



Research Article

Cefuroxime oxidation with new generation anodes: Evaluation of parameter effects, kinetics and total intermediate products

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ABSTRACT

In this study, it was investigated the capability of new generation Sb-SnO₂/Ti anodes, which are well known with their promising results in ozone generation and stability, to remove cefuroxim (CXM) antibiotic from aqueous solution. Comparison of different electrolyte types were performed for this purpose; NaCl and KCl. KCl increased the conductivity and caused to the formation of important oxidants and thus, affected electrochemical oxidation reactions more positively than NaCl. It was obtained that, pH parameter has a very important effect on the removal efficiencies in this process and higher efficiencies were obtained at the natural pH value (pH 7) of the aqueous solution. It was thought that, this was probably because the reactions occurred in aqueous solution mostly instead of anodic surface. Furthermore, the removal efficiencies increased with current density increase and the best results were obtained at 50 mA/cm² current density. As a result of the study, at the end of 60 min of reaction, the aqueous solution containing cefuroxime antibiotic was completely treated without any toxic intermediate product formation with 750 mg/L KCl addition, at pH 7 and 50 mA/cm² current density.

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INTRODUCTION

For many years, antibiotics have been used widely in treatment of bacterial infectious diseases, fish farm practices and animal breeding applications [1, 2]. However, these compounds are considered as the most dangerous types of pharmaceutical pollutants because they cause to the microorganism resistance in the environment and, have some toxicological properties. However, bacteria with antibiotic resistance genes can spread easily and thus, threaten environment and human health, seriously [3].

The risk of contamination of them to the receiving environment is high due to the high rates usage of antibiotics and the lack of treatment technologies and insufficiency of the existing technologies. In this regard, advanced oxidation processes are very advantageous and are one of the most common types of treatment processes those are particularly promising. Because they can eliminate non-biodegradable organic compounds and color and reduce organic pollution and toxicity without producing any process waste [4].

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However, human and veterinary applications of antibiotics are much more higher in Turkey than in northern European countries. Consumption of cephalosporin antibiotics were found very high in Turkey According to the International Medical Statistics (2013–2014) [5, 6]. This group of antibiotics are of a broad spectrum semi-synthetic antibiotic series [7, 8]. Cefuroxime (CXM) is the second generation type of antibiotic belonging to the cephalosporins [9].

Cefuroxime was first patented in 1971 and later used for medical treatment applications in 1977 [10]. However, it became the 291st most prescribed drug in the US in 2016 (more than a million prescriptions) [11]. Chemically, it is originate from 7-aminocephalosporanic acid (7ACA) [12]. International Union of Pure and Applied Chemistry (IUPAC) name is: (6R,7R)-3-(carbamoyloxymethyl)-7-[[2-(furan-2-yl)-2-methoxyiminoacetyl]amino]-8-oxo-5-thia-1-azabicyclo[4.2.0]oct-2-ene-2-carboxylic acid. The molecular formula is defined as: $C_{16}H_{16}N_4O_8S$ [13]. The chemical structure of it is given in Figure 1, 2D and 3D.

The occurrence of cephalosporin antibiotics has been reported in wastewater at $\mu\text{g L}^{-1}$ and ng L^{-1} levels [14, 15]. It is known that 66–100% is excreted into the receiving environment with urine without any change [16]. Furthermore, they cannot be treated efficiently with conventional treatment plants [17, 18]. For the removal of these synthetic antibiotic compounds from wastewaters, it is necessary to use advanced treatment processes (AOPs) such as Fenton, UV, activated sludge carbon nanotubes, nanofiltration and electrochemical processes [19]. Recently, most of the researchers have shown great interest to the electrochemical oxidation processes to remove toxic organic compounds from wastewaters [20, 21].

The main reason for studying with Sn/Sb/Ni-Ti anode is its use on electrochemical ozone generation, commonly and intensely. In general, ozone generation is carried out by CCD (cold corona discharge). However, current efficiency is between 2–4% in ozone production by cold corona discharge method [22]. However, 37% current efficiency is possible with Sn/Sb/Ni-Ti to perform ozone production at room temperature [23]. It is seen the ozone reactions occurred on Sn/Sb/Ni-Ti anode at equation 1 and 2. Furthermore, the cathode reaction is stated at eq. 3 for the electrolysis of water for the electrochemical ozone generation [24]. Ozone could be formed on the anode surface and in water by preventing the oxygen formation (electrochemical oxygen formation occurs at lower voltages) [25, 26]. This is only possible with stable anodes such the Sb-doped SnO_2 anodes.

Ozone reactions on anode



H_2 formation on cathode

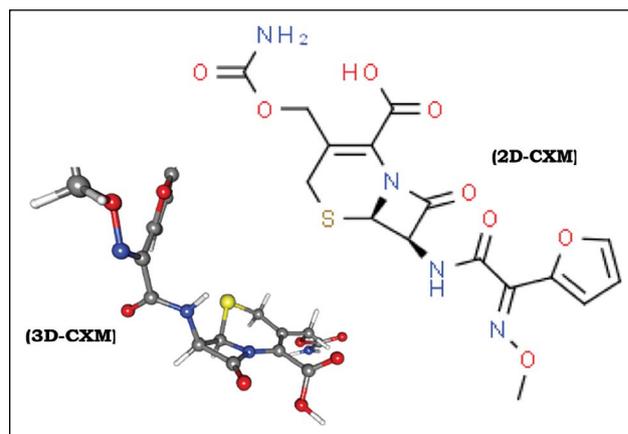


Figure 1. The chemical structure of cefuroxime as 2D and 3D.

Among the factors affecting the electrochemical reactions, anodic material is one of the most important parameter besides, electrolyte, anodic potential and temperature [27]. New anode materials have been investigated include platinum, titanium oxide, graphite, boron-doped diamond, glassy carbon, activated carbon, $\beta\text{-PbO}_2$, IrO_2/Ti etc. [28–30]. However, most of these anode materials are not useful due to having toxicity, instability, and high costs. However, Sb-doped SnO_2 anodes are very useful instead of other type of anodes that having stability, lower costs and non-toxic properties. Furthermore, these type of anodes have shown promising results in electrochemical oxidation processes and ozone production [31–35]. Therefore, it was aimed to investigate the removal of cefuroxime antibiotic from synthetic wastewater with a concentration 50 mg/L CXM with using novel Sb-doped SnO_2 -Ni anodes. Trovo et al. (2011) [36] reported photo Fenton oxidation of amoxicillin with 50 mg/L conc. in synthetic wastewaters. Elmolla and Chaudhuri (2011) experienced UV/ $\text{TiO}_2/\text{H}_2\text{O}_2$ oxidation of amoxicillin and cloxacillin antibiotics with 138 mg/L and 84 mg/L concentrations, respectively in pharmaceutical wastewaters [37].

