

Electrochemical oxidation process of antibiotics in waste water using double layer cathode reactor

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Abstract

This study involves the electrochemical oxidation of antibiotics in a Trickle Bed Electrochemical Reactor (TBER) that was specially built for the purpose. The reactor's anode is formed of stainless steel, and the gas diffusion cathode (GDC) is constructed of carbon black polytetrafluoroethylene (CB-PTFE), which is placed on stainless steel mesh. Studies of tetracycline antibiotics were performed using 0.2M KOH as the supporting electrolyte and concentrations from 100 mg/L to 500 mg/L and (1-5) volts applied. The TC removal rate is governed by the pseudo-first order kinetic removal efficiency which rises with (1) raising the voltage (from 1 to 5) volt, (2) reducing the temperature, and (3) lowering the concentration of TC (to 100 mg/L from 500 mg/L). As compared to amoxicillin and cephalexin, the rate of removing the TC process is quite high, it was discovered that the electrochemical technique, which has a removal rate of 95.97%, is better at getting rid of the antibiotic.

INTRODUCTION

The lack of water has become a major issue. Wastewater can be treated necessary and recycled, it is feasible to satisfy this demand while using less source water [1]. Treating wastewater containing chemicals is a typical measure to reduce the risks of chemicals and the pollution of environmental [2]. Over the previous 50 years, antibiotic use has allegedly increased between about 100,000 to 200,000 tons worldwide. Most drugs, especially antibiotics, are somewhat biodegradable or even resistant to the standard process of activated sludge used in a treatment plant for wastewater, this causes them to be released into environment [3].

Every year, 700,000 people die from illnesses that are drug-resistant, and it is anticipated that there will be 10 million by 2050. Hence, before releasing antibiotics into the environment, they must be removed [1]. In

environment where nearly all types of antibiotics may be found, waste water treatment plant (WWTP) is one of the most significant sources and sinks of antibiotics [4]. The most prevalent of them are tetracyclines, β -lactams, quinolones, and sulfonamides [5]. The typical range of antibiotic concentrations in local WWTP influents is (1 ng/L to > 1 mg/L) [6]. Modern wastewater antibiotic treatment methods include biological, physical, and advanced oxidation processes (AOPs) [7]. Biological processes, which make up a large portion of WWTPs, often have poor antibiotic degradation rate [8]. Under some circumstances, physical methods like membrane and adsorption separation can successfully remove antibiotics from waste water. Although the disposal of membrane and adsorbents concentrates takes time and requires a lot of energy [9]. Antibiotics can be destroyed or reduced to minute molecules

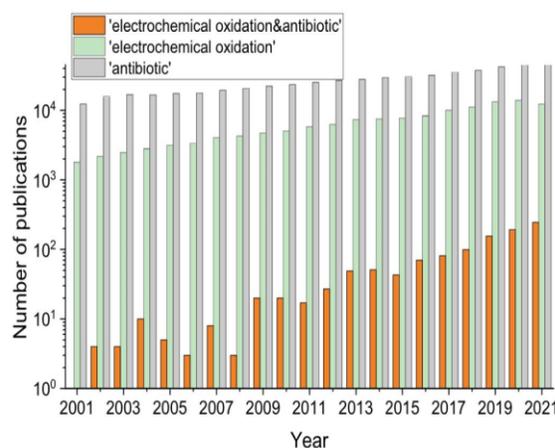
using AOPs. One of the AOPs with the highest efficiency is the electrochemical oxidation (EO) [10]. EO includes both (direct and indirect) oxidation [11]. Recent advancements in electrochemistry include (electrochemically stimulated sulfate reactions, heterogeneous electro-Fenton, and combination methods) [12]. Electrochemical oxidation appears to be one of the AOPs that might be used to purify water [13].

