



Electrooxidation of the Paracetamol on Boron Doped Diamond Anode Modified by Gold Particles

**Kambiré Olló¹, Alloko Kouamé Serge Pacome²,
Pohan Lemeyonouin Aliou Guillaume³,
Koffi Konan Sylvestre² and Ouattara Lassiné^{2*}**

¹*UFR Sciences et Technologies, Université de Man, BP 20 Man, Côte d'Ivoire.*

²*Laboratoire de constitution et réaction de la matière, UFR SSMT, Université Félix Houphouët-Boigny de Cocody, Abidjan, 22 BP 582 Abidjan 22, Côte d'Ivoire.*

³*UFR Sciences Biologiques, Université Peleforo Gon Coulibaly de Korhogo, BP 1328 Korhogo, Côte d'Ivoire.*

Authors' contributions

This work was carried out in collaboration among all authors. Authors KO and AKSP designed the study, wrote the protocol and wrote the first draft of the manuscript. Authors KO and PLAG managed the literature searches and the analyses of the study. Authors OL supervised the work and instructed all the research. All authors read and approved the final manuscript.

Article Information

DOI: 10.9734/IRJPAC/2021/v22i430401

Editor(s):

(1) Dr. Farzaneh Mohamadpour, University of Sistan and Baluchestan, Iran.

Reviewers:

(1) Gülderen Uysal Akkuş, Afyon Kocatepe Üniversitesi, Turkey.

(2) Purvesh Shah, Gujarat University, India.

Complete Peer review History: <http://www.sdiarticle4.com/review-history/68514>

Original Research Article

Received 24 March 2021
Accepted 04 June 2021
Published 10 June 2021

ABSTRACT

The environment pollution, in particular that of the aquatic environment, by wastewater is a reality because it is discharged for the most part without treatment. The presence of pharmaceutical pollutants such as paracetamol in these waters can constitute a risk to human health. The objective of this work is to study the electrochemical oxidation of paracetamol using cyclic voltammetry on the boron doped diamond (BDD) anode and boron doped diamond modified by gold particles (Au-BDD) anode. The Au-BDD electrode was obtained by modifying the surface of BDD with gold particles. This was done by electrodeposition (chronoamperometry) in 0.5 M H₂AuCl₄ and 0.1 M H₂SO₄ using a three pulse nucleation and growth process. Physical characterization with Scanning Electron Microscopy coupled with Dispersive Energy spectroscopy has shown that the Au-BDD surface presents asperities with the presence of microparticles and nanoparticles. The electrochemical

*Corresponding author: E-mail: ouatlassine@yahoo.fr;

characterization made in three electrolytic solutions (H_2SO_4 , NaOH and KClO_4) showed that Au-BDD has a high electroactivity domain than that of BDD. The study of the Benzoquinone-hydroquinone redox couple has shown a quasi-reversible character of these two anodes. It also revealed that Au-BDD has a more accentuated metallic character than BDD. The voltammetric measurements made it possible to show that the paracetamol oxidation is limited by the transport of material on each anode. This oxidation is characterized by the presence of an anodic peak in the support electrolytes stability domain. The paracetamol oxidation is rapid on Au-BDD than on BDD in the various medium explored, thus showing that Au-BDD is more efficient than BDD for the paracetamol oxidation by electrochemical means.

Keywords: Paracetamol; oxidation; cyclic voltammetry, chronoamperometry; gold particles.

1. INTRODUCTION

Organic compounds such as pharmaceuticals present in different spheres of the environment have very varied origins [1,2]. The environmental fate of these compounds as well as their impact on the different ecosystems are very dependent on their interactions with the surrounding environment. Among these organic contaminants, involved in chemical pollution phenomena are antibiotics, analgesics, anti-inflammatories, contrast agents or antiepileptics and pyretics. The latter are introduced into the environment through anthropogenic activities: industrial discharges, animal manure, wastewater from various treatments, waste and unused drugs caused by runoff from landfills household waste or waste water [3-5]. Recent studies claim that hospital wastewater is the pharmaceuticals main source in the aquatic environment [3,6,7]. These substances come essentially from human consumption (prescription in pharmacies, hospital uses) and / or animal (veterinary treatments). They are the cause of various biological anomalies observed in many animal species and in humans [8]. A general awareness is therefore necessary in order to propose methods for treating hospital wastewater. Thus, considerable efforts are made by the implementation of new technologies for the depollution of these contaminated hospital waters before their discharge into the natural environment. The wastewater containing pharmaceutical effluents can be carried out by various processes, including physicochemical and biological treatments, which are currently well controlled at laboratory scale and applied on a large scale in industry [4,9,10]. In this context, several recent advances in the treatment by oxidation of organic contaminants contained in aqueous effluents have been made. These treatment methods are based on the formation of a highly reactive oxidizing chemical species which will transform the most recalcitrant molecules into biodegradable molecules or into

mineral compounds such as CO_2 and H_2O . These processes are called Advanced Oxidation Processes (AOP). They are based on the in situ formation of hydroxyl radicals ($\text{HO}\cdot$) which have an oxidizing power superior to that of traditional oxidants [11-13].

In recent years, there has been a great development of electrochemical methods, such as anodic oxidation and electro-oxidation, for the destruction of toxic and bio-refractory organic pollutants [4,14-16]. In this work, the electrochemical method will be used to oxidize paracetamol (PCM) is one of the most abundant drugs in the environment. It is a method that does not require the use of harmful reagents and its implementation is easy and very effective [17,18]. In particular, it has the advantage of being able to react to pollutants in two different ways, either directly or indirectly. However, the effectiveness of this method is linked to the nature of the electrode used [19]. Thus, this paracetamol oxidation will take place on two electrodes: the boron doped diamond electrode (BDD), known for its interesting electrochemical properties and its stability in highly concentrated acidic and basic medium. It has already been used for this purpose by Gnamba and al [12,20] in our laboratory to degrade pharmaceutical products. Then the several anode modified by gold particles which made headlines for the analytical determination of biomolecules or pollutants, as well as for spectroelectrochemical applications [21-24]. Indeed, the surfaces modification with gold nanoparticles improves the electrochemical response, in particular due to the increase in the active surface and the gold intrinsic properties in the size of the nanometer. It has also been used for the organic molecules reduction such as neuramidase by Wahyuni and al [25].

In this work, a protocol for depositing three nucleation and growth pulses (three pulses deposit) developed by F. Bottari and al is chosen

[26] to modify the surface of BDD by gold particles in order to obtain the BDD anode modified with gold particles (Au-BDD).

To carry out the PCM oxidation by electrochemistry, an exploratory study must be carried out on the optimal conditions for the PCM oxidation on these anodes, in order to avoid trial and error, thus saving time and reducing the cost. It becomes necessary to study this oxidation by voltammetric measurement. Thus, in this work, it will be on the one hand to modify the BDD surface by gold particles and on the other hand to study the voltammetric response of PCM on Au-BDD and BDD anodes.

