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Original scientific paper

Ultrasensitive and selective electrochemical sensor for paracetamol based on thiacloprid nanocomposite

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Abstract

The study introduces a novel electrochemical sensor for paracetamol (PCM) determination based on a nanocomposite composed of zinc layered hydroxide (ZLH) intercalated with sodium dodecyl sulphate and thiacloprid (SDS-THI), integrated with multiwalled carbon nanotubes (MWCNTs). The sensor aims to address limitations of conventional analytical techniques and improve sensitivity, detection limits, and portability. Electrochemical techniques, including electrochemical impedance spectroscopy, square wave voltammetry and cyclic voltammetry, were employed to characterize the sensor performance. The ZLH-SDS-THI/MWCNTs sensor showed superior electrocatalytic activity, with a wide linear range (0.7 to 30 μ M) and a low detection limit (LOD = 0.33 μ M), outperforming several previously reported sensors. The enhanced performance is attributed to the synergistic properties of the composite materials, which offer improved electron transfer, a high surface area, and effective analyte interaction. Importantly, the sensor demonstrated excellent selectivity, as interference studies revealed that common biological and ionic species such as ascorbic acid, glucose, fructose, lysine, chloride, magnesium, and sulphate ions, even when present at 10-, 20-, and 50-fold excess concentrations relative to PCM, caused less than 10 % signal variation. This confirms the sensor's robustness and reliability in complex sample matrices. Overall, this work highlights the potential of incorporating unconventional organic dopants, such as thiacloprid, into layered nanostructures to enhance the performance of electrochemical sensors, offering a promising platform for the selective and sensitive determination of pharmaceutical compounds in environmental and clinical applications.

Keywords

Acetaminophen drug; zinc layered hydroxide; organic dopants; multiwalled carbon nanotubes; voltammetry techniques

Introduction

The increasing presence of pharmaceutical compounds such as paracetamol (PCM) in clinical and environmental samples has raised significant concerns due to potential toxicity, especially in cases of overdose or prolonged exposure. As one of the most commonly used antipyretic and analysis drugs, PCM is frequently monitored in biological fluids and wastewater streams [1,2]. Traditional analytical techniques such as mass spectrometry, spectrophotometry and high-performance liquid chromatography (HPLC) offer excellent sensitivity and specificity but are often limited by their cost, time-consuming sample preparation, and lack of portability for on-site analysis. These limitations have spurred the development of electrochemical sensors, which offer distinct advantages, including high sensitivity, low detection limits, fast response times, and potential for miniaturization [3,4].

A broad spectrum of electrochemical sensors has been developed for the determination of PCM using various nanomaterials and modified electrodes. For instance, glassy carbon electrodes modified with TiO₂-graphene and Nafion exhibited a working range of 1 to 100 μ M and a LOD of 0.21 μ M [5]. Similarly, a carbon paste electrode modified with cork displayed a linear range of 0.025-1.000 mM and an LOD of 2.41 μ M [6]. Other works using MWCNT composites, such as the one by Burc *et al.* [7], reported a linear range of 2.0 to 50 μ M and an LOD of 2.0 μ M. Despite these advancements, many of these sensors still show limitations in terms of sensitivity at low concentrations or have relatively narrow linear ranges, limiting their effectiveness in detecting trace levels of PCM in real samples.

This article presents a novel electrochemical sensor for PCM determination based on a hybrid nanocomposite consisting of zinc layered hydroxide intercalated with sodium dodecyl sulphate and thiacloprid (ZLH-SDS-THI), combined with modified multiwalled carbon nanotubes (MWCNTs). This innovative integration of ZLH, a class of anionic clays with excellent ion-exchange capabilities, with the conductive and high surface area properties of MWCNTs is designed to improve the electrochemical performance of the sensor synergistically. Notably, the incorporation of thiacloprid, which is an insecticide with a unique nitrogen-containing structure, will introduce an unconventional organic dopant [8-10].