As clean reagents are utilized in the electrochemical treatment, it is an intriguing procedure for harmful organic removal. It is simple to alter the process parameters (current density and electrode potential) and the electron transport rate. Furthermore, this procedure may be used with the surrounding pressure and temperature [12]. Organic molecules immediately interact with higher oxides that have developed on the anode surface or with hydroxyl radicals that have been adsorbed. Contaminant direct electro-oxidation rates are influenced by the anode's catalytic efficiency, based on the current density used and the rates of organic compound diffusion towards the anode's active sites. While operating at high electrode potentials, indirect electro oxidation may also happen as a result of the production of secondary bulk oxidizing agents. Its rate is influenced by the rate at which secondary oxidizing agents (such as hydrogen peroxide, persulfates, chlorine species, etc.) diffuse into solutions, as well as by temperature and pH levels [14].

The amount of research on the subjects of EO or antibiotic has been steadily rising over the last 20 years Figure (1). In the past five years, the number of articles on (EO and antibiotics) increased faster by 29.2% per year than (electrochemical oxidation) by 8.6% per year and (antibiotics) by 8.8% per year, indicating a high probability of degradation of antibiotics by EAOPs [15].

Using various anodes and three dimensional electrodes, Carlesi et al. [16] studied electro-oxidation experiments on water solutions contained lincomycin and ofloxacin. While membrane-divided cells have advantages in molecular degradation and energy usage, high electrode voltages permit significant anodic surface activity and less pollutants. On all the anodes that were studied, Ofloxacin oxidizes efficiently, but Lincomycin oxidizes slowly because of deprotonation. Continuous operation is the best usage for a 3-dimensional electrode, however at the applied voltage, the carbon-based electrode displayed undesirable deterioration. Reactive anodic filter (RAF) technology for SnO₂-Sb was developed by Yang et al. [17] for use in electrochemical oxidation cells (EOC). By pushing wastewater through the RAF, the surface area of electro-active surfaces was expanded and mass transfer was improved. Both the CIP decomposition rate constant and the mass transfer rate constant rose by 12 and 5, respectively. During 200 hr ECO test, the CIP decomposition process featured both direct electron transfer and oxidation of generated radicals, yielding a 99% degradation efficiency.

Fig. (1): Number of articles published between 2002 and 2020 on "electrochemical oxidation and antibiotic" and " antibiotic " and " electrochemical oxidation " subjects.



Chloramphenicol (CAP) was decomposed by co-electrochemical oxidation in a three - electrode system cell with two Cu-Ni/graphene cathodes and a Ti/IrO₂/RuO₂ anode, as studied by Xiaozhe et al. [18]. The CAP was removed within 15 minutes using an improved electrolyte that included 0.05 mol/L Na₂SO₄, 50 mg/L CAP, and a current density of 30 mA cm⁻². Due to its rapid oxidation, Mariam et al. used a BDD anode to investigate the oxidation of sulfamethoxazole (SMX) in artificial aqueous. Degradation performance is greater than 95% with a 350mA current. This shows that electro-oxidation using a BDD anode is effective [19].

The following parameters, according to earlier research, influence the removal of antibiotics: (a) current density; (b) pH level; and (c) electrode materials, where Kitazono et al. (2017) [20] studied at the possibility of electrochemical chlortetracycline oxidation utilizing a Ti/PbO₂ anode. because of the greater current density' higher rate of OH• generation. Chlortetracycline's decomposition was modeled using pseudo-first-order kinetics. The effect of current density on the rate of COD removal from wastewater containing antibiotics like trimethoprim and

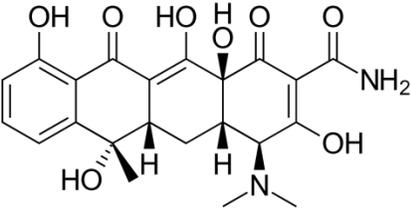
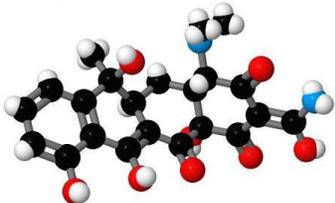
sulfamethoxazole was investigated by Eleoterio and colleagues in 2013 [21]. They indicated that the COD removal efficiency increased as the current density varied between (10 and 100) mA/cm². Using electrochemical oxidation, Moreira et al. (2014) [22] investigated how pH affected the degradation of trimethoprim, at pH 3.0, they discovered that HSO₄ in solution may function as a scavenger for hydroxyl radicals (OH), which may limit trimethoprim's capacity to decay; at pH 4.5, iron precipitate resulted in a decrease in the removal efficiency.

