Performance evaluation of a continuous bipolar electrocoagulation/electrooxidation–electroflotation (ECEO–EF) reactor designed for simultaneous removal of ammonia and phosphate from wastewater effluent

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ABSTRACT

The present study aimed to evaluate the performance of a continuous bipolar ECEO–EF reactor designed for simultaneous removal of ammonia and phosphate from wastewater effluent. The reactor was comprised of two distinct units: electrochemical and separation. In the electrochemical unit, Al, stainless steel, and RuO_2/Ti plates were used. All the measurements were performed according to the standard methods. Maximum efficiency of the reactor for phosphate removal was 99% at pH of 6, current density of 3 A, detention time of 60 min, and influent phosphate concentration of 50 mg/l. The corresponding value for ammonia removal was 99% at a pH of 7 under the same operational conditions as for phosphate removal. For both phosphate and ammonia, the removal efficiency was highest at neutral pH, with higher current densities, and with lower influent concentrations. In addition to removal of phosphate and ammonia, application of the Al^{3+} plates enabled the removal of nitrite and nitrate, which may be present in wastewater effluent and are also products of the electrochemical process. The reactor was also able to decrease the concentrations of phosphate, ammonia, and COD under actual wastewater conditions by 98%, 98%, and 72%, respectively. According to the results of the present study, the reactor can be used for efficient removal of ammonia and phosphate from wastewater.

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1. Introduction

Incompletely treated industrial and municipal wastewaters contain large quantities of nitrogen and phosphorous, which have harmful effects on aquatic life and human health if discharged into the rivers and estuaries [1,2]. Certain forms of water-soluble inorganic nitrogen, such as ammonia, ionized ammonium, nitrite, and nitrate, may form in wastewater, which can reach groundwater and surface water resources [2,3]. Nitrogen is present as ammonia or ionized ammonium in alkaline or acidic conditions, respectively, and both forms deplete dissolved oxygen through oxidation in water bodies [4,5]. Ammonia has toxic effects on aquatic life [4,6,7]; it also encourages eutrophication in receptor water bodies [4,8,9]. Phosphorous is one of the basic nutrients promoting algal blooms in rivers, lakes, coastal areas, and estuaries, which finally leads to eutrophication; this phenomenon causes depletion of oxygen levels in water via algal decay, which has harmful effects on aquatic life [10–13]. The principal phosphorous compounds in the wastewater effluents are generally orthophosphates [14]. Most commonly, phosphate and ammonia nitrogen are simultaneously present in wastewater effluents; therefore, wastewater effluents with high concentrations of ammonia and phosphate must be efficiently treated prior to discharge into the environment.

Various methods have been used for phosphate removal from wastewater, including biological treatment, adsorption, chemical precipitation, ion exchange, electrodialysis, and membrane filtration [10,14]. Because the efficiency of phosphate removal through biological treatment does not exceed 30%, complementary techniques are required for removal of the residual phosphate in wastewater effluent [10]. Chemical precipitation can also be used; however, this method has several disadvantages, including high capital and operational costs, sludge production in high volumes, the requirement for additional manpower, and appli-

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cation of chemical compounds. In addition, when aluminum and iron are used as coagulants in chemical precipitation, alkaline conditions must be maintained during the operation [10,14]. Ammonia removal is also achieved by biological nitrogen removal (BNR), air stripping, and ion exchange [4,9]. BNR is the common treatment for low ammonia concentrations in wastewater. Nevertheless, this method is not effective when high concentrations are present because high levels of ammonia inhibits the process [8]. Moreover, these processes are not capable of significantly reducing the ammonia concentrations. Furthermore, transferring the pollutants to another media poses high operational costs [9]. Therefore, in recent years, there has been an increasing interest in the application of electrochemical technologies for wastewater treatment in the removal of organic compounds and heavy metals [10,14–19].

The most common processes in wastewater treatment based on electrochemical technologies are electrocoagulation (EC), electrooxidation (EO), electroflotation (EF), and electrodecantation [17,20]. Electrocoagulation–electroflotation (EC–EF) has been used by other researchers and was found to be a promising and efficient technology for wastewater treatment [21–23]. Electrocoagulation has also been successfully used for phosphate removal from wastewater effluents [10,11,14]. In addition, ammonia removal from wastewater effluent was achieved through an electrooxidation process [9]. Based on these previous studies, phosphate and ammonia removal can be achieved through electrocoagulation and electrooxidation, respectively, but the removal mechanisms for these two pollutants are different. However, among the studies mentioned only either of the pollutants has been removed by an electrochemical process. In addition, because electrochemical technology has many advantages over the conventional methods, it is necessary to develop a combined system capable of removing both of the pollutants from wastewater effluent simultaneously.