The primary objective of this study is to address current knowledge gaps in the functional integration of organic insecticides within layered hydroxide frameworks for electrochemical sensing. Specifically, this work focuses on (i) the synthesis and structural characterization of ZLH-SDS-THI/MWCNTs nanocomposite materials, (ii) the evaluation of their electrochemical behaviour toward PCM detection via voltammetry and impedance spectroscopy, and (iii) benchmarking their analytical performance against previously reported sensors.

Our sensor demonstrates a broader linear detection range (0.70 to 30 μ M) and a lower LOD (0.33 μ M), positioning it competitively among existing platforms. In contrast, several previous designs fall short of these values, such as a modified carbon paste electrode with a molecularly imprinted polymer with a linear range of 10 to 100 μ M (LOD = 4.63 μ M) [11], and a ZrO₂ NPs modified carbon paste electrode with a range of 10 to 60 μ M (LOD = 068 μ M) [12]. These comparisons highlight the significance of our approach in advancing sensitivity and range in PCM detection.

Ultimately, this research contributes to the evolving landscape of nanomaterial-enabled electrochemical sensors and encourages further exploration of multifunctional composite materials for environmental and biomedical monitoring. It also provides a foundation for rethinking the role of unconventional dopants, such as thiacloprid, in tuning the electrochemical behaviour of sensing platforms.

The novelty of this study lies in the innovative incorporation of thiacloprid, a nitrogen-containing insecticide, as an unconventional organic dopant within the zinc layered hydroxide framework. To the best of our knowledge, this is the first report utilizing thiacloprid-doped layered hydroxide (ZLH-SDS-THI) integrated with multiwalled carbon nanotubes for electrochemical sensing. This unique design strategy offers a novel approach to utilizing organic pesticide molecules as electroactive mediators in nanocomposite electrodes, resulting in superior sensitivity and selectivity for paracetamol detection compared to conventional materials.

Experimental

Reagents and chemicals

Paracetamol (PCM) and potassium chloride (KCl) were purchased from Sigma-Aldrich, USA. Generic paracetamol tablets (500 mg) were procured from a local pharmacy in Tanjong Malim, Perak. Different pH values of phosphate buffer solution (PB) were prepared by mixing stock solutions of K_2HPO_4 (0.1 M) and KH_2PO_4 (0.1 M) (Merck, Germany). Stock solutions of PCM (0.10 mM) were freshly prepared by dissolving PCM in deionized water and stored at 4 °C in a dark place until use. Multiwalled carbon nanotubes (MWCNTs) were obtained from Timesnano, China. The ZLH-SDS-THI nanocomposite was synthesized in the laboratory as previously reported [8]. All reagents used in this experiment were of analytical grade and used without further purification.

Instrumentation

Cyclic voltammetry (CV) and square wave voltammetry (SWV) were conducted using a Potentiostat Series-G750 (USA), while electrochemical impedance spectroscopy (EIS) was performed using a potentiostat/galvanostat model of Ref 3000 (USA). All electrochemical measurements were carried out in a conventional three-electrode cell consisting of a ZLH-SDS-THI/MWCNTs paste electrode as the working electrode, an Ag/AgCl reference electrode (MF-2052 model, Bioanalytical Systems, USA) and a platinum wire as the counter (auxiliary) electrode.

Fabrication of the ZLH-SDS-THI/MWCNTs paste electrode

To fabricate the ZLH-SDS-THI/MWCNTs paste electrode, 0.01 g of ZLH-SDS-THI, 0.09 g of MWCNTs and a suitable amount of paraffin oil were thoroughly mixed using a mortar and pestle until a homogeneous paste was formed. The resulting uniform paste was then packed into a Teflon tube (length: 3 cm and diameter: 2 mm), and a copper wire was inserted at one end of the tube to establish electrical contact. The electrode surface was polished with weighing paper immediately before use. For comparison of electrochemical activity, a bare MWCNT paste electrode was prepared using the same procedure, except without adding the ZLH-SDS-THI modifier.