This study is unique because there is no such a study on electro-oxidation of CFR with these anodes. Most of the studies have focused on fluoroquinolone, trimethoprim, sulfonamide and macrolide group antibiotics for the removal technologies from wastewaters, while, just a little of them have been made for cephalosporin antibiotics [38–41]. However, there are only a few studies about the cefuroxime antibiotic.

MATERIALS AND METHODS

Chemical Agents and Materials

In this study, the removal of cefuroxime (CXM) was investigated by electrochemical oxidation method using new generation Sb- SnO_2 -Ni anodes. Chemicals and materials used in this study are shown in Table 1.

Table 1. List of chemicals and materials used in this study

Chemical agents/materials	Chemical formula	Trade mark
Cefuroxime (CXM)	$C_{16}H_{17}N_3O_4S$	CXM (Sigma-Aldrich)
Titanium mesh	Ti	3Ti7-077FA mesh, Dexmet, USA
Platinized titanium cathode	PtTi	NRK Electrochem DuPont Corp., USA
Antimony (III) oxide	Sb_2O_3	Alfa Aeser Company
Tin (IV) chloride pentahydrate	$SnCl_4 \cdot 5H_2O$	Alfa Aeser Company
Nickel (II) oxide	NiO	Alfa Aeser Company
Potassium chloride	KCl	Alfa Aeser Company
Sodium chloride	NaCl	Alfa Aeser Company
Methanol	CH_3OH	Alfa Aeser Company
Formic acid	CH_2O_2	Emsure
Ethanol	C_2H_5OH	Merck
Hydrochloric acid	HCl	Merck
Sulfuric acid	H_2SO_4	Merck
Oxalic acid	$C_2H_2O_4$	Merck
Ultra pure water	H_2O	Millipore Milli-Q

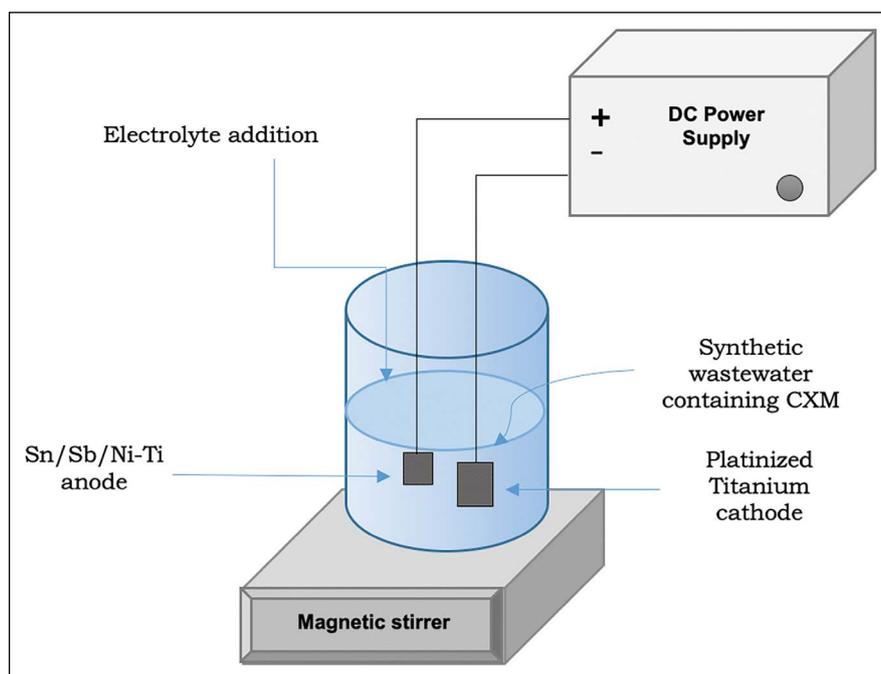


Figure 2. Configuration of the electrochemical reactor.

All of the chemicals were at the purity of $\geq 98\%$ and were used without purification. Aqueous antibiotic solutions were prepared at 20 ± 5 °C. The initial concentration of the CXM was 50 mg/L.

Production of Anodes

Titanium meshes were cut into the dimensions of 2.5 cm x 2.5 cm for the anode preparation. The titanium anode materials were then treated with acetone to remove oil and dirty residues, then they were boiled in 10% oxalic acid ($C_2H_2O_4$) solution for at least 30 min until their color turned to the brown, and then they were left to cool at room tempera-

ture. The cooled titanium meshes were then sonicated for 3 cycles of 15 minutes and dried at room temperature for an hour. Sn/Sb/Ni pyrolysis solution was prepared in the ratio of 500/8/1 according to the prescription of Wang et al. (2005) [42]. Kurt (2020) [43] used Sn/Sb/Ni anodes having 500/8/1 molar rate for anodic oxidation of cefaclor in aqueous solution. The previously prepared titanium meshes were placed into the beaker so that they were all in the solution and the electrodes were started to be coated. One of the most important parameters affecting the tempering process of electrodes is the process temperature. The electrodes were dried in a way that no bubbles were left in the

