode reactions;



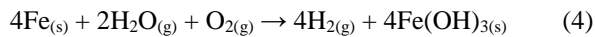
The anode reactions;



In solution;

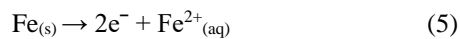


Finally, total reaction can be summarized as;

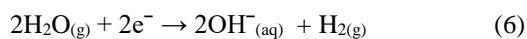


### Second mechanism

The cathode reactions



The anode reactions;



Finally, total reaction can be summarized as;



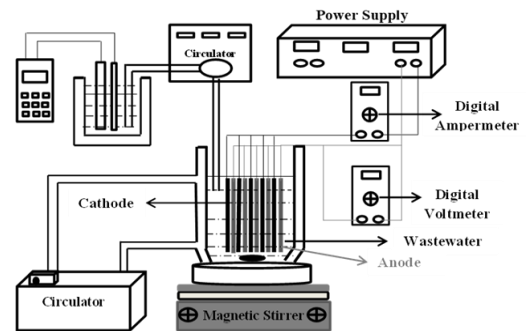
In this study, the effect of the type of supporting electrolyte added to the system in order to increase the conductivity of the wastewater in the electrocoagulation process in the removal of phosphate compounds in high concentrations in wastewater was examined and the most appropriate electrolyte dosage was tried to be determined. In the study, the suitable electrolyte type was examined in terms of both phosphate removal efficiency, energy consumption and reaction rate.

## 2 Material and method

Phosphate solutions used in the experiments were synthetically prepared from solid KH<sub>2</sub>PO<sub>4</sub> with analytical purity. pH adjustments were made using 5M HNO<sub>3</sub> and

NaOH. Electrocoagulation experiments were carried out in the setup shown in Figure 1. The reactor used in the experiments is made of plexiglass material, its volume (100x110x100) is 1100 ml and has a cooling jacket. In the experiments, 850 ml phosphate solution was used. In the experiments, 6 anodes and 6 cathode Fe electrodes of 100x75x3 mm were used. The dimensions of the submerged part of these electrodes are 80x75x3 mm and they are placed at 5 cm intervals.

The experiments were conducted in batch mode. The potential difference applied to the cell and the current flowing through the cell were obtained by using a digitally controlled direct current source (Shenzhen-Mastech HY3005-3) and these parameters were measured with the help of two digital multimeters (Brymen-201). The wastewater in the electrocoagulation reactor was mixed at a constant speed of 150 rpm using a Heidolph MR-3004 brand magnetic stirrer. During the experiments, the temperature, pH and conductivity values of the solution were measured using a WTW Multiline P-4-F-Set-3 multi-parameter device.



**Figure 1.** Schematic view of the experimental system used in the study

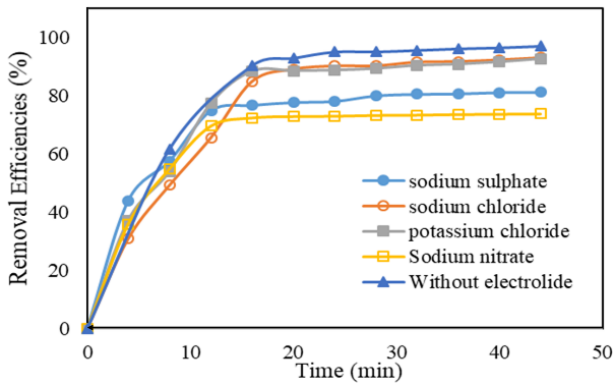
During the experiments, phosphate analyzes were performed spectrophotometrically (Shimadzu UV 160A) at 400 nm wavelength using ammonium vanadomolybdate reagent.

## 3 Results and discussion

### 3.1 The effect of supporting electrolyte type on system variables

Studies examining the effect of supporting electrolyte type on phosphate removal from wastewater by electrocoagulation method, I=0.75 A (CD=0.5 mA cm<sup>-2</sup>), initial pH value of wastewater was chosen as pH<sub>i</sub>=3 and supporting electrolyte concentration CSE=5 mM [19].

