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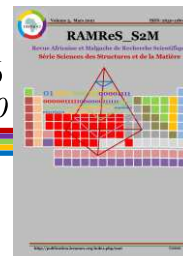
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Oxidation of paracetamol by combination of sonochemistry and electrochemistry on boron-doped diamond electrode

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Abstract:

In this study, sonochemistry (US) was combined with electrochemistry (EO) to improve the degradation of paracetamol on a boron-doped diamond electrode. Voltammetric techniques such as cyclic voltammetry (CV) and differential pulse voltammetry (DPV) were used for the electrochemical analyzes. The study of the electrochemical behavior of the boron-doped diamond electrode, in the presence of the redox couple ferri / potassium ferrocyanide, has made it possible to demonstrate its quasi-metallic character. In addition, this quasi-metallic character of the electrode is improved in the presence of ultrasound. The oxidation of paracetamol was carried out by three methods: sonochemistry, electrochemistry and sonoelectrochemistry (coupling sonochemistry and electrochemistry (US + EO)). The results obtained showed an oxidation rate of paracetamol of 18%; 73% and 89% respectively for sonochemistry, electrochemistry and sonoelectrochemistry after only 1.5 hours. Thus, combining sonochemistry with the preparative electrolysis of paracetamol makes it possible to increase its degradation rate by more than 25%, in the case of this work. The kinetic study was performed according to the pseudo first order model. The apparent rate constants determined are: 0.11 (for US); 0.95 (for EO) and 1.34 h⁻¹ (for US + EO). The combination of sonochemistry and electrochemistry increased the rate of degradation of the organic compound. Sonochemistry and electrochemistry act synergistically in the sonoelectrochemical mineralization of paracetamol, as evidenced by the calculated degree of synergy value S (S = 0.21 > 0).

Keywords: Paracetamol; Sonochemistry; Electrooxidation; Voltammetry; Boron-doped diamond.

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1. Introduction

Paracetamol (acetaminophen or para-acetyl-amino-phenol), reported according to the literature as one of the drugs most frequently consumed in the world, is a metabolite of phenacetin with a strong analgesic and antipyretic effect [1, 2]. Well tolerated pharmaceutical compound that does not have the many side effects of aspirin. It is commonly used for the relief of fever, headache, and pain [3 - 5]. Unfortunately, a considerable increase in the concentration of pharmaceutical contaminants in surface waters has been observed over the past two decades [6, 7]. The presence of these pollutants in the environment creates adverse effects not only on human health, but also affects aquatic organisms [8 - 10].

Taking into account the consequences of the presence of pharmaceutical residues in the environment, even at very low concentrations [11, 12], the need to develop new methods for their elimination seems really important.

The main methods used to remove organic pollutants from wastewater are: aerobic treatment [11], coagulation - flocculation [13], adsorption [14], and filtration [15]. These methods in general are not effective in removing pharmaceutical contaminants, and many of them (pharmaceutical contaminants) persist unaltered after treatment.

Many research groups have started to work on removing these pollutants from the environment

by proposing, developing and adopting new processes known as advanced oxidation processes (AOPs). These processes include photocatalysis [16, 17], electrolysis [18], Fenton processes [19], and electrooxidation [20]. These technologies (AOPs) are based on the formation and use of a powerful oxidant, called the hydroxyl radical (OH^*) [21]. One of the AOPs, electrochemical oxidation (or electrooxidation) is distinguished by its ease of use, its exceptional performance and its ability to combine with renewable energy sources, such as the sun [22].

The combination of several methods of removing pollutants has been the focus of many lines of research within the scientific community over the past ten years [23]. Regarding electrochemistry, its combination with other oxidation processes has made it possible to develop promising treatment processes with many advantages such as high oxidation capacity, fast reaction rate and high efficiency [24]. One of the oxidation processes commonly combined with electrochemistry is sonochemistry.

Since the 1990s, sonochemistry has aroused considerable interest in the proposed technique for the degradation of organic pollutants present in wastewater [25 - 27]. It is worth remembering that this method involves all the chemical processes affected by the cavitation bubbles [28] which can have physical / mechanical or chemical effects. Combining sonochemistry and electrochemistry thus leads to improved mass transport, increased current efficiency and

continuous activation of the working electrode surface [29, 30].

The objective of this work is to study the contribution of sonochemistry in the electro-oxidation of paracetamol on Boron-doped diamond electrode. The choice of this electrode was motivated by its excellent mechanical, optical and electronic properties and above all by its ability to mineralize organic compounds [31, 32]. In this work, the evolution of paracetamol concentration during treatment was monitored by differential pulse voltammetry (DPV).

2. Materials and methods

2.1. Chemical products

All chemicals used in this work were of analytical grade. They are: acetic acid (99.7%), supplied by Emparta; orthophosphoric acid (85%), sulfuric acid (96%), and boric acid (99.5%) supplied by Emsure; potassium ferrocyanide (II) and potassium ferricyanide (III), both supplied by Sigma-Aldrich. Paracetamol was purchased from a pharmacy in Abidjan. Figure 1 shows the structure of paracetamol.

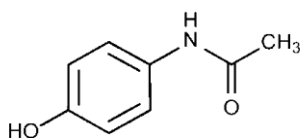


Fig. 1. Structure of paracetamol.

