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*Review*

# Chemically Modified Carbon-based Electrodes for the Detection of Paracetamol: A Short Review

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**Abstract**- Paracetamol (PCM) drug used in the treatment of pain, fever and headache, and it is considered safely for human use subjects. It has been found that the overdose and the chronic use of PCM produces toxic effects of environmental and immediate allergic hypersensitivity. Therefore, it is essential to develop sufficiently sensitive and simple sensors for precise determination of PCM. Chemically modified carbon-based electrodes have been widely used in this approach. This review is focused on the use of various chemically modified carbon-based electrodes such as glassy carbon, carbon paste, carbon screen-printed, graphene paste, and boron doped diamond electrodes in the electrochemical detection of PCM. Finally, we have briefly summarized the recent chemically modified carbon-based electrodes for the determination of PCM using articles encompassing 2018 until June 2020 and the efficiency of sensors are compared in terms of linear range, limits of detection and other proprieties can affect PCM detection as pH, medium, potential oxidation.

**Keywords-** Paracetamol; Carbon-based Electrodes; Electrochemical Sensor

## 1. INTRODUCTION

Paracetamol (PCM) also named acetaminophen is well known analgesic and antipyretic compound, which is widely used as drugs for the treatment of pain, fever, and headache. If the recommended dose is not exceeded, it is considered safely for human use subjects [1-3], but the overdose and the chronic use of PCM produces toxic metabolite accumulation which may result toxic effects of environmental, immediate allergic hypersensitivity and hepatotoxicity [4-6].

Therefore, it is essential to develop sufficiently sensitive and reproducible analytical methods for precise determination of PCM. Over the few years, encompassing 2018 to date, several techniques, have been used for the determination of PCM in different samples including high performance liquid chromatography (HPLC) [7,8], high performance liquid chromatography- ultraviolet spectrophotometry (HPLC-UV) [9-11], high performance thin layer chromatography- densitometric (HPTLC-densitometric) [12], ultra-high performance liquid chromatography-tandem mass spectrometry (UHPLC-MS/MS) [13,14], ultraviolet spectrophotometry (UV) [15-23], thermogravimetric analysis-chemometric approaches (TGA-SPA/PLS) [24], probe electrospray ionization-tandem mass spectrometry (PESI-MS/MS) [25], flow injection analysis-multiple pulse amperometry (FIA-MPA) [26], Excitation emission matrix spectroscopy-parallel factor analysis (EEMs- PARAFAC) [27] and fluorescent [28,29]. However, these techniques have some limitations and disadvantages such as high cost and long analysis time.

At the same time, numerous voltammetric methods, especially coupled with modified electrodes have been developed. Electroanalytical techniques have received tremendous attention, due to their high sensitivity and precision with relatively low instrumental costs. With the development of new materials, researchers have tried chemically modified electrodes for detection of PCM in various samples. Very low detection limits have been achieved with modified electrodes, while such detections were not possible with bare electrodes. Thus, the main goals of this review are to give the most recent advancements on chemically modified carbon-based electrodes for the determination of PCM encompassing 2018 to June 2020 (Science Direct database) and provides a comparable coverage of different targets of interest, linear range and the limit of detection (Tables 1, 2 and 3).

## 2. CHEMICALLY MODIFIED CARBON-BASED ELECTRODES FOR THE DETECTION OF PCM

In recent decades, chemically modified carbon-based electrodes, such as Glassy Carbon (GCE), Boron Doped Diamond (BDD), graphene paste (GPE), graphite (GPE) or carbon paste (CPE) and screen printed (SPE) electrodes have been used widely for the electroanalytical determination of PCM due to their biocompatibility, low cost, and fast electron kinetics.

Carbon-based electrodes are used either alone also as modifiers or in combination with other nanoparticles or organic materials.