2. MATERIALS AND METHODS

2.1 Chemicals

The pharmaceutical product used is paracetamol 500 mg manufactured by the Bailly-Creat France laboratory and purchased from a pharmacy in Abidjan. The chemicals used in this work composed of HAuCl_4 (LDP TORLAN), H_2SO_4 (Sigma-Aldrich), KClO_4 (Merck), NaOH (Panreac), $\text{C}_6\text{H}_4(\text{OH})_2$ (Panreac) and $(\text{C}_6\text{H}_4\text{O}_2)$ (Panreac). All the chemicals were used as received without any further treatment for the experiment. All the solutions used in the current work were prepared with distilled water. All the electrochemical experiments were made at ambient temperature of 25° C.

2.2 Electrodes

This research work was carried out with two working electrodes: the BDD anode and the BDD anode modified with gold particles (Au-BDD). The BDD anode was prepared by the chemical vapor deposition (CVD) method on filament heated between 2440 and 2560 ° C. The gas mixture used is composed of 1 % methane, an excess of dihydrogen and trimethyl boron. The deposit thickness is 1 μm . After doping, the atomic ratio of Boron to Carbon (B)/(C) diamond used is 3500. The Au-BDD anode was obtained by electrodeposition (chronoamperometry) with a protocol which includes three nucleation pulses and growth. The deposit method is as follows:

- BDD was treated by cyclic voltammetry in 0.1 M sulfuric acid with 20 potential scan cycles in a potential domain from - 1.17 to 2.53 V/SCE at 100 mV/s for hydrate the anode surface;

- The electrochemical deposition of gold particles (AuNPs) on BDD surface was carried out using the method of F. Bottari and al [26]. To test the effect of electrode surface terminations on the AuNPs deposition, electrochemical pretreatments were performed after the polishing step. Cathodic pretreatment was carried out at - 2 V for 30 min in 0.1 M phosphate buffer pH 2 and anodic pretreatment at +2 V for 30 min in 0.1 M phosphate buffer pH 2;
- The electrochemical deposition of AuNPs on the surface of BDDs was carried out in a 0.1 M H_2SO_4 solution containing 0.5 M HAuCl_4 . The protocol includes three nucleation growing pulses (three-pulse deposition). The potential and duration of the pulses induce a fast reduction of gold avoiding extensive hydrogen evolution and thus maintaining the surface termination of the electrode. A nucleation pulse consists in 2 s at - 0.7 V, followed by growth pulses at 0 V (150 s, 300 s, 800 s respectively). After rinsing the electrode with H_2O the electrode is further characterized [23].

2.3 Physical Characterization of the Electrodes

The electrodes used for this work were physically characterized from Energy Dispersive X-Ray Spectroscopy (EDX) and from microphotos taken using the scanning electron microscopy (SEM) apparatus.

2.4 Electrochemical Measurements

For the voltammetric study, an ECHOCHEMIE PGSTAT 20 potentiostat autolab connected to a computer and controlled by GPES software was used. A conventional three-electrode cell containing 100 mL of electrolytic solution was used for the electrochemical characterization with BDD and Au-BDD as working electrodes (WE), a platinum wire wound as a counter electrode (CE) and a saturated calomel electrode as reference electrode (RE) (containing saturated KCl solution). The working anode is fixed in a cage with an area of 1 cm^2 , promoting contact between it and the electrolyte through an opening. The reference electrode is placed in a capillary luggin, the end of which is placed about 2 mm from the working electrode in order to avoid the ohmic drop.

3. RESULTS AND DISCUSSION

3.1 Physical Characterization of the Anodes

The BDD anode characterization as well as the prepared electrode (Au-BDD) was carried out by scanning electron microscopy (Fig. 1). On the BDD electrode (Fig. 1.A) one can distinguish randomly oriented poly-dispersed crystals whose size is in the order of a few micrometers whose predominant faces are cubic and triangular. It also shows the polycrystalline nature of BDD with grains strongly bonded to each other [12,20]. The

images of the prepared Au-DDB electrode (Fig. 1.B) show that its surface presents asperities with the presence of microparticles and nanoparticles on its surface. This reflects the modification of the BDD surface by the gold particles (microparticles and nanoparticles). These gold particles are uniformly distributed over the surface of the DDB. However, at the bottom of the gold particles, boron doped diamond crystals are observed. This shows that our electrode is made up of diamond crystals doped with boron on which are fixed gold particles [26].

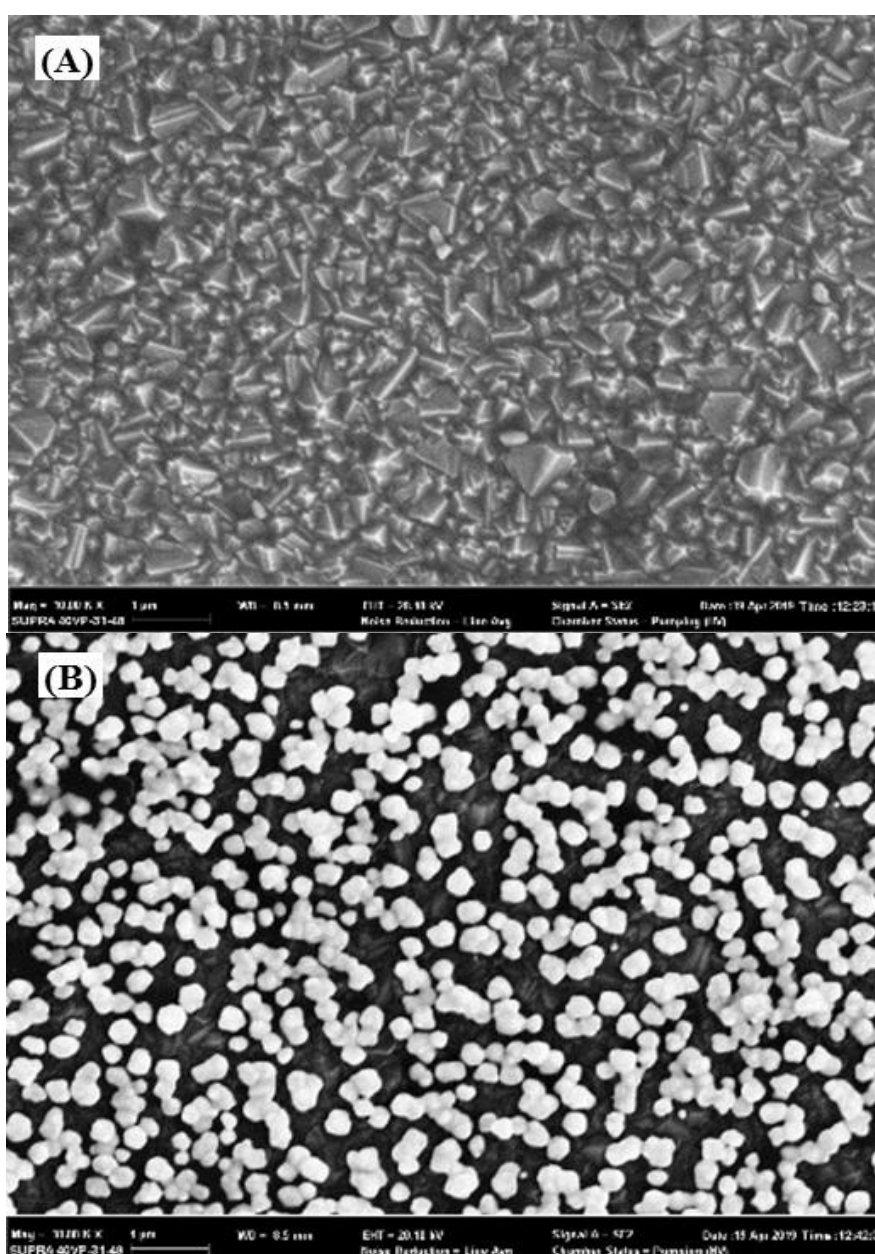


Fig. 1. Scanning electron micrographs of BDD (A) and Au-BDD (B)