Measurement procedure

EIS was conducted in a 4 mM K_3 [Fe(CN)₆ solution containing 0.1 M KCl as the supporting electrolyte. For EIS measurements, an alternating voltage amplitude of 5.0 mV and a frequency range of 1.0 MHz to 1.0 Hz were used. The PCM determination was performed using SWV in 0.1 M PB as the supporting electrolyte. The SWV conditions were as follows: frequency of 120 Hz, step increment of 3 mV, pulse height of 60 mV and potential range of 0.0 to 0.6 V. All electrochemical measurements were carried

out at 25±1 °C and recorded against an Ag/AgCl reference electrode. For real sample analysis, a PCM tablet was accurately weighed, ground into a fine powder, and dissolved in deionized water. The solution was then diluted to volume in a 50 mL volumetric flask prior to analysis.

Results and discussion

Characterization and surface morphology analysis of ZLH-SDS-THI nanocomposite

Transmission electron microscopy (TEM) was used to examine the morphology of ZLH-SDS-THI and ZLH-SDS-THI/MWCNT. Figure 1A indicates an irregular particle size with a plate-like characteristic of ZLH-SDS-THI. The transparent, delicate tube represents MWCNT, which was covered with opaque features of ZLH-SDS-THI (Figure 1B).

Energy-dispersive X-ray (EDX) analysis confirms the successful formation of ZLH-SDS-THI nanocomposite. The EDX spectrum revealed the presence of Zn, C, N, O, S, and Cl in the nanocomposite. The presence of Zn element confirms the formation of host zinc layered hydroxide. Meanwhile, the C and S peaks corresponded to the long-chain alkyl and sulphate groups of the intercalated SDS anions. The incorporation of thiacloprid onto ZLH is confirmed through the appearance of distinct N and Cl peaks (Figure 1C). EDX elemental mapping further demonstrated that Zn was concentrated in the nanocomposite, whereas C, N, S, and Cl were more uniformly distributed, corresponding to the intercalation of SDS and thiacloprid molecules (Figure 1D).

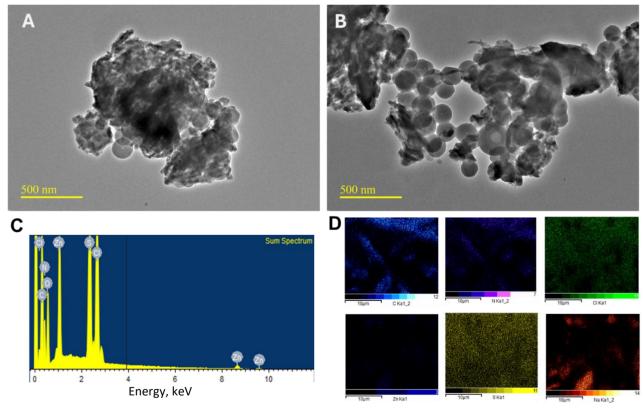


Figure 1. TEM images of (A) ZLH-SDS-THI, (B) ZLH-SDS-THI/MWCNTs, (C) EDX spectrum and (D) elemental mapping of ZLH-SDS-THI nanocomposite

Electrochemical performance of the ZLH-SDS-THI/MWCNTs

CV was carried out to investigate the electrochemical behaviour and electron transfer kinetics of the electrodes using 4.0 mM $K_3[Fe(CN)_6]$ in 0.1 M KCl as a redox probe at a scan rate of 100 mV s⁻¹, as shown in Figure 2. The bare MWCNTs paste electrode (a) exhibited anodic (I_{pa}) and cathodic (I_{pc}) peak currents of 26.98 and 28.17 μ A, respectively, with a peak-to-peak separation (ΔE_p) of 464 mV. In

contrast, the ZLH-SDS-THI/MWCNTs modified electrode (b) showed enhanced redox activity, with I_{pa} and I_{pc} increasing to 34.99 and 36.58 μ A, respectively, and a slightly reduced ΔE_p of 446 mV.

