

# Anodic oxidation of bisphenol A by different dimensionally stable electrodes

Orhan T. Can, Muhammed M. Tutun and Ramazan Keyikoglu

## ABSTRACT

Bisphenol A (BPA) is a known endocrine disrupter and was detected in surface waters. We investigated the mineralization of BPA by electrochemical oxidation. Six different types of electrodes, including the boron-doped diamond (BDD), platinum (Pt), and mixed metal oxide (MMO) electrodes; RuO<sub>2</sub>-IrO<sub>2</sub>, RuO<sub>2</sub>-TiO<sub>2</sub>, IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub>, and Pt-IrO<sub>2</sub>, were compared as the anode material. Total organic carbon (TOC) was analyzed to monitor the mineralization efficiency of BPA. BDD achieved 100% BPA mineralization efficiency in 180 min and at a current density of 125 mA/cm<sup>2</sup>, whereas the TOC removal efficiency of Pt was 60.9% and the efficiency of MMO electrodes ranged between 48 and 54%. BDD exhibited much lower specific energy consumption, which corresponds to a lower energy cost (USD63.4 /kg TOC). The effect of operational parameters showed that the BDD anode was much more affected by the current density, initial BPA concentration, and electrolyte concentration than the other parameters such as the stirring speed and interelectrode distance.

**Key words** | boron-doped diamond, electrochemical oxidation, mineralization, mixed metal oxide anodes, specific energy consumption

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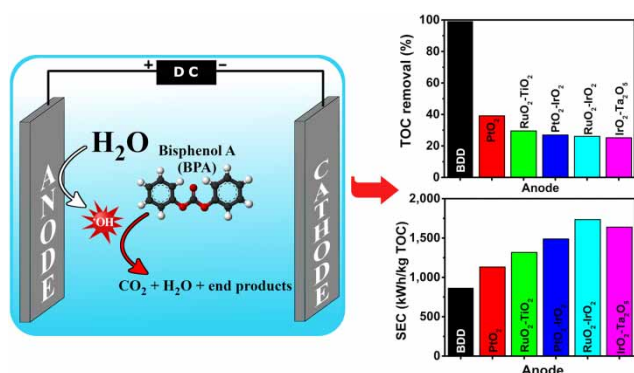
## HIGHLIGHTS

- BDD, Pt, RuO<sub>2</sub>-IrO<sub>2</sub>, RuO<sub>2</sub>-TiO<sub>2</sub>, IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub>, and Pt-IrO<sub>2</sub> were compared as the anode material.
- BDD performed better than MMO anodes for the mineralization of BPA.
- BDD anode showed 100% TOC removal efficiency in 180 min.
- Current efficiency of BDD was 2.53 times higher than Pt and 3.7 times than MMOs.
- The specific energy consumption was higher for MMO electrodes.

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## GRAPHICAL ABSTRACT



## INTRODUCTION

Bisphenol A (BPA) is the precursor compound for the production of epoxy resins and polycarbonates (Prokop *et al.* 2004). Approximately 70% of the produced BPA is used for polycarbonate production, 25% for epoxy resin production, and 5% for the production of various phenoplast products (Tsai 2006). Global bisphenol production, which was 1 million tons in the 1980s, increased to 2.2 million tons in 2009 and to 5.4 million tons in 2015 (Fiege *et al.* 2000). This dramatic increase in the production of BPA requires taking preventive measures for its disposal and release to the environment. BPA is highly toxic in water sources and is known to be a teratogenic and endocrine disruptor in invertebrates (Flint *et al.* 2012). It may cause contamination in water sources as a result of direct discharges from treatment plants and garbage storage facilities (Kang *et al.* 2007).

So far, a wide range of methods has been applied to remove BPA from water such as adsorption (Ahamad *et al.* 2019), photocatalytic oxidation (Dhiman *et al.* 2017; Kumar *et al.* 2018), sonophotolytic (Fadaei & Mardani 2014), ultrasound (Maruyama *et al.* 2006), wet oxidation (Mezohegyi *et al.* 2013), ultraviolet radiation (Neamțu & Frimmel 2006), and ozonation (Oke & Ogundugba 2014) processes. As a conventional treatment method, sodium hypochlorite was shown to be effective at removing BPA with an efficiency of 99% (Bourgin *et al.* 2013). Despite the satisfactory removal efficiency, the formation of chlorinated by-products is an important concern. Oke & Ogundugba (2014) showed that ozonation can be effectively used for the removal of BPA from contaminated water or industrial wastewater. They achieved an 81%

BPA degradation rate at pH 8.25. In 120 min reaction time, the total organic carbon (TOC) removal rate of BPA using 6.67 mg/min O<sub>3</sub> was found to be 41% and 51% at pH 8.25 and pH 11, respectively. The drawback of the ozonation process is considered to be high operating and capital investment costs. The adsorption process is a very effective and low-cost option and widely applied in water treatment (Alshehri *et al.* 2014). However, the low hydrophobicity (log K<sub>ow</sub>) of BPA may restrict its adsorption capacity. Neamțu & Frimmel (2006) investigated the removal of BPA from aqueous solutions by the UV/H<sub>2</sub>O<sub>2</sub> process. In 90 min treatment time, they achieved BPA removal efficiencies of 45% and 60% with 8.5 and 17 mg/L H<sub>2</sub>O<sub>2</sub> concentrations, respectively. However, due to the recalcitrant structure of BPA and its resistance to biological degradation, more powerful and novel treatment methods are required.

