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ORIGINAL PAPER

Electrochemical oxidation of succinic acid in aqueous solutions using boron doped diamond anodes

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Abstract In this work, the electrochemical oxidation of succinic acid on boron-doped diamond (BDD) anodes was investigated. Voltammetric study had shown that no peaks appeared in the region of electrolyte stability which indicates that succinic acid oxidation can take place at a potential close to the potential region of electrolyte oxidation. Galvanostatic electrolyses achieved total chemical oxygen demand (COD) removals and high mineralization yields under different operating conditions (initial COD, current density and nature of supporting electrolyte). Oxalic, glycolic and formic acids were the main intermediates detected during anodic oxidation of succinic acid on BDD electrode and carbon dioxide as the final product. The mean oxidation state of carbon reached the value of 4 at the end of electrolysis which is indicative of mineralization of almost all organics present in aqueous solution. The exponential profile of COD versus specific electrical charge has shown that mass transfer is the limiting factor for the kinetics of electrochemical process. A simple mechanism was proposed for the mineralization of succinic acid. First, hydroxyl radicals attack of succinic acid leading to formation of glycolic, glyoxylic, fumaric and maleic acids. Then, theses acids undergo rapid and non-selective oxidation by hydroxyl radicals to be transformed into oxalic and formic acids which leads to further oxidation steps to mineralize these acids into carbon dioxide and water.

Keywords Anodic oxidation · Carboxylic acids · Chemical oxygen demand · Hydroxyl radicals · Mineralization · Total organic carbon

Introduction

Recently, the electrochemical oxidation on Boron-doped diamond (BDD) anodes has become a promising technique for water and wastewater treatment; this fact is attributed to the production of large amounts of hydroxyl radicals from water on BDD surface (Marselli et al. 2003; Michaud et al. 2003). This electrochemical method has been used at both laboratory and industrial scale to treat wastewaters contaminated with organic pollutants. Numerous previous studies focused on the treatment of wastewaters contaminated with toxic aromatic compounds such as phenol (Hagans et al. 2001; Iniesta et al. 2001; Panizza et al. 2002; Weiss et al. 2008; Yavuz et al. 2008) and phenol derivatives (Bensalah et al. 2009; Bensalah and Gadri 2005; Canizares et al. 2004, 2005a, b; Bensalah et al. 2005; Pacheco et al. 2007; Rodrigo et al. 2001), herbicides (Brillas et al. 2004, 2007; Boye et al. 2006; Sires et al. 2006), dyes (Ahmadi et al. 2007; Butron et al. 2007; Canizares et al. 2006a; Chen and Chen 2006; Saez et al. 2007) and others (Canizares et al. 2006b, 2007; Carter and Farrell 2008; Kapalka et al. 2008a, b; Kraft 2007; Louhichi et al. 2008a, b; Martinez-Huitle and Brillas 2009; Panizza and Cerisola 2009; Prabhakaran et al. 2009; Quiroz Alfaro et al. 2006). Also, electrochemical treatment of other bio-resistant organic pollutants such as aliphatic compounds was investigated by few researches using BDD anodic oxidation. Additionally, the electrochemical oxidation, especially anodic oxidation on BDD and other anodes of oxalic, acetic, maleic, fumaric, formic and tartaric acids

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(Canizares et al. 2003, 2008; Chailapakul et al. 2000; Drogui et al. 2007; Gandini et al. 2000; Ivandini et al. 2006; Kapalka et al. 2008c; Martinez-Huitle et al. 2008; Onofrio et al. 2008) was previously studied. These previous studies (Canizares et al. 2003; Gandini et al. 2000; Ivandini et al. 2006; Martinez-Huitle et al. 2008; Onofrio et al. 2008) showed that oxalic acid was completely transformed into CO_2 and H_2O in most cases, which is encouraging to test the feasibility of this process for destruction of succinic acid.

The goal of the work described here was to study electrochemical oxidation of succinic acid using boron-doped diamond anodes. Carboxylic acids are generally used as additives in food and pharmaceutical industries and are used as corrosion inhibitors, as lubricants; in addition they are main intermediaries formed during the manufacture of plastics (Franch et al. 2002; Rup et al. 2009). The specific objectives of this study were to (a) demonstrate the treatment efficiency of synthetic aqueous solutions containing succinic acid using electrochemical oxidation on BDD electrodes; (b) evaluate the effects of several experimental parameters such as initial Chemical oxygen demand (COD) concentration, current density and nature of supporting electrolyte on the rate and efficiency of the treatment process and (c) establish simple mechanism to describe the mineralization of succinic acid.

The current study was carried out at Laboratory of electrochemical Engineering at the Universtity of Gabes, Gabes, Tunisia during the period March 2010 to September 2010. Chromatography analyses were completed at QWE lab at Texas A&M at Qatar, Doha, Qatar.

Experimental section

Chemicals

Succinic, formic, maleic, fumaric, glycolic, glyoxylic and oxalic acids (>99%) were of analytical grade and obtained from Sigma-Aldrich. The other chemicals were of analytical grade and obtained from Fluka or Merck and used as received. All solutions were prepared with deionized water having 18 $m\Omega\ cm^{-1}$ resistivity from a Mill-Q^{TM} system.

Analytical procedure

The carbon concentration was measured using a Shimadzu TOC-5050 analyzer. Chemical Oxygen Demand (COD) was determined using a HACH DR2000 analyzer. Carboxylic acids were quantified by HPLC using a Supelcogel H column (mobile phase, 0.15% phosphoric acid solution; flow rate, 0.15 ml min^{-1}). The UV detector was set at 210 nm.

Determination of mean oxidation state of carbon

The mean oxidation state of carbon (MOSC) can be calculated from the values of TOC and COD using Eq. 1 (Vogel et al. 2000):

$$\mathrm{MOSC} = 4\left(1 - \frac{\mathrm{COD}}{\mathrm{TOC}}\right) \tag{1}$$

where COD is the chemical oxygen demand (in mol O_2/L) and TOC is the total carbon (in mol C/L), respectively. This parameter oscillates between −4 for the minor oxidation state (CH_4) and +4 for the major oxidation state (CO_2).