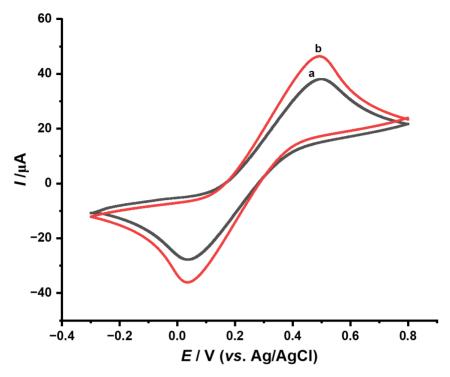


Figure 2. Cyclic voltammograms of 4.0 mM $K_3[Fe(CN)_6]$ in 0.1 M KCl at (a) bare MWCNTs and (b) ZLH-SDS-THI/MWCNTs modified electrodes; scan rate of 100 mV s⁻¹

The increase in peak currents at the modified electrode indicates an improvement in the electroactive surface area and conductivity due to the synergistic effect of the ZLH-SDS-THI nanocomposite and MWCNTs. The decreased ΔE_p compared to the bare electrode suggests a somewhat faster rate of electron transfer at the modified surface, although the separation is still relatively large, indicating quasi-reversible redox behaviour [13]. The improved response is attributed to the layered structure of ZLH, which may facilitate ion exchange and promote redox reactions, while the presence of SDS and thiacloprid may improve dispersion and electron transport [14,15]. These results confirm that the ZLH-SDS-THI/MWCNTs composite provides a more favourable interface for electron transfer processes and is suitable for further electrochemical applications, including the sensing of PCM.

EIS was employed to investigate the interfacial charge transfer properties of the electrodes using $4.0 \text{ mM K}_3[\text{Fe}(\text{CN})_6]$ in 0.1 M KCl as a redox probe. The charge transfer resistance (R_{ct}), derived from the semicircle diameter in the Nyquist plot, provides insight into the electron transfer kinetics at the electrode surface. Figure 3 shows that for the bare electrode, a relatively high R_{ct} value was recorded, indicating considerable resistance to electron transfer and suggesting a less conductive or electrochemically inactive surface. In contrast, the ZLH-SDS-THI-modified electrode exhibited about twice smaller R_{ct} than bare electrode, demonstrating enhanced charge transfer capabilities. This pronounced decrease in R_{ct} can be attributed to the synergistic effect of the ZLH, SDS and THI, which likely improved the surface conductivity and facilitated electron transport. The modification not only increased the electroactive surface area but also introduced more accessible active sites for redox interaction [16,17]. Overall, the EIS results confirm that the ZLH-SDS-THI/MWCNTs-modified electrode provides a more efficient electron-transfer pathway, making it a promising platform for electrochemical sensing applications.

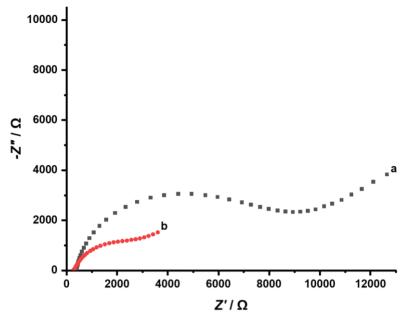


Figure 3. Nyquist plots of 4.0 mM K_3 [Fe(CN)₆ in 0.1 M KCl at (a) bare MWCNTs and (b) ZLH-SDS-THII/MWCNTs

The SWV data for the determination of 1.0 μ M PCM in 0.1 M PB (pH 6.5) reveals significant differences in electrochemical response between the bare and modified electrodes as shown in Figure 4.

