

A Novel High-Performance Non-Enzymatic Electrochemical Glucose Sensor for Aqueous Media Based on MWCNT Modified with Aluminium/Zinc Layered Hydroxide-Ferulate

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A novel non-enzymatic electrochemical glucose sensor based on aluminium/zinc layered hydroxide-ferulate (Al/ZLH-FA)-modified multi-walled carbon nanotubes (MWCNTs) was fabricated as a carbon paste electrode. The composite was morphologically characterized by transmission electron microscopy (TEM), while its electrochemical behaviour was evaluated using cyclic voltammetry (CV), electrochemical impedance spectroscopy (EIS), and square-wave voltammetry (SWV). Under optimized experimental conditions, the sensor exhibited a linear response to glucose concentrations ranging from 3 to 300 $\mu\text{mol L}^{-1}$, with a correlation coefficient (R^2) of 0.9984. The limit of detection (LOD) was calculated to be 0.05 $\mu\text{mol L}^{-1}$. Transmission electron microscopy (TEM) has been used to determine the morphology of Al/ZLH-FA and Al/ZLH-FA/MWCNT, respectively. Because of its remarkable selectivity and sensitivity, the Al/ZLH-FA/MWCNT composite electrode that was produced was useful for analytical applications.

Keywords: Electrochemical sensor, MWCNT, square wave voltammetry, cyclic voltammetry, glucose

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Introduction. The global prevalence of diabetes mellitus is increasing rapidly, with current estimates indicating over 450 million affected individuals and projections suggesting this number will rise to 783 million by 2045 [1, 2]. This trend underscores the urgent need for accurate, sensitive, and user-friendly glucose sensing platforms to manage the disease effectively. Advances in technology, such as continuous glucose monitoring systems, are essential for improving treatment outcomes and reducing complications [3]. The economic and social impact of diabetes is significant, highlighting the importance of comprehensive prevention and control strategies [4, 5]. Minimally invasive and non-invasive technologies such as glucose biosensors are particularly promising, as they enhance patient compliance and provide more convenient monitoring options. The integration of advanced materials and sensing technologies is paving

the way for more effective and user-friendly glucose monitoring systems [6–9]

Traditional enzymatic glucose sensors, though widely used in clinical and personal health monitoring devices, are restricted by numerous limitations. These include enzyme denaturation, temperature and pH sensitivity, short operational life, and complicated immobilization procedures [10]. Non-enzymatic electrochemical glucose sensors (NEEGSs) present a promising alternative to enzymatic glucose sensors, offering enhanced stability, cost-effectiveness, and longer shelf life, supported by advancements in nanotechnology and material science [11–13].

In addition to various strategies for improving NEEGS performance, the incorporation of carbon-based nanomaterials, particularly multi-walled carbon

nanotubes (MWCNTs), has demonstrated great promise. Based on its exceptional electrical conductivity, large specific surface area, and mechanical strength, MWCNTs facilitate rapid electron transfer and provide a robust matrix for anchoring electroactive materials. These characteristics not only enhance the sensor's electrocatalytic activity but also enhance its responsiveness and detection limit [14, 15].

To further enhance the performance of MWCNT-based sensors, researchers have developed their functionalization with metallic or metal hydroxide nanostructures, particularly double hydroxides (LDHs). LDHs composed of aluminium and zinc are particularly beneficial due to their redox behaviour, high surface charge, and environmental compatibility. These materials provide flexibility in interlayer spacing and structural tunability, making them suitable platforms for hosting additional functional molecules. The combination of LDHs with MWCNTs produces a composite that leverages both high conductivity and catalytic reactivity, enabling more effective glucose oxidation at the electrode surface [16].

The incorporation of bioactive organic compounds into layered double hydroxide (LDH) matrices has been shown to enhance electrochemical performance. Ferulic acid, a naturally occurring phenolic antioxidant, is particularly noted for its ability to facilitate electron transfer, enhance surface reactivity, and provide biocompatible functional groups [17, 18]. The antioxidant properties of ferulic acid, combined with the electrochemical benefits of aluminium/zinc LDHs, present a promising avenue for developing high-performance hybrid electrode materials. This combination can potentially enhance the stability and efficiency of electrodes while providing antioxidant protection against ROS and other electroactive species commonly found in biological fluids. Further research in this area could lead to innovative applications in energy storage, biomedical devices, and environmental protection [19–21].

Recent studies have demonstrated the effectiveness of similar hybrid systems in enhancing sensor performance. For example, a non-enzymatic glucose sensor based on poly(cafeic acid)/MWCNTs/CuO nanocomposites achieved a remarkable sensitivity of $2412 \mu\text{A mM}^{-1} \text{cm}^{-2}$ with a detection limit of $0.43 \mu\text{M}$, showcasing the potential of redox-active organic–inorganic hybrids [22]. Likewise, ZnO–CoO/reduced graphene oxide composites have exhibited excellent response characteristics and high selectivity, further affirming the value of combining conductive nanocarbon materials with catalytically active metal oxides or hydroxides [10].

While there are no specific studies on the integration of MWCNTs with aluminium/zinc

LDHs and ferulic acid for glucose sensing, the documented benefits of combining MWCNTs with other materials like ZnO and platinum nanoparticles suggest that such a combination could indeed generate a synergistic effect. This would likely result in improved electron transfer, structural support, and enhanced bioactivity, leading to a highly efficient glucose sensor [23–25].

Therefore, this study aims to develop and characterize a novel high-performance non-enzymatic glucose sensor based on MWCNTs modified with aluminium/zinc layered hydroxide–ferulate (Al/ZLH-FA) nanocomposites. By leveraging the complementary properties of each component, this work seeks to contribute a unique and efficient sensing platform to the growing field of electrochemical biosensors.

EXPERIMENTAL

Chemicals and Reagents

Phosphate buffer solution (PBS); K_2HPO_4 and KH_2PO_4 , glucose, and sodium hydroxide were supplied by Merck (Germany), paraffin oil (for paste electrode construction), and MWCNT from Timesnano (China). The entire electrochemical experiment was carried out with 0.1 mol L^{-1} PBS as the supporting electrolyte. A stock solution of 0.01 mol L^{-1} GLC was freshly produced and kept at 4°C . The modifiers, Al/ZLH-FA nanocomposite, were synthesized accordingly. Using 0.1 mol L^{-1} KCl, a 4 mmol L^{-1} potassium ferricyanide (III) $\text{K}_3[\text{Fe}(\text{CN})_6]$ solution was prepared. All solutions were prepared using deionized water (Barnstead EASY pure LF, Barnstead, Essex, UK).

