

## Fabrication and application of boron doped diamond BDD electrode in olive mill wastewater treatment in Jordan

Inshad Jum'h, Arwa Abdelhay, Hussein Al-Taani, Ahmad Telfah, Mohammad Alnaief and Stefan Rosiwal

### ABSTRACT

A boron doped diamond (BDD) electrode was employed in an electrochemical reactor to oxidize the phenolic content of Jordanian olive mill wastewater. The BDD anode was fabricated using hot filament chemical vapor deposition on niobium and the morphology of the BDD electrode was characterized using an atomic force microscope. Then, electrolysis batch runs were carried out at laboratory scale to test the effect of different process parameters, namely, initial chemical oxygen demand (COD) load (72.9, 33.8, and 0.18 g/L), the addition of Na<sub>2</sub>SO<sub>4</sub> as supporting electrolyte, and adding NaCl along with Na<sub>2</sub>SO<sub>4</sub>, on the efficiency of the treatment process. The results were reported in terms of COD, color and turbidity removal, and pH variation. The experiments revealed that electrochemical oxidation using BDD significantly reduced the COD by 85% with no supporting electrolytes. It was observed that adding Na<sub>2</sub>SO<sub>4</sub> with NaCl brought the COD removal to higher than 90% after 7 hours of treatment for COD loads of 72.9 and 33.8 g/L, and after 2 hours for a COD load of 0.18 g/L. Likewise, color was completely removed regardless of the initial COD load. The turbidity for samples with 72.9 and 33.8 g/L as COD load reached a minimal value of 2.5 and 1 NTU respectively.

**Key words** | advanced oxidation processes, boron-doped diamond electrode, electrochemical oxidation, olive mill wastewater, phenolic compounds

**Inshad Jum'h**<sup>†</sup>  
**Arwa Abdelhay**<sup>†</sup> (corresponding author)  
**Hussein Al-Taani**  
**Mohammad Alnaief**  
German-Jordanian University,  
Amman 11180,  
Jordan  
E-mail: arwa.abdelhay@gu.edu.jo

**Ahmad Telfah**  
Leibniz-Institut für Analytische, Wissenschaften –  
ISAS,  
Bunsen-Kirchhoff-Str.11,  
44139 Dortmund,  
Germany

**Stefan Rosiwal**  
Institute of Metals Science and Technology (WTM),  
Friedrich-Alexander University,  
5,91058 Erlangen,  
Germany

<sup>†</sup>Equal contributors

### INTRODUCTION

During the last century after the industrial revolution, a large amount of industrial wastewater was discharged into rivers, lakes, soils and coastal areas. Industrial water effluents include water for purposes such as processing, washing, extraction, and cooling in facilities that manufacture products (Hanchang 2009). Among these effluents was olive mill wastewater (OMW), which is a liquid by-product generated during olive fruit pressing and processing to extract olive oil. OMW consists of water (80–83%), organic compounds (15–18%), and 2% of inorganic compounds (Chatzisyneon *et al.* 2009). It is also

characterized by very high chemical oxygen demand (COD) (40–200 g/L), biochemical oxygen demand (BOD) (12–60 g/L), total solid contents (40–150 g/L) and a pH of about 5 (Umran *et al.* 2008). The toxicity of the OMW has been attributed to the high degree of phenolic compounds that are biorecalcitrant and accumulate in water bodies. Hence, large amounts of untreated OMW effluent lead to intense phytotoxic phenomena in flora, and quality degradation of ground water and surface aquatic reservoirs.

Jordan is one of the Middle Eastern countries that suffer from contamination by OMW effluents, particularly in the season of olive oil production. In Jordan, the conventional disposal of OMW can affect the limited water resources, livestock resources, and agricultural resources. To tackle this environmental problem, many researchers have proposed various treatment technologies including chemical,

This is an Open Access article distributed under the terms of the Creative Commons Attribution Licence (CC BY-NC-SA 4.0), which permits copying, adaptation and redistribution for non-commercial purposes, provided the contribution is distributed under the same licence as the original, and the original work is properly cited (<http://creativecommons.org/licenses/by-nc-sa/4.0/>).

doi: 10.2166/wrd.2016.062

biological and electrical treatment methods (Lochart 1983; Ju *et al.* 1991; Boari *et al.* 1993; Hayek *et al.* 1996). However, these methods have limitations in their operation. In particular, biological processes need long hydraulic detention times and large reactor volumes (Siringi *et al.* 2013). In this perspective, advanced oxidation processes for OMW treatment have recently attracted great attention due to their potential to destroy toxic phenolic pollutants. More specifically, the efficiency of electrochemical oxidation of phenol in OMW over non-traditional anodes such as boron doped diamond (BDD) electrodes has been proposed and reported in the literature by several researchers (Canizares *et al.* 2006; Chatzisyneon *et al.* 2009; Elaoud *et al.* 2011; Bouhssine *et al.* 2013). Bouhssine *et al.* 2013 obtained a reduction in the COD and of the phenolic compounds of approximately 60 and 63%, respectively, after 30 min of electrooxidation over a BDD anode. Chatzisyneon *et al.* 2009 tested the treatability of undiluted OMW effluent (40,000 mg/L COD) at 20 A for 15 h, yielding 19% COD and 36% phenols' removal respectively. BDD is considered to be an ideal anode for industrial wastewater treatment since it is characterized by high reactivity towards organics oxidation and efficient use of electrical energy (Canizares *et al.* 2006). Moreover, the BDD thin films have outstanding properties, which are significantly different from those of other traditional electrodes such as glassy carbon or platinum electrodes, and therefore they are attractive for electrolysis applications. The BDD electrodes have a large potential window in aqueous solution with low background currents (Martin *et al.* 1996). In addition, the electrodes formed from BDD possess physical properties including hardness, low environmental impact, high hole mobility, and high thermal conductivity (Alfaro *et al.* 2006). Moreover, the surface of BDD electrodes possesses over-potential to oxygen emission (Bouhssine *et al.* 2013) and is stable with little evidence of degradation of electrochemical activity with time (Kraft 2007).

