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Investigation of Electrocoagulation Process for Efficient Removal of Bisphenol A from the Aqueous Environment: Promising Treatment Strategy

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ABSTRACT

Introduction: Endocrine disruptive compounds as a class of organic contaminants in the aquatic environment received severe attention in the last decades. The release of bisphenol A (BPA) as a hazardous organic chemical into the environment has caused high health and environmental concerns. Therefore, its removal from aquatic environments is strongly recommended. The present study deals with BPA removal efficiency from an aqueous environment using the electrocoagulation process (ECP).

Materials and Methods: The effects of parameters including BPA concentration (1-10 mg L^{-1}), current density (3-15 mA cm^{-2}), pH (4-10), and reaction time (5-30 min) on the treatment process were investigated. Response surface methodology (RSM) was employed for optimization of the ECP. The significance of the developed model was investigated by the obtained F-value and P-value.

Results: The maximum BPA removal of 98.2% was attained at pH of 8.5, BPA concentration of 3.25 mg L^{-1}, the current density of 12.0 mA cm^{-2}, and reaction time of 23 min. The significance of the developed model was confirmed by the high F-value of 46.69 and the very low P-value of < 0.0001. Furthermore, the electrical energy consumption of the process was found to be 0.308 kWh m^{-3} in the optimum condition.

Conclusion: The obtained experimental results revealed that the co-precipitation and the adsorption process through the electrostatic interactions as the main removal mechanisms controlled the treatment process.

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Introduction

Recently, new chemical compounds have entered the environment through industrial, agricultural, and other human activities, which caused disruption of the endocrine and subsequently several emerging diseases in human societies. Bisphenol A (BPA) is a well-known one of the emerging pollutants produced and consumed in large volumes annually (38×10^5 tons per year). BPA is used in various industries to produce polysulfones, polycarbonates, plastic precursors, and epoxy resins [1-3]. The US environmental

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protection agency (EPA) and the European Chemical Agency (ECHA) have identified BPA as a hazardous compound with a very high-risk threat to human health and the environment [1].

The excessive use of BPA in industrial activities has led to its widespread distribution in various environments, including soil, water, air, sediments, human tissues, and others [4, 5]. Some studies have reported the amount of BPA in the river waters and coastal waters (≥ 1 μg L^{-1}), stored water (915–1415 μg L^{-1}), groundwater (96.5–170 μg L^{-1}), and landfill leachates (≥ 17 mg L^{-1}) [6, 7]. Studies have revealed that BPA may cause some diseases and disturb hormone function, such as increased sexual dysfunction, sperm count reduction, premature puberty, obesity prevalence, diabetes, brain function disorder, immunodeficiency, hypothyroidism, and hyperthyroidism, breast and prostate cancer, and others. [8-10].

Due to the widespread use of BPA in industrial activities and its essential role in the global market, it seems impossible to prevent its production. Therefore, one of the approaches to prevent and reduce the effects of BPA on human's health and the environment is to effectively treat BPA-contaminated wastewater before discharging it to the environment [4]. During the ECP, coagulation agents are generated by connecting the electric current between the anode and cathode electrodes (iron or aluminum) in electrolytic cell [11, 12]. The coagulation agents (metal hydroxides) generated during the ECP in the electrolytic cell, through flotation and precipitation, removed pollutants. Hydrolysis, electrolysis (formation of the coagulant), and ionization are the relevant reactions that pollutants can experience at this stage [13, 14]. It is recognized that three stages are involved in ECP formation of the coagulant by oxidation of the metal at the anode, destabilization of pollutants and emulsions, and finally, forming of flocs by either aggregation of contaminant particles or adsorption of the contaminant on the coagulant [15, 16].

ECP has extensively been employed due to its efficient treatment of various types of wastewater, including wastewater containing dyes [17], heavy metals [18], and pharmaceutical compounds [19, 20]. The ECP has several advantages, including simple operation, less waste sludge, low use of chemicals, short purification time, low capital costs, and considerable efficiency in wastewater treatment [21]. In the current work, great attempts were made to develop an efficient treatment procedure. The effects of various parameters, including BPA initial concentration, the current density, pH, and reaction time, on the BPA removal efficiency, were investigated.