Electrochemical oxidation (EO) has proved its effectiveness in the treatment of a wide range of wastewaters and the degradation of various recalcitrant pollutants (Ding *et al.* 2012; Gengec & Kobya 2013; Zaouak *et al.* 2013; Can *et al.* 2019a; Turan *et al.* 2020). This process involves two possible mechanisms for the oxidation of pollutants in an aqueous solution: direct and indirect oxidation. In the case of direct oxidation, pollutants are directly oxidized on the anode surface as a result of the electron transfer between the pollutants and electrode. In the latter case, strong oxidants such as hydroxyl radicals (•OHs), HOCl, SO<sub>4</sub><sup>-</sup> are formed on the anode and react with pollutants in the bulk solution (Panizza & Cerisola 2009; Martínez-Huitle *et al.* 2015). The formation of such oxidants depends mostly on

the electrode material, applied current, and electrolyte solution. Electrode material plays a key role in controlling the oxidant generation rate and type of oxidants (Martínez-Huitle & Ferro 2006). Various electrode materials have been developed and they have been roughly classified into two types: 'active' and 'nonactive' by Comninellis (1994). Active electrodes interact strongly with  $\cdot\text{OH}$ , and thus oxidation of organics occurs selectively as the reactions of oxygen evolution proceeds simultaneously. Mixed metal oxide (MMO) electrodes, which are made by forming a thin layer of metal oxides on the substrates such as platinum and titanium, are classified as active anodes (Soni *et al.* 2017). Nonactive anodes, on the other hand, such as boron-doped diamond (BDD), interact weakly with  $\cdot\text{OH}$ . In general, the lower the adsorption strength of the  $\cdot\text{OH}$  on the electrode surface, the higher the oxygen overpotential for a given electrode material (Zaouak *et al.* 2014; Groenen Serrano 2018).

Numerous types of anodes have been employed in the EO process for the removal of BPA. For instance, Dong *et al.* (2019) used BDD anodes for the mineralization of BPA in the presence of  $\text{NaNO}_2$  electrolyte. They obtained 63.7% TOC removal at a current density of  $9.04 \text{ mA/cm}^2$  in 60 min. In another study (Li *et al.* 2017), the EO process using BDD anodes with  $\text{NaCl}$  as the supporting electrolyte was applied for the BPA removal and obtained a TOC removal efficiency of 77.5%. Other studies in the literature have employed BDD anodes in the EO of BPA (Muruganathan *et al.* 2008; Wu *et al.* 2016; Dietrich *et al.* 2017). Regarding the active electrodes, Xue *et al.* (2011) applied a type of DSA electrode of  $\text{Ti/SnO}_2\text{-Sb}_2\text{O}_5/\text{PbO}_2$  for the oxidation of BPA in a sulfate electrolyte solution. Wu *et al.* (2016) compared two nonactive anodes, BDD and  $\text{SnO}_2$ , in the electrooxidation of BPA, and they concluded that both anodes acted highly different because of their different surface characteristics and oxygen evolution potentials. Moreover, other studies have used carbon fiber (Kuramitz *et al.* 2001) and graphite (Govindaraj *et al.* 2013) as electrode materials in the EO of BPA. Overall, these studies employed different electrode materials in various electrolyte mediums, which directly affects the type of oxidant species that will be responsible for the oxidation of BPA. There have been two studies that have investigated the effect of anode material in the EO process for the degradation of BPA (Cui *et al.* 2009; Wu *et al.* 2016). Cui *et al.* (2009) compared the performance of four different titanium (Ti)-based electrodes including Ti/BDD, Ti/Sb-SnO<sub>2</sub>, Ti/RuO<sub>2</sub>, and Ti/Pt in a 0.1 M  $\text{Na}_2\text{SO}_4$  medium. They conducted the cyclic

voltammetry and chrono potentiometry on the anodes and elucidated the degradation pathway of BPA. In another study, Wu *et al.* (2016) compared BDD anode with two different modified  $\text{SnO}_2\text{-Sb}$  anodes in the EO of BPA. They studied the effects of pH and electrolyte medium and found that in 0.1 M  $\text{NaCl}$ , modified  $\text{SnO}_2\text{-Sb}$  anodes exhibited better electrocatalytic activity for BPA oxidation than BDD. Despite the wide range of anode materials employed in the EO of BPA, literature lacks a comprehensive study that investigates the performance of prominent electrode materials. The operation cost is a major concern in electrochemical treatment processes and should be considered along with the process performance.

With these concerns, the present study investigated the performances of different anode materials including BDD, Pt (platinum), and MMO electrodes ( $\text{Ti/RuO}_2\text{-TiO}_2$ ,  $\text{Ti/RuO}_2\text{-IrO}_2$ ,  $\text{Ti/IrO}_2\text{-Ta}_2\text{O}_5$ ,  $\text{Ti/Pt-IrO}_2$ ) in the mineralization of BPA by EO process. Electrodes were compared both in terms of BPA mineralization efficiencies and electrical energy consumptions. For this purpose, the performances of the electrodes on TOC removal were investigated at three different current densities. The effect of various operational parameters (current density, mixing rate, interelectrode distance, initial pollutant concentration, electrolyte concentration) on the process was also determined using the most effective electrode. Finally, specific energy consumption (SEC) was determined based on the amount of consumed electrical energy during TOC removal.

## MATERIAL AND METHOD

### Materials

All the chemicals used in the experiments were of analytical quality and obtained from Sigma Aldrich and Merck. Phosphoric acid was used as the supporting electrolyte during anodic oxidation. BDD, Pt, and MMO electrodes were used as the anode and a stainless-steel electrode was used as the cathode. Pt and MMO electrodes are Ti-based electrodes. BDD electrodes were supplied by DiaCCon GmbH Germany. Pt and MMO electrodes were supplied by Baoji Changli Special Metal Co. Ltd, China. A DC power supply (Agilent 6675A model, USA) was used to provide a maximum output of 20 A-120 V. A magnetic stirrer (Heidolph MR 3000D, Germany) was used to mix the solution.