In order to determine the chemical composition of the electrodes surface and the different proportions of these elements, Energy Dispersive X-Ray Spectroscopy (EDX) measurements were carried out. Fig. 2.A shows the EDX spectrum of the BDD electrode. On this spectrum, two intense peaks at 0.1 KeV and 1.9 KeV are observed which respectively characterize the presence of carbon and silicon. Note also the presence of oxygen on the electrode surface. Fig. 2.B shows the EDX spectrum of the Au-DDB electrode. On this spectrum, we have the presence of the characteristic peaks of carbon and silicon which were present on the spectrum of BDD. Several characteristic peaks of gold on the electrode surface is also observed. This shows the modification of the BDD surface by gold particles. These results confirm those obtained by the SEM.

3.2 Electrochemical Characterization of the Anodes

3.2.1 Benzoquinone-hydroquinone couple

Fig. 3.A shows the cyclic voltammetry of BDD in different concentrations of the benzoquinone-

hydroquinone redox couple at 25 mV/s. Note that the anode and cathode current intensity peaks increases in absolute value with the concentration of the redox couple. The representation of these peaks current densities as a function of the redox couple concentration gives straight lines with correlation coefficients substantially equal to one. Fig. 3.B below gives the results obtained. These investigations show a linearity between the current densities of the peaks (oxidation (j_{pa}) and reduction (j_{pc})) and the concentration. These results give the analytical behavior of the electrode.

Fig. 4.A shows the voltammetric curves of Au-BDD in different concentrations of the redox couple. This figure shows that the intensity of the peak current density increases with the redox couple concentration. The plot of the densities evolution of the anodic and cathodic peak currents as a function of the redox couple concentration, gives lines of correlation coefficients close to 1 (Fig. 4.B). This result shows that the peaks observed are actually linked to the concentration [27]. This shows that the Au-BDD anode can be used for analytical purposes.

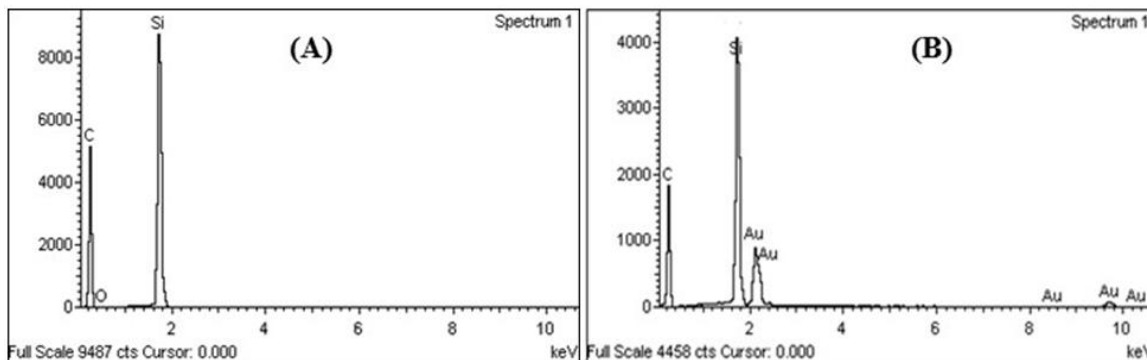


Fig. 2. EDX spectrum of the DDB (A) and Au-DDB (B) anodes

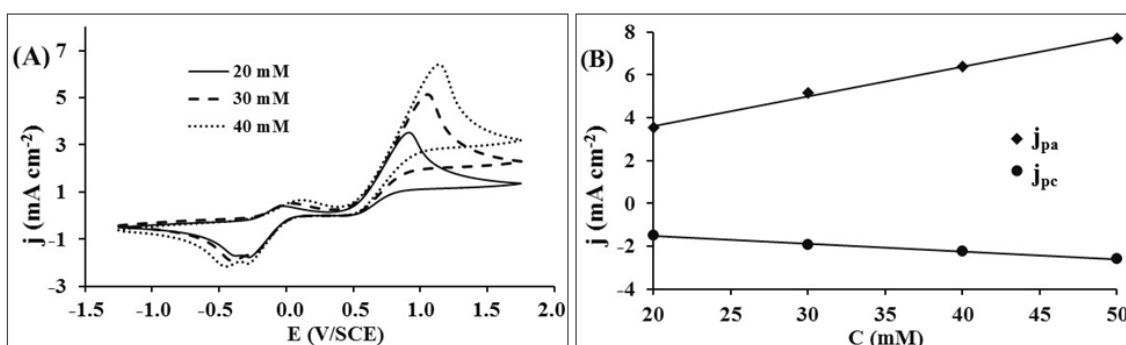


Fig. 3. (A) Cyclic voltammograms of BDD performed at different concentrations of the benzoquinone-hydroquinone couple at 25 mV/s; (B) Trend curves of the anodic and cathodic peak current densities as a function of the benzoquinone-hydroquinone couple concentration.

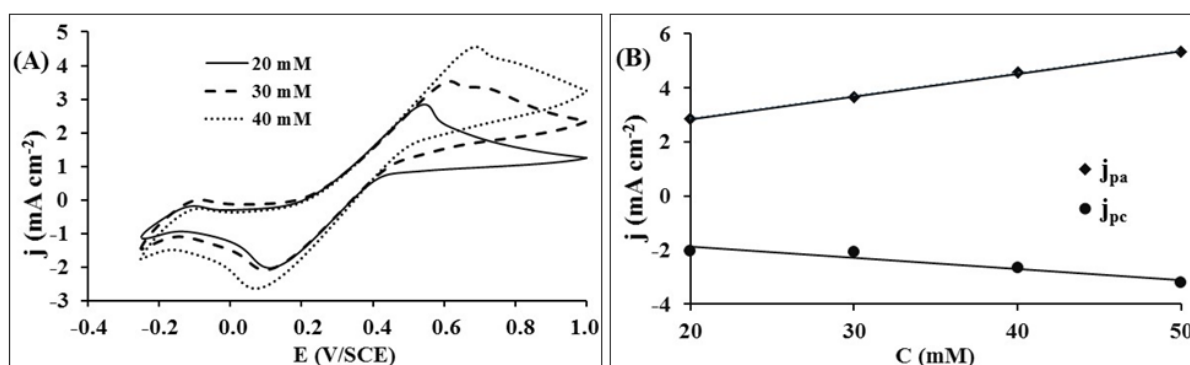


Fig. 4. (A) Cyclic voltammograms of Au-BDD performed at different concentrations of the Benzoquinone-hydroquinone couple at 25 mV/s; (B) Trend curves of the anodic and cathodic peak current densities as a function of the benzoquinone-hydroquinone couple concentration.

