



Concurrent performance improvement and biofouling mitigation in osmotic microbial fuel cells using a silver nanoparticle-polydopamine coated forward osmosis membrane



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ABSTRACT

Osmotic microbial fuel cells (OsMFCs) have demonstrated superior electricity production and effluent quality compared to conventional MFCs having ion exchange membranes. However, according to previous OsMFCs studies, inevitable membrane biofouling led to a severe water flux decline during operation. Here, we demonstrated a substantial reduction in biofouling as well as further improvement in current generation by applying a forward osmosis (FO) membrane modified with silver nanoparticles (nAg) that were deposited on a polydopamine (pDA) coated membrane surface. The OsMFC incorporating an nAg-pDA coated FO membrane exhibited less flux decline (22%) than the OsMFC with a pristine membrane (50%) due to an antibacterial ability of nAg. Additionally, the nAg-pDA coated membrane also decreased the internal resistances of the OsMFC by 35% owing to the enhanced hydrophilicity of the membrane surface, resulting in an improvement in power density (12%). The nAg-pDA coated layer was very thin (c.a. 1.06% of total membrane thickness), so caused no adverse flux decline associated with thickened membrane thickness entailed from the nAg-pDA coating. Instead, it can simultaneously accomplish a reduction in membrane biofouling and an increase in electricity generation in an OsMFC.

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1. Introduction

Microbial fuel cell (MFC) technologies have been regarded as promising approaches for wastewater treatment because they can produce renewable energy (e.g. electricity) from organic matter in wastewater [1,2]. However, the spectra of organic compounds that can be effectively degraded by MFCs are quite limited [3,4], and as such clean water production from wastewater cannot be successfully accomplished using MFCs alone. Recently, in attempts to support and improve the wastewater treatment capability of MFCs, a forward osmosis (FO) process, i.e., a membrane separation technology driven by the osmotic pressure difference between low (feed) and high (draw) concentration solutions across a semi-permeable membrane, has been retrofitted into a MFC, that is referred to as a osmotic microbial fuel cell (OsMFC) [5]. OsMFCs, which have a semipermeable membrane as a separator between anode and cathode chambers, have subsequently achieved water

recovery from wastewater in the anode chamber by using osmotic pressure as the FO process, and concurrently accomplished electricity production from organic matter in wastewater by using electrochemically active bacteria [5–8]. Zhang et al. [5] reported an OsMFC achieved a higher maximum power density (4.74 W/m³) with a water flux of about 3.0 LMH than a conventional MFC equipped with a cation exchange membrane (3.48 W/m³) when used 58 g/L NaCl solution as a catholyte. Ge and He [6] improved a current generation of an OsMFC with NaCl catholyte by adding a small amount of HCl solution, and a water flux of the OsMFC was 2–3 LMH. Ge et al. [7] also reported that an OsMFC treating real wastewater as an anolyte generated a maximum power density of 4.5 W/m³ and achieved a water flux of about 1.06–2.15 LMH. Werner et al. [8] reported that an air-cathode OsMFC obtained a higher maximum power density of 43 W/m³ than a conventional MFC (23 W/m³).

However, it was found that unavoidable FO membrane fouling significantly declined the water flux [6,8]. According to Ge and He [6], fouling reduced water flux performance of FO membranes in OsMFCs. Even osmotic backwashing used to improve water flux recovery failed to efficiently relieve membrane fouling [6]. Werner

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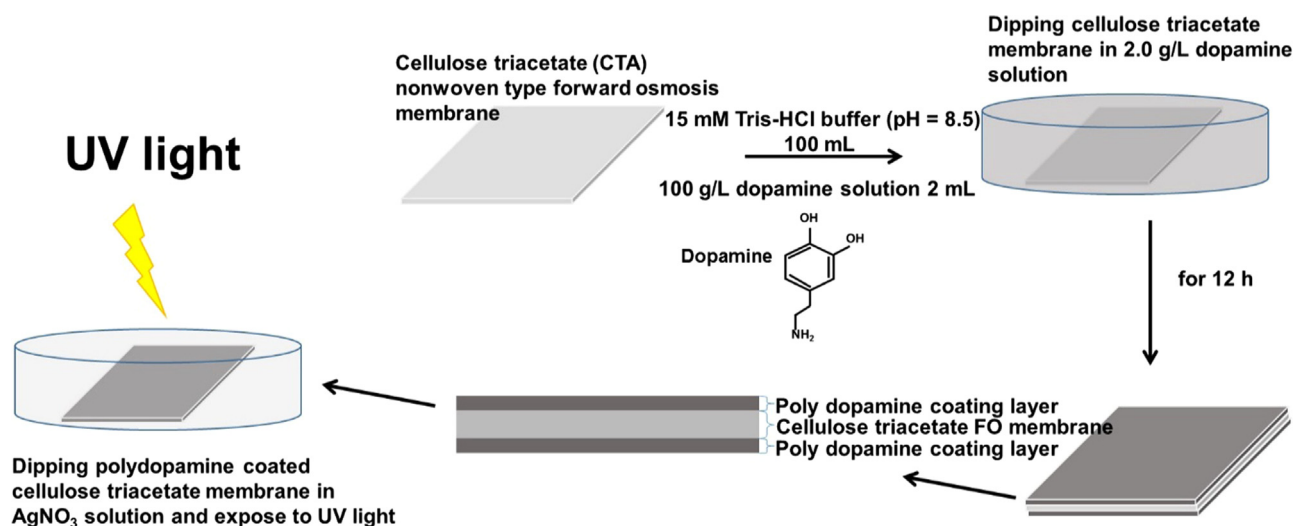