Despite the extensive scientific research on OMW treatment by electrochemical techniques, there are no cited papers to our knowledge on the electrolysis of OMW over BDD with high initial COD load and effluent quality that meet the legal discharge requirements.

This work describes the lab preparation of BDD on niobium substrate and its applications in the electrochemical treatment of OMW in Jordan. The performance of the

process as a function of the operating parameters such as initial COD load, adding supporting electrolyte, and adding chloride ions along with the supporting electrolyte was evaluated. The OMW electrolysis results were reported in terms of COD, turbidity and color removal.

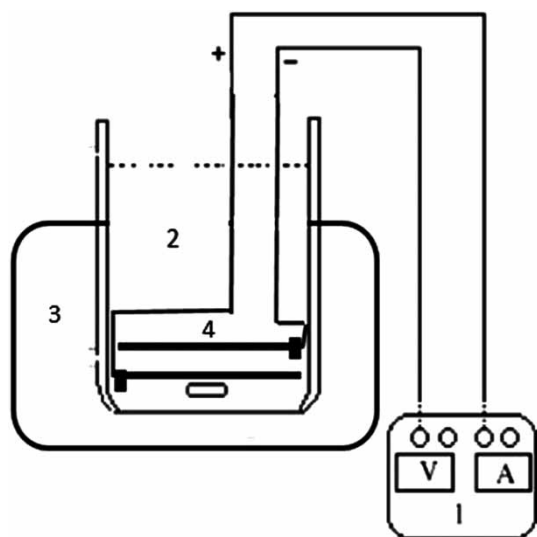
## MATERIALS AND METHODS

### Experimental setup

The BDD electrode consists of two expanded identical and parallel pure niobium circular grids (facilitating the gas bubble flow) separated by 2 mm, having a thickness of 2.8 mm and a diameter of 90 mm (total area of 65 cm<sup>2</sup> per electrode). The electrodes were prepared by hot filament chemical vapor deposition on niobium substrates, which were pretreated by sand blasting to roughen the surface in order to enhance the adhesion strength between the diamond and the substrate as well as to increase the electrochemical performance due to the increase in surface area. The niobium substrates were cleaned twice in ethanol and seeded with diamond powder using an ultrasonic device. The coating process took place in a CVD hot filament coating machine by two parallel filament rows, and the film thickness was measured (8 μm). In order to enhance the doped diamond conductivity, a mixture of CH<sub>4</sub> (1%) and a small amount of B(OCH<sub>3</sub>)<sub>3</sub> were used in the coating process. All the chemical reactions were carried out under galvanostatic conditions, and the characteristic parameters of the diamond films were performed using an atomic force microscope (AFM).

A laboratory-scale electrochemical reactor (Figure 1) was made of Plexiglas with a 1,000 cm<sup>3</sup> capacity to treat OMW. A standard DC power supply was used as the source of electric current for the experiments, and the voltage fluctuations during the reaction were automatically monitored using a digital multimeter. The working electric current was fixed at a constant 3 A. The temperature of the electrolysis reactor was kept constant at room temperature by using an agitated water bath.

Samples were drawn periodically in sealed and clean 20 mL glass vials and analyzed immediately afterwards. The treatment process was tested in terms of COD, color, turbidity removal, and pH variation through the electrolysis time.



**Figure 1** | Schematic diagram of experimental setup: (1) DC power supply; (2) electrochemical cell; (3) water bath; (4) BDD electrodes.

## OMW samples

Three different raw OMW samples (A, B, and C) were collected in sealable metal containers from the northern part of Jordan (Irbid) during the olive fruit harvesting season of the year 2014 and stored at 4 °C. Sample A is the milling wastewater, Sample B is the mixture of olive washing and milling wastewater, and sample C is the washing wastewater. Due to the high content of total suspended solids, the three samples were filtered using a medium crystalline filter paper and then they were centrifuged for 60 minutes at 6,000 rpm. The measurements were repeated to improve averaging. The main physiochemical properties of the

three samples prior to and after filtration processes are given in Table 1.

The tested samples were prepared in 1 L volume, and divided into three groups as follows: the first group was the original samples A, B, and C; the second group were A, B, and C samples mixed with Na<sub>2</sub>SO<sub>4</sub> as a supporting electrolyte; and the third group were A, B, and C with Na<sub>2</sub>SO<sub>4</sub>/NaCl electrolyte.