Instruments

The potentiostat models Series G750 and Reference 3000 (Gamry, USA) were used for cyclic voltammetry (CV), electrochemical impedance spectroscopy (EIS), square-wave voltammetry (SWV) experiments. The working, counter and reference electrodes are the Al/ZLH-FA/MWCNT, platinum wire and Ag/AgCl respectively. A benchtop pH meter model Thermo Scientific Orion 720A (Thermo Fisher Scientific Corp. USA) was used to modify the pH readings after they had been calibrated using a standard pH buffer solution. Al/ZLH-FA and Al/ZLH-FA/MWCNT were investigated morphologically using the transmission electron microscope (TEM) model SU8020 UHR (Hitachi, Japan).

Preparation of the Al/ZLH-FA and Al/ZLH-FA/MWCNT

The proposed electrodes were created by thoroughly combining 0.09 g of MWCNTs and 0.01 g of Al/ZLH-FA with a mortar grinder. Two droplets of paraffin oil were added, and the mixture was stirred to create a homogenous paste. A copper wire was attached to one

end of the Teflon tubes (id = 2.0 mm, 3.0 cm long) that held the homogenised material in purpose of to create electrical connection with the potentiostat. A similar method was used to create a non-modified MWCNT paste electrode without the Al/ZLH-FA modification to assess electrochemical activity.

Measurement of Glucose

Electrochemical measurements for glucose (GLC) determination were carried out using square-wave voltammetry (SWV) with a conventional three-electrode system consisting of an Al/ZLH-FA/MWCNT paste electrode as the working electrode, a platinum wire as the counter electrode, and an Ag/AgCl electrode as the reference electrode. All measurements were performed in 10 mL of 0.1 mol L⁻¹ phosphate buffer solution (PBS) under ambient laboratory conditions (25 ± 2 °C).

Before glucose determination, the effect of solution pH on the electrochemical response was investigated using PBS in the pH range of 6.0–8.0. The optimum pH was selected based on the highest oxidation peak current obtained for glucose. Unless otherwise stated, all subsequent electrochemical measurements were conducted at the optimized pH of 7.0.

Square-wave voltammetry measurements were recorded over the potential range of 0.0 to +0.7 V using a frequency of 120 Hz, a pulse amplitude of 60 mV, and a step potential of 5 mV. Prior to peak-current determination, baseline correction was applied to the SW voltammograms to minimize the contribution of background current and facilitate accurate evaluation of the glucose oxidation peak. Before each measurement, the electrode surface was gently polished on clean weighing paper to obtain a fresh and reproducible surface. After each analysis, the electrode was rinsed thoroughly with deionized water to remove any adsorbed species before subsequent measurements.

The analytical performance of the proposed sensor was evaluated using standard glucose solutions prepared by serial dilution of a freshly prepared 0.01 mol L⁻¹ glucose stock solution with 0.1 mol L⁻¹ PBS. Calibration measurements were performed over the concentration range of 3–300 µmol L⁻¹, and the oxidation peak current was recorded for each concentration to construct the calibration curve. The sensitivity was determined from the slope of the calibration plot, while the limit of detection (LOD) was calculated using the 3σ/m criterion, where σ is the standard deviation of the blank signal and *m* is the slope of the calibration curve.

The selectivity of the proposed sensor was evaluated by investigating the influence of several potentially interfering species commonly encountered in glucose analysis, including fructose, maltose, sucrose, lysine, calcium chloride, magnesium chloride, and sodium chloride. The tolerance limit for each interferent was defined as the maximum concentration producing an analytical error of less than ±10% in the determination of 0.1 mmol L⁻¹ glucose.

To evaluate the analytical applicability of the proposed sensor, recovery studies were carried out using glucose-spiked deionized water. Known amounts of glucose (1.5 × 10⁻⁴, 2.5 × 10⁻⁴, and 3.5 × 10⁻⁴ mol L⁻¹) were added to the aqueous samples, and each concentration was analysed in triplicate under the optimized experimental conditions. The recovery percentage and relative standard deviation (RSD) were calculated to assess the accuracy and precision of the proposed sensing method.

RESULTS AND DISCUSSION

Characterization of the Al/ZLH-FA and Al/ZLH-FA/MWCNT

Figure 1(A) shows the morphology of Al/ZLH-FA which consists mainly of aggregated sheet-like or platelet-like structures, which are characteristic of layered hydroxide materials. Compared with the composite, no tubular carbon nanotube structures are observed, confirming the absence of MWCNTs in this sample.

In contrast, Figure 1(B) shows the TEM image of the Al/ZLH-FA/MWCNT composite. The long, hollow tubular structures (indicated by arrows) correspond to the multi-walled carbon nanotubes (MWCNTs), while the darker aggregated regions attached to or surrounding the nanotubes are attributed to the Al/ZLH-FA nanostructures. The close association between the nanotubes and Al/ZLH-FA suggests that the layered hydroxide-ferulate material was successfully incorporated onto the MWCNT network, forming an interconnected composite structure. Such intimate contact between the conductive MWCNTs and the electroactive Al/ZLH-FA is expected to facilitate electron transfer and increase the electroactive surface area of the modified electrode.

The comparison between Figures 1(A) and 1(B) therefore suggests the successful incorporation of Al/ZLH-FA with the MWCNT framework to form the composite material.

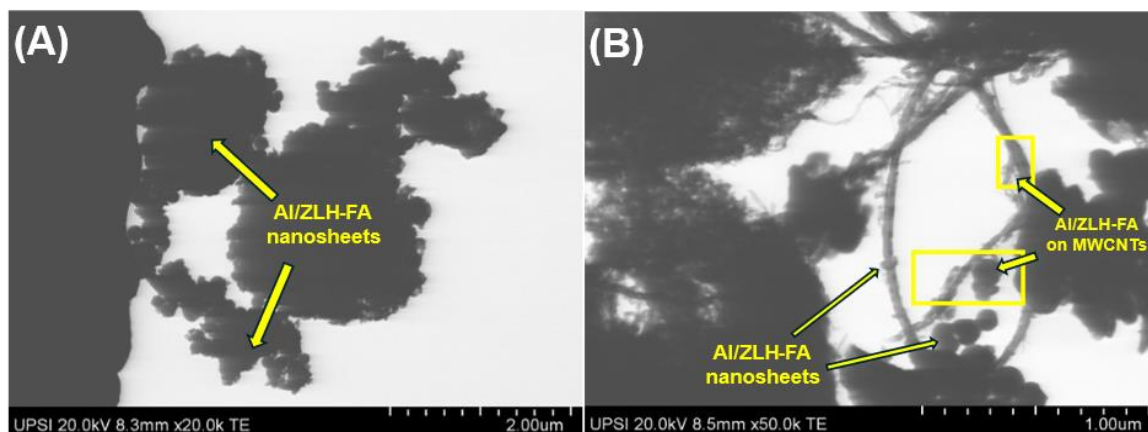


Figure 1. TEM image of (A) Al/ZLH-FA and (B) Al/ZLH-FA/MWCNT.