**Table 1.** Analytical response characteristics of PCM sensor based on chemically modified glassy carbon electrode

| GCE modified by | Method | Linearity (μmol/L) | LOD (nmol/L) | Potential E (V) | pH | Medium (mol/L) | Ref. |
|---|---|---|---|---|---|---|---|
| $MoS_2/TiO_2$ | SWV | 0.5–750 | 100 | 0.39 (vs. SCE) | 7 | 0.1 PBS | [30] |
| f-MWCNTs/CTS-Co | DPV | 0.1-400 | 100 | (vs. Ag/AgCl) 0.32 | 7 | 0.2 PBS | [31] |
| PEDOT/AG | I-T | 0.15-5880 | 41 | (vs. SCE) 0.35 | 7 | 0.2 PBS | [32] |
| GPtNPs | DPV | 5-1490 | 5000 | 0.4 (vs. Ag/AgCl) | 7.4 | 0.1 PBS | [33] |
| FZ-G | SWV | 0.5–200 | 10 | (vs. Ag/AgCl) 0.42 | 4.5 | 0.1 PBS | [34] |
| $Fe_3O_4$/rGO | DP-ASV | 2 - 150 | 720 | (vs. Ag/AgC) 0.37 | 6 | 0.1ABS | [35] |
| GAIN/Cu | DPV | 1-700 | 12 | 0.444 (vs. SCE) | 6 | 0.1 PBS | [36] |
| $La^{3+}$-CuO/MWCNTs | DPV | 0.5-900 | 14 | 0.4 (vs. SCE) | 7 | 0.1 PBS | [37] |
| GO/$Fe_3O_4$@PMDA/Pd | DPV | 0.005-2.5 | 2.1 | 0.4 (vs. Ag/AgC) | 6.5 | 0.2 PBS | [38] |
| $Fe_2O_3$/RGO | DPV | 0.1-74 | 21 | 0.52 (vs. Ag/AgCl) | 4 | 0.1 PBS | [39] |
| $CeO_2$/CNT | DPV | 0.01–900 | 4.4 | 0.72 (vs. SCE) | 7 | 0.1 PBS | [40] |
| (Au/Ag/Pd)NPs/EPGrO | DPV | 5–700 | 120 | 0.38 (vs. Ag/AgCl) | 7 | 0.1 PBS | [41] |
| CuO-Au/MWCNTs | DPV | 0.2 - 6.0 | 16 | 0.25 (vs. SCE) | 7.5 | 0.1 PBS | [42] |
| AuNPs/CNTs-CONH-TAPP | DPV | 4.5–500 | 440 | 0.4 (vs. SCE) | 7 | 0.1 PBS | [43] |
| Pt/NGr | SWV | 0.05–90 | 8 | 0.35 (vs. Ag/AgCl) | 7 | 0.1 PBS | [44] |
| CoPc-flav-f-MWCNTs | SWV | 1–1000 | 1000 | 0.66 (vs. Ag/AgCl) | 7.4 | 0.1 PBS | [45] |
| TC8A/AuNPs/MWCNTs | DPV | 1-150 | 200 | 0.362 (vs. SCE) | 7 | 0.1 PBS | [46] |
| MIP-MWCNTs | DPV | 0.1–2500 | 20000 | 0.385 (vs. SCE) | 7 | 0.1 PBS | [47] |
| PIL-MCNs/CS | DPV | 1-300 | 164 | 0.509 (vs. Ag/AgCl) | 5 | 0.1 PBS | [48] |
| GrNF | DPV | 0.001-150 | 0.43 | 0.19 (vs. Ag/AgCl) | 7 | 0.1 PBS | [49] |
| GI | DPV | 10-500 | 2700 | 0.27 (vs. Ag/AgCl) | 7 | 0.1 PBS | [50] |
| MIP/GO@COF/GCE | DPV | 0.05–20 | 32 | 0.36 (vs. SCE) | 7 | 0.2 PBS | [51] |
| CNT/ILC/RGO/CW | DPV | 0.001-20 | 0.0906 | 403 (vs. Ag/AgCl) | 7.4 | 0.1 PBS | [52] |
| MWCNT-βCD | DPV | 0.05-300 | 11.5 | 0.34 (vs. Ag/AgCl) | 7.4 | 0.01 PBS | [53] |
| NCDs | DPV | 0.5-600 | 157 | 0.34 (vs. Ag/AgCl) | 7 | 0.1 PBS | [54] |
| DNPH | EIS | 0.1-1000 | 250 | - | 9 | $10^{-2}$ HCl | [55] |
| activated GCE | DPV | 0.25-2.5 | - | 0.485 (vs. Ag/AgCl) | 7 | 0.05 PBS | [56] |
| activated GCE | DPV | 5.5-33 μg/L | 1.8μg/L | 0.28 (vs. Ag/AgCl) | 7 | 0.05 BPB | [57] |
| ZKAKC | DPV | 0.01-20 | 4 | 0.405 (vs. Ag/AgCl) | 7.4 | 0.1 PBS | [58] |
| P-NC | DPV | 3–110 | 500 | 0.35 (vs. SCE) | 7 | 0.1 PBS | [59] |

