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Abstract

Petroleum-derived pollutants represent a significant ecological challenge due to their toxicity and the difficulty of their natural degradation processes. Along with the dynamic growth of global demand for fossil fuels, substantial quantities of these substances are released into the atmosphere, water, and soil. Their increasing concentration in the environment necessitates the development of new, more effective removal methods. Currently, three main approaches are used in pollution remediation: physical, chemical, and biological. By leveraging the advantages of microorganisms capable of both catalytically reducing pollutants under anaerobic or microaerophilic conditions, and generating electricity, bioelectrochemical systems may provide an attractive alternative to traditional methods. An additional benefit of these systems is their simultaneous production of electricity and chemical compounds, such as biosurfactants, making them even more appealing in the context of efficient bioremediation. The objective of this doctoral dissertation was to enhance the degradation efficiency of petroleum-based compounds using bioelectrochemical systems in the form of microbial fuel cells.

The literature section of this dissertation includes a review of publications, monographs, legal acts, standards, and patents. It describes issues related to the remediation of petroleum-based compounds, the properties of biosurfactants, their industrial and bioremediation applications, and highlights the key aspects of bioelectrochemical systems. The experimental part consisted of four experiments that utilised different microbial environments as sources of microorganisms capable of degrading petroleum-based compounds and generating electricity.

In the first experiment, external potentials were used to accelerate the growth of microbial communities derived from urban soils contaminated with petroleum-based compounds. For this purpose, a single-chamber bioelectrochemical system with a horizontal air cathode and continuous mixing was designed. Three potential values (-0.3 V, 0.0 V, +0.3 V versus Ag/AgCl) were tested, and their effects on the biofilm development rate were assessed. This experiment demonstrated, for the first time, the positive impact of potential on the performance of bioelectrochemical systems using petroleum compounds as a carbon source. Additionally, the anodic potential was shown to have a strong influence on bacterial community diversity, indicating that the most diverse communities were the most effective in bioelectrochemical conversion of petroleum compounds into electrical energy. The experiment confirmed that a negative potential (-0.3 V versus Ag/AgCl) positively influenced electricity production in bioelectrochemical systems (BES) and promoted the growth of a more biodiverse anodic biofilm.

In the next part of the study, three different anode materials: stainless steel wool, graphite sponge, and carbon fibre veil were compared to analyse their impact on electricity production. The influence of a co-substrate, sodium acetate, and the enzyme laccase on the degradation rate of petroleum components was also investigated. A glass battery reactor containing six anode half-cells was used to ensure identical conditions for the growth of electroactive biofilms on each anode material. The experiment showed that the most energy-efficient anode material was stainless steel wool, which exhibited the highest current density values in each reactor fuelling cycle. Its high efficiency was attributed to its structure, characterized by larger fibres and greater

spaces between them compared to carbon materials. Moreover, the enzyme laccase significantly enhanced the degradation efficiency of petroleum compounds.

The third experiment tested nine microbial consortia from anthropogenic soils, Asian soils, and watercourses for electricity generation and petroleum component degradation. Single-chamber microbial fuel cells with air cathodes were used for this purpose. Two series of measurements were conducted using sodium acetate combined with crude oil or crude oil alone as the carbon source. The experiment showed that soil-derived consortia generated more electricity than watercourse-derived consortia. Furthermore, sodium acetate as a co-substrate positively influenced the growth of electroactive biofilms, contributing to faster adaptation of the consortia and the production of higher power densities. However, in studies on petroleum compound degradation, consortia supplied exclusively with crude oil exhibited higher degradation rates compared to those supplemented with a co-substrate. Based on the results, three microbial consortia were selected for further research.

In the final experiment, three previously selected microbial consortia from a gas station wastewater channel, a petroleum compound separator, and Asian soil from Sri Lanka were tested. Over nine months, the efficiency of electricity production and petroleum compound degradation was evaluated, and potential surfactant-lowering products in the cells were identified. A new type of cell was designed, consisting of one anode chamber and two connected air cathodes. The highest power density (221.68 $mW \cdot m^{-2}$) and coulombic efficiency (55.28 ± 9.01%) were observed for the consortium from the gas station wastewater channel. During the fuelling cycles of each microbial consortium, a reduction in surface tension was noted. Additionally, analyses of the anolytes confirmed the presence of mono- and dirhamnolipids, demonstrating the existence of biosurfactant producers in the studied microbial communities.

The findings presented in this doctoral dissertation provide insights into the application of bioelectrochemical systems in the bioremediation of petroleum-derived pollutants. The studies showed that the processes occurring in these systems can lead to the production of surface-active compounds, such as mono- and dirhamnolipids. The obtained results may serve as a foundation for future research and the implementation of bioelectrochemical systems, contributing to the development of sustainable and effective bioremediation methods.