



Identifying optimized conditions for concurrent electricity production and phosphorus recovery in a mediator-less dual chamber microbial fuel cell

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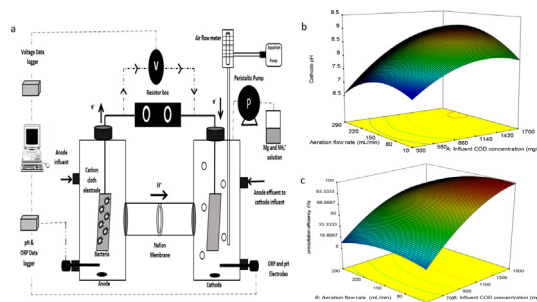


HIGHLIGHTS

- Microbial fuel cell cannot achieve concurrent high-electricity production and phosphorus recovery.
- Optimal predicted values are 184 mW/m² (energy) and 84% (phosphorus recovery).
- Chemical oxygen demand and cathode aeration had great impact on power generation and phosphorus recovery.
- Cathode pH increased with increase in influent chemical oxygen demand, leading to increased phosphorus precipitation.
- High cathode pH (pH > 9) had an impact on purity and size of the struvite crystals.

GRAPHICAL ABSTRACT

(a) Experimental set-up of the dual chamber MFC, (b) Response surface of cathode pH, and (c) Response surface of precipitation efficiency.



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ABSTRACT

Understanding the impact of key operational variables on concurrent electricity production and phosphorus recovery in a microbial fuel cell is required to improve the process and to reduce the operational costs. In this study, a novel mathematical modelling approach, including full factorial design and central composite designs, was employed in a dual-chamber microbial fuel cell to: (i) identify the effect of influent chemical oxygen demand concentration and cathode aeration flow rate on electricity production and phosphorus recovery and (ii) optimise microbial fuel cell power density and phosphorus recovery. Phosphorus was precipitated at the cathode chamber, and the precipitated crystals were verified as struvite using X-ray diffraction and scanning electron microscopy analysis. Response surface methodology showed that influent chemical oxygen demand concentration and cathode aeration flow rate had a joint significant effect on power density, coulombic efficiency, phosphorus precipitation efficiency and phosphorus precipitation rate at the cathode. The effect of varying cathode aeration flow rates on power density and phosphorus recovery was dependent on chemical oxygen demand concentration. Phosphorus precipitation on the cathode electrode was negatively affected by the generated current during batch duration. The response surface mathematical model showed that concurrent high electricity production and high phosphorus recovery cannot be achieved under the same operating conditions; however, operating the microbial fuel cell at high chemical oxygen demand and high cathode aeration flow rate enhanced electricity production and phosphorus recovery. This was confirmed by the experimental results. These findings highlight the importance of operational conditions, such as influent chemical oxygen demand concentration and cathode aeration flow rate, on electricity production and phosphorus recovery.

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1. Introduction

Conventional wastewater treatment is an energy- and resource-intensive process. New, novel and sustainable technologies that are able to generate electricity and, at the same time, recover resources are needed to replace conventional technologies, and these are regarded as the systems of the future. Microbial fuel cells (MFCs) are novel, sustainable systems that convert chemical energy in the organic substrate in wastewater into electrical energy. Besides electricity production, MFCs have been used for wastewater treatment and resource recovery, such as for phosphorus [1,2]. Different physical, chemical and biological approaches, including chemical precipitation, crystallisation, electrochemical and biological processes, have been developed to recover P from wastewater [3–5]. However, most of these approaches are operationally complex and expensive, and they produce low-purity products [6].

In a MFC, P can be recovered exclusively through precipitation because P compounds are not involved in electron transfer via reduction-oxidation (REDOX) reactions [7]. Magnesium ammonium phosphate (MAP) hexahydrate, or struvite, is the most common form of P recovered from wastewater, and it is an efficient slow-release fertiliser. The mechanism of struvite precipitation is highly dependent on the pH of the solution ($\text{pH} > 8$). This precipitation occurs in equimolar concentrations of magnesium (Mg), ammonium (NH_4^+) and P, and these combine with water to form struvite [8]. P recovery as struvite using MFCs involves cathode reactions, whereby water is consumed and hydroxide is generated due to the transfer of electrons from the anode to the cathode. The generated hydroxide leads to increased pH around the cathode, and P starts to precipitate.

The main disadvantage in the MFC's operation is low power output compared with other renewable energies, such as solar and wind power. Therefore, different approaches must be employed to improve the performance of MFCs, for example, modifying the cell design and materials [9], increasing the buffer capacity [10] and modifying the operational conditions that have a significant impact on the system's performance [11]. Operating conditions, such as influent chemical oxygen demand (COD), pH, external load and cathode aeration flow rate, are important factors that influence system performance and microbial communities in MFCs [11]. In particular, influent COD and cathode aeration flow rate have been shown to be important factors limiting electricity production and P recovery in MFCs [1].

