# Investigation of bisphenol A removal using peroxy electrocoagulation method

Füsun Boysan\*, Ayşe Çavunt

*Environmental Engineering, Sakarya University, Sakarya, Turkey, Tel. +90 264 295 5640;*
*emails: fboysan@sakarya.edu.tr (F. Boysan), a.kuzgun@hotmail.com (A. Çavunt)*

## ABSTRACT

Bisphenol A (BPA) is an endocrine-disrupting compound, which is encountered in many areas of our daily life (feeding bottles, automotive parts, etc.). BPA causes a severe health problem and is transmitted to the environment through surface water, groundwater, industrial wastewater, leakage in BPA depots, etc. In this study, the degradation of BPA in synthetically prepared wastewater was investigated by using the peroxy electrocoagulation method with iron electrodes. The effects of pH, amount of hydrogen peroxide, current density and time parameters were investigated on BPA removal. The results of experimental studies indicated that the optimum conditions were 45 ppm (mg/L), 0.1 g/L $Na_2SO_4$, 125 mg/L $H_2O_2$, pH: 2, the current density of 0.11 mA/cm$^2$ and 45 min. Under optimum conditions, BPA removal efficiency was achieved as 80.48%. BPA effluent concentrations were calculated according to the pseudo-second-order kinetic model and the reaction rate constant ($k$) and regression coefficient ($R^2$) in kinetic evaluation demonstrated as 0.0831 and 0.9992, respectively.

*Keywords:* Bisphenol A; Hydrogen peroxide; Iron electrode; Peroxy electrocoagulation

## 1. Introduction

Natural or synthetic chemical compounds that damage the endocrine system by imitating or blocking the endocrine hormones are called endocrine-disruptors (EDCs) [1]. Bisphenols are a quite large group of organic synthetic substances with two hydroxyphenyl functions [2]. Bisphenol A (BPA) is confirmed as an endocrine disruptor with estrogenic activity in many studies. BPA is one of the widely produced and used synthetic endocrine disruptors worldwide, which is used as a raw material for many products such as electronic equipment, food containers, water-pipes, thermal papers, medical products, pacifiers, and toys [3].

The negative effects of BPA on reproductive, nervous, cardiovascular, metabolic, and immune systems, have been proven in the literature both in in-vitro experiments and laboratory animal studies [4–12]. BPA was originally synthesized as a synthetic estrogen, acting as an endocrine disruptor by mimicking estrogen. Some studies have shown that BPA binds not only to estrogen receptors, but also to androgen and thyroid receptors [13].

Despite many studies involving the negative effects of BPA on human health, BPA is still one of the most produced chemical compounds in the world, with 2.2 million tons in 2002 and an estimated 5.5 million tons in 2011 [13].

Over the last 10 y, expended financial resources and academic interests have substantially increased the determination about the possible risks of EDCs in food and drinking water on human health [14]. The main sources of BPA contamination in waters and wastewater are a result of production wastes, leakage of BPA tanks and accidents during transportation [15].

The treatment of BPA contamination in water and wastewater is essential in terms of human health and the environment. BPA can mostly be degraded by microorganisms in conventional wastewater treatment plants [1,16]. Complete elimination of BPA is tough with the conventional treatment systems, this unavoidable situation causes the

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\* Corresponding author.

presence of BPA with a low concentration in surface-waters [17]. Therefore, advanced treatment methods (advance oxidation processes, nanofiltration, adsorption, membrane bioreactor) are required. Depending on the situation, these advanced treatment methods can either be applied individually or in conjunction.

Examples of chemical (advance oxidation) [18–21], physicochemical (adsorption and nanofiltration) [1,17,22,23] and biological [24,25] studies are present in existing literature for BPA treatment.

Peroxy electrocoagulation (PEC) is one of the indirect electrochemical advanced oxidation processes which includes both ferrous ions and hydrogen peroxide [26]. In this process, organic pollutants are degraded with the combination of both electro-Fenton and electrocoagulation [27]. During the process, while $Fe^{2+}$ ion formation occurs at the anode; $OH^-$ ions are produced at the cathode. As a result of the reaction between $Fe^{2+}$ and $H_2O_2$, hydroxyl radicals [$^\bullet OH$] (Fenton reagents) are produced, this is shown in Eq. (1) [28]. In this method, all organic matters are broken down generated by hydroxyl radicals [$^\bullet OH$] until mineralization without exception in the waters [29].