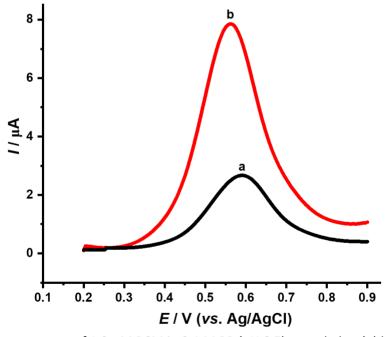


Figure 4. SW voltammograms of 1.0 μ M PCM in 0.1 M PB (pH 6.5) recorded at (a) bare MWCNTs and (b) ZLH-SDS-THI/MWCNTs

At this concentration, the bare electrode exhibited a relatively low oxidation peak current of 2.36 μ A, indicating limited sensitivity and poor electrocatalytic activity toward PCM oxidation. In contrast, the ZLH-SDS-THI/MWCNTs-modified electrode exhibited a much higher current response of 7.31 μ A under the same conditions. This enhancement in peak current is over three times greater than that of the bare electrode, demonstrating the superior electrocatalytic performance of the modified electrode. The incorporation of ZLH provides a high surface area and layered structure that can facilitate electron transfer. SDS, as a surfactant, helps to disperse active materials uniformly and

enhances the adsorption of PCM molecules. THI, an electroactive dye, acts as an electron mediator, promoting redox reactions. MWCNTs contribute excellent electrical conductivity and further increase the electroactive surface area [18,19]. Together, these components create a highly conductive, electrocatalytically active interface that enhances electron-transfer kinetics and improves analyte accessibility. The significant increase in peak current at the modified electrode suggests enhanced sensitivity for paracetamol determination, confirming the potential of the ZLH-SDS-THI/MWCNTs system as an effective electrochemical sensing platform.

Optimization of the parameters for voltammetric analysis

Effect of SWV parameters

The optimization of SWV parameters for the determination of 1.0 μ M PCM was carried out by evaluating the effects of frequency, pulse size and step size on the peak current response (Figure 5). As shown in Figure 5A, for frequency optimization, the peak current increased from 4.95 μ A at 30 Hz to a maximum of 7.35 μ A at 180 Hz, followed by a slight decline to 7.11 μ A at 210 Hz, which is likely due to insufficient time for electron transfer and increased capacitive current at excessively high frequencies. In step-size optimization, the current rose from 4.72 μ A at 1 mV to a peak of 7.37 μ A at 4 mV, then slightly decreased to 6.89 μ A at 5 mV, which may be attributed to signal broadening or overlapping redox processes that reduce resolution (Figure 5B).

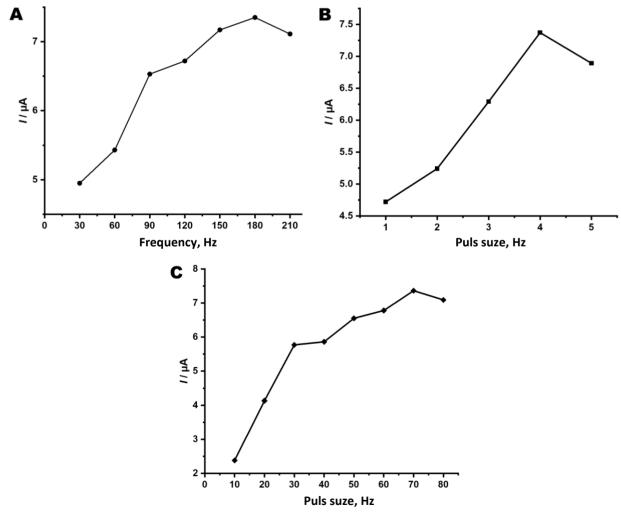


Figure 5. Effects of the SWV parameters (A) frequency, (B) step size and (C) pulse size on current response in solution of 1.0 μ M PCM in 0.1 M PB at pH 6.5

As shown in Figure 5C, the current increased steadily from $2.38~\mu A$ at 10~mV to $7.36~\mu A$ at 70~mV, but dropped slightly to $7.09~\mu A$ at 80~mV, likely due to contributions from non-faradaic (capacitive) currents or distortion of the voltammetric signal. These findings indicate that the optimal parameters, *i.e.* 180~Hz of frequency, 4~mV of step size and 70~mV of pulse size, will enhance the faradaic response while minimizing background interference, thereby improving the sensitivity and reliability of PCM detection [20].

