Plant Archives Vol. 20 Supplement 1, 2020 pp. 831-839 **e-ISSN:2581-6063 (online), ISSN:0972-5210**

STUDY OF THE ENVIRONMENTAL PARAMETERS ON CEPHALEXIN DEGRADATION BY ANODIC OXIDATION WITH BIOLOGICAL INDICATION

Rasha A. Waheeb and Yaaroub Falh Alfatlawy

Department of Biology, College of Science, University of Baghdad, Baghdad, Iraq.

Abstract

This study investigates the Cephalexin antibiotic degradation by the anodic oxidation using an undivided reactor equipped with a graphite plate as an anode electrode and stainless steel as the cathode. Different environmental parameters were examined, such as the solution pH, antibiotic initial concentration, applied current density, solution temperature, and the supporting electrolyte concentration (NaCl). The results demonstrated that the anodic oxidation is more efficient by increasing the initial concentration of supporting electrolyte, applied current density, and solution temperature at the acidic media. At NaCl concentration 1000 mg/l, the higher Cephalexin and COD removal efficiencies 82.75% and 25.98% respectively were obtained for the simulated wastewater and the real wastewater 52.073% and 8.15% respectively. The Cephalexin removal efficiency for simulated wastewater (at Cephalexin initial concentration 100 ppm, current density 0.5 mA/cm^2, pH 6, and NaCl 0 mg/l) were examined by the growth of the Cephalexin sensitive bacteria and it was found that the inhibition zone decreased due to the increase in the Cephalexin removal efficiency. The anodic oxidation was effective for removing the accompanying bacteria to the real Cephalexin contaminated water, where the total dead bacteria was achieved around 10^8CFU/ml by applying current density 0.5 mA/cm^2.

Key words: Environmental Parameter, Cephalexin Degradation, Anodic Oxidation

Introduction

The world faces an environmental crisis of water pollution that comes from the effluents of industries, hospitals, municipal sewage systems, agricultural activities, and others. Water resources must be protected to conserve both human health and current aquatic life by treating the discharged wastewater (Ganiyu *et al.,* 2015), (Ganzenko *et al.,* 2014).

Pharmaceuticals contamination in the water has received the researchers' attention and has been widely documented where it has been found in rivers, lakes, and small creeks (Oturan *et al.,* 2013), (Babu *et al.,* 2009). Among pharmaceuticals types, antibiotics are widespread in human and veterinary use to treat diseases (Oturan *et al.,* 2013). The antibiotics contaminants present in the aquatic environment increases the risks of antibacterial resistance spreading among microorganisms (González *et al.,* 2011).

In the last two decades, Researchers studied many treatment techniques for the removal of antibiotics from wastewater (Yahiaoui *et al.,* 2013), (Sirés and Brillas 2012), (Farhadi *et al.,* 2012). Wastes of antibiotics in water were treated by many traditional techniques which were biological, physical, and chemical with advantages and drawbacks that including efficiency and cost (Farhadi *et al.,* 2012), (Boncukcuoðlu *et al.,* 2016), (Asghar *et al.,* 2015). The conventional treatment methods for wastewater are not effectively degrading the recalcitrant organics matter, as well as the ability to produce another toxic byproduct makes them required subsequent pollution treatment or a combination with other technologies (Homem and Santos 2011), (Coria *et al.,* 2014), (Borghi and Palma 2014), (Morsi *et al.,* 2011), (Khezrianjoo and Revanasiddappa 2015), (Yahiaoui *et al.,* 2013), (Sirés and Brillas 2012).

The constraints of the conventional treatment processes and cost were a significant challenge for the suitable ultimate selection (Asghar *et al.,* 2015). The

***Author for correspondence :** E-mail : rasha.waheeb@yahoo.com, dr.yaaroub@yahoo.com

protection from the antibiotic's harmful effects requires to develop advanced remediation technology that achieves the antibiotics' strict discharge limit (Barhoumi *et al.,* 2015), (Sangeetha *et al.,* 2015). Recently researchers studied advanced oxidation processes (AOPs) as emerging and alternative antibiotics treatment process, *e.g.* ozonation, Fenton and photo-Fenton oxidation, photocatalysis, and electrochemical advanced oxidation (Daghrir and Drogui 2013), (Farhadi *et al.,* 2012), (Derakhshan *et al.,* 2016), (Sirés and Brillas 2012), (Anglada *et al.,* 2009), (Guo *et al.,* 2017), (Ganiyu *et al.,* 2015). AOPs have been employed to generate *in situ* strongly oxidative hydroxyl radicals (•OH) at sufficient concentrations which are reacting non-selectively with the organic contaminations, according to equation (1) (Farhadi *et al.,* 2012), (Oturan and Aaron 2014), (Derakhshan *et al.,* 2016), (Oturan and Aaron 2014), (Ganiyu 2017).

$$AOPs \quad OH \quad ^{Pollutants} \quad CO_2 \quad H_2O \qquad (1)$$

(Farhadi *et al.,* 2012).

AOPs can be implemented to degrade many organic and inorganic compounds at room temperature and atmospheric pressure in an easy, efficient and environmentally friendly manner without the addition of chemical reagents. (Oturan and Aaron 2014), (Rivera-Utrilla *et al.,* 2013), (Oturan and Aaron 2014), (Ganiyu *et al.,* 2015), (Ganiyu 2017). Furthermore, the organic compounds including antibiotics could be mineralized by AOPs methods at a high degree due to the converting toxic byproducts to CO_2 and water, unlike the conventional processes which produce a significant amount of polluted sludge (Daghrir and Drogui 2013), (Feng *et al.,* 2013), (Ganzenko *et al.,* 2014), (Oturan *et al.,* 2013).

The electrochemical advanced oxidation techniques (such as anodic oxidation, electrocoagulation, and electro-Fenton) have been extensively investigated (Askari *et al.,* 2014), (Sirés and Brillas 2012), (Ganzenko *et al.,* 2014), (Ganiyu *et al.,* 2015). In the electrochemical treatment, the combination of chemistry where in situ generate chemical oxidant and electronic science (electron transfer) makes it an effective treatment method that works flexibly and cleanly (Daghrir and Drogui 2013), (García-Gómez *et al.,* 2014), (Särkkä *et al.,* 2015).

