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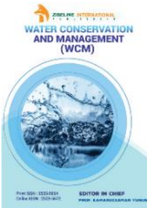

RESEARCH ARTICLE

REMOVAL OF PHARMACEUTICAL COMPOUNDS FROM SECONDARY TREATED DOMESTIC WASTEWATER USING ELECTROCHEMICAL TREATMENT TECHNIQUE

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ABSTRACT

This study explores the electrocoagulation (EC) process for removing six pharmaceutical compounds (PCs) from wastewater. The compounds investigated include azithromycin, diazinon, ibuprofen, ketoconazole, sertraline, and diclofenac. We employed Response Surface Methodology (RSM), specifically a Box-Behnken design, to systematically analyze how temperature, pH, and current density affect the removal efficiency of these compounds. Experimental results indicated that run 11, under optimal conditions of 20°C, a pH of 6.0, and a current density of 29.40 A/m^2, achieved the highest removal efficiencies. Specifically, the removal rates were 65.27% for Azithromycin and 70.09% for diazinon. Statistical analysis using ANOVA showed that current density has a positive influence on removal efficiency, while both temperature and pH have negative impacts. We also explored the interactions among the parameters, revealing significant insights into the operational dynamics of the EC process.

KEYWORDS

Removal efficiency; Electrochemical treatment; Pharmaceutical compounds; Wastewater treatment.

1. INTRODUCTION

The water issue is very critical in Jordan as it is severely impacted by climate change (Matouq et al., 2013). Therefore, Jordan has built many wastewater treatment plants (WWTPs) that produce up to 250 MCM annually (Al-Mubaidin et al., 2022). The consumption and management role for various domestic uses were frequently studied (Al-Hamaiedeh et al., 2023). A lot of attention was paid to enhancing the quality of treated wastewater and other products such as sludge (Aljbour, Al-Hamaiedeh, et al., 2021; Aljbour, El-Hasan, et al., 2021). The persistent presence of pharmaceutical compounds (PCs) in wastewater is a pressing environmental concern due to their potential adverse effects on aquatic ecosystems and human health. Conventional wastewater treatment plants (WWTPs) often fall short inadequately eliminating PCs, resulting in their accumulation in natural water bodies. This underscores the necessity for advanced treatment technologies capable of effectively removing pharmaceutical residues from treated wastewater (Corcoran et al., 2010; Rosal et al., 2010).

Pharmaceutical compounds, including antibiotics, analgesics, and endocrine disruptors, are frequently detected in treated effluent at concentrations ranging from nanograms to micrograms per liter (Behera et al., 2011). These contaminants enter wastewater primarily through human excretion and the disposal of unused medication (Helmecke et al., 2020). Despite their low concentrations, the ecological and health risks associated with these compounds are substantial, as they can disrupt aquatic ecosystems and potentially enter the human food chain (Carballa et al., 2004). Removal of PC from treated wastewater (TWW) is essential for regions where treated wastewater is being reused for irrigation like Jordan to mitigate the environmental and health risks associated with PC.

The removal of PCs from wastewater presents a significant challenge due to their persistence in the environment (Samal et al., 2022; Thakur et al., 2023). Various conventional treatment technologies, such as biological treatment, adsorption using activated carbon, and membrane filtration, have been employed (Eniola et al., 2022; Jelić et al., 2012; Verlicchi et al., 2012). However, these methods have limitations. Biological treatment struggles to efficiently remove hydrophobic or persistent compounds like carbamazepine, while adsorption requires frequent regeneration of the adsorbent, leading to higher operational costs (Gupta, 2009). Membrane filtration, although effective, suffers from membrane fouling and high-energy consumption (Boehler et al., 2012). Advanced oxidation processes (AOPs), such as ozonation and UV/H_2O_2, have shown promise in degrading PCs but often generate toxic by-products and are energy-intensive (Andreozzi et al., 1999). In contrast, electrochemical treatment techniques, including electro-oxidation and electro-coagulation, offer more efficient and selective removal of PCs without the need for chemical additives. Despite challenges such as energy consumption and electrode maintenance, electrochemical methods provide significant advantages, including reduced sludge production and the potential for integration with AOPs to achieve near-complete mineralization of contaminants (Nagarajan et al., 2023). This makes them a promising option for addressing the limitations of traditional methods in pharmaceutical compound removal.

Electrochemical treatment has emerged as a promising solution for removing pharmaceutical compounds from wastewater (Nagarajan et al., 2023). This technique involves applying direct current to electrodes submerged in the wastewater, which facilitates the degradation of

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contaminants through oxidation and reduction reactions or by electrocoagulation, where the coagulants consist of dissolved metals from the electrodes. The benefits of electrochemical treatment include high removal efficiencies, operational simplicity, and the ability to target a wide range of pharmaceutical compounds without requiring chemical additives (Ahmad et al., 2022, Muddemann et al., 2019). Additionally, electrochemical methods, particularly electrocoagulation (EC), are widely employed for treating both domestic and industrial wastewater (Kyzas and Matis, 2016).