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + {}^\bullet OH \qquad (1)$$

Different wastewaters such as detergent water [30], slaughterhouse wastewater [31], leachate [32], oil mill wastewater [33], aniline containing wastewater [34] and textile wastewater included synthetic dyes [35,36] are investigated with peroxy electrocoagulation treatment in many studies, but there is no study in the literature with this method we used on BPA removal. Therefore, the method used in the extent of this study is an innovative method for BPA purification.

The aim of this study is to demonstrate the treatment of BPA-contained wastewater by the peroxy electrocoagulation method. Peroxy electrocoagulation is a method based on the generation of Fenton reagents in the process with the external addition of $H_2O_2$ to the electrocoagulation system. The current study examined that the treatment efficiency of the peroxy electrocoagulation method by using a synthetically prepared 45 mg/L BPA solution with the parameters of pH, hydrogen peroxide dose, current density, and reaction time.

## 2. Materials and methods

### 2.1. Peroxicoagulation process

Peroxicoagulation is a modified electro-Fenton process in which instead of adding extra iron for the production of $Fe^{2+}$, an iron anode is used. These ions react with the $H_2O_2$ formed and form hydroxyl radicals as in the EF process. $Fe(OH)_3$ precipitates are formed due to the increase in the concentration of $Fe^{3+}$ ions in the electrolytic system over time [26]. Pollutants are expected to be removed by the combined effect of homogeneous degradation with hydroxyl radicals and coagulation with the $Fe(OH)_3$ precipitate [34].

Although electrocoagulation uses an iron anode to remove contaminants, it differs from peroxicoagulation by a contaminant removal mechanism. In the traditional electrocoagulation process, contaminants are removed by the separation process, not by the degradation process. However, in the peroxicoagulation process, most of the contaminants are removed by decomposition [26].

The principle of the electrocoagulation process in removing pollutants is based on one or more of the coagulation, adsorption, precipitation and flotation removal mechanisms. Metal ions dissolved from these electrodes form metal-polymer complexes according to the ambient conditions and coagulate by adsorbing the pollutant [37]. Compared to the usual coagulation–flocculation applications, electrocoagulation has the advantage of removing the smallest colloid particles due to the presence of the electrical field [38]. In addition, the sludge formed mainly contains metal oxides or hydroxides and exhibits a structure that can be easily settled and dewatered. The materials can be aluminum or iron in sheet form. Aluminum is generally used in water treatment and iron is used in wastewater treatment. Electrochemical methods are simple, fast, cheap, easily applicable and environmentally friendly in nature. Also, the purified water is drinkable, clear, colorless and odorless with low sludge production. There is no possibility of secondary pollution of water in these techniques [34].

### 2.2. Chemicals

Bisphenol A (97%) was purchased from Acros Organics (Belgium). Sodium sulfate ($Na_2SO_4$), sodium hydroxide (NaOH), hydrochloric acid (HCl) and hydrogen peroxide ($H_2O_2$) was supplied from Merck KGaA (Germany). In the peroxy electrocoagulation process, conductivity was achieved with $Na_2SO_4$ salt as the electrolyte. NaOH was used to stop the reaction in the solution taken into the tubes. Also, HCl and NaOH solutions were used to adjust pH values in the samples. $H_2O_2$ (35%) was used as the peroxy electrocoagulation reagent. All the chemicals used in the study were of analytical grade.

### 2.3. Instruments

BPA concentration analyses were conducted using the Spectroquant Pharo 300 model spectrophotometer from Merck. WTW InoLab pH 720 pH-meter was used for pH measurements. All weighing operations were made using a GR-200 model analytical precision balance from AND.

The pure water, used in the preparation of the samples was obtained from the ultrapure water device (Merck). Magnetic stirrer (BOECO, MSH30), DC power supply (Yıldırım Elektronik Y-0012) and Nüve NF 200 centrifuge were also used for all other applications.