Effect of scan rate

The CV analysis of 4.0 mM K₃[Fe(CN)₆] in 0.1 M KCl was conducted at various scan rates (10-300 mV s⁻¹) using a ZLH-SDS-THI/MWCNTs modified electrode to evaluate the redox behaviour and electrode kinetics. As seen in Figure 6A, the peak current increased progressively with increasing the scan rate, and a linear relationship was observed between the anodic peak current (I_{pa}) and the square root of scan rate ($v^{1/2}$), indicating a diffusion-controlled electron transfer process. The linear regression for I_{pa} vs. $v^{1/2}$ yielded the equation I_{pa} = 1.6328 $v^{1/2}$ + 17.922 with a high correlation coefficient R^2 = 0.9916, confirming excellent linearity as shown in Figure 6B. This strong correlation suggests that the current response is governed primarily by the diffusion of [Fe(CN)₆]^{3-/4-} species to the electrode surface [21]. Additionally, the peak-to-peak separation (ΔE_p) increased slightly with scan rate, indicating quasi-reversible behaviour, which is typical for systems modified with redoxactive mediators like THI. The THI film likely enhances charge transfer kinetics by facilitating electron mediation between the redox species and the electrode surface. Overall, the results confirm that the ZLH-SDS-THI/MWCNTs modified electrode promotes a diffusion-controlled, quasi-reversible redox process with high electrochemical performance.

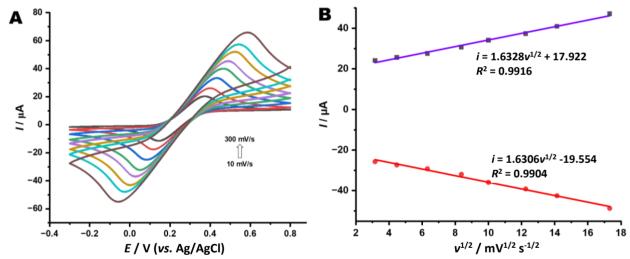


Figure 6. (A) Cyclic voltammograms of ZLH-SDS-THI/MWCNTs with 4.0 mM K_3 [Fe(CN)₆] in 0.1 M KCl at different scan rates (10 to 300 mV s^{-1} and (B) plots of peak currents vs. square root of scan rate

Effect of pH

The SWV investigates the influence of pH on the electrochemical behaviour of 1.0 μ M PCM, evaluated across a pH range of 6.0 to 8.0. The peak potential (*E*) and peak oxidation current (*I*) were measured at each pH to determine the optimal conditions for PCM detection. As shown in Figure 7A, the highest peak current was recorded at pH 6.5 (7.32 μ A), followed by a slight decrease at higher pH values (7.08 μ A at pH 7.0, 6.94 μ A at pH 7.5 and 6.86 μ A at pH 8.0), suggesting reduced electrochemical efficiency in more alkaline conditions. These results indicate that the electrochemical response of PCM is pH-dependent, and pH 6.5 offers the most favourable environment for its oxidation, likely due to an optimal balance of proton availability and electrode kinetics at this point.