Fig. 1. Procedure for applying an nAg-pDA coating onto an FO membrane.

et al. [8] also reported that an initial water flux declined by 28% over three batch cycles due to biofouling in an OsMFC. In addition, for conventional MFCs, membrane fouling degenerated the performance of the MFCs by limiting diffusion of cations [9,10]; those reports suggest that strategies to alleviate membrane fouling are required in order to develop practically feasible OsMFC.

Silver nanoparticles (nAg) have been widely utilized as anti-microbial agents in recent years, although the exact anti-microbial mechanisms of nAg have yet to be fully understood [11,12]. Previous research has shown that nAg has a strong biocidal effect on a broad spectrum of microorganisms, but low cytotoxicity [13–15], and that nAg has been immobilized on various material surfaces where it has successfully performed antimicrobial activities [16–18]. To immobilize nAg on substrates, several coating methods have been employed, including the hydrothermal method [19], ion-beam assisted deposition process [20], in situ interfacial polymerization [17], electroless plating [21], anodic electrochemical technique [22], sol-gel method [23], and layer-by-layer deposition method [24]. However, each of these methods has inherent vulnerabilities, such as the lack of adhesion between nAg and substrates, silver ions being released from the nAg coating layer, and complicated coating procedures [25]. Hence, simpler and more practical methods for nAg coating need to be developed.

Typically, nAg has been simply deposited on the surfaces of diverse substrates by using the metal ion reduction ability of catechols in the polydopamine (pDA) coating layer [25,26], which is easily formed on any surfaces by the self-polymerization of dopamine in oxygenated weak alkaline aqueous conditions [26,27]. pDA is a polymerized catechol amine inspired by the adhesive foot proteins of mussels that are used to cling to wet surfaces [27]. In addition, this pDA coating layer has the versatile capability of not only being able to bind metal nanoparticles, but can also enhance the hydrophilicity of matrix surfaces [28]. It was shown that hydrophilicity of pDA-modified membrane surfaces (e.g., micro-filtration, ultrafiltration, and reverse osmosis membranes) was increased, and that their fouling resistances consequently was improved [29]. In other studies, the power capabilities of electrochemical devices, such as a lithium ion battery and a flow-through type MFC, were improved by making the hydrophobic surfaces of a polyethylene separator and an ultrafiltration membrane more hydrophilic via the application of a pDA coating [30,31].

We herein adopt a simple FO membrane surface modification by using an nAg-pDA coating as a strategy for simultaneous bio-fouling control and electricity production enhancement in an

OsMFC. This surface modification was conducted using a simple two-step procedure. First, a pDA coated FO membrane was prepared by dipping a virgin FO membrane into a mild alkaline dopamine solution (pH 8.5). Next, the nAg particles on the pDA coated FO membrane were immobilized by immersing the membrane into a silver nitrate solution. The physicochemical characteristics of the nAg-pDA coated FO membrane were then investigated, and also intrinsic separation properties, such as pure water permeability, salt rejection, and osmotic water flux, were examined in lab-scale membrane test cells. Finally, an OsMFC equipped with an nAg-pDA coated FO membrane was operated in order to evaluate the effects of the nAg-pDA coating on biofouling formation and the performance (e.g., current generation and water flux) of an OsMFC.

2. Materials and methods

2.1. Silver nanoparticle-polydopamine (nAg-pDA) coating on FO membrane

An nAg-pDA coated FO membrane was prepared as shown in Fig. 1. A cellulose triacetate nonwoven type (CTA NW) FO membrane (Hydration Technology Innovations, LCC, USA) was purchased. Next, 100 g of dopamine hydrochloride (Sigma-Aldrich, USA) was dissolved in 1 L of deionized water to prepare a 100 g/L dopamine stock solution. Then, 2 mL of the dopamine stock solution (100 g/L) was mixed with 100 mL of a Tris-HCl buffer solution (15 mM, pH 8.5) to prepare a 2 g/L of dopamine solution (pH 8.5) for use in the dopamine polymerization process under alkaline pH conditions [31]. The prepared FO membrane was fully immersed into the dopamine solution (2.0 g/L, pH 8.5) in a plastic container for 12 h. During the dopamine polymerization process, this plastic container was mildly shaken using a shaking plate to supply oxygen into the dopamine solution. To immobilize the nAg on the surface of the pDA coated FO membrane, the membrane was immersed in a 0.05 M AgNO₃ solution and exposed to UV light [25].

2.2. FO membrane characterization

To identify the effect of nAg-pDA coating on the FO membrane properties, a virgin FO membrane and the nAg-pDA coated FO membrane were characterized in terms of morphological observation, surface charge, surface hydrophilicity, electrical

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