## Experimental apparatus and procedure

Electrooxidation experiments were performed in a Pyrex glass reactor under galvanostatic conditions. A 500-mL sample of BPA solution was put into the reactor for each experiment. Phosphoric acid was added into the solution as an electrolyte to provide conductivity in the solution. To rule out the additional oxidation that may be caused by the anion of a supporting electrolyte (such as  $\text{Cl}_2$ ,  $\text{SO}_4^{2-}$ , etc.), we used phosphoric acid as a weak acid. For the reactor and experimental setup, see our previous paper (Can et al. 2019b). No pH adjustment was made in the experiments, and the test solution was started at the natural pH (pH 2.5). The pH was measured at the end of the study. The change in the pH at the end of the experiment was found to be no more than 0.2. The increase in temperature during the test period was maintained at 38–42 °C by supplying external air to the outer wall of the reactor.

## Analytical method

Mineralization of organic matters is defined as the conversion of them into the final products of  $\text{CO}_2$  and  $\text{H}_2\text{O}$  by the oxidation process. The mineralization rate of the organic matter content of the solution was determined by monitoring the TOC concentration. The TOC content of the solutions was determined by the non-purgeable organic carbon method using a TOC analyzer with a non-dispersive IR source (Shimadzu, TOC-L model, Japan). Equations (1)–(4) used in the study are listed below, which are commonly used in literature and described in previous studies (Comninellis & Pulgarin 1991; Lee et al. 2017; Can et al. 2019b).

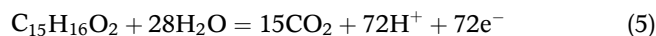
$$\text{TOC removal efficiency, } E, (\%) = \frac{C_i - C_f}{C_i} \times 100 \quad (1)$$

$$\text{SEC} \left( \frac{\text{kWh}}{\text{kg TOC}} \right) = (\text{V.I.t}) / \text{TOC}_{\text{rem}} \quad (2)$$

$$\text{Anode efficiency, } \eta, \left( \frac{\text{mg COD}}{\text{Ahm}^2} \right) = \frac{\Delta(\text{TOC})_{\text{exp}} \times V_s}{\text{ItS}_{\text{anode}}} \quad (3)$$

$$\begin{aligned} \text{Mineralization current efficiency, MCE} (\%) \\ = \frac{n F V_s \Delta(\text{TOC})_{\text{exp}}}{4.32 \times 10^7 \text{mIt}} \end{aligned} \quad (4)$$

where  $n = 72$ , assuming that the overall mineralization of the BPA into  $\text{CO}_2$  occurs as in the following reaction:



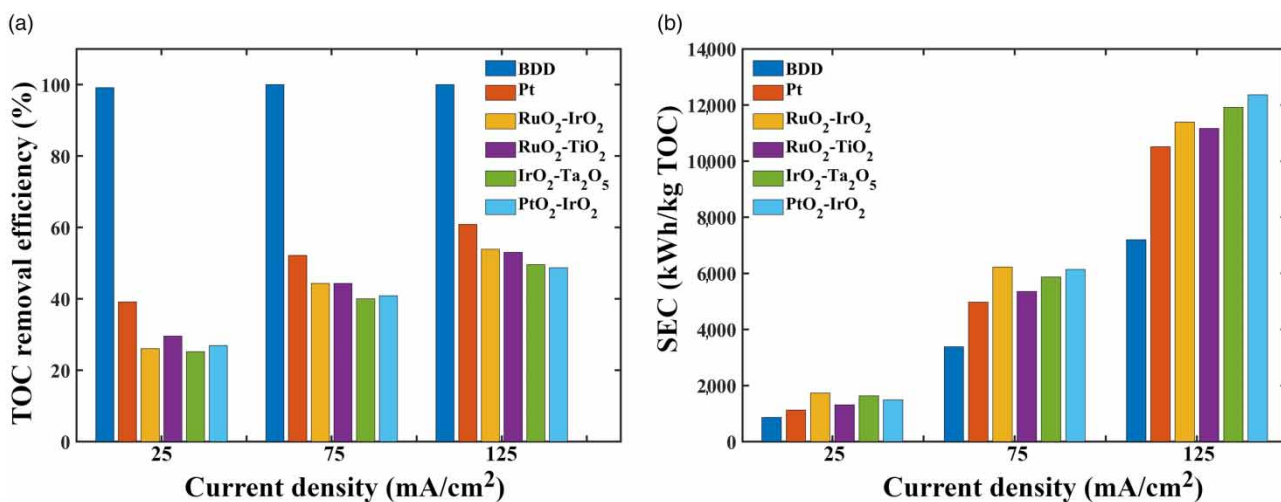
## RESULTS AND DISCUSSION

In this section, the most suitable anode electrode was determined by comparing the BPA mineralization performances at various current densities. Then the effect of the operating parameters such as the current density, stirring speed, inter-electrode gap, initial BPA concentration, and electrolyte concentration on the TOC removal efficiency by the most suitable electrode was determined. It is known that the applied potential and current directly affect the efficiency of the process and the operating cost (Can 2015). Thus, the SEC value that is based on the amount of electricity consumed for TOC removal was also determined.

### The effects of anode type and current density

To investigate the effect of anode type on the TOC removal by the EO process, six different anodes (BDD, Pt, and four MMO electrodes) were compared at different current densities. Figure 1(a) shows the TOC removal efficiencies of different anodes at the current densities of 25, 75, and 125  $\text{mA}/\text{cm}^2$  in 360 min treatment time. There were remarkable differences among the BDD, Pt, and MMO electrodes in their effectiveness at anodic mineralization of BPA. The comparison of TOC removal efficiencies indicated that the BDD electrode was able to mineralize significantly more BPA than Pt and MMO electrodes. Even at the lowest current density of 25  $\text{mA}/\text{cm}^2$ , the BDD electrode achieved almost 100% BPA mineralization efficiency. On the other hand, TOC removal efficiencies by MMO electrodes at the end of the 360-min were obtained as 61% (Pt), 54% ( $\text{RuO}_2\text{-IrO}_2$ ), 53% ( $\text{RuO}_2\text{-TiO}_2$ ), 50% ( $\text{IrO}_2\text{-Ta}_2\text{O}_5$ ), and 49% ( $\text{Pt-IrO}_2$ ).

