

Optimization of pH sensitive polymeric coatings on flexible printed electrodes for biomedical sensing purposes

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Flexible electrochemical pH sensors can play a significant role in healthcare since the pH level affects most biochemical reactions in the human body. Polyaniline (PANI) is one of the most widely used pH-sensitive materials, owing to its strong pH sensitivity, conductivity, chemical stability and low cost. PANI has been widely used in wearable devices due to its easy deposition and high flexibility [1]. Therefore, pairing PANI with additive manufacturing methods, such as printing technologies for electrode fabrication, would facilitate integration of flexible and low-cost electrochemical sensing platforms.

Multiple deposition methods for modifying the electrode surface with polyaniline were tested, including electropolymerization from an aniline monomer solution and drop casting of PANI emeraldine base or PANI/carbon black (CB) composite dispersion directly onto the electrode. Deposition methods were tested on different electrode substrates, including glassy carbon, screen printed carbon and inkjet printed graphene. Electrochemical characterization was conducted through cyclic voltammetry, chronopotentiometry and electrochemical impedance spectroscopy measurements in 0.1 M KCl to determine capacitance of the PANI coatings. Analytical properties were evaluated through potentiometric measurements in phosphate buffers with pH values adjusted to the physiological range (pH 5-8). Hydrogen selective electrodes (H-ISE) were also prepared and characterized for comparison by modifying the electrodes with a plasticized PVC-based membrane containing Hydrogen ionophore I. Aside from optimizing the working electrode, an optimization of reference electrode modification procedure was conducted to ensure a stable reference point in a flexible format. AgCl deposition on printed Ag substrate was conducted chemically (with FeCl₃ solution) or electrochemically (applying constant current of 2.5 mA in 3 M KCl) by varying the duration of the experiment. A stable reference potential was provided by ensuring a constant chloride background in measuring solutions.

With optimal deposition, H-ISE performance showed a reliable Nernstian sensitivity of about 60 mV/pH regardless of substrate, while sensitivity of PANI-modified pH sensors ranges from subnernstian (~40 mV/pH) values for PANI-CB composite to supernernstian values (~70-75 mV/pH) for electropolymerized PANI films. All electrodes exhibited an excellent linearity ($R^2 > 0.99$) in a wide physiologically relevant pH range. Further work includes optimizing the flexible sensor configuration for biomedical sensing purposes, such as testing the permeability of an artificial lung membrane.

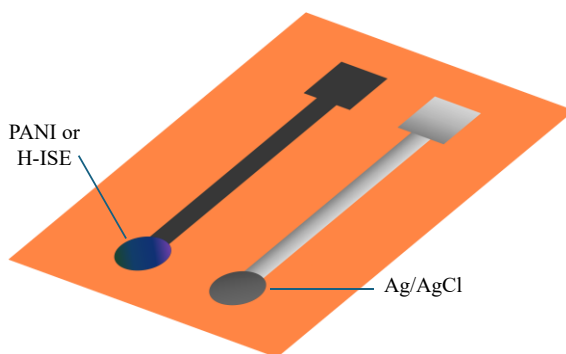


Figure 1: Flexible inkjet printed potentiometric pH sensor.

Acknowledgments

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References

[1] Y. Tang, L. Zhong, W. Wang, Y. He, T. Han, L. Xu, X. Mo, Z. Liu, Y. Ma, Y. Bao et. al., *Membranes* **12**, 504 (2022).