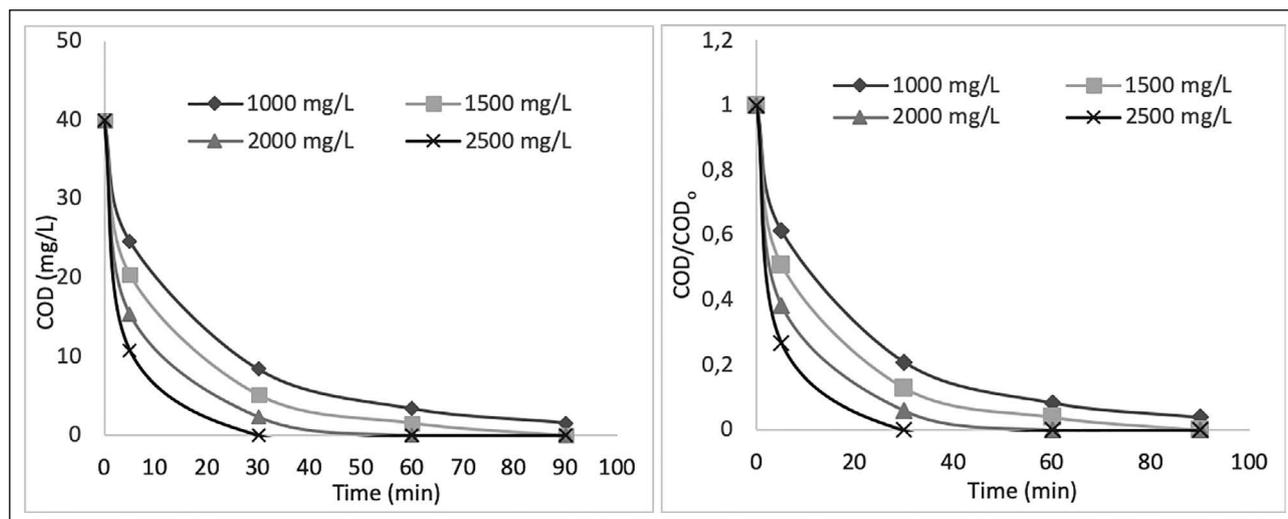


Figure 3. Effect of sodium chloride addition on COD (mg/L) and COD/COD₀ decay (pH 7 and current density: 50 mA/cm²).

gaps of the titanium mesh and kept in an oven heated to 105 °C. In this study, the voltage was kept between 3V and 4V. The distance between the anode and the anode was kept between 1 and 2 cm.

Electrochemical Oxidation Reactor Set-up

In the experiments of electrochemical oxidation processes, a mechanism was created where the anode and cathode are mounted with beakers containing cefuroxime aqueous solution. The anode and cathode were connected to the current supply, Exttech, US, DC power supply (Fig. 2).

Analytical Procedure and Equipment

pH values of the samples were measured by a pH meter (Cyberscan, UK). COD measurements were made according to the APHA (2005) Standard Methods [44]. Total organic carbon (TOC) analysis were performed by a TOC analyzer (TOC-L, Shimadzu, Kyoto, Japan). The residual CXM was determined by Photodiode Detector (PDA) and Ultra Performance Liquid Chromatography (UPLC) (Thermo-Scientific, Massachusetts, USA). 254 and 270 nm wavelengths were selected for the working range of detector. Properties of column used for UPLC are: Hypersil GOLD, C-18 (50 x 2.1 mm; 1.9 μm) (Thermo-scientific, Massachusetts, USA). The column temperature was 35 °C. The mobile phase solution was prepared with water containing 0.1% formic acid and methanol, [MeOH:H₂O]: 40:60 (v/v)]. The analytical process was carried out at a flow rate of 0.2 mL/min. All of the measurements were performed in triplicate.

RESULTS AND DISCUSSION

Effect of Sodium Chloride Addition

The results of this research has shown that salt addition increases conductivity and affects the electrochemical oxidation process positively. However, extra salt addition may

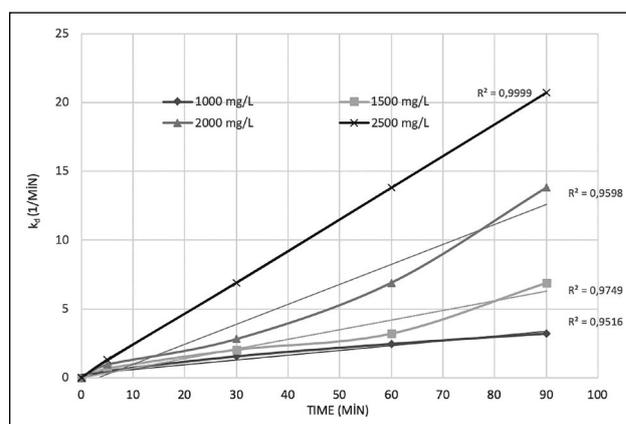


Figure 4. Variation of k_d values with NaCl dose (pH 7 and current density: 50 mA/cm²).

Table 2. Relationship of NaCl doses with first order kinetic values (pH 7, current density: 50 mA/cm² and 60 min reaction time)

NaCl doses (mg/L)	kd (1/min)	R ²
1000	2.4651	0.9516
1500	3.2189	0.9749
2000	11.5129	0.9598
2500	13.8155	0.9999

cause to the environmental problems and increase costs at the same time [45, 46]. NaCl was added to the samples at a concentration of 1000 mg/L, 1500 mg/L, 2000 mg/L and 2500 mg/L, and the electrochemical oxidations were performed at natural pH values of the solution (pH 7) and at a current density of 50 mA/cm². As a result, it was observed that with the increase of salt addition, COD removal efficiencies increased (Fig. 3). However, COD/COD₀ values decreased in parallel with COD values.

However, the amount of NaCl was much higher than KCl needed to removal of COD, thus, there was no need to con-

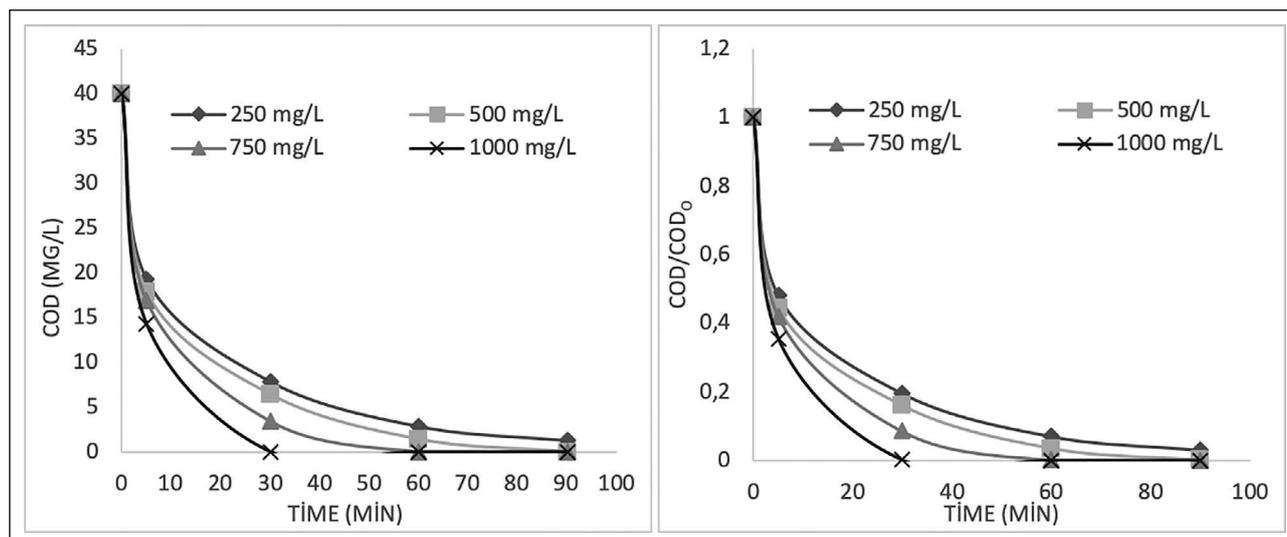


Figure 5. Effect of potassium chloride addition on COD (mg/L) and COD/COD₀ decay (pH 7 and current density: 50 mA/cm²).

tinue to study with this salt type. And thus, there was no need to determine NaCl effect on cefuroxime degradation. For this reason, COD parameter chosen as the main parameter in this study, because it is a clearer and more traditional parameter.