Electrochemical Response of $K_3[Fe(CN)_6]$ at Al/ZLH-FA/MWCNT Paste Electrode

Prior to electrochemical oxidation of analytes, the electrodes' electrochemical properties were compared utilising EIS and CV techniques in a solution containing potassium ferricyanide ($K_3[Fe(CN)_6]$). $K_3[Fe(CN)_6]$ is often used as a benchmark redox probe in CV to evaluate the performance of newly developed sensors and modified electrodes [26]. In this study, the electrochemical properties of Al/ZLH-FA/MWCNT paste electrode was investigated in a 4 mmol L⁻¹ $K_3[Fe(CN)_6]$ probe solution containing 0.1 mol L⁻¹ KCl as supporting electrolyte. The CVs were detected at a scan rate of 150 mV s⁻¹ in the potential range of -0.3 V to +0.9 V. As specified in Figure 2, For the unmodified MWCNT (curve a), the redox peak current response was measured at $I_{pa} = 9.508 \mu A$ and $I_{pc} = 9.297 \mu A$. In the Al/ZLH-FA/MWCNT

paste electrode (curve b), the redox peak current is simultaneously raised at $I_{pa} = 15.61 \mu A$ and $I_{pc} = 15.04 \mu A$. The probe showed a good reversible CV response on the Al/ZLH-FA/MWCNT paste electrode. The increased redox currents recorded for the Al/ZLH-FA/MWCNT paste electrode are indicative of enhanced electron transfer efficiency between the electrode surface and the electrolyte. This enhancement can be attributed to effective surface modification, optimized electron transport pathways, improved mass transfer, and a well-controlled electrochemical environment [27–29]. Additionally, the observed peak-to-peak potential separations (ΔE_p) for both the non-modified MWCNT (0.165 V) and the Al/ZLH-FA/MWCNT paste electrode (0.205 V) exceed the theoretical value of 0.059 V expected for a fully reversible system [30]. This deviation suggests that the electron transfer process at both electrodes follows a quasi-reversible mechanism [31].

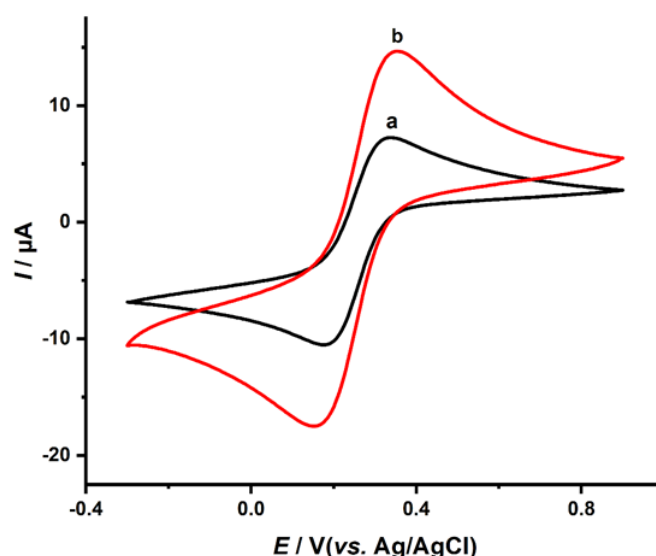


Figure 2. Cyclic voltammograms of (a) non-modified MWCNT and (b) Al/ZLH-FA/MWCNT paste electrodes in 4 mmol L⁻¹ $K_3[Fe(CN)_6]$ containing 0.1 mol L⁻¹ KCl, at a scan rate of 150 mV s⁻¹.

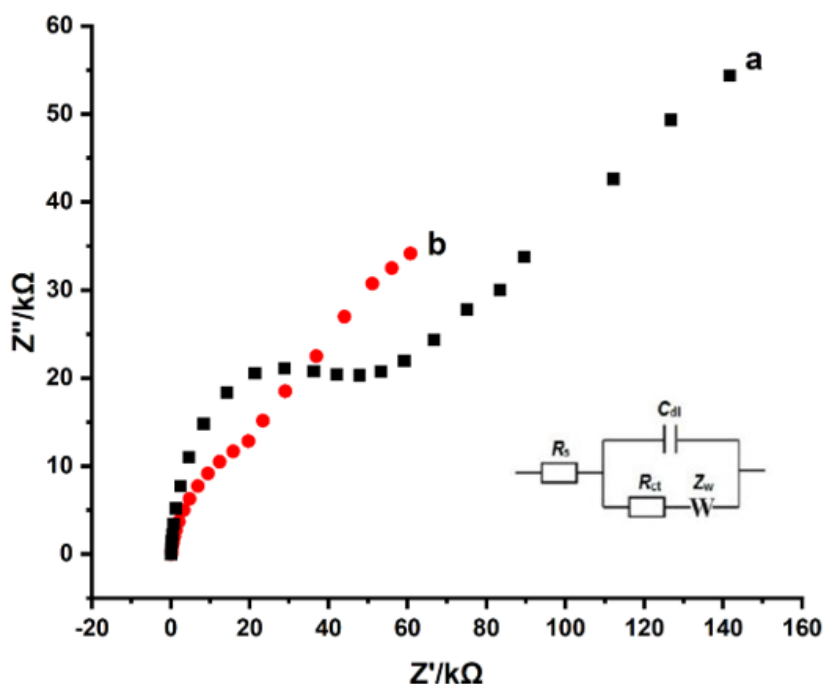


Figure 3. Nyquist plots made with (a) unmodified MWCNT and (b) Al/ZLH-FA/MWCNT paste electrodes in a solution of 4 mmol L⁻¹ K₃[Fe(CN)₆] in 0.1 mol L⁻¹ KCl. The Randles equivalent electrical circuit used for data fitting is shown in the inset.