Here the application of a range of electrochemical sensors to the detection of PCM alone and in mixture for the period from 2018 to 2020 is compared to provide an update to the published articles. As shown in Figure 1, we have classified the various electrodes modified into many sections considering the number of publications.

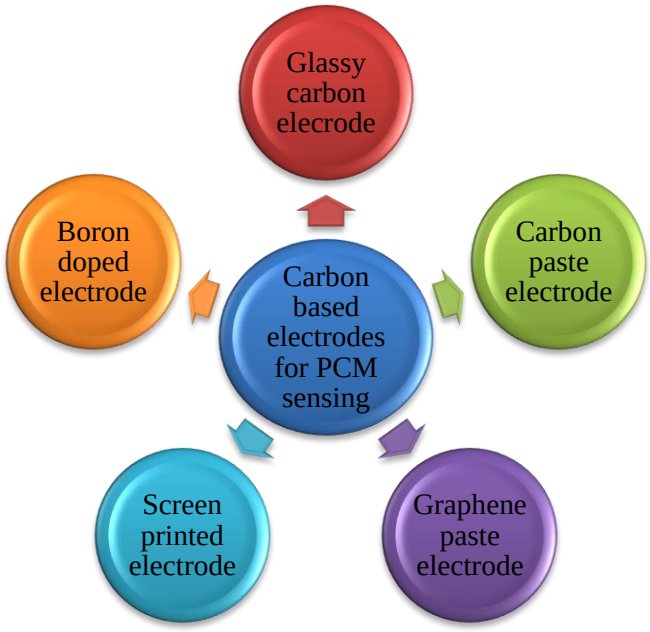


**Fig. 1.** Classified of different carbon-based electrodes for PCM sensing

### 2. 1. Glassy Carbon Electrode

Glassy carbon is widely used as an electrode material in electrochemistry, owing to its physical and chemical properties. The most important properties are high temperature resistance, low density, electrical and thermal resistance. The use of chemically modified glassy carbon electrode (GCE) can help in selective, sensitive, and reproducible detection of PCM or in the presence of other interferences by reduction of ohmic resistance associated. Here we give a brief description of materials such as carbon-based materials and metal nanoparticles, which are commonly used for electrode modification.

Kumar et al reported a sensitive electrochemical detection and efficient photocatalytic degradation of PCM using titanium dioxide-molybdenum disulfide nanocomposite ($MoS_2/TiO_2$ NC). The synthesized nanocomposite showed enhanced photocatalytic activity against PCM and the sensing platform revealed a linear dynamic range from 0.5 to 750 μmol/L with limited detection was 10 nmol/L [30].