### 2.4. Tools and equipment

A 250 mL glass container was used as a reactor for the peroxy electrocoagulation process as seen in Fig. 1. For the peroxy electrocoagulation method, 4 iron plates of 10 cm × 4 cm × 0.1 cm in size were used as anodes and cathodes and the distance between the plates was 1.7 cm. The depth of the electrodes inside the reactor is 5.5 cm. The total active area used for 4 electrodes was calculated as 182 cm².
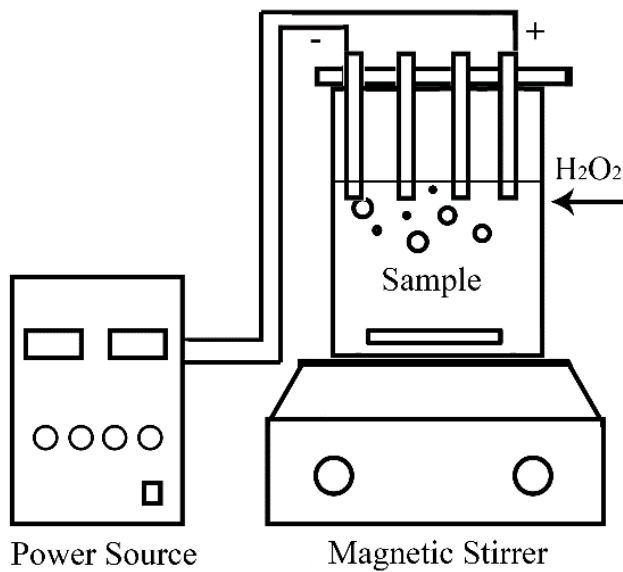
Fig. 1. Schematic diagram of peroxy electrocoagulation.



Fig. 2. Effect of pH on BPA removal efficiency over time.

### 2.5. Preparation and measurement of BPA samples

To find the wavelength of the BPA solution, a spectrum scan was performed on a sample with a concentration of 45 mg/L. According to the result of the spectrum scan, it was determined that BPA amount can be measured at 278 nm, this is in accordance with some literature studies where 278 nm wavelength was used to measure BPA [18,19].

To measure BPA removal in treatment with the peroxy electrocoagulation method, a 45 mg/L stock BPA solution was prepared and a calibration curve was prepared with samples of 5, 15, 25, 35 and 45 mg/L concentrations. In the calibration process, the regression coefficient ($R^2$) was found to be 0.9992 and the linear equation obtained is given in Eq. (2).

$$C = 65.869\,ABS + 2.4597 \tag{2}$$

## 3. Result and discussion

### 3.1. Effect of pH on BPA removal

In the peroxy electrocoagulation process under acidic conditions (2 < pH < 5), it was observed that $^\bullet$OH radicals were formed by catalytic decomposition from hydrogen $H_2O_2$ in the medium where $Fe^{2+}$ ions and organic pollutants were present, therefore, it is an important parameter affecting pH reactions [39–41].

In the study, the initial concentration of BPA ($C_0$) was 45 mg/L and the amount of $H_2O_2$ added was 100 mg/L, the amount of $Na_2SO_4$ was 0.5 g/L, the current density (i) was 0.11 mA/cm$^2$ and the mixing speed was 300 rpm. The experiment was carried out at five different pH values and these were determined as 2, 3, 5, 7 and 9. BPA removal efficiencies at 20, 40 and 60 min at these pH values are given in Fig. 2.

The effect of pH on BPA removal efficiency is shown in Fig. 2. When the BPA removal efficiencies are examined for the 60 min contac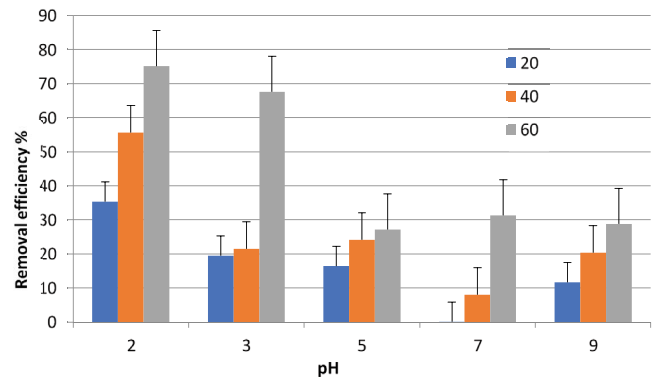t time, it can be seen that the efficiencies increase at low pH values and decrease at high pH values. The removal efficiency of pH 2 at 20, 40 and 60 min were 35.40%, 55.60% and 75.51%, respectively.