Organic matter, measured in terms of COD, plays an important role in providing bacteria with the required carbon source for electricity generation. Increasing the influent COD concentration leads to an increase in the generated voltage. Furthermore, it was found that increasing the COD concentration increases the cathode pH due to the oxygen reduction reaction [12]. In addition, the dissolved oxygen concentration in the cathode chamber, which can be controlled via the cathode aeration flow rate, plays an important role in electricity production, as oxygen acts as an electron acceptor for the transferred electron [13]. However, choosing the optimal aeration flow rate in the cathode compartment can be challenging, especially in dual-chamber MFCs. Operating conditions also influence the purity and size of struvite. Parameters such as pH, molar ratio, mixing energy and aeration rate have an influence on struvite crystallisation [8]. For instance, Durrant et al. [14] showed that high mixing energy could accelerate the nucleation rate, limit crystal growth and increase crystal breakage.

The interplay between the different design and operational variables mentioned above has an impact on the optimal performance of MFCs, and these are just a beginning to be fully understood. This has been noted by several authors, such as Fang et al. [15], who stated that the performance of MFCs is influenced by multiple variables. It has been suggested that a mathematical modelling approach could be a useful alternative for designing and optimising the systems for power generation and wastewater treatment. MFCs are complex hybrid systems involving a number of bioelectrochemical coupling reactions, and

this leads to strong nonlinear characteristics and significant hysteresis properties that make it difficult to directly control and optimise power generation [16]. Consequently, mathematical models are needed to understand the major influencing factors of the whole system, distinguish the main bottlenecks and improve the power generation performance of MFCs [16].

Generally, mathematical modelling can be performed in two ways. The first method is the engineering modelling approach, which is derived from models based on the engineering and physical laws [17]. For instance, a real-time process control system was developed by Tartakovsky et al. [18] to optimise hydrogen production in a microbial electrolysis cell (MEC). The system was able to adjust the energy input by minimising the apparent resistance of the MEC. This approach was efficient to adjust the applied voltage based on the variations in organic load, carbon source properties and hydraulic retention time. However, understanding the effects of these variations on MEC's performance is difficult using this approach.

The second approach is the statistical approach, which was developed using the available experimental data from MFCs. The application of statistical experimental design techniques for the optimisation of MFC operating conditions can result in improved MFC performance, reduced process variability, closer confirmation of the output response to nominal and target requirements, and reduced development time and overall costs. Furthermore, this approach is valuable for characterising the system input-output relationships, especially when there is limited engineering-domain knowledge to characterise the complex mechanisms of electricity generation [19]. This is noted by the authors to have several advantages over the conventional practice of single factor optimisation, which maintains all but one factor at an unspecified constant level and does not depict the combined effect of all factors involved.

Studying the impact of various operational parameters on different responses might be challenging and conflicting. For instance, Yang et al. [20] noted that both experimental and simulation results showed that power output cannot be improved without the sacrifice of the maximal waste removal ratio. In addition, obtaining maximal power output requires degrading the maximal attainable current density. Their findings imply that the objectives of power density, attainable current density and waste removal ratio could conflict, which implies that there is not a single solution that simultaneously optimises each objective. Similarly, concurrent electricity production and P recovery in MFCs have been studied using different cell designs [1,5]. Results from these studies demonstrated that highly defined operating conditions will induce accelerated wastewater treatment rates and enhance energy recovery. In addition, achieving maximum power density and maximum P recovery at the same time might be difficult. Therefore, it is crucial to understand the effects of the different operational conditions on electricity production and P recovery in order to improve the overall performance of MFCs, reduce operational costs and optimise electricity production and P recovery.

Therefore, using statistical optimisation approaches can be helpful for understanding the effects of different parameters within a wide experimental domain with a minimal number of runs. In this way, appropriate operating conditions can be determined, and the corresponding maximum outputs can be obtained. Furthermore, applying a statistical approach, such as central composite design (CCD), helps with identifying the joint effect between various parameters; additionally, the effect of each factor can be evaluated at different levels of the other factors [21]. Different statistical approaches, including full factorial design (FFD), CCD, box-Behnken design (BBD), Plackett-Burman design (PBD), and uniform design (UD), have been used in MFC studies.

Zhang et al. [22] optimized bioelectricity production and sulphate removal in a novel up-flow anaerobic sludge blanket reactor MFC. In this novel study design, the response surface methodology (RSM) approach was used for the first time to optimise sulphate removal. The RSM approach was selected due to the joint effect of the COD/sulphate

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