## Chemicals and analytical methods

Sodium sulfate Na<sub>2</sub>SO<sub>4</sub> (Sigma Aldrich) was used as a supporting electrolyte with a concentration of 0.71% w/v and NaCl (0.06% w/v) (Sigma Aldrich) as a second supporting electrolyte mixed with OMW.

The COD, conductivity, turbidity, pH and absorbance of the samples were investigated and recorded for analysis and comparison.

The pH and conductivity of the samples were measured using a digital calibrated Jenway 3,540 pH/conductivity meter.

The turbidity of the samples was measured with a Wagttech turbidity meter.

The COD values of the initial and treated samples were determined using the dichromate standard method (APHA 1998). Each sample was introduced into commercially available digestion solution (0–1,500 mg/L) containing potassium dichromate (Hach-Germany); afterward, the solution was incubated at 150 °C for 120 minutes in a COD Hach digester (DRB 200-Germany). The COD readings of the electrolyzed samples were measured using a Hach spectrophotometer (DR/2010-Germany).

**Table 1** | Physiochemical properties of OMW samples A, B and C before and after filtration processes

Properties	A		B		C	
	Before	After	Before	After	Before	After
pH	4.84	5.00	4.48	4.33	6.91	7.07
Conductivity (mS)	8.75	11.37	5.82	7.41	1709	1985
Turbidity (NTU)	9,354.2	283.87	2,049.2	85.5	54.9	9.79
Soluble chemical oxygen demand SCOD (g/L)	72.9		33.8		0.18	
Total solids TS (mg/L)	59,815	59,800	27,130	24,875	2,360	2,295
Total volatile solids TVS (mg/L)	37,020	35,350	17,295	15,600	1,220	1,090

## Absorbance

The dark color density of OMW samples is an indicator of the polymeric phenolic compounds' concentration, which is responsible for their dark colors. The discoloration during the treatment process could be measured in terms of absorbance, which was measured during electrolysis time at 450 nm with a Wagtech 7100 photometer.

## RESULTS AND DISCUSSION

### Properties of BDD electrode

The BDD surface was investigated using the AFM technique and characterized by root mean square roughness and average grain size. Figure 2 shows an AFM topography image of the BDD surface. A profile of the AFM image taken along the line in Figure 2(a) is displayed in Figure 2(b). The visible roughness of a few micrometers is a result of surface roughening pretreatment of the niobium substrate. The diamond on the layer surface shows well (111)-faceted crystals with an average grain size of  $0.7 \pm 0.3 \mu\text{m}$ . Importantly, no cracks or holes appear and the peeling phenomenon does not occur, indicating a high quality coating process, which means that the electrode area is totally used for

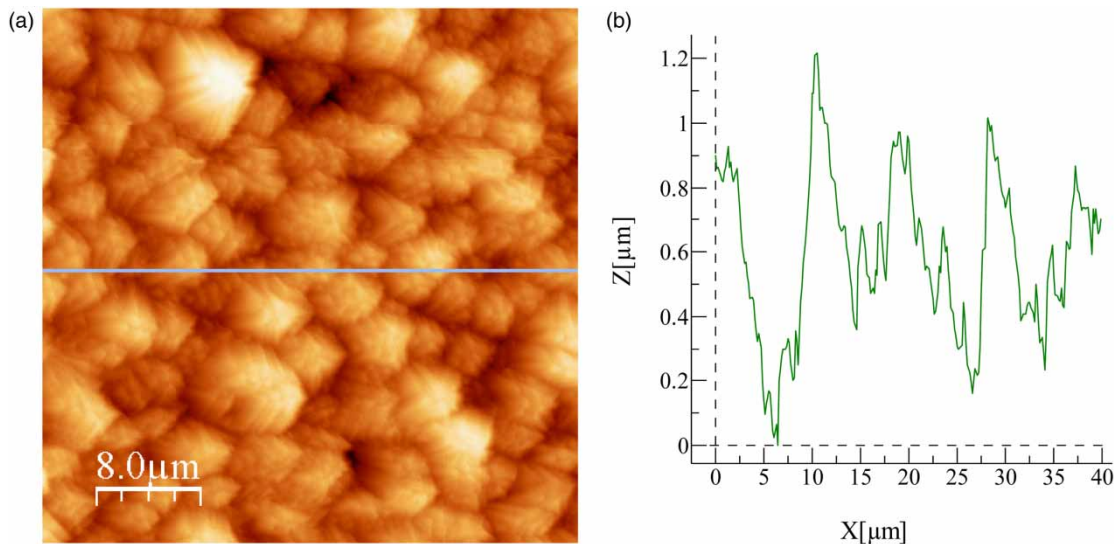
electrochemical oxidation and this consequently increased the electrodes' efficiency.

### OMW electrolysis with no added supporting electrolyte

#### Coloration evolution and absorbance variation during electrolysis

The efficiency of OMW treatment by electrochemical oxidation with a BDD anode was first tested by following up the disappearance of the blackish brown color of the three samples A, B, and C. Results showed that during electrolysis, the dark color of the three samples disappeared gradually and the samples became transparent at the end of the process.