Therefore, the present study sought to develop a combined system based on electrochemical technologies (ECEO–EF) that is capable of removing both phosphate and ammonia from wastewater effluents. Aluminum and iron are the most common sacrificial plates used in the electrocoagulation process, but the higher performance of aluminum plates compared to iron plates has been well documented [14,19,22]. Plates can operate as bipolar or monopolar in the electrocoagulation process, but a simple set up provided by the bipolar mode facilitates easy maintenance during the operation [17,24]. Hence, a bipolar configuration was used in this study. Finally, we aimed to evaluate the performance of the continuous, combinative bipolar ECEO–EF reactor developed for the removal of phosphate and ammonia under different operational conditions of pH, voltage (V), and detention time.

2. Experimental

2.1. Experimental set up

The design of the continuous bipolar ECEO–EF reactor used in the present study is shown in Fig. 1. The plates consisted of nine pieces of Al and 2 pairs of RuO_2/Ti and stainless steel. The bipolar electrochemical cell consisted of three aluminum plates placed between a pairs of an RuO_2/Ti anode and a stainless steel (SS) cathode, and only the two pairs of plates were connected to the D.C. power supply with 0–60 V of electrical potential. The reactor was separated by a glass wall into electrochemical and separation units with 10 and 15 L in volume, respectively. The baffles at the beginning of the first unit and the middle of the second unit were made to equalize the influent velocity and facilitate separation of the scum, respectively. The effective surface area for each plate was 180 cm²
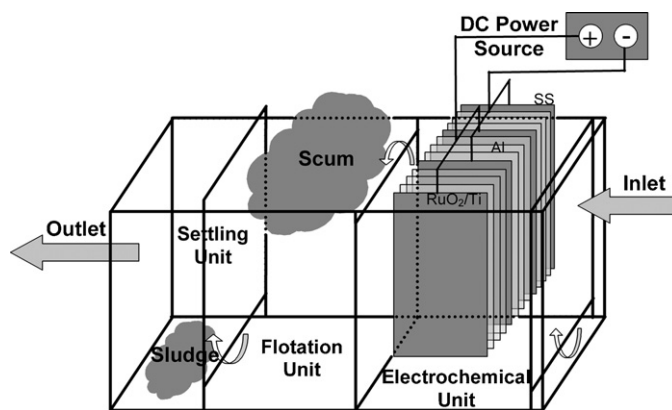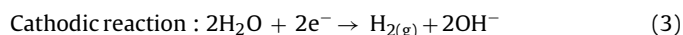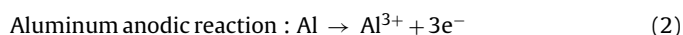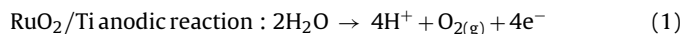

Fig. 1. Schematic design of the ECEO–EF reactor.

(18 cm × 10 cm), and the space between the plates was 8 mm. The efficiency of the reactor was first tested at initial phosphate and ammonia concentrations of 50 mg/l and different operational conditions of pH (5–10), current density (0–3 A), and detention time (10–60 min). After determining the optimum operational conditions, the maximum efficiency of the reactor was tested under different influent concentrations of phosphate and ammonia (15, 30, 50, and 100 mg/l).

2.2. A brief description of the combined ECEO–EF process

When the electric current passed through the two monopolar plates, it charged the neutral sides of each aluminum plate to generate negative and positive regions relative to the adjacent charged sides (Fig. 2). The RuO_2/Ti anode and the SS cathode are non-sacrificial plates and produce hydrogen and oxygen gases under anodic and cathodic reactions in the reactor, respectively. The main reactions occurring at the surface of the plates are as follows:

$$RuO_2/Ti \text{ anodic reaction} : 2H_2O \rightarrow 4H^+ + O_{2(g)} + 4e^- \quad (1)$$

$$\text{Aluminum anodic reaction} : Al \rightarrow Al^{3+} + 3e^- \quad (2)$$

$$\text{Cathodic reaction} : 2H_2O + 2e^- \rightarrow H_{2(g)} + 2OH^- \quad (3)$$

The main reactions occurring in the bulk solution are as follows:

$$Al_{(aq)}^{3+} + 3H_2O \rightarrow Al(OH)_3 + 3H^+ \quad (4)$$

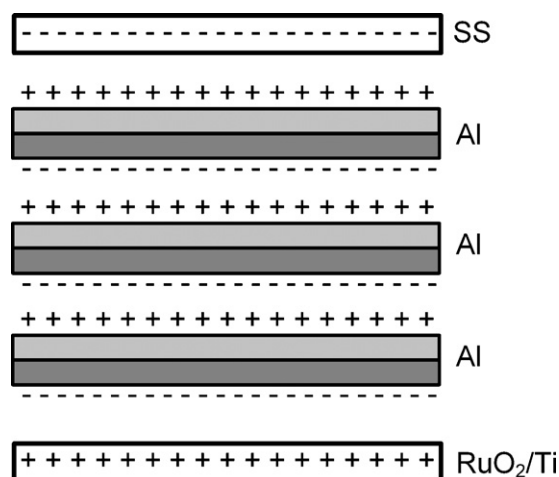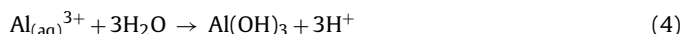$$Al_{(aq)}^{3+} + PO_4^{3-} \rightarrow AlPO_4 \quad (5)$$