Among the electrochemical advanced oxidation processes (EAOPs), the anodic oxidation was extensively investigated at the laboratory scale (Feng *et al.,* 2013). Which have been able to destruct recalcitrant contaminations powerfully from wastewater that exist at low concentrations including pharmaceutical pollutants (Gnamba *et al.,* 2015). In anodic oxidation, the pollutants degradation process involves the oxidation procedure by direct and indirect mechanisms, which depends on the anode properties and process conditions (Feng *et al.,* 2013), (Körbahti and Demirbüken 2017), (Sirés and Brillas 2012), (Boncukcuoğlu *et al.,* 2016), (Carvalho *et al.,* 2011), (Ganzenko *et al.,* 2014), (Rajkumar and Palanivelu 2004), (Stupar *et al.,* 2017), (Särkkä *et al.,* 2015). The advanced oxidation process has been called the clean reagent process due to the use of the electron as main destruction reaction reagent in presence of electrolyte without the use of chemicals or producing polluted by-products or sludge (Morsi *et al.,* 2011), (Carlesi Jara *et al.,* 2007), (Valica and Hostin 2016).

In direct oxidation, the hydroxyl anions have been produced by an electro-oxidation reaction of water, under either acidic, basic, or neutral conditions (Brillas *et al.,* 2005), (Domínguez *et al.,* 2010), (Moura *et al.,* 2016). The produced anions were lost electrons and adsorbed physically at the active anode surface (M) converting to hydroxyl free radical where denoted by (M(•OH)) and described by equation (2) (González *et al.,* 2011), (Domínguez *et al.,* 2010), (Moura *et al.,* 2016), (Ganiyu 2017), (Valica and Hostin 2016), (Chang *et al.,* 2009), (Ganzenko *et al.,* 2014).

$$M + H_2O\ ' \rightarrow M(^{\bullet}OH) + H^+ + e^- \qquad (2)$$

(Brillas *et al.,* 2005), (González *et al.,* 2011).

At the anode surface, The organic pollutants can be activated due to electron extraction by the short live hydroxyl radical forming organic radical which undergo successive rapid degradation reactions resulting in water, carbon dioxide, and inorganics ions (Chang *et al.,* 2009), (Babu *et al.,* 2009), (Moura *et al.,* 2016), (Garcia-Segura *et al.,* 2015), (Ganiyu 2017). So, the direct oxidation is limited to the surface (Ganzenko *et al.,* 2014) and could be described by equation (3) (Chang *et al.,* 2009), (Al-Jawad 2018).

$$M(^{\bullet}OH) + R\ ' \rightarrow M + CO_2 + H_2O + \text{Inorganics Ions} + e^- \qquad (3)$$

The organic direct oxidation and mineralization could be summarized in Fig. 1.

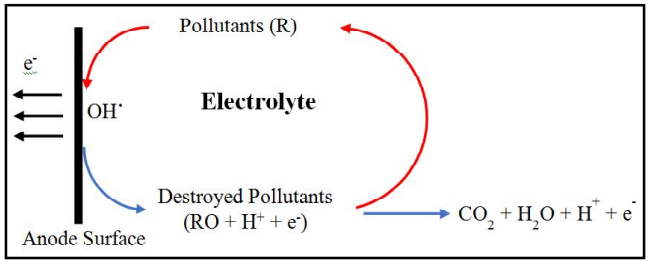

Fig. 1: the direct anodic oxidation of organic matter.

In indirect electro-oxidation, the active oxidation species such as chlorine (Cl), chlorine dioxide (ClO_2), hypochlorite (OCl^-), and hypochlorous acid (HOCl) could be generated in situ on the anode surface by the dissociation of chloride salts in aqueous solution like sodium or potassium chloride which also increase the solution electrical conductivity (Alaani *et al.,* 2018), (Ganiyu 2017), (Al-Jawad 2018), (Ganzenko *et al.,* 2014).

According to equation (4) (Ganzenko *et al.,* 2014), (Al-Jawad 2018), the presence of electrogenerated active chlorine mixture at high concentration as oxidizing species significantly improving the organic compounds degradation where a sequence of oxidizing reactions of organic pollutants occur broach the anode surface or/and in the bulk substrate solution. (Alaani *et al.,* 2018), (Ganiyu 2017), (Al-Jawad 2018), (Ganzenko *et al.,* 2014), (Rajkumar and Palanivelu 2004).

$$\text{Organics} + OCl^- \text{'} \rightarrow \text{intermediate'} \rightarrow CO_2 + H_2O + Cl^- \quad (4)$$

(Shanthi *et al.,* 2011)

The indirect electro-oxidation of organic pollutants could be briefly described by Fig. 2.

As compared with the direct oxidation the indirect process is more efficient in organic degradation (Stupar *et al.,* 2017) due to the effect of reaction kinetics and mass transfer limitations which make the direct oxidation process very slow (Körbahti and Demirbüken 2017).

Researchers were interested in some factors of the organic electrooxidation process due to their significant effect on the extent of pollutant removal, the nature of by-products, operation cost, and reaction time and kinetics. In this study, the effect of the environmental parameters on the Cephalexin and COD removal efficiencies by anodic oxidation were investigated, such as the pH of the media, the Cephalexin initial concentration, the concentration of the supporting electrolyte NaCl, the temperature of the solution, and the applied current density. Moreover, the Cephalexin removal efficiency was indicated by sensitive bacteria. As well by applying the optimal conditions, the Cephalexin and the accompanied bacteria were removed from real wastewater.

Meterials and Methods

Experimental Setup

For antibiotics anodic degradation, all anodic oxidation experiments were conducted in a glass reactor with dimensions 15×15×15 cm containing 2L of antibiotic polluted wastewater as shown in figures 3. Graphite plate and stainless steel (316 grad) were used as an anode electrode and cathode respectively with dimensions 10×13×0.3 cm and 260 cm^2 surface area for each and were implemented at 2 cm from each other. Direct current electrical power was supplied by power source (Yihua/ China) while the applied voltage and current were measured by voltage and current multimeters (Prokit/ Thailand) respectively. The wastewater temperature has been controlled and mixed at 300 rpm by hotplate magnetic stirrer (Chitransh/Korea). A mercury thermometer was used to measure the contaminated solution temperature.

