Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2019.23530

144 (2019) 138–144
March

Optimization of the electrocoagulation process on the effectiveness of removal of Cefixime antibiotic from aqueous solutions

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Received 3 November 2017; Accepted 26 November 2018

ABSTRACT

Nowadays, the remaining of antibiotics in aquatic environments is increasing. Although the amounts of these antibiotics are extremely small (from 1 ng to 1 μg), such tiny amounts can have poisonous effects on humans and aquatic organisms. Accordingly, this study aimed to analyze the effectiveness of the electrocoagulation process on the removal of CFX antibiotic from aqueous solutions. The aim of this study was to the removal of Cefixime antibiotic by electrocoagulation process from aqueous solutions. In a batch mode, several parameters such as pH (3–10), initial concentration (3–100 mg/L), electric current intensity (0.1–1.1 A), and retention time (20–120 min) were investigated on the removal efficiency of Cefixime. The most effective removal (90.1%) was obtained at pH of 6, initial concentration of 5 mg/L, electric current intensity of 0.7 A, and retention time of 60 min. According to the findings of this study, the electrocoagulation process can be considerable as an effective approach to remove Cefixime antibiotic from aqueous solutions.

Keywords: Antibiotic; Cefixime; Electrocoagulation; Aqueous solutions

1. Introduction

Today, the rapid increase of residues of pharmaceutical in surface water and groundwater around industrial and residential communities has caused a major concern. Early concerns about their potential risk were expressed in 1999 [1]. Later then, many environmental researchers became interested in analyzing these hazardous chemical substances [2,3]. Studies show that in human medicine annual drug used about 100,000–200,000 tons of antibiotic to cure and control infection in every around the globe [4–6]. Cefixime is one of the third generation antibiotics of cephalosporin. This drug is used for the treatment of diseases infections including gonorrhea, otitis media, pharyngitis, pneumonia, bronchitis, syphilis, urinary tract infections, etc. [7–9]. According to the studies, this drug has been one of the most intensively used pharmaceuticals in Europe [10]. Antibiotics have discharged by human consumption, agricultural manure, landfill site leachate, and slaughterhouses and sewage sludge as organic fertilizer into aquatic environment [11–13]. Antibiotics are described as environmental contaminants in low concentrations (ng/L to μg/L) but the frequent occurrence of antibiotics occurrence in the aquatic environment has raised concern about their potential impact on the environment [1,14–18]. The release of antibiotics in the environment can effect on human beings and ecosystem. Some of the

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effects caused by antibiotics include aquatic toxicity, the development of resistance in pathogenic bacteria, teratogenic effects in pregnant women and genotoxicity [19–22]. Some of the studies reported which about 80% of a total load of pharmaceuticals entering sewage treatment plants may be discharged into the surface water without any change [23,24]. Given the adverse, irreparable effects of drugs, especially antibiotics, on the environment and the ineffectiveness of conventional wastewater treatment processes in reducing these drugs, it is essential to develop an economical technology to reduce these compounds from aquatic environments [32]. Various methods have been used to remove antibiotics from aqueous solutions such as zonation [25], wetland [26], Fenton process [27], ion exchange [28], membrane processes [29], and adsorption processes like electrocoagulation. Among these methods, adsorption processes are simple and economical; this method can also remove low concentrations of antibiotics from aqueous solutions [30,31]. Adsorption via electrocoagulation is a simple yet effective method for removing Color [32], Fluoride [33], Hexavalent chromium (Cr(VI)) [34], Arsenic [35], suspended solids [36], pesticides, persistent organic compounds [37], and some antibiotics such as tetracycline [38], ciprofloxacin [39], and azithromycin [40]. This method has many advantages as follows: it needs simple equipment and short reaction time, it produces a small amount of sludge, and it does not need any chemical substances [41]. The electrochemical process includes many physical and chemical processes similar to the addition of coagulants such as alum and ferric chloride in water [42]. This process involves applying an electric current to sacrificial electrodes where the current generates a coagulating agent and gas bubble [43]. Factors such as pH, initial concentration, applied current and reaction time heavily affect the effectiveness of this process [34,44]. Iron or aluminum is generally employed as a sacrificial electrode material in EC process [45]. These electrodes are able to produce metal hydroxyls that can absorb pollutants and finally separate them as sediment or floating substances from aqueous solutions [38,46]. The anodic and cathodic reactions for aluminum electrodes can be written as:

Anode reaction:

$$Al_{(s)} \rightarrow Al^{3+}_{(aq)} + 3e^- \quad (1)$$

Cathode reaction:

$$3H_2O + 3e^- \rightarrow \frac{3}{2}H_{2(g)} + 3OH^- \quad (2)$$

Al^{3+} and OH^- ions generated by electrode two above-mentioned reactions react to form various species such as $Al(OH)^+_2$, $Al_2(OH)^{4+}_2$, $Al(OH)^-_4$, $Al_6(OH)^{3+}_{15}$, $Al_7(OH)^{4+}_{17}$, $Al_8(OH)^{4+}_{20}$, $Al_{13}O_4(OH)^{7+}_{24}$, $Al_{13}(OH)^{5+}_{34}$ which transform finally into $Al(OH)^3_{(s)}$ according to the complex precipitation kinetics [38]:

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

To our knowledge, no report is available in the literature dealing with the removing of Cefixime concentration in water using the EC process. Therefore, given the importance of studying pharmaceutical compounds and the advantages of this process, the current study has focused on the optimal conditions for removing Cefixime antibiotic via the electrocoagulation process.

2. Experimental

2.1. Chemical substances and equipment

Cefixime (99%) were purchased from Hangzhou Dayangchem Co. (Hangzhou, China) and used as received. The molecular structure of Cefixime ($C_{16}H_{15}N_5O_7S_2$) is shown in Fig. 1 [47]. Other chemical substances such as HCl, NaOH and KCl were obtained from the Merck Company and used. All the solutions were prepared by using De-ionized water.

A stock solution (1,000 mg/L) was prepared by dissolving an appropriate quantity of Cefixime in sodium hydroxide 0.1 N. Working standard solutions of the required concentration were prepared daily by diluting an appropriate volume of the stock solution with De-ionized water.

The pH of the solutions was adjusted by adding 1 N of HCl or NaOH solutions. The concentration of the electrolyte in EC cell was adjusted by adding 2M KCl, and kept on 1.2 mS/cm, in the during an experiment. Samples were measured for pH by pH meter model (Mettler Toledo), electric conductivity with a conductivity meter (WTW LF09 Model) and Cefixime concentrations with UV/Vis Spectrophotometer (Cecil 7250 Model) at the wavelength of 490 nm.

2.2. Construction of the reactor

Experiments were carried out in a batch electrochemical reactor from Plexiglas with dimensions of 10 cm× 15 cm× 10 cm and 1 L capacity Fig. 2. The electrocoagulation was setup with aluminum/aluminum electrodes (48 cm^2 effective area)

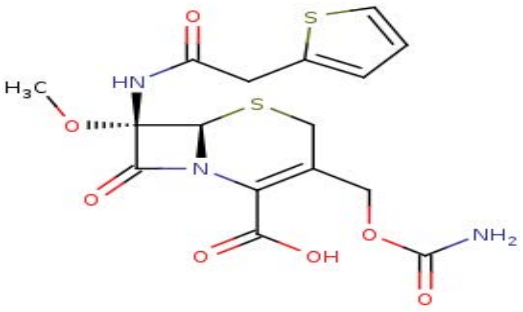

Fig. 1. The molecular structure of Cefixime antibiotic.