Fig. 2. Arrangement of the Al plates between SS and RuO_2/Ti plates.

The products of Eqs. (4) and (5) participate in phosphate removal, while AlPO$_4$ is believed to be more effective at pH values < 6.5 [14].
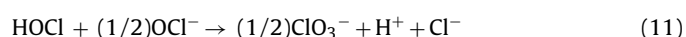
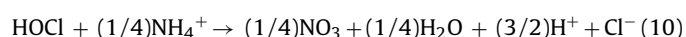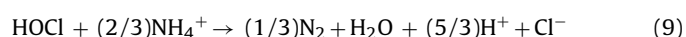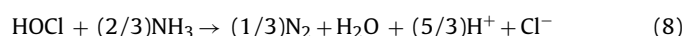Electrolytic oxidation of the anode side of the aluminum plates causes generation of coagulant Al^{3+} in the reactor. In addition, cathodic and anodic reactions generate hydrogen and oxygen gases, respectively, which are involved in the floatation process [25]. These tiny bubbles (average diameter of approximately 20 μm [25]) attach to the flocs formed and float them to the surface of the wastewater. Additionally, the oxygen produced through the anodic reactions improves the anaerobic conditions of the wastewater. Moreover, hydrolysis and polymerization of Al^{3+} lead to formation of gelatinous charged hydroxo-cationic complexes, which are able to remove pollutants through adsorption and charge neutralization [26,27].

The principal mechanisms suggested for ammonia removal are direct and indirect oxidation [9,15,28,29]. Based on studies that have investigated ammonia removal by oxidation, it has been shown that indirect oxidation of ammonia requires the presence of Cl$^-$ in wastewater, while direct oxidation of ammonia occurs at the anode liquid interface [9,30]. It is noteworthy that indirect oxidation is the main process for ammonia removal in the electrochemical oxidation process [31,32].

The following reactions indicate the indirect oxidation of ammonia by HOCl [9]:

$$\text{At the anode}: 2Cl^- \rightarrow Cl_2 + 2e^- \tag{6}$$

In the bulk solution:

$$Cl_2 + H_2O \rightarrow HOCl + H^+ + Cl^- \tag{7}$$

$$HOCl + (2/3)NH_3 \rightarrow (1/3)N_2 + H_2O + (5/3)H^+ + Cl^- \tag{8}$$

$$HOCl + (2/3)NH_4^+ \rightarrow (1/3)N_2 + H_2O + (5/3)H^+ + Cl^- \tag{9}$$

$$HOCl + (1/4)NH_4^+ \rightarrow (1/4)NO_3 + (1/4)H_2O + (3/2)H^+ + Cl^- \tag{10}$$

$$HOCl + (1/2)OCl^- \rightarrow (1/2)ClO_3^- + H^+ + Cl^- \tag{11}$$

In addition, presence of chloride ions (Cl$^-$) has critical advantages, particularly when using aluminum plates. It has been suggested that chloride ions are able to breakdown the alumina film (Al$_2$O$_3$) through pitting corrosion. The alumina film is usually formed at the aluminum anode surface, and it causes increased power consumption and decreased process efficiency because it can inhibit the release of Al^{3+} ions as well as electron transfer. Therefore, the efficiency of the system can be considerably increased via the breakdown of the alumina film (Al$_2$O$_3$) [33].

2.3. Chemicals

All chemical compounds used in the present study were of analytical grade. Phosphate and ammonia solutions were prepared by dissolving KH$_2$PO$_4$ and (NH$_4$)$_2$SO$_4$ in distilled water, respectively. NaCl was used for production of Cl$^-$ ions in the reactor during the experiments. Na$_2$SO$_4$ was added to the synthetic wastewater samples to reach the electrical conductivity of the actual wastewater samples collected. During the study, pH was adjusted by adding 0.1 N H$_2$SO$_4$ and/or 0.1 N NaOH when necessary. Finally, the concentrations of phosphate, ammonia, nitrate, and Cl$^-$ were measured according to the standard methods [34].