Moreover, enormous studies have been proposed the modification of GCE with carbon-based materials, especially CNT and GO combined with metal nanoparticles such as Au, Pt, Pd, Fe and Co [31-46]. The GCE modified exhibited an increase of mass transport, conductivity, rate of electron transfer, electrocatalytic activity and provide a large specific

surface area. Among these, the new nanocomposite based on GO, $Fe_3O_4$, poly-methyldopa and palladium (GO/$Fe_3O_4$@PMDA/Pd) as reported by Lotfi and Veisi. Modification has been performed simply in a simple way by casting of GO/$Fe_3O_4$@PMDA/Pd nanocomposite on the GCE surface. The fabricated sensor exhibited high sensitivity with low detection limits of 2.1 nmol/L in the range of 0.005-2.5 µmol/L [38].

In this approach, some researchers have been examined a novel electrochemical sensor based on the modification of GCE with layered carbon-based materials for sensitive detection of PCM in different matrices [47-54]. The study was employed a novel layered composite based on layer-by-layer modification of a glassy carbon electrode surface with multi-walled carbon nanotubes (CNT), ionic liquid crystal (ILC), graphene (RGO) and 18-Crown-6 (CW) (GCE/CNT/ILC/RGO/CW) showed excellent performance for PCM detection in the presence of dobutamine (DB), amlodipine (AM) and ascorbic acid (AA). This sensor showed a linear range of 0.01-20 µmol/L which is better than many existing sensors and LOD was further improved to 0.0906 nmol/L [52].

In addition, other investigations have been preferred to use GCE as activated, modified with the phenolic compounds and with nitrogen doped carbon dots (NCDs) [55-59]. The modified electrode as noted $ZnCl_2$-KOH activated kelp carbon (ZKAKC/GCE) exhibited the best LD of 4 nmol/L and the range of 0.01-20 µmol/L [58].

Generally, table 1, summarized the LD and range detection for each sensor and the parameter can affect the response of PCM.

### 2.2. Carbon Paste Electrode

The carbon paste electrode (CPE) was first introduced as an electrode in the field of electrochemistry, owing to their properties such as simple preparation, easy renewable surface and economic. Recently, various new materials have been introduced as a modifier for the preparation of conventional CPE such as metal nanoparticles and carbon-based materials to improve the electrochemical performance of the sensor electrodes. We have categorized the various electrodes modified CPE to the detection of PCM into three sections.

Last two years, several studies have been reported the modification of CPE with metal nanoparticles such as Co, Mn, Cu and titanium dioxide ($TiO_2$) for sensitive detection of PCM [60-66]. Azab fabricated a novel sensor by the electrodeposition of cobalt nanoparticles (Nano Co) on the surface of carbon paste electrode (CPE) modified with starch (S) (CPE/S//NanoCo) polymers for the nanomolar detection of paracetamol in presence of warfarin (WA) and caffeine. The fabricated sensor exhibited high sensitivity with low detection limits of 0.99 nmol/L and the linear range of 0.02-150 µmol/L [61].

Furthermore, some work elaborated an electrochemical sensor for sensitive detection of PAM based on the modification of CPE with carbon-based materials such as CNT and graphene oxide (GO) coupled with metal nanoparticles [67-69]. Patil et al proposed a specific,

sensitive and a simple electrochemical sensor MWCNT-ZnO/CPE based on multi-walled nanotubes (MWCNTs) and a graphite electrode, to which ZnO nanoparticles were added to investigate the electrode determination toward paracetamol. It was found that the MWCNT-ZnO/CPE exhibited lower detection limit of 3.32 nmol/L and the linearity observed in the range of 0.01-0.3 µmol/L at the fabricated sensor [68].

Further, other investigations have been used material as a modifier on the CPE [70-73]. The sensor, as named MMIP/MCPE exhibited high electroanalytical detection toward PAM and revealed a linear range from 0.06 to 200 µmol/L with limited detection was 17.3 nmol/L [73].

The electrochemical sensors with detection limits for PCM and their linear ranges are included in table 2.

