

Application of photo-electro oxidation process for amoxicillin removal from aqueous solution: Modeling and toxicity evaluation

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Abstract—The recent increase in the global consumption of antibiotics has led to faster entry of these pollutants into the environment as well as an increase in public concern about its impact on ecosystem and human health. Generally, due to high toxicity of antibiotics, biological methods are not used to treat these pollutants; therefore, advanced oxidation processes are recommended to treat and reduce the toxicity of the wastewater. In this study, we evaluated the efficacy of photo-oxidation (P) and electro-oxidation (E) processes in the removal of amoxicillin (AMX) from wastewater, either as integrated or separate processes. Moreover, the effect of variables, including current density (2-100 mA/cm²), reaction time (2-120 min), and electrolyte concentration (100-1,000 mg/l) on antibiotic removal efficiency were investigated by Box Behnken design under response surface methodology, and optimal conditions were determined for pollutant removal. Then, the effect of AMX concentration and pH variables on the removal efficiency was investigated. The COD removal efficiency was also evaluated under optimal conditions, and eventually the toxicity and bioavailability of the effluent from the combined Photo-Electro oxidation process (PE) were examined. The optimal conditions for variables, including current density, reaction time, and electrolyte concentration for removal efficiency of 62.4%, were 94 mA/cm², 95 min and 997 mg/l, respectively. Investigating the Amoxicillin and pH variables showed that by reducing the contaminant concentration and pH, the antibiotic removal efficiency increased. The toxicity and bioavailability of the final effluent show the reduction of both parameters in the PE reactor effluent. The PE process can provide an appropriate function to reduce the toxicity and antibacterial properties of effluent by removing more than 60% of amoxicillin and 30% of COD from wastewater.

Keywords: Amoxicillin Removal, Box Behnken Design, Modeling, Photoelectrooxidation, Toxicity Evaluation

INTRODUCTION

Humans, during daily activities, discharge a large amount of harmful artificial compounds and chemicals into the environment, which can enter water bodies and threaten health. Treatment of wastewater containing chemical compounds is one of the strategies to reduce these risks as well as environmental pollution [1].

Over the past 15 years, drugs have been highly regarded as potential bioactive chemicals in the environment. These compounds, due to their high persistency, affect the quality of drinking water supplies and can have a long-term impact on human and animal welfare as a potential hazard to the ecosystem [2]. Among drugs, antibiotics are much consumed, which between 2000 and 2010, global

consumption of antibiotics increased by 36% [3]. Annually, 100,000 to 200,000 tons of antibiotics are consumed worldwide; therefore, large amounts of antibiotics in different ways and continuously enter into the aquatic and terrestrial environment [3,4].

Among the various classes of antibiotics, the beta-lactam class is the most consumed worldwide [3]. AMX is a type of semi-synthetic antibiotic that is one of the most widely consumed antibiotics in humans and animals [5]; moreover, the presence of a beta-lactam ring in its chemical structure prevents bacterial cell wall synthesis and causes bacteria to destroy [6,7]. In 2010, the EU listed the maximum standard amount of AMX in food with animal origin at 50 µg/kg [8]. About 60% of AMX consumed by humans is excreted without metabolism through the urine [9]. Discharging wastewater containing antibiotics into the environment causes the deaths of aquatic organisms, including fish [10] and algae [5], and creates drug resistance microorganisms [11].

Biological methods are not suitable for treatment of wastewater

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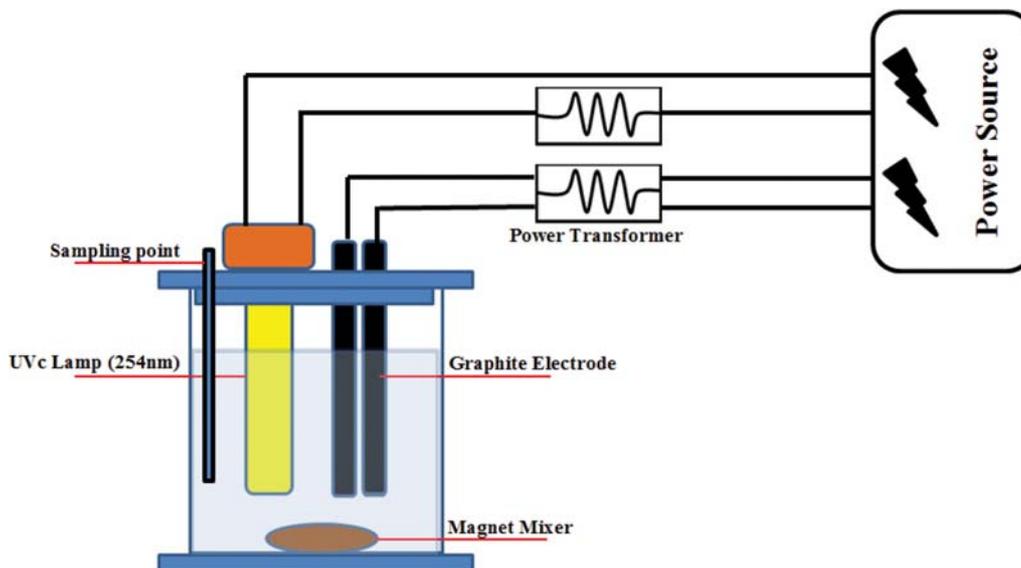