3. Results

3.1. Performance of the reactor for the removal of phosphate

As mentioned in Section 2, the efficiency of the ECEO–EF reactor for phosphate and ammonia removal was tested under different operational conditions of pH, current density, and influent concentrations. The optimum operational conditions are illustrated in the figures presented below. Fig. 3(a) depicts the efficiency of removal of phosphate at different pHs and detention times with an influent phosphate concentration of 50 mg/l and a current density of 3 A. For all detention times, the maximum efficiency of the reactor for phosphate removal was observed in neutral conditions, and the removal efficiency reduced at both lower and higher pH levels. For example, with a detention time of 20 min, a maximum removal efficiency of 90% was observed at a pH of 6, and the efficiency decreased to 85% and 76% at pH values of 5 and 10, respectively. In addition, it can be seen that the average removal efficiency was nearly 50% when the detention time was 10 min, however it increased to over 85% when the detention time was 20 min, and it remained almost constant afterwards. This implies that phosphate removal mainly occurred in the first 20 min of the experiments, and increasing the detention time did not have a considerable impact.
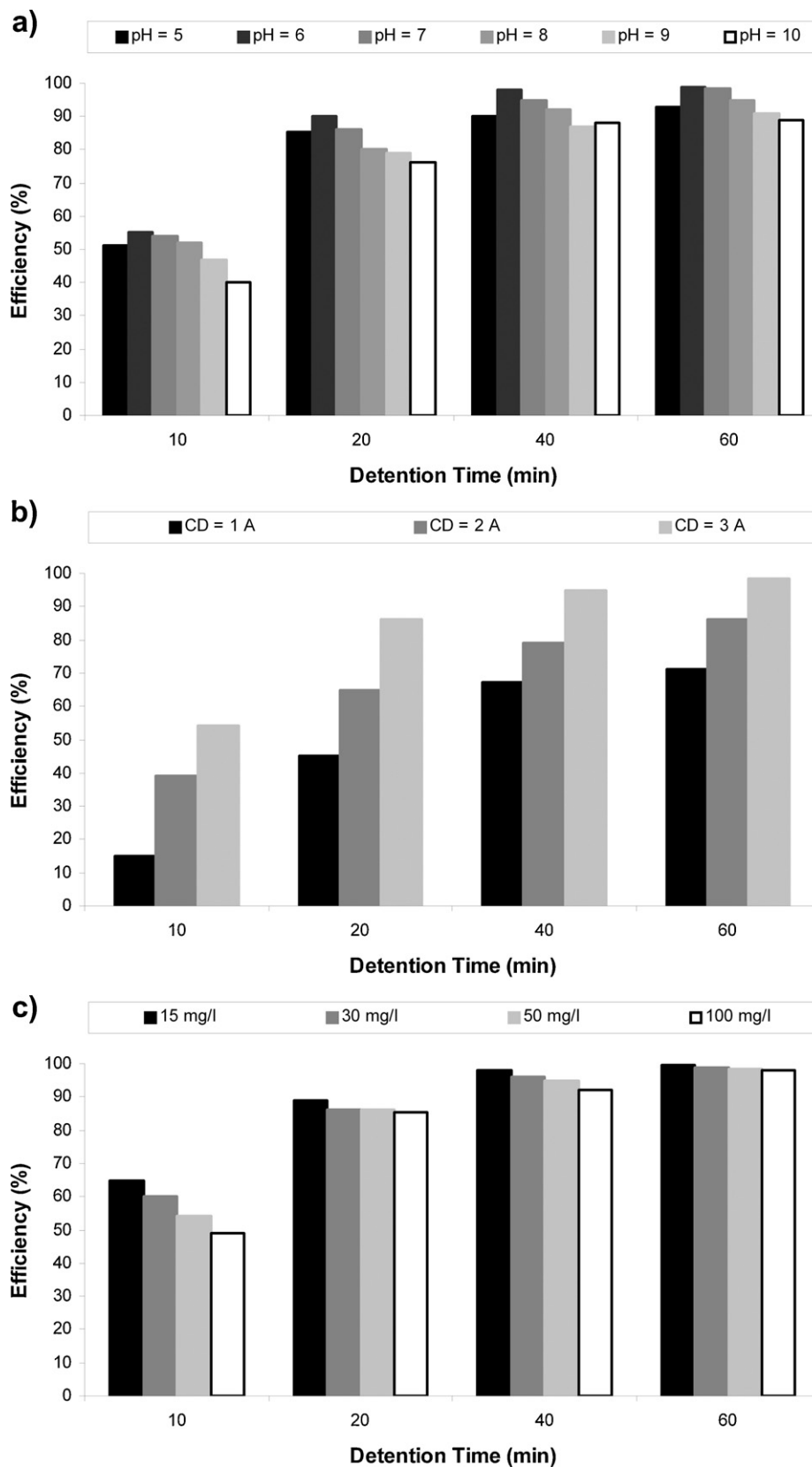
Fig. 3(b) shows the phosphate removal efficiency under different current densities (CD) and at different detention times with an influent phosphate concentration of 50 mg/l and an optimum pH of 7, indicating the effect of current density on phosphate removal. As shown in Fig. 3(b), the removal efficiency of phosphate increased with increasing current density, and the optimum removal efficiency (98.5%) was observed at a current density of 3 A.