**Table 2.** Analytical response characteristics of PCM sensor based on chemically modified carbon paste electrode

| CPE modified by | Method | Linearity (µmol/L) | LOD (nmol/L) | Potential E(V) | pH | Medium (mol/L) | Ref. |
|---|---|---|---|---|---|---|---|
| • np-$CoFe_2O_4$<br>• np-$MnFe_2O_4$ | DPV | • 3–200<br>• 3–160 | • 250<br>• 300 | • 0.602<br>• 0.567 (vs. Ag/AgCl) | 6 | 0.1 PBS | [60] |
| S//NanoCo | DPV | 0.02-150 | 0.99 | 0.4 (vs. SCE) | 2 | 0.1 BR | [61] |
| CuO NPs | CV/ i–t | $10^3$–5x$10^3$ | - | −0.2 (vs. Ag/AgCl) | - | 6 M KOH | [62] |
| ZSM-5/$TiO_2$ | DPV | 2.5-110 | 580 | 0.59 (vs. Ag/AgCl) | 5 | 0.1 PBS | [63] |
| $BF_3$ @MCM 41/DHB | DPV | 1.0 - 101.5 | 330 µM | 0.341 (vs. Ag/AgCl) | 7 | 0.1 PBS | [64] |
| ZIF8@Co-TA | DPV | 0.02–0.44 | 5.1 | 0.46 (vs. SCE) | 6 | 0.1 PBS | [65] |
| IO | DPV | 2-150 | 1160 | 0.458 V (*vs.* silver/silver chloride) | 7 | 0.1 PBS | [66] |
| $Nd_{OX}$-SWCNT | SWV | 0.10-9.5 | 50 | 0.57 V (vs. Ag/AgCl) | 3.2 | 0.01 PBS | [67] |
| MWCNT-ZnO | DPV | 0.01-0.3 | 3.32 | 0.613 (vs. Ag/AgCl) | 5 | 0.2 PBS | [68] |
| GO-Y | DPV | 7-400 | 1450 | 0.55 (vs. Ag/AgCl) | 7 | 0.1 PBS | [69] |
| WPE | DPV | 0.50-100 | 53.6 | 0.279 (vs. silver ink) | 6 | 0.1 PBS | [70] |
| NC | DPV | 0.2-1.3 | 3710 | 0.561 (vs. Ag/AgCl) | 5 | 0.2 PBS | [71] |
| Sl | CV | 1–160 | 21 | 0.561 (vs. Ag/AgCl) | 5 | 0.2 PBS | [72] |
| MMIP | DPV | 0.06-200 | 17.3 | 0.44 (vs. SCE) | 6.5 | 0.2 PBS | [73] |

### 2.3. Screen-Printed, Boron Diamond and Graphene Paste Electrodes

Graphene As shown in table 3, we compare the linear ranges and detection limits of each electrode.

Screen-printed electrodes (SPEs) have been applied in biosensor applications, due to their advantages over the traditional electrodes such as low in cost and easily disposable. Despite the large attention, a little of studies have been cited the use of SPEs as modified toward PAM detection [74-77]. Zhang et al developed a facile and sensitive sensor based on MXene modified screen-printed electrode (MXene/SPE) for the detection of PAM and isoniazid (INZ). The sensor showed excellent electrocatalytic activity toward the detection of PAM with wide linear range from 0.25 to 2000 μmol/L and low detection limits of 48 nmol/L [76].

**Table 3.** Analytical response characteristics of PCM sensor based on chemically modified screen-printed, boron diamond and graphene paste electrodes