2.2. Electrochemical experiments

The electrochemical experiments were carried out using an AUTOLAB PGSTAT-20 (potentiostat / galvanostat) equipped with a USB electrochemical interface and driven by a GPES 4 (Eco Chemie) software package in conjunction with a system with three electrodes and a computer for data storage and processing. A three-electrode cell system composed of a saturated calomel electrode (SCE) as a reference electrode, a platinum wire as a counter electrode and boron-doped diamond (DDB) (geometric area = 1 cm²) as an electrode working was used for the electrochemical studies. In this work, all the measured potentials were related to the standard hydrogen electrode (SHE) from the relation:

$$E_{\text{SHE}} = E_{\text{SCE}} + 0.25 \quad (1)$$

2.3. Electrolysis

For the preparative electrolysis, the boron-doped diamond electrode was used as the anode and a zirconium plate was used as the cathode. These two electrodes, separated by 2 cm and each having a surface area of 45.37 cm², delimit a volume of 90.74 cm³. In this volume, the solution is recirculated using a mini peristaltic pump of the WATSON MARLOW 505U type. A FABRIMEX 141 type voltage generator allowed us to keep the current constant during the electrolysis.

2.4. Sonochemistry and sonoelectrochemistry

The sonochemistry and sonoelectrochemistry experiments were carried out in an ultrasound tank (model: Super RK 255, BANDELIN SONOREX) with a frequency of 35 kHz. This tank has been filled with distilled water to an optimum level, so as not to damage the ultrasonic system. It consists of three piezoelectric wave-producing transducers; which by propagating in the system contribute to the mineralization of the pollutant. The electrochemical cell was placed inside the ultrasonic tank. In the case of sonoelectrochemistry, the ultrasonic tank was coupled to the electrolyser.

2.5. Electrochemical analysis

The analytical method used to assess the degradation of paracetamol on the boron-doped diamond electrode is differential pulse voltammetry. This method made it possible to determine the evolution of the paracetamol concentration from a pre-established calibration curve. Before use, BDD was pretreated in sulfuric acid first and then in Britton-Robinson buffer solution. The purpose of these pretreatments was to purify the electrode. Regarding the first pretreatment, several voltammetric cycles (seven cycles) were carried out on the surface of the electrode, in a sulfuric acid solution (0.5 M), under a potential of -2 V and a pretreatment time 90 s [33]. As for the second pretreatment was carried out in a Britton-Robinson buffer solution (0.03 M), under a

potential of 2 V for 15 s. After these various pretreatments, the boron-doped diamond electrode was used for the electrochemical analysis of solutions containing paracetamol, by differential pulse voltammetry. The differential pulse voltammetric curves were obtained under the following conditions: $E_p = -2$ V; $t_p = 90$ s; modulation time ≥ 0.05 s; potential scan rate = 70 mV/s.

3. Results and discussion

3.1. Characterization of the boron-doped diamond electrode (BDD)

3.1.1. Behavior of the electrode in the presence of ferri / potassium ferrocyanide

The electrochemical characterization of the BDD electrode was carried out in a solution of sulfuric acid (0.3 M), used as a support electrolyte and in the presence of the couple ferri/potassium ferrocyanide (10, 20 and 50 mM). Voltammetric measurements were performed in the absence and / or presence of ultrasound (35 kHz). Figure 2a shows the intensity voltammetric curves (for different concentrations of the redox couple $Fe(CN)_6^{3-/4-}$) as a function of the potential scanning speed. As for Figure 2b, this concerns the evolution of the peak currents (anodic (I_{pa}) and cathodic (I_{pc})) as a function of the concentration of the redox couple. All these measurements were carried out in the presence of ultrasound.

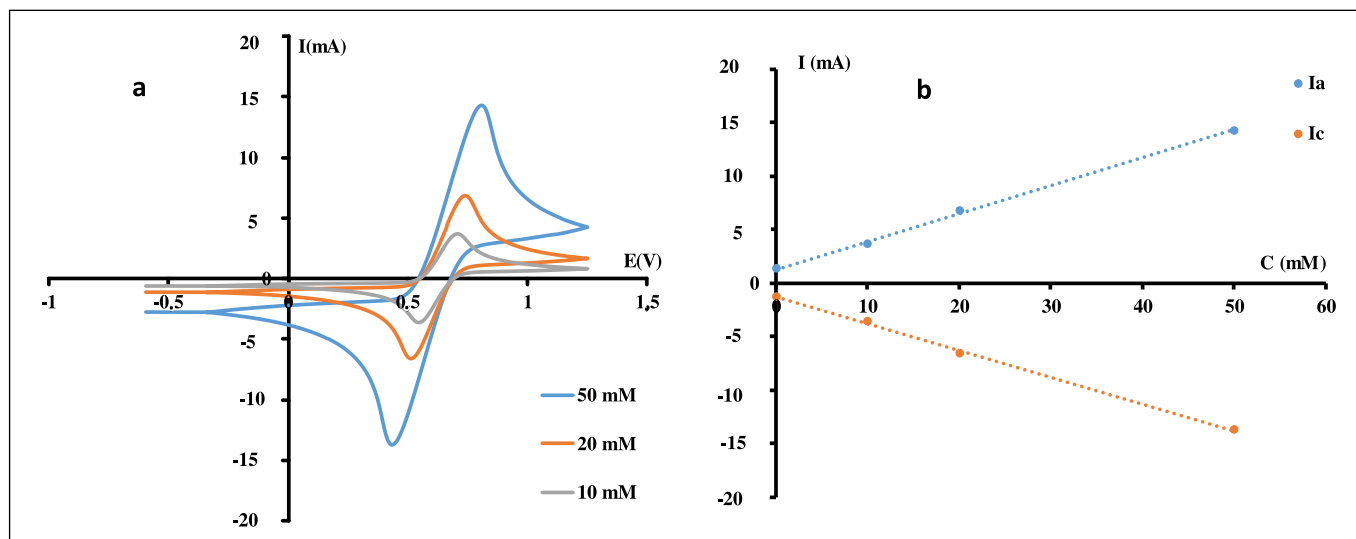


Fig. 2. (a) Voltammetric curves ($= 50 \text{ mV/s}$) at different concentrations of the redox couple $\text{Fe}(\text{CN})_6^{3-/4-}$; (b) Current intensity of anode and cathode versus the concentration of the redox couple; experimental conditions: $E_p = -2 \text{ V}$; $US = 35 \text{ kHz}$; WE: BDD, RE: SHE, CE: Pt wire, $T = 28 \text{ }^\circ\text{C}$; $\text{pH} = 6.0$.

