Development of Flexible Electrode for Bioelectrochemical Systems

Abstract

The growing global demand for sustainable wastewater treatment necessitates energy-efficient and resource-recovering technologies that can address pollution while generating bioelectricity. Bioelectrochemical systems (BES), including microbial fuel cells (MFCs), offer a promising solution but suffer from key challenges such as low power output, inefficient electron transfer, and the rigidity of conventional electrodes. The development of flexible, high-performance electrodes is crucial to enhance BES efficiency and scalability, especially for treating real textile wastewater a complex industrial effluent with high organic load and synthetic dyes.

This PhD research focuses on the development of flexible electrode materials tailored for BES applications. Three different fabrication strategies were explored: (i) polycarbonate embedded with carbon black using twin-screw melt extrusion, (ii) gold-sputtered paper-based substrates followed by electrochemical polymerization of polyaniline (PANI), and (iii) polyester microfiber nonwoven (PMN) substrates dip-coated with carbon nanotube (CNT) ink followed by PANI electrochemical polymerization. Among these, the PANI/CNT/PMN electrode demonstrated superior flexibility, electrical conductivity, and microbial compatibility, making it the most effective for real wastewater treatment applications.

The PANI/CNT/PMN flexible electrode was optimized using AC electrochemical polymerization, resulting in a mesoporous structure (BET surface area: 3.2212 m^2/g, pore diameter: 65.96 nm) that facilitated enhanced biofilm attachment, electron transfer, and mass transport. Electrochemical characterization confirmed significant improvements in charge storage capacity, reduced impedance ($R_{ct} = 2.08\ \Omega$), and high electrocatalytic activity. Cyclic Voltammetry (CV), Differential Pulse Voltammetry (DPV), and Electrochemical Impedance Spectroscopy (EIS) demonstrated the electrode's redox stability and fast electron transfer kinetics, surpassing traditional carbon cloth electrodes.

The developed electrode was tested in a Multielectrode Assembly Bioelectrochemical Reactor (MEABR) coupled with a Biophotovoltaic (BPV) cell for real textile wastewater treatment. The system achieved power and current densities of 1500 ± 4.5 mW/m^2 and 500 ± 6.5 mA/m^2, representing a nearly 2000-fold improvement over carbon cloth electrodes. The pollutant removal efficiency was also significantly enhanced, with 85.9% COD removal, 81.06% TOC removal, and 82.7% dye decolorization.

Microbial community analysis using 16S rRNA sequencing revealed that the biofilm on the PANI/CNT/PMN electrode was dominated by an uncultured filamentous, nanowire-producing *Lysinibacillus* strain, known for its high extracellular electron transfer (EET) efficiency. Advanced biofilm characterization, including Time-of-Flight Secondary Ion Mass Spectrometry (ToF-SIMS), provided 3D imaging and depth profiling, identifying malonic acid secretion as a key metabolic byproduct, indicating enhanced biofilm activity and electrochemical performance.

To further optimize biofilm-electrode interactions, twelve different nanomaterials and thin films were integrated into the PANI/CNT/PMN electrode, leading to material-specific microbial enrichment and enhanced biodegradation in the MEABR system. The integration of the BPV cell enabled an energy-positive configuration, demonstrating the feasibility of a self-sustaining wastewater treatment system.

This research presents a scalable and cost-effective approach for integrating flexible electrodes into BES for industrial wastewater remediation, bridging laboratory research with real-world applications. The findings contribute to the advancement of microbial electrochemical technologies, offering a sustainable solution for energy generation, pollutant degradation, and resource recovery. Future research should focus on further optimizing electrode architecture, microbial community engineering, and long-term system stability, particularly exploring the synergistic role of *Lysinibacillus* and other electroactive microbes to enhance BES performance and commercialization potential.