| **Sensors** | **Method** | **Linearity (μmol/L)** | **LOD (nmol/L)** | **Potential E (V)** | **pH** | **Medium (mol/L)** | **Ref.** |
|---|---|---|---|---|---|---|---|
| GNPs-Naf/SPE | DPV | 0.25–30 | 250 | 0.374 (vs. Ag/AgCl) | 7.4 | 0.1 PBS | [74] |
| Carbon screen-printed electrodes (SPEs) | DPV | 0.5 - 10 | 218 | 0.205 (vs. Ag/AgCl) | 7 | 0.1 PBS | [75] |
| MXene/SPE | DPV | 0.25-2000 | 48 | 0.5 (vs. Ag/AgCl) | 1 | 0.1 M $H_2SO_4$ | [76] |
| SPCE/CB-ERGO | SWV | 9.9–95 | 5300 | 0.286 (vs. Ag/AgCl) | 7 | 0.1 PBS | [77] |
| CeBiO*X* NFs modified SPE | DPV | 130 -500 | 200 | 0.5 (vs. Ag/AgCl) | 7.4 | 0.1 PBS | [78] |
| BDDE | DPV | 0.1 - 200 | 13.5 | 0.4 V (vs. silver/silver) | 8.3 | 0.1 ammonium buffer | [79] |
| CP–BDD | • CV<br>• FIA–MPA | 30.08-100 | 30 | • 0.75<br>• 0.95 (vs. Ag/AgCl) | 4 | • 0.2 PBS<br>• 0.05 $H_2SO4$ | [80] |
| • BDD<br>• B:CNW | DPV | • 0.065-32<br>• 0.032-32 | • 430<br>• 281 | 0.44 (vs. Ag/AgCl) | 7 | BR | [81] |
| poly-L-Asp/GPE | Amperometry | 0.05-108.25 | 11 | 0.464 (vs. Ag/AgCl) | 7 | 0.1 PBS | [82] |

Boron-doped diamond (BDD) is an electrode material with the excellent properties, owing to their stability, high chemical resistance, and good repeatability of response as presented all carbon-based electrodes. Recently, a few reports developed the electrochemical sensor based BDD for the PAM detection. Last two years, only three works were cited the use of BDD to

detection of PAM [78-80]. Among of these, an unmodified boron doped diamond electrode (BDDE) was applied for the first time toward dopamine (DA) and PCM detection. This electrode showed a linear range of 0.1 to 200 µmol $L^{-1}$ and very low detection limits were achieved, equaling 13.5 nmol/L [79].

The graphene paste electrode (GPE) has been widely applied in electrochemical applications due to high electron transport rate and large surface area. It has become an interesting alternative for the electrochemical sensors. However, a very little works was employed GPE in the detection of PCM. Luo et al created a novel poly-L-Asp modified GPE for the electrochemical determination of acetaminophen (AC) and revealed a linear range from 0.05 to 108.25 µmol/L with limits of detection was 11 nmol/L [82].

## 3. A COMPARATIVE OVERVIEW OF CHEMICALLY MODIFIED CARBON-BASED ELECTRODES AND FUTURE PERSPECTIVES

Chemically modified carbon-based electrodes provided excellent linear working range and significantly low LOD. As we cited above, they are many works proposed by the researchers on the use of carbon-based materials and metal nanoparticles as a modifier toward of the fabrication of sensors and amelioration of linear range and limit of detection in terms of PAM detection.

Hence, we give a comparison of the best sensor for GCE, CPE, SPE, BDD and GPE as shown in figure 2.

GCE present numerous publications nearly 31 (56%), from 2018 to 2020 (June). In many instances, CNT, GC, and metal oxide nanoparticles are present on the surface of GCE as modifier that enhance selectivity toward PCM, each electrode provides a very good linear range and LOD. In the case of both electrodes such as GC/CNT/ILC/RGO/CW/GCE and GO/$Fe_3O_4$@PMDA/Pd/GCE, a linear range of 0.01-20 µmol/L and of 0.05-2.5 µmol/L respectively, as well as LOD of 0.0906 nmol/L and 2.1 nmol/L were obtained .

Furthermore, CPE present an important number of publications 14 (28%) in the same period. As well as the modification of electrode by CNT and metal oxide nanoparticles that enhance selectivity toward PCM. For example, MWCNT combined with ZnO modified CPE (MWCNT-ZnO/CPE) provided an excellent linear working range 0.02-150 µmol/L and significantly low LOD of 0.99 nmol/L. In addition, CPE/S//NanoCo provided a very good working range 0.01-0.3 µmol/L and significantly low LOD of 3.32 nmol/L .

