

Laser-induced graphene paper-based electrodes for detection of the pancreatic cancer biomarker CA19-9 in real samples

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Summary: Pancreatic cancer has a high mortality rate due to challenges in early detection. This research reports the optimization, structural characterization, and biosensing performance of laser-induced graphene (LIG) electrodes fabricated on paper substrates for CA19-9 detection. Preliminary analyses of urine samples suggest that the paper electrodes show promising results for point-of-care applications.

Keywords: Biosensor, pancreatic cancer, CA19-9, Laser-induced graphene (LIG), paper-based electrodes.

Introduction

Pancreatic cancer remains one of the most significant challenges in modern oncology due to its high mortality and late diagnosis, which limit treatment options and reduce survival rates [1]. According to the American Society of Clinical Oncology (2023) [2], the 5-year relative survival rate is only 3% when diagnosed at an advanced stage. The absence of sensitive, selective, and practical early detection methods hinders effective treatment. Innovative, accurate, and non-invasive diagnostics offer strong potential to improve outcomes and support public healthcare [3]. Functional material-based biosensors, especially using the EIS technique, offer new paths for early diagnosis [4]. Beyond sensitivity and specificity, these devices offer portability, flexibility, and multifunctionality [3].

Experimental Procedure

Paper substrates were treated with sodium tetraborate, dried, and pressed, followed by hydrophobization using rosin in ethanol and a second pressing step. Laser processing was applied to form interdigitated graphene electrodes, with parameters optimized by varying power and speed. Morphological and structural characterization was performed using SEM-EDS and Raman spectroscopy. Capacitance measurements were assessed via EIS (Solartron SI 1260). Data

analysis was conducted using Interactive Document Maps (IDMAP) generated with the Projection Explorer Sensors software (PEX-Sensors). For biosensor validation, urine samples from patients were analyzed and compared with ELISA reference measurements. Measurement conditions are still being optimized to improve sensitivity and calibration. All procedures involving human samples were ethically approved.

Results

The formation of interdigitated electrodes (IDEs) was optimized by systematically varying laser power (3–8% of 22 W) and scanning speed (300–800 mm/min). Based on this, the best condition was achieved at 7% power and 300 mm/min for digit formation (Fig. 1), while for the terminals and contacts, the optimized parameters were 6% power and 500 mm/min (Fig. 2), with sheet resistance of $230.12 \pm 28.07 \Omega \text{ sq}^{-1}$ (Fig. 3). Morphological evaluation via scanning electron microscopy (SEM) revealed well-defined porous graphene structures. Energy-dispersive spectroscopy (EDS) confirmed the presence of carbon, oxygen, and calcium, principally. Raman spectroscopy further validated the synthesis of multilayer porous graphene, evidenced by the characteristic D, G, and 2D bands (Fig. 4). Electrochemical impedance spectroscopy (EIS) demonstrated that electrodes (terminal and contacts) produced

at 6% power and 500 mm/min exhibited the lowest resistivity values when compared to other conditions. Furthermore, a post-synthesis mechanical pressing step was found to significantly reduce real impedance by increasing the packing density of the graphene sheets. The optimized platform was applied as a capacitive biosensor for the CA19-9 biomarker. The sensing data followed a Langmuir–Freundlich model ($R^2 = 0.99$), yielding a limit of detection (LOD) of 2.38 U/mL and a limit of quantification (LOQ) of 7.86 U/mL. Preliminary analyses of urine samples (Fig. 5) show promising results for point-of-care applications. Enhancements in the measurement protocol are needed to develop a calibration curve that can more accurately quantify the biomarker, aligning more closely with the results obtained from the ELISA technique.

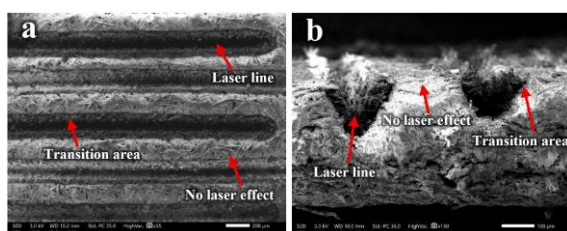


Fig. 1. SEM images of (a) top view and (b) cross-section of the interdigitated area.

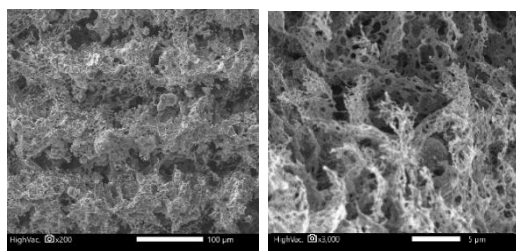


Fig. 2. SEM micrographs of LIG electrodes fabricated under 6.0% power, 500 mm min⁻¹.

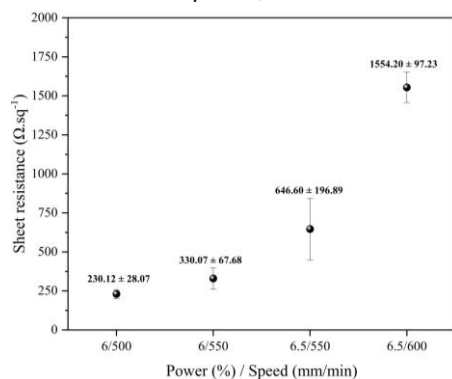


Fig. 3. Sheet resistance of different conditions of electrodes processing

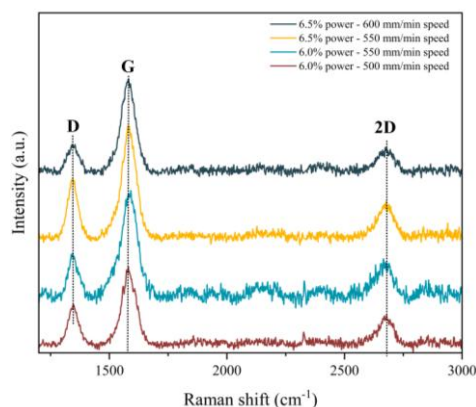


Fig. 4. CA19-9 measurements of urine samples using ELISA and Paper-based biosensor analysis.

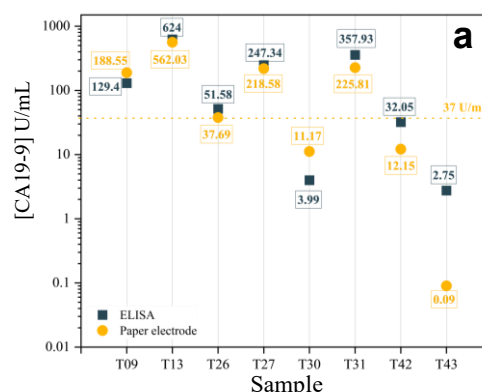


Fig. 5. CA19-9 measurements of urine samples using ELISA and Paper-based biosensor analysis.

Conclusions

Preliminary results show that LIG paper-based electrodes were successfully optimized. Urine sample analyses indicate promising performance for point-of-care applications.

Acknowledges

National Council for Scientific and Technological Development (CNPq, Brazil) (grant numbers 446650/2024-2, 107555/2024-7); National Institute of Organic Electronics INEO/FAPESP/INCT (grant number 314845/2025-9) and São Paulo Research Foundation (FAPESP) (grant number 2024/06362-6).

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