

Urea removal using electrocoagulation process with copper and iron electrodes

Mostafa Mamdouh^a, Safwat M. Safwat^{b,*}, Hisham Abd-Elhalim^b, Ehab Rozaik^b

^aEnvironmental Engineering Department, Dar Al-Handasah, Cairo, Egypt, email: mos.mamdouh@gmail.com

^bSanitary and Environmental Engineering Division, Faculty of Engineering, Cairo University, Giza, Egypt, Tel. +201092525159; emails: safwat@eng.cu.edu.eg (S.M. Safwat), hishama.halim2011@gmail.com (H. Abd-Elhalim), ehab.helmy@dar.com (E. Rozaik)

Received 5 February 2020; Accepted 13 October 2020

ABSTRACT

The ability to remove urea from wastewater was investigated using an electrocoagulation process (EC). Two electrodes were examined: iron and copper. Several parameters were investigated to demonstrate the performance of the system. These parameters include voltage, electrolyte type, and the gap distance. Results showed that the removal efficiencies reached 40.15% at 6 V for the copper electrode and 51% at 12 V for the iron electrode, after 90 min. In a comparison of EC with chemical coagulation, EC was found to perform better. Fourier transform infrared and scanning electron microscopy were performed to investigate the characteristics of the produced sludge and the electrode surface. The electric energy consumption for iron and copper electrodes were 112.8 and 32.4 kWh/m³, respectively. Results showed that more iron electrode was consumed than copper electrode under the same conditions.

Keywords: Electrocoagulation; Chemical coagulation; Urea; Voltage; Wastewater

1. Introduction

Urea is present in the ecosystem as a contaminant not only from wastewater production but from many other sources, such as leakage from farms, effluent from plants using it as a raw material, and as a final product of mammalian protein metabolism [1,2]. While urea itself has low toxicity, it decomposes into ammonia and hydrogen, which can be toxic in natural marine environments. Many techniques have been used to remove urea from wastewater, including the hydrolysis process, enzymatic decomposition, adsorbent removal, decomposition by a strong oxidant, removal by biological treatment, and use of a catalyst [1,3]. Electrochemical oxidation of urea has various advantages over other techniques, as it does not require the addition of large amounts of chemicals to wastewater or a supply of oxygen for the cathode. Furthermore, the production of

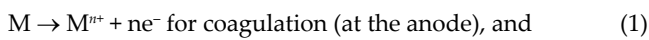
secondary pollution is unlikely to occur, and the system setup is simple. Because of these benefits, electrocoagulation is considered a better technology than other oxidation processes [4,5].

The theory behind the electrocoagulation process depends on destabilizing repulsive forces that keep particles suspended. In addition, the mechanism for removing these particles from wastewater begins when the flocs have formed because of the destabilization of repulsive forces, and these flocs induce the suspended particles to settle out of solution [4–7].

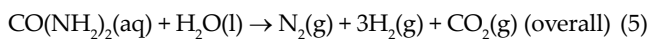
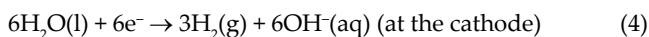
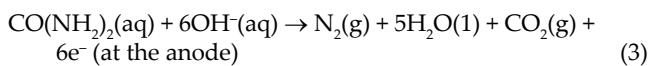
Throughout the electrocoagulation process, a solution containing an electrolyte is used as a conductive medium. Two electrodes are immersed in this solution, and a direct voltage is applied to the electrodes [4–7]. The electrocoagulation process consists of three main phases: (i) anode oxidation, (ii) cathode gas bubble formation, and

* Corresponding author.

(iii) sedimentation and flotation of the generated flocs. Two reactions occur as soon as an electric current act on the electrodes: an oxidation reaction occurs at the anode that generates cations, and reduction reactions occur at the cathode [4–7]. The cations produced in the oxidation reactions form metal hydroxides, which are responsible for the destabilization of suspended solids. At the cathode, hydrogen gas is generated continuously, leading to particle flocculation. The floatation or washing out of the pollutants is achieved by the flocculated particles [4–7]. Charge neutralization, adsorption, or sweep coagulation are considered other pollutant removal techniques [4–7]. The reactions occurring at the electrodes during the electrocoagulation are as follows (where M refers to the metal in the electrodes):



Similarly, the urea electro-oxidation reactions occur as follows [4]:



Electrocoagulation was able to remove several types of pollutants from aqueous solutions, such as phenols, heavy metals, domestic wastewater, industrial wastewater, and other pollutants [8–11]. Several electrodes can be used as anodes for the electrocoagulation process, such as aluminum, iron, copper, zinc, and titanium [5,10,12,13]. In most modern literature reviews of the electrochemical treatment of urea, special attention is given to the use of various types of anode materials such as boron-doped diamond, nickel, platinum, ruthenium-titanium oxide, or other boron-doped electrodes. Studies have investigated the ability of the electrocoagulation process to remove urea from aqueous solutions. When using aluminum and titanium electrodes, the removal efficiencies were 40% and 59%, respectively [14]. When using zinc electrodes, the removal efficiency was 66% [2]. Since electrocoagulation showed promising results regarding the ability to reduce the concentration of urea in aqueous solutions, it is important to further investigate this process to reach the optimum design that maximizes the removal efficiency of urea [2,14]. One of the parameters that widely affects the performance of the electrocoagulation process is the types of electrodes. Therefore, it is necessary to investigate the performance of new anode materials for urea removal. To the best of our knowledge, this research is the first to investigate iron and copper anodes for the treatment of urea-contaminated water streams by electrocoagulation. In this study, tests and experiments were conducted using a bench-scale electrocoagulation cell to study the removal of urea. The removal rates of urea and pH were investigated by studying the effects of changing voltage, the type of the electrolyte, the anode material, and gap distance

between electrodes when the urea is dissolved in the synthetic or real wastewater.

2. Materials and methods

2.1. Characteristics of wastewater

Two wastewater samples were used in this research. The first sample of synthetic wastewater was created by mixing and dissolving urea (99% purity) and sodium chloride (99% purity) into distilled water to obtain urea and sodium chloride concentrations of 1 and 0.40 g/L, respectively. The second sample was collected from a wastewater treatment plant in Cairo, Egypt. The characteristics of the real wastewater containing urea are shown in Table 1.