Determination of average current efficiency (ACE)

The chemical oxygen method was used for the current efficiency determination of the succinic acid oxidation. In this method, the COD was measured during electrolysis and the average current efficiency (ACE) was calculated using Eq. 2 (Canizares et al. 2005a; Louhichi et al. 2008a):

$$\mathrm{ACE} = \frac{(\mathrm{COD}_0 - \mathrm{COD}_t)FV}{8I\Delta I} \tag{2}$$

where COD_0 and COD_t are the chemical oxygen demands (in g O_2./L) at times 0 and t (in seconds), respectively; I is the current intensity (in A), F is Faraday constant (96,487 C./mol), V is the volume of the electrolyte (in L) and 8 is a dimensional factor for unit consistency {[(32 g O_2/(mol of O_2)]/[4 mol of e^-/(mol of O_2)]}.

Preparation of boron-doped diamond electrode

Boron-doped diamond films were provided by CSEM (Switzerland) and synthesized by the hot-filament chemical vapor deposition technique (HF CVD) on single-crystal p-type Si <100> wafers (0.1 cm, Siltronix). The temperature range of the filament was 2,440–2,560°C and that of the substrate was 830°C. The reactive gas was methane in excess hydrogen (1% CH_4 in H_2). The dopant gas was trimethylboron with a concentration of 3 ppm. The gas mixture was supplied to the reaction chamber at a flow rate of 5 dm^3/min, providing a growth rate of 0.24 μm/h for the diamond layer. The resulting diamond film thickness was about 1 μm. This HF CVD process produces columnar, random texture and polycrystalline films with an average resistivity of 0.01 Ω·cm. Prior to being used in galvanostatic electrolysis assays, the electrode was polarized for 30 min with a 1-M H_3PO_4 solution at 50 mA cm^{-2} to remove any impurity from its surface.

Voltammetry experiments

Electrochemical measurements were performed using a conventional three-electrode cell in conjunction with a computer-controlled potentiostat/galvanostat (Autolab Model PGSTAT 30, Eco Chemie B.V, Utrecht, Netherlands). Diamond was used as the working electrode, $Hg/Hg_2Cl_2 \cdot KCl$ (sat) as a reference and platinum as a counter electrode. The BDD electrode was circular with a cross-sectional area of 1 cm^2. Voltammetry experiments were performed in unstirred solutions (100 ml). Anode was anodically polarized for 5 min with a 1 M H_2SO_4 solution at 0.1 A prior to each experiment.

Galvanostatic electrolysis set-up

The galvanostatic electrolyses were carried out in a single-compartment electrochemical flow cell (Fig. 1). Diamond-based material was used as anode and stainless steel (AISI 304) as the cathode. Both electrodes were circular (100 mm diameter) with a cross-sectional area of 78 cm^2 each and an electrode gap of 9 mm. The electrolyte was stored in a glass container (500 ml) and circulated through the electrolytic cell using a centrifugal pump. The linear velocity of the fluid was 2.31 cm/s. A heat exchanger was used to maintain the temperature at the desired value. The experimental setup also contained a cyclone for gas–liquid separation and a gas absorber to collect the carbon dioxide contained in the gases evolved from the reactor into sodium hydroxide.

Results and discussion

Voltammetric study

Figure 2 presents oxidative voltamograms of succinic acid (1 mM) on BDD anode in an aqueous solution containing 0.1 M Na_2SO_4 at a scan rate of 100 mV/s. A cyclic voltammogram without organic matter is shown in Fig. 2a for the sake of comparison. As can be observed, polarization curves showed that no oxidation peaks appeared in the potential region of electrolyte stability. In addition, no oxygen evolution potential shifts were observed in the presence of succinic acid as was observed in the case of other carboxylic acids and glycols (Canizares et al. 2003; Gandini et al. 2000; Ivandini et al. 2006; Louhichi et al. 2008b; Martinez-Huitle et al. 2008; Onofrio et al. 2008). This could be due to the fact that succinic acid oxidation on BDD surface takes place at a potential, very close to that of the electrolyte oxidation. The absence of anodic peaks evidences that the succinic acid oxidation on BDD anodes is much more difficult than aromatic compounds' oxidation such as phenols, herbicides, dyes and surfactants (Bensalah et al. 2009; Bensalah and Gadri 2005; Boye et al. 2006; Canizares et al. 2006b; Louhichi et al. 2008b; Martinez-Huitle and Brillas 2009; Bensalah et al. 2005; Rodrigo et al. 2010; Sires et al. 2006; Weiss et al. 2008). Similar results were reported by other studies on aliphatic carboxylic acids and alcohols (Canizares et al. 2003, 2008; Gandini et al. 2000; Ivandini et al. 2006; Louhichi et al. 2008b; Martinez-Huitle et al. 2008; Onofrio et al. 2008).

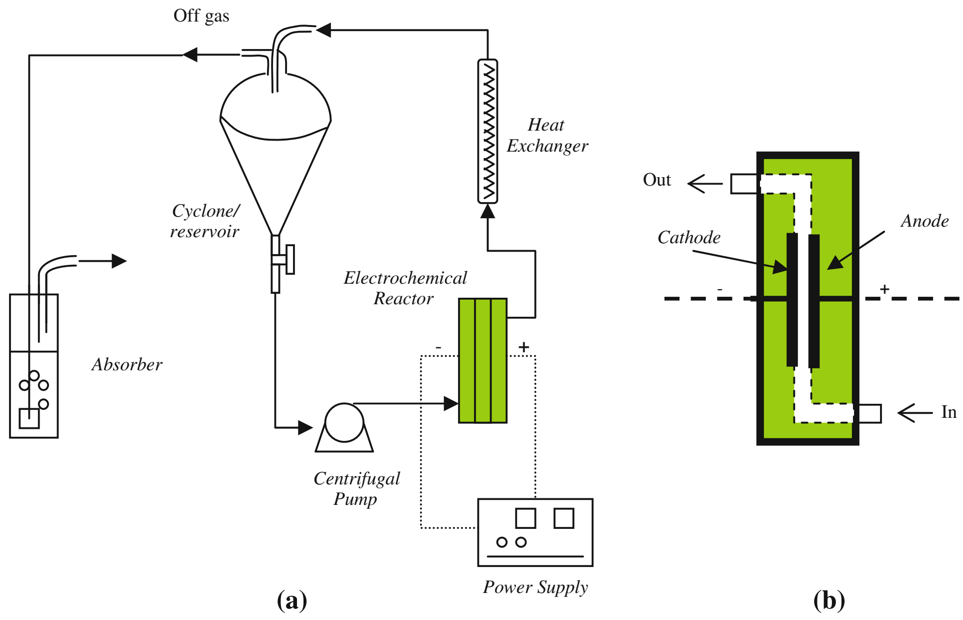

Fig. 1 Experimental setup: **a** layout of semi-pilot plant; **b** details of the electrochemical cell

