Is it worth the use of bipolar electrodes in electrolytic wastewater treatment processes?

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Abstract

In this work, it is studied the advisability of the application of bipolar electrodes (consisting of Boron Doped Diamond coatings) for the electrolytic treatment of wastewater. Results are discussed in terms of the success in two different reactions: the depletion of an organic pesticide (2,4-dichlorophenoxyacetic acid) and the oxidation of chlorides to perchlorates. This later reaction should be prevented in an actual environmental remediation process because of the hazardousness of perchlorate, but it has been investigated in this work because it gives valuable information about formation of oxidants during electrolytic processes as chlorates and perchlorates behaves as inert species in the oxidation of 2,4-D. Results pointed out the importance of the optimization of the number of bipolar electrodes to be included in each cell for a given application. Bipolar configuration can help to improve the results obtained in the removal of organic compounds by direct processes and it also influences on the formation oxidizing species that can improve the results of this electrolytic process. However, it increases significantly the cell voltage and hence the power consumption. In this work, it is demonstrate that each bipolar electrode saves around 22% of the electric charge provided

by the single cell to deplete 2,4-D and to generate oxidants, although this positive effect is masked by the increase in the cell voltage. Energy required to oxidize organic increases with the number of bipolar electrodes installed in the cell while energy efficiency in the production of oxidants shows a maximum for the cell configuration with 1 bipolar electrode.

Keywords

Bipolar electrode, wastewater, electrolysis, perchlorate, pesticide

Highlights

- Application of bipolar electrodes can help to optimize electrolytic treatment processes
- Current efficiency and energy efficiency depend on the concentration of compound.
- Bipolar configurations show advantageous when the mediated oxidation becomes relevant.
- Electrolysis of wastewater carried out in the lab scale should be easily scaled-up

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1. Introduction

During the last decades, electrochemical oxidation of wastewaters has been a hot topic in environmental electrochemistry thanks to the development of novel anode materials. Among them, boron doped diamond (BDD) has become the most promising electrode for this application. These electrodes have shown very high efficiencies in the use of electricity and high chemical and electrochemical stability [1, 2]. The hydroxyl radicals, and many other oxidants produced at high concentrations on the surface of diamond, are responsible for the harsh oxidation conditions attained and help to explain the higher efficiencies obtained in the degradation of organics in comparison with other well-known Advanced Oxidation Processes [3, 4]. Their main drawback seems to be their bad mechanical properties but lack of scale-up studies makes difficult to determine how they could perform in an industrial process. Most papers related to the electrochemical oxidation of wastewater polluted with organics with BDD are carried out at a lab or bench scale, and they are mainly focused on the reaction of oxidation, particularly on:

- the study of the chemical species involved in the reaction of removal of a pollutant, trying to elucidate reaction mechanisms and occurrence of hazardous intermediates or final products in the reaction medium, and
- the characterization of the effect on efficiency of the treatment of single operating parameters (such as temperature and current density) or waste composition parameters (pH, electrolyte composition) in very simple electrochemical cells, typically consisting of single mixed tanks or one-compartment flow pass cell.

For this reason, they typically show the changes during the progress of the treatment of the concentration of pollutants, intermediates and final products and, sometimes, they propose a very preliminary calculation of efficiency. Little attention is paid to the use of commercial cells and even less to the effect of the cell design on the results of a treatment.

As a consequence, most of these studies should be classified as lab-scale regardless of the size of the cell used. It is important to take in mind that scale-up should not be understood as a single increase in the volume or flow rate of wastewater treated, but as the study of other aspects of the processes such as flow-conditions, electrode arrangements, auxiliary services, construction materials, etc.

Hence, it is clear that many efforts have to be done during the next years in the scale-up of wastewater electrolytic treatment processes, in order to determine if the technology could be applied at the full scale. Next step should be to improve system performance by a proper cell design, mainly using commercial cells. This stage is carried out at the bench (and sometimes pilot plant) scale and up to now very few works have been focused on this topic [5-7].