The discoloration of sample A is shown in Figure 3. This observation was confirmed by the absorbance results presented in Figure 4. As illustrated in Figure 4, the absorbance decreased gradually to reach an absorbance reduction of 80, 97, and 98% for samples A, B, and C, respectively. These findings are in accordance with the results of Bouhssine *et al.* (2013). The initial concentration of organic pollutants is a key factor that affects, to a great degree, the efficiency of OMW electrolysis. Obviously, the maximum absorbance reduction was obtained with the least polluted sample, which is sample C.



**Figure 2** | (a) AFM topographies of the BDD layer on niobium substrate. (b) Profile taken along the line in Figure 1(a).



**Figure 3** | Discoloration of sample A during electrolysis with BDD anode.

On the other side, the minimum absorbance removal was observed with sample A. This could be attributed to the fact that sample A is heavily polluted, which means implicitly that it contains a higher concentration of phenolic compounds.

### COD reduction during electrolysis

In order to verify if the absorbance reduction of the three samples is concomitant with COD removal, the COD reduction was measured against time, as depicted in Figure 5. It is obvious that in all cases the COD removal increased with time to attain a maximum removal of 87, 92, and 89% for samples A, B, and C, respectively. This COD reduction during electrolysis using no supporting electrolyte could be attributed to the generation of hydroxyl

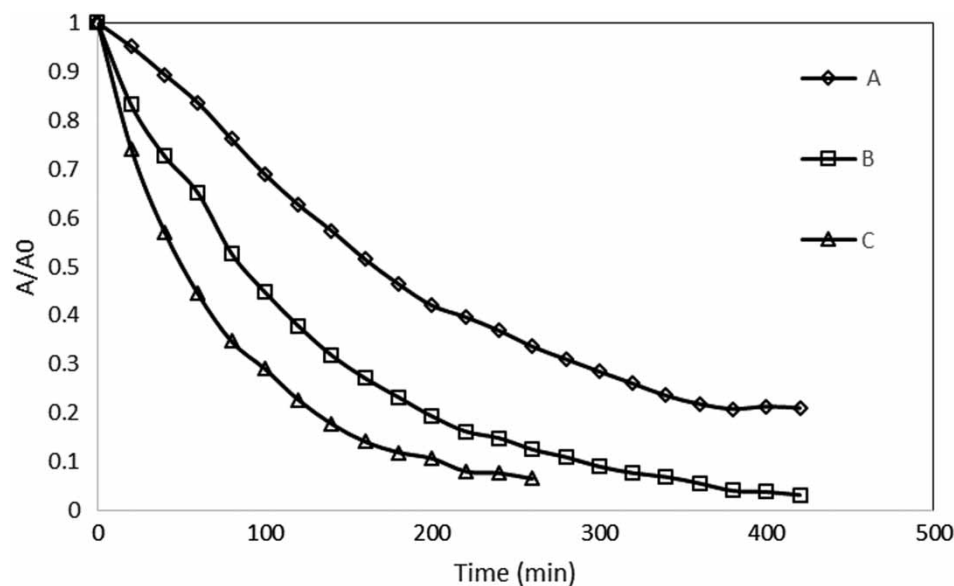
radicals (Elaoud *et al.* 2011) that reacts with polyphenols on the BDD electrode (Martínez-Huitle & Ferro 2006; Panizza *et al.* 2008).

### Turbidity variation during electrolysis

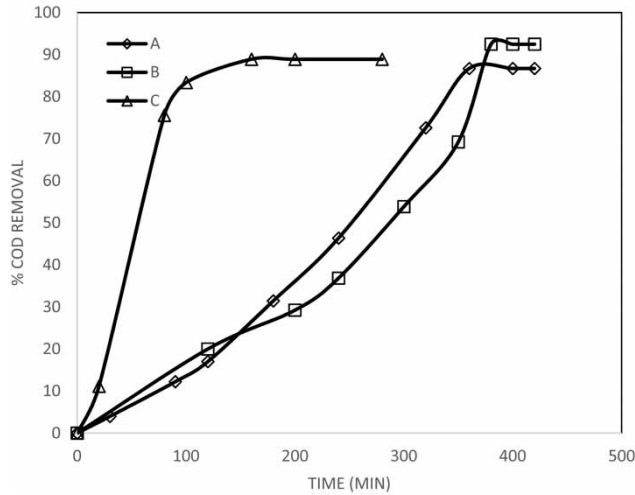
Turbidity was also monitored during the process time. For both samples A and B, the turbidity had a tendency to decrease with time as shown in Figure 6 and it reached values of 10 and 3.6 NTU, respectively. This trend could be justified by the oxidative polymerization of the phenols and tannins originally present in OMW (Assas *et al.* 2002). However, for sample C, the turbidity fluctuated around an average value of 25 NTU. This instability in turbidity readings could be justified by the formation of small oily micelles in sample C, as it has the lowest oil content (Yang 2007).