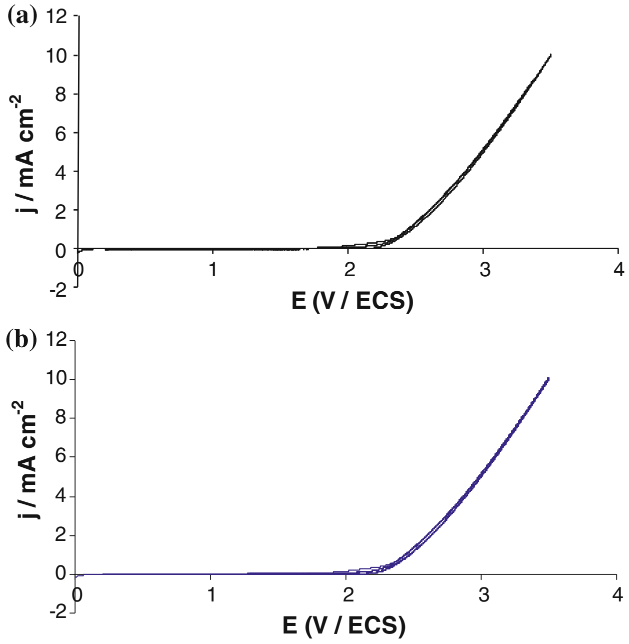

Fig. 2 Cyclic voltammograms on BDD anode (1 cm^2) of (**a**) 0.1 M Na_2SO_4, (**b**) 0.1 M Na_2SO_4 + 1 mM succinic acid. Experimental conditions: pH = 2; scan rate 100 $mV \cdot s^{-1}$; auxiliary electrode: Platinium; reference electrode: SCE

Moreover, voltammetric behavior of succinic acid indicates that succinic acid is much more difficult to oxidize than aromatic acids such as benzoic and salicylic acids (Montilla et al. 2002; Louhichi et al. 2006) which can be explained in terms of chemical structure (aromatic or aliphatic). It appears that radical intermediates of aromatics are more stable than those of aliphatics because of double-bond delocalization which facilitates electrochemical oxidation of aromatics. Moreover, anodic oxidation of succinic acid on BDD electrode is more difficult than that of aliphatic carboxylic monoacids (oxalic, fumaric, maleic, formic), glycols and aliphatic alcohols (Canizares et al. 2003; Gandini et al. 2000; Ivandini et al. 2006; Kapalka et al. 2008c; Louhichi et al. 2008b; Martinez-Huitle et al. 2008; Onofrio et al. 2008). This is probably due to the presence of two carboxyl groups in its chemical structure. The electronic withdrawing effect of these groups makes the oxidation of succinic acid more difficult.

According to our previous studies (Bensalah et al. 2009; Bensalah and Gadri 2005; Louhichi et al. 2008a, b), the indirect oxidation by strong oxidizing reagents such as hydroxyl radicals which can be electrogenerated from the oxidation of electrolyte can take place and play an important role in electrochemical process with BDD anodes. Galvanostatic electrolyses using BDD anodes can be used to mineralize succinic acid into CO_2 and H_2O or transform it to easily bio-degradable substances.

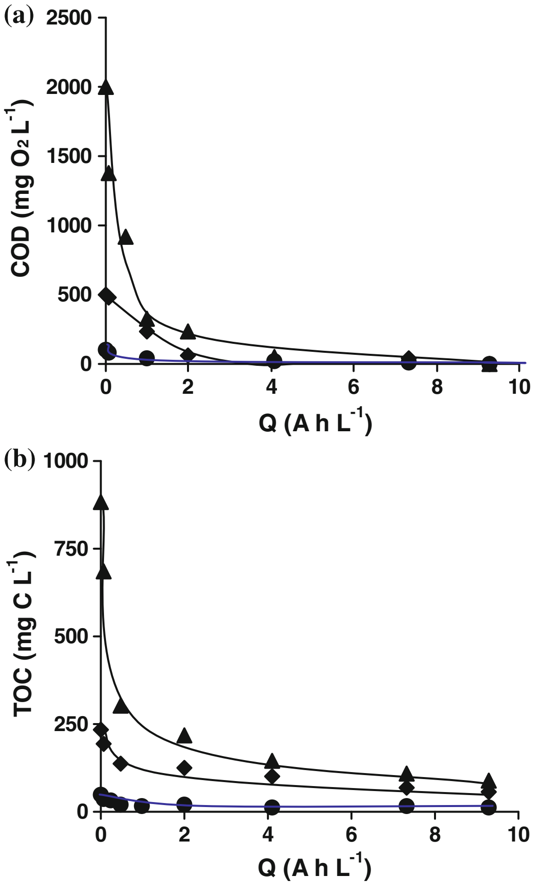

Fig. 3 Influence of initial COD concentration on the changes of (**a**) COD and (**b**) TOC with specific electrical charge during galvanostatic electrolyses of succinic acid. Experimental conditions: Current density: $j = 30$ mA cm^{-2}; $T = 25°C$; pollutant composition: $COD_0 = 2{,}000$ mg O_2 dm^{-3} (*filled triangle*), $COD_0 = 500$ mg O_2 dm^{-3} (*filled diamond*) et $COD_0 = 100$ mg O_2 dm^{-3}(*filled circle*), 0.1 M de Na_2SO_4 and pH = 2

Galvanostatic electrolyses

Figure 3 shows the effect of initial COD concentration on the changes of COD and TOC with specific electric charge during galvanostatic electrolyses ($j = 30 mAcm^{-2}$) of aqueous solutions (0.1 M Na_2SO_4) containing different concentrations of succinic acid. COD and TOC were sufficiently destroyed towards the end of electrochemical treatment independently of succinic acid concentration. The continuous decrease of TOC indicated that galvanostatic electrolysis of succinic acid on BDD anodes leads to the formation of CO_2 from the beginning of electrolysis. Furthermore, the anodic oxidation on DDB has completely eliminated COD contained in the aqueous solution, but small amounts of refractory carbon have been detected especially when high succinic acid concentrations were electrolyzed. COD and TOC decay was very fast at the beginning of electrolysis up to 2 A h/L, and then it becomes slower as specific electric charge increases. An exponential profile of COD versus Q was observed and similar amount of electric charge were consumed in all cases. The results of these experiments can be summarized as follows:

- Mineralization of succinic acid can be accomplished by galvanostatic electrolysis on BDD anode. In contrast, voltammetric results showed absence of anodic peaks in the potential region of electrolyte stability.
- The electrochemical oxidation of succinic acid leads to the accumulation of several aliphatic intermediates difficult to be oxidized.
- The overall kinetics of electrochemical process is controlled by mass transfer for the range of concentration used in this study.

