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Electrochemical Degradation of Bisphenol A in Water with/without Ce(IV) Addition

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This study investigates the degradation of bisphenol A (BPA) by electrooxidation on a boron-doped diamond (BDD) anode with/without Ce(IV) addition in 1 M Na₂SO₄ and real environmental water matrices: municipal wastewater treatment plant primary and secondary effluents (MWTPPE and MWTPSE, respectively), river water (RW), and groundwater (GW). The results show that the BPA degradation and TOC removal increased along with the current density (0.25–0.75 A/cm²), while the BPA degradation decreased but the TOC removal increased when the temperature rose (25°C-75°C). Similar efficiencies of BPA degradation or TOC removal were observed in pH 3–10 phosphate buffer solutions. At 0.5 A/cm² and 75°C in 1 M Na₂SO₄, complete BPA degradation was achieved using electrolysis (E), E+Ce(IV), Ce(IV); however, the BPA degradation or TOC removal was in the order Ce(IV) > E+Ce(IV) > E, and complete TOC removal was only attained using E alone. In MWTPSE, the BPA electrochemical degradation efficiency achieved by adding chemical Ce(IV) was similar to that by adding Ce(IV) regenerated (R-Ce(IV)) from real spent thin-film transistor liquid-crystal display (TFT-LCD) Cr-etching solutions; nevertheless, the TOC removal was better for the former than for the latter. The rates of formation and degradation of p-benzoquinone (an intermediate from BPA electrochemical degradation) followed the order E+Ce(IV) > E > E+R-Ce(IV). Without electrolysis, the *p*-benzoquinone could almost not be degraded when using Ce(IV) alone. Similar degradation efficiencies of BPA in MWTPPE, MWTPSE, RW, and GW were observed ($k = (3.68-6.07) \times 10^{-3} \text{ s}^{-1}$), but the complete TOC removal could not be obtained in RW, despite reaching complete pbenzoquinone degradation in the four matrices.

Keywords: electrochemical degradation; bisphenol A; *p*-benzoquinone; boron-doped diamond (BDD) electrode; mineralization

1. INTRODUCTION

The global production of bisphenol A (BPA) reaches several million tons annually [1,2], and is often found in CDs, sunglasses, bottles, cans, and containers for food and water [1]. However, it is regarded as an emerging pollutant which may have adverse impacts on the environment and human health [1]. BPA is also used in the manufacturing of epoxy resin-based paints, medical devices, surface coatings, printing inks and flame retardants [3], while the major use of BPA is the production of epoxy resins and polycarbonate plastics (e.g., bottles, tableware, cookware, food-packaging materials), which has raised global concern for being the major route for human exposure [4]. Recently, BPA has also been a concern due to its use as a color developer in the manufacturing of thermal paper, with concentrations up to 42,600 mg/g [2].

The widespread and frequent use of BPA in consumer products has resulted in its migration from materials to the environment and humans [2]. It has been reported that the BPA levels are in the ranges of 1.0-628 ng/L, 10-1080 ng/L, and 0.42-25,600 ng/g d.w. in river waters, wastewater treatment plant (WWTP) effluents, and sewage sludge, respectively [2]. In France, the BPA concentrations downstream from WWTP outputs range from 0.11 to 0.79 µg/L, a 3.8-fold increase compared to the normal levels [5]. The concentration of total BPs in Taihu Lake ($81-3.0\times10^3$ ng/L) was found to be significantly higher than that in Luoma Lake (range: $1.5\times10^2-1.9\times10^3$ ng/L) in China [6]. In Taiwan, the BPA concentrations in river waters were in the range 0.01-44.7 ng/L [7]. There is thus an urgent need to develop effective treatment processes for BPA removal in water matrices.

The removal of BPA has been studied using different approaches, such as a hot persulfate process [8], zero valent aluminum-acid system [9], electrochemical oxidation (using BDD/Si [10], BDD/Ti [11], or with the aid of graphite [12]). Moreover, tetra-valent cerium (Ce(IV)) was reported to be useful for the destruction of phenolic compounds [13,14]. So far, however, little attention has been paid to the electrochemical degradation of BPA on BDD/Nb with/without Ce(IV) addition in real environmental matrices and with the variation of *p*-benzoquinone (BPA-derived intermediate). These issues are thus the focus of the current work.