Chemical Materials

The anodic oxidation experiments were carried out to remove Cephalexin antibiotic in wastewater as real and synthetic wastewater. To prepare the synthetic wastewater, the antibiotics were supplied from The General Company for Pharmaceutical Industry (SDI) and mixed with deionized water (local). Hydrochloric acid (HCl) and sodium hydroxide (NaOH) (Sigma-Aldrich) were used to adjust the initial pH, meanwhile sodium chloride (NaCl) salt was used as supporting electrolyte and as chlorine source for indirect process. The real wastewater that polluted by Cephalexin was supplied

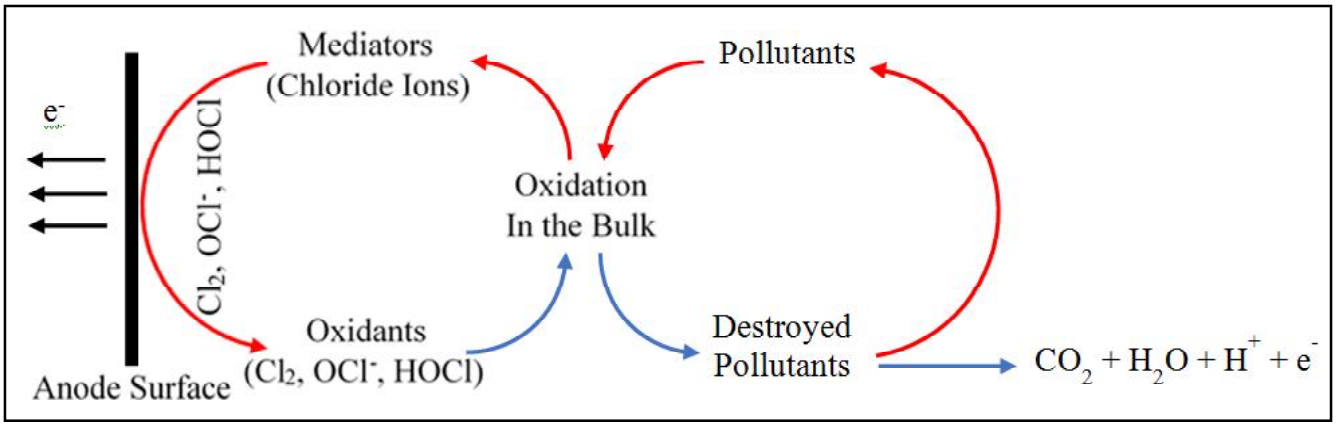

Fig. 2: the indirect anodic oxidation of organic matter.

from the effluent of the batch process of preparation Cephalexin syrup.

The Effective Parameters

The experiments were carried out under many different environmental parameters to study their effect on the antibiotic and COD removal efficiencies and the physical and chemical properties. Where, the concentrations of antibiotic were indicated and measured using the High-Performance Liquid Chromatography (HPLC) and the COD was measured by the DR5000 COD Spectrophotometer / Lovibond. the antibiotic and COD removal efficiencies were calculated according to equation (5)

$$\text{Removal Efficiency (\%)} \quad \frac{C_0 \quad C}{C_0} \quad 100 \qquad (5)$$

Preparation of Culture Media:

For the growth test of bacteria, two culture medias were used which are Muller Hinton agar and nutrient agar. According to the instructions of manufacture companies, the culture medias were prepared in the laboratory and sterilized by the autoclave (15 lbf/ inch^2) at 121 for 15 minutes.

The Biological Indication of Antibiotic Anodic Degradation

The extent of the anodic degradation of the antibiotics in the wastewater was indicated by testing the growth of gram-positive bacteria (*Staphylococcus aureus* which is sensitive to the antibiotic) on Muller-Hinton media as a sign of the degradation of antibiotics (Abd-alwahab and Rasheed 2015).

Total Viable Bacteria Count

This measurement was conducted to count the validity of bacteria in real pharmaceutical wastewaters before and after degradation. The enumeration of the bacteria was done by using indirect cell count (spread plate method) on solid nutrient agar media.

Results and Discussion

The first group of the experiments were carried out by using simulated contaminated water by Cephalexin to study the effect of anodic oxidation parameters on antibiotic removal efficiency, such as antibiotics initial concentration, pH, wastewater temperature, NaCl supporting electrolyte concentration, and the applied current density. Furthermore, the removal of the antibiotics from the simulated wastewater were indicated by the bacteriological test at Cephalexin concentration of 100 ppm. The second group were carried out by treating a real wastewater contaminated by the antibiotics by the two removal processes (direct and indirect) and studying the effect of the anodic oxidation on the combined bacteria in the real wastewater.

The Effect of Initial pH

The results show the maximum removal efficiency of the Cephalexin was (63%) at pH = 3, where the experiments were conducted at 100 ppm initial Cephalexin concentration and 130 mA/cm^2 applied current density, and solution temperature 20ºC (Fig. 4). While, the maximum COD removal efficiency was (19.83%) (Table 1).

The results show that the removal of the Cephalexin is more efficient in acidic medium than the neutral and alkaline medium, where the neutral and alkaline medium nearly have the same effect on antibiotic degradation efficiency. This behavior can be attributed to the high formation of the hydroxyl radicals in the acidic medium, which is a strong oxidizer to the organic materials.