Materials and Methods

Bisphenol A ($(CH_3)_2C(C_6H_4OH)_2$) was obtained from Sigma-Aldrich. Sodium hydroxide (NaOH), potassium chloride (KCl), hydrochloric acid (HCl), and HPLC grade methanol (CH_3OH) were obtained from Merck. All solutions were prepared with double-distilled water.

Treatment procedure

Plexiglas reactor with a useful volume of 250 mL and two aluminum electrodes with immersed dimensions of 4 cm × 2 cm × 1 mm were employed. By applying the direct current (Megatek power supply 30D5), the desired current density was adjusted. The constant electrode inter distance of 2.5 cm and 50 mM KCl electrolyte were considered during the treatment process. The pH of the samples was adjusted using NaOH and HCl. The residual of BPA in the treated samples was evaluated using HPLC system (Knauer Smartline) by C_{18} column (250 mm × 4.6 mm × 5.0 μm) and employing isocratic elution of methanol and water (30:70) with a flow rate of 1.2 mL min^{-1}, the temperature of 30 °C, and wavelength set at 276 nm. The removal of the BPA and the electrical energy consumption were calculated using Eq. 1 and Eq. 2, respectively [22-24]:

$$\text{BPA Removal (\%)} = \frac{C_I - C_F}{C_I} \times 100 \quad (1)$$

$$\text{Electrical energy consumption} = \frac{UIT}{V} \quad (2)$$

Where C_I and C_F (mg L^{-1}) denote the BPA concentration before and after the ECP, respectively. U, I, t, and V denote the applied voltage (V), electrical current (A), reaction time (h), and the volume of sample (L), respectively.

Process variables and experimental design

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The design of the treatment process was performed by design expert software. Response surface methodology (RSM) was employed to evaluate the association between the variables. The BPA concentration (X_1), current density (X_2), pH (X_3), and reaction time (X_4) were considered as the independent variables. Also, the removal efficiency of BPA was considered as the response of the developed model. The level of -α, -1, 0, + 1, and + α for all variables was demonstrated in table 1.

Table 1: Level of independent variables for the BPA removal

Variables (X_i)	Level				
	-α	**-1**	**0**	**+1**	**+α**
(A), X_1 = BPA concentration (mg L^{-1})	1.0	3.2	5.5	7.7	10.0
(B), X_2 = Current density (mA cm^{-2})	3.0	6.0	9.0	12.0	15.0
(C), X_3 = Solution pH	4.0	5.5	7.0	8.5	10.0
(D), X_4 = Reaction time (min)	5.0	11.2	17.5	23.7	30.0

The polynomial regression equation of Eq. 3 was employed to predict the removal efficiency of contaminant by considering the input variables [23, 24].

$$Y = \beta_0 + \sum_{i=1}^{n} \beta_i X_i + \sum_{i=1}^{n} \beta_{ii} X_i^2 + \sum_{i=1}^{n-1} \sum_{j=i+1}^{n} \beta_{ij} X_i X_j + \varepsilon \quad (3)$$

Where *Y* denotes the predicted response of contaminant removal efficiency. The parameters of X_i and X_j denote the coded values of independent factors. The coefficient parameters of β_0 (the intercept parameter), β_i (the linear coefficients), β_{ii} (the quadratic coefficients), and β_{ij} (the interaction coefficients) expressed in the mentioned equation. The parameters of n and ε denote the investigated independent variables and the experimental error, respectively.

Ethical issue

The current work was conducted in the spring of 2018, after receiving approval from the ethics committee of Kerman University of Medical Sciences [IR.KMU.REC.1397.385].