In Fig. 5 is presented the cyclic voltammograms of BDD and Au-BDD in the benzoquinone-hydroquinone couple of 20 mM concentration. This figure shows that the potential of the Au-BDD oxidation peak is lower than that of BDD. However, that of its reduction peak is higher than that of BDD. It is also note that the transport of the charges is faster with Au-BDD than BDD. This shows that Au-BDD has a more almost metallic character than BDD.

3.2.2 Sulfuric acid medium

In Fig. 6 are shown the cyclic voltammograms of our electrodes in acidic medium (0.1 M H_2SO_4). The BDD voltammogram presents three regions. From -1 to -0.67 V/SCE, there is a rapid increase in absolute value of the current density corresponding to the hydrogen evolution. Then from -0.67 V/SCE to 1.82 V/SCE, an absence of electrochemical reaction translated by an almost zero current density is observed. In fact, there was no change (characteristic peak in the formation of an oxide layer) of the anode surface in the electrolytic solution. This zone is called the anode electroactivity domain or the support electrolyte stability domain with a potential window of $\Delta E = 2.49$ V reflecting its stability with respect to acid corrosion. From 1.82 V/SCE, a rapid increase in the current density characterizing the oxygen evolution reaction is observed. The characteristic $j = f(E)$ of Au-BDD has, in the forward potential scan, a peak characteristic of the gold oxidation at 1.22 V/SCE. We note the presence of a couple of peaks in the return potential scan at 0.53 and -0.046 V/SCE, reflecting the gold oxides reduction. The presence of these peaks shows that the anode surface contains gold particles [26]. Au-BDD would therefore have more active

surfaces than BDD. From 1.91 V/SCE, there is a rapid increase in the current density corresponding to the oxygen evolution reaction.

Fig. 6 shows that BDD produces oxygen more quickly than Au-BDD, on the other hand it produces hydrogen less quickly. Indeed, the hydrogen evolution reaction and oxygen evolution reaction are respectively at -0.67 V/SCE and 1.82 V/SCE for BDD; -0.6 V/SCE and 1.91 V/SCE for Au-BDD. Therefore BDD is more electrocatalytic for the oxygen evolution than Au-BDD. On the other hand, it is less so for the hydrogen evolution.

In order to verify the relationship between the oxidation and reduction peaks, a study was carried out by varying the potential of the anodic potential zone in an H_2SO_4 solution (0.1 M) at 25 mV/s (Fig.7). By reducing the positive limit of the potential scanning area, it is we found that the intensity of the reduction peak current densities decreases in absolute value. This result shows that the oxidation and reduction peaks are linked. The oxides formed in the forward potential scan are reduced in the return potential scan.

3.3 Paracetamol Oxidation

3.3.1 Paracetamol oxidation in sulfuric acid medium

Fig. 8.A shows the cyclic voltammograms of the anodes in the absence and in the presence of PCM in H_2SO_4 (0.1 M). In the tablet presence, a change in the voltammograms appearance is observed. With regard to the BDD anode, there is the appearance of an oxidation peak at a potential of 0.9 V/SCE of current density 2 mA.cm⁻² for 1.5 g/L. The PCM addition also resulted in a lowering of the oxygen evolution

reaction current density and an increase in the potential for the oxygen evolution reaction start. In the absence of PCM, the O₂ evolution reaction begins at 1.82 V/SCE, on the other hand in the

presence of PCM the O₂ evolution reaction begins at 2.2 V/SCE. As the paracetamol concentration increases, the intensity of the oxidation wave current density increases [28].

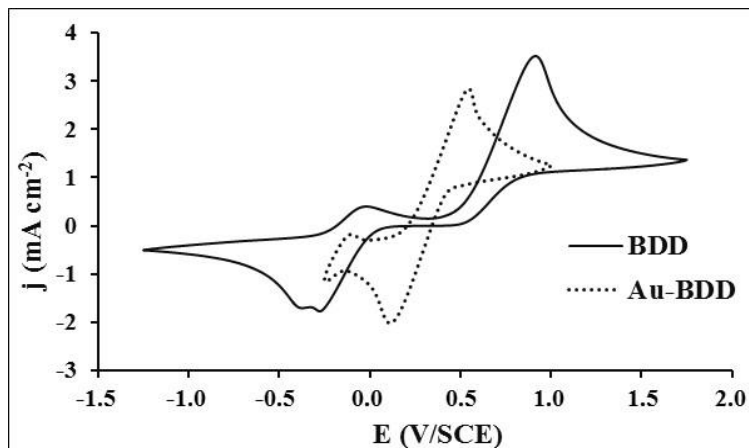


Fig. 5. Cyclic voltammograms of the anodes in the benzoquinone-hydroquinone couple 20 mM at 25 mV/s

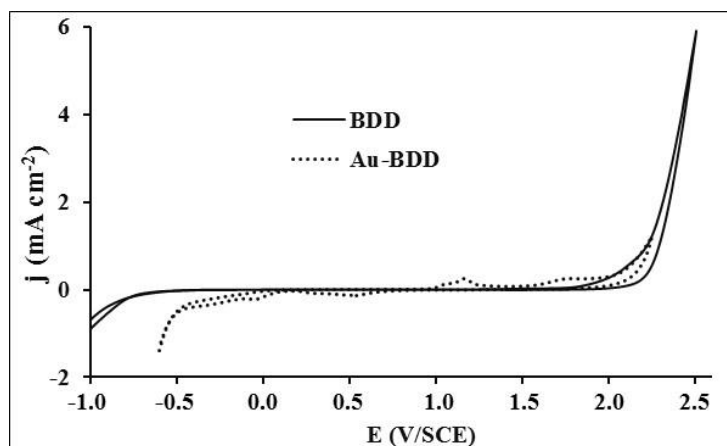


Fig. 6. Cyclic voltammograms of the anode in H₂SO₄ (0.1 M) at 25 mV/s

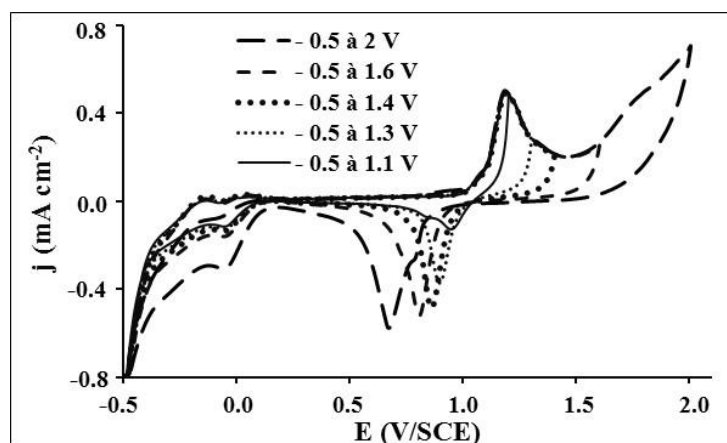


Fig. 7. Cyclic voltammograms of Au-BDD in the H₂SO₄ solution (0.1 M) at 25 mV/s with different potential windows