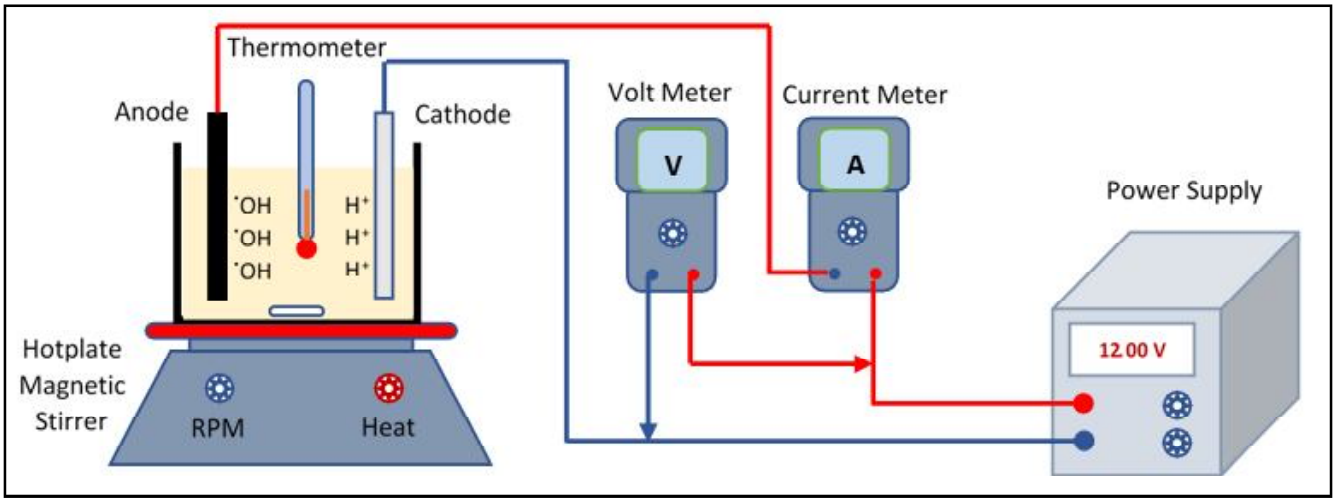

Fig. 3: Schematic of experimental setup.

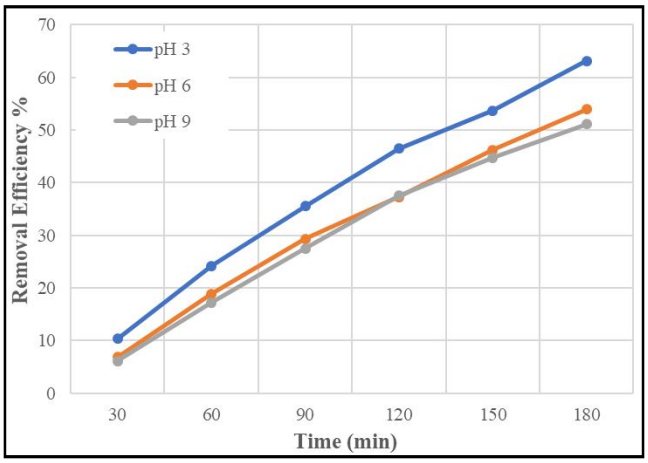

Fig. 4: the effect of pH on the removal of Cephalexin (100 ppm, 20 ºC, and 130 mA/cm^2).

Table 1: The influence initial pH on COD removal efficiency at (100 ppm, 20 ºC 130 mA/cm^2)

pH	COD (mg/l)		
	Before	After	Removal (%)
3	121	97	19.83
6	119	97	18.49
9	120	102	15.00

Whereas, in the neutral and alkaline medium, it is tending to increase the rate of oxygen disturbing the diffusion of the antibiotic to the anode surface. These results agree with (Garcia-Segura *et al.,* 2015), (Sangeetha *et al.,* 2015).

The Effect of Antibiotics Initial Concentration

The maximum Cephalexin removal efficiency was 63% which was obtained at Cephalexin initial concentration 100 ppm, solution temperature 20 ºC, and applied current density 130 mA/cm2 (Fig. 5). The maximum COD removal efficiency was 18.49% (Table 2).

In this study, antibiotics removal efficiency is significantly influenced by antibiotics' initial concentration. The antibiotic removal efficiency increases with the initial antibiotic concentration decreasing. This behavior is ascribed to the limited amount of hydroxyl free radical produced by the electrooxidation process, which is insufficient to oxidize higher concentration at the same conditions (Yahiaoui *et al.,* 2013), (Chen *et al.,* 2014) and this behavior agrees with (Dirany *et al.,* 2012).

The Effect of Electrolyte Temperature

For Cephalexin, the maximum degradation efficiency was obtained at 40ºC. It was 74.33% which was obtained at 100 ppm Cephalexin initial concentration and applied current density 130 mA/cm2 (Fig. 6). As well, the maximum COD removal efficiency for Cephalexin was

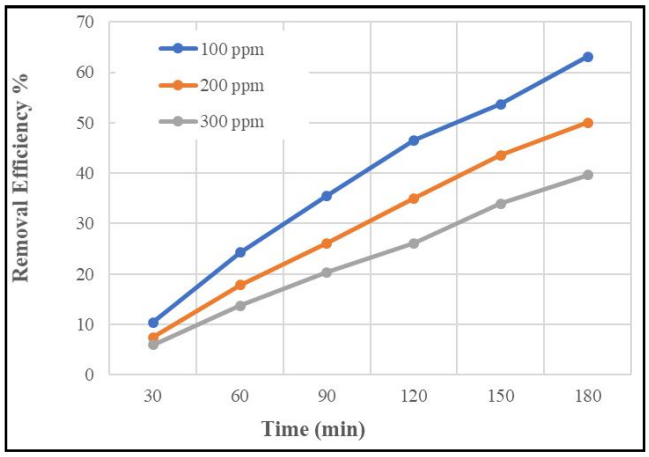

Fig. 5: the effect of Cephalexin initial concentration on the removal efficiency (20 ºC and 130 mA/cm^2).

Table 2: The influence of antibiotic initial concentration on COD removal efficiency at (20 ºC, 130 mA/cm2).

Initial Concentration (ppm)	COD (mg/l)		
	Before	After	Removal (%)
100	119	97	18.49
200	244	208	14.75
300	371	328	11.59

obtained at 40ºC and it was 18.49% (Table 3).