### 3.2. Effect of $H_2O_2$ amount on BPA removal

A variety of $H_2O_2$ concentrations, 10, 25, 50, 75, 100, 125 and 150 mg/L, were studied to examine the effect of $H_2O_2$ concentration in BPA removal by peroxy electrocoagulation.

The experiment conditions remained consistent, all studies were conducted with $Na_2SO_4$ amount of 0.5 g/L, current density (i) 0.11 mA/cm$^2$ and mixing speed of 300 rpm. BPA removal efficiencies are given in Fig. 3.

Fig. 3 shows the effect of $H_2O_2$ dose on BPA removal efficiency over time. According to the results of BPA removal efficiencies, while the removal efficiency at low $H_2O_2$ amount is less, it appears that as the $H_2O_2$ dose increases, the removal efficiency increases as well. While $H_2O_2$ amount is 125 mg/L, the removal efficiency in the 20th min is 78.58%, 81.07% in the 40th min and 79.16% in the 60th min. Removal efficiencies increased rapidly until the amount of $H_2O_2$ reached 125 mg/L, and after this value, the increase of removal efficiency decreased. Therefore, $H_2O_2$ amount was chosen as 125 mg/L as a result of the experiment.

### 3.3. Effect of conductivity on BPA removal

Conductivity must be present between the electrodes in order to perform the peroxy electrocoagulation process, and various electrolytes, salts soluble in aqueous medium, can be used to achieve this conductivity. In this study, sodium sulfate ($Na_2SO_4$) was used to ensure conductivity for two main reasons. First of all, it is easy to supply. Secondly, sodium sulfate does not produce by-products such as BPACl, which are more dangerous than BPA since they form a chlorinated compound with BPA.

In order to determine the optimum electrolyte dose of the initial concentration of 45 mg/L BPA solution using the peroxy electrocoagulation process, the power supply was operated at the maximum current with the reaction time of 1 min and the sodium sulfate doses of 0.1, 0.25, 0.5, 0.75 and 1 g/L as experimental conditions. The results are shown in Table 1.

When the results in Table 1 are examined, current density of 1 mA/cm$^2$ was obtained with 0.1 g/L $Na_2SO_4$,
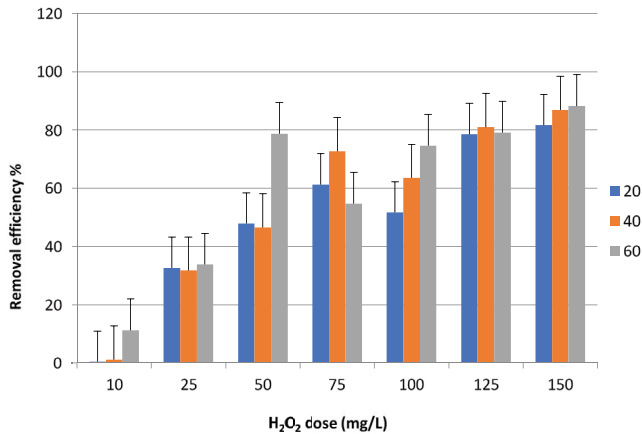
Fig. 3. Effect of $H_2O_2$ amount on BPA removal efficiency over time.

therefore it was decided that 0.1 g/L salt would be used for the current density in the study.

### 3.4. Effect of current density on BPA removal

Current density is one of the important operating parameters for BPA removal with peroxy electrocoagulation. When excessive current intensity is applied, the increase in the discharge may increase as well as the sludge formation [42]. In this study, 4 different experiments were carried out at 0.11, 0.27, 0.38 and 0.55 mA/cm². Experiments were carried out with $Na_2SO_4$ amount of with 0.1 g/L, $H_2O_2$ amount of 125 mg/L, pH value of 2 and mixing speed of 300 rpm and contact time study was examined as 20 and 40 min. The removal efficiencies obtained are shown in Fig. 4.