According to literature (Martinez-Huitle and Brillas 2009; Panizza and Cerisola 2009; Quiroz Alfaro et al. 2006; Rodrigo et al. 2010), these results can be explained by mediated electrochemical oxidation processes which play an important role in the anodic oxidation of succinic acid. Mediated oxidation of succinic acid by hydroxyl radicals and other oxidants electrogenerated from supporting electrolyte oxidation on BDD appears to be more important than the direct oxidation mechanism in the case of succinic acid. Similar results were previously observed in several cases during galvanostatic electrolyses of organics on BDD anodes (Canizares et al. 2006b; Louhichi et al. 2008a; Martinez-Huitle and Brillas 2009; Rodrigo et al. 2010; Saez et al. 2007).

Figure 4 presents the behavior of COD and TOC (Fig. 4a) and MOSC (Fig. 4b) with specific electrical charge during galvanostatic electrolysis ($j = 30$ mA cm^{-2}) on BDD anode of succinic acid aqueous solution (0.1 M Na_2SO_4) containing 2,000 mg O_2 L^{-1}. Experimental results shown in Fig. 4 show that COD decreased more rapidly than TOC towards the end of electrolyses. This indicates that a diversity of intermediates was formed and CO_2 was not the final product of succinic acid oxidation on BDD anodes. Figure 4b shows in the behavior of MOSC (mean oxidation state of carbon) during the process. This parameter ranges between −4 for the minor oxidation state of carbon (CH_4) and +4 for the major oxidation state (CO_2) and it is used to qualitatively assess the nature of the organics using measurements of TOC and COD. This parameter can give valuable information concerning the reaction progress during oxidation processes. In this work, MOSC was used mainly to confirm the formation of carbon dioxide during oxidation of organics when it reaches its maximum value of +4 during oxidation of different organics. Figure 4b shows that different intermediates can be formed during succinic acid oxidation because different values of MOSC were observed during the electrochemical oxidation process. Towards the end of electrolysis, MOSC reached a value close to +4 confirming that almost complete mineralization of organic carbon and only negligible amounts of refractory organics remain in water during the anodic oxidation of succinic acid on BDD electrodes.

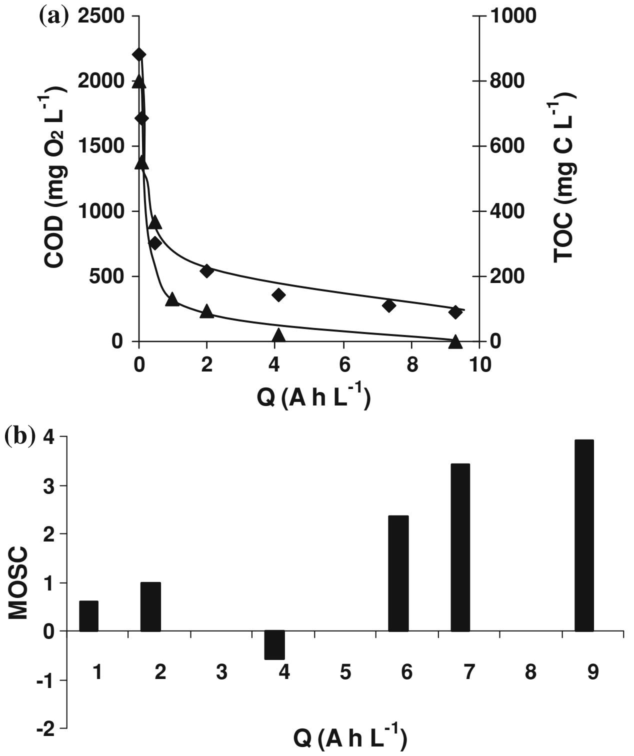

Fig. 4 Changes of (**a**) COD (*filled triangle*) and TOC (*open triangle*) with specific electrical charge; and (**b**) MOSC with COD during galvanostatic electrolyses of succinic acid. Experimental conditions: Current density: $j = 30$ mA cm^{-2}; $T = 25$°C; pollutant composition: $COD_0 = 2{,}000$ mg O_2 dm^{-3}, 0.1 M de Na_2SO_4 and pH = 2

A hypothesis was made that the electrochemical oxidation of succinic acid can lead to the formation of carboxylic acids such as glycolic, glyoxylic, fumaric, maleic, oxalic and formic acids. In order to test this hypothesis, identification and quantification of succinic acid BDD-anodic oxidation intermediates were carried out using high-performance liquid chromatography (HPLC). Figure 5 shows the effect of specific electric charge on the formation of these intermediates, during galvanostatic electrolysis ($j = 30$ mA cm^{-2}) on BDD anode of succinic acid aqueous solution (0.1 M Na_2SO_4) containing 2,000 mg O_2/L. As can be observed from Fig. 5a, the decay of succinic acid concentration is more rapid than that of TOC. More than 95% of succinic acid removal was achieved at specific electric charge of 5 A h/L while at this specific electrical charge, the TOC removal was only about 75%. It can be also observed that oxalic, glycolic and formic acids were the main intermediates formed with very low concentrations compared with the initial concentration of pollutant. Among these intermediates, oxalic acid was found to be