Results

Fit Model Analysis and Analysis of variance (ANOVA)

According to the experimental design, 30 experiments were conducted to find out the optimum treatment condition and to understand the effect of the main operating parameters of X_1 (BPA concentration), X_2 (current density), X_3 (solution pH), and X_4 (reaction time) on the removal efficiency of BPA. The obtained results are given in table 2.

Table 2: Experimental results of BPA removal

Run	Actual value				Coded value				Experimental removal (%)	Experimental removal (%)
	A (mg L^{-1})	**B (mA cm^{-2})**	**C**	**D (min)**	**X_1**	**X_2**	**X_3**	**X_4**		
1	5.5	9	7	17	0	0	0	0	71.6	70.8
2	3.25	12	5.5	23	-1	1	-1	1	79.4	80.1
3	7.75	12	8.5	23	1	1	1	1	85.2	84.7
4	7.75	6	5.5	23	1	-1	-1	1	58.2	58.9
5	3.25	6	8.5	11	-1	-1	1	-1	76.5	75.8
6	7.75	6	5.5	11	1	-1	-1	-1	54.7	54.5
7	5.5	3	7	17	0	-2	0	0	64.8	64.1
8	7.75	6	8.5	11	1	-1	1	-1	66.1	66.7
9	5.5	9	10	17	0	0	2	0	84.2	85.1
10	7.75	12	8.5	11	1	1	1	-1	75.8	76.3
11	5.5	9	7	17	0	0	0	0	71.1	70.5
12	5.5	15	7	17	0	2	0	0	83.4	83.7
13	1	9	7	17	-2	0	0	0	89.7	88.3
14	3.25	12	8.5	23	-1	1	1	1	94.3	94.8
15	3.25	6	5.5	23	-1	-1	-1	1	65.2	65.1

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Run	Actual value				Coded value				Experimental removal (%)	Experimental removal (%)
	A (mg L^{-1})	B (mA cm^{-2})	C	D (min)	X_1	X_2	X_3	X_4		
16	10	9	7	17	2	0	0	0	60.1	60.5
17	5.5	9	4	17	0	0	-2	0	56.1	55.4
18	3.25	12	8.5	11	-1	1	1	-1	75.7	57.1
19	3.25	6	5.5	11	-1	-1	-1	-1	65.0	65.7
20	7.75	6	8.5	23	1	-1	1	1	71.9	72.4
21	5.5	9	7	5	0	0	0	-2	47.2	47.8
22	3.25	12	5.5	11	-1	1	-1	-1	69.2	69.7
23	7.75	12	5.5	23	1	1	-1	1	69.1	69.4
24	5.5	9	7	30	0	0	0	2	67.5	67.1
25	3.25	6	8.5	23	-1	-1	1	1	84.5	84.0
26	7.75	12	5.5	11	1	1	-1	-1	61.3	61.9
27	5.5	9	7	17	0	0	0	0	72.1	72.5
28	5.5	9	7	17	0	0	0	0	71.4	72.0
29	5.5	9	7	17	0	0	0	0	72.8	71.6
30	5.5	9	7	17	0	0	0	0	71.2	71.6

Based on the performed runs, the following model was developed by RSM as a function of main operating parameters within the designated range.

$$\text{Removal BPA}(\%) = 74.08 - 5.28X_1 + 4.38X_2 + 6.84X_3 + 4.34\,X_4 + 1.78X_2X_4 - 3.48X_4^2 \quad (4)$$

In this equation, *Y* is the removal rate of BPA (%), and X_1 to X_4 represents the coded independent factors of BPA concentration, the current density, pH, and reaction time, respectively. The adequacy of the proposed model was investigated using the analysis of variance (ANOVA) test and summarized in table 3.