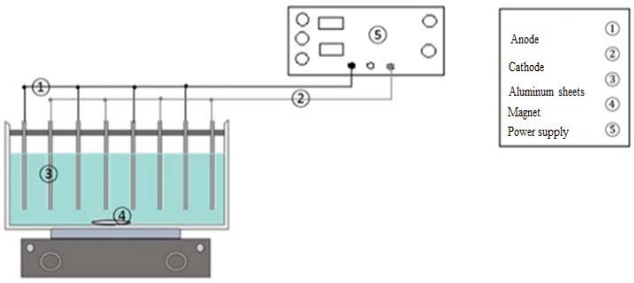

Fig. 2. The schematic representation of the electrocoagulation process.

immersed vertically, monopole configuration and in parallel. The distance between electrodes (anode and cathode) was a value of 1.5 cm during experiments. A direct current was supplied by a DC power source.

2.3. The operation of the reactor

Process variables were chosen on the basis of trial experiments and available literature. pH (3–10), initial Cefixime concentration (3–100 mg/L), current density (0.1–1.1 A), and reaction time (20–120 min) were analyzed. Successful mixing was achieved using a magnetic plate with a stirrer. The stirrer speed was controlled at 400 rpm. All the runs were performed at room temperature (20°C ± 1°C).

Experiments were calculated by three repetitions and the error was found to be below 5%. Removal efficiency of Cefixime was calculated by following formula:

$$R = \frac{C_t - C_0}{C_0} \times 100 \quad (4)$$

C_0 = initial CFX concentration (mg/L); C_t = CFX concentration in time t (mg/L)

3. Result and discussions

3.1. Characterization

To magnify and observe the morphology of the surface of Cefixime powder and sludge of the process in an optimal condition, electronic microscopic images were taken. These images are shown in Fig. 3. Image B shows the sludge of the process after the reaction: a huge part of Cefixime reacts with aluminum hydroxyls and turns into sludge.

3.2. Effects of pH

Experiments were conducted based on pH 3–10 to analyze the effects of pH. The relevant results of the effects of pH changes on Cefixime removal are presented in Fig. 4. According to the findings, the effectiveness of the removal process increased as the pH level increased from 3 to 6, and then the effectiveness decreased as the pH level went higher. The effectiveness amount of Cefixime removal reached 8.1% in pH 10. The most efficient amount was 53.7% at pH 6.

One of the most effective parameters in chemical processes, especially the process of coagulation and electrocoagulation, is the pH level [38]. The pH 6 to 8 has been usually considered as the practical pH level in the process of coagulation and electrocoagulation [48]. In the current study, the effectiveness of Cefixime removal increased as the pH increased from 3 to 6, yet the effectiveness decreased when the pH level went beyond that level (from 6 to 10). Experimental results show that in acidic pH, the main products of hydrolyzed aluminum are AL^{+3}, ALO^{+}, and $ALOH^{2+}$ compounds that are less able to adsorb pollutants. On the contrary, at lower pH levels (5–8), these compounds are weaker, so the adsorption level increases drastically [49]. In a study by Yoosefian et al. the highest effectiveness level of ciprofloxacin removal via the electrocoagulation process was observed at pH 7.5. In their study, increasing pH level from 4 to 7 increased the effectiveness level from 40%–100%. Nevertheless, moving to higher pH levels decreased the effectiveness down to 82% [39]. In another study, Amrane et al. reported the pH level of 3–10 as the best range for tetracycline removal; within

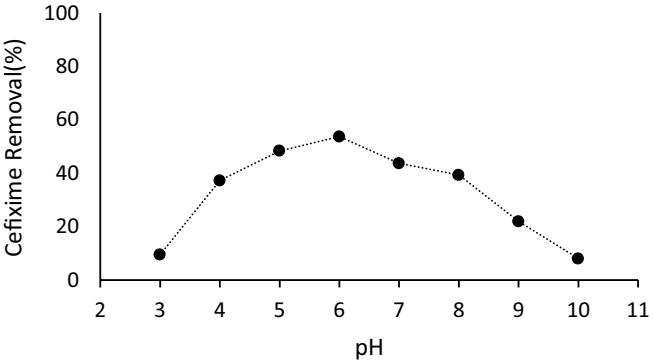

Fig. 4. Effects of pH changes on the effectiveness of Cefixime removal (Cefixime initial concentration: 50 mg/L, electrical conductivity: 1.2 ms/cm, electric current intensity: 0.5 A, retention time: 20 min, reaction temperature: 20°C ± 1°C).