In the experiment performed with addition of 2000 mg/L NaCl, COD value decreased to the 2,4 mg/L at the end of 30 min, and it was completely consumed at the end of 60 min (Fig. 3) at the solution’s natural pH value (pH 7) of the solution and 50 mA/cm² current density. In Table 2, it was shown the relationship of different NaCl doses with first order kinetics for the electrochemical oxidation of CXM. In Figure 4, it was given k_d values depending on COD variation with NaCl doses (pH 7 and current density: 50 mA/cm²).

According to the results, it was seen that the most efficient electrochemical oxidation processes was found with addition of 2000 mg/L and 2500 mg/L NaCl.

Effect of Potassium Chloride Addition

KCl was added to the samples at a concentration of 250 mg/L, 500 mg/L, 750 mg/L and 1000 mg/L. The electrochemical oxidations were performed at natural pH values of the solution (pH 7) and at a current density of 50 mA/cm². According to the Figure 5 it was seen that the COD value decreased to the 3,4 mg/L just after 30 min reaction and reached to the zero after 60 min with 750 mg/L KCl addition. However, it reached to the zero after 30 min with 1000 mg/L KCl. Although a better efficiency was possible with excessive amounts of the electrolyte, the use of excess chemicals may increase the savings. Thus, the optimum amount of KCl was found to be 750 mg/L. As a result, it was observed that with the increase of salt addition, COD removal efficiencies increased, and at the same time COD/COD₀ values decreased in parallel with COD values (Fig. 5). With the increase of salt amount, resistance of

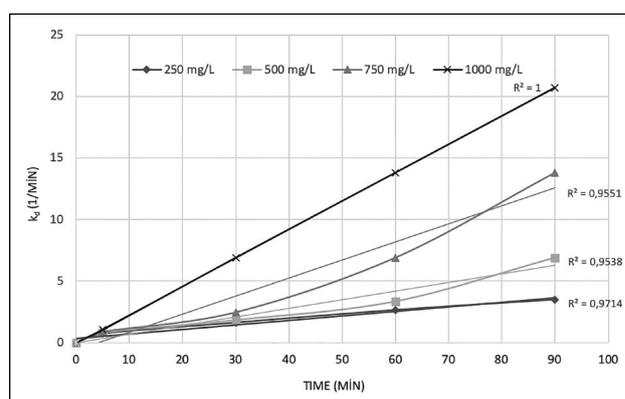


Figure 6. Variation of k_d values with KCl dose (pH 7 and current density: 50 mA/cm²).

Table 3. Relationship of KCl doses with first order kinetic values (pH 7, current density: 50 mA/cm² and 60 min reaction time)

NaCl doses (mg/L)	kd (1/min)	R ²
250	2.6593	0.9714
500	3.3524	0.9538
750	6.9078	0.9551
1000	13.8155	1

the solution decreases. As a result of resistance decrease higher potential difference occurred on the electrodes and the antibiotic compounds were degraded faster [47]. Furthermore, hypochloric acid and chlorine gas increase with increase of salt [46].

In Table 3, it was shown the relationship of different KCl doses with first order kinetic values for the electrochemical oxidation of CXM. In Figure 6, it was given k_d values depending on COD (assumed as major parameter) variation with KCl dose (pH 7 and current density: 50 mA/cm²). According to the k_d values stated in Figure 6, it was seen that KCl addition

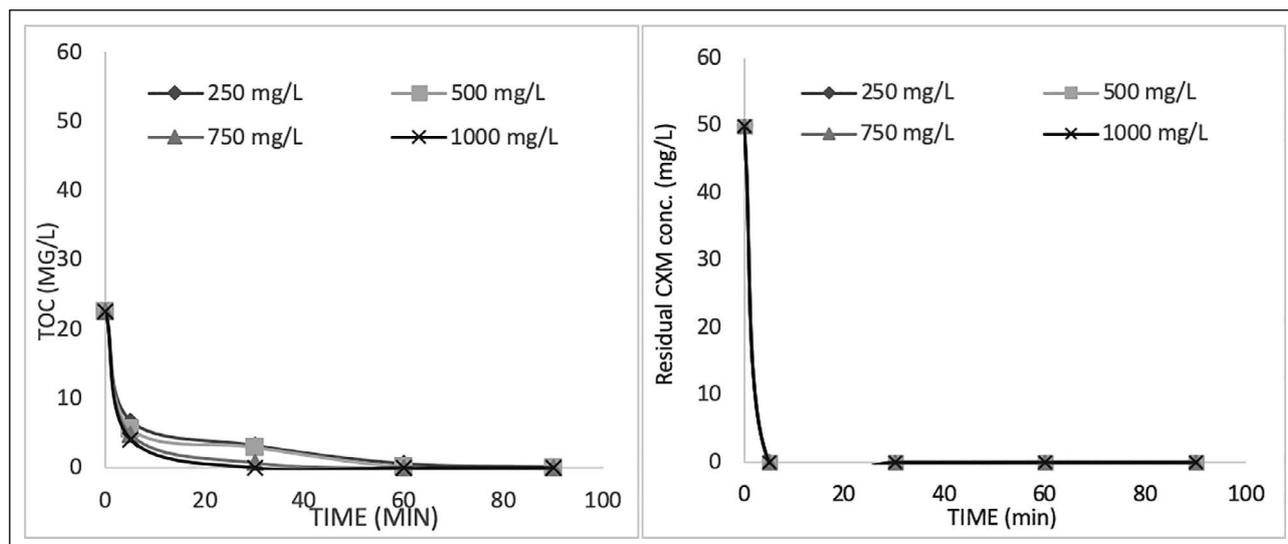


Figure 7. Effect of potassium chloride addition on TOC (mg/L) and residual CXM conc. (mg/L) decay (pH 7 and current density: 50 mA/cm²).

affected the electrochemical oxidation processes significantly, due to increase of conductivity and thus, hypochlorid acid. According to the results, it was seen that the most efficient CXM oxidation was carried out with 750 mg/L and 1000 mg/L KCl addition, at pH 7 and 50 mA/cm² current density.