The study observes that the degradation of the antibiotic is more efficient at high temperatures, and increases with the solution temperature increasing. As the solution temperature increases, the viscosity decreases and facilitates the diffusion of the antibiotic molecules to

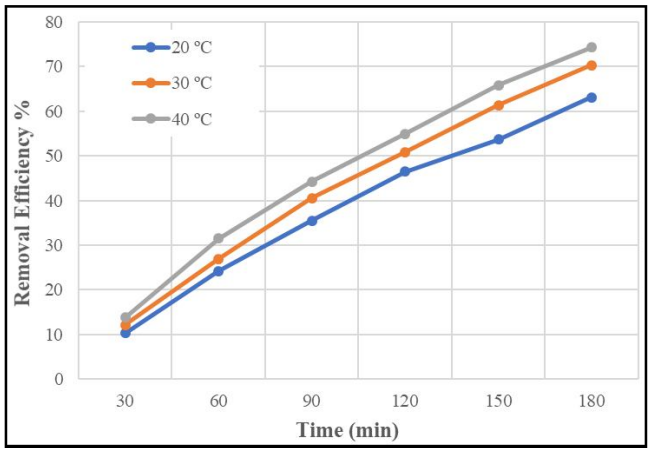

Fig. 6: The effect of solution temperature on the antibiotic removal efficiency (100 ppm and 130 mA/cm^2).

Table 3: The influence of solution temperature on COD removal efficiency at (100 ppm, 130 mA/cm^2).

Temperature (ºC)	COD (mg/l)		
	Before	After	Removal (%)
20	119	97	18.49
30	120	95	20.83
40	119	94	21.01

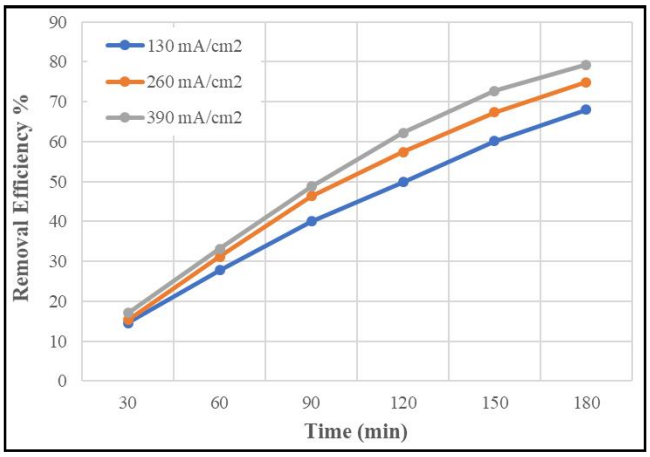

Fig. 7: the effect of applied current density on the Cephalexin removal efficiency (100 ppm and 20ºC).

Table 4: The influence of applied current density on COD removal efficiency at (100 ppm, 20ºC).

Applied Current Before (mA/cm^2)	COD (mg/l)		
	Before	After	Removal (%)
0.5	119	97	18.49
1.0	123	99	19.51
1.5	124	96	22.58

the anode surface and increasing the reaction rate at the surface. It is also increasing the reaction kinetics by increasing the reaction constant, which depends on the reactant temperature (Yahiaoui *et al.,* 2013), (Chen *et al.,* 2014).

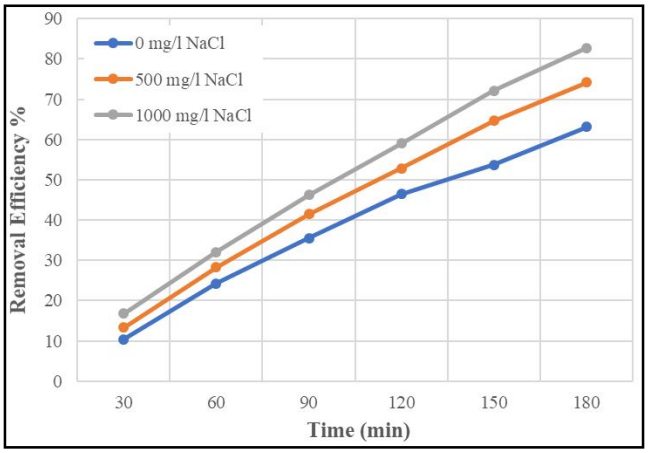

Fig. 8: the effect of NaCl concentration on the removal Cephalexin efficiency (100 ppm, 20ºC and 130 mA/cm^2).

Table 5: The influence of NaCl concentration on COD removal efficiency at (100 ppm, 20 ºC 130 mA/cm^2).

NaCl Concentration (mg/l)	COD (mg/l)		
	Before	After	Removal (%)
0	119	97	18.49
500	124	96	22.58
1000	127	94	25.98

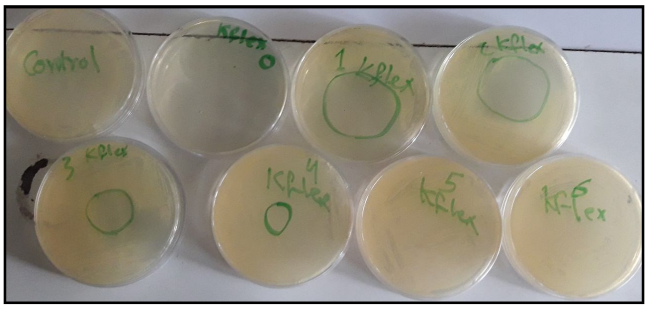

Fig. 9: The bacterial inhibition zone test for the indication of Cephalexin antibiotic anodic degradation.

The Effect of Applied Current Density

The maximum degradation of Cephalexin was 73.7% at higher applied current density 390 mA/cm^2, 100 ppm initial concentration for each and 20ºC solution temperature (Fig. 7). Also, at higher applied current density, the maximum COD removal efficiency was 22.58% (Table 4).

The study shows that the increase of current density improves the antibiotics removal efficiency due to increasing the hydroxyl free radical production rate at the anode surface. More amount of hydroxyl radicals degrades more amount of antibiotics molecules and decreasing the antibiotics removal time (Coria *et al.,* 2014), (Yahiaoui *et al.,* 2013).

The Effect of the Concentration of NaCl Supporting Electrolyte

In the indirect anodic oxidation, the maximum antibiotic removal percent was 82.75, at Cephalexin initial concentration 100 ppm and NaCl concentration 1000 mg/l (Fig. 8). Meanwhile, the maximum COD removal efficiency was 25.98% (Table 5).