### pH variation during electrolysis

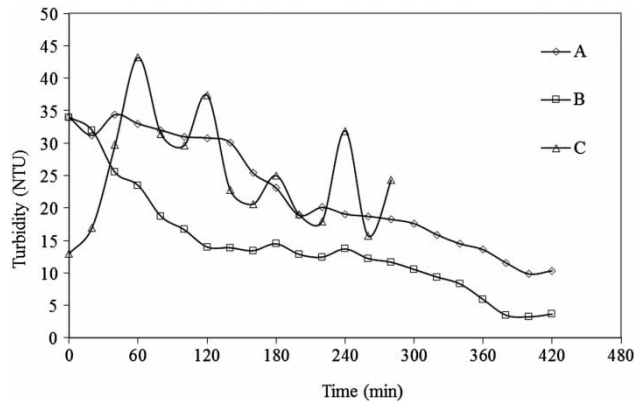
Figure 7 shows the pH variation during electrolysis. The pH increase observed during OMW electrolysis is a proof of the formation of  $\text{OH}^-$ . Unbalanced  $\text{OH}^-$  and  $\text{H}^+$  formation takes place due to the hydrogen evolution reaction and oxygen evolution reaction occurring at the



**Figure 4** | The reduction of absorbance during oxidation on BDD anode with no supporting electrolyte (current: 3 A).



**Figure 5** | COD removal during electrolysis with no supporting electrolyte (current: 3 A).



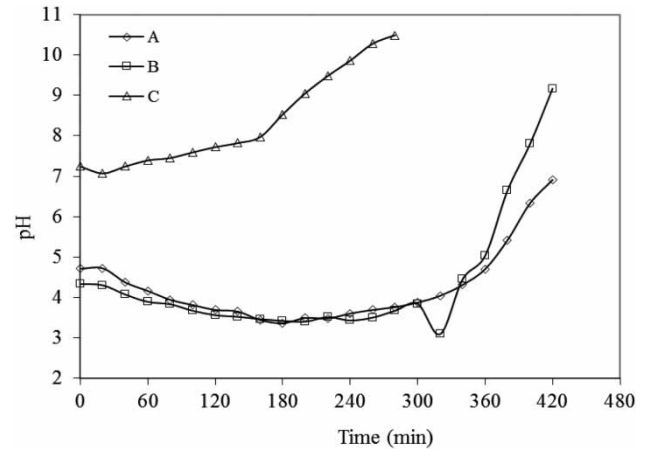
**Figure 6** | Turbidity variation during the oxidation process on BDD anode with no supporting electrolyte (current: 3 A).

cathode and anode, respectively (Alencar de Souza & Ruotolo 2013).

### OMW electrolysis using $\text{Na}_2\text{SO}_4$ and $\text{NaCl}$ as supporting electrolytes

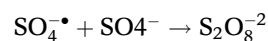
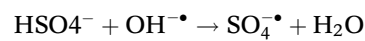
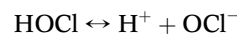
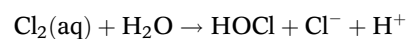
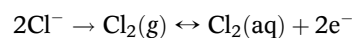
#### Absorbance variation during electrolysis

Figure 8(a)–8(c) depict the effect of  $\text{Na}_2\text{SO}_4$ , when used as supporting electrolyte alone and with  $\text{NaCl}$ , on absorbance variation during electrolysis. It can be seen that for the three samples A, B, and C the absorbance abatement was enhanced by adding  $\text{Na}_2\text{SO}_4$  and even more when adding  $\text{Na}_2\text{SO}_4$  and  $\text{NaCl}$  simultaneously. It was very likely that the absorbance



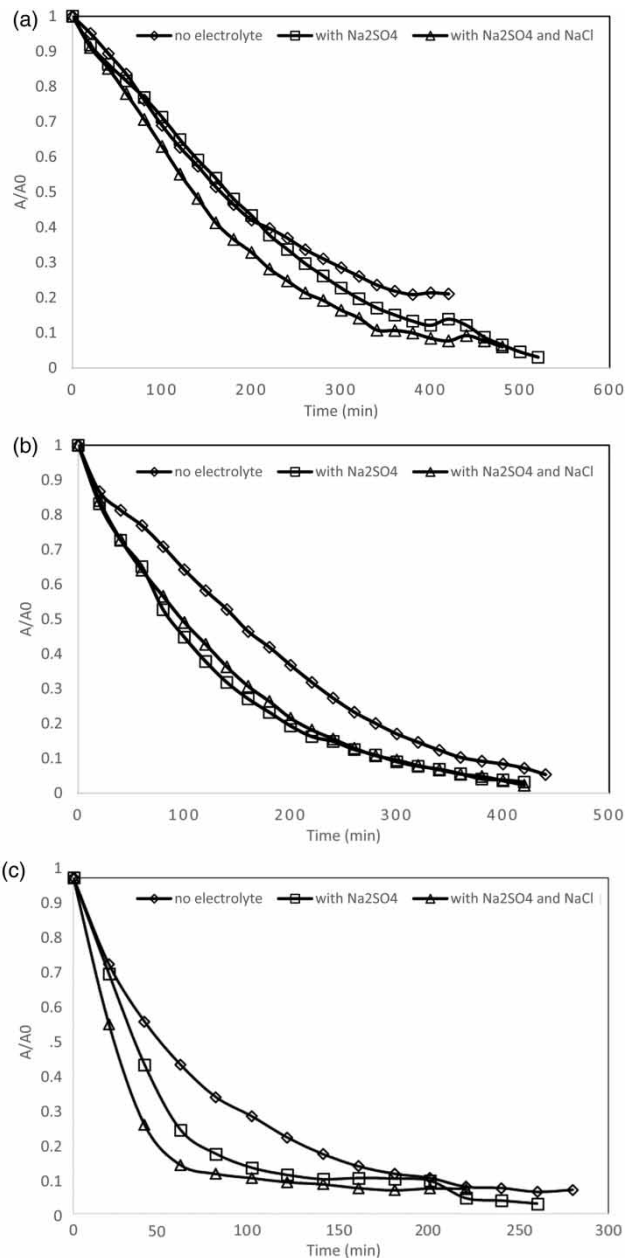
**Figure 7** | pH variation during OMWW electrolysis on BDD anode with no supporting electrolyte (current: 3 A).