2.2. Electrocoagulation system setup

The experiment was processed in a batch reactor consisting of a glass beaker with magnetic stirring for 90 min at atmospheric pressure and a temperature of $22^{\circ}C \pm 2^{\circ}C$. A DC power supply (3–12 V) was used to control the electrical current applied to the two electrodes that were parallel to each other with a gap distance of 4.50 cm. A surface area of 30 cm² was used for the iron, copper, and stainless steel (SS) electrodes, and 21 cm² of this area was immersed in the solution. Stainless steel was used as the cathode material, and either iron (Fe) or copper (Cu) was used as the anode material. In order to prevent the shearing of the flocs, the stirring speed was kept at a low level of 100 rpm. The electrolysis duration period was 5–90 min. Distilled water was used to wash and clean the electrodes before they were used to remove any impurities on the surface of the electrodes. Samples were withdrawn periodically: every 5 min for the first 20 min of the experiment, then every 10 min for the 40 min following the first 20 min, at 60 min, and finally at 90 min. Filtration of the withdrawn samples was necessary to remove any sludge formed during electrolysis. All experiments were conducted for each anode type to assess and examine the effect of changing the spacing between the electrodes from 4.5 to 3 cm, changing electrolyte type, increasing the surface area of the anode, and finally using the system with real wastewater for urea removal.

2.3. Analysis

Several analytical measurements were performed on the influent and effluent to assess the removal treatment of urea over time: (i) calculating the urea removal efficiency (percentage) after the process of electrocoagulation according to removal efficiency percentage = $(C_0 - C_e) \times 100/C_0$

Table 1
Characteristics of real wastewater

Parameter	Unit	Value
pH	–	7.68
COD	mg/L	150
Urea	mg/L	1,000
Total dissolved solids (TDS)	mg/L	570

where C_0 represents the influent concentration of urea, while C_e represents the effluent concentration of urea; (ii) analysis of the sludge formed using a Fourier-transform infrared (FTIR) spectrometer; and (iii) inspection of the anode morphologies using a scanning electron microscopy (SEM). Conventional chemical coagulation has been conducted and was used for a comparative analysis with the electrocoagulation process using jar tests. Copper sulfate and ferric sulfate were used in chemical coagulation to simulate copper and iron electrodes. Conventional chemical coagulation included flash mixing for 90 s at 100 rpm followed by a gentle mixing for 20 min at 30 rpm, and a 20 min settling duration. Samples were then collected for further analysis [15]. Fig. 1 shows the setup of the electrocoagulation unit.

3. Results and discussion

3.1. Dependence on voltage for the EC process

The voltage applied between the electrodes is responsible for generating hydrogen bubbles, and the accumulation and growth of flocs encourages the electrode dissociation in order to generate coagulant species [4–7,15]. The performance of the iron and copper electrodes was analyzed at 3, 6, 9, and 12 V. Fig. 2 shows the variation and comparison of the urea removal efficiencies vs. the treatment time for the two electrodes at all operational voltages. As presented in these Fig. 2, for all applied voltages, the rate of urea removal rapidly increased during the first 30 min of the electrocoagulation process for both electrode types. After 30 min, the desorption phenomenon reduces the rate of urea removal as well as the removal efficiencies of the electrocoagulation process. In addition, the presence of oxide layers on the anode surface causes the passivation effect, which affects the efficiency of the electrocoagulation process [4–7,15]. These oxide layers form because of the oxidation reactions. Although the oxidation reaction encourages the corrosion phenomena, it may also stabilize the oxide layers on the anode surface. The greatest removal efficiencies for urea occurred after 90 min with values of 51% at 12 V for iron and 40.15% at 6 V for copper. Thus, the iron electrode performed better than the copper electrode. The current efficiencies were found to be 52% and 72% for iron and copper electrodes, respectively. The applied voltage affects the rate at which ions are released in the form of Cu^{2+} or Fe^{2+} , which affects the resulting coagulation

rate. Consequently, the metal hydroxide generation rate increases in the wastewater, leading to an increase in urea removal efficiencies. The pollutant adsorption phenomena will also occur on the surfaces of metal hydroxides, oxides, and oxyhydroxides [4–7,15]. Pollutant reduction may also occur as a result of the destabilization mechanism, which consists of three procedures: compression of the double layer, charge neutralization, and floc formation [4–7,15]. In addition, the production of hydrogen bubbles into solution helps to remove the pollutants through the flotation process. Furthermore, increasing the voltage applied to the anode leads to an increase in the rate of hydrogen bubble production, as well as a reduction in the bubble size. When the copper electrode was used as an anode, the maximum efficiency of urea removal did not occur at the maximum voltage. One explanation of this phenomenon might be that when a higher voltage is applied to the system, the turbulence of the system increases. When the turbulence in the system is very high, the process of coagulation is unstable and the agglomeration of the particles is affected by this instability, as the particles do not have enough time to accumulate and remove pollutants. During the electrocoagulation process, pH values increased slightly for all voltages applied to the system with respect to time. A possible reason for this pH increase may be the reactions that take place at the cathode, which are responsible for the formation of hydroxyl ions and hydrogen bubbles [4–7,15].

3.2. Effect of gap distance between electrodes

The gap distance between electrodes has an important effect on the electrocoagulation process and affects the ohmic potential related to the electrocoagulation cell and energy depletion [4,5]. In this study, the effects of electrode gap distance were tested at 12 and 6 V for the iron and copper electrodes, respectively. Fig. 3 illustrates the urea removal efficiencies at various electrode spacing intervals (3.0 and 4.5 cm). The higher removal efficiencies of urea were obtained at 3.0 cm spacing. The system configuration may play the most important role in causing this phenomenon. With a gap distance of 3 cm, the distance between the electrodes is similar to the spacing value between the electrodes and the beaker walls, and the similarity of the distances is related to the reactor cross-section, as a circular reactor was used. Thus, a gap distance



Fig. 1. Setup of electrocoagulation unit.