EC involves immersing metal electrodes, typically aluminum or iron, into wastewater and passing an electrical current through the system (Ghernaout et al., 2011; Islam, 2019). This induces electrolysis, causing the metal at the anode to dissolve and release metal cations into the water. These metal cations neutralize and coagulate suspended solids, colloids, heavy metals, and organic pollutants. The resulting flocs, which are larger and heavier than the individual particles, settle more easily at the bottom of the treatment vessel, forming sludge. Concurrently, hydrogen bubbles generated at the cathode help float additional aggregates to the surface. In the case of aluminum electrodes, the electrolysis process releases aluminum hydroxide, which further aids in coagulation by adhering to suspended particles and forming dense flocs. These flocs can then, be removed from the treated water through sedimentation, filtration, or other separation techniques, contributing to the purification process (Garcia-Segura et al., 2017; Mollah et al., 2004; Mollah et al., 2001).

The specific objectives of this study are to analyze the effectiveness of the electrocoagulation process in removing pharmaceutical compounds (PCs) from secondary treated domestic wastewater in Jordan. This will involve investigating the influence of key operational parameters, such as initial temperature, initial solution pH, and current density, on treatment efficiency. Finally, the study aims to design and optimize a factorial experimental setup to systematically assess the interactions between these variables.

2. MATERIAL AND METHODS

2.1 Description of the study area

The study was conducted using treated wastewater (TWW) from the Ain Albasha wastewater treatment plant (ABWWTP), which is located along the Amman-Jerash main road. The plant has been operating since 1988. The initial design capacity of the plant was 4,000 cubic meters per day. In 2010, the design capacity was expanded to 14,900 cubic meters per day. The average influent flow reached 17,350 cubic meters day daily in 2010 (MWI, 2018).

2.2 Sample Collection, Preparation, and Characterization

To evaluate the EC process for removing (PCs) from wastewater, samples of secondary TWW were collected from the outlet of the secondary settling tank at the ABWWTP. During collection, the samples were placed in sterile glass jars that had been pre-cleaned with distilled water and kept on ice during transport to the laboratory, following standard laboratory procedures to ensure sample integrity. The TWW samples were analyzed to quantify 40 PCs using Liquid Chromatography-Mass Spectrometry (LC-MS) with a Shimadzu LC system. The PCs included a variety of categories from different pharmaceutical classes. Samples were stored at -2°C and cleaned using liquid-liquid extraction and solid-phase extraction methods. This methodology ensures accurate measurement of PC concentrations, providing insights into their removal efficiency during wastewater treatment.

2.3. Experimental Setup

The experimental setup included two 1 mm thick rectangular aluminum electrodes, a 2-liter open-top glass beaker, and a magnetic hot stirrer plate for mixing and temperature control. Analytical instruments used for monitoring included calibrated pH probes, conductivity meters, a DC voltmeter, an ammeter gauge, a timer, and a thermometer. A DC power supply provided a constant voltage of 21.8 V, with the current varying based on the wastewater sample's components. The electrodes were washed with distilled water and dried to remove impurities before each experiment. The EC cell was filled with 1.5 liters of wastewater, ensuring proper immersion of the electrodes. Initial parameters such as pH, temperature, and current density were recorded before starting the EC process. After the EC process, the same parameters were recorded again to assess the changes. Figure 1 shows a schematic for the EC batch reactor.

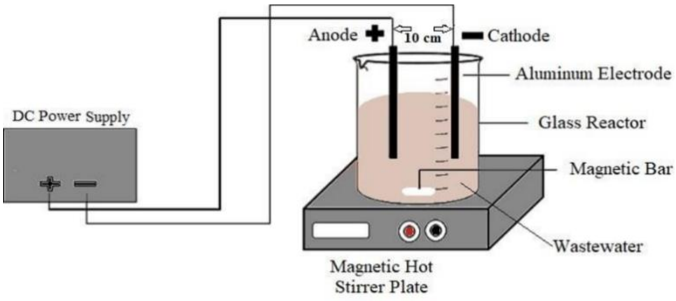

Figure 1: Experimental setup

2.4 Effect of process parameters

The study employed a surface response methodology to evaluate the impact of three factors which are the initial pH, current density (CD), and temperature on the degradation of pharmaceuticals in secondary treated domestic wastewater.

To identify the optimal conditions for PC removal, the Box-Behnken design method was utilized, resulting in 15 experiments conducted under uniform conditions for 45 minutes each. The design involved three levels for each factor (-1, 0, +1), facilitating the creation of a quadratic model to study the interactions between temperature, pH, and CD. This approach allowed for the generation of effect and interaction plots and ANOVA analysis using Minitab 19, providing a comprehensive understanding of how these factors influence the electrocoagulation (EC) process. The methodology ensured a robust experimental design, enabling the optimization of EC tests and the effective removal of PCs from secondary treated wastewater (Al-Mrayat et al., 2022; Aljbour, 2019; Aljeradat et al., 2022). Table 1 shows the actual values of the independent variables (temperature, pH, and CD) utilized in the optimization of the overall EC tests, employing a three-factor Box-Behnken design in this study.

