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# Zeolite-assisted electrocoagulation for remediation of phosphate from calcium-phosphate solution

Ahmad K. Alenezi<sup>1\*</sup>, Hussein A. Hasan<sup>2</sup>, Khalid S. Hashim<sup>3,4</sup>, Joseph Amoako-Attah<sup>3</sup>, Michaela Gkantou<sup>3</sup>, Magomed Muradov<sup>3</sup>, Patryk Kot<sup>3</sup>, Bareq Abdulhadi<sup>3,4</sup>

<sup>1</sup>BSc student, Civil Engineering Department, Liverpool John Moores University, UK.

<sup>2</sup>Department of Chemistry, College of Science, University of Babylon, Iraq

<sup>3</sup>Department of Civil Engineering, Liverpool John Moores University, UK

<sup>4</sup>Environmental Engineering Department, College of Engineering, University of Babylon, Iraq

\*Email: [A.K.Alenezi@2016.ljmu.ac.uk](mailto:A.K.Alenezi@2016.ljmu.ac.uk)

**Abstract.** Phosphorus is crucial to the growth of the skeletons and the living cell. However, elevated phosphate concentrations represent a problem due to its grave impacts on human health, animal and also because of its effects on water quality and economy. This research studies the applicability of filtration-electrocoagulation (filtration - EC) system to remove phosphate from wastewater when calcium is present as competitor ions. In laboratory batch trials, the ability of zeolite filter-electrocoagulation system to eliminate phosphate from artificial wastewater samples, in the presence of calcium as competitor ions, was tested. The research was initiated by studying the ability of the electrocoagulation method (EC), as a separated treatment unit, to remove phosphate from solutions that containing calcium ions. In this phase of the study, the influence initial pH, current densities and electrolysing period. Then, the new method, filtration-EC, was applied to treat solutions (with identical chemical composition to that used in the first phase) considering the effects of the initial pH, current densities, contact periods (in the filter), electrolysing period (in the EC cell) and zeolite dosage. The results of this research evidenced that the filtration - EC system completely removes phosphate from calcium-phosphate solution at pH, current density, dosage of zeolites, contact period and electrolysing period of 7, 2.0 mA.cm<sup>-2</sup>, 150.0 g, 30.0 minutes and 20.0 minutes, respectively.

## 1. Introduction

Phosphorus can be existed at elevated concentration in the crust of Earth (such as rocks and other natural geological formations that explains its unacceptable concentration in freshwater resources [1, 2]. Beside the presence of phosphate in the natural formations, effluents that resulted from man activities have increased the concentration of phosphate in water sources [2-5]. In addition, climate changes maximises water consumptions that limits the availability of freshwater [6-10]. Although, this element can be found in freshwater resources in different dissolved forms, including orthophosphate polyphosphate and organic phosphate, the most predominant and thermodynamically stable form of phosphate is the orthophosphate [11]. Where, the published articles indicate that the orthophosphate forms half of the total quantity of phosphates in the nature [12, 13]. On the top of that, polyphosphate is not stable in the aquatic environment, which results in a conversion of this type of phosphate to orthophosphate form due to series natural chemical reactions [12]. Hence, in the laboratory studies, orthophosphate is the common indicator for phosphate pollution [2].



Phosphate is acknowledged as a crucial element for the health and growth of mankind, animal and living cells [2]. At the same time, when the concentration of phosphate increased more than the necessary concentration for the health of mankind and other organisms, it causes wide ranges of health, environmental and economic problems [11]. For example, considerable literature evidenced that 100µg/L of phosphorus is quite enough to trigger a revolution in the algal growth in the sources of freshwater (known as the eutrophication phenomenon) that results in a substantial deterioration of the quality of freshwater. Where, the eutrophic water body experiences severe lack of sunlight, depletion of oxygen, and substantial increase in the concentrations carbon dioxide that cause rapid death of aquatic animals and plants and consequently generation of toxic chemicals, colours and odours [12, 13]. Economically, the relevant investigations evidenced that high concentration of phosphate causes fouling of pipes and inhibition or minimising the removal of water pollutants, which substantially increase the cost of regular maintenance of water supply facilities and the cost of water treatment process [11, 14, 15]. On the top of that, phosphate, at high concentrations, leads to various health problems [15].