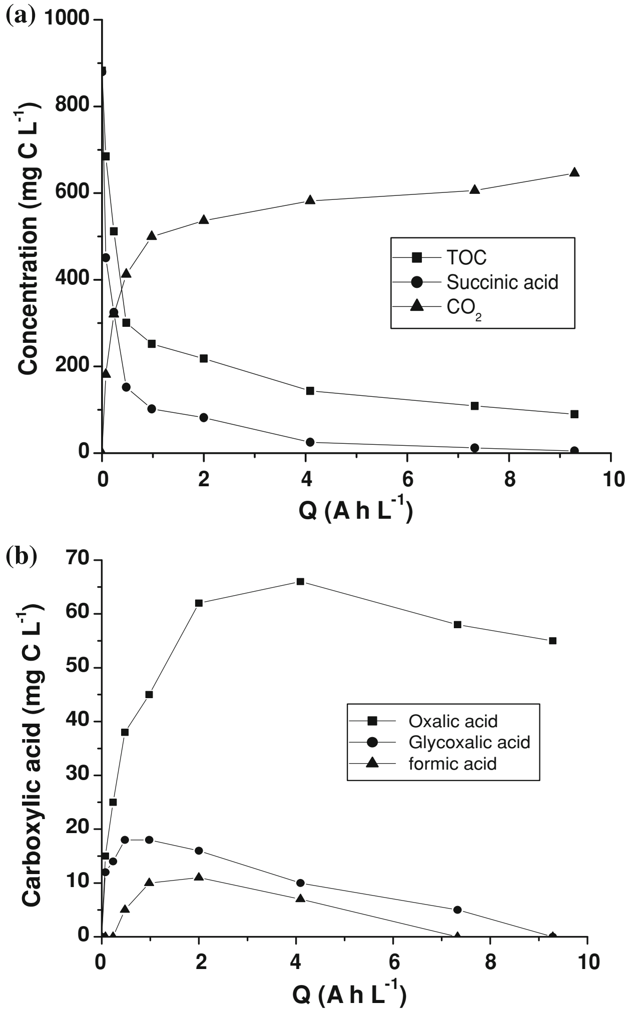

Fig. 5 Changes of (**a**) TOC, succinic acid and CO_2 and (**b**) carboxylic acid intermediates with specific electrical charge during galvanostatic electrolyses of succinic acid. Experimental conditions: Current density: $j = 30$ mA cm^{-2}; $T = 25°C$; pollutant composition: $COD_0 = 2{,}000$ mg O_2 dm^{-3}, 0.1 M de Na_2SO_4 and $pH = 2$

formed with higher concentration compared with glycolic and formic acids and it was persistent up to the end of electrolysis. The electrochemical oxidation of succinic acid using a BDD electrode leads to the rapid sequential formation of carboxylic acids (oxalic, glycolic, formic) and carbon dioxide; the latter was the sole final product. Little amounts of glyoxylic and maleic acids were also detected. The small concentration of intermediates suggests that, once the electrochemical oxidation of succinic acid starts, this process proceeds to the complete mineralization of the molecule in a zone close to the anode surface. This behavior was previously reported for the oxidation of other organics on BDD anodes (Canizares et al. 2006b; Louhichi et al. 2008a; Martinez-Huitle and Brillas 2009; Rodrigo et al. 2010; Saez et al. 2007) and supports the hypothesis that the oxidation of the organics on these electrodes occurs directly on the anode surface or at a small distance very close to this surface, mediated by hydroxyl radicals generated by water oxidation.

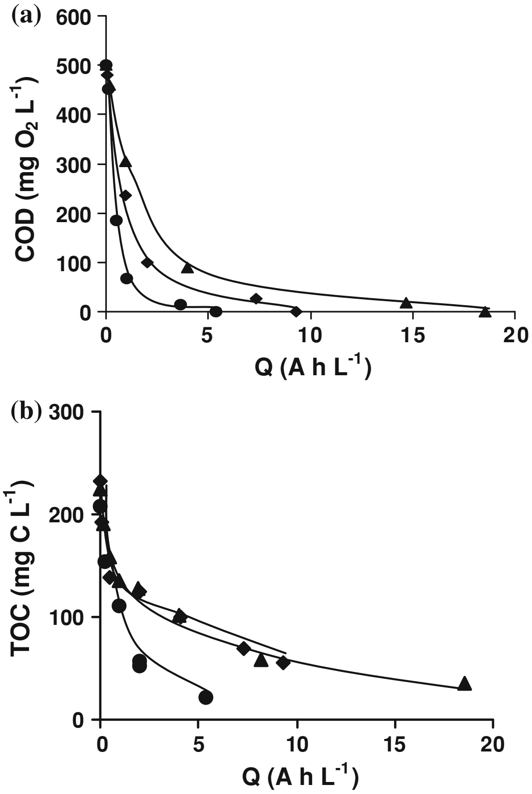

Fig. 6 Influence of current density on the changes (**a**) COD and (**b**) TOC with specific electrical charge of during galvanostatic electrolyses of succinic acid. Experimental conditions: Current density: $j = 15 mAcm^{-2}$ (*filled circle*), $j = 30$ mA cm^{-2} (*filled diamond*) and $j = 60$ mA cm^{-2} (*filled triangle*); $T = 25°C$; pollutant composition: $COD_0 = 500$ mg O_2 dm^{-3}, 0.1 M de Na_2SO_4 and $pH = 2$

Figure 6 shows the influence of current density on COD and TOC destruction with specific electrical charge during galvanostatic electrolyses of succinic acid aqueous solutions (0.1 M Na_2SO_4) containing 500 mg O_2/L. COD and TOC were totally eliminated from the solution regardless of the applied current density. Increase in current density did not significantly affect the rate and efficiency of succinic acid electrochemical oxidation on BDD anode in the range 15–60 mA cm^{-2}. Furthermore, the influence of the current density on the changes of ACE with COD (Fig. 7) leads to the following observations:

- For COD concentrations higher than 100 mg O_2/L, a significant influence of current density on ACE was observed. The increase of current density from 15 to 60 mA cm^{-2} resulted in decreasing ACE from 1 to 0.4. This can be explained by the fact that increase in current density favors side reactions of oxygen evolution and electrolyte oxidation but without affecting degradation of the succinic acid. At high current

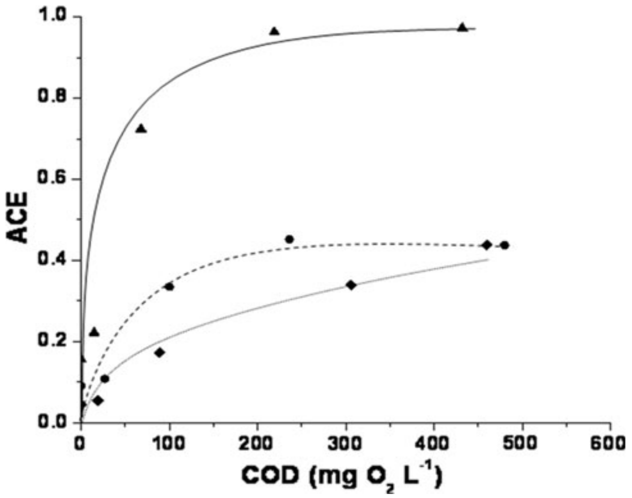

Fig. 7 Changes of ACE with COD during galvanostatic electrolyses of succinic acid. Experimental conditions: Current density: $j = 15$ mA cm^{-2} (*filled triangle*), $j = 30$ mA cm^{-2} (*filled circle*) and $j = 60$ mA cm^{-2} (*filled diamond*); $T = 25$°C; pollutant composition: $COD_0 = 500$ mg O_2 dm^{-3}, 0.1 M de Na_2SO_4 and pH = 2

densities, electrical charge consumed is mostly used by water oxidation into oxygen which increases unnecessarily electrochemical treatment costs.