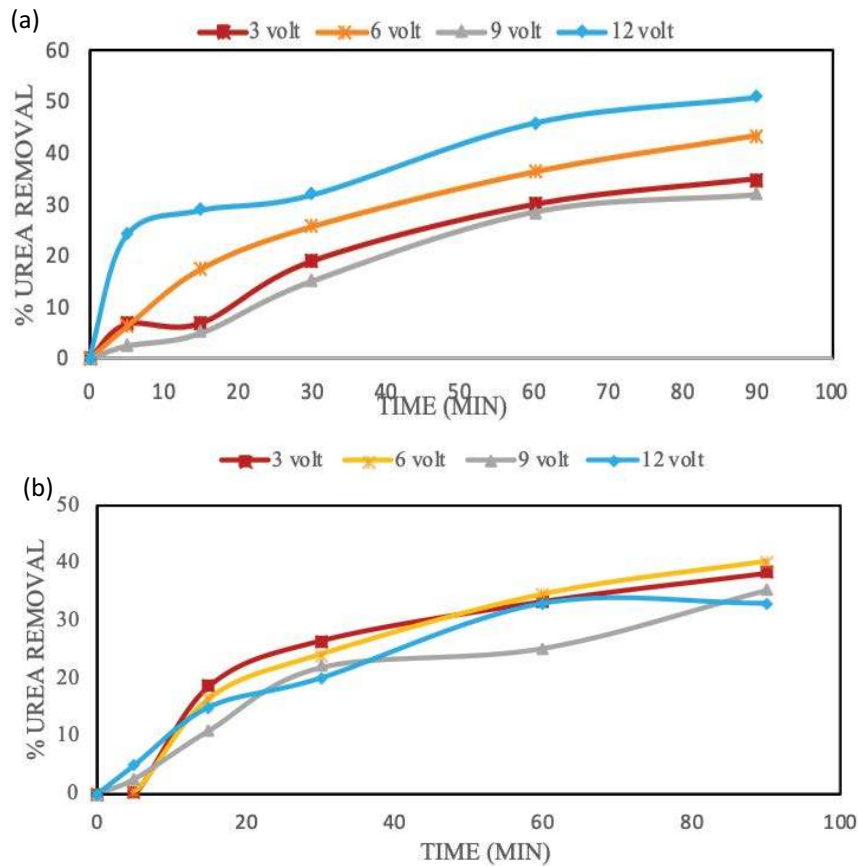


Fig. 2. Urea removal efficiencies vs. time at various voltage values: (a) Fe electrode and (b) Cu electrode.

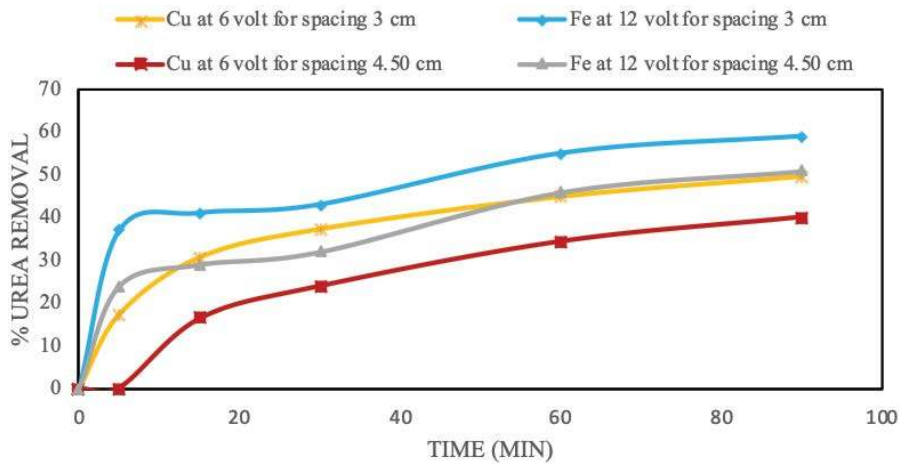


Fig. 3. Urea removal efficiencies at various electrode spacing for the optimum voltages.

of 3 cm leads to the following: (i) a uniform distribution of the flocs inside the beaker and (ii) a decrease in floc turbulence (because of the small separation between electrodes) that might occur through mixing, leading to an increase in the electrostatic effect that prevents particle collision. The behavior of the floc formation is reversed with a large spacing, as a strong reduction in floc formation was observed [7,15–17]. For all spacings, the level of pollutant removal

was similar. For the first 30 min of the experiment, pollutant removal occurred rapidly, and the removal rate decreased thereafter. The highest removal efficiencies detected for urea removal with a gap distance of 3 cm were 49.65% for the copper electrode and 59% for the iron electrode. The removal efficiencies when using the copper electrode were greater than those obtained with the iron electrode, possibly because the rate of floc generation was higher when using

the iron electrode. The removal efficiencies demonstrate that the electrocoagulation process is much better with the iron electrode than with the copper electrode.

3.3. Influence of changing the surface-area-to-volume ratio

One of the most important factors in the reactor design for the electrocoagulation process is the ratio of the immersed electrode surface area to the volume of the treated solution (S/V ratio) [18–20]. As described in the literature, there is a direct relation between the S/V ratio and the current density consumption. The current density decreases when the S/V ratio increases; therefore, the S/V parameter has a major effect on the performance of the electrocoagulation process. In this research, an increase in the S/V ratio was studied by increasing the surface area of the anode and determining the effect on the efficiency of urea removal. Surface areas of 30 and 60 cm² were tested. Fig. 4 shows the influence of changing the S/V ratio on the urea removal efficiency vs. time for the optimum voltage for the copper and iron electrodes. The highest removal efficiencies were obtained with an anode surface area of 60 cm²: 46.80% for the copper electrode and 57% for the iron electrode. The larger anodes may have greater removal efficiency because a larger surface area motivates electrical transport, which leads to an increase of the resistance of the electrochemical cell. Consequently, urea dissolves more readily, and the overall removal efficiency of urea increases.

3.4. Influence of electrolyte type on the electrocoagulation performance

The electrolyte has a major effect on the removal efficiency of urea. The availability and suitability of an electrolyte material, as well as its effect on human health, must be considered. Calcium chloride (CaCl₂) and sodium chloride (NaCl) meet the above-mentioned criteria, as they are both non-toxic for human health and regularly available. The major supporting role of the electrolyte during the electrocoagulation process is to increase the conductivity of the treated solution. The type of electrolyte also affects

the generation rate of several oxidants in the treated solution [21]. To study the effect of electrolyte type on the urea removal efficiency, two electrolytes were tested: NaCl and CaCl₂. The concentration used for both electrolytes was 0.40 g/L. Fig. 5 shows the removal efficiencies of urea over time with CaCl₂ and NaCl at the optimum voltages for each type of electrodes. A higher removal efficiency with CaCl₂ as an electrolyte was observed possibly because CaCl₂ has higher conductivity than NaCl, resulting in the improved chemical dissolution of urea, which encourages electrical transport in the electrochemical cell. Greater electrical transport increases the electrochemical cell resistance and the dissolution of urea, ultimately increasing the urea removal efficiency. Removal efficiencies were found to be 46.16% and 58.65% for copper and iron electrodes, respectively.