Hence, a number of treatments, such as adsorptions, cultivations of a microalgae, chemical precipitations, membranes filtration and electrocoagulation, were suggested to eliminate or minimise phosphate in waters or wastewaters [15-18]. Although these methods were practiced in the literature, the electrocoagulation method (EC) has attracted an increasing interest as a phosphate removal/minimising method because this method is environmental-friendly, cheap, and it consumes low energy [19]. Additionally, the EC method requires, comparing to traditional approaches, short period to attain good removal level of the targeted pollutant(s) [20], and it eliminates the need for chemicals coagulants that prevents the generation of secondary pollutants [21, 22]. Additionally, the EC method showed the potential to remove a broad range of pollutants from solutions, such as phenols [18], heavy metals [23, 24], nitrate [25], pathogens [26, 27], textile dyes [28] and organic matter [29]. More importantly, dense with low water content sludge is produced from the EC plants [23, 25], that facilitates its recycling [30-33] or disposal in low-cost manner [34-37]. At the same time, like other technologies, the EC suffers from a number of key drawbacks, such as the sensitivity to the chemical composition of solutions, where the literatures proved that some ions, like silica and organic matters, exert extra stress on the efficiency of the EC systems that limits its ability to eliminate other contaminants [38, 39]. For instance, the published articles proved that the efficiency of the EC systems in the elimination of fluorides from solutions decreases with the occurrence of sulphate [38].

The current research is therefore aiming at combining two eco-friendly methods, namely zeolite filter and EC cell (aluminium-based cell) to eliminate or minimise phosphate from water in the existence of competitors (calcium ions).

## 2. Materials and methods

### 2.1. Solutions and materials

Initially, to prepare synthetic calcium-phosphate solution, one litre of deionised water was decant into a glass beaker that have 1500 millilitre capacity; then 87.870 milligrams of  $\text{KH}_2\text{PO}_4$  was added to this beaker and stirred for few minutes to produce 20 mg/l phosphate solution. 5 mg.L<sup>-1</sup> of calcium was added to the samples using the required weights of  $\text{CaCl}_2$ . NaCl was used to increase the conductivity of this solution to 320 µS/m, while the variation of the pH was achieved using proper amounts of hydrochloric acid or sodium hydroxide. Three samples of prepared samples, after modifying the pH to the desire level, were used in each experimental trial.

The electrocoagulation cell was manufactured using four aluminium plates (as electrodes) that have net area of 245 cm<sup>2</sup>; these electrodes were located inside a 1400 milliliters plastic vessel.

Zeolite samples, which have average particle size of 0.1 to 0.3 cm, were supplied by Liverpool John Moores University. Aluminium and iron content, in this zeolite sample, is about 0.130 (wt/wt).

### 2.2. Batch experimental trials

Batch experimental trials, in this study, was commenced according to published methodologies [40, 41]. The solution was treated using the zeolite filter, EC and filtration-EC method separately. In the zeolite filter, various dosages of zeolite particles were placed in the bottom of the beaker; then calcium-

phosphate solution was decanted into the beaker and mechanically stirred for 20 minutes at speed of 110.0 rpm. Then, the stirred solution was rested, at the laboratory temperature, for the required period (detention time), then filtrated and tested for the residual phosphat concentration (using Hach Lange spectrophotometer and phosphate cuvetts). The removal of phosphat by the EC method, separately, was tested by electrolysing the calcium-phosphate solution at various current densities, electrolysing period and pH. Finally, the filtration-EC methods was used to treat calcium-phosphate solution at the best levels of the current densities, electrolysing period, detention period, pH and dosage of zeolite.