Materials and methods

1. Chemicals and analytical procedure

Tetracycline antibiotic (98% purity) was used, lists some of its properties in Table (1) [23]. The supporting electrolyte used in this study is potassium hydroxide, the molecular formula and molecular weight of which are (KOH and 56.11g/mol), respectively. To clarify the degradation of TC from the electrochemical oxidation process, a UV-vis spectrophotometer was used for this purpose at a wavelength of TC 276 nm [24].

Table 1: Some properties of tetracycline

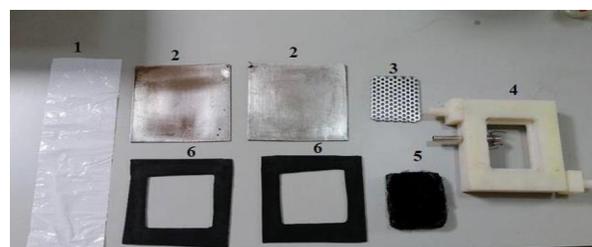
Tetracycline	properties
Molecular formula	C ₂₂ H ₂₄ N ₂ O ₈
2D structure	
3D structure	
Molecular weight (g/mol)	444.435

physical shape	Crystalline powder
Purity	98%
pH	3 - 7
Appearance	Pale yellow
Melting Point (°C)	172 – 174
Storage temperature (°C)	2 - 8
λ max (maximum wavelength) (nm)	359

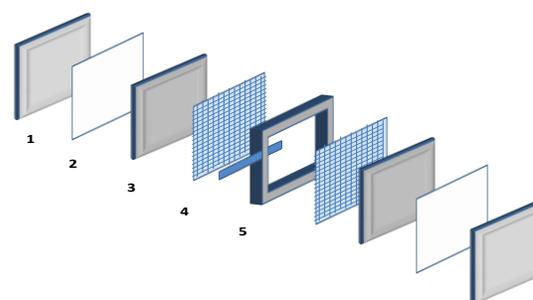
2. Experimental procedure and setup

A TBER reactor consisting of: (1) an anode and a cathode separated by a Cigard membrane to prevent circuit breakage, (2) supported by two stainless steel plates, (3) two stainless steel nets were use, (4) cathode frame, (5) use two C-PTFE block beds as a cathode, and (6) two rubber rings were positioned between the cathode, the anode, and the membrane in order to electrically separate the components and prevent gas and liquid leaks. TBER parts before they are installed are shown in Figure (2). Most of these items were produced local. TC and KOH are dissolved in deionized water in a glass flask and the temperature of the solution is controlled by a water bath from (10-50) c°. Experiments are carried out as shown in Figure (3). For waste removal, a dosing pump, and a liquid flow meter were used to control the liquid flow at a constant flow rate of 200 ml/min. A gas flow meter was used to constant flow rate at 7 L/min.

Fig. (2): Trickle Bed Electrochemical Reactor (TBER) (a) Parts of TBER, (b) Schematic representations of TBER components are shown: 1. the cell body, 2. the membrane, 3. the C-PTFE block, 4. the stainless steel mesh, and 5. the cathode frame.