3.5. FTIR spectral analysis for the by-products from the electrocoagulation process

FTIR spectral analysis was carried out to investigate and clarify the by-products generated from the electrocoagulation process. For each electrode, two sludge samples were analyzed using FTIR spectral analysis. Thus, the analysis was performed on a total of four sludge samples. The first two sludge samples contained only the electrolyte (blank sample), and the second two samples contained both the electrolyte and urea. Figs. 6 and 7 show the FTIR spectral analysis of the by-products for the blank and urea-containing sludge samples (obtained at 6 and 12 V using copper and iron electrodes), respectively. There are differences between the FTIR spectra for the various sludge samples. The FTIR spectra of the two blank samples obtained with different anodes show differences between 400 and 4,000 cm⁻¹, proving the presence of dissimilar components. These dissimilar components occurred as a consequence of the dissolution of either copper or iron electrodes during the electrocoagulation process [22]. The spectra for the other two urea-containing sludge samples show an enormous-ranging and maximum band between 1,350 and 1,850 cm⁻¹ which occurred again between 2,850 and 3,850 cm⁻¹, representing the presence of an OH group. When the OH group exists,

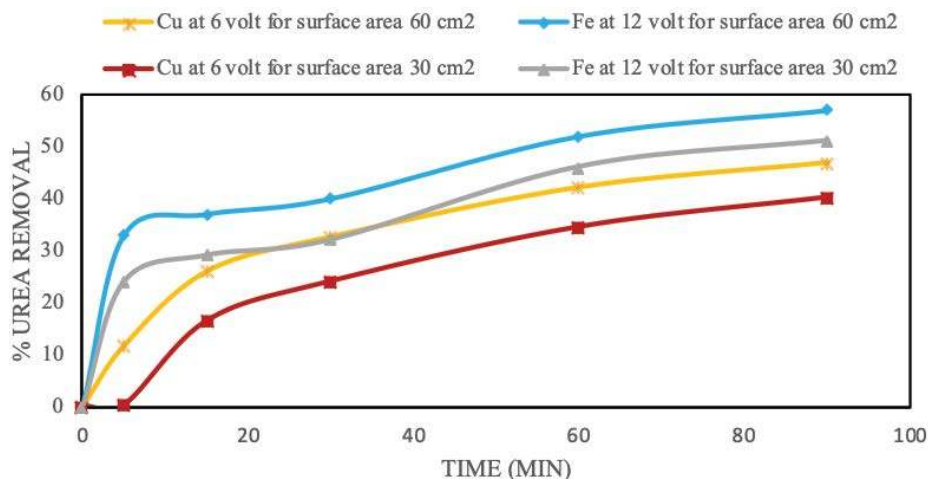


Fig. 4. Influence of changing the S/V ratio on the urea removal efficiency vs. time.

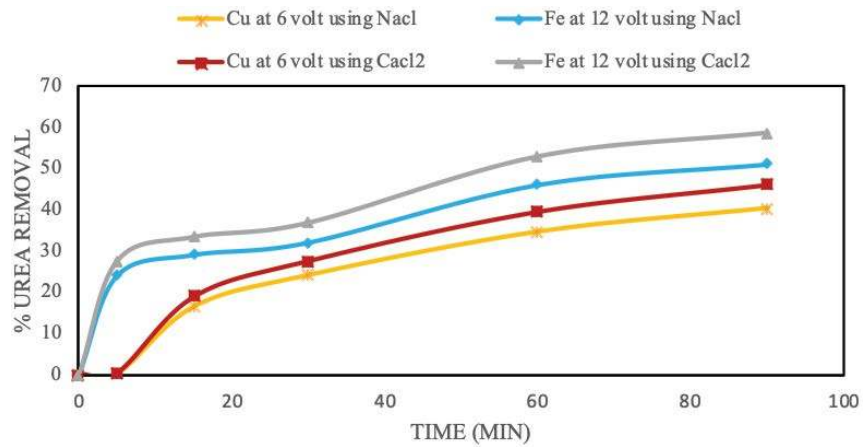


Fig. 5. Influence of changing the electrolyte type on the urea removal efficiency vs. time.

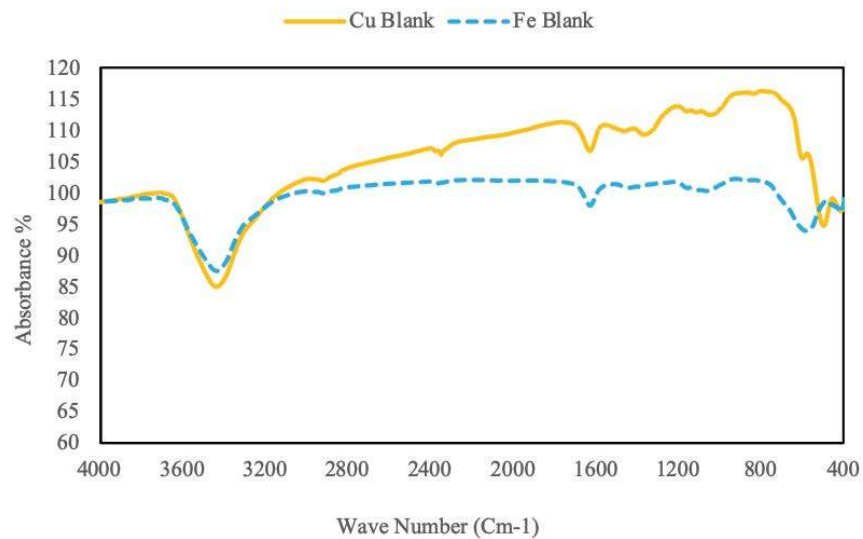


Fig. 6. FTIR spectral analysis of the sludge sample without urea.