Table 1: Temperature, pH, and CD in the optimization of overall EC tests at a constant contact time of 45 minutes under uniform conditions, using a three-factor Box-Behnken design

Run	X1	X2	X3
1	-1	-1	0
2	-1	1	0
3	1	-1	0
4	1	1	0
5	-1	0	-1
6	-1	0	1
7	1	0	-1
8	1	0	1
9	0	-1	-1
10	0	-1	1
11	0	1	-1
12	0	1	1
13	0	0	0
14	0	0	0
15	0	0	0

Run	T (C°)	pH	CD (A/m²)
1	10	6	16.75
2	10	10	16
3	30	6	24.25
4	30	10	21.25
5	10	8	11.7
6	10	8	20.94
7	30	8	11
8	30	8	27.8
9	20	6	13.75
10	20	6	29.4
11	20	10	15.2
12	20	10	25.3
13	20	8	17.25
14	20	8	18
15	20	8	17.75

The selected temperature values were (10°C, 20°C, and 30°C) which reflect the average ambient temperatures in Jordan throughout the year, while pH values (6.0, 8.0, and 10.0) correspond to the acidic and basic conditions typical of treated sewage at the WWTP. Current density values were determined based on sewage salinity and conductivity.

2.5 Assessment of PC Degradation in the EC Process

The degradation of PCs was evaluated in terms of removal efficiency (R%). The EC process is pivotal in assessing the efficacy of removing PCs from a wastewater sample. The following equation is used to calculate the % R (Aliedeh et al., 2021; Aljbour et al., 2017):

$$\% R = [\frac{C_0 - C_f}{C_0}] \times 100 \quad (1)$$

C_o and C_f are the initial and final concentrations of selected PCs (ng/L).

To assess the suitability of TWW for purposes such as drinking water supply, recreation, and sustaining diverse ecosystems, the evaluation of the TWW also involved assessing the individual concentration of each PC and comparing it to the Predicted No-Effect Concentration (PNEC). The PNEC represents the concentration below which a substance is not expected to pose any adverse effects on the environment. It is derived

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from ecotoxicological studies and is commonly used in environmental risk assessments to ensure that the presence of pharmaceutical compounds in water bodies remains at safe levels. Table 2 shows the PNEC values for each selected PC obtained from the NORMAN Ecotoxicology database (Spurgeon et al., 2022).

Table 2: PNEC values due to the NORMAN Ecotoxicology Database

Pharmaceutical Compound	Lowest PNEC (ng/L)	Pharmaceutical Compound	Lowest PNEC (ng/L)	Pharmaceutical Compound	Lowest PNEC (ng/L)
1,7-Dimethylxanthine	21400	Ketorolac	370	Erythromycin	300
Acetaminophen	46000	MDA	830	Fluoxetine	100
Amlodipine	230	MDMA	30100	Ibuprofen	11
Amphetamine	24800	Metformin	160000	Ketoconazole	8.1
Atenolol	150000	Methamphetamine	9740	Ketoprofen	2100
Azithromycin	19	Metoprolol	8600	Sulfachloropyridazine	730
Trimethoprim	120000	Morphine	5380	Sulfamethazine	30000
Caffeine	1200	Naproxen	1700	Sulfamethoxazole	600
Carbamazepine	2000	Ofloxacin	1390	Thiabendazole	1200
Cimetidine	176000	Oxazepam	370	Sertraline	9.4
Citalopram	16000	Paracetamol	14100	Diphenhydramine	990
Cotinine	10000	Phenazone	1100	Risperidone	380
Diazinon	10	Propranolol	200	Diclofenac	50

3. RESULTS AND DISCUSSION

3.1 Classification of PCs

In a study by Al-Qaisy, a questionnaire was distributed to pharmacies in Karak/Jordan to identify prevalent medications consumed by the population (Al-Qaisi, 2023). These medications excreted through urine and feces, enter wastewater treatment plants (WWTPs) in varying concentrations. The PCs were categorized into 14 groups based on their effects on different bodily systems, following Nawaz and Sengupta's classification (Nawaz & Sengupta, 2019). At the (ABWWTP), over 500 emerging contaminants, including pharmaceuticals and pesticides, were detected in TWW, which was analyzed using LC-MS. The study focused on groups namely central nervous system stimulants and depressants, analgesics, antipyretics, anticonvulsants, cardiovascular medications, antimicrobial agents, antipsychotics, NSAIDs, azole antifungals, anthelmintics, and nicotine metabolites, as detailed in Table 2. The analyses result for secondary TWW at the ABWWTP, collected on March 2024 are shown in Table 3.