At this point, the use of bipolar configuration is a matter of the major significance in the scale up of electrochemical processes and hence on those processes based on the electrochemical removal of pollutants. The objective of the present investigation is the study of the bipolar cell configuration for the treatment of wastewaters using two reactions that are going to develop simultaneously in an electrolytic cell with diamond electrodes: the depletion of an organic pesticide and the oxidation of chlorides to perchlorates. The aim is not to look for the optimization of the environmental remediation process (obviously in such an application production of perchlorate should be prevented) but simply to increase knowledge about scale up of this type of processes taking advantage of the simultaneous occurrence of both reactions during the electrolysis of a synthetic wastewater.

2. Materials and methods

2.1 Experimental setup

All electrolyses were conducted in a DiaCell® type 401 supplied by Adamant Technologies (Switzerland) (Figure 1). The DiaCell® module can be assembled with one, two, three and four compartments, each of them being fed with wastewater by internal parallel inlets. The boundary electrodes are monopolar Si/BDD and in case of using multiple compartments, the separating electrodes are floating bipolar Si/BDD electrodes. All electrodes are circular (100 mm diameter) and the inter electrode gap is 1 mm. Regarding the BDD electrodes, resistivity of the Si wafers was 100 mΩ cm and the thickness of the BDD coatings was 2-3 μm. Boron concentration in the coatings was 500 ppm and the ratio sp^3/sp^2 around 150. Prior to electrolysis assays the BDD electrodes were cleaned during 10 min in a 1 M Na₂SO₄ solution at 15 mA cm⁻².

2.2 Analysis procedures and methods

The electrolyses were carried out galvanostatically (external current applied was 10 A) using 1.2 dm³ of a solution containing 100 mg dm⁻³ 2,4-dichlorophenoxyacetic acid (2,4-D) and 3 g dm³ NaCl at natural pH (3.5). The electrolyte was stored in a glass tank and circulated through the electrolytic cell by means of a centrifugal pump. The electrolyte flow rate through the cell was 26.4 dm³ h⁻¹. A heat exchanger coupled with a controlled thermostatic bath was used to maintain the temperature at 25 °C. All electrolyses were performed in the same operating conditions in order to evaluate the efficiency of the different electrode arrangements.

All the samples extracted from electrolyzed solutions were filtered with 0.45 μm nylon filters from Whatman before their analysis. The decay of 2,4-D and the evolution of organic intermediates were followed by reversed-phase chromatography, total organic

carbon (TOC) and Chemical Oxygen Demand (COD). The chromatography system was an Agilent 1100 series coupled a UV detector. The analytical column Phenomenex Gemini 5 μm C18 was used. The analytics conditions were a mobile phase consisting of 60 % acetonitrile and 40 % water to 2 % acetic acid with a flowrate of $0.4 \text{ cm}^3 \text{ min}^{-1}$, UV detection wavelength of 280 nm, temperature of the oven at 25 °C, and volume injection of 20 μL . The Total Organic Carbon (TOC) concentration was monitored using a Multi N/C 3100 Analytik Jena analyzer. Measurements of pH were carried out with an InoLab WTW pH-meter. The Chemical Oxygen Demand (COD) was monitored using a HACH DR200 analyzer.

Chlorine species (Cl^- , ClO_3^- and ClO_4^-) were determined by ion chromatography using a Metrohm 7330 IC Separation Center coupled with a 752 pump unit and a 732 IC detector. The column used was a Metrosep A Supp 4. The mobile phase consisted of 1.8 mM Na_2CO_3 and 1.7 mM NaHCO_3 with a flow rate of 1.0 ml min^{-1} .