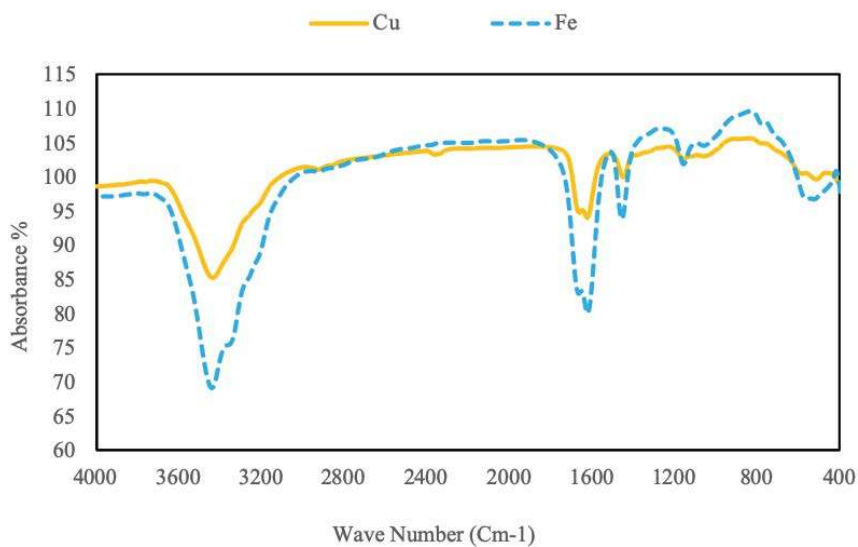


Fig. 7. FTIR spectral analysis of the sludge sample with urea.

the adsorption of the reverse ions improves throughout the settling process. Thus, adsorption is one of the removal methods throughout the electrocoagulation process.

3.6. Investigation of electrode morphologies

The morphologies of the copper and iron electrodes were investigated before and after the electrocoagulation process at a voltage of 6 V for copper and 12 V for iron. Fig. 8 presents the SEM images of the two types of electrodes before and after the electrocoagulation process. For each of the copper and iron anodes, corrosion occurred because of the electrocoagulation process, providing evidence for the treatment. The outer surfaces of both types of anode were investigated using SEM, and cracks and small voids were observed on the copper electrode surface.

Nevertheless, the outer surface of the iron anode was rough with large voids. The large number of cracks and voids on the outer surfaces of both electrodes result from the consumption of the electrode materials on the active side of the anodes, which is a consequence of the creation of oxygen at the anode surface [5,22]. The corrosion that occurred on the surface of the copper electrode was uniform, while non-uniform corrosion occurred on the iron electrode as a result of the surface pitting. Uniform corrosion is considered better than non-uniform corrosion here because it can be predicted more easily.

3.7. Performance of chemical coagulation

We examined the influence of various concentrations (10–160 g/L) of copper sulfate and ferric sulfate on urea

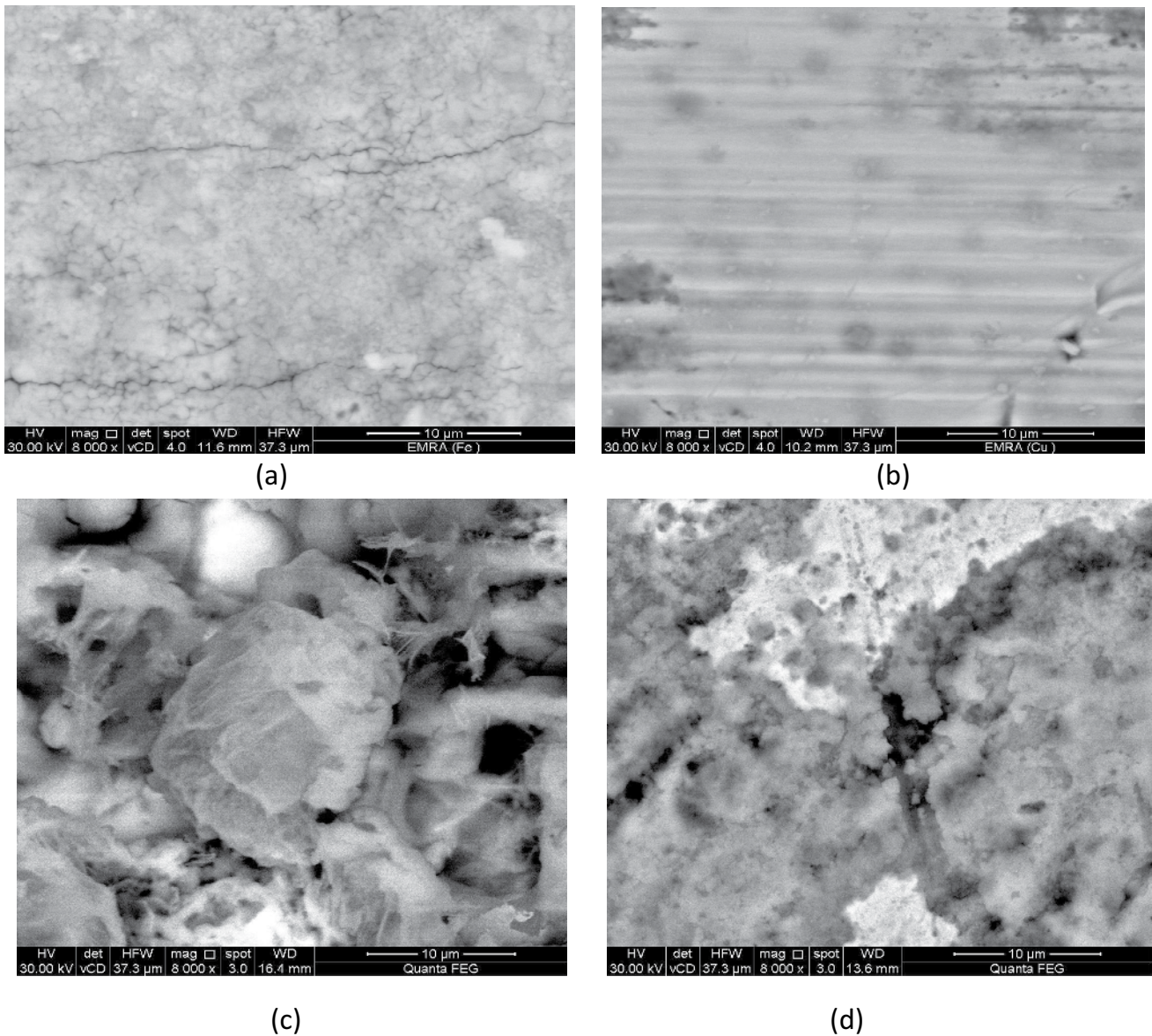


Fig. 8. SEM image: (a) Fe electrode, (b) Cu electrode prior to the EC process, (c) Fe electrode and (d) Cu electrode following the EC process.