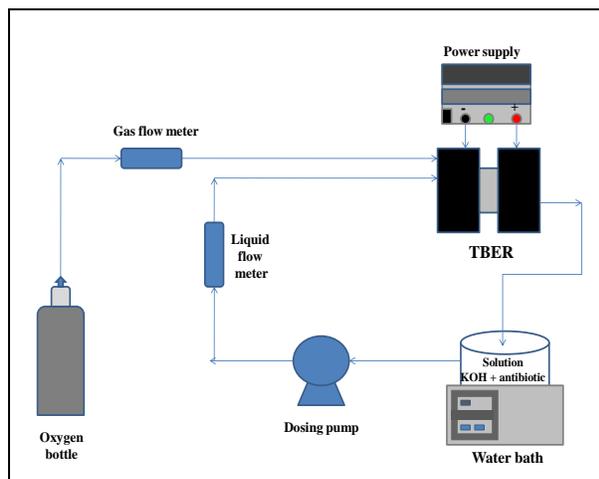


a



b

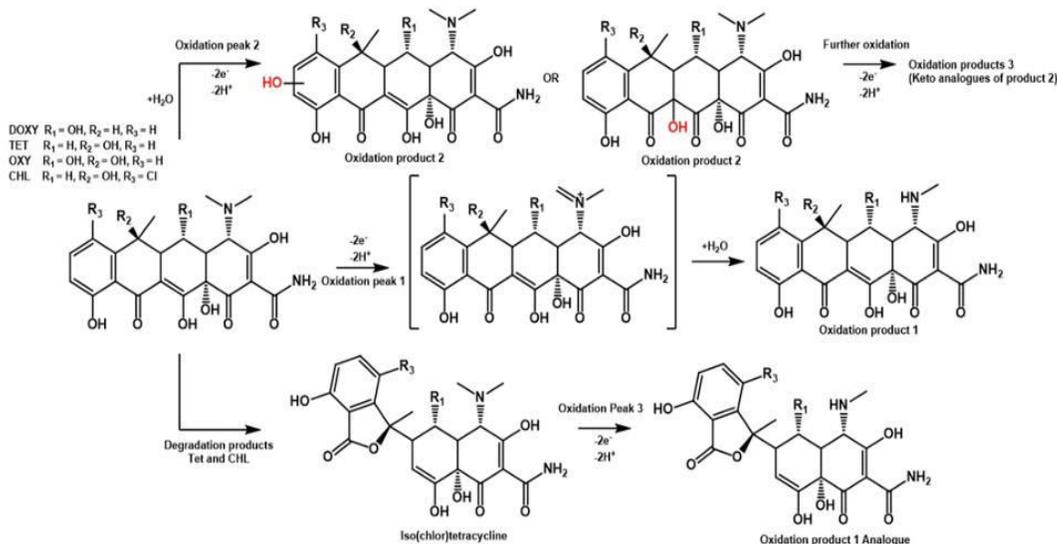
Fig. (3): Diagrammatic representation of the TBER unit's experimental.



3. Intermediates

A common example of an aromatic chemical is tetracycline, which degrades in three stages

Fig. 4: The tetracycline pathway in the electrochemical oxidation process



[25]: (i) the conversion of an aromatic compound to a quinonic compound, (ii) creating aliphatic acids that involves opening rings (oxalic acid is considered as predominant), and (iii) mineralization of CO₂ and water. Three different types of functional groups, including double bonds, phenol groups, and amine groups, are present in TC with very high electron concentrations. These groups are rapidly damaged by radicals produced during electrochemical oxidation. A most reactive of them all, a double bond reacts with radicals preferentially and generates intermediates [26]. The oxidation products of tetracycline that result from the electrochemical oxidation processes are shown in Figure (4) [27].