2. MATERIALS AND METHODS

2.1. Chemicals and materials



Figure 1. Molecular structure of BPA.

The BPA (with two aromatic rings shown in Fig. 1) was purchased from Chem Service Inc. (USA). Na₂SO₄ (Showa) was used to prepare the supporting electrolyte, while *p*-benzoquinone (Alfa) was used for the quantification of this intermediate. The BDD electrode (substrate: Niobium; BDD coating thickness: 2.0 mm) was supplied by Diachem (CONDIAS GmbH, Germany). For the Ce(IV) addition tests, we dissolved diammonium cerium(IV) nitrate ((NH₄)₂Ce(NO₃)₆) (Showa) in different electrolytes. The Ce(IV) regenerated (R-Ce(IV)) from real spent thin-film transistor liquid-crystal display (TFT-LCD) Cr-etching solutions was also used in the tests; the details of these solutions were stated in our earlier works [15–17].

2.2. Cyclic voltammetry (CV) of BPA

The electrochemical characteristics of BPA on BDD were studied with cyclic voltammetry (CV) in 1 M Na₂SO₄, and the potential scan range was $0.0 \leftrightarrow 2.0$ V (starting/ending at 0.0 V vs Ag/AgCl and scan rate = 100 mV/s). The CV measurements were conducted using a CHI 660B electrochemical work station. The BDD electrode tested in electrolytic measurements was also used as the working electrode, and the counter electrode was a platinum wire. An Ag/AgCl electrode (3 MKCl) was used as the reference electrode.

2.3. Degradation experiments

The anolyte (100 mL) was 1 M Na₂SO₄ or real water matrix (from the local area) spiked with 100 mg/L BPA, while the catholyte was only 1 M Na₂SO₄ solution. The BDD electrode (2 cm²) was used as the anode, while a stainless-steel plate acted as the cathode for the degradation of BPA at different operating conditions galvanostatically performed in a thermostatted divided cell (separator: Nafion-212 (Dupont)) with magnetic stirring. A DC power supply (Good Will Instrument Co. Ltd. GPS-2303) was used for the electrolytic experiments. The cell voltage and current were monitored over time based on the readings of the DC power supply. During the experiments, samples were collected from the anolyte at intervals and detected for the concentrations of BPA, *p*-benzoquinone, and total organic carbon (TOC).

The BPA degradation efficiency, TOC removal efficiency, and pseudo first-order rate constant (*k*) of BPA degradation can be calculated as follows:

BPA degradation efficiency = $[1 - (C_t/C_o)] \times 100\%$	(1)
TOC removal efficiency = $[1 - (TOC_t/TOC_o)] \times 100\%$	(2)
$Ln(C_o / C_t) = kC_t$	(3)

where C_t is the residual concentration of BPA at a given electrolytic time, C_o is the initial concentration of BPA, TOC_t is the concentration of TOC at a given electrolytic time, and TOC_o is the initial concentration of TOC.

The real environmental water matrices were collected in a local area, and these waters were municipal wastewater treatment plant primary and secondary effluents (MWTPPE and MWTPSE, respectively), river water (RW), and groundwater (GW). The magnitude of original TOC, biological

oxygen demand (BOD), or chemical oxygen demand (COD) concentration was in the order MWTPPE > MWTPSE > RW> GW (Table 1). Although these matrices had different values of original conductivity, oxidation–reduction potential (ORP), dissolved oxygen (DO), and pH, their conductivity data were similar after adding 1 M Na₂SO₄. BPA (100 mg/L) was spiked into the different matrices before electrolysis.

Table 1. Characteristics of the tested environmental matrices, namely the municipal wastewater treatment plant primary and secondary effluents (MWTPPE and MWTPSE, respectively), groundwater (GW), and river water (RW) before BPA addition.