Fig. 1. Schematic of the batch reactor used in the study.

containing these materials due to their sensitivity to toxic and emergent organic contaminants [12]. Recently, several treatment methods have been developed based on advanced oxidation processes (AOPs) to remove toxic compounds and organic pollutants that are resistant to degradation. These processes are capable of producing OH° radical species that can oxidize resistant organic compounds [13-15]. The produced OH° in the AOPs can non-selectively react, oxidize, and mineralize any persistent organic pollutants [1,16,17]. Many studies have used electrochemical processes as a simple, economical, safe, and environmentally compatible technology for the removal of various pollutants from aquatic environments [18-20].

Most of the electrochemical processes used to treat wastewater include two electrocoagulation and electrolysis processes (electro-oxidation) [21-23]. The electrocoagulation process is based on the coagulant production due to the reduction of the electrode in anode, and the produced coagulant reacts with the pollutant and deposits [24,25]. In electrolysis, anodic oxidation process degrades and removes the contaminants from the solution; moreover, while destructing the contaminants, the amount of produced sludge is much lower compared to the electrocoagulation process. The electrochemical process based on electrolysis can be combined with other AOPs to increase the amount of pollutant removal efficiency [26]. Many studies have investigated the use of photo-electrolysis process [25,26], processes based on Fenton [27] and photocatalysts [16,27,28], the interaction effect of irradiation using UV rays on chemical and electrochemical processes. The photo-oxidation process is one of the advanced oxidation processes; and if it is combined with electro-oxidation, it will increase the potential of the products in removing pollutants due to the production of free radicals from the products of the electro-oxidation process [15].

The present study investigated the effectiveness of E and P processes separately and in a combined way to remove AMX antibiotic from aqueous environments.

Also, the effect of different operational parameters and their interaction on AMX removal was investigated, and an appropriate sta-

tistical model was suggested for AMX removal by PE process. Toxicity and bioavailability of final effluent were also examined.

MATERIALS AND METHODS

1. The Reactor

The reactor used in this study is a batch reactor with a volume of 300 ml. Inside the reactor, a 5-watt 45-volt UVC lamp (Philips, Poland) with a wavelength of 254 nm, and a length of 10 cm, was placed, as well as two graphite rod electrodes with a length of 10 cm and a diameter of 0.8 cm with a distance of 1 cm from each other. The input current density to the reactor was controlled by a DC power supply (DAZHENG PS-305D, China). The solution was mixed by a magnet at a constant speed of 150 rpm. An outlet was used to sample the reactor content at the specified times. Fig. 1 represents the schematic of the reactor.

2. Chemical and Reagents

Pure AMX (CAS Number: 26787-78-0) obtained from Sigma Aldrich Co. (St. Louis, MO, USA) was used to prepare the synthetic wastewater. A stock solution (1,000 mg/l) was prepared and applied for experiments. A pre-determined dose of NaCl (from Merck GmbH Darmstadt, Germany) was added to the solution as electrolyte. To adjust the pH, HCl and NaOH 0.1 N were used. To determine the toxicity and bioavailability of the final effluent, *Staphylococcus aureus* PTCC 1112 bacteria and *Saccharomyces cerevisiae* PTCC 5052 yeast, prepared from the collection center of industrial microorganisms (Iran), were respectively used.

3. HPLC-UV Analysis

To measure the amount of remaining AMX the HPLC (YL9100-Technolab system ©) equipped with YL9110 quaternary pump, YL9160PDA detector, wavelength 235 nm with 1 ml/min flow rate, high-purity grade methanol (MeOH) (from Merck GmbH (Darmstadt, Germany), and distilled HPLC grid (Sigma Aldrich Co. (St. Louis, MO, USA)) with a ratio of 75 to 25 were used for methanol and distilled water, respectively. The analytical column (Brisa LC2

Table 1. Variables and levels considered based on Box Behnken design

Variable	Level		
	-1	0	1
Electrolyte concentration (mg/l)	100	550	1000
Current density (mA/cm ²)	2	51	100
Reaction time (min)	2	61	120

C18, 4.6×250 mm, 5 μm (© Teknokroma Análítica, SA)) was used to determine the remaining AMX concentration in the solution (retention time 5 min, temperature 25 °C).