Results and Discussion

Effect of operating conditions on tetracycline oxidation

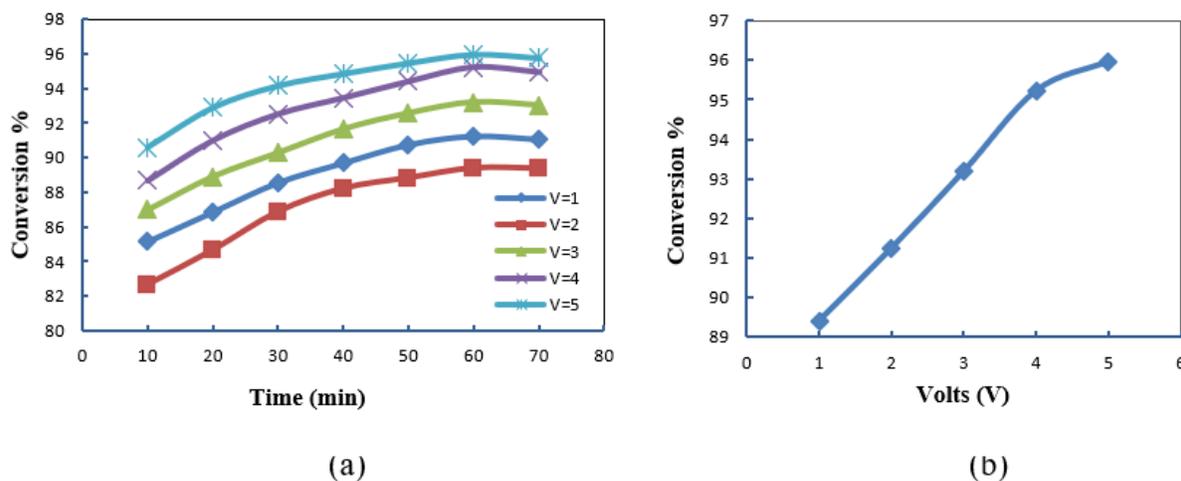
1-Effect of the applied potential

The voltage used has an impact on tetracycline conversion, as seen in Figure (5.a). The conversion of tetracycline increased when the

applied voltage increased from (1-5) volts. Conversion increased from (89.4 to 95.97). The in-situ oxidation of TC is influenced by time when various voltages are applied. It is evident that during the first 60 min of operation, the rate of tetracycline removal increased with increasing time at all voltages. This is as a result of the voltage increasing the

production of hydrogen peroxide, which accelerates the oxidation of tetracycline [28]. Tetracycline conversion achieves its steady-state value and is essentially constant for the remainder of the electrolysis duration after 60 minutes due to the presence of H₂O₂ concentration [29]. It can be pointed out to an important point of the effect of the applied voltage on the rate of removal of tetracycline that as the voltage increases, its effect on the rate of removal weakens because the concentration of hydrogen peroxide decreases,

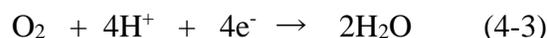
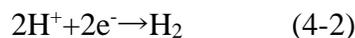
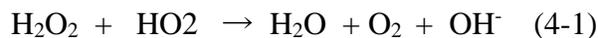
Fig. 5: Shows the effect of the applied potentials (1, 2, 3, 4, and 5 V) on the conversion of tetracycline 100 mg/L tetracycline concentration, and 10 °C reaction temperature (a) by the time (b) 60 min



2-Effect of the tetracycline initial concentration

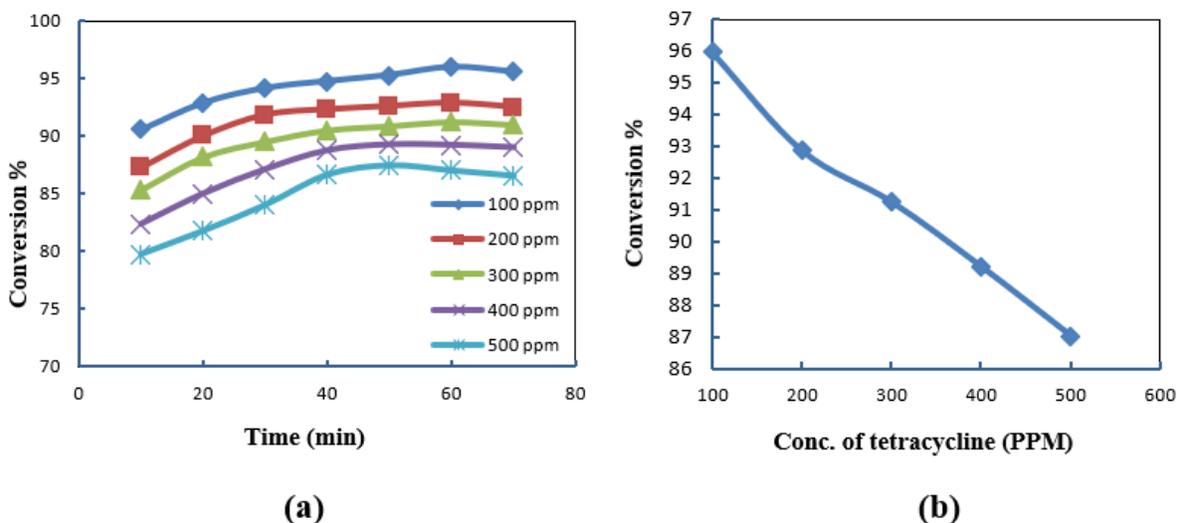
According to Figure (6.a), the conversion of TC decreases as the initial concentration of tetracycline rises. For a constant reaction time of 60 minutes, Figure (6.b) illustrates the impact of the initial tetracycline concentration on the in situ oxidation of TC. It is evident that the conversion of TC reduced with increasing TC initial concentration, peaking at 95.97% at 100 mg/L and continuing to decline with successive increases to 500 mg/L. The findings of the current investigation supported Zhang's findings [32] by showing that when