Figure 1(b) displays the SEC values calculated based on the amount of TOC removed by each anode at the current densities of 50, 75, and 125  $\text{mA}/\text{cm}^2$ . The lowest SEC values were achieved for the BDD electrode in the range between 866 and 7,200  $\text{kWh}/\text{kg TOC}$ . SEC values of Pt and MMO electrodes, from smallest to largest, occurred in the order of  $\text{Pt} > \text{RuO}_2\text{-TiO}_2 > \text{Pt-IrO}_2 > \text{IrO}_2\text{-Ta}_2\text{O}_5 > \text{RuO}_2\text{-IrO}_2$ . It is worth noting that the SEC value of the  $\text{IrO}_2\text{-Ta}_2\text{O}_5$  electrode was lower than that of the  $\text{RuO}_2\text{-IrO}_2$



**Figure 1** | (a) TOC removal efficiencies and (b) SEC values of BDD, Pt, and MMO electrodes at different current densities (experimental conditions: reaction time: 360 min, [BPA]<sub>0</sub> = 150 mg/L, stirring speed = 500 rpm, interelectrode distance: 15 mm, [H<sub>3</sub>PO<sub>4</sub>] = 0.25 M, conductivity 22.1 mS/cm).

electrode, despite its lower TOC removal efficiency. This was due to the different potential difference (V) of the electrodes measured during the experiment with each electrode. As the current applied to the electrooxidation cell increases, the potential (V) between the electrodes increases because the solution conductivity is constant. Since the amount of TOC removed does not increase proportionally to the applied current density, an increase in the SEC was observed with an increase in the applied current.

Due to the inherent characteristics of BDD electrodes, which had the highest removal efficiencies, lower voltage values were obtained compared with the other electrodes. Only the BDD electrode was able to achieve the complete mineralization of BPA. At the current density of 75 mA/cm<sup>2</sup>, 240 min treatment time was sufficient to obtain 100% TOC removal efficiency for the BDD electrode. At the highest current density (125 mA/cm<sup>2</sup>), this duration decreased to 180 min. Pt performed relatively better than the MMO electrodes, achieving 39% and 43% TOC removal. In contrast, the MMO electrodes achieved 34% and 35% TOC removal on average under the same conditions.

The degradation capacity of the anodes depends on their capabilities to produce •OHs. BDD electrodes have extraordinary properties such as the wide potential window, high electrochemical stability, and negligible adsorption of organic molecules (Enache *et al.* 2009). Diamond is an exceptional hard crystalline form of carbon, with sp<sup>3</sup> hybridized carbon atoms bonded together. Boron doping into diamond gives BDD electrodes p-type semiconducting characteristics by acting as an electron acceptor because

of the electron deficiency in its outer shell. With these properties, the BDD electrode is capable of generating high amounts of •OH radicals and other active intermediates through water oxidation. Since these active species are loosely adsorbed on the surface of BDD, they readily attack and cause oxidative mineralization of organic pollutants (Zhi *et al.* 2003). In the case of MMO electrodes, coating an oxide layer causes the formation of a deactive region that increases the anode potential and leads to mechanical losses at the interface. This results in a decreased electrical conductivity and efficiency in the degradation of pollutants (Moradi *et al.* 2020).

Another important factor that determines the efficiency of anodes in the degradation of pollutants is their oxidation potentials. Anode electrodes are divided for simplicity into two classes: active and non-active electrodes (Comminellis 1994). Active anodes, which present low oxygen evolution overpotential, are good electrocatalysts for the oxygen evolution reaction (OER) and, consequently, lead to selective oxidation of the organic materials due to chemisorbed •OH. Therefore, the oxidation power of this type of anodes is generally low. On the other hand, non-active anodes are weak electrocatalysts for the OER, and direct EO is supposed to occur at these electrodes. A higher oxidation state is unavailable and the organic species are directly oxidized by a physisorbed •OH. Pt-based, Ru-based oxides, and Ir-based oxides are considered active anodes with low oxidation potential (RuO<sub>2</sub>-TiO<sub>2</sub> = 1.4–1.7 V, IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub> = 1.5–1.8 V, Pt = 1.7–1.9 V). On the other hand, BDD is considered non-active, thus presenting high oxidation potential (SnO<sub>2</sub> = 1.9–2.2 V, PbO<sub>2</sub> = 1.8–2.0 V, BDD = 2.2–2.6 V).

The production of quasi-free  $\bullet\text{OH}$  on the surface of BDD has been confirmed by electron spin resonance measurements with spin traps. These explain the lower performance of MMO electrodes compared with the BDD anode, on which the  $\bullet\text{OH}$  is loosely attached. This electrogenerated  $\bullet\text{OH}$  on the BDD anode is highly reactive and thus can mineralize the organics with a high-current efficiency (Martínez-Huitle & Andrade 2011).

Table 1 shows the electrolysis parameters during the mineralization of BPA by different anodes. As the current density increased, the TOC removal efficiency, average voltage, and SEC all showed an increase, while a decrease with the anode and mineralization efficiency was observed. The higher production rate of  $\bullet\text{OH}$  with increasing current density can explain the increase in TOC removal efficiency at higher current densities. During electrooxidation, part of the applied electric energy is transformed into heat. Therefore, when the current density (applied current) is increased, the voltage also increases, and more heat is generated. A significant portion of the energy is converted into heat rather than producing  $\bullet\text{OH}$ . This conversion reduces the efficiency of the process.

