



Albumin fouling-free electrochemical sensors based on polyaniline single walled carbon nanotube composites for precise purine metabolomics

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ABSTRACT

Electrochemical sensors enable rapid and accurate detection of targets. However, fouling is a burden that restricts sensor performance in complex biofluids and fouling resistant or fouling-free property is paramount to guarantee the reliable operation of sensors. Single-walled carbon nanotubes (SWCNTs) possess an exceptional catalytic capability owing to the weak molecular adsorption of sp³ carbon. Poly(aniline-N-propane sulfonic acid) (PAPS) polymer dispersed SWCNTs with a high conductivity (4.184 S/cm), and hydrophilicity were prepared to circumvent the fouling issues. The length of the PAPS dispersed SWCNTs were $3.1 \pm 1.0 \mu\text{m}$ and the modified screen-printed carbon electrodes (PAPS-SWCNTs/SPCEs) demonstrated efficient electro-oxidation of purines, such as, uric acid (UA), xanthine (XA), and hypoxanthine (HX) after exposure to high concentrations of a common foulant, serum albumin. The peak-to-peak separations of UA–XA, XA–HX, and UA–HX were 0.396 V, 0.352 V, and 0.748 V, respectively. The detection limits of UA, XA, and HX were 0.047, 0.049, and 0.052 μM respectively. The practical applicability of the sensor was established using human serum and synthetic urine samples. The fabricated sensor is fouling-free and could serve as a potential diagnostic device for the early detection of renal diseases, such as renal calculi, chronic kidney diseases, and renal failure in resource-limited settings, since it does not require scrupulous sample pretreatment, frequent recalibration or prolonged waiting times. Moreover, the developed sensor adheres to ASSURED criteria, which is crucial for the diagnosis of renal diseases in resource-limited settings.

1. Introduction¹

The frequent and existing challenge of electrochemical sensors is fouling or interference, leading to poor sensitivity and selectivity. In biological samples, the common foulant is serum albumin. To overcome this constraint, fouling resistant or fouling-free sensor materials are paramount to guarantee their reliable operation [1–4]. Hence, stable, and cost-effective fouling free materials are crucial in fouling free sensors fabrication. To date, different macromolecular materials such as

zwitterionic materials, poly(ethylene glycol), poly(3-octylthiophene), proteins, and peptides have been explored to deal with fouling issues [5,6]. To completely circumvent fouling, well-dispersed single-walled carbon nanotubes (SWCNTs) are ideal electrode modifiers [7]. Hence, Pristine SWCNTs have captivated considerable practical interest owing to their unique quasi-one-dimensional structure, mechanical strength, electrical conductivity, and chemical stability. Nonetheless, pristine SWCNTs have high surface energy to form bundles or entangled ropes. The hydrophobicity and significant inter-tube van der Waals interaction

Abbreviations: (SPCE), screen-printed carbon electrode; (SWCNT), single-wall carbon nanotubes; (FE-SEM), field-emission scanning electron microscope; (EDX), energy-dispersive X-ray spectroscopy; (TEM), transmission electron microscopy; (XPS), X-ray photoelectron spectroscopy; (CV), cyclic voltammetry; (ASA), active surface area; (XA), xanthine; (HX), hypoxanthine; (UA), uric acid; (AA), ascorbic acid; (DA), dopamine; (Cr), creatinine; (I_{pa}), anodic peak current; (I_{pc}), cathodic peak current; (EIS), electrochemical impedance spectroscopy; (DPV), differential pulse voltammetry; (LOD), limit of detection.

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SWCNTs [67]. Despite considerable advancements, conjugated polymers still face several challenges on their path towards biosensing applications. These challenges include their complex structure, synthesis, and tendency to phase separate, resulting in multiphase arrangement with limited morphological stability.

Recently, efficient and simple methods for dispersing SWCNTs in aqueous media have focused on hydrophilizing CNTs with molecules that non-covalently bind to the CNT surface. This non-covalent functionalization holds great promise, as it minimizes the modification-induced changes in the electronic and mechanical properties of CNTs. Previously, we have utilized poly(2-dimethylaminoethyl methacrylate-co-styrene) poly(sodium 4-styrene sulfonate-r-LAHEMA) to increase the aspect ratio of SWCNTs [68]. Considering all these aspects, in this study, a simple PANI polymer, PAPS was employed not only to disperse SWCNTs but also to achieve fouling-free sensing of the targets in varied biological matrices.

4. Conclusions

A highly conductive PAPS-SWCNTs-2 was prepared to establish an ultrasensitive electrochemical sensor that could be applied for the selective and simultaneous quantification of purine metabolites, UA, XA, and HX. The PAPS-SWCNTs-2 provided abundant electrochemically active sites, and extraordinary electrical conductivity, which permitted the ultrasensitive quantification of purines. The selectivity results verified that the tested interferents had no apparent influence on the synchronous estimation of purine metabolites. Furthermore, the repeatability and stability of the sensor are high. Moreover, the quantification of UA, XA, and HX in human serum and synthetic urine samples showed appreciable recoveries of approximately 100 %. This is a conceptual study for the fouling free, ultrasensitive and convenient sensor preparation technique. It takes less than 10 min because PAPS-SWCNTs-2 is drop-casted directly on the electrode surface. A further advantage of our approach is the simple and simultaneous purine detection in human serum and synthetic urine. This involves a simple sample dilution with no requirements for separation or purification steps. The results indicate the favorable properties of the PAPS-SWCNTs-2/SPCE sensor for determining small purine metabolites in the presence of large albumin biomolecules, which do not influence the analysis.

CRedit authorship contribution statement

Thenmozhi Rajarathinam: Writing – original draft, Software, Methodology, Investigation, Data curation, Conceptualization. **Sivaguru Jayaraman:** Validation, Methodology, Investigation, Conceptualization. **Devaraju Subramani:** Validation, Methodology, Investigation, Data curation. **Suraj Aswale:** Validation, Methodology, Investigation, Data curation. **Jaewon Lee:** Writing – original draft, Validation, Supervision. **Hyun-jong Paik:** Validation, Supervision, Methodology. **Chang-Seok Kim:** Validation, Supervision, Methodology, Funding acquisition. **Jang-Hee Yoon:** Validation, Methodology, Investigation. **Seung-Cheol Chang:** Writing – review & editing, Supervision, Funding acquisition, Conceptualization.

Declaration of competing 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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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.coco.2025.102363>.

Data availability

Data will be made available on request.

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