4. Design of Experiments

This study was conducted in two stages. In the first stage, the optimal conditions were evaluated for the three variables: electrolyte concentration, current density, and reaction time. To determine the value of these variables, Design Expert Software (version 10), Box Behnken design under response surface methodology was used. By determining the ranges of each variable, three levels were considered and a total of 17 runs were performed. Each run was repeated three times and the mean was reported. The variables and their levels are presented in Table 1. At this stage, the concentration of AMX was fixed as 50 mg/l. Response surface regression was used to analyze the experimental results and the optimal conditions for removing AMX by the polynomial second order model were obtained according to Eq. (1) [29].

$$Y = \beta_0 + \sum_{i=1}^k \beta_i X_i + \sum_{i=1}^k \beta_{ii} X_i^2 + \sum_{i=1}^k \sum_{j=1}^k \beta_{ij} X_i X_j + \varepsilon \quad (1)$$

where Y is the AMX removal efficiency (%), β_0 is the fixed coefficient of the model, X_i and X_j are the coded variables, β_0 , β_{ii} , and β_{ij} are linear, quadratic, and interaction coefficients, respectively.

In the first stage, the optimal conditions for electrolyte concentration, current density, and reaction time in each reactor were separately determined. The second stage was conducted to evaluate the effect of AMX concentration and pH on the efficiency of processes. At this stage, and in optimal conditions for three variables, the AMX synthetic samples were fed into the reactor with four different concentrations of 10, 30, 70, and 100 mg/l and three-times repetition with fixed pH. Then, the tests were conducted with fixed concentration at pH 2, 4, 8, and 12 with three-times repetition.

5. COD Measurement

The capability of the reactor to remove COD was also examined. The COD test was carried out using the closed reflux colorimetric method according to the standard instruction of the method No. 5220D [30]. The removal efficiency of COD in neutral pH conditions (pH=7), reaction time 10, 30, 60, and 90 minutes, and the optimum conditions obtained for electrolyte concentration and current density were investigated at the first stage in the PE reactor and compared with the amount of AMX removal. At this stage, in order to investigate the capability of combined PE reactor compared to the reactor with the E process alone and P process alone (current density 94 mA/cm², electrolyte concentration 997 mg/l, AMX concentration 10 mg/l, and pH 7), the reactor was used once only by the E process and once only by the P process in the above mentioned conditions, and the results were compared with each other.

6. Investigating Antimicrobial and Toxicity Properties

The antimicrobial property of the effluent from the reactor under optimal conditions was investigated with the inhibition zone method using *Staphylococcus aureus* ATCC 6538 as an indicator microorganism and based on the study of Serna-Galvis et al. [31].

S. aureus is used as an indicator microorganism in the antimicrobial investigation test, since this microorganism is an important human pathogen with high stability against antibiotics [32].

The toxicity of effluent was examined using *Saccharomyces cerevisiae* yeast, which is a eukaryotic organism. According to previous studies, this yeast is a suitable indicator for the presence of hazardous compounds in aquatic environments [33]. To investigate the toxicity, a suspension of *S. cerevisiae* was prepared using 0.005 g of granular yeast and 0.5 ml of distilled water, then 100 μl of yeast suspension was added to 900 μl of the sample. The existing culture medium was approximately 1.6×10⁶ cells per ml. The control sample in this test included a solution containing 100 μl of yeast suspension and 900 μl of distilled water free of antibiotic. The sample was contacted with the yeast suspension for an hour, and then 50 μl of the sample with 50 μl of methylene blue (0.01% m/v, pH 4.6) were combined and deposited on a Neubauer chamber and examined under a microscope (LEICACME). The colorless live cells and colored dead cells were counted in 64 houses of the Neubauer chamber. Accordingly, the toxicity (death of *S. cerevisiae*) is expressed by the percentage of dead cells (100×total cells/dead cells). Moreover, the number of dead cells in the control sample should be included in the calculation and the percentage of toxicity should be reduced in the final report. Note that the toxicity of the compounds remaining from the electrolysis process, such as chlorine compounds, was neutralized by adding sodium sulfite.