The SEC is one of the most critical factors dictating the economics of the electrooxidation process. The smaller the SEC, the more efficient the process. In the study, the SEC value increased when the current density was increased due to a reduction in the conversion rate of applied energy to  $\bullet\text{OH}$ . The anode efficiency and mineralization current efficiency are the other important factors governing the economics of the electrooxidation process (Table 1). Although the removal efficiency of pollutants increases with increasing current density, a decrease in the current and anode efficiency is observed due to the mass transfer limitations and wasted energy (Comminellis *et al.* 2008). Both the anode efficiency and the mineralization current efficiency of the EO declined when the current density was increased for all anode materials. Overall, the BDD anode exhibited the highest mineralization efficiency even at the highest current density. Table 1 shows the strong role of anode type on the process efficiency as well as on the economy. As expected, for a current density of 25 mA/cm<sup>2</sup> and 360 min process time, the anode efficiency of the BDD anode was 2,375 mg TOC/Ahm<sup>2</sup>, and the mineralization current efficiency was 10.2%. These values are 2.53

**Table 1** | Electrolysis parameters during the mineralization of BPA by different anodes (experimental conditions: reaction time: 360 min,  $[\text{BPA}]_0 = 150$  mg/L, stirring speed = 500 rpm, interelectrode distance: 15 mm,  $[\text{H}_3\text{PO}_4] = 0.25$  M, conductivity 22.1 mS/cm)

Anode	J (mA/cm <sup>2</sup> )	TOC <sub>rem</sub> (%)	U (V)	SEC (kWh/kg TOC <sub>rem</sub> )	Cost (\$/kg TOC)	$\eta$ (mg TOC/Ahm <sup>2</sup> )	MCE (%)
BDD	25	99.1	8.2	865.9	63.4	2,375.0	10.2
	75	100.0	10.8	3,385.3	247.7	798.6	3.4
	125	100.0	13.8	7,200.0	526.92	479.2	2.1
Pt	25	39.1	4.2	1,132.8	82.9	937.5	4.0
	75	52.2	8.3	4,978.8	364.4	416.7	1.8
	125	60.9	12.3	10,515.4	769.5	291.7	1.3
Ti/Pt-IrO <sub>2</sub>	25	27.0	3.8	1,488.8	108.95	645.8	2.8
	75	40.9	8.0	6,144.5	449.7	326.4	1.4
	125	48.7	11.5	12,362.1	904.7	233.3	1.0
Ti/RuO <sub>2</sub> -TiO <sub>2</sub>	25	29.6	3.7	1,317.2	96.4	708.3	3.0
	75	44.3	7.6	5,352.0	391.7	354.2	1.5
	125	53.0	11.4	11,163.9	817.0	254.2	1.1
Ti/RuO <sub>2</sub> -IrO <sub>2</sub>	25	26.1	4.3	1,734.4	126.9	625.0	2.7
	75	44.3	8.8	6,228.7	455.8	354.2	1.5
	125	53.9	11.8	11,388.4	833.4	258.3	1.1
Ti/IrO <sub>2</sub> -Ta <sub>2</sub> O <sub>5</sub>	25	25.2	4.0	1,637.8	119.8	604.2	2.6
	75	40.0	7.5	5,874.3	429.9	319.4	1.4
	125	49.6	11.3	11,915.8	872.03	237.5	1.0

U: average voltage between electrodes,  $\eta$ : anode efficiency, MCE: mineralization current efficiency, t for 360 min.

times higher than that of Pt electrodes and 3.7 times higher than other MMO electrodes. The electrical energy cost of the process was also calculated by taking into account the unit electrical energy price in Turkey (according to a February 2020 survey in the Turkish market, the price of electricity was 0.07318 US\$/kWh). Moreover, the BDD anode exhibited a much lower energy consumption cost of US\$65.40 to remove 1 kg TOC. In conclusion, the BDD electrode was selected as the most suitable electrode material due to the highest removal efficiency with the lowest energy consumption.

### The effect of operating parameters on BPA mineralization using BDD electrode

#### Effect of current density

The effect of the current density on the TOC removal efficiency was investigated at five different current densities (25, 50, 75, 100, and 125 mA/cm<sup>2</sup>) for a reaction time of 90 min. Figure 2(a) presents the effect of the current density on TOC removal efficiency. There was a direct relationship between the current density and TOC removal efficiency. As the current density applied to the electrooxidation cell was increased, the amount of TOC removed increased as well. At the end of the 90 min treatment time, TOC removal efficiency values for the current densities of 25, 50, 75, 100, and 125 mA/cm<sup>2</sup> was 41%, 54%, 70%, 81% and 91%, respectively. The oxidative degradation of organics at the BDD anode has been reported to only take place in the potential region of water decomposition (Boye *et al.* 2006). As can be seen in Figure 2(a), the percentage of BPA mineralization

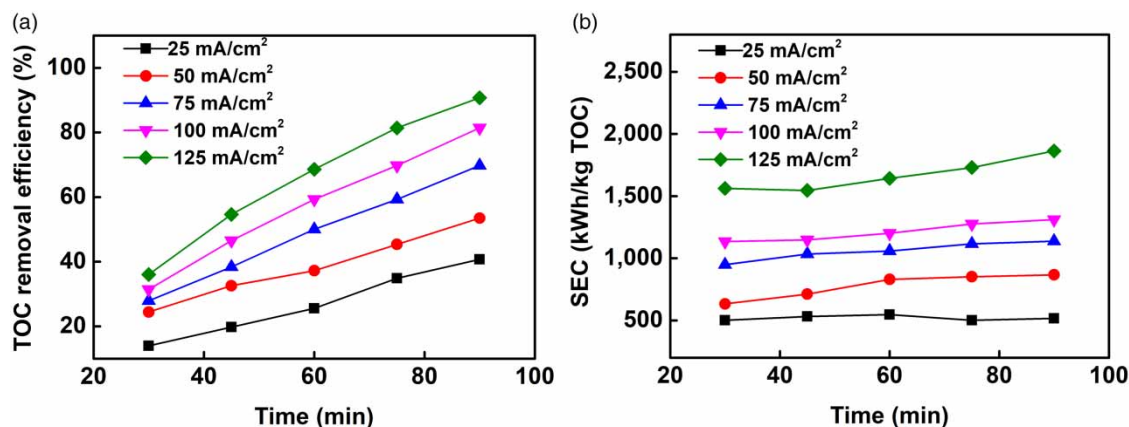
was directly dependent on the applied current, with a higher value leading to increased TOC removal efficiencies.