Na<sub>2</sub>SO<sub>4</sub>, NaNO<sub>3</sub>, NaCl and KCl salts, which are abundant and have high water solubility, were used as support electrolytes and the effects of these electrolytes on phosphate removal efficiency, energy consumption and reaction rate were investigated. The variation of phosphate removal efficiency and energy consumption of the system versus time for different types of supporting electrolytes are given in Figure 2 and Figure 3, respectively.



**Figure 2.** The change of removal efficiencies vs time for PO<sub>4</sub>-P ions in different types of supporting electrolyte

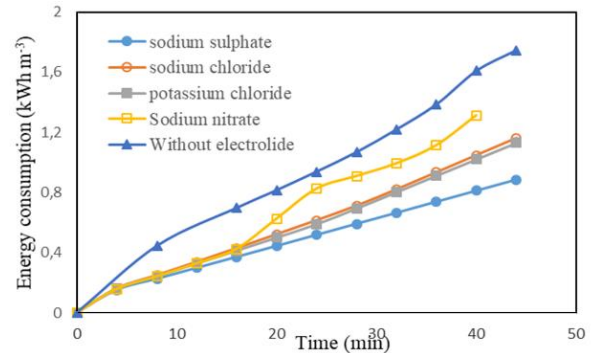
When Figure 2 is examined, it is seen that relatively higher removal efficiency is obtained in experiments without using any electrolyte. This is thought to be due to the competition between PO<sub>4</sub><sup>3-</sup> ions in the environment and Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup> ions added to the water. It has been determined that this decrease in the removal efficiency is less in Cl<sup>-</sup> ions because of the oxidation of chlorine in water to Cl<sub>2</sub> and other forms. Reactions of chlorine in water are shown Equation (8), (9) and (10) [20]. In studies, NaCl and KCl showed almost the same effect in terms of phosphate removal efficiency. The highest removal efficiencies were obtained with NaCl and KCl, while the lowest removal efficiency was obtained with NaNO<sub>3</sub> [21]. In addition, a decrease in efficiency is observed as a result of forming a complex with Fe<sup>3+</sup> in the presence of SO<sub>4</sub><sup>2-</sup> ions and affecting the current efficiency by adhering to the electrode surface [22]. At the end of the reaction period of 20 minutes, the phosphate removal efficiencies of the system for Na<sub>2</sub>SO<sub>4</sub>, NaNO<sub>3</sub>, NaCl and KCl were found to be 78%, 73.2%, 93.03% and 92.58%, respectively.



As can be understood from these equations, the reactions that occur in the presence of Cl<sup>-</sup> ions in the environment are equilibrium reactions and change according to the pH of the wastewater. At low pHs, the HOCl molecule is dominant, while at high pH, the OCl<sup>-</sup> molecule is dominant. Since the oxidation power of HOCl is much higher than that of OCl<sup>-</sup>, it is desirable that the pH of the wastewater be low in order for the oxidation to occur in the water to occur faster and more effectively.

As seen in Figure 3, the highest energy consumption occurs when electrolyte is not used. NaCl and KCl showed the same effect in energy consumption as in efficiency. However, the lowest energy consumption was obtained when Na<sub>2</sub>SO<sub>4</sub> was used [23]. Because while other electrolytes give water two ions, one positive and one

negative, sodium sulphate gives 3 ions, 2 positive and negative. At the end of the reaction period of 20 minutes, the energy consumptions of the system for without electrolyte, Na<sub>2</sub>SO<sub>4</sub>, NaNO<sub>3</sub>, NaCl and KCl were found to be 0.815, 0.443, 0.624, 0.520 and 0.500 kWh m<sup>-3</sup>, respectively. Electronegativity is a relative concept that varies from compound to compound. Since the electronegativity value of SO<sub>4</sub><sup>2-</sup> ion is higher than other anions, it increases the conductivity at a higher rate. For this reason, the Na<sub>2</sub>SO<sub>4</sub> compound provides a lower energy consumption.