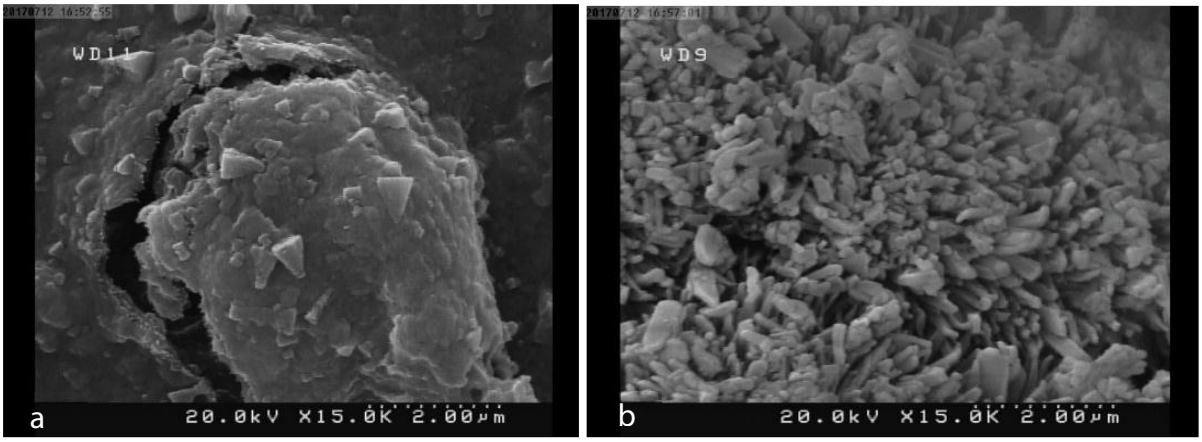

Fig. 3. SEM images (a: Cefixime powder, b: sludge of the process in an optimal condition).

this range, the process could remove 99% of this compound while in pH 2 this effectiveness decreased to 10% [38]. Finally, Parsa et al. found that the highest level of effectiveness in removing ciprofloxacin by the electrocoagulation process through a direct current was achieved at pH 5–6 [50].

3.3. Effects of initial concentration

Fig. 5 shows the results of the effects of changes in the initial concentration of Cefixime on the effectiveness of Cefixime removal. Based on these findings, the most effective amount (75.66%) was observed in 5 mg/L concentration level.

The other factor that increased the effectiveness of Cefixime removal was increasing the initial concentration level of Cefixime up to 5 mg/L. Higher concentration levels, however, lowered the effectiveness. Increasing the initial concentration of Cefixime causes an increase in the needed concentration level of aluminum hydroxyls in the environment. Given the similarity of conditions in higher initial concentrations, the ratio of aluminum hydroxyl to the pollutants decreases and weakens the effectiveness of its adsorption. Likewise, the findings of this study showed that if the initial concentration of Cefixime increased from 5 to 100 mg/L, the effectiveness of the removal process decreased down to 47.19%. As reported by Yoosefian et al., increasing the initial concentration of ciprofloxacin from 57.67 to 112.33 mg/L lowered the effectiveness of electrocoagulation down to 13.64% [34]. Hence, the findings of this study totally agree with Yoosefian et al.'s results.

3.4. Effects of changes in electric current intensity

Fig. 6 represent the amount of Cefixime removal in the electric current intensity levels of 0.1–1.1 A. According to this diagram, the effectiveness of removal increased as the electric current intensity increased up to 0.7 A and reached 77.23%. The removal amount decreased when the electric current intensity went higher than 0.7 A so that in 1.1 A, the effectiveness of removal reached 53.09%.