Table 2. Removal efficiency based on Box Behnken design for the three variables of current density, reaction time, and electrolyte concentration

Run number	Current density (mA/cm ²)	Reaction time (min)	Electrolyte concentration (mg/l)	Removal (%)
1	100	61	100	5.5
2	60	2	100	1.54
3	60	61	550	40
4	20	61	100	2.5
5	100	2	550	8.5
6	60	61	550	32
7	100	61	1000	61
8	20	2	550	2.5
9	60	61	550	34.5
10	60	61	550	31.5
11	100	120	550	27.5
12	20	120	550	13.5
13	60	120	100	7
14	20	61	1000	41
15	60	120	1000	52.5
16	60	2	1000	9.5
17	60	61	550	33.8

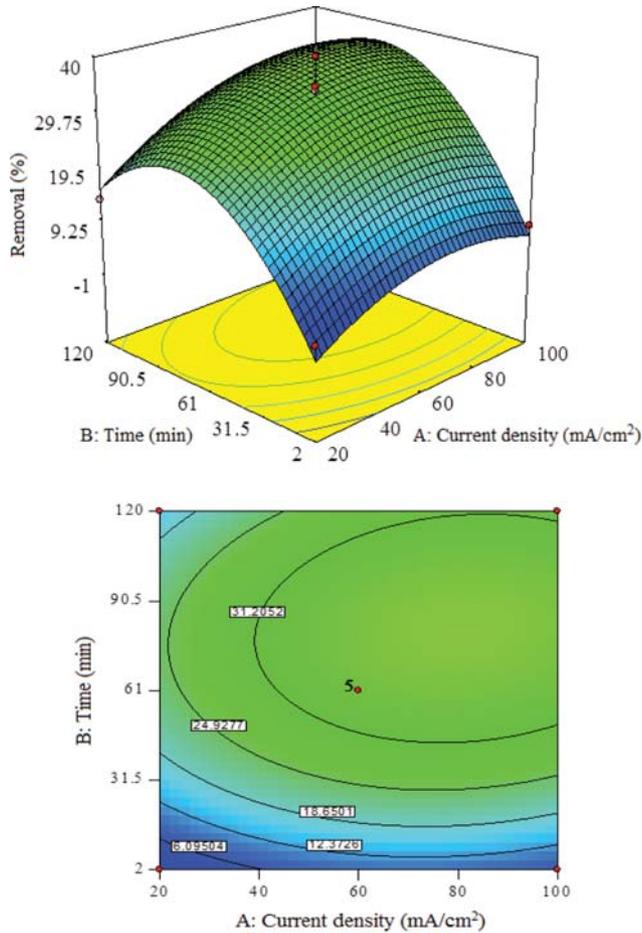


Fig. 2. Removal efficiency based on current density and time (AMX concentration: 50 mg/l, electrolyte concentration: 550 mg/l, pH: 7, UV radiation).

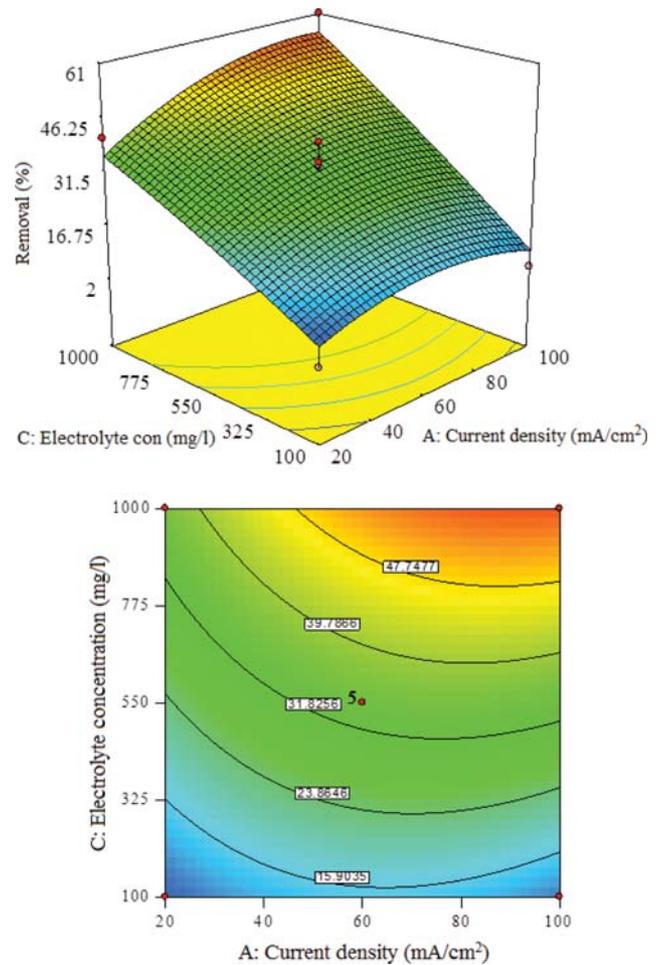


Fig. 3. Removal efficiency based on current density and electrolyte concentration (AMX concentration: 50 mg/l, time: 61 min, pH: 7, UV radiation).

RESULTS AND DISCUSSION

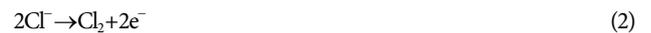
1. The Effect of Current Density, Reaction Time, and Electrolyte Concentration

As mentioned in the method section, in the first step, the effect of three variables of current density, reaction time, and electrolyte concentration on the removal efficiency was investigated in 17 runs. The results of AMX removal based on the variations of the three variables are presented in Table 2.