Figure 2(b) shows the SEC values calculated based on the amount of TOC removed. The SEC values increased with the increasing current density applied. At the end of the 90 min treatment time, the SEC value for the current density of 25 mA/cm<sup>2</sup> was 516 kWh/kg TOC and it was 1,864 kWh/kg TOC for the current density of 125 mA/cm<sup>2</sup>. At this stage, an increase in the current density led to an increase in TOC removal efficiency. However, SEC values are adversely affected by this. For instance, by increasing the current density five-fold, the TOC removal efficiency increased 2.25-fold; however, the SEC value increased by 3.6-fold. Therefore, it would be more economical to operate the process at a more extended refining period at a lower current density in order to achieve an acceptable SEC value.

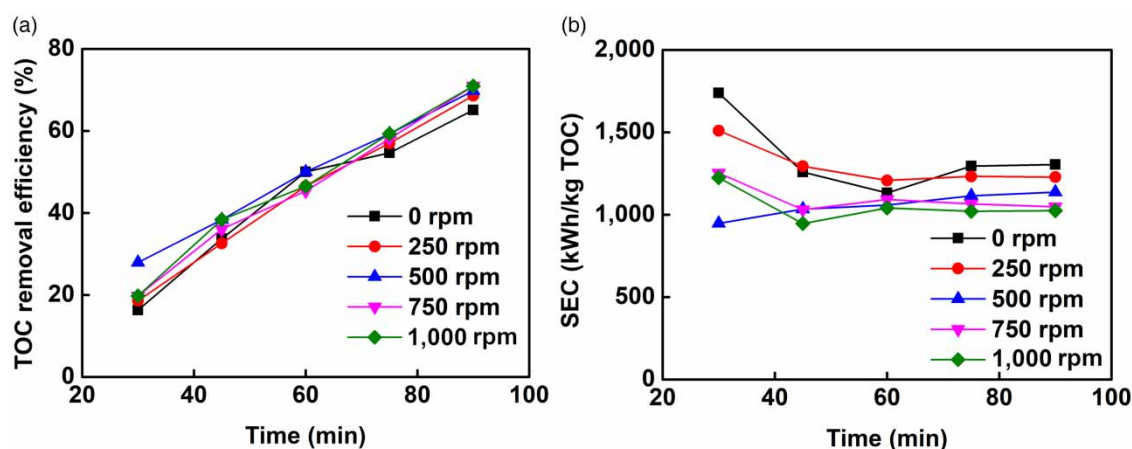
#### Effect of stirring speed

The effect of stirring speed on TOC removal efficiency was investigated at five different stirring speeds (0, 250, 500, 750, and 1,000 rpm). Figure 3(a) demonstrates the effect of stirring speed on TOC removal efficiency from a BPA solution. There was no significant variation among the different stirring speeds in terms of TOC removal efficiency. Nevertheless, a slight improvement can be observed compared with the non-stirred condition. At the end of the 90 min treatment time, TOC removal efficiency values for the stirring speeds of 0, 250, 500, 750, and 1,000 rpm were 65%, 69%, 70%, 71%, and 71%, respectively.

Figure 3(b) shows SEC values calculated depending on the amount of TOC removed for five different stirring speed values. As the figure indicates, the SEC value decreases as



**Figure 2** | (a) The effect of the current density on the TOC removal and (b) on SEC (experimental conditions: [BPA]<sub>0</sub> = 150 mg/L, stirring speed = 500 rpm, interelectrode distance = 15 mm, [H<sub>3</sub>PO<sub>4</sub>] = 0.25 M, conductivity 22.1 mS/cm, and anode = BDD).



**Figure 3** | (a) The effect of the stirring speed on the TOC removal and (b) on SEC (experimental conditions:  $[BPA]_0 = 150$  mg/L, current density =  $75$  mA/cm<sup>2</sup>, interelectrode distance =  $15$  mm,  $[H_3PO_4] = 0.25$  M, and anode = BDD).

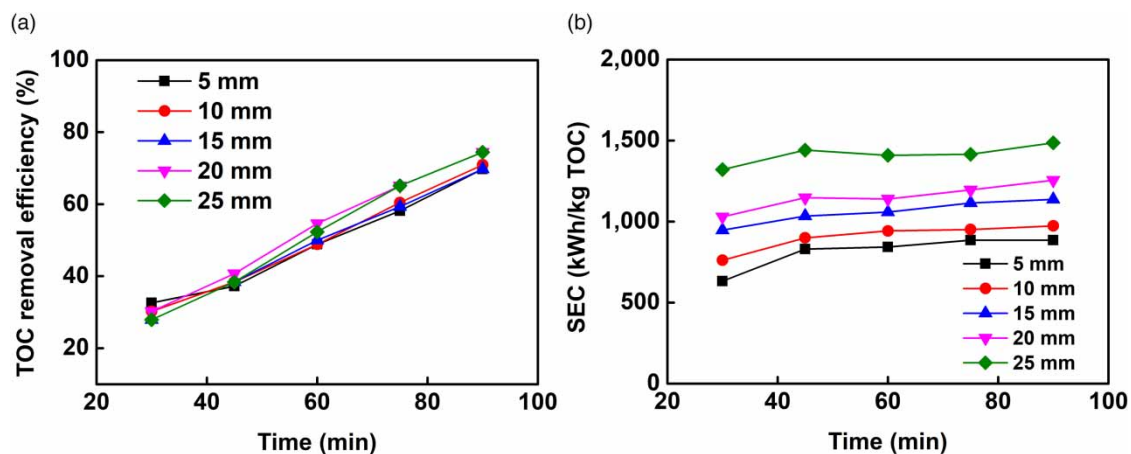
the stirring speed increases. The SEC value for 0 rpm and TOC efficiency of 65% is  $1,304$  kWh/kg TOC, and is  $1,024$  kWh/kg TOC for 1,000 rpm and TOC efficiency of 71%. The difference in TOC removal efficiency between 1,000 rpm (the highest stirring speed) and 0 rpm is about 6%, and in parallel with this, the SEC value is about 27% less. This result shows that it is economically more advantageous to carry out the process at a high stirring speed. However, it should be kept in mind that there are also costs involved in carrying out the stirring process in the reactor.