Table 3: ANOVA results for the developed quadratic model

Source	Sum of squares	df	Mean square	F-value	p-value Prob > F
Model	3102.21	1	517.03	46.69	<0.0001
X_1	668.87	1	668.87	60.40	<0.0001
X_2	460.25	1	460.25	41.56	<0.0001
X_3	1122.03	1	1122.03	101.33	<0.0001
X_4	451.53	1	451.53	40.78	<0.0001
X_5	50.77	1	50.77	4.58	<0.0001
$X_2\,X_4$	348.75	23	348.75	31.50	0.0431
X_4^2	668.87	18	11.07	46.69	<0.0001
Residual	254.68	5	13.25	-	-
Lack of Fit	238.58	29	3.22	4.12	0.0621
Pure Error	16.10	1	517.03	-	-
Cor Total	3356.89	1	-	-	-
$R^2 = 0.9741$	Adjusted $R^2 = 0.9543$		Predicted $R^2 = 0.9022$	Adequate precision = 25.96	

The correlation between the predicted values versus the experimental values of BPA removal efficiency and the normal probability versus the externally studentized residuals of the developed model was demonstrated in figure 1 The observed satisfactory agreement between the actual and the predicted values confirmed the normality of the obtained results and the applicability of the developed polynomial model.

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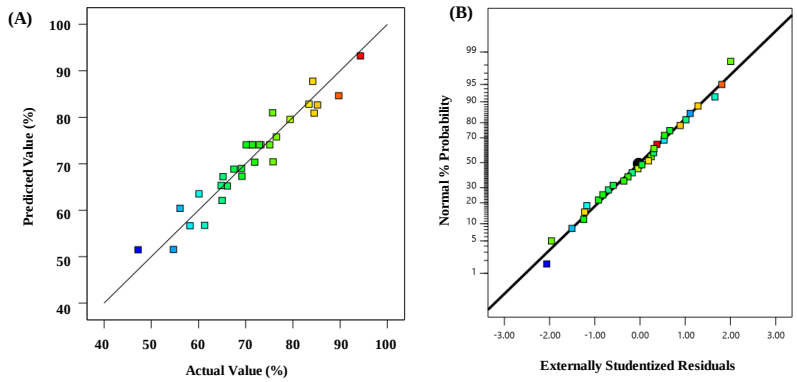


Figure 1: (A) Predicted values versus actual values and (B) normal probability plot of studentized residuals' model for the proposed model

The effect of investigated parameters on the BPA removal

The obtained results showed that the initial BPA concentration had a negative effect on the removal of BPA so that by increasing the initial BPA concentration from 1 to 10 mg L^{-1}, the removal efficiency of BPA was decreased from 84.6 to 63.5, where other variables, including the current density, pH, and reaction time kept constant at their central points of 9.0 mA cm^{-2}, 7.0, 17 min, respectively.

According to the results, the current density revealed a positive effect on the BPA removal efficiency. Accordingly, under the constant condition of central points for other variables, including the initial BPA concentration of 5.5 mg L^{-1}, pH of 7.0, and reaction time of 17 min, the current density of 3 mA cm^{-2} resulted in the removal efficiency of 65.1%. While by increasing the current density to the value of 15 mA cm^{-2}, the removal efficiency of 82.3% was achieved. The contour plot and 3D plot of BPA removal efficiency as a function of BPA concentration and current density were demonstrated in figure 2.

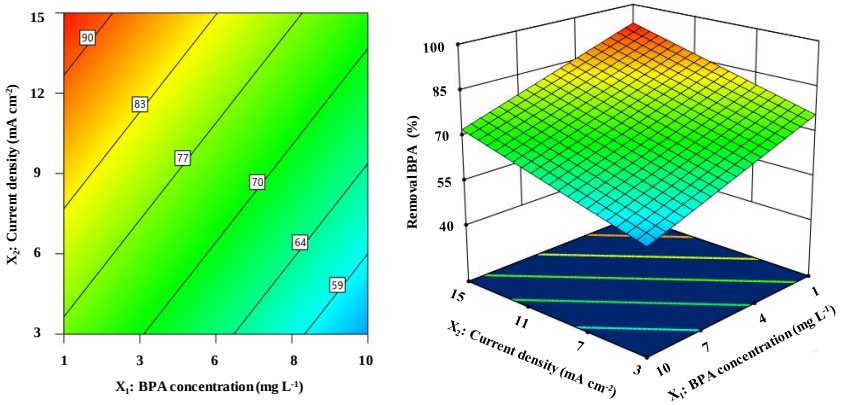


Figure 2: Contour and 3D surface plot for the combined effect of BPA concentration and current density