EIS is frequently employed to evaluate the kinetics of electron transfer between the sensor surface and the electrolyte. The electronic characteristics of the modified Al/ZLH-FA/MWCNT paste electrode in 4 mmol L⁻¹ K₃[Fe(CN)₆] solution were assessed in this investigation using EIS over a frequency range of 1 MHz to 1 Hz. The EIS spectra are categorised into two segments, as illustrated in Nyquist plots: a semicircular section at elevated frequencies and a linear section at reduced frequencies. The semicircle diameter indicates electron transfer resistance (R_{ct}), which works as a kinetic control parameter for interfacial electron transfer at the electrode, while the linear part reflects the diffusion process [32]. Figure 3 depicts the Nyquist plots, which demonstrate the unique of the electrodes' impedimetric performances under study and the related Randles equivalent circuit model. Non-modified MWCNT had a high R_{ct} (43.53 kΩ), but modified Al/ZLH-FA/MWCNT had a lower R_{ct} (7.93 kΩ). This implies that compared to bare MWCNT, the Al/ZLH-FA/MWCNT electrode has higher conductivity and faster electron transport.

In addition, the R_{ct} values are used to calculate the electron transfer apparent rate (k_{app}) in order to determine the redox pair kinetic facility using the following equation (Eq. 1)

$$k_{app} = \frac{RT}{F^2 R_{ct} C} \quad (\text{Eq. 1})$$

where, R is the gas constant (8.314 J mol⁻¹ K⁻¹), T is the absolute temperature (298 K), F is the Faraday's constant (96485 C mol⁻¹) and C is the K₄[Fe(CN)₆] solution concentration. This study found that the calculated k_{app} values for bare MWCNT and Al/ZLH-FA/MWCNT were 1.53 × 10⁻⁶ cm s⁻¹ and 8.39 × 10⁻⁶ cm s⁻¹, respectively. The high k_{app} and low R_{ct} values for Al/ZLH-FA/MWCNT indicating a fast electron transfer process and good conductivity compared to the non-modified MWCNT [33]. The EIS results support the outcome of the CV studies.

The correlation between peak current, peak potential, and scan rate provides valuable information about electrochemical mechanisms. As a result, the influence of scan rate was investigated on the Al/ZLH-FA/MWCNT paste electrode using a CV experiment with scanning potentials ranging from -0.3 V to +0.9 V. The peak current increased with the scan rate from 10 mV s⁻¹ to 300 mV s⁻¹. Figure 4 shows the corresponding CV diagram. The regression equations for anodic peak current $I_{pa} = 0.0426v + 8.4612$ ($R^2 = 0.9948$) and cathodic peak current $I_{pc} = -0.0413v - 7.7308$ ($R^2 = 0.9924$) showed substantial linear correlation with scan rates (Figure 5A). A surface-controlled quasi-reversible process on the Al/ZLH-FA/MWCNT electrode surface is indicated by the slight shift in the redox potentials (E_{pa} and E_{pc}) that occurs as the scan rate increases.

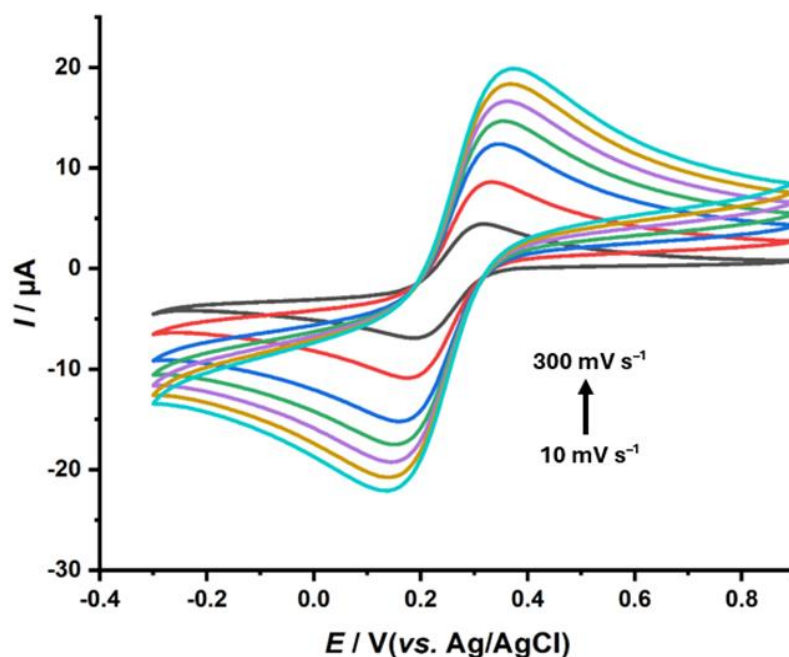


Figure 4. Cyclic voltammograms in the solution of $4 \text{ mmol L}^{-1} \text{K}_3[\text{Fe}(\text{CN})_6]$ in $0.1 \text{ mol L}^{-1} \text{KCl}$ at scan rates of 10, 40, 70, 100, 150, 200, 250, and 300 mV s^{-1} .