removal performance by conventional coagulation methods. Results were compared with those obtained from the electrocoagulation process [5,23]. Fig. 9 shows the jar tests using copper sulfate and ferric sulfate. Fig. 10 shows the performance of conventional coagulation for urea removal at various chemical coagulant dosages. When copper sulfate is used, the urea removal efficiency increased with increasing dosage, indicating better performance than ferric sulfate for all dosages, except 160 g/L, for which the behavior was reversed. The highest urea removal efficiency achieved was 70% for ferric sulfate and 58% for copper sulfate when using an elevated coagulant dose of 160 g/L. The performance when using copper sulfate as the coagulant was better than that obtained via the electrocoagulation process, indicating that the electrocoagulation process is less efficient than chemical coagulation when using a copper anode for urea treatment. In contrast, results obtained with the iron anode in the electrocoagulation treatment are better than those obtained using chemical coagulation at all dosages, except 160 g/L, where the removal efficiency by chemical coagulation was much better.

3.8. Performance of EC with real wastewater

In this study, a comparison was conducted with synthetic wastewater using real wastewater for both copper (at 6 V) and iron (at 12 V) electrodes to study the effect of the electrocoagulation process with real wastewater. Both urea and chemical oxygen demand (COD) were measured for the real wastewater samples. Urea was measured to facilitate the comparison with synthetic wastewater, while COD was measured because real wastewater can contain other pollutants that can compete for the flocs formed and so affect the efficiency of urea removal. The highest COD removal efficiencies after 90 min were 25% and 32% for the copper and iron electrodes, respectively. The COD removal efficiency was much higher for the iron electrode than the copper electrode for most of the electrocoagulation process. Fig. 11 shows urea removal efficiencies vs. time for copper and iron electrodes at optimum voltages when using real wastewater. The highest rate of urea removal was achieved with the iron electrode, indicating that the performance of the iron electrode is better than that of the copper

electrode for the treatment of urea. The removal efficiencies obtained after 90 min were observed to be 32% in the case of iron and 25% in the case of copper. There are many reasons for the increase in COD removal efficiency. First, the metal hydroxide generation rate increases as a result of the increased metal ion dissolution rate [8,24]. Second, the pollutant adsorption phenomenon occurs on the surfaces of metal hydroxides, oxides, and oxyhydroxides [25–28]. Third, the quantity of pollutants is reduced when the production of hydrogen bubbles in the solution increases, as the bubbles help to remove the pollutants through the flotation process. During the electrocoagulation process, pH values increased slightly for all applied voltages with respect to time, possibly because of reactions at the cathode that transform water into hydroxyl ions and hydrogen bubbles [29–32]. After 90 min, the pH values for both copper and iron anodes are similar, suggesting that the water molecule separation occurs equally with both electrodes. The dissolution of copper produces divalent Cu^{2+} ions that form the copper hydroxide, the thermodynamically favored compound at a pH higher than 7. The dissolution of iron produces Fe^{2+} at pH ranges between 7 and 9. The formation of these hydroxides is preferred at higher pH values, and the hydroxides trap colloids/contaminants, removing them from the treated solution through the settling process, thereby increasing the rate of COD removal. The urea removal efficiency in real wastewater may be lower than in synthetic wastewater because the presence of additional pollutants can affect the electrocoagulation process.

3.9. Economic evaluation of electrocoagulation process

To optimize the electrocoagulation process, both the electrical energy cost (EEC) and the consumption of the electrode material (EMC) should be calculated. EEC can be calculated using the following equation (Eq. (6)):

$$\text{EEC} \left(\text{kWh/m}^3 \right) = \frac{I \times U \times t}{V} \quad (6)$$

where the current intensity, the cell voltage, the electrolysis time, and the active volume of the reactor are represented by I (A), U (V), t (h), and V (m^3), respectively. The EEC for

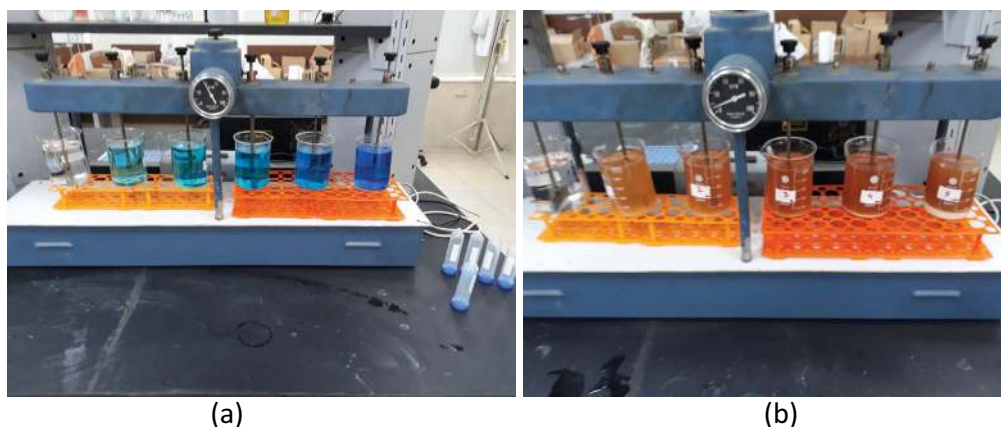


Fig. 9. Jar test using (a) cupric sulfate (pentahydrate) and (b) ferric sulfate hydrate.