Table 3: The concentrations of selected PCs measured in ng/L within the raw sample from the secondary TWW collected at the ABWWTP

No.	Pharmaceutical Compound	Raw sample Conc. (ng/L)	No.	Pharmaceutical Compound	Raw sample Conc. (ng/L)
1	1,7-Dimethylxanthine	30.3	21	Diclofenac	75.7
2	Acetaminophen	32.5	22	Ketorolac	81.2
3	Amphetamine	35.1	23	Ketoprofen	36.6
4	Caffeine	140.4	24	Amlodipine	39.3
5	Carbamazepine	144.8	25	Atenolol	32.5
6	Cotinine	37.3	26	Bisprolol	52.7
7	Phenazone	42.4	27	Citalopram	31.4
8	Sulfamethoxazole	41.5	28	Diazinon	57.5
9	Sulfamethazine	35.3	29	Fluoxetine	44.8
10	Trimethoprim	39.6	30	Ketoconazole	39.9
11	Cimetidine	36.5	31	Metoprolol	32.5
12	Diphenhydramine	41.4	32	Metformin	53.4
13	MDA	38.8	33	Oxazepam	144.1
14	MDMA	36.3	34	Paracetamol	172.2
15	Thiabendazole	34.4	35	Propranolol	185.5
16	Sulfachloropyridazine	32.5	36	Risperidone	46.6
17	Morphine	36.6	37	Sertraline	38.3
18	Methamphetamine	85.8	38	Ofloxacin	57.7
19	Erythromycin	77.7	39	Azithromycin	180.8
20	Ibuprofen	63.3	40	Naproxen	82.8

The concentrations of selected PCs measured in the raw sample from the secondary TWW at ABWWTP, as shown in Table 2, indicate the potential for toxicity effects of six compounds (Diclofenac, Sertraline, Ketoconazole, Ibuprofen, Diazinon, and Azithromycin). This inference is drawn from a comparison with the compounds listed in Table 2. These results align with the results of previous research, which also observed high toxicity associated with these compounds in wastewater (Beril & Celik, 2017; Khasawneh & Palaniandy, 2021). Therefore, during EC treatment the focus was on these six PCs. Figure 2 shows a summary risk ranking of the six selected PCs in the raw sample that exceed PNECs.

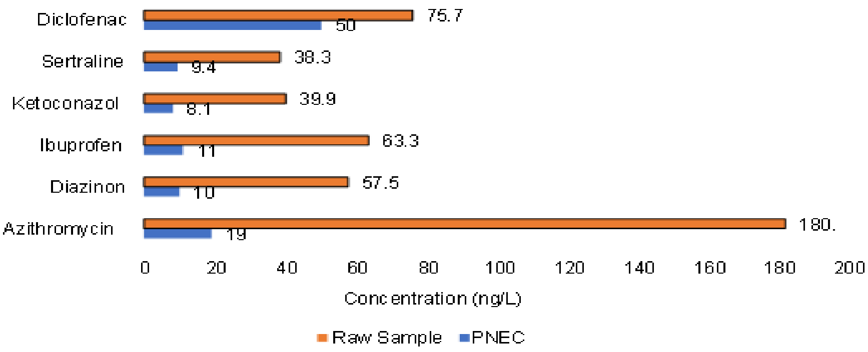

Figure 2: Summary risk ranking of the six selected PCs in the raw sample that exceeds the PNECs

3.2 Effect of electrolysis time

The study aimed to determine the optimal electrolysis time for the (EC) process to remove selected PCs from secondary TWW. EC experiments were conducted at ambient temperature with constant pH and agitation speed. Electrolysis times ranging from 5 to 45 minutes were tested, chosen for their short duration and economic feasibility, especially for future application in WWTPs in developing countries. Figure 3. shows the effect of electrolysis time on the % R of PCs.

The results presented in Fig. 3 show that longer electrolysis times resulted in higher removal efficiencies. Extending the electrolysis time enhances the removal efficiency. However, this also leads to increased electricity consumption for the EC process, which raises the associated costs. Considering electricity consumption, the optimal electrolysis time for effectively removing PCs while balancing economic factors was determined to be 45 minutes.