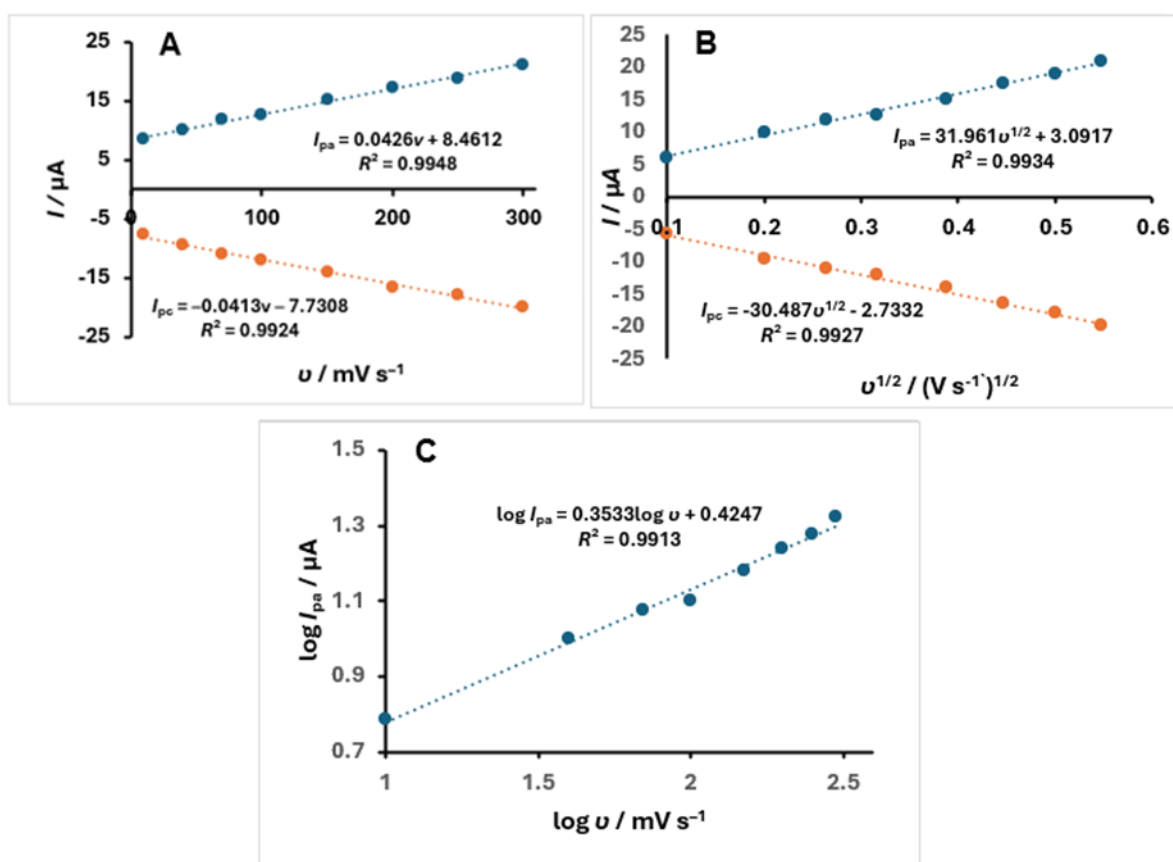


Figure 5. Relationship between (A) peak currents and potential scan rates, (B) peak currents and square root of scan rates, and (C) logarithmic values of the anodic peak current ($\log I_{pa}$) against the logarithm of the scan rate ($\log v$).

A linear plot of the voltammetric peak current against the square root of the scan rate was constructed, displaying a linear dependence (Figure 5B). The anodic peak current, $I_{pa} (\mu A) = 31.961v^{1/2} + 3.0917$ ($R^2 = 0.9934$) and cathodic peak current, $I_{pc} (\mu A) = -30.487v^{1/2} - 2.7332$ ($R^2 = 0.9927$) show that, within the studied scan rate range, a diffusion-controlled mechanism governs the electrochemical reaction of $K_3[Fe(CN)_6]$ at the modified electrode. The CV data were further analyzed by plotting the logarithmic values of the peak current ($\log I_p$) against the logarithm of the scan rate ($\log v$). A straight line was obtained (Figure 5C), which can be expressed by the equation $\log I_p (\mu A) = 0.3553 \log v + 0.42$ surface 47 ($R^2 = 0.9913$). The electrode process may be regulated by a diffusion-controlled mechanism, as indicated by the slope of 0.3553, which is near the theoretical value of 0.5 [34].

To further electroactive characterizations, the electroactive surface area of Al/ZLH-FA/MWCNT paste electrode was analysed in 4 mmol L⁻¹ $K_3[Fe(CN)_6]$ using the effect of different scan rates. The electroactive surface area of the Al/ZLH-FA/MWCNT composite electrode was therefore determined using the Randles-Sevcik equation (Eq. 2) and the linear regression equation of the previously described curve:

$$I_p = (2.69 \times 10^5) n^{3/2} A D^{1/2} C v^{1/2} \quad (\text{Eq. 2})$$

where I_p is the peak current (μA), n is the number of electron involved in the redox reaction ($n = 1$ for $K_3[Fe(CN)_6]$), A is the electroactive surface area (cm^2), D is the diffusion coefficient of the $K_3[Fe(CN)_6] = 7.6 \times 10^{-6} cm^2 s^{-1}$, C is the $K_3[Fe(CN)_6]$ concentration (4 mmol L⁻¹), and v is the scan rate ($V s^{-1}$). The amount of $I_{pa}/v^{1/2}$ was driven from the slope of the corresponding plots for the electrode

and subsequently, the value of A was mathematically measured by (Eq. 2). Therefore, the A value of Al/ZLH-FA/MWCNT paste electrode was defined to be 0.01 cm^2 .

Additionally, the heterogeneous standard speed constant (k^0), a crucial kinetic parameter to assess the quasi-reversible behaviour and the speed of electron transfer between an electrode and electroactive factors, was estimated using the electrode's recorded cyclic voltammogram at various scan rates [35]. Nicholson's method was used for this purpose as the following equation 3 (Eq. 3):

$$\Psi = k^0 [\pi D n F v / RT]^{-1/2} \quad (\text{Eq. 3})$$

where Ψ , D , n , F , v , R , and T denote the kinetic parameter, diffusion coefficient, number of electrons, Faraday constant (96,485 Cmol⁻¹), scan rate, gas constant (8.314 J K⁻¹ mol⁻¹), and temperature (298 K), respectively. The electrochemical reversibility of a single electrode is represented by the symbol ψ , which stands for Nicholson's dimensionless kinetic parameter. If $\psi \geq 7.0$, a system is deemed reversible; if $\psi < 7.0$, a quasi-reversible process. To determine the values of ψ , Lavagnini et al. [36] determined the validity ranges for the experimental ΔE_p and x values (mV). The following equation was used to apply Nicholson's technique for this purpose. Equations 4 (Eq. 4) and 5 (Eq. 5) were used to determine ψ values with differences in the redox peak potential of < 200 mV and > 200 mV, respectively.

$$\Psi = (-0.6288 + 0.0021 \Delta E_p) / (1 - 0.017 \Delta E_p) \quad (\text{Eq. 4})$$

$$\Psi = 2.18 [D/\pi]^{1/2} \exp[(D^2 F / RT) n \Delta E_p] \quad (\text{Eq. 5})$$

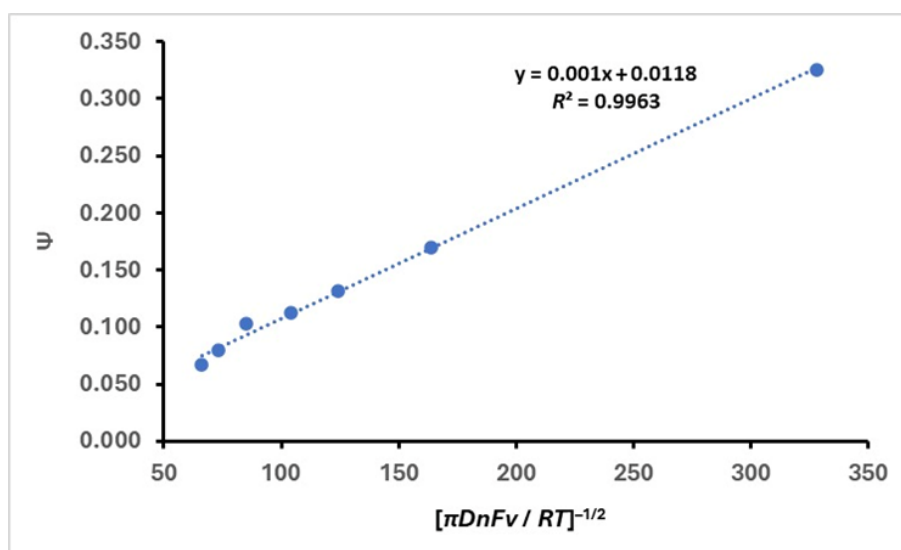


Figure 6. Plot of ψ versus square root of $\pi D n F v / RT$ for Al/ZLH-FA/MWCNT paste electrode.