As it is seen in Figure 7, TOC decreased to at the end of 30 min to 0,78 mg/L with the addition of 750 mg/L KCl, and it was completely consumed at the end of 60 min. According to these results, it was accepted that the most efficient treatment was carried out in 60 min with 750 mg/L KCl, at pH 7 (natural pH value) and 50 mA/cm² current density (Fig. 7). Tu et al. (2015) [48], compared the removal efficiencies for the removal of antibiotic active substance with NaCl and Na₂SO₄ and it was obtained much better results with NaCl. However, in this study, it was observed that the addition of KCl increased conductivity and caused to the formation of important oxidants such as chloride gas and hypochloric acid, affecting electrochemical oxidations more positively than NaCl addition [46].

pH Optimization

The pH parameter plays an important role in electrochemical oxidation processes. However, it was observed that, in electrochemical oxidation processes, the reactions are affected positively sometimes at acidic and sometimes at alkaline conditions [49]. Formation of ozone, hydroxyl radicals and chlorine gas bounded to the electrolyte and related oxidants may cause to this result, while the formation of hydrogen peroxide and related oxidation reactions may occur as a result of cathodic reactions at basic pH values [49].

It was carried out in the pH range of 3–9. In the experiment performed at neutral pH value, 7 with 750 mg/L KCl salt addition, COD value decreased to 3.4 mg/L at the end of

30 min, and was consumed completely at the end of 60 min (Fig. 8). COD/COD₀ values decreased in parallel with COD values (Fig. 8).

In Table 4, it was shown the relationship of different pH values with first order kinetic values for the electrochemical oxidation of CXM. In Figure 9, it was given the k_d values depending on COD variation with pH variation. According to the results, it was seen that the most efficient CXM oxidation was obtained at natural pH value, 7 of the solution (KCl conc.: 750 mg/L and current density: 50 mA/cm²).

It has been observed that pH has a very important effect on the removal efficiency of the process and higher efficiencies were obtained at the natural pH (pH 7) of the solution with addition of 750 mg/L KCl and at 50 mA/cm² current density (Fig. 8–10). This is probably because the reactions occurred in the solution mostly instead of anodic surface.

Wang et al. (2016) [50] investigated the removal of ciprofloxacin with a Sb-doped SnO₂/Ti anode and they saw that, the removal rates were higher at higher pH values. Sivrioğlu and Yonar (2016) studied treatment of textile wastewater with Sn/Sb/Ni anode and they obtained that, the COD and colour removals were found to be 98% and 99%, respectively at pH 3. Although pH 7.2 showed relatively lower efficiency compared to acidic conditions, pH 7.2 was chosen as the optimum value to avoid extra pH adjustment step and cost. Kurt (2020) [43] investigated the electrochemical oxidation of cefaclor with Sn/Sb/Ni anode, and pH 7 was obtained as the best. Jojoa-Sierra et al. (2017) [51] reported the electro-oxidation of norfloxacin with Ti/IrO₂ anode at different pH values and the removal efficiencies of the process followed the pH values of 9.0>7.5>6.5>3.0. At alkaline conditions, the potential of chlorine gas and hypochloride ion formation could sup-

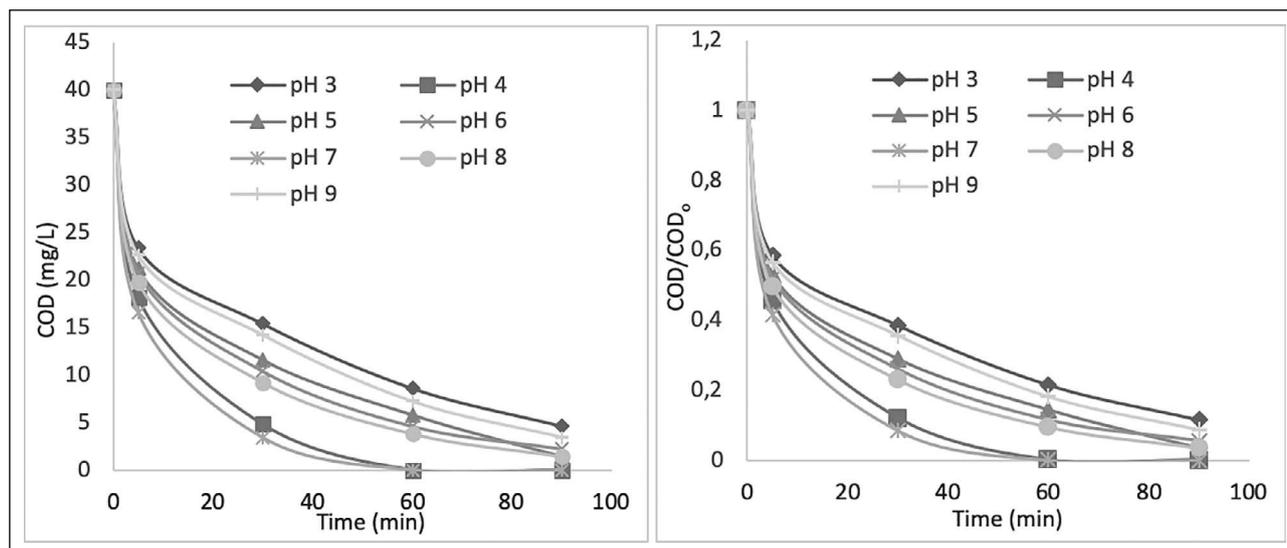


Figure 8. Effect of pH variation on COD (mg/L) and COD/COD₀ decay (KCl conc.: 750 mg/L and current density: 50 mA/cm²).

port the removal efficiencies [52, 53]. However, according to some of the researchers, acidic pH values could be efficient. Under acidic conditions, chlorine gas is exposed that is able to generate HOCl and elimination of ·OH scavengers could be possible at acidic conditions.

Determination of Optimum Current Density

Current density is the another important parameter for electrochemical processes because it has have an active role in reaction kinetics [53]. It is one of the factors affecting decomposition rate positively as well as the operating cost. However, it may affect the strength of the anode and cathode negatively in case of presence very high values. In our study, it was observed that the anodes with a current density of 75–100 mA/cm² could not withstand high current levels and they were broken. Thus, it was studied between 10–50 mA/cm² current densities.