### *2.2.1. Filtration experiments*

#### *2.2.1.1. Effects of pH*

To investigate the impact of pH a solution on phosphates adsorption by zeolites, at pH 3, 7 or 8, 150g.L<sup>-1</sup> of zeolite were applied for 30 minutes to 20 mg.L<sup>-1</sup> phosphate samples. The obtained removal at each value of the pH was recorded, and the whole results were compared later. As it was earlier mentioned that HCl or NaOH was used, in this paper, to adjust the pH.

#### *2.2.1.2. Effects of adsorbent dosages*

The impact of the adsorbent dosages on the remediation of phosphates was checked against three weights of zeolites, namely 37.0, 75.0 and 150.0 grams per one litre of calcium-phosphate solution. As it was earlier mentioned, in this paper, the required weight of zeolite and phosphate-containing water sample were mixed together in a 1400 millilitre beaker, and then stirred for 20.0 minutes at speed of 110.0 rpm. The stirred solution was left to rest to the required period. The obtained removal at each weight of zeolite was recorded, and the whole results were compared later.

#### *2.2.1.3. Effects of contact period*

Treatment time was extended from 10.0 to 120.0 minutes to check the effects of the contact time of the ability of the zeolite filter to remediate phosphate from water. These experiments were commenced at the optimum pH and the zeolite dosage that were obtained in the previous stages. The obtained removal at each period was recorded, and the whole results were compared later.

### *2.2.2. EC experiments*

#### *2.2.2.1. Effects of current densities on phosphates remediation*

1.0, 2.0 and 3.0 mA.cm<sup>-2</sup> was applied to calcium-phosphate solutions, 20.0 mg.L<sup>-1</sup>, to check the effects of the current densities on the remediation of phosphate from calcium-phosphate solution. Each experiment was run for 20.0 minutes. The obtained removal at each current density was recorded, and the whole results were compared later.

#### *2.2.2.2. Effect of electrolyzing time*

Electrolyzing time was extended from 5.0 to 30.0 minutes to check the effects of the treatment time of the ability of the EC system to remediate phosphate from water. These experiments were commenced at the optimum pH optimum current density obtained in the earlier experiments, and constant space between electrodes. The obtained removal at each period was recorded, and the whole results were compared later.

#### *2.2.2.3. Filtration-EC experiments*

In this last part of the research, the zeolite filter was applied to the calcium-phosphate solution, and then the solution was transferred to the EC system. These experiments were performed at the optimum conditions observed in filtration and EC experiments.

## **3. Results and discussion**

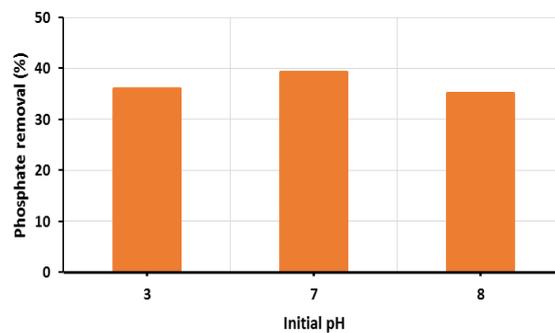
### 3.1. Filtration experiment

#### 3.1.1. Effects of pH

As it is mentioned earlier in the methodology section of this research, pH of 4, 7 and 8 were used to check the impact of pH on the adsorption of phosphates on zeolite particles.

Figure 1 displays the relationship between the pH and phosphates adsorption; results of these experiments indicated that the phosphate removal was increased as the pH grew from 4.0 to 7.0, then it declined again as the pH grew to 8.0.