After determining the optimum operational conditions with respect to pH (neutral) and current density (3 A), these parameters were held constant and the removal efficiency of phosphate was assessed under different influent phosphate concentrations, and the results are illustrated in Fig. 3(c). For low detention times, the reactor had higher efficiencies with lower influent concentrations of phosphate. For example, with a detention time of 10 min, the removal efficiency of the reactor was slightly under 50% for an influent concentration of 100 mg/l, while it increased to well over 60% for an influent concentration of 15 mg/l. On the other hand, with higher detention times, increasing the influent phosphate concentrations did not have a significant effect on the performance of the reactor. For example, with a detention time of 60 min, it can be seen that regardless of the influent phosphate concentration, the removal efficiency was quite high (approximately 90%). Hence, it can be implied that the reactor works well even under high influent concentrations of phosphate, provided that the necessary detention time is provided.

3.2. Performance of the reactor for the removal of ammonia

Fig. 4(a) illustrates the effect of pH on the efficiency of the reactor for ammonia removal at different detention times with an influent ammonia concentration of 50 mg/l and a current density of 3 A. As with phosphate, the maximum removal efficiency was observed at a neutral pH, though no significant difference was observed over the entire range of pH values from 5 to 10. For instance, with a detention time of 60 min, the maximum removal efficiency of 99% was observed at a pH of 7, and it remained as high as approximately 95% over the entire range of pH values mentioned above. In contrast to phosphate, however, higher detention times were required to obtain the optimum removal efficiency. As can be seen from Fig. 4(a), the average removal efficiency for ammonia was as low as slightly below 60% with a detention time of 20 min, while it significantly increased to over 90% at 40 min and remained almost unchanged afterwards. This indicates that 40 min (vs. 20 min for phosphate) is the optimum detention time for ammonia removal.

Fig. 4(b) shows the effect of current density (CD) on the removal of ammonia with different detention times at an influent ammonia concentration of 50 mg/l and optimum pH of 7. Similar to phosphate, higher current densities led to increased removal efficiencies for all detention times. For example, with a detention time of 40 min, the maximum removal efficiency of 97% was observed with
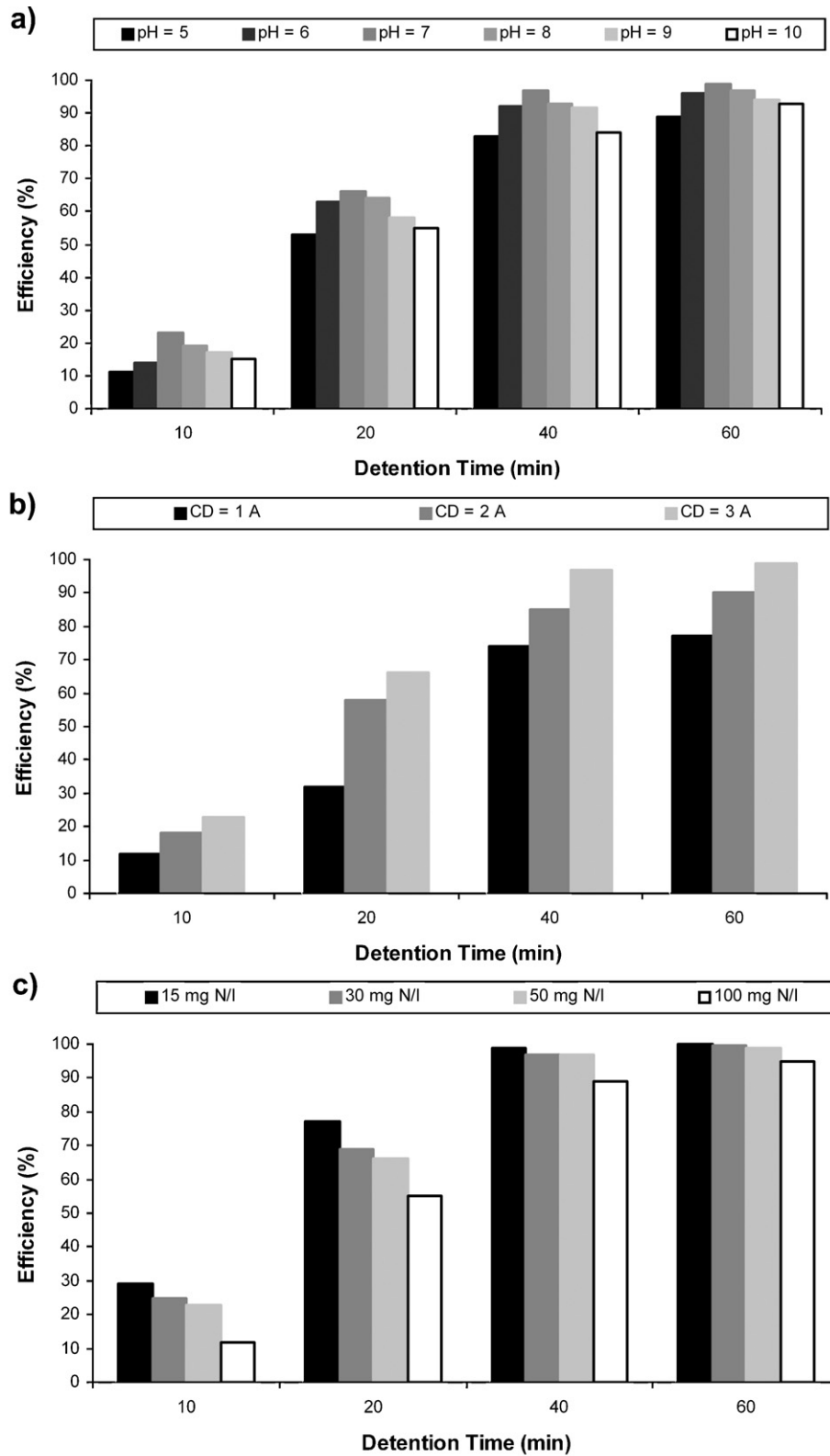
Fig. 3. Phosphate removal efficiencies of the reactor with different detention times under different operational conditions of pH (a), current density (b), and influent phosphate concentrations (c).