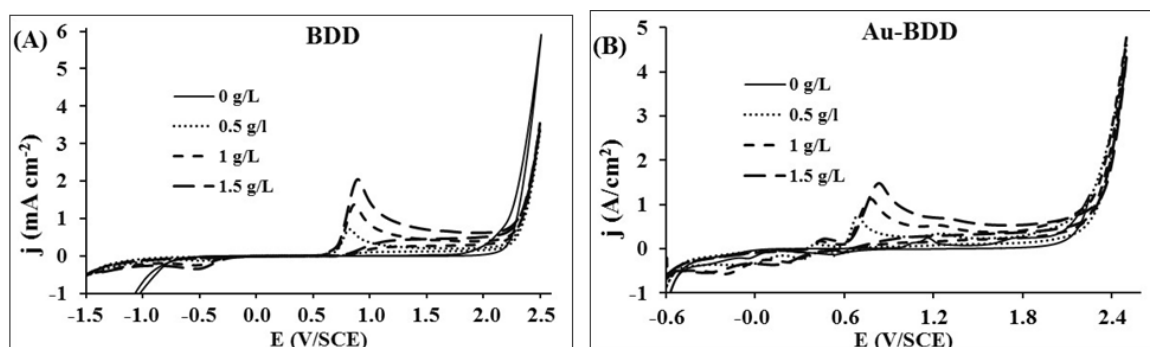


Fig. 8. Cyclic voltammograms of the anodes containing different concentrations of PCM at 25 mV/s

In Fig. 8.B (Au-BDD anode), in the presence of PCM, two oxidation peaks at 0.53 and 0.87 V/SCE are observed, then an oxidation wave around 1.37 V/SCE in the forward potential scan. In the presence of PCM, the potential for the start of oxygen evolution reaction increases. It goes from 1.91 in the absence of PCM to 2.2 V/SCE in the presence of PCM 1.5 g/L. This shows that the oxygen evolution reaction is delayed by the increase in the paracetamol concentration. In the return potential scan of the voltammogram, there are two reduction peaks at 0.2 and at -0.35 V/SCE. These voltammograms show an increase of the oxidation peak current density with the PCM concentration. The increase in the oxidation wave current density with the PCM concentration is confirmed by the plotting of the oxidation wave current density as a function of the concentration which gives lines of correlation coefficient close to 1 (Fig. 9). The proportionality between the oxidation peak current density and the PCM concentration confirms that these peaks are linked to the PCM oxidation [27]. PCM oxidation takes place in the stability domain of the supporting electrolyte. This indicates that the PCM oxidation on BDD and Au-BDD in H_2SO_4 medium is direct [28].

Fig. 10 presents the cyclic voltammograms of BDD and Au-BDD in H_2SO_4 medium (0.1 M) containing PCM 1.5 g/L at 25 mV/s. The Au-BDD oxidation peak current density is lower than that of BDD. It is also observed that the oxidation peak potential of Au-BDD is lower than that of BDD. This result shows that the PCM oxidation in H_2SO_4 medium with Au-BDD requires less energy than with BDD.

The paracetamol oxidation in H_2SO_4 medium on our anodes was explored by varying the scanning rate at potential. The results obtained

made it possible to show that the intensities of the oxidation current densities vary with the scan rates with the two electrodes. Plotting the oxidation peaks current densities as a function of the scan rate square root (Fig. 11) gives lines with correlation coefficients close to 1 for the two electrodes. There is therefore a linear relationship between these two parameters. This reflects the fact that the oxidation process of paracetamol is controlled by diffusion [27].

The temperature influence of the support electrolyte on the PCM oxidation has been studied. These studies show a decrease in the value of the gas evolution potential as well as an increase in the oxidation current density with temperature for the two types of electrodes. On the other hand, the plot of the natural logarithm as a function of the temperature inverse $\ln(I_p) = f(1/T)$ (Fig. 12) gives lines. The calculation of the activation energies of this reaction (paracetamol oxidation) from the slope of the line $\ln(I_p) = f(1/T)$ gives $E_a = 6,815 \text{ kJ} \cdot \text{mol}^{-1}$ with the BDD anode and $E_a = 12.437 \text{ kJ} \cdot \text{mol}^{-1}$ with the Au-BDD anode. These values are less than $40 \text{ kJ} \cdot \text{mol}^{-1}$, this confirms that the reaction is controlled by diffusion on these electrodes [29,30].

3.3.2 Paracetamol oxidation in neutral (KClO_4) medium

The cyclic voltammograms recorded in KClO_4 medium (0.1 M) with BDD and Au-BDD are presented in Fig. 13.A. With the BDD electrode we have the hydrogen evolution from -1.5 to -0.2 V/SCE. On this voltammogram, a rapid evolution of the current density from 1.6 V/SCE is observed. The electroactivity domain of the electrode is 3.1 V. In this domain, there is an almost zero current density followed by an oxidation wave (1.6 to 2.2 V/SCE) which would be due to the presence of impurities or an oxide

layer on the anode surface. Indeed the presence of oxide layer would be responsible for an increase in the current density as well as redox surface transitions. In addition, the surface oxide

formation would cause a decrease in the anode stability. Hence the electrode would be more stable in the potential range from - 0.2 to 1.6 V/SCE.

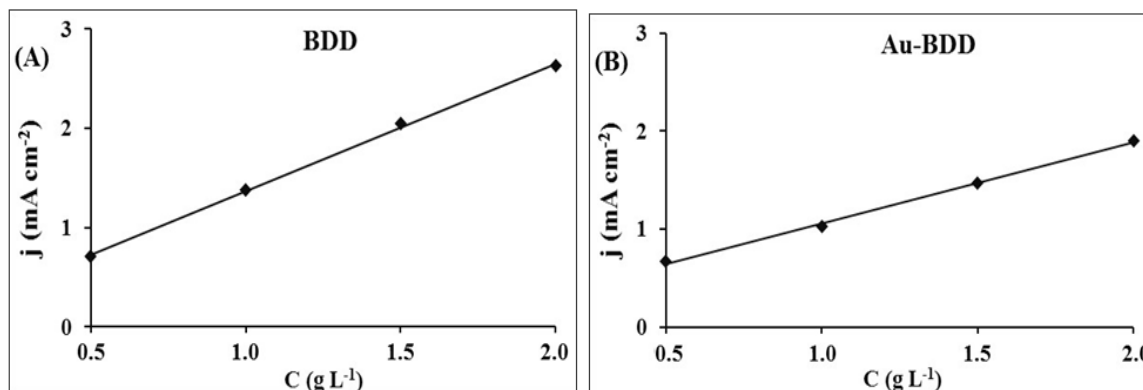


Fig. 9. Evolution of the anodic wave current densities resulting from the PCM oxidation as a function of the PCM concentration at 25 mV/s