and this can be seen through the values of the removal rate at 4 and 5 volts where they are close, and to improve efficiency, the voltage is set at specific value [30]. It's possible that this is due to the increasing side reactions at higher voltages. The following reactions are shown [31]:



TC starting concentration grew from (20 to 100) mg/L by utilizing H₂O₂ as an oxidant, the removal rate of TC reduced from (75% to 64%). The conversion may have been reduced because hydroxyl radicals were still available in sufficient quantities despite changes in tetracycline concentration, or raising the initial concentration could improve the side reactions, resulting in the production of additional intermediates such oxalic acid, carbamic acid, formic acid, and auronon oxalic acid.

Fig. 6: Shows the effect of the initial concentration of TC (100, 200, 300, 400, and 500 mg/L) on the conversion of tetracycline at 5V, and 10 °C reaction temperature (a) by the time (b) 60 min



3-Effect of the reaction temperature

Temperature has an impact on TC conversion. When the reaction temperature drops, Figure (7.a) demonstrates that the conversion of TC increases. TC conversion at a constant reaction time of 60 minutes is shown in Figure (7.b) as a function of reaction temperature. As can be observed, the conversion of tetracycline rose as reaction temperature declined, peaking at 95.97% at 10 °C, and then decreased as reaction temperature increased further. This finding indicates that as the temperature drops, more oxygen dissolves in the electrolyte and

more hydrogen peroxide is produced, increasing the rate at which tetracycline will be oxidized [33,34,35]. High temperatures cause hydrogen peroxide to break down and lower oxygen's solubility in the electrolyte, both of which reduce TC oxidation conversion. Furthermore, by delaying the non-faradic reaction below [36], the self-decomposition of hydrogen peroxide is prevented at lower reaction temperatures:

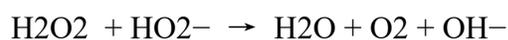
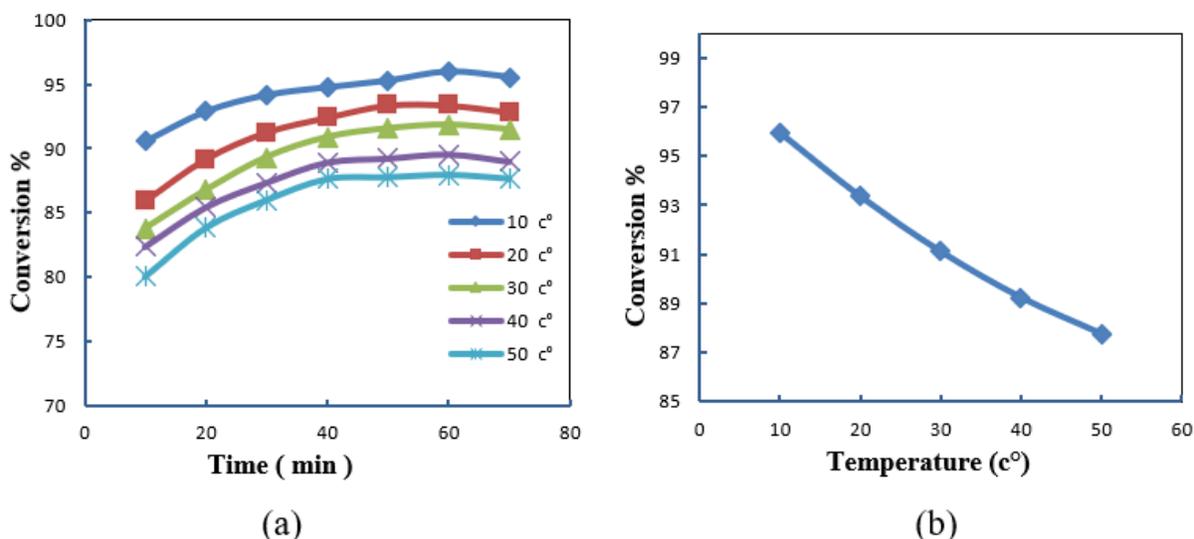