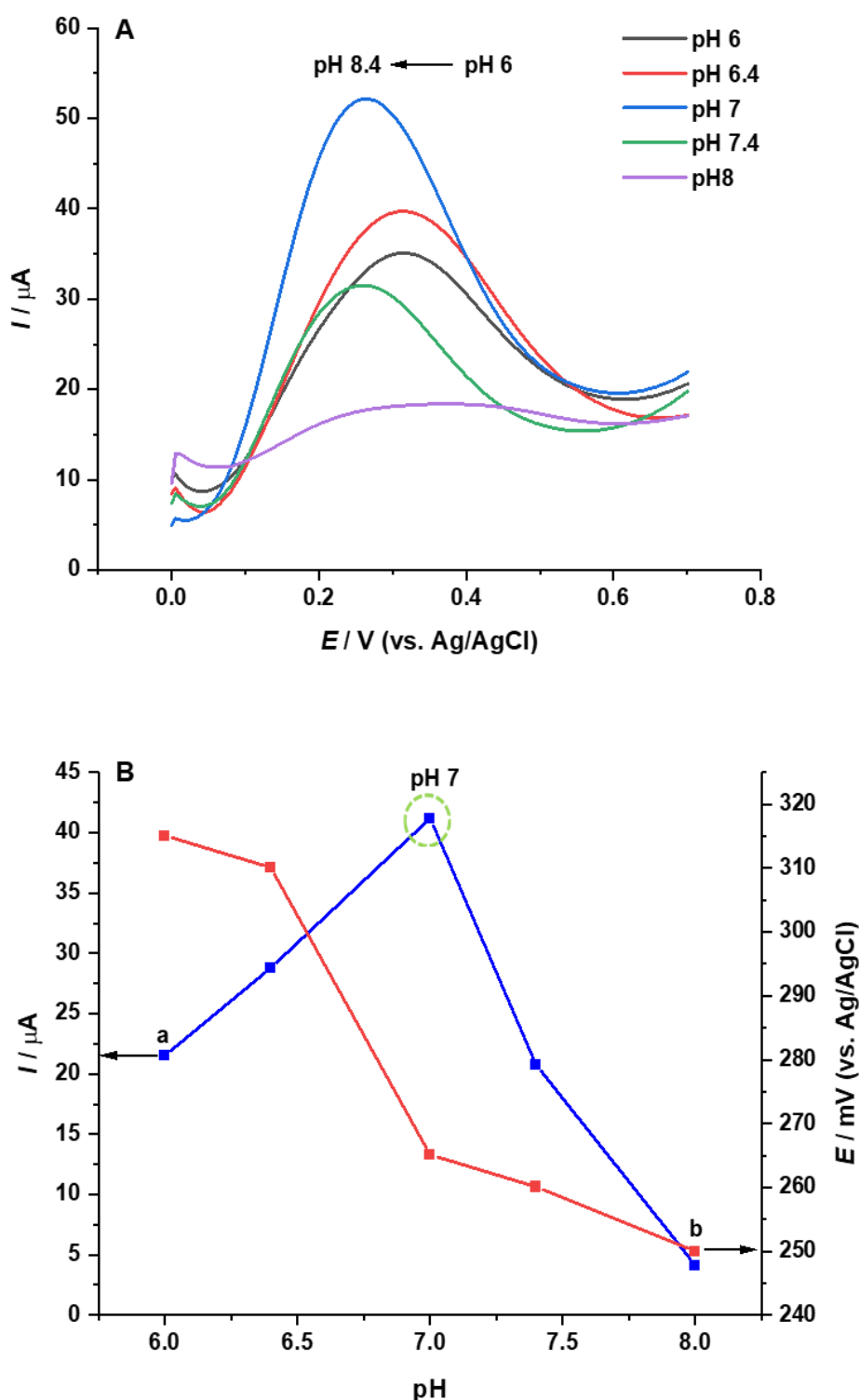


Figure 7. (A) SW voltammograms at different pH and (B) Effect of buffer pH on the SWV response of GLC oxidation towards (a) peak current, I (μA), and (b) peak potential, E (mV).

As a result, the slope of the ψ versus $[\pi\text{DnFv}/RT]^{-1/2}$ plots (Figure 6) was used to calculate the value of k^0 , which was found to be $1.1 \times 10^{-3} \text{ cm s}^{-1}$ for the Al/ZLH-FA/MWCNT paste electrode. The modified electrode displayed a quasi-reversible

electron transport process with a relatively high k^0 ($k^0 \geq 2.0 \times 10^{-5} \text{ cm s}^{-1}$).

The addition of Al/ZLH-FA as a MWCNT modifier improves the modified electrode's

conductivity performance, electroactive surface area, and electron transfer rate.

Optimization of Detection Condition: The Solution pH Effect

In most circumstances, the electrolyte pH is a critical parameter in the electrochemical reaction. SWV characterisation was used to investigate the pH-dependent current response to the oxidation of 0.1 mmol L⁻¹ GLC in the pH range 6-8 using phosphate buffer (Figure 7A). Figure 7B illustrates the peak currents and potentials measured in analyte solutions at various pH levels. The GLC oxidation peak potential dropped as the pH value increased. The negative shift in peak potential with increasing pH buffer indicates that protons are involved in GLC oxidation. The peak oxidation current was highest at pH 7 and declined as the solution diverged from neutral. The results could be related to the low pH environment, which decreases GLC oxidation. Both acidic and alkaline environments rendered GLC oxidation less favourable, resulting in a lower current peak height. As a result, pH 7 was chosen as the GLC detection threshold.

Electrochemical Oxidation of Glucose at Al/ZLH-FA/MWCNT Paste Electrode

Using the SWV technique, the electrochemical behaviour of the Al/ZLH-FA/MWCNT paste electrode against glucose oxidation was investigated in the potential window between 0 and +0.7 V at a scan rate of 100 in 0.1 M PBS (pH 7). Figure 8 shows that the peak current of GLC oxidation on Al/ZLH-FA/MWCNT is 54 times greater ($I_{pa} = 15.67 \mu A$) than the peak currents of non-modified MWCNT ($I_{pa} = 0.288 \mu A$). The inclusion of Al/ZLH-FA increased the electrode's electroactive surface area, resulting in a higher current level. Therefore, the electrochemical sensor utilising Al/ZLH-FA/MWCNT represents a promising alternative to GLC electroanalysis.