Another category of carbon-based electrodes as SPE, BDD and GPE showed a good sensitivity for the detection of PCM. The electrode as MXene/SPE, BDDE and poly-L-Asp/GPE exhibited a good linearity from 0.25 to 2000 µmol/L, 0.1 to 200 µmol/L and 0.05 to 108.25 µmol/L respectively. As well as a low LOD of 48 nmol/L, 13.5 nmol/L and 11 nmol/L were obtained.

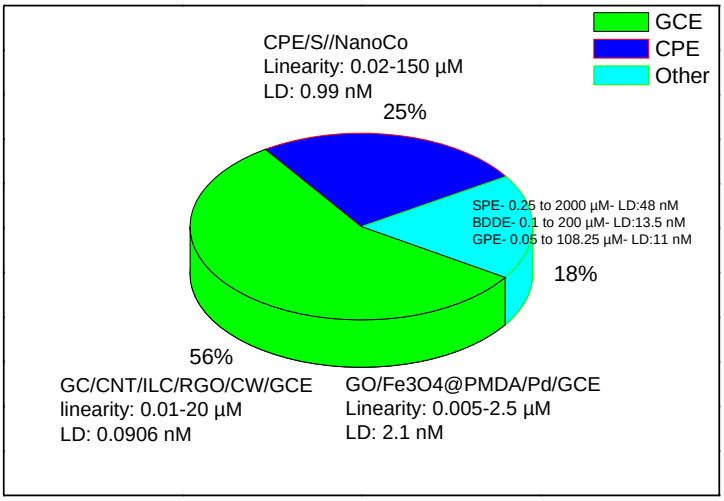


**Fig. 2.** Comparison of the best sensor for GCE, CPE, SPE, BDD and GPE

There is an increasing current in the application of carbon-based electrodes, especially GCE due to the important benefits that are produced forward by the hybridization of polymer materials and nanoparticles. The use of different types of nanoparticles in combination with the CNT, GC, SPE and BDD in the fabrication of electrochemical sensors conduct to high sensitivity. However, the enormous interest and the extensive study achieved in the development of chemically modified carbon-based electrodes such as CNT SPE, BDD and GPE for the detection of PCM are insufficient. Thus, more reproducible results of these electrodes are highly challenging. On the other hand, to develop the sensors for PAM detection, researchers were testing the performance of these sensors in the various samples like commercial tablets and biological fluids, but only four works have cited PAM detection in the water analysis such as river water [43], surface waters [56,57], and tap water [75]. Then, the growing production and the enormous consummation of PAM for human health, it's not controlled after usage, and can be caused the troubles of the aquatic environment. Over there, the enzymes produced by microorganisms existing in the aquatic environment play an essential role in the degradation and transformation of the paracetamol to nontoxic compounds but are insufficient to remove this substance completely. The development of sufficiently sensitive and reproducible analytical methods for precise determination of PCM is essential for the further protection of water quality. Thus, another challenge take place is the application of electrochemical methods for detection of PCM in wastewater.

## 4. CONCLUSION

In this brief review, we have summarized the recent production of the publications put into the development of PCM detection using carbon-based electrodes. As shown in the review,

there has been a promising regarding the development of chemically modified carbon-based electrodes in combination with nanoparticles and carbon-based materials for the detection of PCM. It is clearly that the application of carbon-based electrodes, especially GCE exhibit a high selectivity toward PCM detection. On the other hand, we bring out that, other carbon-based electrodes such as CNT, GPE, SPE and BDD electrodes provide many potentials, but is not sufficiently developed toward detection of PAM .

Also, the carbon-based electrodes used as sensors and their application in water analysis are not sufficiently developed compared with in commercial tablets and biological fluids. The development of innovative methods is essential to ensure the complete elimination of paracetamol for the further protection of water quality. For these reasons, another challenge take place is the application of electrochemical methods for detection of PCM in wastewater.

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