Fig. 7: Shows the effect of temperature reaction (10, 20, 30, 40, and 50 c°) on the conversion of tetracycline at 5V, and 100 mg/L initial concentration of TC (a) by the time (b) 60 min



Comparison removal of antibiotics by electrochemical oxidation process

The structure of TC, which consists of four linearly oriented hexagonal rings with different double bonds and many functional groups, must be emphasized since it lends the compound its robust and complex features [37]. In this investigation, two other antibiotics, amoxicillin (AMX) and cephalexin (CFX), were employed to compare how well they worked with tetracycline under ideal circumstances discovered in earlier studies utilizing the TC antibiotic, which, under similar circumstances, were carried out on amoxicillin and cephalexin at wavelengths (265 and 261, respectively) [38,39]. The three antibiotics' structures share a considerable deal in common, having the identical phenolic and di(methyl) amino substituents [40] as seen in Figure (8), as shown in Figure (9), where the maximum removal of TC, AMX, and CFX after 1 hour was 95.97%, 32.85%, and 40.5%, respectively, it permits straightforward electrochemical discrimination from other antibiotic types, such as CEF and AMX. The aforementioned moieties are therefore expected to be involved in the electrochemical

oxidation of TC. This is in accordance with Cánovas et al. [41], which examined antibiotics using electrochemical oxidation and found that tetracycline was more effectively removed than amoxicillin and cephalexin.

Table 2: Tetracycline electrochemical oxidation conversion with amoxicillin and cephalexin under the optimum conditions (5V, 100 mg/L antibiotic, and 10 °C).

Time (min)	TC	AMX	CF
10	90.57	13.69	18.69
20	92.88	19.15	24.06
30	94.15	22.15	27.13
40	94.75	25.92	30.69
50	95.27	28.23	34.94
60	95.97	32.85	40.5

Fig. 8: Chemical comparisons between the TC, AMX, and CEX

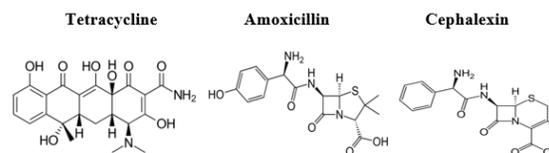
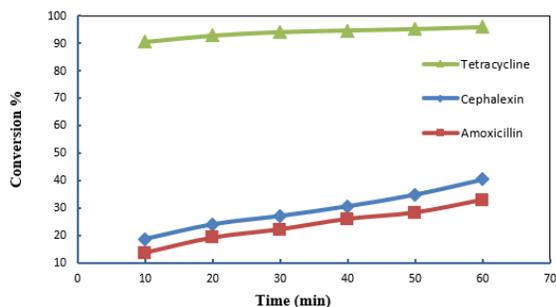