Furthermore, SWV was selected for subsequent evaluation of the sensor performance because it provided a stronger analytical signal. Minor variations in the background current were observed between independently prepared electrodes, which may be attributed to variations inherent in the manual fabrication and surface renewal of the paste electrodes. Nevertheless, comparable glucose oxidation responses were obtained, indicating satisfactory reproducibility of the modified electrodes.

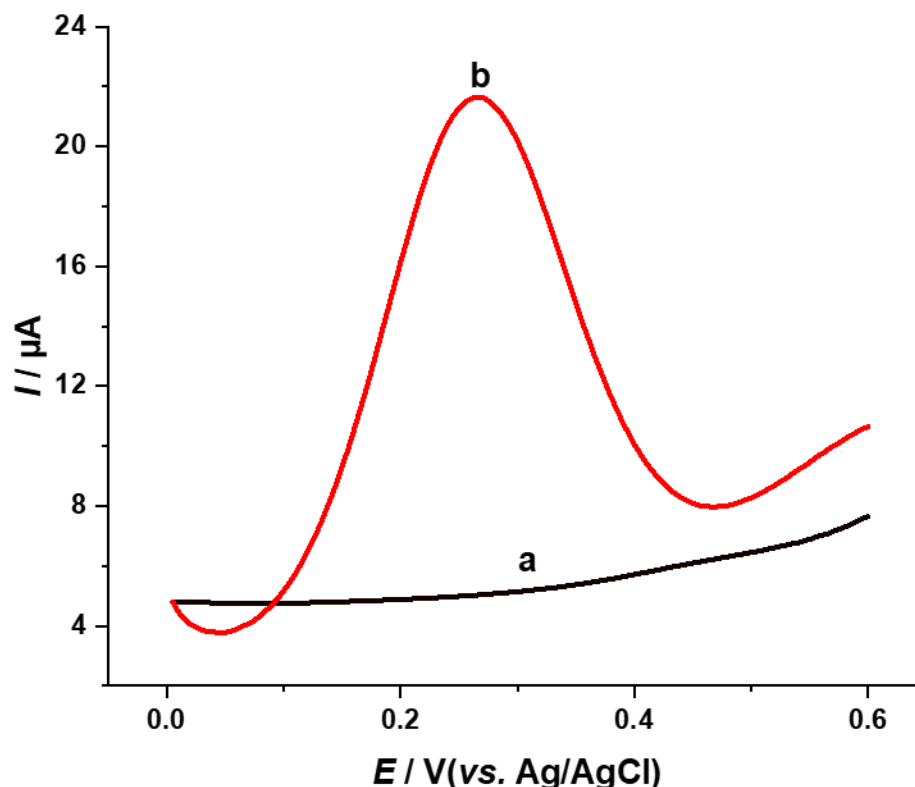


Figure 8. SW voltammograms in the solution of 0.1 mmol L⁻¹ GLC in 0.1 mol L⁻¹ PBS at pH 7 using: (a) non-modified MWCNT and (b) Al/ZLH-FA/MWCNT.

Table 1. Determination of GLC in comparison of various modified electrodes.

Electrode	LWR (μM)	LOD (μM)	Method	Ref.
rGO/NiO/GCE	$2 \times 10^1 - 8 \times 10^1$	1.93×10^1	SWV	[37]
MPBA/NH ₂ @Pt/AgVO ₃	$5 \times 10^1 - 2.2 \times 10^4$	6.13×10^1	SWV	[38]
SPCE/MWCNT/ZnO/MIP	$0 - 5 \times 10^3$	1.81×10^3	CV	[39]
Cu(II)-GO/SPCE	$1 \times 10^2 - 1 \times 10^3$, $1 \times 10^3 - 1.25 \times 10^4$	1.20×10^2	Amp	[40]
CuO/ITO electrode	$1 \times 10^1 - 4 \times 10^3$	3	BS-CV	[41]
Graphene/MIL-101(Cr)-NH ₂ /SS	$1 \times 10^1 - 2 \times 10^3$	4	SWV	[42]
Al/ZLH-FA/MWCNT/CPE	$3 - 3 \times 10^2$	5×10^{-2}	SWV	This work