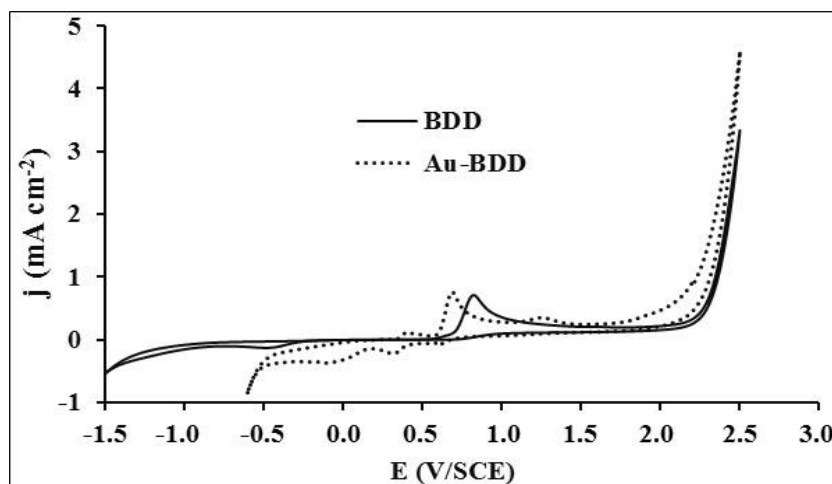


Fig. 10. Cyclic voltammograms of the anodes in H₂SO₄ with PCM 1.5 g/L at 25 mV/s

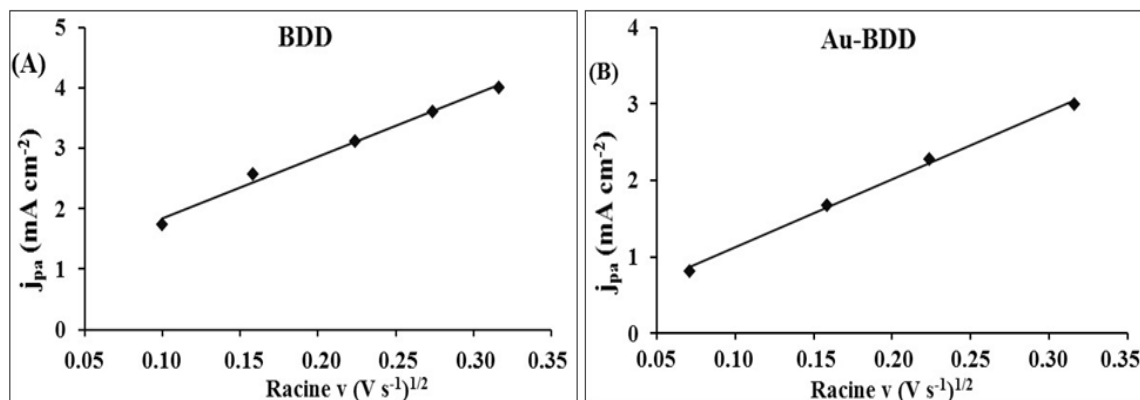


Fig. 11. Evolution of the PCM oxidation currents density as a function of the scan rate square root (A) and as a function of the natural logarithm (B) in H₂SO₄ (0.1 M), 2 g/L of PCM

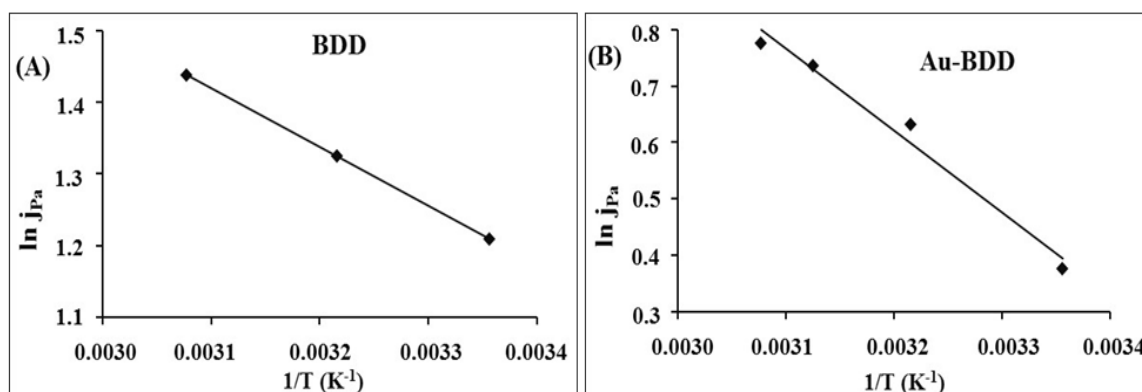


Fig. 12. Curve $\ln(j_p) = f(1/T)$ of DDB and Au-DDB anode in H_2SO_4 with PCM 1.5 g/L at 25 mV/s, CE: Pt, ER: SCE

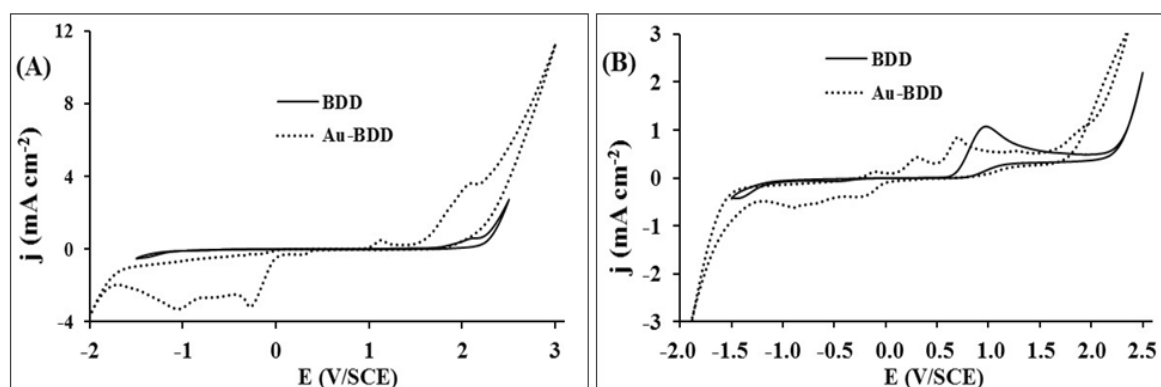


Fig. 13. Cyclic voltammogram of the anodes in $KClO_4$ (0.1 M) and in $KClO_4$ containing 1 g/L of PCM at 25 mV/s

The cyclic voltammogram of BDD undergoes modifications in the presence of gold. With Au-BDD, a hydrogen evolution reaction from -1.8 V/SCE followed by a diffusion plateau between -1.8 and 0 V/SCE which would come from the reduction of dissolved oxygen. There is also observed from 0 to 1 V/SCE an almost zero current density domain followed by a peak at 1.2 V/SCE which would be related to the gold oxide formation. The oxygen evolution reaction begins at 1.7 V/SCE characterized by a rapid increase in current. Between the oxygen and hydrogen evolution potential domain, the potential window is $\Delta E = 3.5$ V. On return potential scan, we have two peaks at -0.21 and -1.06 V/SCE which would be due to the reduction gold oxides. The phenomena observed attest that the BDD surface has been modified.

Fig. 13.B shows that the Au-BDD electrode has oxidation peaks in the presence of PCM, on the other hand the BDD electrode has a single

oxidation peak. These observations state that the electronic transfer takes place on the electrode surface. The large number of oxidation peaks observed suggests that this electrode could be useful in the marking and detection of organic compounds [28]. The presence of these oxidation peaks shows a production, and then a possible intermediate compounds oxidation during the PCM oxidation. It is also observed that these oxidation peaks observed with Au-BDD all have lower potentials than that obtained with BDD. These results lead us to say that Au-BDD would be more effective than BDD for the PCM oxidation in the neutral medium.