Figure 3 demonstrates the 3D plot of BPA removal efficiency as a function of BPA concentration and pH. As seen, the pH solution showed a significant effect on the removal efficiency of BPA, so that, if other parameters were kept constant at their center points BPA concentration of 5.5 mg L^{-1}, current density of 9.0 mA cm^{-2}, and the reaction time of 17 min), by increasing the pH level from –α to +α (4 to 10), the removal efficiency of BPA increased from 60.1% to 77.8%.

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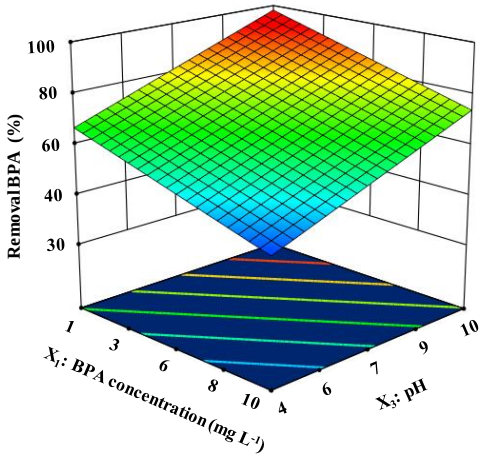


Figure 3: 3D surface plot for the combined effect of BPA concentration and pH.

Figure 4 illustrates the contour and 3D plot of BPA removal efficiency as a function of the reaction time and the current density. As seen, the reaction time revealed a significant effect on the BPA removal efficiency. If other operating parameters were fixed at their center points, including BPA concentration of 5.5 mg L^{-1}, the current density of 9.0 mA cm^{-2}, and pH of 7, the removal efficiency was increased from 51.4% to 69.0%, while the reaction time increased from –α (5 min) to +α (30 min). Moreover, according to the quadratic effect of reaction time (X_4^2), by increasing the reaction time from 5 min to 23 min, the removal efficiency of BPA was improved from 20% to 73%. However, by a further increase in reaction time from 23 min onwards, the removal efficiency was decreased to 61.1%.

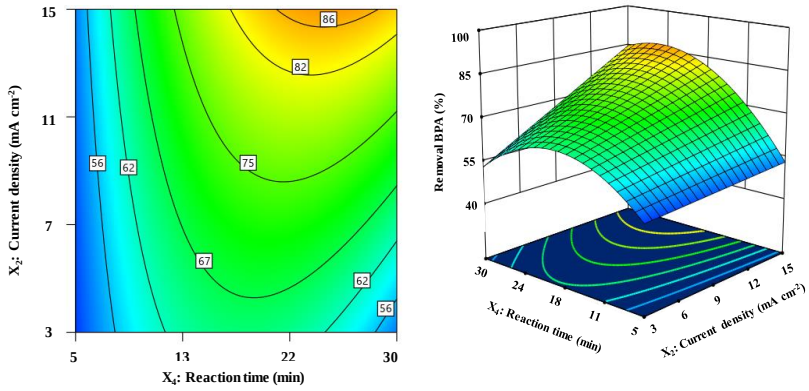


Figure 4: Contour and 3D surface plot for the combined effect of current density and reaction time Optimization process

RSM provides a fast, is cost-effective, and efficient treatment process by optimizing the values of operating variables to the desired level. The level of all main parameters was set on the maximum desirability within the investigated range. The best operating condition with a satisfactory removal efficiency of 98.2% was achieved under the BPA concentration of 3.25 mg L^{-1}, the current density of 12.0 mA cm^{-2}, pH of 8.5, and reaction time of 23 min. The electrical energy consumption (EEC) under optimum conditions for the treatment process was found to be 0.308 kWh m^{-3}.