The effect of current density on BPA removal efficiency is shown in Fig. 4. When the BPA removal efficiencies are examined at 20 min, it can be seen that removal efficiency is highest at low current density and decreases as the current density increases. When the current density was 0.11 mA/cm², the removal efficiency after 20 min was 82.09% and after 40 min the removal efficiency was 75.94%. Thus, it was determined that the maximum removal efficiency for BPA occurred at 20 min and at a current density of 0.11 mA/cm².

### 3.5. Effect of reaction time on BPA removal

In order to determine the optimum reaction time, experiments were conducted at the initial BPA concentration ($C_0$) of 45 mg/L, $Na_2SO_4$ amount of 0.1 g/L, $H_2O_2$ amount of 125 mg/L, pH 2 and mixing speed 300 rpm. To examine the effect of contact time on BPA removal efficiency using peroxy electrocoagulation process, 7 different time values of 5, 10, 15, 20, 30, 45 and 60 min were studied. As a result of the experiments, the BPA removal efficiencies are shown in Fig. 5.

Fig. 5 shows the effect of reaction time on BPA removal. In this experiment, all parameters used for removal reaction were kept at an optimum level and it was determined that the maximum removal efficiency for BPA was 80.48% in the 45 min time interval.

Table 1
Voltage (V) and current density (mA/cm²) values over $Na_2SO_4$ concentration

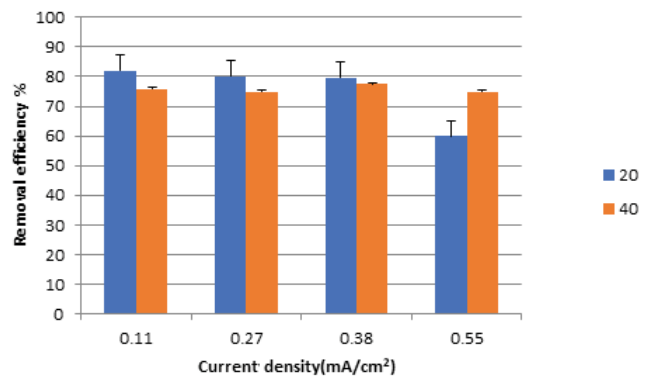| $Na_2SO_4$ (g/L) | V | mA/cm² |
|---|---|---|
| 0.1 | 27.8 | 1.09 |
| 0.25 | 28.1 | 1.20 |
| 0.5 | 28.1 | 1.48 |
| 0.75 | 28.1 | 1.75 |
| 1 | 28.1 | 2.14 |



Fig. 4. Effect of current density on BPA removal efficiency over time.
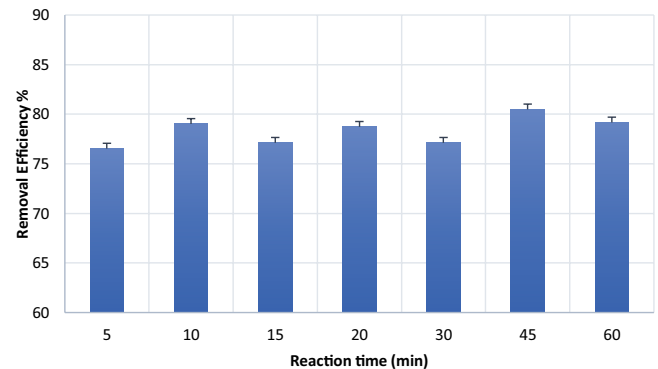


Fig. 5. Effect of reaction time on BPA removal efficiency.

Various studies have been conducted on BPA removal with the discovery of its toxic feature, and new studies are still ongoing. Since the extent of BPA's toxic effect on the environment was identified, various studies have been conducted on methods of BPA removal and new studies are emerging.

A literature review was conducted to explore existing studies and identify the different processes related to BPA removal. The findings of studies with different processes related to BPA removal are summarized in Table 2. The relevant table contains the methods used, the initial concentration of BPA, the reaction volume, pH, chemicals used, current density, reaction time and BPA removal efficiency.