Parameter	TOC	BOD	COD	Conductivity	ORP	DO	all
	(mg/L)	(mg/L)	(mg/L)	(µs/cm)	(mV)	(mg/L)	рн
GW	1.2	0.3	2.3	190	301	8.2	6.69
RW	3.0	2.4	5.3	545	277	7.5	7.29
MWTPPE	68	63	83	792	-40.0	6.6	7.41
MWTPSE	7.8	8.0	14	790	162	8.8	7.43

2.4. Analysis

The concentration of BPA in each sample was monitored by HPLC (Hitachi Chromaster 5420). A Shishedo C18 column (250 mm×4.6 mm, particle size, 5µm) was used for separation. The mobile phases were water/methanol (30:70, v/v) (ECHO Chemical Co) and water/acetonitrile (55:45, v/v) (ECHO Chemical Co) for the analyses of BPA and *p*-benzoquinone, respectively. The flow rate was 1.0 mL/min and the injection volume was 20 µL. For the detection of BPA (225 nm) and *p*-benzoquinone (245 nm), the retention times were 6.3 and 4.3 min, respectively. Mineralization of BPA was evaluated based on the abatement of corresponding TOC determined using a Shimadzu 5000 Model TOC analyzer at 680°C of combustion temperature. The carrying gas was purified air with a flow rate of 150 mL/min. Details of the analyses of water quality parameters such as BOD, COD, DO, ORP, and pH were stated elsewhere [18,19].

3. RESULTS AND DISCUSSION

3.1. Cyclic voltammetric (CV) analysis of BPA on BDD electrode

Figure 2 shows the cyclic voltammograms of 1 M Na₂SO₄ aqueous solutions with/without 100 mg/L BPA on BDD at a scan rate of 100 mV/s. The anodic peak at ~1.53 V vs. Ag/AgCl of 1 M Na₂SO₄ is associated with the oxidation of sulfate to persulfate [19,20]. When BPA was present in the 1 M Na₂SO₄, the peak current at the same potential was smaller and the anodic peak current decreased with the increase of the scanning cycle number, which is attributed to the partial coverage of the intermediate-deactivated layer [21,22] or polymeric product deposition on the surface of BDD [10]. This phenomenon can be alleviated by anodic polarization in the potential region of water decomposition (>2.3 V) [21,22]. However, no peak for BPA oxidation was identified, so the BPA

degradation in subsequent electrolysis experiments should be contributed mainly by hydroxyl radicals (•OH), and not by the direct electro-oxidation of BPA on BDD. This CV result on BDD/Nb is similar to that on BDD/Si reported in a paper which regards the anodic peak (sulfate oxidation to persulfate) as that for BPA oxidation [10].



Figure 2. Cyclic voltammograms of 1 M Na₂SO₄ aqueous solutions with/without 100 mg/L BPA on BDD at 25° C (scan rate = 100 mV/s).

3.2. Effect of current density on BPA electrochemical degradation

The degradation of BPA on BDD in 1 M Na₂SO₄ under different current densities (0.25-0.75 A/cm²) was performed at 25°C. The degradation efficiency of BPA increased as the current density increased (Fig. 3a). However, the BPA degradation curves were similar at 0.50 and 0.75 A/cm². This trend is also true for TOC removal (Fig. 3b). In general, the increase in current density may increase the production of •OH and thus improve the degradation of target compounds. Therefore, the BPA was completely degraded for 15 min electrolysis at 0.50 and 0.75 A/cm², while it took more 5 min at 0.25 A/cm². However, Murugananthan et al. [10] reported that the electrochemical degradation of BPA on BDD/Si in 0.1 M Na₂SO₄ increased with increasing current density at a lower initial BPA concentration (C₀ = 20 mg/L) and smaller current densities (0.014–0.036 A/cm²); moreover, the oxidation reaction of BPA was limited by current density. The *k* values of BPA degradation were 3.12×10^{-3} , 5.39×10^{-3} , and 6.44×10^{-3} s⁻¹ at 0.25, 0.50 and 0.75 A/cm² respectively (Fig. 3a inset). Therefore, the *k* value at 0.50 A/cm² on BDD/Nb (5.39×10^{-3} s⁻¹) obtained in this study was significantly greater than that at 0.036 A/cm² on BDD/Si (1.28×10^{-4} s⁻¹) [10].