reduction was boosted by using  $\text{Na}_2\text{SO}_4$  electrolyte due to the formation of oxidizing species like  $\text{SO}_4^-/\text{S}_2\text{O}_8^{2-}$  (Saracco *et al.* 2000). A further decrease in absorbance was achieved by adding both  $\text{Na}_2\text{SO}_4$  and  $\text{NaCl}$ , which could be an indication of the generation of chloro-oxidant species such as  $\text{ClO}^-$  (Cheng & Kelsall 2007; Alencar de Souza & Ruotolo 2013). In other words, two secondary oxidants ( $\text{S}_2\text{O}_8^{2-}$  and  $\text{ClO}^-$ ) are most probably electro-generated on the BDD surface upon the addition of  $\text{Na}_2\text{SO}_4$  and  $\text{NaCl}$ , according to the following chemical reactions:



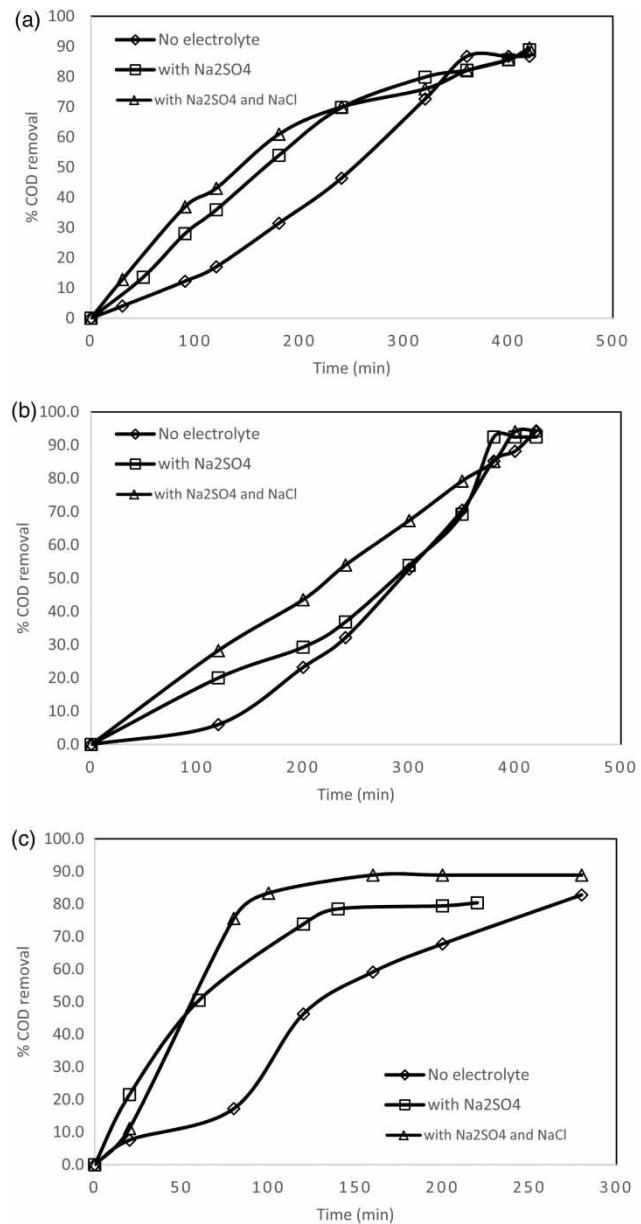
#### COD reduction during electrolysis

The effect of adding supporting electrolyte on the COD removal % was examined for the three types of OMW A, B, and C. Figure 9(a)–9(c) show the COD removal %



**Figure 8** | Effect of  $\text{Na}_2\text{SO}_4$  (0.71% w/v) and  $\text{NaCl}$  (0.06% w/v) electrolytes on absorbance reduction during electrolysis (current 3 A): (a) samples A, (b) samples B, and (c) samples C.

during treatment time for three cases: no added electrolyte, with  $\text{Na}_2\text{SO}_4$ , and with  $\text{Na}_2\text{SO}_4$  and  $\text{NaCl}$ . It can be noticed that with the three types of OMW, whether electrolyte has been added or not a COD removal % of about 90% was achieved. This percentage is much higher than the removal percentage reported by *Giannis et al. (2007)* and



**Figure 9** | Effect of  $\text{Na}_2\text{SO}_4$  (0.71% w/v) and  $\text{NaCl}$  (0.06% w/v) electrolytes on COD removal % during electrolysis (current 3 A): (a) samples A, (b) samples B, and (c) samples C.