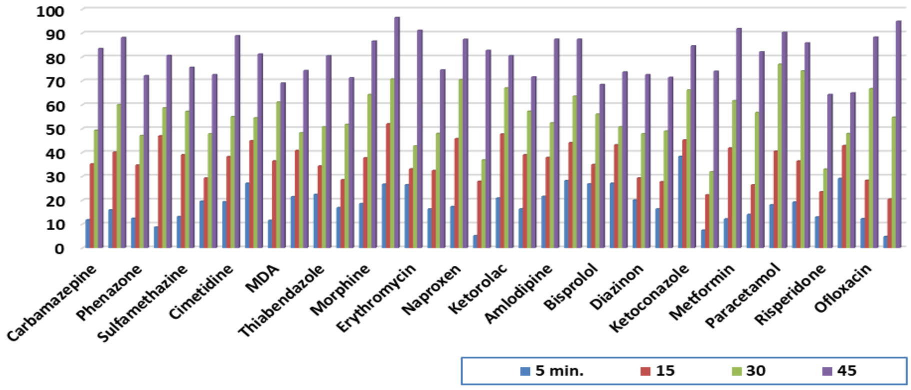

Figure 3: Effect of electrolysis time on removal efficiency of pharmaceuticals

3.3 Surface Response Analysis of Removal Efficiency in Electrocoagulation Process Experiments

The surface response analysis of the R% for the EC process was performed using the Box-Behnken design. The experimental design included 15 runs, and we assessed the removal efficiencies for six pharmaceuticals: Azithromycin, Diazinon, Ibuprofen, Ketoconazole, Sertraline, and Diclofenac. These compounds were chosen because they are found in high concentrations in treated wastewater, often exceeding the PNEC, which highlights the need for effective removal during treatment. In these experiments, the electrolysis time was consistently set at 45 minutes. The results of the removal efficiencies for each experimental run are presented in Table 4.

Table 4: The R% for the selected PCs under different experimental conditions

Run	Experimental Condition			% R					
	X1	X2	X3	PC1	PC2	PC3	PC4	PC5	PC6
1	-1	-1	0	62.78	64.87	60.19	58.40	45.69	54.16
2	1	-1	0	61.95	62.96	57.98	52.63	41.78	57.07
3	-1	1	0	27.38	29.74	12.32	36.84	31.59	21.40
4	1	1	0	26.11	30.61	15.48	30.58	24.80	22.32
5	-1	0	-1	20.30	28.52	12.32	26.82	21.93	19.29
6	1	0	-1	22.40	31.65	15.80	27.82	32.90	19.95
7	-1	0	1	51.99	43.83	36.97	46.62	46.21	35.54
8	1	0	1	31.91	41.57	21.17	41.10	44.39	32.50
9	0	-1	-1	24.17	48.00	42.18	41.10	39.16	27.08
10	0	1	-1	18.85	22.78	7.58	19.05	15.14	16.38
11	0	-1	1	65.27	70.09	64.61	69.17	56.66	58.92

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Table 4 (Cont): The R% for the selected PCs under different experimental conditions

12	0	1	1	30.48	36.35	19.27	46.87	41.51	25.23
13	0	0	0	28.15	36.87	17.54	34.34	32.64	26.68
14	0	0	0	28.04	35.13	13.90	28.32	38.12	25.63
15	0	0	0	26.38	30.61	10.58	27.07	40.73	22.32

PC1 = Azithromycin, PC2 = Diazinon, PC3 = Ibuprofen, PC4 = Ketoconazole, PC5 = Sertraline, and PC6 = Diclofenac

The experimental results for the EC process of six PCs indicate significant variations in the percentage of removal (% R) across different runs. Notably, run 11 yielded the highest removal efficiency, underscoring the effectiveness of the operational conditions applied during this run. The results from run 11 not only confirm the efficacy of the chosen conditions but also provide a valuable reference point for future studies and practical applications in wastewater treatment processes. Continued research in this area is essential to further refine the parameters for optimal performance across a broader range of contaminants.

The main effect plots in Figure 4 illustrate the influence of the three factors, namely: temperature, pH, and current density on the % R of the six PCs (Azithromycin, Diazinon, Ibuprofen, Ketoconazole, Sertraline, and Diclofenac). The results show a consistent qualitative trend for all six compounds. The results indicate that temperature has a moderate effect on removal efficiency, with slight improvements at lower temperatures. This effect is likely due to the enhanced stability of coagulant species and reduced side reactions at cooler temperatures, which improves the performance of the EC process. However, the overall effect is not dramatic, suggesting that temperature is not the primary limiting factor in this system.

The pH shows a negative impact on % R, particularly when the pH deviates from neutral (pH 6). The decrease in % R at more acidic or alkaline conditions can be attributed to the destabilization of the coagulant flocs and the altered solubility of pharmaceuticals under extreme pH conditions. Neutral pH is optimal as it supports the effective formation of metal hydroxide flocs, which are necessary for the adsorption and removal of pharmaceuticals from the solution.

Current density has a positive impact on the % R. As the current density increases, more metal ions are released from the electrodes, which enhances the formation of flocs and improves the % R. However, this effect plateaus at higher current densities due to limitations in the system, such as excessive heating or the saturation of reactive species in the solution.

The overall trends for these effects are qualitatively similar for all six pharmaceutical compounds, indicating that despite their chemical differences, the electrocoagulation process is broadly effective for their removal when these parameters are optimized.