In Figure 2a, the anode and cathode peaks observed correspond respectively to the oxidation and to the reduction of the redox couple. These observed peaks show that our working electrode (BDD) is a good electronic conductor [32, 33]. The heights of the anode and cathode peak currents increase with the concentration of the redox couple. The representation of the intensities of the anodic and cathodic peak currents versus the concentration of the redox couple, leads to straight lines substantially at the origin of the axes (Figure 2b). The slopes of the plots obtained are $0.3 \times 10^{-3} \text{ A mM}^{-1}$, with $R^2 = 0.99$ and $0.2 \times 10^{-3} \text{ A mM}^{-1}$, with $R^2 = 0.99$ for the peak intensities anodic and cathodic, respectively. The values of the slopes of the lines $I_{pa} = f([\text{Fe}(\text{CN})_6^{3-/4-}])$ and $I_{pc} = f([\text{Fe}(\text{CN})_6^{3-/4-}])$ are therefore different in the presence of

ultrasound. This difference is due to the existence of two competing reactions, the reduction of $\text{Fe}(\text{CN})_6^{3-}$ ions and the production of hydrogen, resulting from the decomposition of water. The presence of ultrasound is rather favorable to the release of hydrogen [34, 35]. In addition, the values of the cathode peak potentials decrease when the sonochemistry intervenes. Indeed, for ferri / ferro concentrations of 20 and 50 mM, the cathode peak potentials go from 0.57 and 0.46 V to 0.52 and 0.44 V respectively in the absence and in the presence of ultrasound. Despite these observations, the surface of the diamond did not undergo any modification. The result of the representation of the peak current (anodic or cathodic) as a function of the concentration of the redox couple $\text{Fe}(\text{CN})_6^{3-/4-}$, confirms the quasi-metallic character of BDD [31, 32].

3.1.2. Influence of sonochemistry on the behavior of BDD

To study the influence of sonochemistry on the behavior of the boron-doped diamond electrode, we compared the voltammograms obtained in the absence and then in the presence of ultrasound, for 50 mM of potassium ferri / ferrocyanide (Figure 3).

Figure 3 shows an almost overlay of the voltammograms in the absence and in the presence of ultrasound. However, the intensities of the anode peak currents in the presence of ultrasound are slightly higher. For a potential of 0.8V / SHE, the intensities of the anode peak current are 15.26 mA and 14.10 mA in the

presence and in the absence of ultrasound, respectively. This small difference is due to the low applied frequency (35 kHz). Indeed, the applied frequency conditions lead to the formation of radicals. These radicals participate to the degradation of organic compound, which is favorable to the increase in the anodic peak current. Frequencies between 100 and 1000 kHz are ideal for heavy radical formation, as the number of bubbles and their stability increase [35]. Above 1000 kHz, the bubbles formed are small and the production of radicals much lower [36]. Therefore, sonochemistry improves the electrochemical properties of the boron-doped diamond electrode.

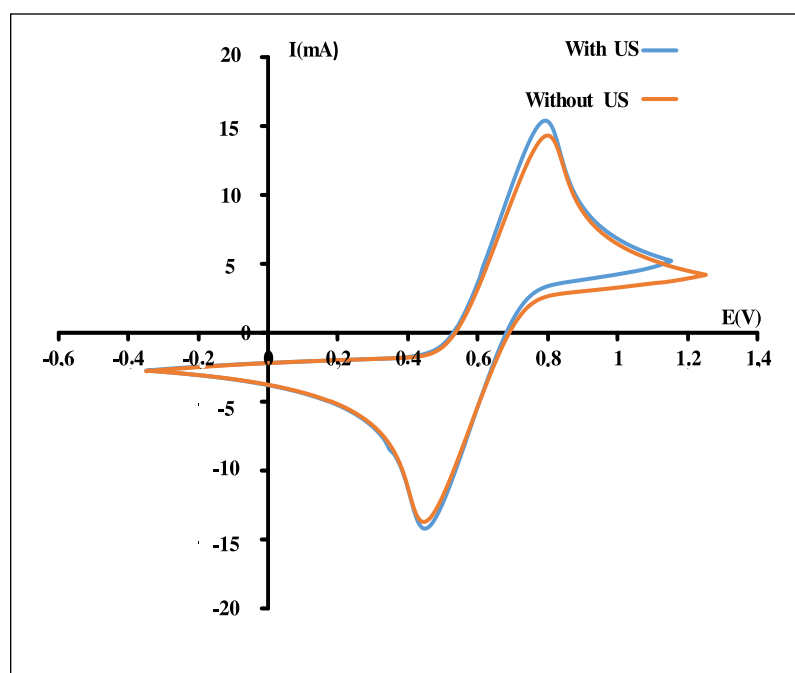


Fig. 3. Voltammetric curves (50 mV/s) of the redox couple $\text{Fe}(\text{CN})_6^{3-/4-}$ in the absence and in the presence of ultrasound; experimental conditions: $E_p = -2$ V; US = 35 kHz; WE: BDD, RE: SHE, CE: Pt wire, $T = 28$ °C; pH = 6.0.