The current efficiency (CE) was calculated with Equation 1 where COD_t and $\text{COD}_{t+\Delta t}$ are the chemical oxygen demand (in $\text{g O}_2 \text{ dm}^{-3}$) at times t and $t+\Delta t$ (in seconds), respectively, I is the current intensity (A), F is the Faraday constant (96487 C mol^{-1}), V is the volume of the electrolyte (dm^3) and 8 is a dimensional factor for unit consistence ($\frac{32 \text{ g O}_2 \cdot \text{mol}^{-1} \text{ O}_2}{4 \text{ mol e}^- \cdot \text{mol}^{-1} \text{ O}_2}$).

$$CE = \frac{[(COD)_t - (COD)_{t+\Delta t}] F V}{8 I \Delta t} \quad (1)$$

The specific energy consumption (w , kWh m^{-3}) was calculated by Equation 2, where U is the cell potential (V):

$$w = \frac{UIt}{V} \quad (2)$$

3. Results and discussion

3.1 Electrolysis of 2,4-D solutions with bipolar electrodes

Figure 2a shows the changes in the cell potential during the galvanostatic electrolysis of the 2,4-D synthetic wastewater with the different electrode configurations studied in this work for the Diacell. As it can be observed, cell voltage is kept almost constant during the four electrolyses, regardless of the electrode configuration tested. In fact, just a small decrease is observed at the beginning of the electrolysis, which can be related to the slight increase in the temperature caused by the ohmic losses or with the effect on conductivity of the production of protons and hydroxyl ions caused by water oxidation and reduction. On the other hand, as it can be seen in Figure 1b, cell voltage increases importantly with the number of bipolar electrodes in a linear fashion with a slope of approx. 5.9 V/bipolar electrode. This increase has a direct impact on the energy consumption of the electrochemical process and it means a 66% increase of the cell voltage of the single cell with every bipolar electrode connected.

Figure 3 focuses on the influence of the number of bipolar electrodes mounted in the cell on the efficiency of the oxidation of the organic pollutant 2,4-D. Particularly, it shows the changes of the raw pollutant, intermediates (globalized as total chromatographic area) and carbon dioxide with the external charge applied to the cell. This parameter is directly related to the electrolysis time because externally applied intensity is kept constant in all the electrolysis tests ($I = 10.0$ A). In Part a, it can be observed that 2,4-D is completely mineralized (transformed into carbon dioxide) and that externally applied electric charge required to attain complete depletion of the organic compounds decreases with the

number of bipolar electrodes operating in the cell. Likewise, Part b shows that oxidation conditions are also harsher because the peak that totalized the chromatographic area of intermediates decreases with the number of electrodes. These observations can be easily explained by taking into account the internal electric charge supplied by the bipolar electrodes, transferring electrons from one face of the electrode to the other, without passing through an external circuit. Taking into account the almost linear trend of the TOC removal vs Q_{ext} , external electric charge required for the total depletion of 2,4-D ($Q_{ext_depletion}$) can be used for comparison purposes. Thus, differences in the external electric charge required for total depletion can be used to estimate in a preliminary way the internal electric charge passed in every bipolar electrode, simply by obtaining the additive effect. This value can be estimated in nearly $5.9 \text{ kAh m}^{-3}/\text{bipolar electrode}$. This value is very interesting because it means that every bipolar electrode provides around 22.5% of the electric charge provided by the single cell to deplete the organic pollutant studied (26.31 kAh m^{-3})

Obviously, there should be established a balance between the positive effect of the current applied and the negative effect of the cell voltage on the progress of the oxidation and efficiency. Thus, when results obtained in the electrolyses are plotted vs the energy applied the clear advantages of connecting bipolar electrodes that seems to be obtained according to results of Figure 4 are diluted and, in fact, it can be observed that energy efficiency decreases in the sequence 2,3,0,1 bipolar electrode in the electrochemical cell. The same compartment was obtained by Abdessamad et al. 2013 when studied the electrochemical degradation of dye solutions using both monopolar and bipolar electrochemical cells.