Fig. 2 shows the removal efficiency based on the current density and reaction time. Moreover, in Figs. 3 and 4, the removal efficiency is, respectively, shown on the basis of current density and electrolyte concentration (Fig. 3), and reaction time and electrolyte concentration (Fig. 4).

Current density is one of the key parameters in electro-chemical advanced oxidation processes (EAOPs), since it is related to species that play the oxidation role in the process. Furthermore, in all EAOPs, controlling the amount of $M(OH^{\bullet})$ according to Eq. (1) and indirect production of oxidizing agents such as chlorine active species produced by Eqs. (2) and (3) are important [34]. Generally, the amount of pollutants degradation increases by increasing the current density and production of oxidizing species, due to the

presence of electrolyte in the medium [35-38].



The type of electrolyte used not only affects the transfer of electrons in the environment, but also can produce potent oxidizing agents such as chlorine active species [39].

NaCl and Na_2SO_4 are usually used to provide electrolytes in electrochemical processes. In the processes of AO and AO- H_2O_2 , organic compound removal and their mineralization occur more rapidly in the presence of NaCl compared to Na_2SO_4 [40,41] due to the degradation of organic compounds in the presence of active chlorine species. Furthermore, hydroxyl radical removal process is faster in the presence of sulfate compared to chloride. Therefore, studies have shown that organic compounds are more likely to degrade in the presence of NaCl [40,42]. Table 3 indicates the ANOVA test for analyzing the data. Eq. (4) shows the suggested removal efficiency equation based on the model and regression coefficients.

In this Equation A, B, and C are, respectively, current density, reaction time, and electrolyte concentration. The coefficients in Eq.

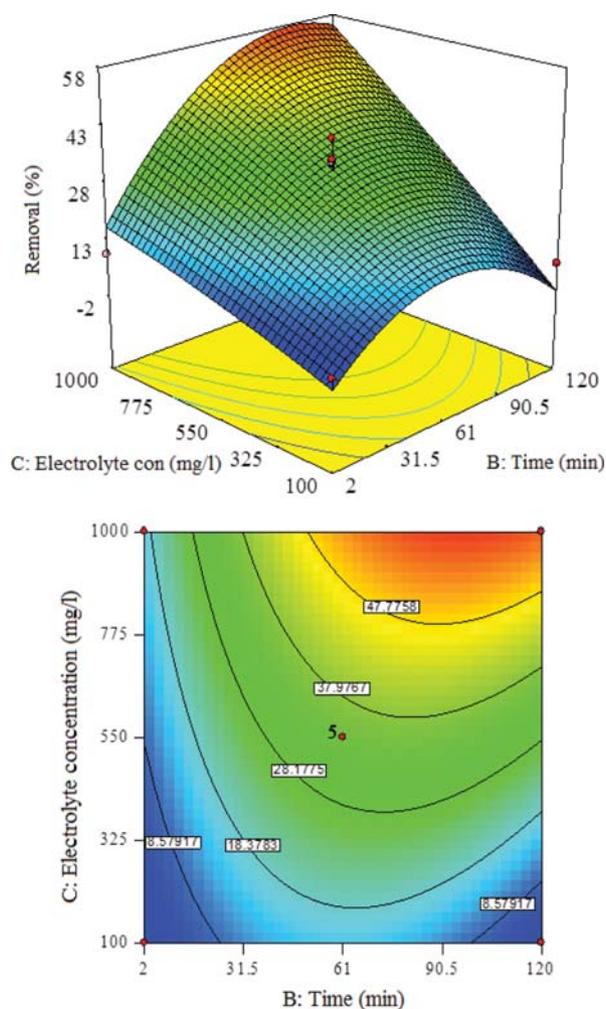


Fig. 4. Removal efficiency based on time and electrolyte concentration (AMX concentration: 50 mg/l, current density: 60 mA/cm², pH: 7, UV radiation).

(4) show the positive effect of all the three variables on the AMX removal efficiency.

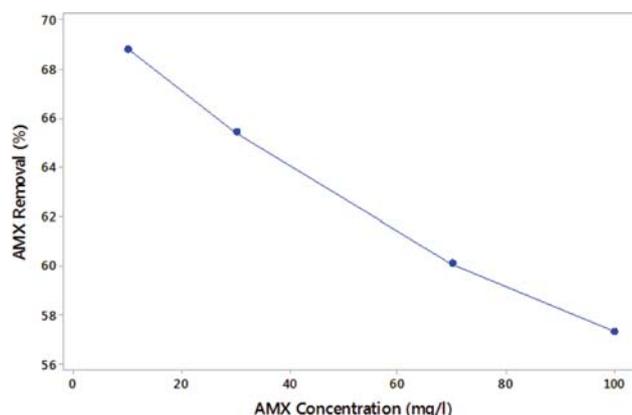


Fig. 5. AMX removal efficiency at different concentrations of the drug (current density: 94 mA/cm², time: 95 min, electrolyte concentration: 997 mg/l, and pH: 7).