a current density of 3 A, however it was as low as 74% with a current density of 1 A.

After obtaining the optimum operational conditions with regard to pH (neutral) and current density (3 A), the removal

efficiency of ammonia was evaluated under different influent ammonia concentrations. Fig. 4(c) illustrates the effect of influent ammonia concentrations on the removal efficiency of the reactor. Although the performance of the reactor was quite

Fig. 4. Ammonia removal efficiencies of the reactor in different detention times under different operational conditions of pH (a), current density (b), and influent phosphate concentrations (c).

sensitive to the influent ammonia concentrations when low detention times were used, higher detention times resulted in a sufficiently high removal efficiency that was independent of variations in the influent concentrations. For example, with a detention time of 10 min, the removal efficiency was nearly 30%

when the influent concentration was 15 mg/l, and it decreased to approximately 10% when the influent concentration was 100 mg/l. In contrast, with a detention time of 60 min, the removal efficiency remained over 95% regardless of the influent concentration.
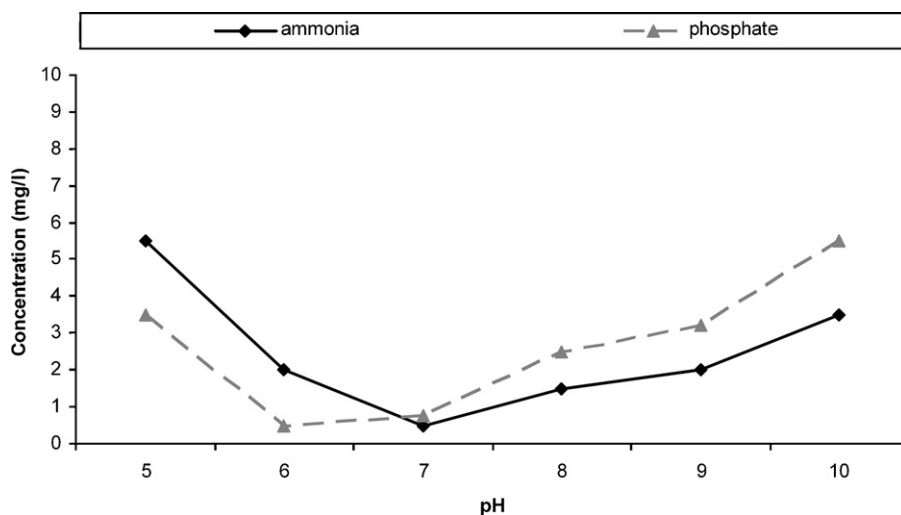
Fig. 5. The effect of pH on the removal of phosphate and ammonia from wastewater.

Fig. 5 provides a different view of the effect of pH on the removal efficiencies for both phosphate and ammonia. As previously mentioned, the maximum phosphate removal efficiency was observed at a pH of 6, while the optimum pH for ammonia removal was 7. However, because both phosphate and ammonia are to be removed in the same reactor, 7 can be considered as the optimum pH for removal of both pollutants. This is acceptable because phosphate removal was quite effective, even at a pH of 7.