**Figure 3.** The change of energy consumptions vs time for PO<sub>4</sub>-P ions in different types of support electrolyte

Although the most suitable supporting electrolyte in terms of power consumption is sodium sulphate, considering the removal efficiency, it is seen that NaCl and KCl are the most suitable electrolytes. NaNO<sub>3</sub> is not preferred due to both the lowest removal efficiency and the highest energy consumption. The rate constants calculated according to first and second order reaction kinetics for different support electrolyte types are shown in Table 1.

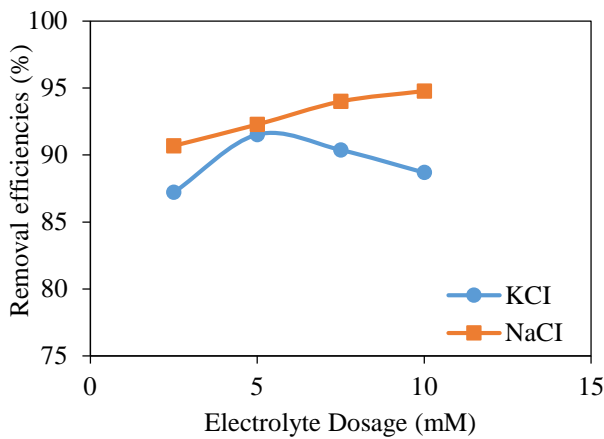
When Table 1 is examined, it is seen that the reaction proceeds according to the 2nd degree when the electrolyte is not used. The highest removal rate was achieved when no electrolyte was used. In both reaction degrees, the lowest R<sup>2</sup> values were obtained in Na<sub>2</sub>SO<sub>4</sub> and NaNO<sub>3</sub> electrolytes. In terms of reaction rate, it is seen that the most suitable electrolytes are KCl and NaCl.

### 3.2 The effect of supporting electrolyte concentration on system variables

While examining the effect of supporting electrolyte concentration on phosphate removal from wastewater by electrocoagulation method, current intensity was chosen as 0.75 A (current density 0.5 mA cm<sup>-2</sup>) and initial pH of 3. The experiments were performed at 2.5 mM, 5 mM, 7.5 mM and 10 mM electrolyte concentrations using NaCl and KCl which gave similar results with other types of support electrolyte. The change of phosphate removal efficiency for different KCl and NaCl concentrations is given in Figure 4, and the change of the energy consumption of the system is given in Figure 5

**Table 1.** Reaction rate constants for different types of support electrolyte according to first and second order reaction kinetics

Supporting electrolyte type	$k_1$ (min <sup>-1</sup> )	R <sup>2</sup>	$k_2$ (L mg <sup>-1</sup> min <sup>-1</sup> )	R <sup>2</sup>
Without electrolyte	0.0764	0.8562	0.0073	0.9785
Na <sub>2</sub> SO <sub>4</sub>	0.0305	0.7071	0.0008	0.8192
NaNO <sub>3</sub>	0.0250	0.5932	0.0005	0.6761
NaCl	0.0619	0.8547	0.0031	0.9468
KCl	0.0557	0.7985	0.0025	0.9234