In Table 2, the advantages and disadvantages of different methods are compared for the removal of BPA in an aqueous medium. Through this comparison it can be seen

Table 2
Studies on BPA removal

| Method used | BPA amount ($C_0$) (mg/L) | $V$ (mL) | pH | Chemicals | $I$ (mA/cm$^2$) | $t_r$ (min) | Efficiency (%) | Reference |
|---|---|---|---|---|---|---|---|---|
| Peroxy electro-coagulation | 45 | 200 | 2 | $H_2O_2$ (125 mg/L) $Na_2SO_4$ (0.5 g/L) | 0.11 | 45 | 80.48 | This study |
| Membrane (NF270)-Fenton | 20 | 2,000 | 7 | $H_2O_2$ (10 mg/L) Ferrous (0.4 mg/L) Horseradish (HRP) (7.5%) Lakkaz (0.12 U/mL) | NDA | 180 | 89 | [17] |
| Micro-wave-Mn-Fen-ton (300 W) | 100 | 500 | 4 | $H_2O_2$ (34 mg/L) $Fe^{2+}$ (2.1 mg/L) $Mn^{2+}$ (2.7 mg/L) | NDA | 6 | 99.7 | [19] |
| Membrane (electrochemical microfiltration) | 50 | 250 | NDA | $Na_2SO_4$ (0.1 mol/L) | NDA | 0.88 | 97 | [43] |
| UV | 10 | NDA | NDA | Hg-lamp (400 W) | NDA | 30 | 100 | [44] |
| Photo-Fenton | 10 | NDA | NDA | Hg-lamp (400 W) $H_2O_2$ (100 mg/L) | NDA | 15 | 100 | [44] |
| $O_3$ | 10 | 500 | NDA | $O_3$ (4.05 mg/min) | NDA | 10 | 100 | [45] |
| Sono-Fenton (300 kHz; 80 W) | 27 | 300 | NDA | $FeSO_4$ (100 μmol/L) $H_2O_2$ ($35 \times 10^{-3}$ mol/L) | NDA | 90 | 100 | [47] |
| Photocatalytic | 40 | 100 | NDA | $TiO_2$ (1 g/L) Hg/Xe-lamp (10 mW cm$^{-2}$) | NDA | 15 h | 99 | [48,49] |
| Electro-Fenton | 10 | 1,500 | 2.9 | $Na_2SO_4$ (0.05 M) $Fe_2SO_4$ (0.5 mM) | NDA | 50 | >90 | [50] |

$C_0$: initial concentration; $V$: reaction volume; $i$: current density; $t_r$: reaction time; sono: ultrasound; NDA: no data available.

that Microwave-Mn-Fenton analysis by [19] and membrane (electrochemical microfiltration) by [43] obtained better efficiency in a shorter time than other processes [19].

It can be considered that photo-Fenton processes [44] and ozone oxidation processes [45] are more efficient than the other BPA removal methods in aquatic environments. The major disadvantages of these methods are undoubtedly the use of chemicals that require activation of the catalysts, pH control and/or neutralization steps. In addition, the use of ozone can lead to the production of many oxidation products that may have a higher toxic potential than BPA itself and whose dangerous properties have not been clarified yet [46].

### 3.6. Kinetic examination

In this study, 3 different kinetic models for BPA removal were examined and equations of first-degree, second-degree and pseudo-second-degree kinetic models are given below.

$$\ln \frac{C_0}{C} = kt \qquad (3) \quad [51]$$

$$\frac{1}{C} - \frac{1}{C_0} = kt \qquad (4) \quad [52]$$

$$\frac{t}{C} = \frac{1}{k_2 C_e^2} + \frac{1}{C_e} t \qquad (5) \quad [53]$$

where $C_0$ = initial BPA concentration (mg/L); $C$ = final BPA concentration (mg/L); $k$ = mean mass transport coefficient (1/min); $t$ = reaction time (min); $C_e$ = concentration coefficient (mg/L); $k_2$ = rate constant (L/mg min).

As it can be seen in Table 3, the most favorable model that obtained the kinetics of the PEC process is the pseudo-second-order kinetic model, where the regression coefficient ($R^2$) is 0.99995 and the reaction rate is 0.0831.

BPA effluent concentrations were calculated according to the pseudo-second-order kinetic model. Fig. 6 shows the theoretical and experimental removal efficiencies calculated in PEC processes with pseudo-second-order kinetics. As seen from the figures, a rapid removal efficiency was obtained in the first 10 min. After 45 min, the theoretical removal efficiency and the experimental removal efficiency obtained were very close.