3.3.3 Paracetamol oxidation basic (NaOH) medium

Fig. 14.A shows the cyclic voltammogram of BDD and Au-BDD in NaOH medium (0.1 M). In this medium, the BDD electroactivity domain is 1.31 V/SCE and from -0.72 to 0.59 V/SCE. It also

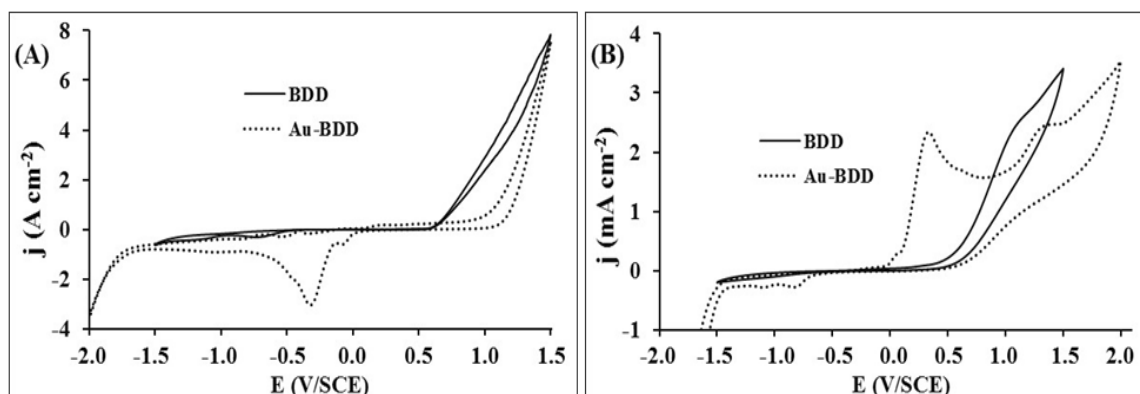


Fig. 14. Cyclic voltammograms of the anodes in NaOH (0.1 M) and in NaOH containing 1.5 g/L of PCM at 25 mV/s

has an almost zero current, thus showing its stability with respect to basic corrosion in this area. On the characteristic $j = f(E)$ of Au-BDD, we observe a hydrogen evolution at -1.67 V/SCE followed by a plateau (-1.67 to -1.2 V/SCE) reduction of dissolved oxygen. This plateau is upstream of an increase in current (-1.2 to -0.7 V/SCE). This is the area of mixed kinetics.

From - 0.7 to 0.95 V/SCE, in the forward potential scan we have a domain where the current is almost zero which corresponds to the double layer region. The result is a rapid increase in current density which marks the start of oxygen evolution reaction at 0.6 V/SCE. The support electrolyte stability domain is $\Delta E = 2.37$ V. On the return potential scan, it is observed a peak at -0.6 V/SCE which would come from the gold oxides reduction on the electrode surface characteristic of the electrode surface modification. Compared to BDD, Au-BDD oxygen evolution reaction less quickly in basic medium. Oxygen is produced from 0.59 V/SCE with BDD on the other hand with Au-BDD the oxygen evolution reaction begins at 0.98 V/SCE. The Au-BDD electrode would be more capable of oxidizing a variety of organic compounds in this solution because its field of electroactivity is wider.

Fig. 14.B shows the behavior of BDD and Au-BDD in the presence of PCM in NaOH (0.1 M). In the absence and presence of PCM, BDD has the same behavior up to approximately 0.7 V/SCE. From this value, we note a delay in the oxygen evolution reaction as well as a lowering of the oxygen evolution reaction current density in the presence of PCM 1.5 g/L. This shows that the PCM oxidation requires a high overvoltage. In

the domain -0.5 to 0.7 V/SCE we have a domain with an almost zero current which translates an absence of reaction on the anode surface in this domain. Then a rapid increase in current density is observed from 0.7 V/SCE. These results reflect the paracetamol oxidation on the anode surface. The absence of a peak could suggest that the PCM oxidation would be difficult in the basic medium. On the cyclic voltammogram of Au-BDD in the presence and in the absence of PCM a change in the shape of the curves in the presence of paracetamol is observed.

Indeed, we have the appearance of a peak followed by an oxidation wave in the forward potential scan in the presence of PCM. These results reflect the paracetamol oxidation on the anode surface. On the backwards potential scan, a reduction peak which is characterized by the reduction of the gold oxides. The oxidation wave current density at 1.2 V/SCE increases with the PCM concentration. This shows that this oxidation wave could be that of an intermediate compound. This figure shows that the oxidation peak potential on Au-BDD is low compared to the PCM oxidation potential on BDD. This shows that the Au-BDD anode oxidizes the PCM faster than the BDD anode in basic medium.

4. CONCLUSION

The deposit was made by electro-deposition by chronoamperometry made it possible to deposit gold particles (microparticles and nanoparticles) on the BDD anode surface. The physical characterization of the two electrodes confirmed the presence of the gold particles on Au-BDD surface. The electrochemical characterization measurements carried out in various support electrolytes, in particular the

hydroquinone/benzoquinone medium, have made it possible to show that these electrodes have almost metallic properties. Investigations with the redox couple also show that Au-BDD has a higher reaction kinetics compared to BDD. These electrodes have different kinetics at the level of the oxygen evolution reaction and hydrogen evolution reaction. Au-BDD has a wider of electroactivity domain than that of BDD. The voltammetric study carried out in the presence of paracetamol has shown that its oxidation results in an irreversible anode peak on the voltammogram in the support electrolyte stability domain. The process of oxidizing organic matter is limited by diffusion with the two electrodes. In the three medium explored (acid, neutral and basic), the PCM oxidation potential with the modified electrode is lower than that of BDD. It emerges from this study that the two electrodes are adequate for the PCM oxidation. However, the gold-modified electrode oxidizes organic matter faster than BDD with less energy. So it is more efficient and economical for the PCM oxidation.

ACKNOWLEDGEMENTS

We are thankful to the Swiss National Funds for its financial support. They funded the project (IZ01Z0_146919) which durability helped this work to be undertaken.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