Complete TOC removal (mineralization) needed 60, ~90, and 120 min at 0.25, 0.50, and 0.75 A/cm², respectively. Similar results were also observed in our earlier works when using BDD for the degradation of different target compounds, such as acesulfame [19], *N*,*N*-diethyl-*m*-toluamide (DEET) [23], and caffeine [24]. Similarly, Murugananthan et al. [10], Cuiet al. [11], and Govindaraj et al. [12] also observed the increase in TOC or COD removal with increasing current density on BDD/Si,

BDD/Ti, and graphite, respectively. However, the time needed for complete mineralization of TOC in this study was shorter than that (14 h) needed on the BDD/Si at $C_0 = 20 \text{ mg/L}$ and 0.036 A/cm² in 0.1 M Na₂SO₄ and that (>2 h) on the graphite at $C_0 = 500 \text{ mg/L}$ and 0.020 A/cm² in 0.05 M NaCl. Only ~78% TOC removal was achieved on the BDD/Ti at $C_0 = 100 \text{ mg/L}$ in 0.1 M Na₂SO₄ [11]. The different results found in this study and the earlier works are mainly due to the difference in operating conditions. Accordingly, the current density of 0.50 A/cm² was used in the subsequent experiments.



Figure 3. Effect of current density on (a) BPA degradation (inset: ln(C₀/C) against time) and (b) TOC removal on BDD in 1M Na₂SO₄ at 25°C.

3.3. Effect of temperature on BPA electrochemical degradation

With increasing temperature (25°C, 50°C and 75°C), the degradation of BPA decreased (Fig. 4a) but the TOC removal increased (Fig. 4b). Complete BPA degradation took five minutes longer at

75°C than at 25°C and 50°C. Nevertheless, an opposite trend was found for complete TOC removal, which needed 40, 90, and ~90 min at 25°C, 50°C and 75°C, respectively. The results for TOC removal are associated with the fact that an increase in temperature decreases solution viscosity and increases mass transfer [25], while those for BPA degradation are related to the polymeric product deposition on the surface of BDD [10]. It is also possible that more •OH was generated at a higher temperature to enhance the TOC removal. The pseudo first-order rate constants (*k*) of BPA degradation were 5.39×10^{-3} (> 1.28×10^{-4} s⁻¹ on BDD/Si [10]), 4.10×10^{-3} , and 1.97×10^{-3} s⁻¹ at 25°C, 50°C and 75°C, respectively. Therefore, the temperature of 75°C was used in subsequent experiments.



Figure 4. Effect of temperature on (a) BPA degradation (inset: $ln(C_0/C)$ against time) and (b) TOC removal on BDD at 0.50 A/cm².

3.4. Effect of pH on BPA electrochemical degradation

Figure 5 shows the solution pH (3, 7, and 10 buffered by phosphate buffer) on BPA degradation and TOC removal on BDD at 0.5 A/cm² and 75°C. At 15 min, the BPA degradation

efficiency was 100% at pH 3 and 10, while five more minutes were needed to achieve the same BPA degradation efficiency at pH 7. Furthermore, the electrolysis time required for complete TOC removal was 40 min at pH 3 and 50 min at both pH 7 and 10. The *k* values of BPA degradation were $(3.00-6.06)\times10^{-3}$ s⁻¹ in these solutions.

The slight influence of solution pH on the BPA degradation and TOC removal is likely related to the difference in solution phosphate concentration. It was reported that •PO4²⁻ radicals and peroxophosphate (P₂O₈⁴⁻) might be formed during electrolysis [26]. Although the formation of •SO4⁻ radicals and peroxosulfate (S₂O₈²⁻) might also affect the result [27], the original sulfate concentrations were the same (1 M) in the three phosphate buffer solutions. It was reported that a BPA molecule has two ionizable hydrogen atoms with a pK_a value of 9.59–11.30 [10,28], exists predominantly in the ionized form at pH 10, which could be more readily attacked by the electrophilic •OH and rapidly oxidized [10]. However, this is not the case in the current study, so the molecular or dissociated forms of BPA did not play an important role in the BPA degradation. This is also true when considering the shift of equilibrium potential (E_{eq}) for the electrochemical •OH generation on BDD at the three pH according to the reaction of BDD + H₂O \rightarrow BDD(•OH) + H⁺ + e⁻ (weakly adsorbed •OH formed by water electrolysis at the anode surface) [29] and the corresponding Nernst equation of E_{eq} = E_{eq}^o– (2.303RT/F) pH (where R, T, and F have their usual meanings).