In a study of Sivrioğlu and Yonar (2016), it was obtained that the current density strongly affected COD and color removal with Sn/Sb/Ni anodes, but it was observed that increasing of the current density caused to the energy loss. While it was expected high efficiency at high current values, the durability of the anodes were decreased [54].

Figure 11 shows the effect of current density on COD and COD/COD₀ parameters at the optimum pH value and KCl concentration. According to the Figure 11. COD/COD₀ values decreased in parallel with COD values. In the experiment performed at neutral pH value with 750 mg/L KCl addition, COD value was consumed completely at the end of 60 min.

In Figure 12 and Table 5 it was given the kd values depending on current density variation. According to the results, it was seen that the most efficient CXM oxidation was found at 50 mA/cm² current density (pH 7 and KCl conc.: 750 mg/L).

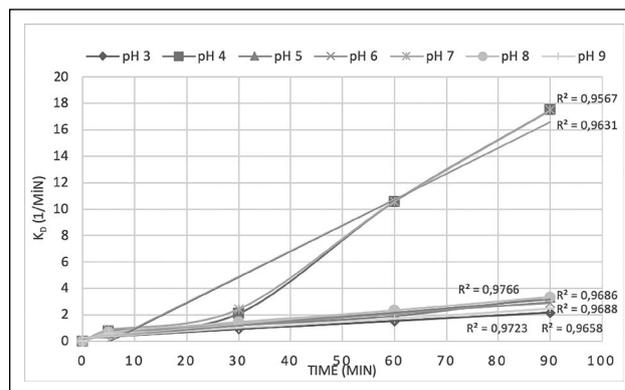


Figure 9. Variation of k_d values with pH variation (KCl conc.: 750 mg/L and current density: 50 mA/cm²).

Table 4. Relationship of pH variation with first order kinetic values (KCl conc.: 750 mg/L and current density: 50 mA/cm² and 60 min reaction time)

NaCl doses (mg/L)	kd (1/min)	R ²
3	1.5371	0.9658
4	10.5966	0.9567
5	1.9310	0.9686
6	2.1628	0.9688
7	10.5966	0.9631
8	2.3538	0.9766
9	1.7148	0.9723

Figure 13 shows the effect of current density on TOC and residual CXM conc. at the optimum pH and KCl concentration. According to the graphs in Figure 13 it was observed that the removal efficiencies increased with current density increase and the most efficient results were obtained at 50 mA/cm² current density.

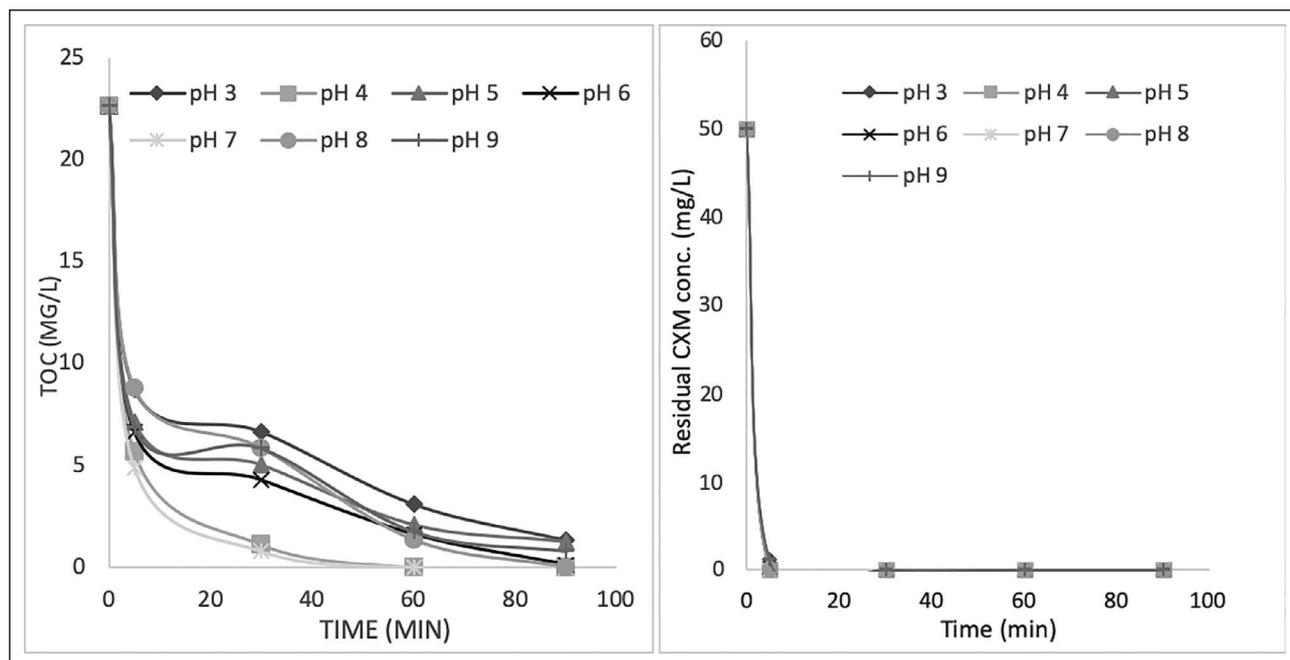


Figure 10. Effect of pH variation on TOC (mg/L) and residual CXM conc. (mg/L) decay (KCl conc.: 750 mg/L and current density: 50 mA/cm²).