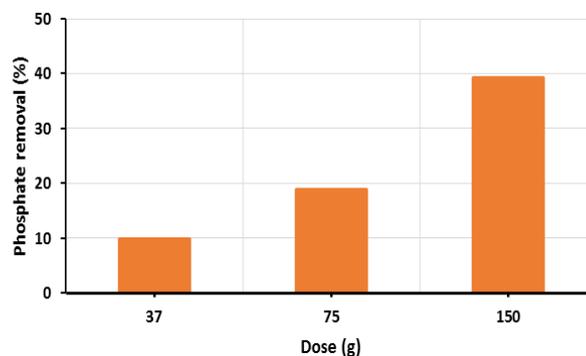
The most acceptable explanation to this variation in the adsorption of phosphates with the pH is the concentrations of  $H^+$  and  $OH^-$  in the bulk solutions substantially increase at high and low pH, respectively, which results in a competition between phosphates and these ions for the adsorption sites and consequently minimises the removal of phosphate [2, 23]. Hence, the authors adopted pH of 7.0 to complete the experimental work.



**Figure 1.** Variation of phosphates adsorption with the pH of water.

#### 3.1.2. Effects of adsorbent dosage

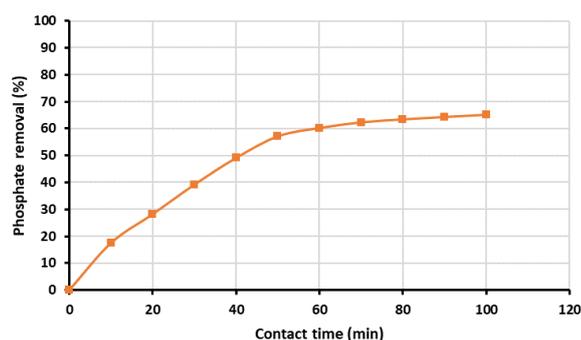
At pH of 7.0, various dosages of zeolites, from 37.0 to 150.0 grams, were added to each one litre of calcium-phosphate solution to check whether it affects the adsorption of phosphate on the zeolite particles or not. A significant link between the dosage of zeolites and adsorption phosphate was observed during the experimental work, as shown in Figure 2. Where, the removal of phosphates was escalated from about 10 to 40% when the dosages of zeolites increased from 37 to 150 grams per litre, respectively. The most acceptable explanation to this variation in the adsorption of phosphates with the pH is the substantial increase in the adsorption area (when the dosage increased) that consequently maximises the adsorption of phosphates [40]. Hence, 150 grams of zeolite (per one litre of phosphate) was used to complete the experimental work.



**Figure 2.** Impacts of the dosages of zeolites on phosphates adsorption.

#### 3.1.3. Effect of contact time

Calcium-phosphate solution ( $20.0 \text{ mg.L}^{-1}$ ) and zeolite particles (150 grams) were contacted in a glass beaker for 100 minutes in order to monitor the progress of adsorption process with detention time. The change in phosphates removal, from calcium-phosphate solution, with the increase of the contact period is displayed in Fig. 3; a substantial increase in the adsorption was detected with the progress of contact time up to 60 minutes. Then, a slight increase was observed over the last 40 minutes. The obvious increase in the adsorption of phosphate during the first 60 minutes is availability of adsorption sites, however over time, these sites became occupied by phosphates or other competing ions that resulted in a decrease in the removal efficiency over the last 40 minutes of treatment [42]. These results evidenced that the reasonable contact time is 60 minutes, which was adopted by the authors for the rest of study.