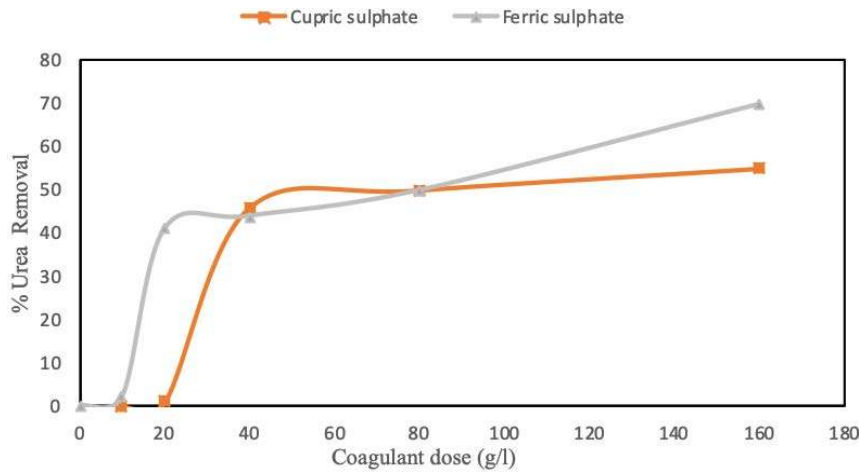


Fig. 10. Urea removal efficiencies with various coagulant doses.

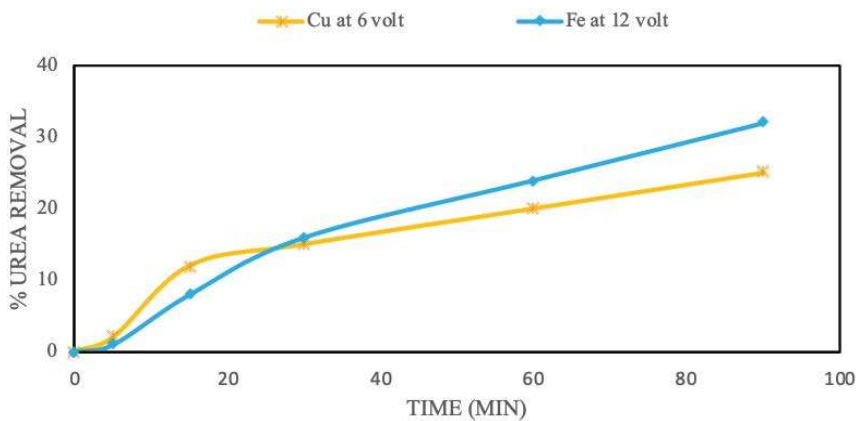


Fig. 11. Urea removal efficiencies vs. time for Cu and Fe electrodes at optimum voltages when using real wastewater.

iron and copper electrodes were 112.8 and 32.4 kWh/m³, respectively. The electrical energy consumption for the copper electrode was lower than that for the iron electrode. EMC can be obtained using the following equation (Eq. (7)):

$$EMC \left(\text{g/m}^3 \right) = \frac{M \times I \times t}{(z \times F \times V)} \quad (7)$$

where the molar mass of the electrode, the number of electrons, and the Faraday constant are represented by *M* (g/mol), *z*, and *F* (96,500 c/mol), respectively. The values of electrode consumption at maximum removal were 9.8 and 6.4 g/m³ for iron and copper electrodes, respectively. Thus, a greater mass of iron electrodes was consumed than of copper electrodes.

4. Conclusions

This study was carried out to evaluate the removal efficiency of urea using the electrocoagulation process. This evaluation used two types of anodes: copper and iron. The obtained results indicated that the iron electrode provides much higher urea removal efficiencies than

the copper electrode. The highest urea removal efficiency obtained was around 51% at 12 V when using an iron electrode, and 40.15% at 6 V when using a copper electrode. Higher removal efficiencies were obtained with a gap distance of 3 cm than with a distance of 4.50 cm. When CaCl₂ was used as an electrolyte, removal efficiencies were greater than those obtained when using NaCl as an electrolyte. The FTIR spectra of the two samples show that the fingerprints for the two sludge samples drawn from the electrocoagulation cell for the two anodes were not identical, providing proof of the presence of dissimilar components. The morphologies of the copper and iron electrodes were investigated before and after the electrocoagulation process at a voltage of 6 V for copper and 12 V for iron using the SEM. Corrosion occurred to the anodes because of the electrocoagulation process, providing evidence of the treatment. The urea removal efficiency using the electrocoagulation process with the iron electrode is much higher than using conventional chemical coagulation, while the reverse is true when using copper electrodes. The urea removal efficiency in real wastewater was lower than that obtained in synthetic wastewater because of the presence of additional pollutants that can affect the electrocoagulation process.