Figure 5 presents the interaction effect plots between the three parameters (temperature, pH, and current density) on the removal efficiency. Interaction plots illustrate how the combined effect of two factors influences the response, providing insight into whether changes in one factor alter the effect of another.

The interaction plots in Figure 5 reveal relatively weak interaction effects among the parameters studied. This suggests that the influence of one factor on the removal efficiency does not significantly depend on the levels of the other factors. For example, temperature changes do not significantly alter the impact of pH or current density on removal efficiency. The near-parallel lines in the plots indicate minimal interaction between the parameters.

The weak interactions observed could be due to the dominant effects of individual parameters, such as pH and current density, which exert stronger, more direct influences on the electrocoagulation process than the combined effects of temperature with the other factors. The process might also be less sensitive to combined parameter variations due to the consistent behavior of the pharmaceuticals across the different experimental conditions, or because the parameters are being studied within a range that minimizes their interaction effects.

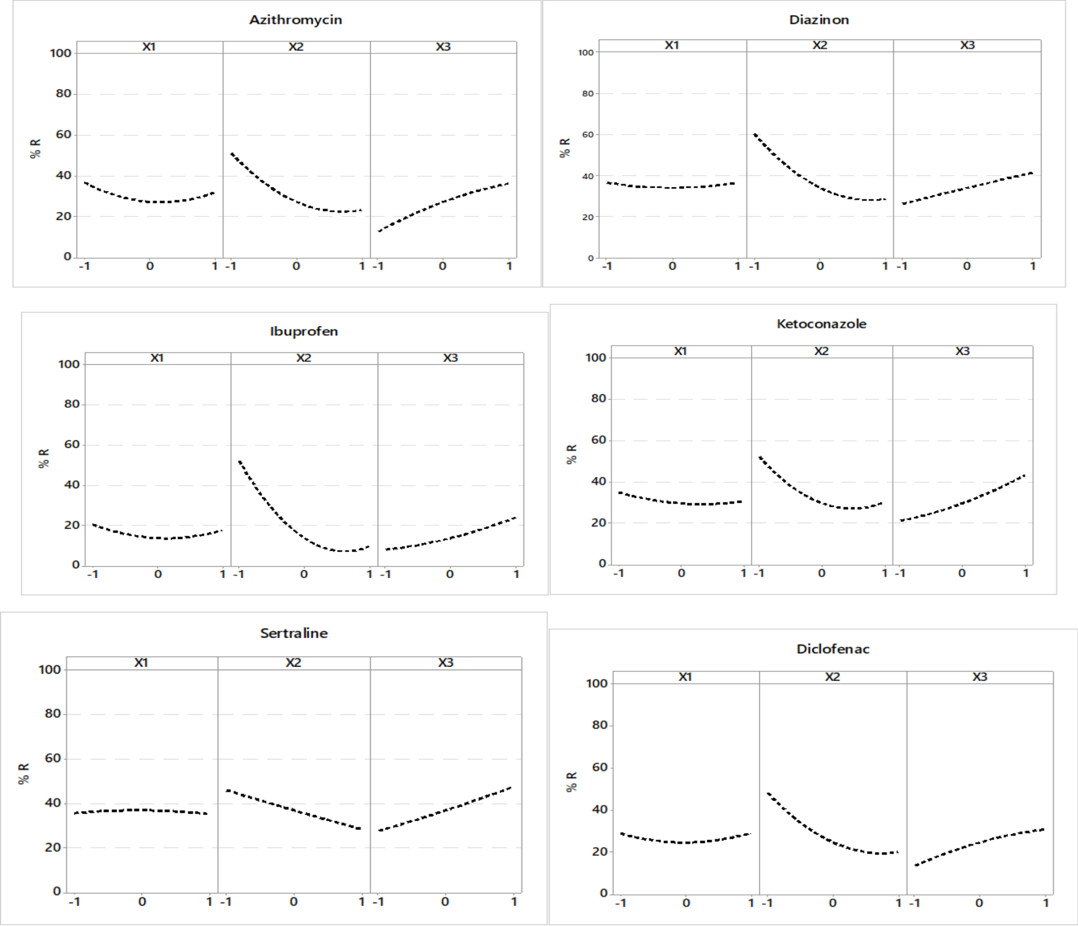

Figure 4: Main effect plots for % R of six PCs