- At low COD concentrations, no significant influence on the efficiency of anodic oxidation was observed. This has been explained in literature (Canizares et al. 2006b; Louhichi et al. 2008a; Martinez-Huitle and Brillas 2009; Rodrigo et al. 2010; Saez et al. 2007) in terms of the controlling mechanisms of the electrochemical process (electrodic or mass transfer). In these cases mass transfer mainly controls the overall oxidation kinetics.

Figure 8 shows the evolution of COD and TOC with specific electrical charge during galvanostatic electrolyses ($j = 30$ mA cm^{-2}) on BDD anode of succinic acid aqueous solutions ($COD_0 = 500$ O_2 mg/L) in the presence of three different supporting electrolytes: $NaClO_4$, Na_2SO_4 and NaH_2PO_4. High COD and TOC removals were obtained in the presence of three salts and the nature of salt did not have significant influence on the rate or efficiency of BDD-anodic oxidation of succinic acid. These results are different from the previously reported results obtained with aromatic compounds' (phenols, dyes and surfactants) degradation, leading to the conclusion that electrogenerated oxidants from the anodic oxidation of the electrolyte play a very important role in the oxidation process. In this context, it was previously reported (Martinez-Huitle and Brillas 2009; Martinez-Huitle and Ferro 2006; Rodrigo et al. 2010) that the electrochemical oxidation of wastewaters on conductive-diamond anodes can lead to the electrosynthesis of different oxidants such as peroxosulphates (Michaud et al. 2000), peroxophosphates (Canizares et al. 2005b), hypochlorite (Fernandes et al. 2004), etc. The type of these oxidants depends on the waste composition (e.g., pH and electrolytes) and on the operating conditions. As can be observed from Fig. 8, similar results were obtained with Na_2SO_4, $NaClO_4$ and NaH_2PO_4. In the presence of perchlorate ions (ClO_4^-), OH· radicals are the main electrogenerated oxidants on BDD surface from water discharge. However, in solutions containing sulfate and phosphate ions, in addition to hydroxyl radicals, other oxidants may be electrogenerated (e.g., persulfate and perphosphate ions, respectively). This indicates that hydroxyl radical are mainly responsible for the mineralization of succinic acid and its oxidation intermediates.

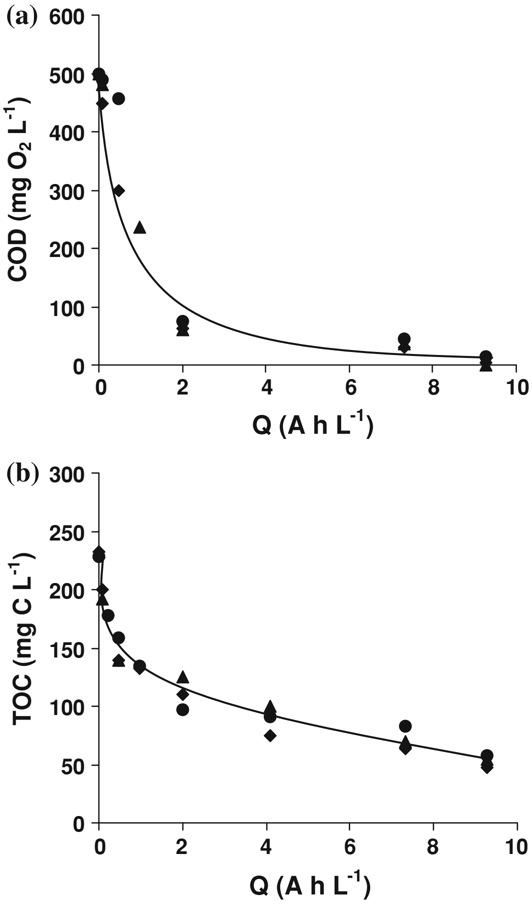

Fig. 8 Influence of supporting electrolyte on the changes of (**a**) COD and (**b**) TOC with specific electrical charge of during galvanostatic electrolyses of succinic acid. Experimental conditions: Current density: $j = 30$ mA cm^{-2}; $T = 25$°C; pollutant composition: $COD_0 = 500$ mg O_2 dm^{-3}, 0.1 M de Na_2SO_4 (*filled triangle*), 0.1 M de $NaClO_4$ (*filled circle*) et 0.1 M de NaH_2PO_4 (*filled diamond*) and pH = 2

Based on these results, a simple mechanism is proposed for succinic acid mineralization by electrochemical oxidation with BDD and is presented in Fig. 9. Hydroxyl

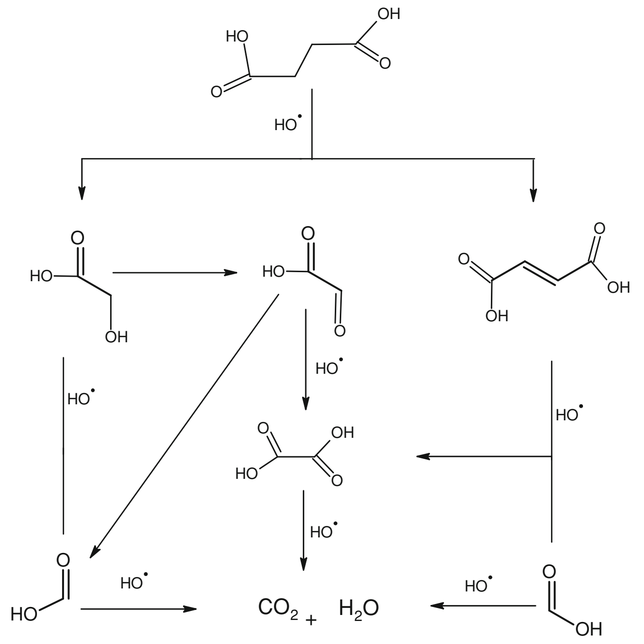

Fig. 9 Simple mechanism proposed for succinic acid mineralization by electrochemical oxidation on BDD anodes

radicals electrogenerated at the surface of BDD anode react with succinic acid to produce glycolic, glyoxylic, fumaric and maleic acids. Then, these intermediates undergo a rapid transformation into oxalic and formic acids which are mineralized to CO_2 and H_2O at the end of the process.