Additionally, the oxidation peak potential shifts to more negative values with the increasing pH, from 592.1 mV at pH 6.0 to 489.1 mV at pH 8.0. This linear negative shift in peak potential with pH suggests that protons are directly involved in the redox process of PCM. The relationship between the peak potential (E_{pa}) and pH is described by the linear regression equation: E_{pa} = -50.02 pH + 888.86 with a strong correlation coefficient (R^2 = 0.9942), indicating excellent linearity. The slope of -50.02 mV pH⁻¹ is close to the theoretical value of -59 mV pH⁻¹ expected for an equal number of electrons and protons involved in the redox mechanism, further supporting the proton-coupled electron transfer (Figure 7B) [22]. As illustrated in Scheme 1, PCM was electrochemically oxidised to produce N-acetyl-p-quinoneimine (NAPQI), releasing two protons and two electrons. Overall, these results confirm that pH 6.5 is optimal for detecting PCM and that the electrochemical oxidation process is governed by both pH-dependent kinetics and proton availability.

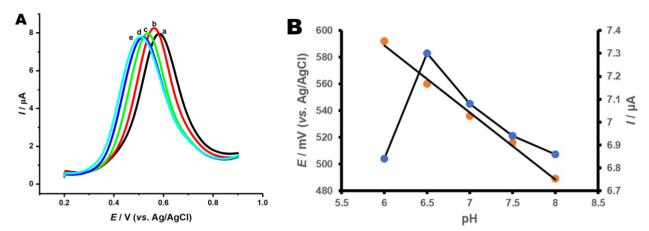


Figure 7. (A) SW voltammograms profile of 1.0 μ M PCM in 0.1 M PB at different pH (a-e: 6.0 to 8.0) recorded using ZLH-SDS-THI/MWCNTs and (B) the plot of peak current and peak potential vs. pH

Scheme 1. Proposed mechanism of the PCM and N-acetyl-p-quinoneimine (NAPQI)

Effect of concentration changes

Figure 8A demonstrates the calibration performance of an electrochemical sensor for determination of PCM, showing a linear response within the concentration range of 0.7 to 30.0 μ M, with a LOD of 0.33 μ M. The current responses recorded at various PCM concentrations show a clear, proportional increase, confirming good sensor linearity. Specifically, the sensor produced currents of 6.71 μ A at 0.7 μ M, 7.32 μ A at 1 μ M, 8.92 μ A at 3 μ M, 18.82 μ A at 7 μ M, 24.42 μ A at 10 μ M and 54.6 μ A at 30 μ M. Based on these values, the calibration curve exhibits a linear regression equation of I = 0.0165 $C_{PCM} + 5.9311$ with a correlation coefficient of $R^2 = 0.9937$, indicating excellent linearity and reliable quantitative performance for PCM detection as seen in Figure 8B. These results are more significant than the previously reported findings, as shown from the comparative data given in Table 1.

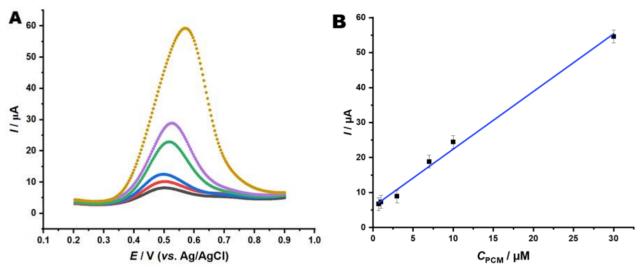


Figure 8. (A) SWV profiles of ZLH-SDS-THI/MWCNTs for different PCM concentrations and (B) corresponding calibration curve of 0.70 to 30.0 μ M PCM in 0.1 M PB (pH 6.5)

Table 1. Comparison of electroanalytical parameters for PCM determination obtained in this study with different electrochemical sensors previously published

Sensor	Method	Range, μM	LOD, μM	Ref.
PMBP/MWCNT/CPE	SWV	1.0 to 1000.0	0.245	[4]
Nafion/TiO₂-graphene-modified GCE	CV	1.0 to 100.0	0.210	[5]
SWCNT/Ni/GCE	CV	50.0 to 500.0	0.117	[23]
GCPE/RGO	SWV	4.0 to 22.0	0.307	[24]
Spectroscopic graphite	CV	5.0 to 150.0	0.200	[25]
SWCNT/PVP-modified PGE	SWV	1.0 to 500.0	0.380	[26]
CSS/CoPc-modified SPCE	PEC	1.50 to 500.0	0.850	[27]
aGCE	DPV	4.00 to 60.0	0.320	[28]
ZLH-SDS-THI/MWCNT	SWV	0.70 to 30.0	0.330	This work