In addition to ammonia, concentrations of nitrate and nitrite were also measured for different detention times, and the results are illustrated in Fig. 6. It can be seen that ammonia concentrations diminished with increasing detention times. With a detention time of 10 min, the ammonia concentration was slightly below 40 mg/l, while it decreased to below 20, 2, and 1 mg/l with detention times of 20, 40, and 60 min, respectively. In addition, nitrite and nitrate concentrations were negligible and below detection limits for all of the detention times.

3.3. Performance of the ECEO–EF reactor under actual wastewater conditions

For evaluating the performance of the ECEO–EF reactor under actual wastewater conditions, samples were taken from the effluent of an anaerobic reactor. The mean characteristics of the wastewater were as follows: an ammonia concentration of 28 mg/l, a phosphate concentration of 48.3 mg/l, a Cl^- concentration of 910 mg/l, and an electrical conductivity of 1600 $\mu s\,cm^{-1}$. The performance of the reactor was tested under the optimum operational conditions determined for the synthetic wastewater samples (pH of 7, current density of 3 A, and detention time of 60 min). It was found that the reactor was able to decrease the ammonia concentration by 98% (effluent concentration of approximately 0.56 mg/l) and the phosphate concentration by 98% (effluent concentration of 0.96 mg/l). In addition, the reactor was able to decrease the chemical oxygen demand (COD) from 35 to 10 mg/l (efficiency of approximately 72%).

4. Discussion

The present study sought to evaluate the performance of a continuous bipolar electrocoagulation/electro-oxidation–electroflotation (ECEO–EF) reactor designed for simultaneous removal of ammonia and phosphate from wastewater effluent under different operational conditions of pH, voltage (V), detention time, and influent concentrations of either of the pollutants. According to the results of this study, although the optimum operational conditions might be different for removal of phosphorous and ammonia, the reactor was found to be capable of effectively removing both the pollutants from wastewater.

For phosphate removal, the optimum efficiency of the reactor was obtained at a pH of 6, a current density of 3 A, an influent phosphate concentration of 15 mg/l, and a detention time of 20 min. A neutral pH was found to be optimal for phosphate removal, and phosphate precipitation was mainly due to formation of $AlPO_4$ (as mentioned in Section 2.2) [14]. This is consistent with the results of previous studies. For example, it has been suggested that phosphate removal efficiencies higher than 90% can be obtained over a wide range pH values from 5 to 9. In addition, in a study that investigated phosphate removal by electrocoagulation, the optimum removal efficiency was achieved at a neutral pH. Furthermore, the reaction time for efficient phosphate removal was only 20 min [10]. In the case of current density, higher removal efficiencies can be attributed to increased release of Al^{3+} ions when higher current densities are used [11]. Similar results were obtained in other studies. In a study that assessed the effect of current density and phosphate concentration on the efficiency of phosphate removal by an electrocoagulation process, it was found that the current density has a positive impact on the removal of phosphate from wastewater [11], though increased current density may increase power consumption. Regarding influent phosphate concentrations, the reactor was more efficient at lower influent concentrations. However, increasing the influent phosphate concentrations did not have a significant effect on the reactor performance, implying that the reactor works well even with high influent concentrations if the necessary detention time is provided. It was also found that the majority of the phosphate was removed from wastewater with a detention time of 20 min, which is consistent with the results from the study of Irdemez et al. [11].

For ammonia removal, the optimum efficiency of the reactor was obtained at a pH of 7 (generally high efficiency was observed over the pH range from 5 to 10), a current density of 3 A, an influent phosphate concentration of 15 mg/l, and a detention time of 40 min. With respect to the optimum pH range for phosphate removal, similar results were obtained by the study of Vanlangendonck et al. [35]. In this study, we found that the optimum pH range for removal of ammonia is 5.5–10. Similar results were also observed by Chiang et al. [32] and Li and Liu [9]. However, according to Eq. (11), in strongly alkaline conditions, HOCl, which is a strong oxidizing agent, is changed to ClO_3^-, which has a lower oxidizing potential than HOCl. Therefore, strong alkaline conditions decrease the
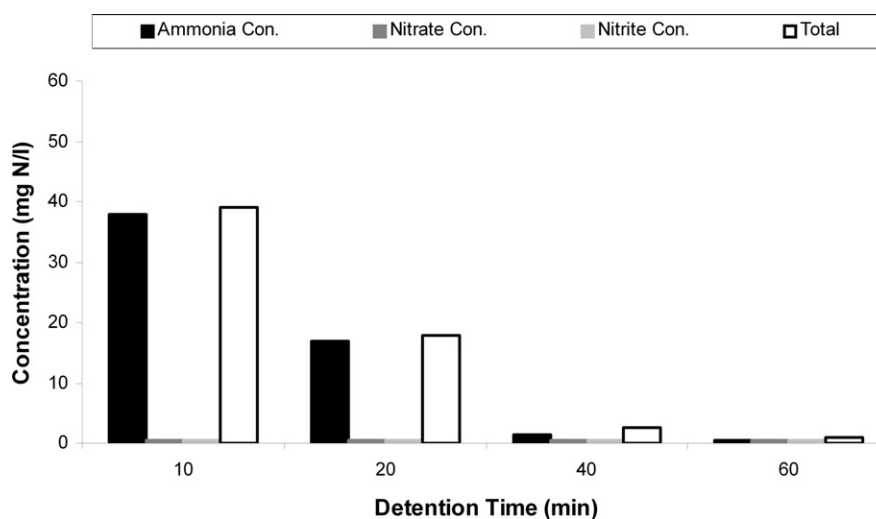
Fig. 6. Concentrations of nitrate, nitrite, ammonia, and total nitrogen in the reactor effluent with different detention times.