The results show that antibiotics removal efficiency is highly increasing as the NaCl concentration in the solution increases. The anodic oxidation was improved by the presence of chloride ions in the electrolyte due to

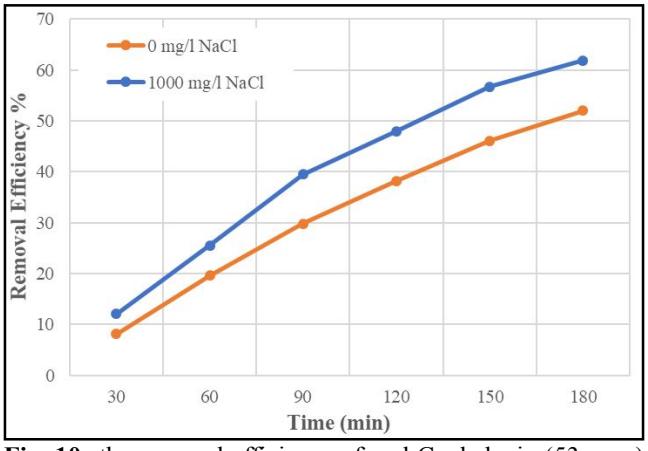

Fig. 10: the removal efficiency of real Cephalexin (53 ppm) contaminated wastewater (20 ºC, and 130 mA/cm^2).

Table 6: The COD removal efficiency of real wastewater.

NaCl Concentration (mg/l)	COD (mg/l)		
	Before	After	Removal (%)
0	834	766	8.15
1000	834	701	15.95

Table 7: The number of bacterial total viable count.

Total Viable Count (Cell×10^{-8}) (cfu/ml)	
Before Treatment	After Treatment
1.07	0.08

the production of chlorine, which oxidizes the organics indirectly and degrades the antibiotics in the bulk of the electrolyte (Babu *et al.,* 2009), (Indu *et al.,* 2014).

Bacterial Detection for The Antibiotic Anodic Degradation

The inhibition zone test was carried out to detect the extent of the antibiotic anodic degradation using simulated wastewater at Cephalexin initial concentration 100 ppm, pH (6), and current density 0.5 mA/cm^2. The test reveals that the inhibition zone decreases as the anodic degradation time increasing (Fig. 9). This behavior indicates that the bacteria growth has been affected by the presence of the antibiotics in water, so the decrease in the inhibition zone is attributed to the effectiveness of the antibiotic degradation by anodic oxidation to the intermediate compounds that also do not affect the bacteria growth.

The Removal of Cephalexin from The Real Wastewater

The real contaminated water by Cephalexin was supplied from The General Company for Pharmaceutical Industry (SDI) with Cephalexin concentration 53ppm and COD 834 mg/l. For direct removal process, the results show that the maximum antibiotics removal efficiency is 52.073% (see Fig. 10) and COD removal efficiency was 8.15% (Table 6). Whereas, for the indirect removal process, the higher antibiotics removal efficiency was 61.87% (Fig. 10) and the COD removal efficiency was 15.95% (Table 6).

The Cephalexin removal efficiency for real wastewater is less than the simulated as well as the COD removal percent due to the high initial COD (there is another degradable organic) which needs very high production hydroxyl free radical for the anodic degradation.

The Wastewater Sterilization by Anodic Oxidation

During the experiments of antibiotic direct oxidation, the number of bacteria colonies (maybe the antibiotic resistance bacteria) were counted in the real Cephalexin contaminated water. The results show that the number of the bacteria colonies decreases during the anodic oxidation experiment due to non-selective oxidation by the powerful oxidant hydroxyl free radical that may oxidizes the bacteria cell membrane resulting in the damage of essential cell functions and causing death (Bensalah and Abdel-Wahab 2013), (Jeong *et al.,* 2007).

Conclusion

The anodic degradation of the antibiotic and the COD removal efficiency increases with increasing the solution temperature, the supporting electrolyte concentration, and the applied current density. Whereas, the antibiotic and COD removal efficiencies decrease with the increase in the initial concentration of the antibiotics and the pH media.

The anodic oxidation is very efficient and cost-effective as compared with the traditional processes for antibiotic degradation. Even though the antibiotics removal efficiency is favorable in the presence of NaCl salt and the acidic media, it has a bad impact on the aquatic environment due to the increase in the salinity and acidity of water.

The results of the anodic degradation of the antibiotics do not affect the sensitive bacteria, and therefore the generation of new strains of antibiotic-resistant bacteria is not permitted.

The anodic oxidation of the antibiotic is effective in sterilizing the wastewater from the accompanying bacteria, which may be the antibiotic-resistant bacteria.

References

Abd-alwahab, Zainab I. and Bassam G. Rasheed (2015). "Antibacterial Silver Nanoparticles Produced by Nd/ : *YAG Laser,*" **18(2):** 315–21.

Al-Jawad, Noor Hashim Jawad (2018). "Experinemtal Study on The Effective Parameters for The Removal of Methylene Blue Electrochemically from Aqueos Solutions." Al-Nahrain University.

Alaani, Hashem, Shahir Hashem and Francois Karabet (2018). "Electrochemical Removal of Hydrocortisone from Aqueous Environments Using Aluminum Electrodes." *Oriental Journal of Chemistry,* **34(1):** 203–13.

Anglada, Ángela, Ane Urtiaga and Inmaculada Ortiz (2009). "Contributions of Electrochemical Oxidation to Waste-Water Treatment: Fundamentals and Review of Applications." *Journal of Chemical Technology and Biotechnology,* **84(12):** 1747–55.

Asghar, H.M.A., T. Ahmad, S.N. Hussain and H. Sattar (2015). "Electrochemical Oxidation of Methylene Blue in Aqueous Solution." *International Journal of Chemical Engineering and Applications,* **6(5):** 352–55.

Askari, Masoomeh, Mahmood Alimohammadi, Mohammad Hadi Dehghani, Mohammad Mahdi Emamjomeh and Shahrokh Nazmara (2014). "Removal of Natural Organic Matter from Aqueous Solutions by Electrocoagulation." *Journal of Advances in Environmental Health Research,* **2(2):**

Babu, B. Ramesh, P. Venkatesan, R. Kanimozhi and C. Ahmed Basha (2009). "Removal of Pharmaceuticals from Wastewater by Electrochemical Oxidation Using Cylindrical Flow Reactor and Optimization of Treatment Conditions." *Journal of Environmental Science and Health - Part A Toxic/Hazardous Substances and Environmental Engineering,* **44(10):** 985–94.