3.2. Oxidation of paracetamol

3.2.1. Electrooxidation of paracetamol assisted by ultrasound

To study the influence of ultrasound on paracetamol mineralization (10 mM), we performed a comparative study. First, we worked under the singular effect of ultrasound and electrooxidation, and second, under the combined effect of ultrasound - electrooxidation. This work was carried out at a frequency of 35 kHz, a current density of 70 mA/cm² and in a solution of sulfuric acid (0.3 M). The results obtained have been presented in Figure 4. This shows differential pulse voltammograms of the change in paracetamol concentration during degradation by sonochemistry (a),

electrochemistry (b) and sonoelectrochemistry (c). In this figure, the peak oxidation current of paracetamol is observed at 0.9 V, regardless of the treatment method applied. This peak current decreases with the treatment time. This decrease in the intensity of the peak current is very slow for the degradation of paracetamol by sonochemistry, rapid for the degradation by electrooxidation (preparative electrolysis) and very rapid for the degradation by sonoelectrochemistry (sonochemistry combined with electrooxidation). Thus, combining sonochemistry and electro-oxidation leads to better mineralization of paracetamol than these methods taken separately.

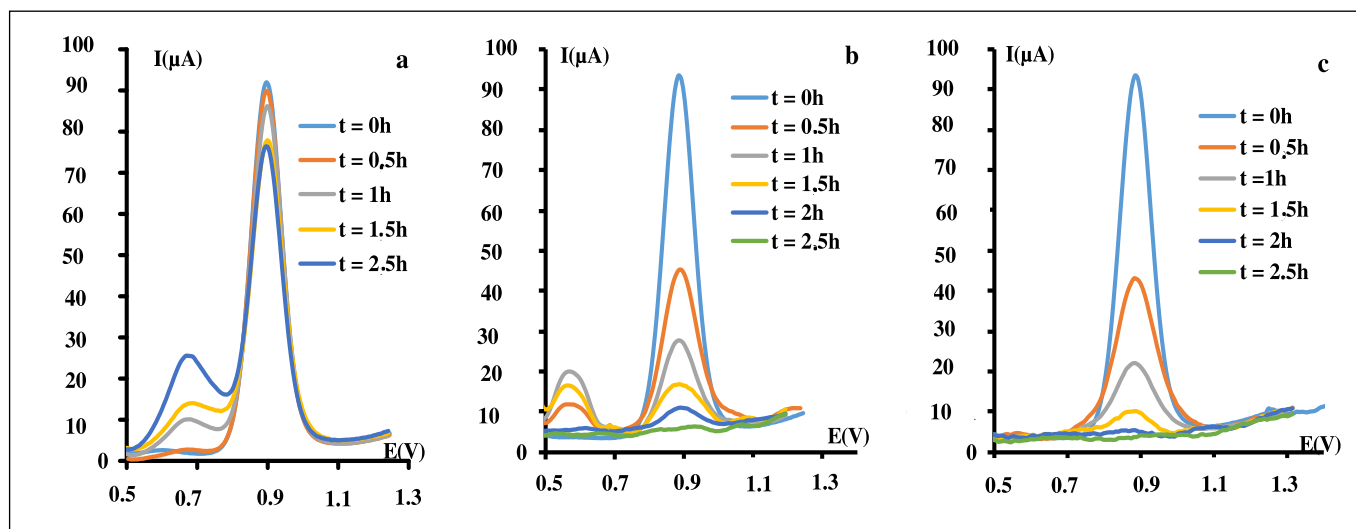


Fig. 4. Differential pulse voltammetric curves of sonochemical (US) (a) Electrochemical (EO) (b) and sonoelectrochemical (US + EO) (c) oxidation of paracetamol (10 mM) in sulfuric acid (0.3 M); $j = 70 \text{ mA/cm}^2$; WE = BDD; RE = SHE; CE = Pt wire; T = 28 °C; flow rate = 2.7 mL/s.

In-depth analyzes of the previous results made it possible to represent the evolution of the C/C_0 ratio (instantaneous concentration of paracetamol/initial concentration of paracetamol) over time and to determine its rate of degradation ($\eta = (1 - \frac{C}{C_0}) \times 100$), for the different treatment methods (Figures 5a and 5b). According to Figure 5a, the electrochemical and sonoelectrochemical methods lead to good degradation of paracetamol, unlike the sonochemical method. These results are in agreement with those of the literature, researchers during their work showed that sonochemistry alone is ineffective when it comes to treating wastewater containing complex organic pollutants [37]. In the case of this work, the degradation efficiencies are 18%; 73% and 89%, for sonochemistry, electrochemistry and sonoelectrochemistry after only 1.5h,

respectively (Figure 5b). Thus, combining sonochemistry with the electrolysis preparation of paracetamol made it possible to increase its degradation efficiency by more than 25%. This improvement in the degradation of paracetamol by the “sonochemistry-electrochemistry” combination is due to the large presence of hydroxyl radicals. Indeed, during the electrooxidation process using a non-active electrode (such as boron-doped diamond), hydroxyl radicals form on the surface thereof. These hydroxyl radicals thus formed participate in the degradation of paracetamol. The high degradation efficiency obtained in the case of sonoelectrochemistry is explained by the increase in mass transfer, the minimization of the fouling of the electrodes and the combined generation of radicals by ultrasound and the polarized surface of the DDB [38].