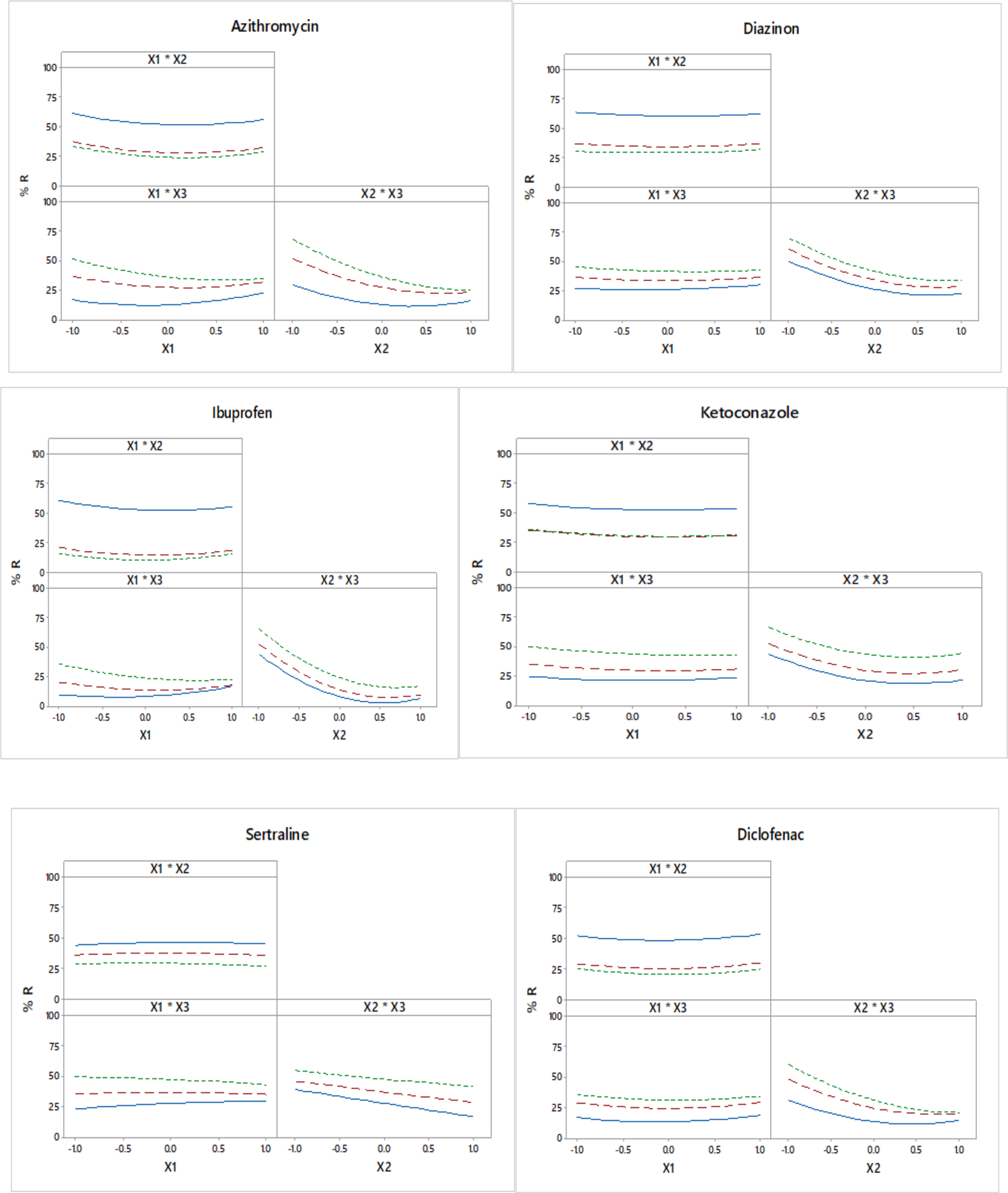

Figure 5: Interaction effect plots for % R of six PCs

Table 5: ANOVA results for the RSM

	Azithromycin			Diazinon			Ibuprofen			Ketoconazole			Sertraline			Diclofenac		
	Effect	Coef	P-Value	Effect	Coef	P-Value	Effect	Coef	P-Value	Effect	Coef	P-Value	Effect	Coef	P-Value	Effect	Coef	P-Value
Constant	27.52	3.38		34.2	1.71		14.01	2.03		29.91	2.61		37.16	2.58		24.88	2.58	
X1	-5.02	-2.51	0.279	-0.04	-0.02	0.985	-2.84	-1.42	0.305	-4.14	-2.07	0.252	-0.39	-0.19	0.907	0.36	0.18	0.913
X2	-27.84	-13.92	0.001	-31.61	-15.81	0.000	-42.58	-21.29	0.000	-21.99	-11	0.001	-17.56	-8.78	0.003	-27.97	-13.99	0.000
X3	23.48	11.74	0.002	15.22	7.61	0.001	16.04	8.02	0.001	22.24	11.12	0.001	19.91	9.95	0.001	17.37	8.69	0.003
X1xX1	13.99	6.99	0.070	4.93	2.46	0.171	10.64	5.32	0.034	6.24	3.12	0.242	-2.96	-1.48	0.553	8.78	4.39	0.118
X2xX2	20.07	10.04	0.022	20.75	10.38	0.001	34.33	17.17	0.000	23.16	11.58	0.004	0.56	0.28	0.909	18.94	9.47	0.010
X3xX3	-5.74	-2.87	0.390	-0.55	-0.28	0.865	4.48	2.24	0.276	5.11	2.56	0.327	1.35	0.67	0.784	-4.89	-2.45	0.341
X1xX2	-0.22	-0.11	0.971	1.39	0.7	0.659	2.68	1.34	0.480	-0.25	-0.12	0.959	-1.44	-0.72	0.760	-1	-0.5	0.832
X1xX3	-11.09	-5.54	0.117	-2.69	-1.35	0.405	-9.64	-4.82	0.041	-3.26	-1.63	0.504	-6.4	-3.2	0.212	-1.85	-0.93	0.696
X2xX3	-14.73	-7.37	0.053	-4.26	-2.13	0.211	-5.37	-2.68	0.188	-0.13	-0.06	0.979	4.44	2.22	0.366	-11.49	-5.75	0.050