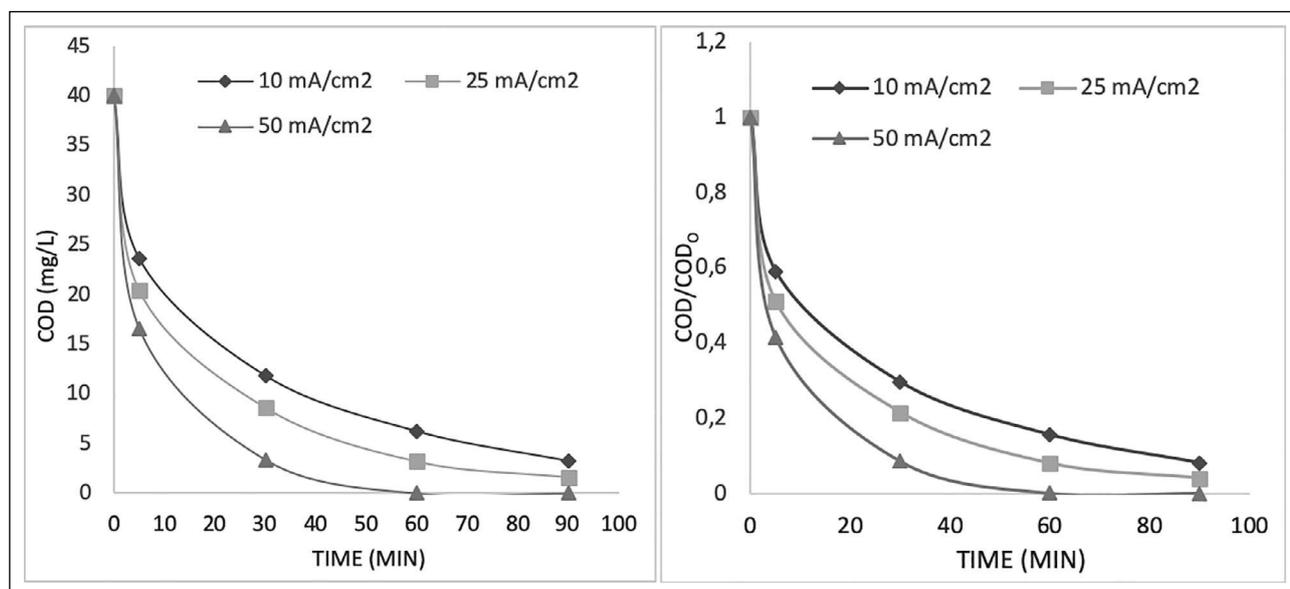


Figure 11. Effect of current density variation on COD (mg/L) and COD/COD₀ decay (pH 7 and KCl conc.: 750 mg/L).

Wang et al. (2005) investigated electrochemical ozone production at Ni-Sb-doped SnO₂ anode in acidic aqueous solution at room temperature with a current efficiency up to 35%, after a short time, it was reported current efficiencies up to 50 % confirmed by Christensen et al. (2009) [34, 42]. Researches continue to try and find more active and efficient anodes generating ozone that can be operate at room temperature with low cell voltages; therefore, Wang et al. (2005) proved the studies at Hong Kong University are newsworthy [42].

Total Intermediate Products Evaluation

The use of KCl or NaCl as the additional electrolyte may cause to the occurrence of chlorinated intermediates, as mentioned by Sirés et al. (2014) [55]. At this study, HPLC chromatograms of the samples taken during electrolysis showed the gradual disappearance of CXM antibiotic, and the formation of a series of organic/aromatic intermediates with chromatogram peaks formed at different retention times. At this way, it was calculated the total intermediate products from the peaks formed at different retention times.

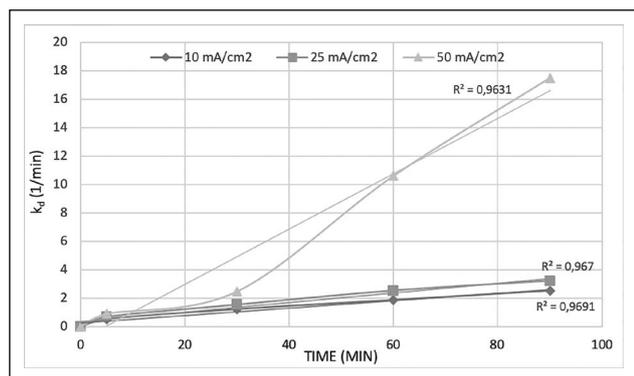


Figure 12. Variation of k_d values with current density parameter (pH 7 and KCl conc.: 750 mg/L).

According to the Figure 14, toxic intermediate formation ended at the end of 30 min reaction time with the addition of 1000 mg/L KCl and at the end of 60 min with the addition of 750 mg/L KCl at pH 7 and 50 mA/cm² current density. However, while the formation of organic/aromatic intermediates ended just after 60 min of electrochemical reaction time at pH 4 and pH 7, it continued throughout the reaction at pH 3, pH 8 and pH 9. It was thought that increase of the formation of •OH radicals in water under the acidic and alkaline conditions may lead to the increase of toxic organic intermediates formation. Low current densities (10 and 25 mA/cm²) resulted in incomplete oxidation of organic compounds and the formation of new intermediates. As a result, at the end of 60 min reaction time synthetic wastewater containing cefuroxime antibiotic was completely treated without any toxic intermediate product formation with the addition of 750 mg/L KCl, at pH 7 and 50 mA/cm² current density.

Table 5. The relationship of current density variation with first order kinetic values (pH 7, KCl conc.: 750 mg/L and 60 min reaction time)

Current density (mA/cm ²)	kd (1/min)	R ²
10	1.8643	0.9691
25	2.5257	0.9670
50	10.5966	0.9631

Duan et al. (2020) [56] reported electro-oxidation of ceftazidime antibiotic in real municipal wastewaters with PbO₂-Ce and SnO₂-Sb anodes. While 99.37% of ceftazidime degradation and 95.52% COD removal was achieved with Ti/SnO₂-Sb anode, 75.15% ceftazidime degradation and 83.54% COD removal was obtained with Ti/PbO₂-Ce anode, under 4 mA cm⁻¹ current. Yahya et al. (2016) [57] investigated the ability of Electro-Fenton process with carbon-felt cathode and Pt anode for degradation and mineralization of levofloxacin (LEV) in aqueous solution. The absolute rate constant was found to be $(2.48 \pm 0.18) \times 10^9 \text{ M}^{-1} \text{ s}^{-1}$. 400 mA current value and 0.1 mM catalyst Fe²⁺ loading were observed to be optimum. Chemical oxygen demand and mineralization degree was reached to >91% at the end of 6 h. A number of intermediate products were identified using HPLC and LC-MS. N atoms in LEV were released as NH⁴⁺ and NO⁻³ ions. Nitrogen atoms mainly transformed into NH⁴⁺ rather than in NO⁻³. The concentration of NH⁴⁺ reached 0.28 mM at 300 min, while that of NO⁻³ reached to the zero at 300 min. The nitrogen loss could be explained by the formation of volatile nitrogen compounds and the presence of oxamic acid that is hardly oxidizable by hydroxyl radicals.