Figure 5. Effects of solution pH on BPA degradation and TOC removal (inset) on BDD at 0.5 A/cm² and 75°C.

3.5. Effects of Ce(IV) addition on BPA electrochemical degradation in 1M Na₂SO₄ and MWTPSE

According to our earlier study, Ce(IV) is a suitable oxidant to be used for the degradation of organic emerging contaminants (e.g., acetaminophen) via metal ion mediated electrochemical

oxidation [13]. Therefore, in this work we examined the effects of Ce(IV) addition on BPA electrochemical degradation in 1 M Na₂SO₄ and MWTPSE. At 0.5 A/cm² and 75°C 1 M Na₂SO₄, complete BPA degradation was achieved at 20 min on BDD for electrolysis (E) alone ($k = 4.10 \times 10^{-3} \text{ s}^{-1}$), whereas the corresponding time was only 5 min when the BPA electrolysis was performed with the addition of 250 mg/L Ce(IV); moreover, this time was even shorter (one minute) when Ce(IV) alone was used as the oxidant without electrolysis (Fig. 6a). On the other hand, total TOC removal was only achieved when using electrolysis alone at 60 min, and the other two operation modes merely achieved ~85–90% TOC removal at 60–120 min, although the TOC removal rate was in the order of Ce(IV) >E+Ce(IV) > E within 30 min (Fig. 6b). This finding is associated with the fact that •OH is more powerful than Ce(IV) for mineralization of TOC, because the oxidation potential is higher for the former (2.80 V) than for the latter (1.44–1.61 V). Additionally, organic acid intermediates might form before mineralization to CO₂ and H₂O [11]. It is thus inferred that Ce(IV) failed to oxidize these acid intermediates.

Similar tests were also conducted in municipal wastewater treatment plant secondary effluent (MWTPSE) spiked with 1 M Na₂SO₄ and 100 mg/L BPA (Fig. 7a). The trend of BPA electrolysis with/without250 mg/L Ce(IV) addition in MWTPSE was similar to that in 1 M Na₂SO₄. The BPA electrochemical degradation efficiency when adding chemical Ce(IV) ($k = 7.67 \times 10^{-3} \text{ s}^{-1}$) was similar to that when adding Ce(IV) regenerated (R-Ce(IV)) from real spent thin-film transistor liquid-crystal display (TFT-LCD) Cr-etching solutions ($k = 8.34 \times 10^{-3} \text{ s}^{-1}$) [15–18]. However, the TOC removal was better when adding chemical Ce(IV) than R-Ce(IV), because the regenerated Cr-etching solutions contained abundant NH₄⁺ and its oxidation might compete with •OH production on BDD (Fig. 7b).

This result is consistent with the concentration variation of *p*-benzoquinone formed during electrolysis (Fig. 7a inset).





Figure 6. Effects of Ce(IV) addition on (a) BPA electrochemical degradation and (b) TOC removal in 1 M Na₂SO₄ (E: electrolysis; E + Ce(IV) (250 mg/L): electrolysis plus Ce(IV) oxidation; Ce(IV): Ce(IV) alone (250 mg/L)) at 0.5 A/cm² and 75°C.

A similar curve of *p*-benzoquinone formation and degradation at BPA $C_o = 100 \text{ mg/L}$ and 0.01 A/cm²in 0.1 M Na2SO₄ was also reported by Cui et al., who used a BDD/Ti anode [11].





Figure 7. Effects of Ce(IV) addition (250 mg/L) on (a) BPA electrochemical degradation (inset: *p*-benzoquinone) and (b) TOC removal in municipal wastewater treatment plant secondary effluent (MWTPSE) (E: electrolysis; E + Ce(IV): electrolysis plus Ce(IV) oxidation; E + R-Ce(IV): electrolysis plus regenerated (R) Ce(IV) oxidation; Ce(IV): Ce(IV) oxidation) at 0.5 A/cm² and 75°C.

p-benzoquinone is regarded as one of the important one-ring aromatic intermediates [11] during the electrochemical degradation of phenolic chemicals (e.g., BPA), which may be formed from the cleavage of the isopropylidene bridge [30], and can then be further oxidized to aliphatic acids and finally mineralized to carbon dioxide [11]. The formation and degradation rate of *p*-benzoquinone followed the order E+Ce(IV) > E > E+R-Ce(IV). Most of the *p*-benzoquinone was not degraded when using Ce(IV) alone without electrolysis, which also supports the results shown in Fig. 6b.