$$\text{Removal (\%)} = +33.89 + 5.38 \times A + 9.81 \times B + 18.43 \times C + 9.38 \times B \times C - 5.81 \times A^2 - 15.67 \times B^2 \quad (4)$$

As shown in Table 3, the P-value of the three studied variables in this study is significant (P-value < 0.005). Among the variables studied, based on the values of F value, the greatest effect is related to electrolyte concentration [29] due to the high impact of indirect anodic oxidation electrolyte concentration resulting from the products of electrochemical process produced in the cathode electrode. The anodic oxidation process can be improved through the active species of chlorine and H₂O₂ produced by electrochemical processes in the cathode [43]. When chlorine is produced by anodic oxidation, H₂O₂ is produced by the reduction process in cathode. In the presence of these indirect oxidizing agents, the anodic oxidation is called “mediated oxidation” [34]. Chlorine active species are the most important indirect oxidizing agents used in wastewater treatment and are among the first products of electrode, even at low current densities. Mediated oxidation by active chlorine is dependent on the direct oxidation of chloride ions in anode and production of chlorine (Cl₂) through Eq. (4). Chlorine produces

Table 3. ANOVA for response surface reduced quadratic model

Source	Sum of squares	df	Mean square	F value	p-Value
Model	5296.69	6	882.78	22.72	<0.0001
A-current density	231.13	1	231.13	5.95	0.0349
B-time	769.50	1	769.50	19.81	0.0012
C-electrolyte conc	2718.06	1	2718.06	69.96	<0.0001
BC	352.31	1	352.31	9.07	0.0131
A ²	142.33	1	142.33	3.66	0.0846
B ²	1036.90	1	1036.90	26.69	0.0004
Residual	388.51	10	38.85		
Lack of fit	342.62	6	57.10	4.98	0.0711
Pure error	45.89	4	11.47		
Cor total	5685.20	16			
	R-squared		0.9317		
	Adj R-squared		0.8907		

Table 4. Optimal conditions for the three variables of current density, reaction time, and electrolyte concentration

Parameter	Current density (mA/cm ²)	Reaction time (min)	Electrolyte concentration (mg/l)	Removal (%)	
				Predicted	Experimental
Optimum condition	94	95	997	62.47	63.02

hydrochloric acid (HClO) in accordance with Eq. (5) [44]. Table 4 shows the optimal conditions predicted by the model.

2. The Effect of Antibiotic Concentration

After determining the optimal conditions for the three variables (current density, reaction time and electrolyte concentration), in the second stage, the effect of AMX concentration on the removal efficiency was investigated. Fig. 5 shows the removal efficiency of AMX at various concentrations of the AMX. By increasing the AMX concentration, the removal efficiency decreases.

According to other studies, in all EAOPs, an increase in the organic compound concentration requires an increase in treatment time to achieve acceptable treatment levels [45-49]. By increasing the organic compound concentration, their removal efficiency by the process is reduced in the same conditions [50-52]. By increasing the amount of contaminant concentration, the amount of contaminant removal increases per unit of reaction time [53,54]. The results of the study by Zuorro et al. on the removal of reactive green 19 dye by the UV/H₂O₂ process, as well as the study done by Chen et al. on the removal of Red Amine Blue dye by the H₂O₂/Fe₃O₄ process, are consistent with the results of the present study [55,56].

3. The Effect of pH

The effect of solution pH on the AMX removal efficiency was evaluated under optimal conditions for the variables, including current density, reaction time, electrolyte concentration, and AMX concentration. Fig. 6 shows the effect of pH on the AMX removal efficiency in optimal conditions. By increasing pH, the antibiotic removal efficiency decreases. The highest removal rate is 71.43% at pH 2.

Increasing the AMX removal efficiency at lower pH is due to the existence of dominant active chlorine species in this pH [44]. HClO can be converted to ClO⁻, depending on pH, based on Eq.

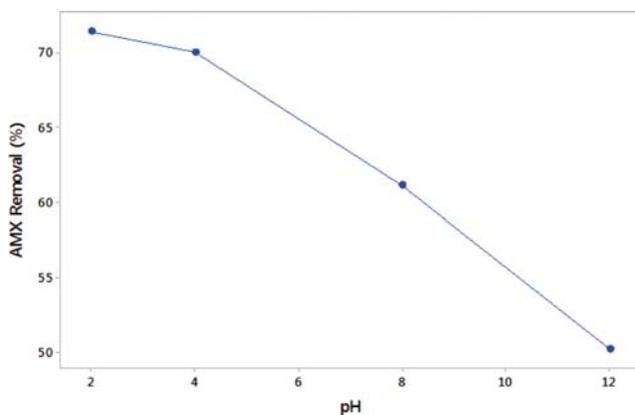


Fig. 6. AMX removal efficiency at different pH (current density: 94 mA/cm², time: 95 min, electrolyte concentration: 997 mg/l, and AMX concentration: 10 mg/l).