Conclusion

The main conclusions of this work can be summarized as follows:

- Voltammetric results have shown that electrochemical oxidation of succinic acid is difficult to be realized directly on BDD anode. This could be explained by the absence of anodic peaks in succinic acid cyclic voltammograms on the potential region of electrolyte stability.
- Galvanostatic electrolyses of succinic acid on BDD anodes allow achieving high removal efficiencies of both succinic acid and its intermediates.
- The main intermediates detected during galvanostatic electrolyses of succinic acid are oxalic, glycolic and formic acids. Changes in MOSC during galvanostatic electrolyses indicated the formation of different intermediates and confirmed that electrochemical oxidation of succinct acid on BDD electrodes leads to almost complete mineralization of organic carbon (MOSC close to +4).
- Current density and supporting electrolyte have no significant influence on kinetic and efficiency of electrochemical oxidation of succinic acid using BDD anodes. These results can be explained by mass transfer limitations and mediated oxidation of organics by hydroxyl radicals.

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References

Ahmadi MF, Bensalah N, Gadri A (2007) Electrochemical degradation of anthraquinone dye Alizarin Red S by anodic oxidation on boron-doped diamond. Dyes Pigments 73(1):86–89

Bensalah N, Gadri A (2005) Electrochemical oxidation of 2,4,6-trinitrophenol on boron-doped diamond electrodes. J Electrochem Soc 152(6):D113–D116

Bensalah N, Gadri A, Cañizares P, Sáez C, Lobato J, Rodrigo MA (2005) Electrochemical oxidation of hydroquinone, resorcinol, and cathecol on boron-doped diamond anodes. Environ Sci Tec 39(18):7234–7239

Bensalah N, Trabelsi H, Gadri A (2009) Electrochemical treatment of aqueous wastes containing pyrogallol by BDD-anodic oxidation. J Environ Manag 90(1):523–530

Boye B, Brillas E, Marselli B, Michaud PA, Comninellis Ch, Farnia G, Sandona G (2006) Electrochemical incineration of chloromethylphenoxy herbicides in acid medium by anodic oxidation with boron-doped diamond electrode. Electrochim Acta 51(14):2872–2880

Brillas E, Boye B, Sirés I, Garrido JA, Rodríguez RM, Arias C, Cabot PL, Comninellis Ch (2004) Electrochemical destruction of chlorophenoxy herbicides by anodic oxidation and electro-Fenton using a boron-doped diamond electrode. Electrochim Acta 49(25):4487–4496

Brillas E, Banos MA, Skoumal M, Cabot PL, Garrido JA, Rodriguez RM (2007) Degradation of the herbicide 2, 4-DP by anodic oxidation, electro-Fenton and photoelectro-Fenton using platinum and boron-doped diamond anodes. Chemosphere 68(2):199–209

Butron E, Juarez ME, Solis M, Teutli M, Gonzalez I, Nava JL (2007) Electrochemical incineration of indigo textile dye in filter-press-type FM01-LC electrochemical cell using BDD electrodes. Electrochim Acta 52(24):6888–6894

Canizares P, Garcia-Gomez J, Lobato J, Rodrigo MA (2003) Electrochemical oxidation of aqueous carboxylic acid wastes using diamond thin-film electrodes. Ind Eng Chem Res 42(5):956–962

Canizares P, Saez C, Lobato J, Rodrigo MA (2004) Electrochemical oxidation of polyhydroxybenzenes on boron-doped diamond anodes. Ind Eng Chem Res 43(21):6629–6637

Canizares P, Lobato J, Paz R, Rodrigo MA, Saez C (2005a) Electrochemical oxidation of phenolic wastes with boron-doped diamond anodes. Water Res 39(12):2687–2703

Canizares P, Larrondo F, Lobato J, Rodrigo MA, Saez C (2005b) Electrochemical synthesis of peroxodiphosphate using Boron-Doped Diamond anodes. J Electrochem Soc 152(11):D191–D196

Canizares P, Gadri A, Lobato J, Bensalah N, Rodrigo MA, Saez C (2006a) Electrochemical oxidation of Azoic Dyes with Conductive-Diamond Anodes. Ind Eng Chem Res 45(10):3468–3473

Canizares P, Martinez L, Paz R, Saez C, Lobato J, Rodrigo MA (2006b) Treatment of Fenton-refractory olive-oil-mills wastes by electrochemical oxidation with Boron Doped Diamond Anodes. J Chem Technol Biotech 81(8):1331–1337

Canizares P, Louhichi B, Gadri A, Bensalah N, Paz R, Rodrigo MA, Saez C (2007) Electrochemical treatment of the pollutants generated in an ink-manufacturing process. J Hazard Mater 146(3):552–557

Canizares P, Paz R, Saez C, Rodrigo MA (2008) Electrochemical oxidation of alcohols and carboxylic acids with diamond anodes: A comparison with other advanced oxidation processes. Electrochim Acta 53(5):2144–2153

Carter KE, Farrell J (2008) Oxidative Destruction of Perfluorooctane sulfonate using boron-doped diamond film electrodes. Environ Sci Tech 42(16):6111–6115

Chailapakul O, Popa E, Tai H, Sarada BV, Tryk DA, Fujishima A (2000) The electrooxidation of organic acids at boron-doped diamond electrodes. Electrochem Comm 2(6):422–426

Chen X, Chen G (2006) Anodic oxidation of Orange II on Ti/BDD electrode: Variable effects. Sep Pur Technol 48(1):45–49

Drogui P, Blais JF, Mercier G (2007) Review of electrochemical technologies for environmental applications. Recent patents on engineering 1:257

Fernandes A, Morao A, Magrinho M, Lopes A, Goncales I (2004) Electrochemical degradation of C.I. acid Orange 7. Dyes and Pigments 61(3):287–296

Franch MI, Ayllon JA, Peral J, Domenech X (2002) Photocatalytic degradation of short-chain organic diacids. Catalysis Today 76(2–4):221–233

Gandini D, Mahe E, Michaud PA, Haenni W, Perret A, Comninellis Ch (2000) Oxidation of carboxylic acid at boron-doped diamond electrodes. J Appl Electrochem 30(12):1345–1350

Hagans PL, Natishan PM, Stoner BR, O'Grady WE (2001) Electrochemical oxidation of phenol using boron-doped diamond electrodes. J Electrochem Soc 148(7):E298–E301