Barhoumi, Natija, Lazhar Labiadh, Mehmet A. Oturan, Nihal Oturan, Abdellatif Gadri, Salah Ammar and Enric Brillas. (2015). "Electrochemical Mineralization of the Antibiotic Levofloxacin by Electro-Fenton-Pyrite Process." *Chemosphere,* **141:** 250–57.

Bensalah, Nasr and Ahmed Abdel-Wahab (2013). "Electrochemical Inactivation of P. Aeruginosa, A. Hydrophila, L. Pneumophila Using Boron Doped Diamond Anodes." *Journal of Advanced Oxidation Technologies,* **16(1):** 9–15.

Boncukcuoðlu, Recep, Baybars Ali Fil, Alper Erdem Yýlmaz, and Alper Nuhoðlu (2016). "Electro-Oxidation Treatment for Turbidity in Pistachio Processing Industry Wastewater (PPIW)." *Proceedings of the 2nd World Congress on New Technologies,* **1(8):**

Borghi, Alexandre Augusto and Mauri Sergio Alves Palma (2014). "Tetracycline: Production, Waste Treatment and Environmental Impact Assessment." *Brazilian Journal of Pharmaceutical Sciences,* **50(1):** 25–40.

Brillas, Enric, Ignasi Sires, Conchita Arias, Pere Lluis Cabot, Francesc Centellas, Rosa Maria Rodrýiguez and Jose´ Antonio Garrido (2005). "Mineralization of Paracetamol in Aqueous Medium by Anodic Oxidation with a Boron-Doped Diamond Electrode." *Chemosphere,* **58:** 399–406.

Carlesi Jara, C., D. Fino, V. Specchia, G. Saracco and P. Spinelli (2007). "Electrochemical Removal of Antibiotics from Wastewaters." *Applied Catalysis B: Environmental,* **70:** 479–87.

Carvalho, D.A., J.H. Bezerra Rocha, N.S. Fernandes, D.R. DA SILVA and C.A. MARTÍNEZ-HUITLE (2011). "Application of Electrochemical Oxidation as Alternative for Removing Methyl Green Dye from Aqueous Solutions." *Latin American Applied Research,* **41:** 127–33.

Chang, Ming, Chengyao Gao and Juyuan Jiang (2009). "Electrochemical Oxidation of Organic Compounds Using Boron-Doped Diamond Electrode." *Journal of The Electrochemical Society,* **156(2):** E50–54.

Chen, Te San, Ren Wei Tsai, Yu Syuan Chen and Kuo Lin Huang (2014). "Electrochemical Degradation of Tetracycline on BDD in Aqueous Solutions." *International Journal of Electrochemical Science,* **9(12):** 8422–34.

Coria, Gabriela, José L. Nava and Gilberto Carreño (2014). "Electrooxidation of Diclofenac in Synthetic Pharmaceutical Wastewater Using an Electrochemical Reactor Equipped with a Boron Doped Diamond Electrode." *Journal of the Mexican Chemical Society,* **58(3):** 303–8.

Daghrir, R. and P. Drogui (2013). "Tetracycline Antibiotics in the Environment: A Review." *Environmental Chemistry Letters,* **11(3):** 209–27.

Derakhshan, Zahra, Mehdi Mokhtari, Fatemeh Babaei, Roya Malek Ahmadi, Mohammad Hassan Ehrampoush and Mohammad Faramarzian (2016). "Removal Methods of Antibiotic Compounds from Aqueous Environments – A Review." *Journal of Environmental Health and Sustainable Development,* **1(1):** 43–62.

Dirany, Ahmad, Ignasi Sireìs, Nihal Oturan, Ali O Zcan and Mehmet A. Oturan (2012). "Electrochemical Treatment of the Antibiotic Sulfachloropyridazine: Kinetics, Reaction Pathways, and Toxicity Evolution." *Environmental Science and Technology,* **46(7):** 4074–82.

Domínguez, Joaquín R., T. González, P. Palo and J. Sánchez-Martín. (2010). "Anodic Oxidation of Ketoprofen on Boron-Doped Diamond (BDD) Electrodes. Role of Operative Parameters." *Chemical Engineering Journal,* **162(3):** 1012–18.

Farhadi, Sajjad, Behnoush Aminzadeh, Ali Torabian, Vahid Khatibikamal and Mohammad Alizadeh Fard (2012). "Comparison of COD Removal from Pharmaceutical Wastewater by Electrocoagulation, Photoelectro-coagulation, Peroxi-Electrocoagulation and Peroxi-Photoelectrocoagulation Processes." *Journal of Hazardous Materials,* **219(220):** 35–42.

Feng, Ling, Eric D. van Hullebusch, Manuel A. Rodrigo, Giovanni Esposito and Mehmet A. Oturan (2013). "Removal of Residual Anti-Inflammatory and Analgesic Pharmaceuticals from Aqueous Systems by Electrochemical Advanced Oxidation Processes. A Review." *Chemical Engineering Journal,* **228:** 944–64.

Ganiyu, Soliu (2017). "Electrochemical Advanced Oxidation Processes for Removal of Pharmaceuticals from Water/: Performance Studies for Sub-Stoichiometric Titanium Oxide Anode and Hierarchical Layered Double Hydroxide Modified Carbon Felt Cathode." Paris-Est University.

Ganiyu, Soliu O., Eric D. Van Hullebusch, Marc Cretin, Giovanni Esposito and Mehmet A. Oturan (2015). "Coupling of Membrane Filtration and Advanced Oxidation Processes for Removal of Pharmaceutical Residues: A Critical Review." *Separation and Purification Technology,* **156:** 891–914.

Ganzenko, Oleksandra, David Huguenot, Eric D. van Hullebusch, Giovanni Esposito and Mehmet A. Oturan (2014). "Electrochemical Advanced Oxidation and Biological Processes for Wastewater Treatment: A Review

of the Combined Approaches." *Environmental Science and Pollution Research,* **21(14):** 8493–8524.

García-Gómez, C., P. Drogui, F. Zaviska, B. Seyhi, P. Gortáres-Moroyoqui, G. Buelna, C. Neira-Sáenz, M. Estrada-Alvarado and R.G. Ulloa-Mercado (2014). "Experimental Design Methodology Applied to Electrochemical Oxidation of Carbamazepine Using Ti/PbO2 and Ti/BDD Electrodes." *Journal of Electroanalytical Chemistry,* **732:** 1–10.