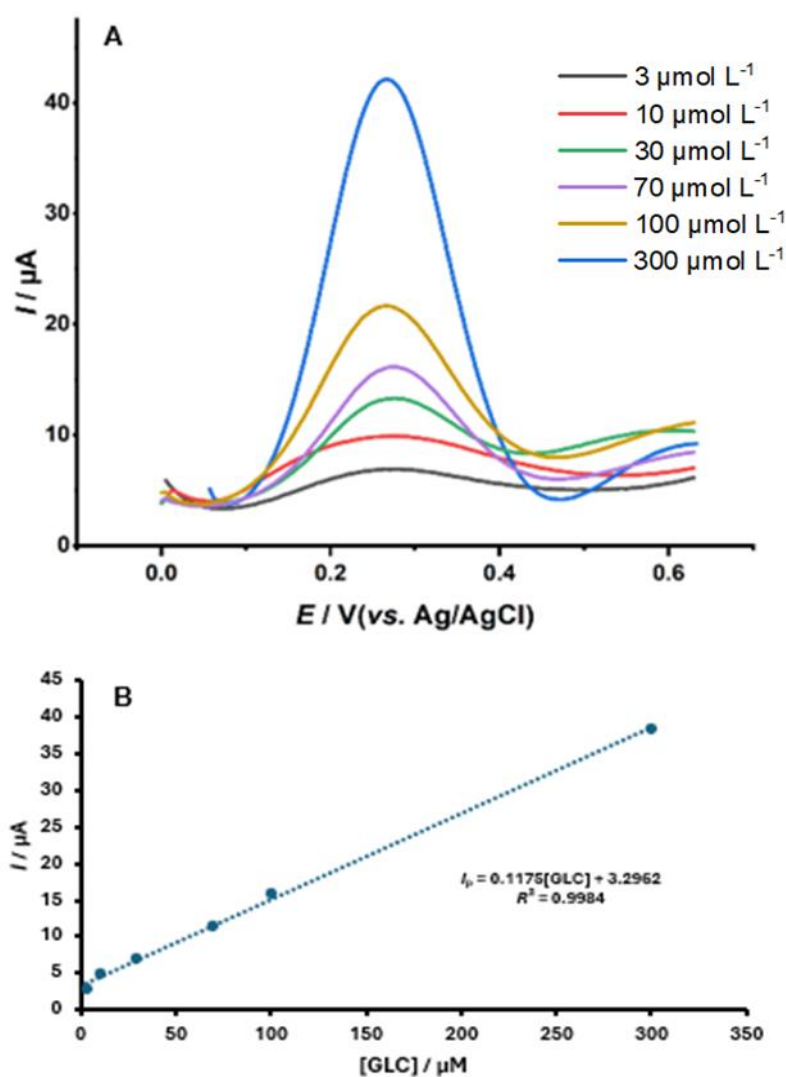


Figure 9. (A) SWV voltammograms of different concentration of GLC and (B) linear relationship between peak currents (I_p) and GLC concentration ($[\text{GLC}]$) in the range 3 – 300 $\mu\text{mol L}^{-1}$. Measurement conditions: pH = 7, pulse size = 60 mV, step size = 5, and frequency = 120 Hz over the potential range of 0 to +0.7 V.

Analytical Characteristics

To determine the quantitative capabilities of the suggested electrode, Al/ZLH-FA/MWCNT paste electrode, SWV was utilized to generate a calibration curve for electrochemical detection of GLC as a simple and sensitive approach. Diluting the standard stock yielded a series of GLC solutions at varying concentrations in 0.1 M PBS (pH 7). Following each measurement, the modified electrode was rinsed with distilled water. Figure 9A depicts SWV voltammograms with varying GLC concentrations on the modified electrode's surface. Under optimal conditions, when GLC concentrations increase, the SWV oxidation peaks gradually increased. Figure 9B shows a linear relationship between the peak current (I_p) and GLC concentration ([GLC]) ranging from 3–300 $\mu\text{mol L}^{-1}$, and sensitivity of 0.1175 $\mu\text{A}\mu\text{mol L}^{-1}$ which can be described by Equation 6.

$$I_p (\mu\text{A}) = 0.1175[\text{GLC}] (\mu\text{mol L}^{-1}) + 3.2962 (R^2 = 0.9984) \quad (\text{Eq. 6})$$

The proposed modified electrode's limit of detection (LOD) for GLC was determined to be 0.05 $\mu\text{mol L}^{-1}$ using the $3\sigma/m$ ratio, where σ represents the standard deviation and m is the slope of the linear segment of the calibration curve. Table 1 compares the analytical performance of the Al/ZLH-FA/MWCNT

paste electrode to several earlier electrodes used in the previous works to determine GLC. This comparison indicates that the proposed modified electrode possesses good glucose detection performance including fast response, high sensitivity and low detection limit.

Interference Effect

The selectivity of the proposed Al/ZLH-FA/MWCNT paste electrode was evaluated by examining the influence of several potentially interfering species commonly encountered in glucose analysis. Fructose, maltose, sucrose, lysine, calcium chloride, magnesium chloride, and sodium chloride were individually introduced into a solution containing 0.1 mmol L^{-1} glucose under the optimized experimental conditions. The tolerance limit was defined as the maximum concentration of each interfering species that produced a deviation of less than $\pm 10\%$ in the glucose oxidation peak current. As summarized in Table 2, fructose and maltose were tolerated at concentrations up to **50-fold** higher than that of glucose, whereas sucrose, lysine, calcium chloride, magnesium chloride, and sodium chloride were tolerated at **10-fold** excess without causing significant interference. These findings demonstrate the satisfactory selectivity of the proposed sensor for glucose determination.

Table 2. Tolerance limits of potential interfering species for the determination of 0.1 mmol L^{-1} glucose under the optimized experimental conditions. The tolerance limit is expressed as the maximum fold excess relative to the glucose concentration producing an analytical error of less than $\pm 10\%$.

Interfering species	Maximum tolerance (fold excess relative to glucose)
Fructose	50-fold
Maltose	50-fold
Sucrose	10-fold
Calcium chloride	10-fold
Lysine	10-fold
Magnesium chloride	10-fold
Sodium chloride	10-fold

Table 3. Quantification of GLC samples in spiked deionized water.

Sample	GLC added (M)	GLC found (M)	Recovery (%)	RSD (%)
Spiked deionized water	1.50×10^{-4}	1.47×10^{-4}	98	0.29
	2.50×10^{-4}	2.36×10^{-4}	94	0.33
	3.50×10^{-4}	3.32×10^{-4}	95	0.37

Recovery Study in Glucose-Spiked Deionized Water

For the recovery study, known amounts of analytical-grade glucose were spiked into the deionized water to obtain final concentrations of 1.5, 2.5 and 3.5 x 10⁻⁴ M. Each spiked sample was prepared in triplicate and analysed immediately via SWV at Al/ZLH-FA/MWCNT paste electrodes. The results were tabulated in Table 3.

The recoveries obtained for glucose ranged from 94% to 98%, indicating good agreement between the added and measured concentrations in glucose-spiked deionized water. The low RSD values (<0.4%) demonstrate excellent repeatability of the electrochemical measurements under the optimized experimental conditions. These findings indicate that the proposed Al/ZLH-FA/MWCNT sensor is capable of accurately determining glucose in a simple aqueous medium with minimal matrix interference. The recovery study provides an initial evaluation of the analytical performance of the sensor and serves as a baseline for future application to more complex biological or environmental samples.

CONCLUSION

In this study, an Al/ZLH-FA/MWCNT paste electrode was successfully fabricated for the development of a non-enzymatic electrochemical glucose sensor. The incorporation of Al/ZLH-FA into the MWCNT matrix significantly enhanced the electrochemical performance of the modified electrode by improving electron-transfer characteristics, as confirmed by cyclic voltammetry and electrochemical impedance spectroscopy. Under the optimized experimental conditions, the proposed sensor exhibited a wide linear range (3–300 µmol L⁻¹), high sensitivity, a low detection limit (0.05 µmol L⁻¹), satisfactory selectivity against common interfering species, and excellent recovery (94–98%) with good precision (RSD < 0.4%) for glucose determination in glucose-spiked deionized water (aqueous medium). These findings demonstrate that the Al/ZLH-FA/MWCNT paste electrode is a promising platform for glucose determination in aqueous media and provides a useful foundation for future evaluation in more complex biological sample matrices.

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- 111 Mohamad Hafiz Ahmad Tajudin, Mohamad Syahrizal Ahmad, Illyas Md Isa, Emi Norzehan Mohamad Mahbob, Norhayati Hashim, Anwar Ul-Hamid, Mohamad Idris Saidin, Rahadian Zainul, Suyanta M. Si and Sofian Ibrahim
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