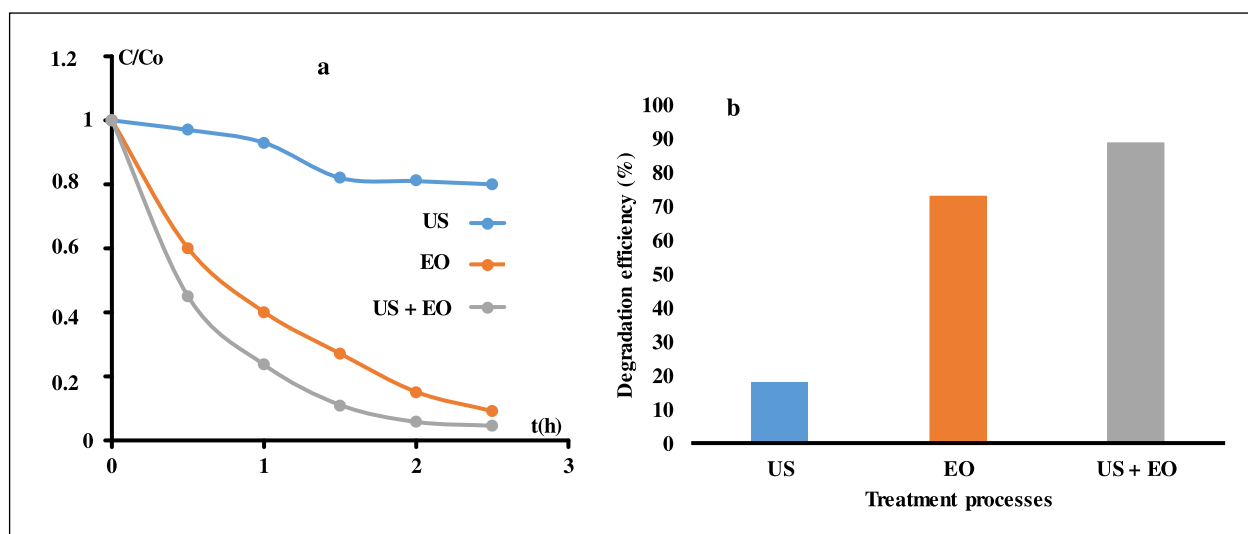
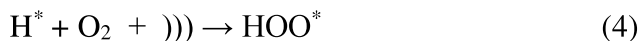
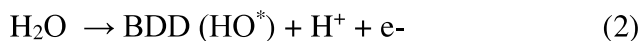
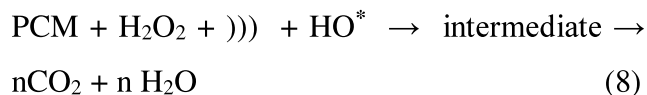


Fig. 5. (a) Comparative study of sonochemical (US), electrochemical (EO) and sonoelectrochemical (US + EO) oxidations of paracetamol (10 mM) in sulfuric acid (0.3 M); $j = 70 \text{ mA/cm}^2$; $T = 28 \text{ }^\circ\text{C}$; flow rate = 2.7 mL/s; (b) Degradation efficiency of paracetamol after 1.5 h of treatment, for the different methods used.

The reaction mechanism of the degradation of paracetamol would be [39, 40], where “)))” denotes US (Ultrasounds):



In addition, we are witnessing the production of H_2O_2 by recombination of the HO^* radicals, which will lead to complete mineralization of paracetamol:



The influence of sonochemistry on the evolution of the oxidation current efficiency of paracetamol was also studied (Figure 6). From this study, it emerges that combining sonochemistry with electrochemistry contributes to reducing current losses.

Sonochemistry has positive effects on the process of electrochemical degradation of organic matter. It contributes to the activation of the electrodes, the increase of the mass transfer and the increase of the current efficiency [28, 30, 38, 40 - 43].

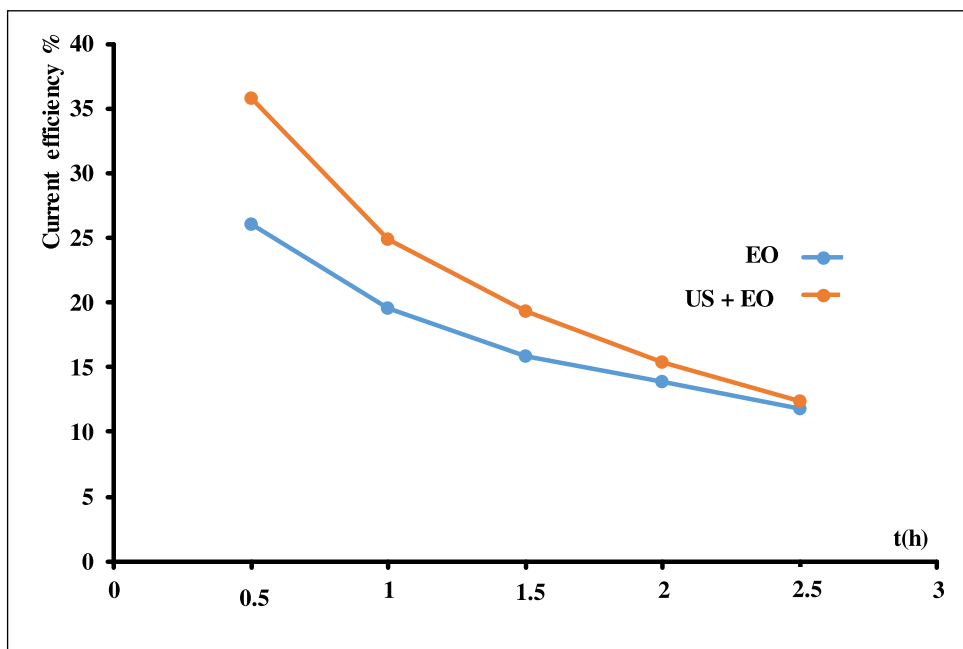


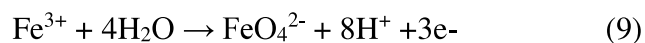
Fig. 6. Effect of sonochemistry on evolution of current efficiency versus the treatment time for a solution containing 10 mM of PCM; Anode: DDB (63 cm²) and cathode: Zr. $i = 70 \text{ mA/cm}^2$. $V = 250 \text{ mL}$.

