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Feasibility study on a double chamber microbial fuel cell for nutrient recovery from municipal wastewater

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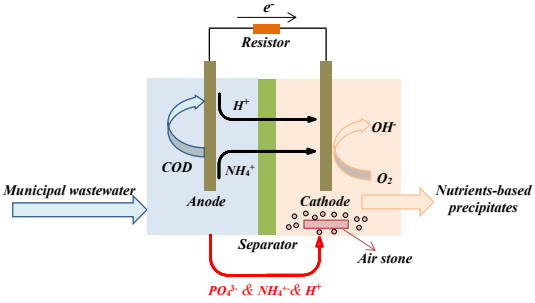
HIGHLIGHTS

- Double chamber MFC can achieve effective nutrient recovery from wastewater.
- Phosphate is removed by microbial absorption and recovered by chemical precipitation.
- Ammonium is accumulated by current generation and recovered as precipitates.
- MFC with CEM as separator is the best option for nutrient recovery.

GRAPHICAL ABSTRACT



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ABSTRACT

Microbial fuel cell (MFC) is currently considered a promising technology for wastewater treatment. This study aims to evaluate the feasibility of a double-chamber MFC in terms of: (i) operating mode (batch mode, self-circulation mode, single-continuous mode) of anolyte on the nutrient accumulation in the catholyte, (ii) aeration conditions (anode effluent with aeration supplied in catholyte; anode effluent without aeration supplied in catholyte; cathode effluent with aeration supplied in catholyte and cathode effluent without aeration supplied in catholyte) on the nutrient recovery and (iii) types of separators (cation exchange membrane (CEM), forward osmosis (FO), and nonwoven (NW)) to remove nutrients toward their recovery from municipal wastewater. Results showed that there was no negligible increase in the phosphate concentration of the catholyte at the three different modes but accumulation of ammonium. At different aeration conditions, nutrients can be recovered by chemical precipitation at high pH generated by the MFC itself. Basically, phosphate was removed by microbial absorption and recovered by chemical precipitation while ammonium was accumulated by current generation and recovered as precipitates. It was found that double-chamber MFC with the CEM as the separator reported the

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best nutrients removal with > 97.58% of NH_4^+-N and > 94.9% of PO_4^{3-}-P removed/recovered, followed by the MFC with the nonwoven and FO membrane, respectively. Thus, the double-chamber MFC is feasible for recovering nutrients in a comprehensive bioelectrochemical system.

1. Introduction

Phosphorus (P) and nitrogen (N) as nutrients are responsible for several serious environmental problems such as eutrophication in water bodies, especially if both are in quantities in the aquatic environment. In water, P and N exist in phosphate and ammonium ions, respectively. It should be noted here that phosphate and ammonium are also essential to plants and crops in agriculture and always used for direct land application in the form of chemical fertilizers. Phosphate as the non-renewable resource is mainly derived from natural phosphate-based rocks, but it has been estimated that the global phosphate deposits will completely run out in 30–300 years [1]. This means the production of phosphate-based fertilizers will end at some point. More importantly, the role of phosphate in the fertilizer production will not be replaced by any other materials [2].

Ammonium utilized for the purposes of fertilizer production is usually achieved through the industrial Haber-Bosch process via the conversion of nitrogen gas in the atmosphere to reactive ammonium [3]. However, this process is energy-intensive and its excessive application may disturb the natural N-cycle and hence cause problems for the environment and people. For this reason, nutrient recovery is more valuable than nutrient removal because it does not only minimize the pollution risk to receiving waters; also it contributes to sustainable resource management [4,5]. Currently, the concept of municipal wastewater and its treatment has shifted from a human health concern and environmental hazard to one where it is considered to be an untapped source. Firstly, it contains a large amount of valuable components such as nutrients (phosphate and ammonium ions), heavy metals, fresh water and chemical energy stored in the organics. Secondly, it exists in large quantities [6].

Understandably, recovering nutrients from municipal wastewater is a promising research approach with a practical benefit. Several methods including adsorption, chemical precipitation and biological processes have been developed or are still in the development stage for nutrients recovery from municipal wastewater [7–10]. For example, biological processes such as an enhanced biological phosphorus removal system can efficiently recover nutrients in the form of excessive sludge. However, the surplus sludge contains toxic substances and pathogens [11], so this method has been banned for direct land application in some European countries such as Switzerland [12].