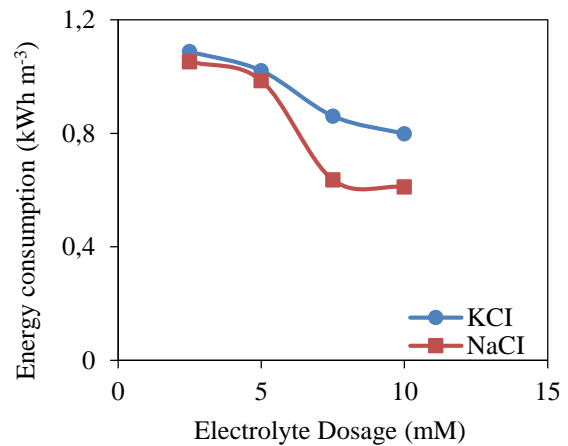


**Figure 4.** The change of phosphate removal efficiencies vs different supporting electrolyte concentrations

When KCl and NaCl are compared, it can be said that NaCl is a little more suitable in terms of phosphate removal efficiency. As the NaCl concentration increases, the phosphate removal efficiency increases, even if only a small amount. However, this increase is not high enough when the cost of the chemicals used is considered. Therefore, an electrolyte concentration at which both energy and chemical substance consumption will be low must be applied.

Figure 5 show that NaCl reduces energy consumption a little more compared to KCl. The increase in the electrolyte concentration does not affect the efficiency, but significantly reduces the energy consumption. At the end of the 40-minute reaction time, the energy consumption of the system 2.5 mM, 5 mM, 7.5 mM and 10 mM KCl are 1.088, 1.021, 0.8611 and 0.798 kWh m<sup>-3</sup>, respectively. This values for NaCl were determined as 1.052, 0.986, 0.6364 and 0.6117 kWh m<sup>-3</sup>. As the conductivity of the wastewater increases with the addition of electrolyte, its electrical resistance decreases, which causes a decrease in the potential difference applied to the system at constant current density. The reduction of this potential difference means that the energy consumption of the system is reduced.

The rate constants calculated according to the second order reaction kinetics are also given in Table 2. According to the data in Table 2, it is seen that NaCl is a more suitable electrolyte in terms of reaction rate.



**Figure 5.** The change of energy consumptions vs different supporting electrolyte concentrations

**Table 2.** Second order reaction rate constants at different KCl and NaCl concentrations

C <sub>SE</sub> (mM)	KCl		NaCl	
	$k_2$ (L mg <sup>-1</sup> min <sup>-1</sup> )	R <sup>2</sup>	$k_2$ (L mg <sup>-1</sup> min <sup>-1</sup> )	R <sup>2</sup>
0	0.002	0.91	0.002	0.91
2.5	0.0018	0.91	0.0032	0.79
5	0.0024	0.88	0.0708	0.76
7.5	0.0014	0.91	0.0791	0.63
10	0.0021	0.93	0.1417	0.90

#### 4 Results

While examining the effect of electrolyte type and concentration in studies of removing phosphate ions by electrocoagulation, Na<sub>2</sub>SO<sub>4</sub>, NaCl, NaNO<sub>3</sub> and KCl were used as electrolytes. Studies with iron electrodes have shown that the use of electrolyte does not affect the phosphate removal efficiency much. However, since the use of supporting electrolyte increases the conductivity of the wastewater, the energy consumption of the system is greatly reduced. When the electrolyte is not used, while the energy consumption of the system to achieve 90% efficiency is 1.626 kWh m<sup>-3</sup>, when 5 mM NaCl and 5 mM KCl was used, these values are determined as 0.614 and 0.692 kWh m<sup>-3</sup>, respectively.

In studies of removal of phosphate ions from wastewater by electrocoagulation method, it is seen that the most

suitable electrolyte type is NaCl and KCl when both disposal efficiency, energy consumption and reaction rate are taken into consideration. Although Na<sub>2</sub>SO<sub>4</sub> is more suitable in terms of energy consumption, it reduces phosphate removal efficiency. NaNO<sub>3</sub>, on the other hand, is not considered suitable in terms of both energy consumption and removal efficiency. The optimum electrolyte concentration was determined as 5 mM. Although the use of more electrolyte reduces the energy consumption of the system, it is not considered appropriate in terms of removal efficiency and total cost.

#### Conflicts of interest

No conflict of interest was declared by the authors.

Similarity rate (iThenticate): %18

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