The ANOVA analysis for the EC process, as summarized in Table 5, reveals critical insights into the factors influencing the removal efficiency of the six PC). The constant values indicate the baseline removal efficiency when all independent variables are set to zero. These values vary among the different PCs, reflecting their inherent differences in removal efficiency in the EC process.

The effect of temperature on the removal efficiency shows a negative coefficient across all PCs. However, the P-values indicate that this effect is not statistically significant ($P > 0.05$) for most compounds. This suggests that while temperature variations may influence the process, they do not significantly affect removal efficiency within the tested range. The lack of significance may imply that the EC process is relatively robust against temperature changes.

The pH exhibits a highly significant negative effect on removal efficiency for all PCs, with P-values < 0.001. This indicates a strong inverse relationship, where increases in pH lead to decreased removal efficiency. The current density shows a consistently positive and statistically significant effect on removal efficiency ($P < 0.05$ for all PCs). This suggests that higher current densities enhance the removal efficiency, likely due to increased production of coagulant species and improved floc formation during the electrocoagulation process.

The quadratic term for temperature ($X1^2$) shows marginal significance for some compounds ($P = 0.070$). This suggests a potential non-linear relationship where extreme temperature values may negatively influence removal efficiency, although this effect requires further investigation to confirm its practical relevance. The quadratic effect of pH ($X2^2$) is significant for most compounds ($P < 0.05$), indicating that the relationship between pH and removal efficiency is not linear. This reinforces the finding that pH adjustments can substantially impact removal efficiency, emphasizing the importance of optimizing this parameter in the EC process. The quadratic term for current density ($X3^2$) does not exhibit significance ($P > 0.05$), suggesting that the effect of current density on removal efficiency is linear within the tested range.

The interaction between temperature and pH shows no significant effect ($P > 0.05$) for any compound. This indicates that temperature does not influence the effect of pH on removal efficiency, suggesting independent effects of these parameters. The interaction between temperature and current density is marginally significant for some compounds, indicating that the effect of temperature on removal efficiency may slightly depend on the current density level, though this relationship is not strong. The interaction between pH and current density shows a marginal significance ($P \approx 0.053$ for some compounds). This suggests that while these factors predominantly affect removal efficiency independently, there may be scenarios where their combined effects warrant further exploration.

4. CONCLUSIONS

This study successfully employed response surface methodology to investigate the electrocoagulation process for the removal of six pharmaceutical compounds from wastewater. The experimental results revealed significant differences in removal efficiencies across various operational conditions, with run 11 demonstrating the highest overall removal percentages for all compounds tested.

The optimal conditions for maximizing removal efficiency were identified as a specific combination of temperature, pH, and current density. The conditions of run 11' proved to be the most effective, achieving maximum removal rates of 65.27% for Azithromycin, 70.09% for Diazinon, and notable efficiencies for other compounds.

The analysis indicated that while current density positively influenced removal efficiency, temperature, and pH exhibited negative impacts. Higher current densities enhanced the generation of coagulants, thus improving the removal of contaminants. In contrast, the negative effects associated with pH suggest the need for careful monitoring and adjustment during the EC process to achieve optimal results.

Utilizing RSM facilitated a comprehensive understanding of the interactions between the selected parameters, allowing for an effective optimization of the EC process. The statistical analysis provided insights into the main effects and interactions, aiding in the identification of optimal conditions for the removal of pharmaceuticals from wastewater.

The research contributes valuable knowledge to the field of wastewater treatment, particularly regarding the electrocoagulation of pharmaceutical contaminants. The insights gained from this study can guide future investigations and practical implementations, ultimately enhancing the efficiency of EC systems in addressing the challenges posed by pharmaceutical pollutants in wastewater. Further studies are recommended to explore the scalability of these findings and to investigate the long-term performance and sustainability of the electrocoagulation process in real-world applications.

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DECLARATION OF GENERATIVE AI IN SCIENTIFIC WRITING

During the preparation of this work, the author used GPT in order to improve the readability and the language of the manuscript. After using this tool, the author reviewed and edited the content as needed and took full responsibility for the content of the publication.

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