#### Effect of interelectrode distance

The impact of the interelectrode distance on TOC removal efficiency was investigated at five different

values (5, 10, 15, 20, and 25 mm). Figure 4 shows that the TOC removal efficiency for the distances of 5, 10, and 15 mm was almost the same, which was about 70%. When the distance was increased to 20 mm, TOC removal reached about 74%. However, an increase in the distance from 20 to 25 mm yielded no improvement in TOC removal efficiency.

Figure 4(b) shows that SEC values increased as the interelectrode distance was increased. At the end of 90 min treatment time, although the TOC removal efficiency for the distances of 5, 10, and 15 mm was almost identical, a significant increase in the SEC values of respective distances was observed. The SEC value was  $885$  kWh/kg TOC for 5 mm distance and  $1,137$  kWh/kg TOC for 15 mm distance at the end of 90 treatment time.



**Figure 4** | (a) The effect of the interelectrode distance on the TOC removal efficiency and (b) on SEC (experimental conditions:  $[BPA]_0 = 150$  mg/L, current density =  $75$  mA/cm<sup>2</sup>, stirring speed =  $500$  rpm,  $[H_3PO_4] = 0.25$  M, conductivity =  $22.1$  mS/cm, and anode = BDD).



The increase in the SEC values was due to the increase in resistance and potential difference, which are directly related to interelectrode distance. As the interelectrode distance increases, the electrical resistance passing through the solution increases and leading to an increase in the potential difference, which causes the SEC value to increase. The increase in SEC value can be up to twice as much. This is undesirable for the cost-effectiveness of the process.

### Effect of BPA concentration

The effect of the concentration of BPA on TOC removal efficiency was investigated at five different initial concentrations (50, 100, 150, 200, and 250 mg/L) and the results are presented in Figure 5. As can be seen from the figure, there was a significant decline in the TOC removal efficiency as the initial BPA concentration was increased. At the end of 90-min treatment time, TOC removal efficiency values for initial BPA concentrations of 50, 100, 150, 200, and 250 mg/L were 100%, 74%, 70%, 62% and 57%, respectively. Complete mineralization was obtained at the initial concentration of 50 mg/L with a 100% TOC removal rate. However, in relation to the amount (mg/L) of TOC removed, the mineralization performance was greater at higher initial concentrations. This was mainly due to the utilization of  $\cdot\text{OH}$  in the presence of more pollutant molecules, thus avoiding the  $\text{O}_2$  evolution reaction (Brillas *et al.* 2004).

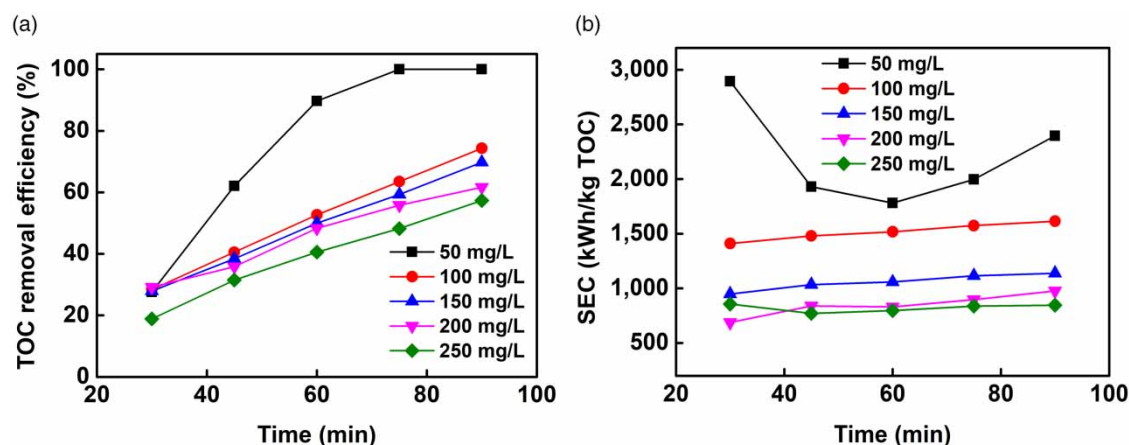
As Figure 5(b) shows, the SEC values decreased with increasing BPA concentrations. This occurrence was due to the difference in the amount of the substance removed for the same amount of electricity. Reduction in the

mineralization efficiency with increasing BPA concentrations was an expected outcome because the process under the same conditions had to deal with more pollutants. Although BPA was entirely mineralized at an initial concentration of 50 mg/L, only 57% of BPA was mineralized at a concentration of 250 mg/L. However, this efficiency value (57%) corresponds to 143 mg/L BPA, which is almost three times the concentration of 50 mg/L. When considered together with the SEC parameter, operating the process at high concentrations is desirable for SEC value. However, it is thought that extremely high concentrations will inhibit the process by causing electro-polymerization in the anode electrode.