Iniesta J, Michaud PA, Panizza M, Cerisola G, Aldaz A, Comninellis Ch (2001) Electrochemical oxidation of phenol at boron-doped diamond electrode. Electrochim Acta 46(23):3573–3578

Ivandini TA, Rao TN, Fujishima A, Einaga Y (2006) Electrochemical oxidation of oxalic acid at highly Boron-Doped Diamond electrodes. Anal Chem 78(10):3467–3471

Kapalka A, Foti G, Comninellis Ch (2008a) Kinetic modeling of the electrochemical mineralization of organic pollutants for wastewater treatment. J Appl Electrochem 38(1):7–16

Kapalka A, Lanova B, Baltruschat H, Foti G, Comninellis Ch (2008b) Electrochemically induced mineralization of organics by molecular oxygen on boron-doped diamond electrode. Electrochem. Comm 10(9):1215–1218

Kapalka A, Lanova B, Baltruschat H, Foti G, Comninellis Ch (2008c) DEMS Study of the acetic acid oxidation on Boron-Doped Diamond electrode. J Electrochem Soc 155:E96–E100

Kraft A (2007) Doped Diamond: a compact review on a new versatile electrode material. IntJ Electrochem Sci 2(5):355–385

Louhichi B, Bensalash N, Gadri A (2006) Electrochemical oxidation of benzoic acid derivatives on Boron Doped Diamond: voltammetric study and galvanostatic electrolyses. Chem Eng Tech 29(8):944–950

Louhichi B, Ahmadi MF, Bensalah N, Gadri A, Rodrigo MA (2008a) Electrochemical degradation of an anionic surfactant on boron-doped diamond anodes. J Hazard Mater 158(2–3):430–437

Louhichi B, Bensalah N, Gadri A (2008b) Electrochemical oxidation of glycols on boron-doped diamond anode. J Environ Eng Manage 18(3):231–237

Marselli B, Garcia-Gomez J, Michaud PA, Rodrigo MA, Comninellis Ch (2003) Electrogeneration of hydroxyl radicals on boron-doped diamond electrodes. J Electrochem Soc 150(3):D79–D83

Martinez-Huitle CA, Brillas E (2009) Decontamination of wastewaters containing synthetic organic dyes by electrochemical methods: a general review. Appl Catal B: Environ 87(3–4):105–145

Martinez-Huitle CA, Ferro S (2006) Electrochemical oxidation of organic pollutants for the wastewater treatment: direct and indirect processes. Chem Soc Rev 35(12):1324–1340

Martinez-Huitle CA, Ferro S, Reyna S, Cerro-Lopez M, De Battisti A, Quiroz MA (2008) Electrochemical oxidation of oxalic acid in the presence of halides at Boron Doped Diamond electrode. J Braz Chem Soc 19(1):150–156

Michaud PA, Mahe E, Haenni W, Perret A, Comninellis Ch (2000) Preparation of peroxodisulfuric acid using boron-doped diamond thin-film Electrodes. Electrochem Solid State Lett 3(2):77–79

Michaud PA, Panizza M, Ouattara L, Diaco T, Foti G, Comninellis Ch (2003) Electrochemical oxidation of water on synthetic boron-doped diamond thin film anodes. App Electrochem 33(2):151–154

Montilla F, Michaud MA, Morallon E, Vazquez JL, Comninellis Ch (2002) Electrochemical oxidation of benzoic acid at boron-doped diamond electrodes. Electrochim Acta 47(21):3509–3513

Onofrio S, Alessandro G, Chiara G, Serena R, Giuseppe F (2008) Electrochemical incineration of oxalic acid at boron doped diamond anodes: role of operative parameters. Electrochim Acta 53(5):2095–2108

Pacheco MJ, Morao A, Lopes A, Ciriaco L, Goncalves I (2007) Degradation of phenols using boron-doped diamond electrodes: a method for quantifying the extent of combustion. Electrochim Acta 53(2):629–636

Panizza M, Cerisola G (2009) Direct and mediated anodic oxidation of organic pollutants. Chem Rev 109(12):6541–6569

Panizza M, Michaud PA, Iniesta J, Comninellis Ch, Cerisola G (2002) Electrochemical oxidation of phenol at boron-doped diamond electrode: application to electro-organic synthesis and wastewater treatment. Ann Chim 92(10):995–1006

Prabhakaran D, Kannadasan T, Ahmed Basha C (2009) Treatability of resin effluents by electrochemical oxidation using batch recirculation reactor. Int. J. Environ. Sci. Tech 6(3):491–498

Quiroz Alfaro MA, Ferro S, Martinez-Huitle CA, Vong YM (2006) Boron doped diamond electrode for the wastewater treatment. J Braz Chem Soc 17(2):227–236

Rodrigo MA, Michaud PA, Duo I, Panizza M, Cerisola G, Ch Comninellis (2001) Oxidation of 4-chlorophenol at boron-doped diamond electrodes for wastewater treatment. J Electrochem Soc 148(5):D60–D64

Rodrigo MA, Canizares P, Sanchez-Carretero A, Saez C (2010) Use of conductive-diamond electrochemical oxidation for water treatment. Catalysis Today 151(1–2):173–177

Rup S, Zimmermann F, Meux E, Schneider M, Sindt M, Oget N (2009) The ultrasound-assisted oxidative scission of monoenic fatty acids by ruthenium tetroxide catalysis: Influence of the mixture of solvents. Ultrasonics Sonochem 16(2):266–272

Saez C, Panizza M, Rodrigo MA, Cerisola G (2007) Electrochemical incineration of dyes using a boron-doped diamond anode. J Chem Technol Biotech 82(6):575–581

Sires I, Cabot PL, Centellas F, Garrido JA, Rodriguez RM, Arias C, Brillas E (2006) Electrochemical degradation of clofibric acid in water by anodic oxidation: comparative study with platinum and boron-doped diamond electrodes. Electrochim Acta 52(1):75–85

Vogel F, Harf J, Hug A, Von Rohr PR (2000) The mean oxidation number of carbon (MOC)- A useful concept for describing oxidation processes. Water Res 34(10):2689–2702

Weiss E, Groenen-Serrano K, Savall A (2008) A comparison of electrochemical degradation of phenol on boron doped diamond and lead dioxide anodes. J Appl Electrochem 38(3):329–337

Yavuz Y, Koparal AS, Outveren UB (2008) Phenol degradation in a bipolar trickle tower reactor using boron-doped diamond electrode. J Environ Eng 134:24–31