Garcia-Segura, Sergi, Jürg Keller, Enric Brillas and Jelena Radjenovic (2015). "Removal of Organic Contaminants from Secondary Effluent by Anodic Oxidation with a Boron-Doped Diamond Anode as Tertiary Treatment." *Journal of Hazardous Materials,* **283:** 551–57.

Gnamba, Corneil Quand-Meme, Foffié Thiéry Auguste Appia, Evelyne Marie Hélène Loba, Ibrahima Sanogo and Lassine Ouattara (2015). "Electrochemical Oxidation of Amoxicillin in Its Pharmaceutical Formulation at Boron Doped Diamond (BDD) Electrode." *Journal of Electrochemical Science and Engineering,* **5(2):** 129–43.

González, Teresa, Joaquín R. Domínguez, Patricia Palo, Jesús Sánchez-Martín and Eduardo M. Cuerda-Correa (2011). "Development and Optimization of the BDD-Electrochemical Oxidation of the Antibiotic Trimethoprim in Aqueous Solution." *Desalination,* **280:** 197–202.

Guo, Wan-Qian, Yao-Yao Wu and Juan-Shan Du (2017). "Advanced Oxidation Processes for Antibiotics Removal: A Review." **21:** 1–14.

Homem, Vera and Lúcia Santos (2011). "Degradation and Removal Methods of Antibiotics from Aqueous Matrices - A Review." *Journal of Environmental Management,* **92(10):** 2304–47.

Indu, M.S., A.K. Gupta and C. Sahoo (2014). "Electrochemical Oxidation of Methylene Blue Using Lead Acid Battery Anode." *APCBEE Procedia,* **9:** 70–74.

Jeong, Joonseon, Jee Yeon Kim, Min Cho, Wonyong Choi and Jeyong Yoon (2007). "Inactivation of Escherichia Coli in the Electrochemical Disinfection Process Using a Pt Anode." *Chemosphere,* **67(4):** 652–59.

Khezrianjoo, Sajjad and Hosakere Doddarevanna Revanasiddappa (2015). "Electrochemical Oxidation of M-Cresol Purple Dye in Aqueous Media." *Water Quality Research Journal of Canada,* **50(4):** 305–13.

Körbahti, Bahadýr K. and Pelin Demirbüken (2017). "Electrochemical Oxidation of Resorcinol in Aqueous Medium Using Boron-Doped Diamond Anode: Reaction Kinetics and Process Optimization with Response Surface Methodology." *Frontiers in Chemistry,* **5(October):** 1–14.

Morsi, M.S., A.A. Al-Sarawy and W.A. Shehab El-Dein (2011). "Electrochemical Degradation of Some Organic Dyes by Electrochemical Oxidation on a Pb/PbO2 Electrode." *Desalination and Water Treatment,* **26:** 301–8.

Moura, Dayanne Chianca De, Marco Antonio Quiroz, Djalma Ribeiro Da Silva, Ricardo Salazar and Carlos Alberto Martínez-Huitle (2016). "Electrochemical Degradation of Acid Blue 113 Dye Using TiO2-Nanotubes Decorated with PbO2 as Anode." *Environmental Nanotechnology, Monitoring and Management,* **5:** 13–20.

Oturan, Mehmet A. and Jean Jacques Aaron (2014). "Advanced Oxidation Processes in Water/Wastewater Treatment: Principles and Applications. A Review." *Critical Reviews in Environmental Science and Technology,* **44(23):** 2577–2641.

Oturan, Nihal, Jie Wu, Hui Zhang, Virender K. Sharma and Mehmet A. Oturan. (2013). "Electrocatalytic Destruction of the Antibiotic Tetracycline in Aqueous Medium by Electrochemical Advanced Oxidation Processes: Effect of Electrode Materials." *Applied Catalysis B: Environmental,* **140–141:** 92–97.

Rajkumar, D. and K. Palanivelu (2004). "Electrochemical Treatment of Industrial Wastewater." *Journal of Hazardous Materials,* **113:** 123–29.

Rivera-Utrilla, José, Manuel Sánchez-Polo, María Ángeles Ferro-García, Gonzalo Prados-Joya and Raúl Ocampo-Pérez (2013). "Pharmaceuticals as Emerging Contaminants and Their Removal from Water. A Review." *Chemosphere,* **93(7):** 1268–87.

Sangeetha, V., V. Sivakumar, A. Sudha and K. Kannan (2015). "Electrochemical Degradation of Sago Wastewater Using Ti/PbO2 Electrode: Optimisation Using Response Surface Methodology." *International Journal of Electrochemical Science,* **10(2):** 1506–16.

Särkkä, Heikki, Amit Bhatnagar and Mika Sillanpää (2015). "Recent Developments of Electro-Oxidation in Water Treatment - A Review." *Journal of Electroanalytical Chemistry,* **754:** 46–56.

Shanthi, V., K. Ramanathan and C. Ahmed Basha (2011). "Domestic Sewage Treatment Using Batch Stirred Tank Electrochemical Reactor." *International Journal of ChemTech Research,* **3(3):** 1711–21.

Sirés, Ignasi and Enric Brillas (2012). "Remediation of Water Pollution Caused by Pharmaceutical Residues Based on Electrochemical Separation and Degradation Technologies: A Review." *Environment International,* **40(1):** 212–29.

Stupar, S. Lj, B.N. Grgur, A.E. Onjia and D. Mijin (2017). "Direct and Indirect Electrochemical Degradation of Acid Blue 111 Using IrOX Anode." *International Journal of Electrochemical Science,* **12(9):** 8564–77.

Valica, Martin and Stanislav Hostin (2016). "Electrochemical Treatment of Water Contaminated with Methylorange." *Nova Biotechnologica et Chimica,* **15(1):** 55–64.

Yahiaoui, Idris, Farida Aissani-Benissad, Florence Fourcade and Abdeltif Amrane (2013). "Removal of Tetracycline Hydrochloride from Water Based on Direct Anodic Oxidation (Pb/PbO2 Electrode) Coupled to Activated Sludge Culture." *Chemical Engineering Journal,* **221:** 418–25.