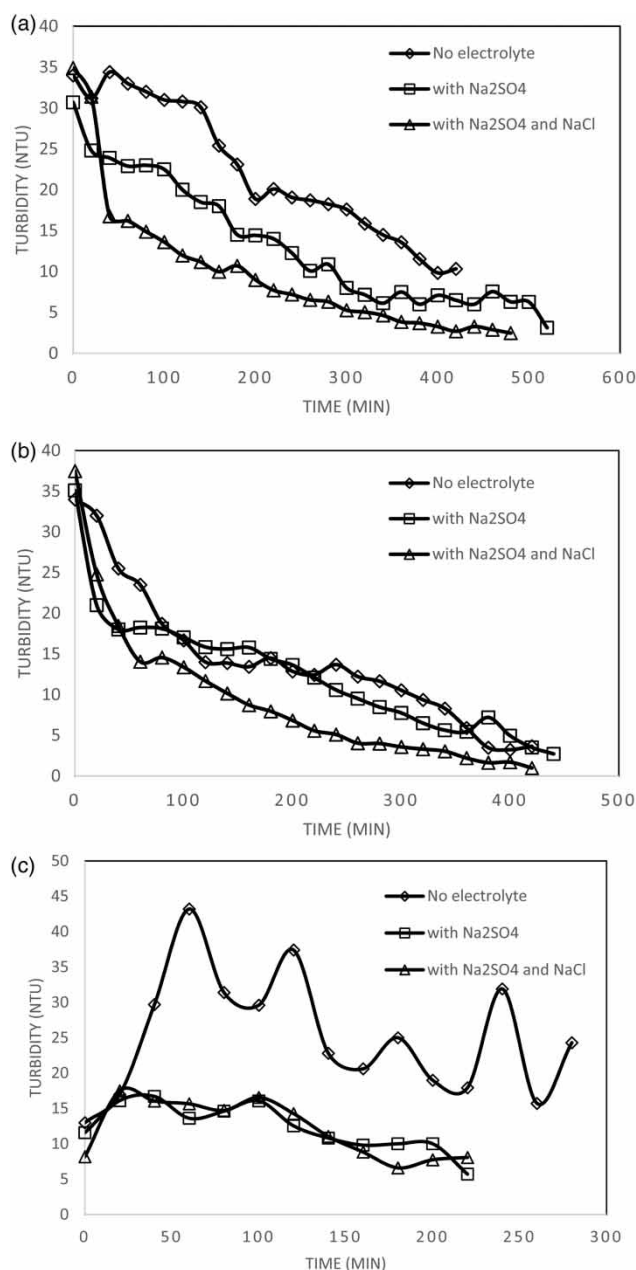
*Chatzisyneon et al. (2009)*. However, for all samples (A, B, and C) the kinetics of the COD removal was apparently enhanced by the addition of  $\text{Na}_2\text{SO}_4$  and better enhancement was achieved by the concomitant addition of  $\text{Na}_2\text{SO}_4$  and  $\text{NaCl}$ . Hence, the most effective electrolyte for COD removal is the  $\text{Na}_2\text{SO}_4/\text{NaCl}$ , which might be a result of the electro-generation of new oxidant species

along with hydroxyl radicals such as peroxodisulfate and chloro oxidant species (Bouhssine *et al.* 2013). The final COD values for samples A, B and C supplemented with  $\text{Na}_2\text{SO}_4/\text{NaCl}$  electrolytes were 70, 76, 21 mg/L, respectively. The COD removal % – time plots are in good

accordance with the absorbance results found in the previous section, and they follow the same trend.

### Turbidity variation during electrolysis

The impact of the electrolyte type on the turbidity of the three samples (A, B, and C) is shown in Figure 10(a)–10(c). It is observed that for all samples the turbidity decreased with electrolysis time. However, the lowest values throughout the process time were obtained when adding together  $\text{Na}_2\text{SO}_4$  and NaCl as supporting electrolytes. The turbidity of sample A dropped from 10 to 3 NTU, sample B from 2.7 to 0.98 NTU, and sample C from 15 to 8 NTU when using no electrolyte and with  $\text{Na}_2\text{SO}_4/\text{NaCl}$  electrolyte, respectively. This result could be justified by the increase in oxidative polymerization of phenols due to the electro-generation of new oxidant species (Elaoud *et al.* 2011).



**Figure 10** | Effect of  $\text{Na}_2\text{SO}_4$  (0.71% w/v) and NaCl (0.06% w/v) electrolytes on turbidity removal during electrolysis (current: 3 A): (a) samples A, (b) samples B, and (c) samples C.

### CONCLUSIONS

The electrochemical treatment of OMW has been investigated using BDD anodes. This method was an efficient and promising technology for OMW treatment even with no supporting electrolyte. Further enhancement in the electrochemical treatment was gained once a combination of two electrolytes,  $\text{Na}_2\text{SO}_4$  and NaCl, was added. This combination of electrolytes was efficient to reach a COD removal higher than 90% after 7 hours of treatment time for samples A and B and 2 hours for sample C. In addition, the dark color of the OMW was completely removed and the three samples turned transparent at the end of the electrolysis. The turbidity of the three samples was also strongly affected by the addition of  $\text{Na}_2\text{SO}_4$  and NaCl electrolytes. The turbidity of samples A, B, and C reached minimal values (2.5, 1, and 8 NTU) when both  $\text{Na}_2\text{SO}_4$  and NaCl electrolytes were added.