REFERENCES

- Coulibaly B, Pohan LAG, Kambiré O, Kouakou LPS, Goure-Doubi H, Diabaté D, Ouattara L, Valorization of Green Clay from Bouaflé (Ivory Coast) in the Simultaneous Elimination of Organic Pollutants and Metallic Trace Elements by Adsorption: Case of Methylene Blue and Cadmium Ions. *Chemical Science International Journal*. 2020;29(8): 37-51.
- Vlachogianni T, Valavanidis A. Pharmaceuticals and Personal Care Products As Contaminants in the Aquatic Environment. A category of organic wastewater pollutants with special characteristics. *PHARMAKEFTIKI*. 2013;25(1):16-23.
- Hélène C-J, Dimitri S, Nabaz M, Frédéric H, Guillaume B, Véronique L, Philippe LC. Evaluation of pharmaceuticals in surface water: Reliability of PECs compared to MECs. *Environment International*. 2014;73:10–21.
- Appia FTA, Gnamba CQ-M, Kambiré O, Berté M, Sadia SP, Sanogo I, Ouattara L. Electrochemical Oxidation of Amoxicillin in Its Commercial Formulation on Thermally Prepared RuO₂/Ti. *J. Electrochem. Sci. Technol*. 2016;7(1):82-89.
- Dalecka B, Oskarsson C, Juhna T, Rajarao GK. Isolation of Fungal Strains from Municipal Wastewater for the Removal of Pharmaceutical Substances. *Water*. 2020;12(2):524.
- Bendz D, Paxeus NA, Ginn TR, Loge FJ. Occurrence and fate of pharmaceutically active compounds in the environment, a case study: Höje River in Sweden. *J. Hazard Mater*. 2005; 122(3):195-204.
- Angeles LF, Mullen RA, Huang IJ, Wilson C, Khunjar W, Sirotkin HI, McElroy AE, Aga DS. Assessing pharmaceutical removal and reduction in toxicity provided by advanced wastewater treatment systems. *Environ. Sci.: Water Res. Technol*. 2020;6:62–77.
- Heron RJL, Pickering FC. Health effects of exposure to active pharmaceutical ingredients (APIs). *Occupational Medicine*. 2003;53(6):357-362.
- Placide SP, Berté M, Loba EMH, Appia FTA, Gnamba CQ-M, Sanogo I, Ouattara L. Assessment of the Physicochemical and Microbiological Parameters of a Teaching Hospital's Wastewaters in Abidjan in Côte d'Ivoire. *Journal of Water Resource and Protection*. 2016;8:1251-1265.
- Chen W, Chang L, RenS-B, He Z-C, Huang G-B, Liu X-H. Direct Z-scheme 1D/2D WO_{2.72}/ZnIn₂S₄ hybrid photocatalysts with highly-efficient visible-light-driven photodegradation towards tetracycline hydrochloride removal. *Journal of Hazardous Materials*. 2020;384:121308.
- Kambiré O, Pohan LAG, Sadia SP, Kouadio KE, Ouattara L. Voltammetric study of formic acid oxidation via active chlorine on IrO₂/Ti and RuO₂/Ti electrodes. *Mediterranean Journal of Chemistry*. 2020;10(8):799-808.
- Quand-Meme GC, Auguste AFT, Hélène LEM, Mohamed B, Placide SS, Sanogo I, Ouattara L. Electrooxidation of Ceftriaxone in Its Commercial Formulation on Boron Doped Diamond Anode. *Journal of*

- Advanced Electrochemistry. 2016;2(2):85-88.
13. Pohan ALG, Ouattara L, Kondro KH, Kambiré O, Trokourey A. Electrochemical Treatment of the Wastewaters of Abidjan on Thermally Prepared Platinum Modified Metal Oxides Electrodes. *European Journal of Scientific Research*. 2013;94(1):96-108.
 14. Burgos-Castillo RC, Sirés I, Sillanpää M, Brillas E. Application of electrochemical advanced oxidation to bisphenol A degradation in water. Effect of sulfate and chloride ions. *Chemosphere*. 2018;194:812-820.
 15. He J, Li N, Zhang D, Zheng G, Zhang H, Yu K, Jiang J. Real-time monitoring of ciprofloxacin degradation in an electro-Fenton-like system using electrochemical-mass spectrometry. *Environ. Sci.: Water Res. Technol.* 2020;6:181-188.
 16. Hamous H, Khenifi A, Bouberka Z, Derriche Z, Electrochemical degradation of Orange G in K₂SO₄ and KCl medium. *Environmental Engineering Research*. 2020;25(4):571-578.
 17. Dewil R, Mantzavinos D, Poullos I, Rodrigo MA. New perspectives for Advanced Oxidation Processes. *Journal of Environmental Management*. 2017;195(2):93-99.
 18. Kouadio KE, Kambiré O, Koffi KS, Ouattara L. Electrochemical oxidation of paracetamol on boron-doped diamond electrode: analytical performance and paracetamol degradation. *J. Electrochem. Sci. Eng.* 2021;11(2):71-86.
 19. Pulgarin C, Alder N, Péringer N, Comninellis C. Electrochemical detoxification of a 1,4-benzoquinone solution in wastewater treatment. *Water Research*. 1994;28(4):887-893.
 20. Gnamba CQ-M, Appia FTA, Loba EMH, Sanogo I, Ouattara L. Electrochemical oxidation of amoxicillin in its pharmaceutical formulation at boron doped diamond (BDD) electrode. *J. Electrochem. Sci. Eng.* 2015;5(2):129-143.
 21. Liu G, Lu M, Huang X, Li T, Xu D. Application of Gold-Nanoparticle Colorimetric Sensing to Rapid Food Safety Screening. *Sensors*. 2018;18(12): 4166.
 22. Graczyk A, Pawlowska R, Jedrzejczyk D, Chworos A. Gold Nanoparticles in Conjunction with Nucleic Acids as a Modern Molecular System for Cellular Delivery. *Molecules*. 2020;25(1):204.
 23. Vidotti M, Carvalhal RF, Mendes RK, Ferreira DCM, Kubota LT. Biosensors based on gold nanostructures. *J. Braz. Chem. Soc.* 2011;22(1):3-20.
 24. Khongkow M, Yata T, Boonrungsiman S, Ruktanonchai UR, Graham D, Namdee K, Surface modification of gold nanoparticles with neuron-targeted exosome for enhanced blood-brain barrier penetration. *Scientific Reports*. 2019;9(1):8278.
 25. Wahyuni WT, Ivandini TA, Saepudin E, Einaga Y. Development of neuraminidase detection using gold nanoparticles boron-doped diamond electrodes. *Analytical Biochemistry*. 2016;497:68-75.
 26. Bottari F, De Wael K. Electrodeposition of gold nanoparticles on boron doped diamond electrodes for the enhanced reduction of small organic molecules. *Journal of Electroanalytical Chemistry*. 2017;801:521-526.
 27. Kambire O, Pohan LAG, Appia FTA, Ouattara L. Anodic Oxidation of Chlorides on Platinum Modified by Metallic Oxides. *Int. J. Pure Appl. Sci. Technol.* 2015;27(1):27-43.
 28. Koffi KM, Ouattara L. Electroanalytical Investigation on Paracetamol on Boron-Doped Diamond Electrode by Voltammetry. *American Journal of Analytical Chemistry*. 2019;10:562-578.
 29. Samet Y, Elaoud SC, Ammar S, Abdelhedi R. Electrochemical degradation of 4-chloroguaiacol for wastewater treatment using PbO₂ anodes. *Journal of Hazardous Materials*. 2006;138(3): 614-619.
 30. Belhadj-Tahar N, Savall A. Mechanistic Aspects of Phenol Electrochemical Degradation by Oxidation on a Ta/PbO₂ Anode. *J. Electrochem. Soc.* 1998;145:3427-3434.

© 2021 Olló et al.; This is an Open Access article distributed under the terms of the Creative Commons Attribution License (<http://creativecommons.org/licenses/by/4.0>), which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

Peer-review history:

The peer review history for this paper can be accessed here:

<http://www.sdiarticle4.com/review-history/68514>