The electrical current intensity is another important factor in the electrocoagulation process. As the electric current increases, more OH^- and AL^{3+} ions are produced; therefore, there are more metal hydroxyls and flocs that are able to adsorb pollutants [34]. This process increases the efficiency of the removal process. Moreover, the higher current intensity increases the H_2 bubbles that can remove some pollutant flocs [29].

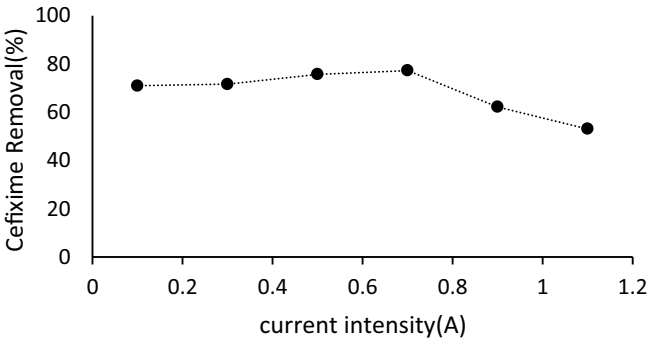

Fig. 6. Effects of changes in electric current intensity on the effectiveness of Cefixime removal (Cefixime initial concentration level: 5 mg/L, pH 6, electrical conductivity: 1.2 sm/cm, retention time: 20 min, reaction temperature: 20°C ± 1°C).

In a study by Amrane et al., the retention time needed for removing 99% of tetracycline via the electrocoagulation process decreased from 15 to 5 min by increasing the current intensity from 0.1 to 0.8 A [28]. In the same vein, Yazdanbakhsh et al. also found that increasing the current intensity up to 20 mA/cm could improve the effectiveness of azithromycin removal via the process of proxy electrocoagulation. On the contrary, current intensities beyond 20 mA/cm decreased the effectiveness level due to the destruction of OH^- radical [43].

The findings of this study showed that an increase in the electrical current intensity from 0.1 to 0.7 A could improve the effectiveness of the process of Cefixime removal from 70.96% to 78.5%. Higher current intensities (up to 1.1 A), however, decreased the effectiveness down to 53.09%.

3.5. Effects of changes in retention time

Fig. 7 shows changes in the effectiveness of Cefixime removal in different retention times. According to these results,

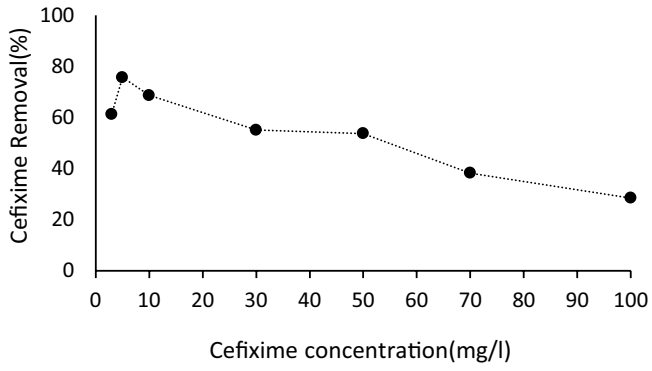

Fig. 5. Effects of changes in the initial concentration of Cefixime on the effectiveness of Cefixime removal (pH amount: 6, electrical conductivity: 1.2 sm/cm, electric current intensity: 0.5 A, retention time, 20 min, reaction time: 20°C ± 1°C).