(5), and vice versa.



Up to pH 3 the dominant chlorine species in the solution is Cl₂, from pH 3 to 8 HClO, and at pH above 8 the dominant species is ClO⁻ [57]. Given that HClO (E°=1.49 V/SHE) and Cl₂ (E°=1.36 V/SHE) show a higher redox potential than OCl⁻ (E°=0.89 V/SHE), the oxidation of organic compounds in acidic conditions is faster than alkaline conditions [58]. According to Eq. (6), HClO can be converted to ClO³⁻, which reduces organic compounds degradation [44].



Previous studies emphasized the increase in the efficiency of AO and AO-H₂O₂ processes at pH of about 3 compared to higher pH [59,60]. This issue is explained by the presence of stronger oxidizing species as anionic radicals, as well as the availability of active chlorine species with higher oxidation potential in acidic pH [58]. Generally, the best performance for electrochemical processes has been reported at pH 3 [61,62]. It could be due to the absence of carbonate and bicarbonate species, which reduces the process efficiency due to the degradation effects on OH[•], [63] and the spontaneous degradation of H₂O₂ to water and oxygen, which usually occurs at pH above 5 [62]. On the other hand, some studies have reported the maximum amount of organic pollutants mineralization at pH 2-4 [64,65], 2-3 [66] and 3-4 [67].

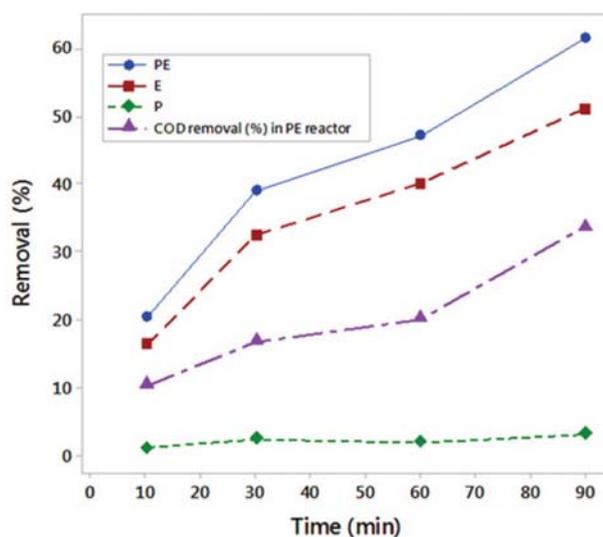


Fig. 7. AMX removal efficiency in the PE, E, and P reactors, and COD removal efficiency in the PE reactor at different reaction times (current density: 94 mA/cm², electrolyte concentration: 997 mg/l, AMX concentration: 10 mg/l, and pH: 7).

4. Comparison of the Process Efficiency Separately and Combined

In this study, the AMX removal efficiency was evaluated and compared separately in the P, E, and combined PE reactors (Fig. 7). Moreover, the COD removal efficiency was evaluated in the PE reactor.

As Fig. 7 indicates, the most effective removal efficiency is related to the E reactor compared to the P reactor. The P reactor alone has an average of 5% AMX removal efficiency, which in the PE combined reactor alone increases the AMX removal efficiency by 17.23% compared to the E reactor. It shows the synergistic effect of the two processes of E and P in the combined process. Increasing the AMX oxidation is due to the presence of UV light and its role as a potential activation technology, referred to in various studies [68,69]. UV rays produce free radicals that can oxidize organic compounds by affecting some of the E process products [15]. The free radicals produced in the combined process include those listed in Eqs. (7) to (9) [70].



5. Antimicrobial and Toxicity Test Results

One of the methods for determining the amount of contaminant activity is microbial testing, determining the antimicrobial property and toxicity of the effluent after the process [71]. In this stage, the bioavailability and toxicity of the final effluent from the reactor were evaluated in optimal conditions, respectively, on *Staphylococcus aureus* bacteria and *Saccharomyces cerevisiae* yeast. Fig. 8 shows the results of the final effluent bioavailability test using the inhibition zone method test. Fig. 9 represents the toxicity of final effluent on *S. cerevisiae* yeast.