3.2 Electrolysis of chloride solutions with bipolar electrodes

1

2 Although direct and especially hydroxyl radicals mediated oxidation may have an
3 important role in electrochemical oxidation [8, 9], the most relevant processes in
4 electrochemical oxidation of wastewater sometimes are associated to mediated oxidation
5 by species formed from the oxidation of salts contained in wastewater. Typically these
6 species have a very important role on the oxidation of organics during electrochemical
7 treatment of wastewater and many papers have shown the effect of the supporting
8 electrolyte [10-12] on the efficiency, and others even have focused on the production of
9 oxidants from chloride [3], sulfate [13], phosphate [14] [15] and even carbonates[16]
10 solutions. Respect to the organic contained in a typical wastewater concentration of salts
11 is much higher and this means that in case of having an electrochemical reactivity (like
12 happens with chloride), influence on results should be taken into account.

13 Final product in the oxidation of chlorides with BDD anodes is not hypochlorite but
14 perchlorate [3] and main intermediate is chlorate if current density is high enough. This
15 species is a very strong oxidant from the thermodynamic point of view according to its
16 great reduction potential but kinetically is very slow at room temperature and, hence, it
17 behaves as an inert substance. For this reason, it is a good case of study to check the effect
18 of using bipolar cells, especially taking into account that chlorides are easily oxidized by
19 diamond electrodes either by direct or hydroxyl radicals mediated oxidation. Formation
20 of chlorates and perchlorates explains the lower efficiency in the removal of organics
21 observed when diamond anodes are used in a supporting electrolyte containing chloride
22 [17].

23 Figure 5 shows the changes in the concentration of chloride, chlorate and perchlorate.
24 Almost nil concentrations of hypochlorite were detected and these low concentrations can
25 be explained by the high current densities studied and also because this species can

1 rapidly combine with 2,4-D or with intermediates formed during the electrolysis. Again,
2 the efficiency of the system (in terms of reaction progress for a given external electric
3 charge applied) increases with the number of bipolar electrodes connected. Including
4 more bipolar electrodes leads to: (i) a faster removal of chlorides, (ii) a faster formation
5 of perchlorates and (iii) a much lower concentration of chlorates.

6 To oxidize completely chlorides to perchlorates around 54.05 kAh m^{-3} are required in a
7 single cell with no bipolar electrodes. Every electrode decreases the Q_{ext} required by 12.25
8 kAh m^{-3} / bipolar electrode. This means that, within the range of bipolar electrodes
9 connected to the diacell, approximately 22.65% of the electric charge can be provided by
10 each bipolar electrode installed. This percentage is very similar to the reduction observed
11 in the oxidation of organics but should be balanced with the negative effect on the cost of
12 the cell voltage increase. Again, the balance between the increase in the cell voltage and
13 the improvement in the charge is almost compensated in the case of the oxidation of
14 chlorides to perchlorates as it is clearly observed in Figure 6 in which all point seems to
15 lay over the same line.

16 Hence, bipolar configuration can help to improve the results obtained in the removal of
17 organics compound by direct processes and it also influence on the formation oxidants
18 that can improve the results of this electrolytic process although the increase in the cell
19 voltage can be negative.

20 **3.3 Current efficiency of electrolytic processes with bipolar electrodes**

21 To summarize all these results and get relevant conclusions, Figure 7 shows the effect of
22 the number of bipolar electrodes connected in the cell in the current efficiency (define as
23 the ratio between electrons supplied and used successfully in the oxidation process) and
24 in the energy efficiency (defined as the ratio between the COD removed and the energy

1 applied). As it can be observed, in the case of the oxidation of organics both parameters
2 has an opposite trend clearly explained by the low current efficiencies obtained (lower
3 than 5%) associated to the low concentration of organics.