### REFERENCES

Alencar de Souza, R. B. & Ruotolo, L. A. 2013 Phenol electrooxidation in different supporting electrolytes using



- boron-doped diamond anodes. *Int. J. Electrochem. Sci.* **8**, 643–657.
- Alfaro, M. A., Ferro, S., Martínez-Huitle, C. A. & Vong, Y. M. 2006 [Boron doped diamond electrode for the wastewater treatment](#). *J. Braz. Chem. Soc.* **17**, 227–236.
- APHA 1998 *Standard Methods for the Examination of Water and Wastewater*, 20th edn. American Public Health Association, Washington, DC, USA.
- Assas, N., Ayed, L., Marouani, L. & Hamdi, M. 2002 [Decolorization of fresh and stored-black olive mill wastewaters by \*Geotrichum candidum\*](#). *Process Biochem.* **38**, 261–265.
- Boari, G., Mancini, I. & Trulli, E. 1993 Anaerobic digestion of olive oil mill effluent pretreated and stored in municipal solid waste sanitary landfills. *Water Sci. Technol.* **28**, 27–34.
- Bouhssine, I., Tazi, A. & Azzi, M. 2013 Treatment of olive mill wastewater by electrolysis on boron doped diamond (BDD) electrode. *J. Mater. Environ. Sci.* **4**, 354–361.
- Canizares, P., Martinez, L., Paz, R., Saez, C., Lobato, J. & Rodrigo, M. A. 2006 [Treatment of Fenton-refractory olive oil mill wastes by electrochemical oxidation with boron-doped diamond anodes](#). *J. Chem. Technol. Biotechnol.* **81**, 1331–1337.
- Chatzisyneon, E., Xekoukoulotakis, N. P., Diamadopoulos, E., Katsaounis, A. & Mantzavinos, D. 2009 [Boron-doped diamond anodic treatment of olive mill wastewaters](#). *Water Res.* **43**, 3999–4009.
- Cheng, C. Y. & Kelsall, G. H. 2007 [Models of hypochlorite production in electrochemical reactor with plate and porous anodes](#). *J. Appl. Electrochem.* **37**, 1203–1217.
- Elaoud, S., Panizza, M., Cerisola, G. & Mhiri, T. 2011 [Veratic acid treatment by anodic oxidation with BDD anode](#). *J. Chem. Technol. Biotechnol.* **87**, 381–386.
- Giannis, A., Kalaitzakis, M. & Diamadopoulos, E. 2007 [Electrochemical treatment of olive mill wastewater](#). *J. Chem. Technol. Biotechnol.* **82**, 663–671.
- Hanchang, S. 2009 *Point Sources of Pollution: Local Effects and Control*, Vol. 1, Industrial Wastewater-Types, Amounts and Effects. Encyclopedia of Life Support Systems (EOLSS), Beijing, China.
- Hayek, B., Mosa, M. & Halasah, N. 1996 An experimental method for treatment of olive mills wastewater utilizing up flow anaerobic sludge blanket (UASB) reactor. Presented at Jordan Chemical Engineering Conference II, Jordan, 2–4 September. Jordan Engineers Association, Jordan, pp. 64–81.
- Ju, S., Weber, M. E. & Mujumdar, A. 1991 [Electro-osmotic dewatering of bentonite suspensions](#). *Sep. Technol.* **1**, 214–221.
- Kraft, A. 2007 Doped diamond: a compact review on a new, versatile electrode material. *Int. J. Electrochem. Sci.* **2**, 355–385.
- Lochart, N. C. 1983 [Electro-osmotic dewatering of clays. II. Influence of salt, acid, and flocculants](#). *Colloids Surf.* **6**, 239–251.
- Martin, H. B., Argoitia, A., Landau, U., Anderson, A. B. & Angus, J. C. 1996 [Hydrogen and oxygen evolution on boron-doped diamond electrodes](#). *J. Electrochem. Soc.* **143**, 133–136.
- Martínez-Huitle, C. A. & Ferro, S. 2006 [Electrochemical oxidation of organic pollutants for the wastewater treatment: direct and indirect processes](#). *Chem. Soc. Rev.* **35**, 1324–1340.
- Panizza, M., Brillas, E. & Comninellis, C. 2008 Application of boron-doped diamond electrodes for wastewater treatment. *J. Environ. Eng. Manage.* **18**, 139–153.
- Saracco, G., Solarino, L., Aigotti, R., Specchia, V. & Maja, M. 2000 [Electrochemical oxidation of organic pollutants at low electrolyte concentrations](#). *Electrochim Acta.* **46**, 373–380.
- Siringi, D., Home, P. G., Chacha, J. & Mulimi, L. 2013 Wastewater remediation using electrochemically produced layered double hydroxides. *IJEIT* **2**, 61–64.
- Umrán, U., Umit, A., Savas, A. K. & Ulker, O. 2008 [Complete treatment of olive mill wastewaters by electrooxidation](#). *Chem. Eng. J.* **139**, 445–452.
- Yang, C. 2007 [Electrochemical coagulation for oily water demulsification](#). *Sep. Purif. Technol.* **54**, 388–395.

First received 28 March 2016; accepted in revised form 15 July 2016. Available online 26 August 2016