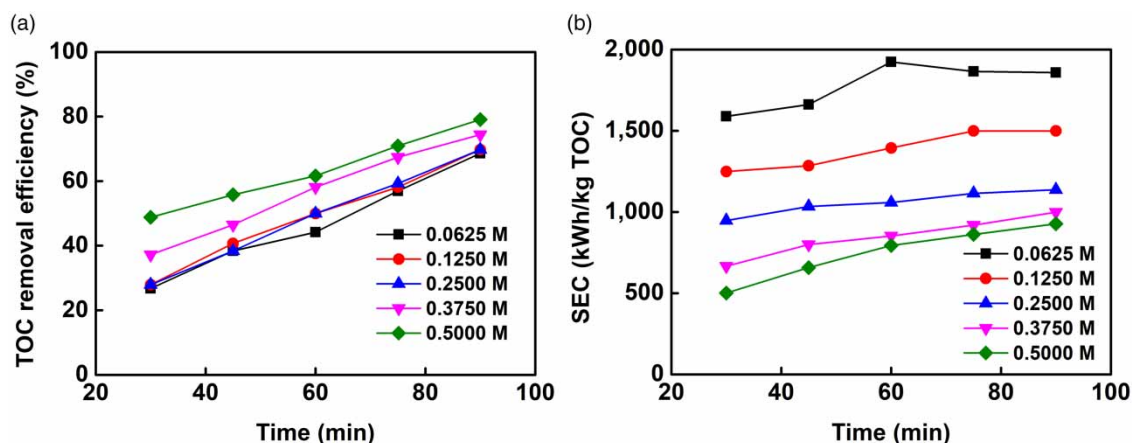
### Effect of electrolyte concentration

Electrochemical processes are strongly affected by the type and the concentration of the supporting electrolytes used (Keyikoglu *et al.* 2019). The effect of the electrolyte concentration on the TOC removal efficiency was investigated at five different  $\text{H}_3\text{PO}_4$  concentrations (0.0625, 0.125, 0.25, 0.50, and 0.75 M). As displayed in Figure 6(a), TOC removal efficiency was almost the same around 69–70% for the concentrations of 0.0625, 0.125, and 0.25 M. For the electrolyte concentrations of 0.375 and 0.5 M, there was an increase in the TOC removal efficiency, which reached 74% and 79%, respectively.

Figure 6(b) shows that the SEC value decreased as the electrolyte concentration was increased. Although the TOC removal efficiencies with electrolyte concentrations of 0.0625, 0.125, and 0.25 M were almost the same, the SEC values decreased at the same electrolyte concentrations. This was due to the decreased resistance and



**Figure 5** | (a) The effect of BPA concentration on the TOC removal efficiency and (b) on SEC (experimental conditions: interelectrode distance = 15 mm, current density = 75 mA/cm<sup>2</sup>, stirring speed = 500 rpm,  $[\text{H}_3\text{PO}_4] = 0.25 \text{ M}$ , conductivity = 22.1 mS/cm, and anode = BDD).



**Figure 6** | (a) The effect of the electrolyte (phosphoric acid) concentration on the TOC removal efficiency, (b) on the SEC experimental conditions =  $[BPA]_0 = 150$  mg/L, current density =  $75$  mA/cm<sup>2</sup>, stirring speed =  $500$  rpm, interelectrode distance =  $15$  mm, and anode = BDD).

potential difference that resulted from the increase in the interelectrode conductivity. At the end of 90 min treatment time, the SEC value for the electrolyte concentration of  $0.0625$  M and TOC removal efficiency of  $69\%$  was  $1,859$  kWh/kg TOC and  $928$  kWh/kg TOC for the concentration of  $0.5$  M electrolyte and TOC removal efficiency of  $79\%$ . An eight-fold increase in the amount of electrolyte led to an increase of about  $10\%$  in the removal efficiency and a decrease in the SEC value of about  $50\%$ . A high electrolyte concentration had a positive effect on the SEC value by reducing the electrical resistance by decreasing the potential difference.

This study investigated the performance of different anode materials on the mineralization of BPA by an EO process. The results indicated that, unlike the chemical bonding of  $\cdot OH$  by MMO electrodes, BDD electrodes physically bind  $\cdot OH$  and thus provide high oxidation capacity. The possible degradation pathway of BPA by electrocatalytic oxidation was reported to occur via the attack of  $\cdot OH$  to the aromatic ring in the BPA molecule (Wu *et al.* 2016). The formation of BPA 3,4-quinone was shown to cause DNA damage in the liver (Sakuma *et al.* 2010), and any treatment technology must therefore warrant complete mineralization of BPA to avoid the formation of toxic by-products. In the present study, the BDD electrode was the only one to achieve complete mineralization of BPA, which ensures the safe discharge of BPA-containing wastewaters. Even though BDD costs 18 times more than Pt/O<sub>2</sub> and 36 times more than MMO electrodes, the high mineralization of BDD compensated for the material cost and showed considerably lower SEC values while yielding a higher current efficiency than other anode materials tested. However, the major

disadvantage of these electrodes is the material cost, which must be taken into account in the cost assessment of the overall process.

## CONCLUSIONS

In this study, we examined the mineralization of BPA by the EO process using different anode materials. The comparison was made between BDD, Pt, and several MMO electrodes in terms of mineralization efficiency of BPA. BDD electrode was the only anode that achieved  $100\%$  mineralization of BPA in 180 min and at the current density of  $125$  mA/cm<sup>2</sup>. Pt appeared to be relatively better than the MMO electrodes, achieving  $60.9\%$  TOC removal efficiencies compared with the MMO electrodes, whose TOC removal efficiency ranged between  $48\%$  and  $54\%$ . BDD electrode was also more advantageous than the other electrodes for energy efficiency, with an SEC value of  $865.9$  kWh/kg TOC, which corresponds to a treatment cost of  $63.4$  US\$/kg TOC. In comparison, the treatment cost was calculated for Pt, Ti/Pt-IrO<sub>2</sub>, Ti/RuO<sub>2</sub>-TiO<sub>2</sub>, Ti/RuO<sub>2</sub>-IrO<sub>2</sub>, Ti/IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub> as  $82.9$  US\$/kg,  $108.95$  US\$/kg,  $96.4$  US\$/kg,  $126.9$  US\$/kg, and  $119.8$  US\$/kg, respectively. Similarly, the current efficiency of BDD anodes was 2.53 times higher than that of Pt electrodes and 3.7 times higher than other MMO electrodes. Both the process performance and the energy consumption of the electrooxidation process were influenced by the operating conditions. The effect of current density, initial BPA concentration, and the electrolyte concentration was more significant than the stirring speed and interelectrode distance. In this regard, more

attention should be given to the parameters that have the most effect on process performance.

## DECLARATION OF COMPETING INTEREST

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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