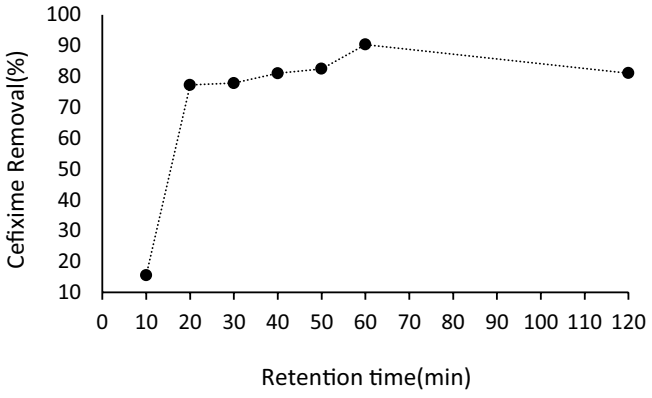

Fig. 7. Effects of retention time on the effectiveness of Cefixime removal (Cefixime initial concentration level: 5 mg/L, pH 6, electrical conductivity: 1.2 sm/cm, electric current intensity: 0.7 A, retention time: 20 min, reaction temperature: 20°C ± 1°C).

Table 1
Comparison of various methods for removal of Cefixime

Process	Removal (%)	Time (min)	References
UV/H_2O_2	100	180	The removal of the cefixime antibiotic from aqueous solution using an advanced oxidation process (UV/H_2O_2) [51]
WO_3/UV photocatalyst	83.3	100	Application of response surface methodology on Cefixime removal from aqueous solution by ultrasonic/photooxidation [52]
Iraqi bentonite	88.68	15	Adsorption of Cefixime on to Iraqi bentonite [53]
UV-LED/S2O82⁻	91.97	30	Application of ultraviolet light-emitting diodes to the removal of cefixime trihydrate from aqueous solution in the presence of peroxydisulfa [54]
Modified bionanocomposite	90.6	30	Efficient and selective removal of cefixime form aqueous solution by a modified bionanocomposite [55]
Electrocoagulation process	90.1	60	This study

increasing the retention time from 20 to 60 min increased the effectiveness level from 77.23% to 90.1%. However, after 60 min, the effectiveness of removal decreased in a way that after 120 min the effectiveness of removal was 81.08%.

Based on the findings of this study, increasing the retention time from 20 to 60 min increased the effectiveness of Cefixime removal from 77.23% to 90.31%, but increasing the retention time beyond 60 min up to 120 min decreased the effectiveness level down to 9.23%. Such a decrease in effectiveness level could be attributed to the disposal phenomenon. In this regard, Yoosefian et al. found that the effectiveness of ciprofloxacin removal via the electrocoagulation process significantly increased by 10.9% as the retention time increased from 12.24 to 22.76 min and decreased by 5.73% as the retention time increased to 112.32 min. Yoonesian regards 20–23 min as the most appropriate time for removing ciprofloxacin [34]. Yazdanbakhsh et al. also found that increasing the retention time from 10 to 60 min could improve the effectiveness of azithromycin removal through the process of proxy electrocoagulation. Yet, moving beyond 60 min (70–80 min) did not change the effectiveness level.

3.6. Comparison with other studies

A comparison between the results of this study with other published researches on the removal of Cefixime is presented in Table 1. As can be seen, the removal efficiency of electrocoagulation process in equilibrium time 60 min is near or higher than other procedures (such as WO_3/UV photocatalyst). So, this method can be used an effective process for the removal of Cefixime from polluted water.

4. Conclusion

The findings of this study showed that the electrocoagulation process can remove 90.1% of Cefixime antibiotic in the following optimal condition: pH 6, the initial concentration of 5 mg/L, the electric current intensity of 0.7 A, and the retention time of 60 min. Thus, given the inappropriateness of the current removal processes in wastewater treatment plants, the advantages of the electrocoagulation process make this process an appropriate method for Cefixime removal. Nonetheless, since there are various wastewater types and antibiotics, it is suggested that the effectiveness of this process should be reanalyzed for other antibiotics and aqueous solutions to reach more generalized conclusions.

Acknowledgment

This study is a result of a research proposal by the Vice Chancellor for Research and Technology, Qom University of Medical Sciences, with the ethics code: IR.MUQ.REC.139.51. We hereby express our gratitude to the Vice Chancellor for Research and Technology for their financial support and cooperation.

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