4 Opposite, when the compound that is oxidized is contained in large concentrations (case
5 of chloride), current efficiency and energy efficiency increase with the use of a bipolar
6 electrode and this last parameter, which is the most relevant from the viewpoint of
7 economy, shows a clear maximum. When the role of these oxidants in the electrolytic
8 process is important, a clear advantage arises with the use of bipolar electrode
9 configurations. This is especially important because a very interesting hot topic nowadays
10 in electrolysis of wastewater is the activation of oxidants produced anodically [18] [19]
11 either by chemical interaction with other oxidants [20], ultraviolet light [21] or ultrasound
12 irradiation [22].

13 This means that cells with bipolar electrodes can become a good choice for the removal
14 of compounds contained at large concentrations but not at low concentration without a
15 significant contribution of mediated oxidation processes. Results also demonstrates that
16 studies carried out in the lab scale should be scaled-up and a first scale up study could be
17 the feasibility of connecting bipolar electrodes in the type of cell that is pretended to be
18 used at the full scale.

20 **4. Conclusions**

21 From this work, the following conclusions can be drawn:

- 22 - Bipolar configuration can help to improve the results obtained in the removal of
23 organics compound by direct processes and it also influence on the formation

oxidized chloro-species that can improve the results of this electrolytic process although the increase in the cell voltage can be negative.

- Every bipolar electrode provides around 22% of the electric charge provided by the single cell to deplete 2,4-D and to generate perchlorate. Thus, the externally applied electric charged required in both processes decreases with the number of bipolar electrodes used.
- The lower electric charge required does not compensate the increase in the cell voltage observed with the number of bipolar electrodes in the case of 2,4-D depletion and hence it is observed a negative effect on energy efficiency of increasing the number of bipolar electrodes in the electrochemical cell.
- Energy efficiency in the production of oxidants shows a maximum for the cell configuration with 1 bipolar electrode. This fact clearly pointed out that concentration of species to be oxidized has a very important effect on the choice of the cell configuration.

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Figure captions

Figure 1: Arrangement of the experimental set up. Detail of the electrochemical cell section.

Figure 2. a) Changes in the cell potential during the electrolyses of a 100 mg dm⁻³ 2,4-D and 3000 mg dm⁻³ of NaCl with and diacell equipped with (◆) No bipolar electrodes; (●) 1 bipolar electrode; (■) 2 bipolar electrodes; (▲) 3 bipolar electrodes. b) Plot of the steady state cell voltage with the number of bipolar electrodes installed.

Figure 3. a) Changes in the concentration of 2,4-D, mineralization and b) intermediates during the electrolysis with an external current intensity of 10 A. (full points: 2,4-D; empty points: carbon dioxide (calculated by TOC depletion); grey points: sum of chromatographic peaks areas of intermediates). (▲) 3 bipolar electrodes; (■) 2 bipolar electrodes; (●) 1 bipolar electrode; (◆) No bipolar electrodes.

Figure 4. Changes in the 2,4-D and mineralization with energy applied for electrolyses at a current intensity of 10 A. (full points: 2,4-D; empty points: carbon dioxide (calculated by TOC depletion). (▲) 3 bipolar electrodes; (■) 2 bipolar electrodes; (●) 1 bipolar electrode; (◆) No bipolar electrodes.

Figure 5. Changes in the chlorine speciation with the external electric charge passed during the electrolysis with an external current intensity of 10 A. (full points: choride; empty points: perchlorates; grey points: chlorates). (▲) 3 bipolar electrodes; (■) 2 bipolar electrodes; (●) 1 bipolar electrode; (◆) No bipolar electrodes.

Figure 6. Changes in the chlorine speciation with energy applied for electrolyses at a current intensity of 10 A. (full points: choride; empty points: perchlorates; grey points: chlorates). 3 bipolar electrodes; (■) 2 bipolar electrodes; (●) 1 bipolar electrode; (◆) No bipolar electrodes.

1 **Figure 7.** Influence of the number of bipolar electrodes on the efficiency (Δ) and energy
2 consumption ($\square$) of a diacell for the removal of organics (a) and the oxidation of chlorides
3 (b).

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