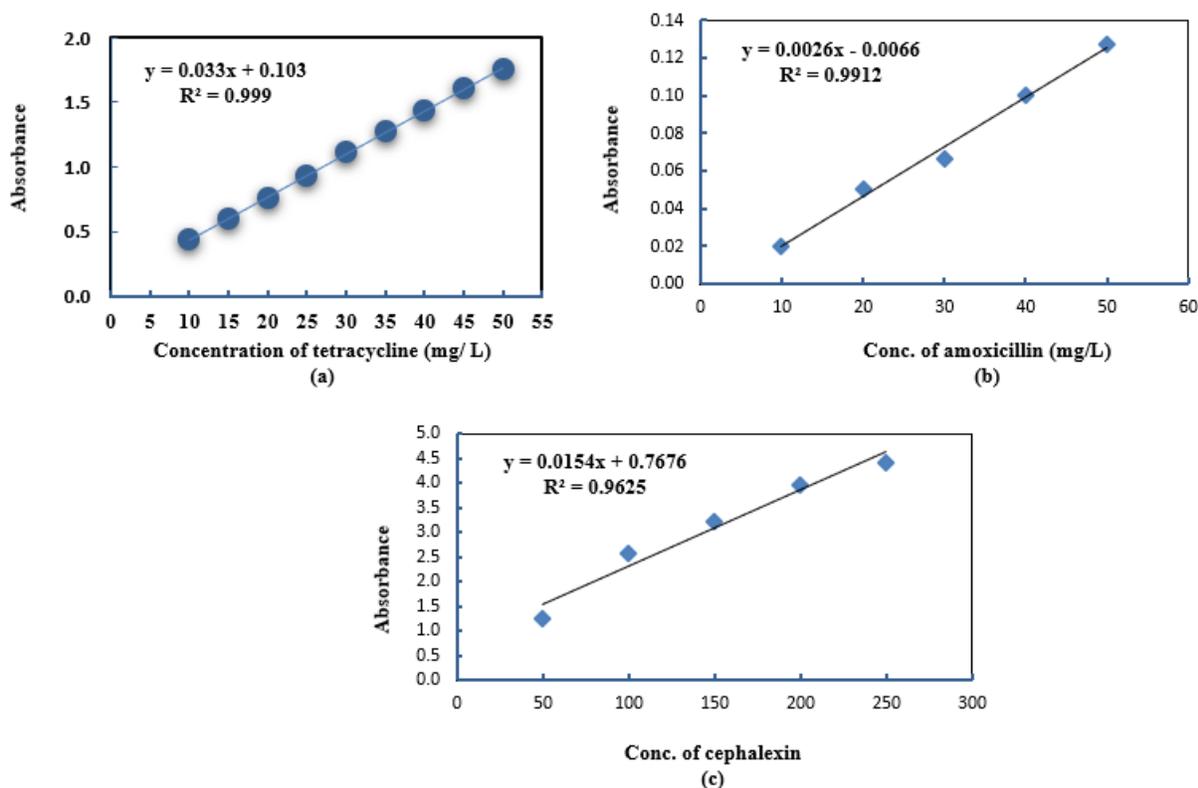
Fig. 9: Tetracycline is removed and oxidized using the electrochemical oxidation method with H₂O₂ and other antibiotics at the following conditions: 5 V voltage, 100 mg/L tetracycline, amoxicillin, and cephalixin concentration, 60 minutes of reaction time, and 10°C.



Calibration curve

A calibration curve for TC, AMX, and CFX was developed using samples of the three antibiotics at different concentrations Figure (10). After determining the absorbance for each concentration, the concentration data was plotted against it. TC, AMX, and CFX are measured at a wavelength of (276, 265, 261) nm, respectively.

Fig. 10: UV-VIS/1800 calibration curve for: (a) rang concentration TC (10-50) mg/L (b) rang concentration AMX (10-50) mg/L (c) rang concentration CFX (50-250)mg/L.



Conclusion

The following are the key conclusions of this study:

1. The findings demonstrated that at 5V, 100 mg/L tetracycline concentration, and 10°C reaction temperature, the maximum

electrochemical oxidation conversion of tetracycline (95.97%) was achieved.

2. Raising the applied voltage increased the electrochemical oxidation of tetracycline, but only to a point; beyond this point, the oxidation of tetracycline decreases.

3. Tetracycline's electrochemical oxidation is decreased by raising the temperature, and tetracycline concentration.

4. Tetracycline's electrochemical oxidation at the ideal temperature was studied kinetically, and the results showed that the system followed pseudo-first-order kinetics, with apparent reaction rate constants k_{app} falling from 0.016 min⁻¹ at 10 °C to 0.01 min⁻¹ at 40 °C.

5. The tetracycline oxidation process at TBER has a negative activation energy (E) of -11.3236 KJ/mol.

6. Based on our findings and comparisons with earlier studies, it was determined that the design we presented here is a successful and efficient design for the on-site oxidation of tetracycline. It is also a good process economically, meaning that it is affordable and environmentally friendly because H₂O₂ is produced locally and used directly in the oxidation process.

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