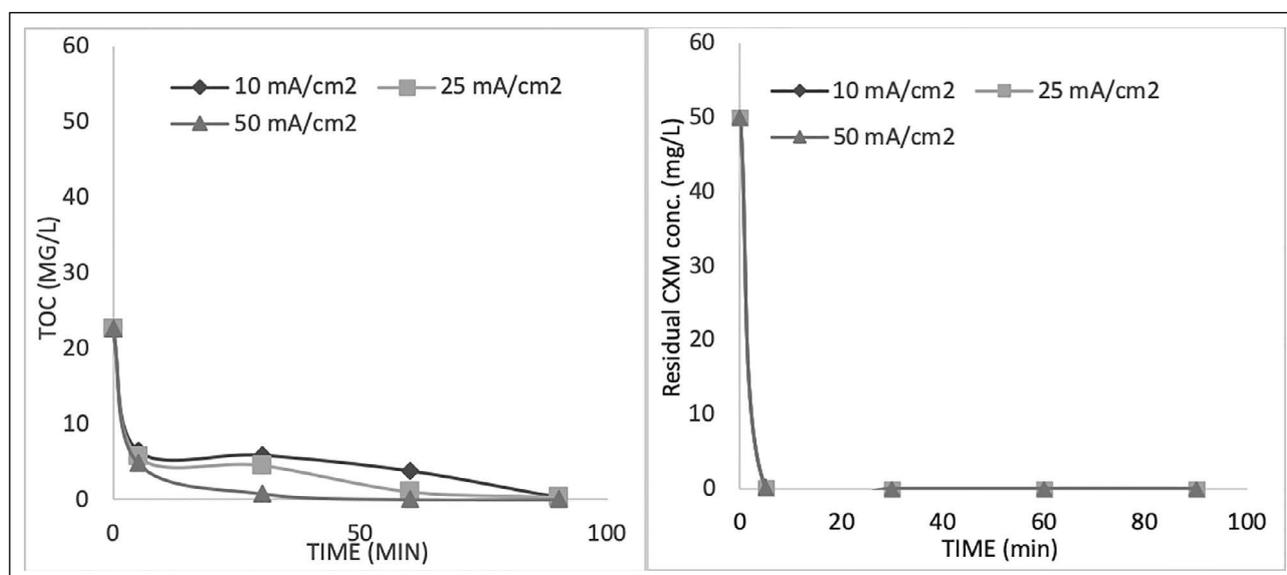


Figure 13. Effect of current density variation on TOC (mg/L) and residual CXM conc. (mg/L) decay (pH 7 and KCl conc.: 750 mg/L).

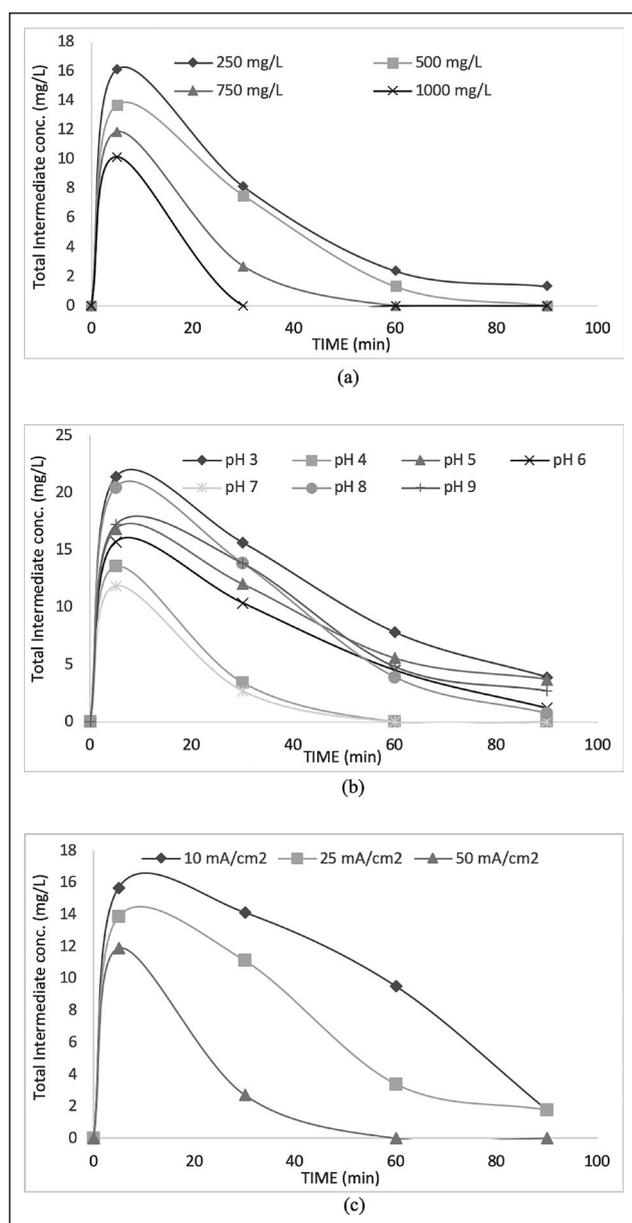


Figure 14. Occurring of total intermediate products in electrochemical oxidation processes with Sb-SnO₂-Ni anodes (a) KCl effect (b) pH effect (c) current density effect (pH 7 and KCl conc.: 750 mg/L).

CONCLUSIONS

In this study, it was investigated the removal of cefuroxim (CXM) from aqueous solution with novel Sb-SnO₂/Ti anodes. Comparison of different electrolyte types were made (NaCl and KCl). KCl increased the conductivity and caused to formation of important oxidants such as chloride gas and hypochlorite acid and affected the reactions more positively than NaCl addition. The optimum results were observed with 750 mg/L KCl addition. KCl was found as the optimum electrolyte type affecting the electrochemical reactions positively the most even in lower concentrations. Thus, it could

be possible to obtain higher removal efficiencies with real wastewaters assuming they include Cl ions highly, without addition extra chemicals. pH has a very important effect on the removal efficiencies and pH 7 was considered as the optimum, which is the natural pH value of the solution. This was probably because the electrochemical oxidation reactions occurred in the aqueous solution mostly instead of on the anodic surface. As a result of the study, it could be possible to operate process easier and more economically by working at neutral pH values, due to there is no need to chemical addition and extra cost. However, the removal efficiencies increased with current density increase and the optimum results were obtained at 50 mA/cm² current density. Because active oxidants occurred increasingly at higher values.

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DATA AVAILABILITY STATEMENT

The author confirm that the data that supports the findings of this study are available within the article. Raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

CONFLICT OF INTEREST

The author declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

ETHICS

There are no ethical issues with the publication of this manuscript.

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