Fig. 7 shows that the ratio of inhibition zone diameter in different percentages of AMX antibiotic removal is different from that of the conditions the *S. aureus* bacteria is contacted with 10 mg/l of pure AMX solution. This ratio decreases by increasing removal percentage; therefore, in the removal efficiency of 60% this ratio decreases to 0.26, which indicates the reduction of antibacterial

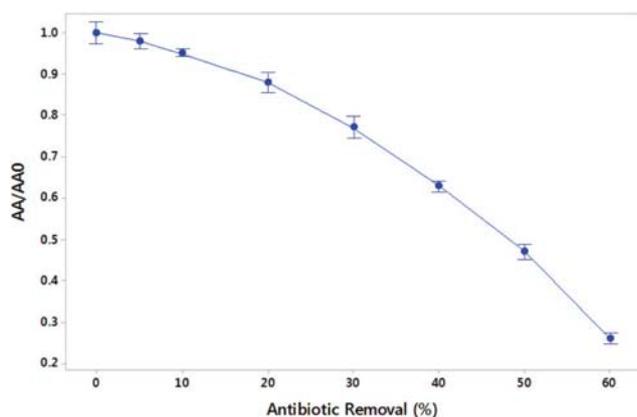


Fig. 8. Final effluent bioavailability resulting from PE process on *S. aureus* bacteria (current density: 94 mA/cm², electrolyte concentration: 997 mg/l, AMX concentration: 10 mg/l, and pH: 7).

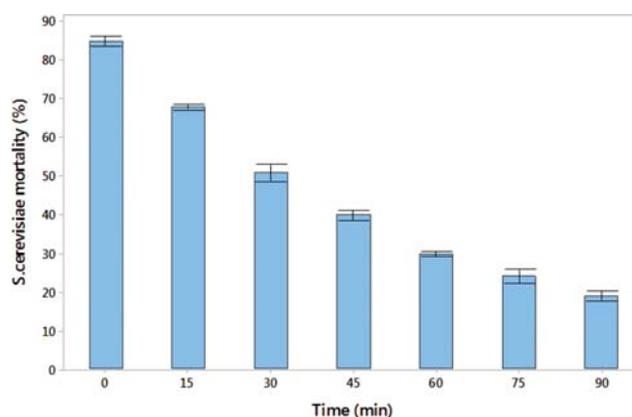


Fig. 9. Final effluent toxicity from PE process on *S. cerevisiae* yeast (current density: 94 mA/cm², electrolyte concentration: 997 mg/l, AMX concentration: 10 mg/l, and pH: 7).

properties or the bioavailability of the products in the final effluent resulting from the PE process. This decreasing trend is also observed in the final effluent toxicity. The mortality rate of *S. cerevisiae* yeast in a solution containing 10 mg/l of AMX is 85% at zero reaction time, which decreases to 19% in 90 minutes with the PE process. This result is due to the reduction of final products toxicity in the PE reactor effluent. In previous studies, advanced oxidation has been able to effectively reduce toxicity [17].

6. Anodic Oxidation

AMX removal by anodic oxidation was investigated in this study. Anodic oxidation can be conducted through direct electron transfer from the surface of the anode, or through heterogeneous reactive oxygen species (ROS), that have been created as intermediate products resulting from water oxidation. These intermediate products have hydroxyl radicals and are physically located at the surface of the anode (with M(^oOH) shown in Equation 10). Furthermore, weaker oxidants, such as H₂O₂ produced by dimerization of M(^oOH) in Eq. (11), play a role in anodic oxidation. The other oxidant is O₃ which is produced by Eq. (12) at the surface of the anode [34].



CONCLUSION

The present study examined the effect of P and E processes (separately and combined) on the AMX removal efficiency from aquatic environment. The results showed that under neutral pH conditions, the optimum value of AMX removal was 62.47% at current density, reaction time, and electrolyte concentration of 94 mA/cm², 95 min and 997 mg/l, respectively. Examining the effect of other variables showed that by increasing the AMX concentration, the removal efficiency decreases. Furthermore, evaluating the pH parameter showed that pH reduction to 2 increases AMX removal to 71.43%. Investigating the type and comparison of the efficiency of

the processes used in this study showed that the AMX removal efficiency in the PE combined reactor is higher than that of the P and E processes alone. This comparison also showed that the combination of the P and E processes caused a synergistic effect that improves the removal efficiency in the PE combined reactor. Investigating the COD removal rate showed that the maximum COD removal efficiency in 95 min was 33.51% in the PE combined reactor. Studying the bioavailability and toxicity of the final effluent from the combined reactor in optimal conditions showed that both toxicity and bioavailability parameters decreased, indicating the low toxicity and antimicrobial properties of the final products resulting from AMX degradation compared to the pure AMX solution.

Based on the results, it is concluded that the PE process is able to remove more than 60% of AMX from aquatic environments in optimal conditions as well as removing more than 30% of COD. By decreasing the toxicity and antimicrobial effect of the final effluent, it can be treated by biological wastewater treatment processes.

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