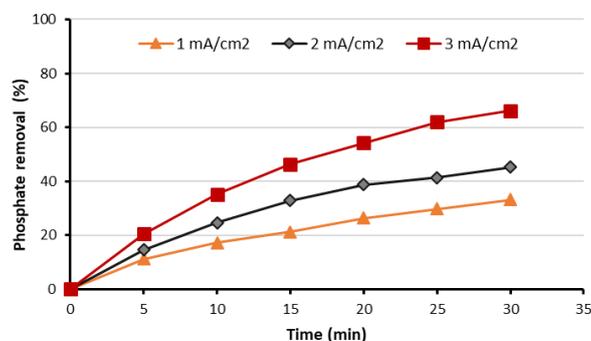


**Figure 3.** Impacts of the contact period on adsorption of phosphate on zeolites.

### 3.2. EC experiments

#### 3.2.1. Effect of current densities and electrolyzing period

In order to study the impacts of both electrolyzing time and current densities on the remediate of phosphates by the aluminium-based EC system form calcium-phosphate solution, three levels of currents ( $1.0$ ,  $2.0$  and  $3.0 \text{ mA.cm}^{-2}$ ) were applied for 30 minutes to the calcium-phosphate solution. All experiments were run at pH of 7. Figure 4 depicts the change in phosphate remediation with both current density and electrolyzing time. Obviously, remediation of phosphate is strongly depending on these two parameters, where the removal of phosphate escalated from 30s% to 60s% (after 30 minutes) when the current density changed from  $1.0$  to  $3.0 \text{ mA.cm}^{-2}$ , respectively. The same behaviour was observed with the progress of electrolysing period. The most acceptable explanation to this variation in the remediation of phosphates with the electrolyzing time and current density is increase in the anodic dissolution (production of coagulants) and consequently it improves the remediation of phosphates [41]. To avoid high power consumption, the authors considered current density of  $2 \text{ mA.cm}^{-2}$  and electrolyzing period of 30 minutes as the reasonable EC conditions, in this research, for phosphate removal.



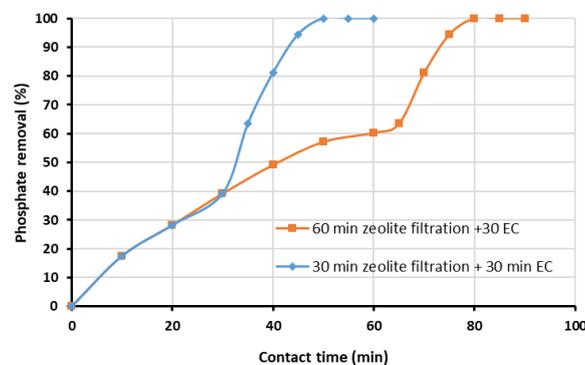
**Figure 4.** Change in phosphate removal with the change in current density and EC time.

### 3.3. Filtration-EC method

The outcomes of the above experiments highlighted an important fact that the goal of a complete removal of phosphates from phosphate-calcium solution is too difficult to be accomplished using a single treatment method. A filtration-EC system therefore has been applied, in the present study, to achieve this goal. This system was operated using the optimum conditions from the filtration and EC experiments.

It was obviously noticed that the filtration-EC system possesses the capacity to completely remediate phosphate (100%) from calcium-phosphate solution after 50.0 minutes, see Figure 5. These outcomes evidence that combination of EC and filtration technologies could be a cost-effective alternative for water treatment in those areas that facing a lack of drinking water due to economic reasons. Additionally, the simplicity of the filtration-EC method could enable the researchers to develop it, for example remote control of this method could be achieved by using sensing and computerising technologies [43-47].

## 4. Conclusions



**Figure 5.** Remediation of phosphate from calcium-phosphate solution by filtration-EC system.

The suggested combined method, filtration-EC method, according to the final outcomes of this research, demonstrated high efficiency in the remediation of phosphates from calcium-phosphate solution. It was found that the efficiency of the filtration - EC system to eliminate phosphate from calcium-phosphate solution is improved when the dosage of zeolite, contact period, electrolysing period and/or current densities are increased. Conversely, the significant increase or decrease of pH negatively influenced the remediation of phosphate using the filtration - EC system. Generally, it was found that neutral pH, 150 mg of zeolite, 30.0 minutes of electrolyzing, 30 minutes of contacting with zeolite and current density of 2 mA. cm<sup>2</sup> are the optimum operation conditions for complete removal of phosphates from calcium-phosphate solution.

Although the this study evidenced the efficiency of the filtration - EC system, more researches are required to evidence the efficiency of this system on large scales as the present research was performed using lab-scale system.

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