Acknowledgments

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

References

- [1] E. Urbańczyk, M. Sowa, W. Simka, Urea removal from aqueous solutions—a review, *J. Appl. Electrochem.*, 46 (2016) 1011–1029.
- [2] S.M. Safwat, M.E. Matta, Performance evaluation of electrocoagulation process using zinc electrodes for removal of urea, *Sep. Sci. Technol.*, 55 (2019) 2500–2509.
- [3] S.M. Safwat, M.E. Matta, Adsorption of urea onto granular activated alumina: a comparative study with granular activated carbon, *J. Dispersion Sci. Technol.*, 39 (2018) 1699–1709.
- [4] M.Y.A. Mollah, P. Morkovsky, J.A.G. Gomes, M. Kesmez, J. Parga, D.L. Cocke, Fundamentals, present and future perspectives of electrocoagulation, *J. Hazard. Mater.*, 114 (2004) 199–210.
- [5] S.M. Safwat, A. Hamed, E. Rozaik, Electrocoagulation/electroflotation of real printing wastewater using copper electrodes: a comparative study with aluminum electrodes, *Sep. Sci. Technol.*, 54 (2019) 183–194.
- [6] M.M. Emamjomeh, M. Sivakumar, Review of pollutants removed by electrocoagulation and electrocoagulation/flotation processes, *J. Environ. Manage.*, 90 (2009) 1663–1679.
- [7] J.N. Hakizimana, B. Gourich, M. Chafi, Y. Stiriba, C. Vial, P. Drogui, J. Naja, Electrocoagulation process in water treatment: a review of electrocoagulation modeling approaches, *Desalination*, 404 (2017) 1–21.
- [8] A.S. Fajardo, R.F. Rodrigues, R.C. Martins, L.M. Castro, R.M. Quinta-Ferreira, Phenolic wastewaters treatment by electrocoagulation process using Zn anode, *Chem. Eng. J.*, 275 (2015) 331–341.
- [9] F. Akbal, S. Camcıotless, Copper, chromium and nickel removal from metal plating wastewater by electrocoagulation, *Desalination*, 269 (2011) 214–222.
- [10] S.M. Safwat, Treatment of real printing wastewater using electrocoagulation process with titanium and zinc electrodes, *J. Water Process Eng.*, 34 (2020) 101137, 1–9, doi: 10.1016/j.jwpe.2020.101137.
- [11] P.I. Omwene, M. Kobya, Treatment of domestic wastewater phosphate by electrocoagulation using Fe and Al electrodes: a comparative study, *Process Saf. Environ. Prot.*, 116 (2018) 34–51.
- [12] X. Chen, G. Chen, P.L. Yue, Department, Investigation on the electrolysis voltage of electrocoagulation, *Chem. Eng. Sci.*, 57 (2002) 2449–2455.
- [13] T.R. Devlin, M.S. Kowalski, E. Pagaduan, X. Zhang, V. Wei, J.A. Oleszkiewicz, Electrocoagulation of wastewater using aluminum, iron, and magnesium electrodes, *J. Hazard. Mater.*, 368 (2018) 862–868.
- [14] S.M. Safwat, M. Mamdouh, E. Rozaik, H. Abd-Elhalim, Performance evaluation of electrocoagulation process using aluminum and titanium electrodes for removal of urea, *Desal. Water Treat.*, 191 (2020) 239–249.
- [15] D.T. Moussa, M.H. El-Naas, M. Nasser, M.J. Al-Marri, A comprehensive review of electrocoagulation for water treatment: potentials and challenges, *J. Environ. Manage.*, 186 (2017) 24–41.
- [16] W. Simka, J. Piotrowski, G. Nawrat, Influence of anode material on electrochemical decomposition of urea, *Electrochim. Acta*, 52 (2007) 5696–5703.
- [17] Z. Al-Qodah, M. Al-Shannag, Heavy metal ions removal from wastewater using electrocoagulation processes: a comprehensive review, *Sep. Sci. Technol.*, 52 (2017) 2649–2676.
- [18] S. Ahmed, E. Rozaik, H. Abdelhalim, Effect of configurations, bacterial adhesion, and anode surface area on performance of microbial fuel cells used for treatment of synthetic wastewater, *Water Air Soil Pollut.*, 226 (2015) 300, 1–13, doi: 10.1007/s11270-015-2567-3.
- [19] B. Khaled, B. Wided, H. Béchir, E. Elimame, L. Mouna, T. Zied, Investigation of electrocoagulation reactor design parameters effect on the removal of cadmium from synthetic and phosphate industrial wastewater, *Arabian J. Chem.*, 12 (2019) 1848–1859.
- [20] S. Garcia-Segura, M.M.S.G. Eiband, J.V. de Melo, C.A. Martínez-Huitle, Electrocoagulation and advanced electrocoagulation processes: a general review about the fundamentals, emerging applications and its association with other technologies, *J. Electroanal. Chem.*, 801 (2017) 267–299.
- [21] A.C. Ndjomgoue-Yossa, C.P. Nansu-Njiki, I.M. Kengne, E. Ngameni, Effect of electrode material and supporting electrolyte on the treatment of water containing *Escherichia coli* by electrocoagulation, *Int. J. Environ. Sci. Technol.*, 12 (2015) 2103–2110.
- [22] T.Y. Nayir, S. Kara, Container washing wastewater treatment by combined electrocoagulation – electrooxidation, *Sep. Sci. Technol.*, 53 (2017) 1592–1603.
- [23] S.Y. Lee, G.A. Gagnon, Comparing the growth and structure of flocs from electrocoagulation and chemical coagulation, *J. Water Process Eng.*, 10 (2016) 20–29.
- [24] S. Ahmadzadeh, A. Asadipour, M. Pournamdari, B. Behnam, H.R. Rahimi, M. Dolatabadi, Removal of ciprofloxacin from hospital wastewater using electrocoagulation technique by aluminum electrode: optimization and modelling through response surface methodology, *Process Saf. Environ. Prot.*, 109 (2017) 538–547.
- [25] G. Busca, S. Berardinelli, C. Resini, L. Arrighi, Technologies for the removal of phenol from fluid streams: a short review of recent developments, *J. Hazard. Mater.*, 160 (2008) 265–288.
- [26] Y.X. Zhao, B.Y. Gao, B.C. Cao, Z.L. Yang, Q.Y. Yue, H.K. Shon, J.H. Kim, Comparison of coagulation behavior and floc characteristics of titanium tetrachloride (TiCl₄) and polyaluminum chloride (PACl) with surface water treatment, *Chem. Eng. J.*, 166 (2011) 544–550.
- [27] M. Bennajah, M. Maalmi, Y. Darmane, M.E. Touhami, Defluoridation of drinking water by electrocoagulation/electroflotation: kinetic study, *J. Urban Environ. Eng.*, 4 (2010) 37–45.
- [28] M. Yoosefian, S. Ahmadzadeh, M. Aghasi, M. Dolatabadi, Optimization of electrocoagulation process for efficient removal of ciprofloxacin antibiotic using iron electrode; kinetic and isotherm studies of adsorption, *J. Mol. Liq.*, 225 (2017) 544–553.
- [29] I. Ali, M. Asim, T.A. Khan, Arsenite removal from water by electro-coagulation on zinc-zinc and copper-copper electrodes, *Int. J. Environ. Sci. Technol.*, 10 (2013) 377–384.
- [30] C. An, G. Huang, Y. Yao, S. Zhao, Emerging usage of electrocoagulation technology for oil removal from wastewater: a review, *Sci. Total Environ.*, 579 (2017) 537–556.
- [31] R. Kamaraj, S. Vasudevan, Evaluation of electrocoagulation process for the removal of strontium and cesium from aqueous solution, *Chem. Eng. Res. Des.*, 93 (2015) 522–530.
- [32] X. Zhang, H. Lin, B. Hu, The effects of electrocoagulation on phosphorus removal and particle settling capability in swine manure, *Sep. Purif. Technol.*, 200 (2018) 112–119.