3.2.2. Optimization of the quantity of ferrous ions in the electrooxidation of paracetamol

In order to determine the optimum amount of Fe^{2+} ions for efficient mineralization of paracetamol, electrolysis was performed in the presence of 0 mM; 10 mM; 50 mM and 100 mM Fe (II), in sulfuric acid (0.3 M), at a current density of 70 mA/cm^2 , on the boron-doped diamond electrode. Figures 7a and 7b show the results obtained.

In Figure 7a we observe a degradation over time of paracetamol, regardless of the concentration of ferrous ion added to the treatment. According to Figure 7b, the mineralization rate of paracetamol is 73; 80; 84 and 62%, respectively for the ferrous ion concentrations of 0; 10; 50 and 100 mM. Thus, the degradation of paracetamol is significant for Fe^{2+} concentrations of between 10 and 50 mM. For ferrous ion concentrations greater than or equal to 100 mM,

this is less important. The concentrations of ferrous ions between 10 and 50 mM, produce a sufficient quantity of Fe^{3+} ions which react with water molecules to form ferrate ions (FeO_4^{2-}). These ions participate in the degradation of paracetamol [44]. The formation of ferrate ions is described using the following equation:



On the other hand, the excess of ferrous ions causes a significant formation of Fe^{3+} . These ions formed (Fe^{3+}) would form a complex with the organic compound [45], thus delaying its degradation. The optimum concentration of ferrous ions for efficient degradation of paracetamol is 50 mM.

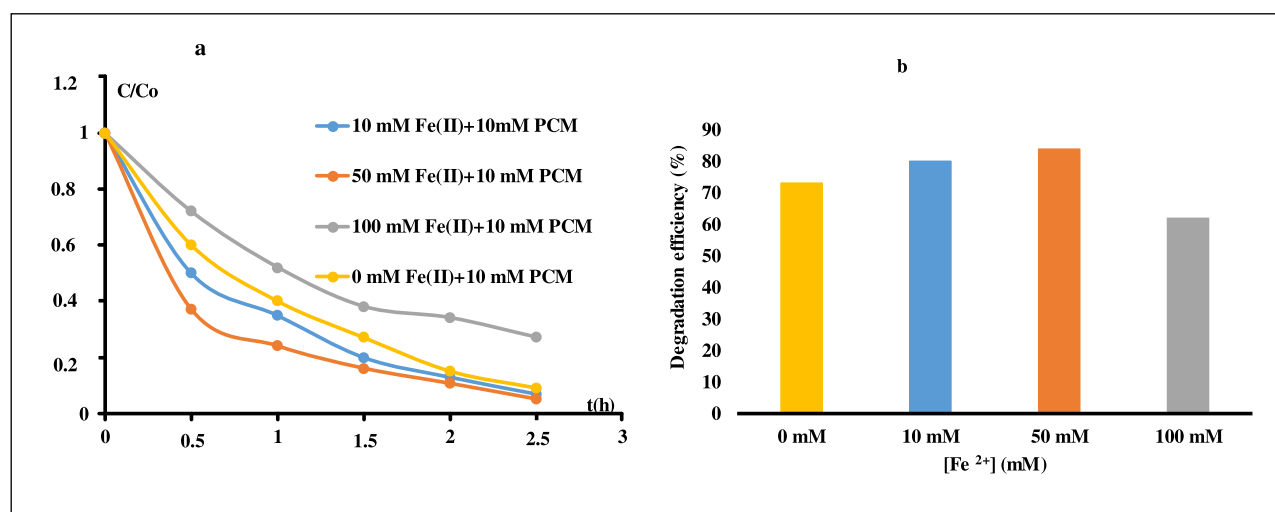


Fig. 7. (a) Evolution of the C/C_0 ratio versus the time; (b) Degradation efficiency of paracetamol after 1.5 h of treatment, for the different concentrations of ferrous ions used; $j = 70 \text{ mA/cm}^2$; $T = 28 \text{ }^\circ\text{C}$; WE = BDD; RE = SHE; CE = Pt wire.

3.2.3. Sonoelectrooxidation of paracetamol in the presence of Fe^{2+}

The degradation of paracetamol by sonoelectrochemical was carried out in the presence of 50 mM of Fe^{2+} , at a frequency of 35 kHz and a current density of 70 mA/cm². The variation in the amount of organic pollutant was followed by DPV method. The voltammograms obtained in the absence and then in the presence of ultrasound are shown in Figures 8a and 8b, respectively.

In Figures 8a and 8b, there are two current peaks, one at 0.6 V, corresponding to the peak oxidation current of Fe^{2+} ions and the other at 0.9 V, corresponding to that of paracetamol. Also, the oxidation peak of paracetamol tends to decrease a little more rapidly over time, in the case of sonoelectrochemical treatment. The oxidation peak of the ferrous ions decreases very slowly during these two treatments (electrochemical and sonoelectrochemical).

Figures 9a and 9b show the evolution of the concentration of the pollutant over time and the different oxidation rates, for electrochemical and sonoelectrochemical treatment, respectively. Figure 9a shows that the concentration of

paracetamol decreases rapidly in the presence of ferrous ions. This reduction is even more rapid when the electrochemical treatment is assisted by ultrasound. After 1.5 h of treatment, the rate of degradation of paracetamol which was 73%, in the absence of ferrous ions, increases to 84% in the presence of 50 mM of Fe^{2+} , in the case of electrochemistry not assisted by ultrasound. In the presence of ultrasound, the presence of ferrous ions does not significantly increase the rate of degradation. There is 89% degradation of paracetamol in the absence of ferrous ions and 92% when these ions are present in solution (Figure 9b). This variation would be due to the setting up in solution of the reaction of Fenton (Fe^{2+} / H_2O_2).