Discussion

Statistical analysis

The obtained results confirmed that the proposed model was significant at a confidence

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level of 95%. The correlation coefficient between the predicted and actual values of BPA removal efficiency was computed to be 0.9741, implying that the model could not explain only 2.6% of the total variance in the response. Moreover, the observed variation of less than 0.20 between the adjusted R^2 (0.9543) and the predicted R^2 (0.9022) indicates the significance of the model. The obtained value of 25.96 for adequate precision represents a desirable signal-to-noise ratio.

Effect of independent variables

According to the obtained results, increasing the initial BPA concentration revealed a diverse effect on the BPA removal efficiency. It could be attributed to the constant amount of adsorption sites provided by the constant quantity mass of produced coagulant agents. The generated flocs at higher concentrations of BPA were not enough to adsorb and coagulate all the BPA molecules. Therefore, BPA removal efficiency was decreased [25, 26]. The amount of generated coagulant agents as the key parameter of ECP is directly related to the reaction time and current density.

It is confirmed that the current density strongly affected the coagulant agents and gas bubble generation rate and floc distribution [27, 28]. The higher removal efficiency at higher current density was likely because of the more intensive release of aluminum cations from the anode surface, which resulted in the formation of a greater amount of insoluble form of aluminum precipitates [29-31]. Moreover, based on Faraday's law, reaction time affected the number of aluminum cations released into the treatment reactor.

Furthermore, one more important parameter that was affected the removal of BPA was the pH level of the treated samples. It was reported that the pH of the treated samples could be decreased or increased due to the reactions involved in the ECP [32, 33]. In the current work, the pH of the treatment condition was investigated in the range of 4 to 10. The obtained results confirmed that the pH of the sample revealed a significant effect on the BPA removal. Reaction (1) and (2) describes the release of aluminum ions from the anode electrode (oxidation reaction), and hydrolysis of H_2O molecules, and production of hydrogen gas from the cathode electrode (reduction reaction), respectively. The released aluminum ions from the anode electrode convert to aluminum hydroxide (main coagulants agents) throughout a series of reactions with the H_2O and the generated products of its hydrolysis, as shown in reaction (3) [15, 28].

$$Al_{(s)} \leftrightarrow Al^{+3}_{(aq)} + 3e^- \quad \text{(Anode)} \quad (1)$$

$$2H_2O + 2e^- \leftrightarrow H_{2(g)} + 2OH^-_{(aq)} \quad \text{(Cathode)} \quad (2)$$

$$2Al_{(aq)} + 6H_2O \leftrightarrow 2Al(OH)_{3(s)} + 3H_{2(g)} \quad \text{(Overall reaction)} \quad (3)$$

The dominant mechanisms of BPA removal are attributed to co-precipitation, adsorption, or concurrent co-precipitation and adsorption processes. The generated complexes of aluminum polyhydroxides in the alkaline pH range of 8 to 10 revealed efficient behavior as the coagulant agents. The aluminum ions released from the anode electrode during the ECP could be converted into the low soluble form of $Al(OH)_3$, which is finally polymerized in $Al_n(OH)_{3n}$ forms and resulted in the formation of dense flocs.

Conclusion

The current work revealed that ECP successfully eliminated the BPA contaminant from the aqueous environments. The obtained lack of fit of 0.0620 confirmed that the model was significant, and the obtained experimental results were accurate and reliable. The impact of the variables and their interactions were evaluated by ANOVA. The obtained results confirmed that BPA initial concentration and solution pH revealed a greater influence on the removal efficiency of the process. The optimum condition for BPA removal of 98.2% was attained at pH of 8.5, BPA concentration of 3.25 mg L^{-1}, the current density of 12.0 mA cm^{-2}, and reaction time of 23 min. The electrical energy consumption of the ECP was found to be 0.308 kWh m^{-3}.

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Conflict of interest

The authors declare that they have no conflict of interest regarding the publication of the current paper.

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