oxidation rate of ammonia and reduce its removal efficiency [36]. Therefore, neutral pH is suggested for ammonia removal due to the presence of higher concentrations of HOCl. As with phosphate, ammonia removal was also more efficient with higher current densities. This is mainly due to the fact that higher current density leads to increased production of Cl_2 and a subsequent increase of HOCl, which leads to higher ammonia oxidation rates [9]. This is also in agreement with results described in other studies. In a study investigating the removal of ammonia by electrochemical oxidation, a higher current density was found to lead to increased Cl_2 production at the anode surface according to Eq. (6), which increased the ammonia oxidation rate [9]. With respect to influent ammonia concentrations, the ammonia removal efficiency increased with decreasing influent concentrations, though removal efficiencies for high concentrations were still desirable, especially when higher detention times were used (>40 min), implying the high efficiency of the reactor for ammonia removal even in high influent concentrations of ammonia.

In addition, it can be implied from Fig. 4(a) that removal rate for ammonia is lower than for phosphate since the majority of the ammonia was removed in 40 min (compared to 20 min for phosphate (Fig. 3(a)). There are several explanations for this result. First, because the presence of phosphate has a negative effect on ammonia removal due to oxygen evolution [35], the removal rate of ammonia remains low when significant phosphate concentrations exist in the reactor (detention time of 20 min). However, when the majority of the phosphate is removed (detention time of >20 min), ammonia removal reaches higher levels. Second, the longer detention time required for ammonia removal may be due to the slower reaction rate of ammonia with HOCl. Third, because phosphate transfers more electric current than Cl^- due to its higher electric charge, Eq. (6) cannot effectively occur at the anode surface when high concentrations of phosphate are present. This reduces the amount of HOCl production (Eq. (7)), which in turn reduces the oxidation rate of ammonia by HOCl [35].

As with phosphate, sulfate ions have the same inhibitory effect on the removal of ammonia from wastewater. However, this effect was negligible due to low sulfate concentrations in the samples. In addition, a high chloride concentration of 1000 mg/l was applied in the present study to neutralize the negative effect of phosphate and sulfate ions on ammonia oxidation. Because chloride ions act as a catalyst, they are not consumed in the reaction (Eq. (6)–(11)). Therefore, the chloride concentration in the effluent was almost equal to that in the influent (940 mg/l). This difference might cause chloride ions to be released as gas or to participate in the reactions, leading to the formation of chloramines [9].

Finally, it is noteworthy that the reactor was effective under actual wastewater conditions. Therefore, this reactor can be effectively used for the simultaneous removal of phosphate and ammonia from wastewater effluents. In addition, because of the especial design of the reactor (i.e., continuous flow), it is more applicable (compared to batch reactors) for use in wastewater treatment plants, in which continuous flow usually exists.

5. Conclusions

The main objective of the present study was to evaluate the performance of the ECEO–EF reactor for the removal of ammonia and phosphate from wastewater effluent. According to the results of this study, we found that the reactor is capable of sufficiently removing both phosphate and ammonia, which are believed to be two of the most important pollutants present in wastewater. For both phosphate and ammonia, the performance of the rector was optimum at a neutral pH. Increasing the current density and detention time had positive impacts on the overall performance of the reactor. In addition, it was found that the reactor had high performance even when high concentrations of phosphate and ammonia are present in the influent. The reactor efficiently removed phosphate, ammonia, and COD under actual wastewater conditions. Finally, based on the advantages of electrochemical technologies for the removal of a wide range of environmental pollutants, it is suggested that further studies should be conducted to obtain the optimum operational conditions for removal of a variety of pollutants that can be removed by the same mechanism.

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