Even though the effective recovery of N and P could be obtained via adsorption, the desorption of nutrient-loaded adsorbents may be required to achieve the desired recovered nutrients. As for nutrients recovery through chemical precipitation, this has been deemed one of the most promising methods due to its high stability and efficiency. In this scenario, magnesium and calcium materials are usually utilized as the precipitator. Struvite ($MgNH_4PO_4{\cdot}6H_2O$) employed in stoichiometric precipitation of magnesium, ammonium and phosphate has proved to be an efficient fertilizer when directly applied in agriculture [13–17]. Similarly, calcium can react with phosphate to form hydroxyapatite ($Ca_5[OH][PO_4]_3$, HAP) which also works as an efficient fertilizer supplement. The reaction between nutrient and magnesium/calcium ions could be described as follows:

$$Mg^{2+} + PO_4^{3-} + NH_4^+ + 6H_2O \rightarrow MgNH_4PO_4{\cdot}6H_2O\downarrow \quad (1)$$

$$5Ca^{2+} + 3PO_4^{3-} + 3OH^- \rightarrow Ca_5(OH)(PO_4)_3\downarrow \quad (2)$$

The chemical precipitation requires high pH (> 8), for which the pH value is the most important influencing factor. More importantly, the chemicals added to increase the pH, could represent a high percentage of the cost of the nutrient recovery process [18–20].

Microbial fuel cell (MFC) is currently considered a promising technology in the wastewater treatment due to its production of electricity and wastewater purification [21,22]. Typically, the MFC has an anode chamber and a cathode chamber, respectively, and a cation-exchange membrane (CEM) is installed to separate the two chambers. In the anode chamber, the anaerobic microorganisms are catalysts that convert chemical energy stored in organics directly into electricity [23,24]. MFC can use sewage sludge as a substrate, which is considered a kind of biomass [25,26]. Recently, nutrients recovery through the MFC process has attracted much attention [4,5,8] since the pH value of electrolyte can be raised without adding alkaline chemicals [27,28]. In this scenario, the costs of pH adjustment can be reduced. Furthermore, the energy recovered in the MFC may be used for its own operation and maintenance, which indicates that the MFC may be a neutral/positive energy balance system.

The single- and double-chamber MFCs constitute the most common types of MFC. Recently, the single-chamber MFC with an air-cathode electrode and multi-chamber MFC coupled with CEMs and/or anion-exchange membranes (AEMs) have been developed for recovering nutrients from wastewater [2,29–31]. For example, Chen et al. [29] proposed a new MFC that simultaneously uses CEMs and AEMs to recover nutrients and purify wastewater. In their study, wastewater was circulated between the anode and cathode chambers and this particular configuration facilitated nutrient accumulation. According to their findings, the concentrations of ammonium and phosphate ions were condensed to 1.5 and 2.2 of their initial concentrations. This caused the removal of > 64% of PO_4^{3-}-P and 96% of NH_4^+-N.

Even though the single-chamber MFC could reduce the overall operating costs to some extent, integrating the anolyte and catholyte may buffer the pH of electrolyte and thus inhibit the pH elevation of the electrolyte. Referring to the multi-chamber MFC, using several chambers may increase the maintenance costs and operation complexity. However, a very few articles have studied on nutrient recovery using a conventional double-chamber MFC.

This paper investigated the feasibility of the conventional double-chamber MFC for nutrient recovery from municipal wastewater. The concentrations of NH_4^+-N, PO_4^{3-}-P and COD along the treatment process and the recovery process were evaluated. Furthermore, the sediment deposits in the cathode chamber were analysed by energy dispersive spectroscopy (EDS) and scanning electron microscopy (SEM). More importantly, a new design of double-chamber MFC with the forward osmosis (FO) membrane or nonwoven as the separator was also evaluated.

2. Materials and method

2.1. MFC construction

The double-chamber MFC was made of organic glass (i.e., poly [methyl methacrylate]) and consisted of an anode chamber and a cathode chamber in a rectangular form. The effective liquid volumes of each chamber were both 350 mL. A CEM (CMI7000, Membranes International Inc., USA) was used to separate the anode chamber and cathode chamber. A cylinder-shaped graphite felt (Sanye Carbon Co. Ltd., Beijing, China) 3 cm in diameter and 0.6 cm in thickness served as the anode electrode in the double-chamber MFC to hold the electrically active biofilms. The cathode electrode was made of a carbon-fiber brush (~3 cm in length and ~3 cm in diameter) coated with a titanium bar. The anode electrode and cathode electrode were placed on opposite sides of the CEM and connected by a copper wire. Moreover the

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