### 3.7. Cost analysis

The factors determining the cost in electrochemical processes can be listed as electrical energy, used electrode material and used reagents. In this study, the usage of electrical energy, iron electrode and, $Na_2SO_4$, HCl and $H_2O_2$

Table 3
$k$ and $R^2$ values of kinetic models

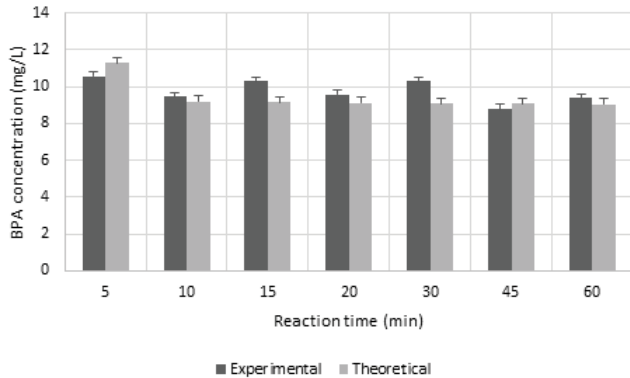| Kinetic model | $R^2$ | $k$ |
|---|---|---|
| First-order | 0.3614 | 0.002 |
| Second-order | 0.4142 | 0.0003 |
| Pseudo-second-order | 0.9995 | 0.0831 |



Fig. 6. Experimental and theoretical values of pseudo-second-order model.

chemicals were determined to calculate the cost of wastewater treatment per m³. The cost analyses calculations can be seen in Table 4.

The electrical energy required (kWh) (kg BPA)$^{-1}$ for the wastewater containing 1 kg of BPA treatment with the peroxy electrocoagulation method was determined as a function of time using Eq. (6), which can be seen below [54]:

$$\text{EEC} = \frac{\int UIdt}{(C_0 - C_t)V\,3.6} = \frac{I\int Udt}{(C_0 - C_t)V\,3.6} \tag{6}$$

where $U$, is the applied voltage (V), $I$ is the current (A), $t$ is the electrolysis time (min), $C_0$, is the initial concentrations of BPA, $C_t$, (mg/L) is the BPA concentration at time $t$ and $V$ (L) is the volume of treated wastewater.

As seen from the results, it can be said that the PEC process indicated an effective performance for the decontamination of BPA contaminated wastewaters by providing superior removal efficiency and cost effectiveness.

## 4. Conclusion

This study aimed to contribute to the literature on BPA removal by investigated the removal of BPA toxic substances using the peroxy electrocoagulation method. $H_2O_2$ was added to the BPA solution and the removal was done using the peroxy electrocoagulation process method. As a result of the analysis, it was determined that the removal efficiency was 80.48% when the optimum conditions were met. The experimental conditions that resulted in the highest efficiency was determined as pH 2, $H_2O_2$ dose of 125 mg/L, $Na_2SO_4$ dose of 0.1 g/L, current density of 0.11 mA/cm² and reaction time of 45 min.

Table 4
Cost analyses of per m³ BPA wastewater treatment

| Parameter | Consumption (m⁻³ WW) | Unit price ($) | Total cost ($) |
|---|---|---|---|
| Electrical energy | 0.214 kWh m$^{-3}$ | 0.12 kWh$^{-1}$ | 0.256 |
| Fe-electron | 0.20 g/L | 0.76 kg$^{-1}$ | 0.152 |
| $Na_2SO_4$ | 0.1 g/L | 0.230 kg$^{-1}$ | 0.023 |
| HCl | 1.1 L/m³ | 78 L$^{-1}$ | 85.8 |
| $H_2O_2$ | 125 mg/L | 29 L$^{-1}$ | 9.135 |
| | | Total cost/m³ | 95.866 |

Bisphenol A, which shows toxic properties as an endocrine disrupting compound worldwide, appears in many objects and materials we use in daily life. As a result of this wide area of use, there is an increased risk of people coming into contact with BPA. In addition to this, various pollutant sources such as discharge of wastewater treatment plants and leachate in solid waste storage areas causes the environment to also be contaminated. The fact that there are so many areas of its use and its discharge into the environment, the lives of many people and living creatures are in danger due to exposure to BPA. In an attempt to prevent this, various studies have been conducted and published globally and in Turkey for possible regulatory restrictions on the use of these toxic substances.

## Acknowledgment

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