3.6. Effects of environmental water matrix on BPA electrochemical degradation

To further explore the effects of the environmental water matrix on BPA electrochemical degradation, four different types of water matrices (municipal wastewater treatment plant primary and secondary effluents (MWTPPE and MWTPSE, respectively), groundwater (GW), and river water (RW)) were tested in this work. Regardless of the similar degradation efficiencies of BPA in all tested matrices ($k = (3.68-6.07)\times10^{-3}$ s⁻¹) (Fig. 8a), the completion of total TOC removal was achieved in MWTPPE, MWTPSE, and GW at 40–50 min, but it could not be obtained in RW, even at 120 min (Fig. 8b). Furthermore, complete *p*-benzoquinone degradation was observed in the four matrices, although it was faster (by 15–50 min) in MWTPPE and MWTPSE than in GW and RW. According, it is inferred that BPA and its degradation intermediates were easier to be degraded than the inherent organic compounds in RW.



Figure 8. BPA electrochemical degradation (inset: *p*-benzoquinone) (a) and TOC removal (b) in municipal wastewater treatment plant primary effluent (MWTPPE), municipal wastewater treatment plant secondary effluent (MWTPSE), river water (RW), and groundwater (GW) at 0.5 A/cm² and 75°C.

4. CONCLUSIONS

In this study we tested the BPA degradation and TOC removal on BDD in different water matrices. No peak for BPA oxidation in CV analysis was identified, so the BPA degradation in electrolysis should be attributed mainly to •OH, and not the direct electro-oxidation of BPA on BDD. The BPA degradation and TOC removal at 25°C increased along with the current density (0.25-0.75 A/cm²); however, the BPA degradation or TOC removal was similar at 0.50 and 0.75 A/cm². With increasing temperature (25° C, 50° C and 75° C), the degradation of BPA decreased but the TOC

removal increased. The pseudo first-order rate constants (*k*) of BPA degradation were 5.39×10^{-3} , 4.10×10^{-3} , and 1.97×10^{-3} s⁻¹ at 25°C, 50°C and 75°C, respectively. The BPA degradation (*k* = (3.00–6.06)×10⁻³ s⁻¹) or TOC removal at 0.5 A/cm² and 75°C in pH 3–10 phosphate buffer solutions was similar.

At 0.5 A/cm² and 75°C in 1 M Na₂SO₄, the complete BPA degradation was achieved at 20 min for electrolysis (E) alone ($k = 4.10 \times 10^{-3} \text{ s}^{-1}$); however, the BPA degradation efficiency and TOC removal rate was in the order Ce(IV) >E+Ce(IV) > E for these three different processes, while complete TOC removal was only achieved using electrolysis (E) alone. The trend of BPA electrolysis with/without Ce(IV) addition in MWTPSE spiked with 1 M Na₂SO₄ and 100 mg/L BPA was similar to that in 1 M Na₂SO₄. The BPA electrochemical degradation efficiency when adding Ce(IV) (k =7.67×10⁻³ s⁻¹) was similar to that when adding R-Ce(IV) ($k = 8.34 \times 10^{-3} \text{ s}^{-1}$). However, the TOC removal was better when adding chemical Ce(IV) than when adding R-Ce(IV). The rates of *p*benzoquinone formation and degradation followed the order E+Ce(IV) > E > E+R-Ce(IV). Most of the *p*-benzoquinone was not be degraded when using Ce(IV) alone without electrolysis. Similar degradation efficiencies of BPA in MWTPPE, MWTPSE, RW, and GW were observed ($k = (3.68-6.07)\times 10^{-3} \text{ s}^{-1}$), but the complete TOC removal could not be obtained in RW. Furthermore, the complete *p*-benzoquinone degradation was observed in the four matrices, although it was faster in MWTPPE and MWTPSE than in GW and RW. According, it is inferred that BPA and its degradation intermediates were easier to be degraded than the inherent organic compounds in RW.

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