The acidic nature of the reaction medium promotes the establishment of the Fenton reaction which will considerably contribute to the oxidation of PCM. The determination of the current yields, for the degradation methods used, shows a large amount of current applied to sonoelectrochemical oxidation compared to electrochemical oxidation (Figure 10).

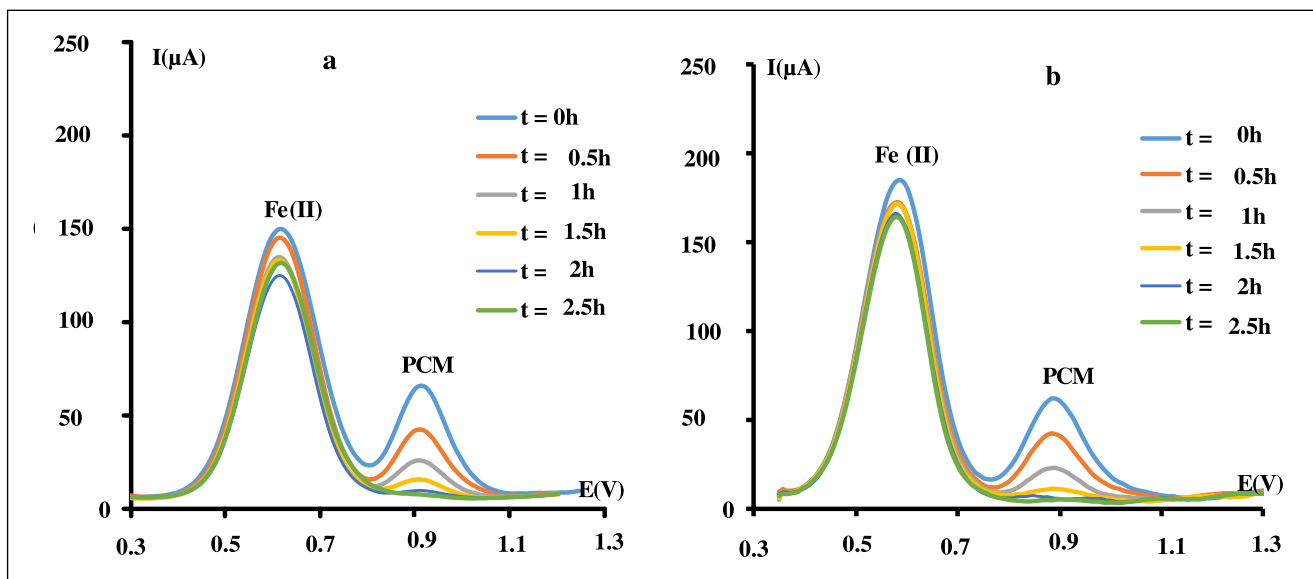


Fig. 8. Differential pulse voltammetric curves of the electrochemical (EO) (a) and sonoelectrochemical (US + EO) (b) oxidation of paracetamol (10 mM) in the presence of ferrous ions (50 mM); $j = 70 \text{ mA/cm}^2$; $T = 28 \text{ }^\circ\text{C}$; flow rate = 2.7 mL/s.

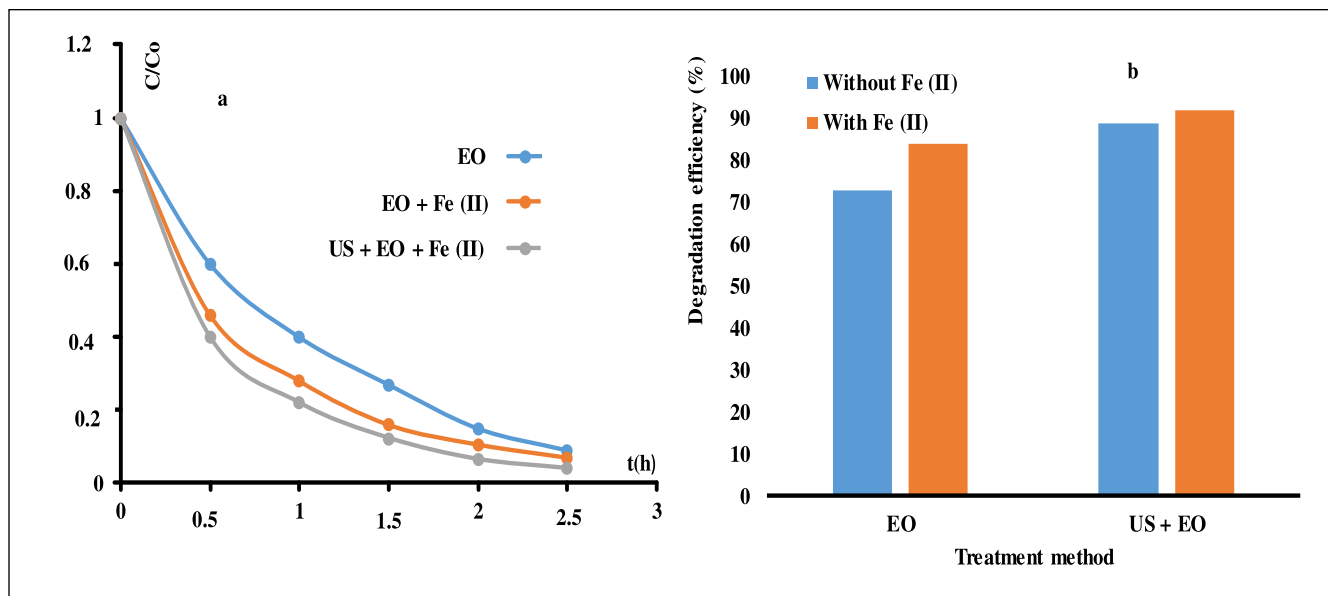


Fig. 9. (a) Degradation of paracetamol (10 mM) over time; (b) degradation rate for the treatment methods used; Fe (II) (50 mM); $j = 70 \text{ mA/cm}^2$; $T = 28 \text{ }^\circ\text{C}$; flow rate = 2.7 mL/s.