PMBP: 1-phenyl-3-methyl-4-benzoyl-5-pyrazolone; RGO: reduced graphene oxide; PVP: polyvinylpyrrolidone; PGE: pencil graphite electrode; CSS: carbon spherical shells; CoPc: cobalt(II) phthalocyanine; SPCE: screen printed carbon electrode

Interference study

The interference study was conducted using common biological and ionic substances, including ascorbic acid, fructose, glucose, lysine, chloride ions, magnesium ions, and sulphate ions, tested at 10-, 20- and 50-fold excess concentrations relative to PCM (Figure 9).

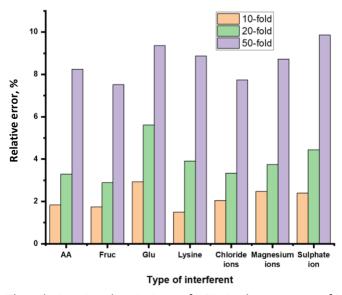


Figure 9. The relative signal variations of PCM in the presence of interferents

The results demonstrated that all tested interferents contributed less than 10 % signal change, indicating no significant effect on the sensor's performance. At the 10-fold level, the current responses were minimal, showing negligible interference. Slight increases were observed at the 20-fold concentration, yet still remained well within acceptable limits. Even at the highest concentration tested (50-fold), the sensor maintained its selectivity, with all interferences causing current responses below the 10 % threshold. These findings confirm the developed electrochemical sensor's strong anti-interference capability and high selectivity for accurate PCM detection in complex biological environments.

Real sample analysis

The applicability of ZLH-SDS-THI/MWCNTs paste electrode was investigated for the PCM determination in pharmaceutical tablets. As shown in Table 2, the recoveries between 98.6 % and 102.1 % indicate that the ZLH-SDS-THI/MWCNTs paste electrode can be implemented efficiently for PCM analysis in real samples.

Table 2. Quantification of PCM samples recorded using ZLH-SDS-THI/MWCNTs paste electrode in pharmaceutical tablets

Content uM

Sample Initial		Content, μM		
	Initial	Added	Found	Recovery (%)
1		5.0	70.2	98.6
2	66.2	10.0	77.8	102.1
3		15.0	80.7	99.4

Reproducibility, repeatability and stability

To evaluate the reproducibility of the ZLH-SDS-THI/MWCNTs paste electrode, the electrochemical response toward 1.0 μ M PCM was measured using five independently fabricated modified electrodes. The relative standard deviation (RSD) obtained was 3.24 %, confirming excellent reproducibility of the proposed electrode. Furthermore, ten successive PCM measurements yielded an RSD of 3.79%, demonstrating good repeatability and indicating that the electrode surface was not fouled by PCM oxidation products. The long-term stability of the ZLH-SDS-THI/MWCNTs paste electrode was also examined after storage at room temperature for 4 weeks, during which it retained approximately 95.3 % of its initial current response, indicating outstanding storage stability and reusability of the developed electrode.

Conclusion

The ZLH-SDS-THI/MWCNTs nanocomposite electrode demonstrated high sensitivity for paracetamol with a wide working range (0.70 to 30.0 μ M) and a low detection limit (0.33 μ M). It showed excellent selectivity against common interferents (<10 % variation) and reliable recoveries in pharmaceutical samples (98.6 to 102.1 %). These results highlight THI-doped layered nanostructures as a promising platform for electrochemical sensing in clinical and environmental applications.

Conflict of interest: The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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