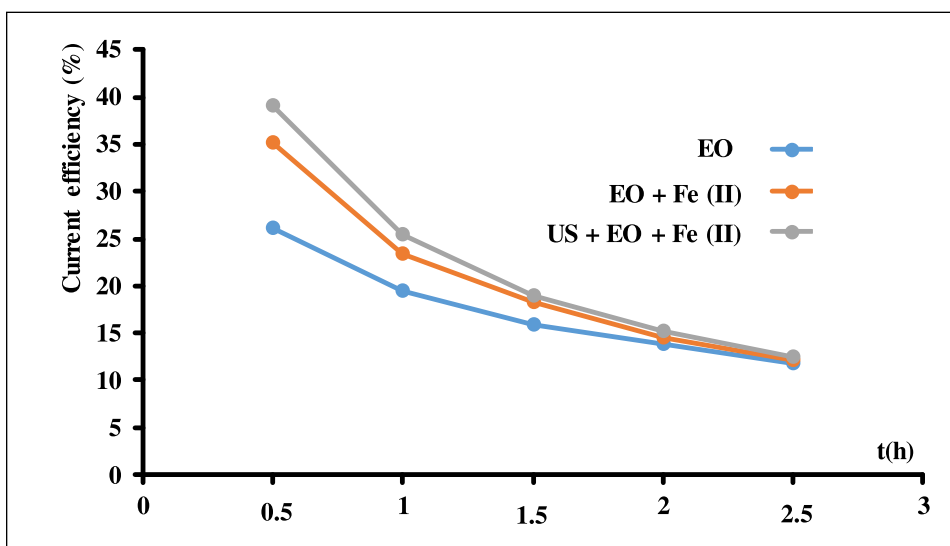


Fig. 10. Effect of sonochemistry on the change in current efficiency as a function of treatment time for a solution containing 10 mM of PCM and 50 mM of Fe^{2+} ; Anode: DDB (63 cm^2) and cathode: Zr. $i = 70 \text{ mA/cm}^2$; $V = 250 \text{ mL}$.

3.2.4. Synergistic effect (S)

In the case of treatment by sonoelectrochemistry, to verify if the different processes, namely sonochemistry and electrochemistry, act synergistically in the degradation of paracetamol, we have assumed that these two processes follow a pseudo-kinetics first order. Under these conditions, the synergistic effect is calculated according to Eq. (10) [46]:

$$s = 1 - \frac{k_{\text{US}} + k_{\text{EO}}}{k_{(\text{US} + \text{EO})}} \quad (10)$$

where k_{US} : apparent rate constant for the sonochemical treatment; k_{EO} : apparent rate constant for the treatment by electro-oxidation and $k_{(\text{US} + \text{EO})}$: apparent rate constant by treatment by sono-electrochemistry. If $s > 0$: synergistic effect; $s = 0$: cumulative effect and $s < 0$: antagonistic effect. Table 1 presents the

values of the apparent rate constants and the value of s obtained.

Table 1

Kinetic parameter and synergistic effect	US	EO	US + EO
	$k \text{ (h}^{-1}\text{)}$	0.11	0.95
S		0.21	

The values of the apparent rate constants show that by combining sonochemistry and electrooxidation, the rate of degradation of the organic pollutant is increased. In the case of this work, the degradation rate of paracetamol in the sonoelectrochemical treatment process is 1.4 times higher than that of electrooxidation and 12 times higher than that of sonochemistry.

$s = 0.21 > 0$, there is therefore a synergistic effect between sonochemistry and electro-oxidation in the sonoelectrochemical degradation of paracetamol.

4. Conclusion

In this study, synthetic wastewater, contaminated with paracetamol (10 mM), was treated with sonoelectrochemistry method, on a diamond electrode doped with boron. The first part of this work focused on the electrochemical characterization of the working electrode (DDB). The analysis of the voltammetric curves of the potential sweep in a solution of ferri / potassium ferrocyanide, made it possible to show the quasi-metallic character of the diamond electrode doped with boron. This metallic character of the electrode is reinforced in the presence of ultrasound. In the last part of this work, the characterized electrode was used in the sonoelectrochemical degradation of paracetamol. In order to demonstrate the contribution of ultrasound in sonoelectrochemical treatment, the degradation of paracetamol was carried out by sonochemistry (US), electrooxidation (EO) then by ultrasound coupling and electrooxidation (US-EO). The levels of mineralization of the paracetamol obtained are 18, 73 and 89% for the treatment by sonochemistry, electrooxidation and sonoelectrooxidation, respectively. Thus, coupling sono-chemistry to the preparative electrolysis of paracetamol makes it possible to increase its degradation rate by more than 25%, in the case of this work. The combination of ultrasound and electrooxidation can effectively degrade paracetamol than the two methods taken separately. The calculated degree of synergy value of 0.21 indicates that the elimination of

paracetamol is higher when the two processes (EO and US) are coupled, due to the synergistic effect between these processes.

Aknowledgements

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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