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Communication

Development of Microbial Fuel Cell Using Onion Skin as Disposable Membrane

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Abstract

Microbial fuel cells (MFCs) are promising technologies for generating bioelectricity from organic waste through microbial catalysis. However, the high cost of membranes used in MFCs remains one of the major concerns limiting their widespread application. In this study, a low-cost and readily available bio-membrane fabricated from onion skin was developed and applied as the membrane component in MFCs. By using Shewanella oneidensis MR-1 as the model exoeletrogen, the onion skin membrane equipped MFC delivered even higher bioelectricity output (~133 mW/m^2) compared to that equipped with the costly Nafion membrane (~74 mW/m^2). This research demostrated that it is possible to use onion membrane as a cost-effective alternative to Nafion membrane for bioelectrochemical systems.

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1. Introduction

Microbial fuel cell (MFC) is an innovative device in which the chemical energy stored in the organic waste can be efficiently converted to bioelectricity with the assist of the metabolic catalysis by microorganisms (Lovley 2006, Zhao, Slade et al. 2009, Yong, Dong et al. 2012, Sun, Peter Kingori et al. 2015). Thus, MFC is considered to be a promising technology for energy-saving wastewater treatment (Yong, Yu *et al*. 2011, Tao, Wei *et al*. 2013, Xu 2013, Yong, Yu *et al*. 2013, Yong, Yu *et al.* 2014). More impressively, series of important and interesting applications derived from MFC have been developed in recent years, which including microbial electrolysis cells for efficient H_2 production (Cheng and Logan 2007), microbial electrosynthesis cells for CO_2 upgrading (Lovley and Nevin 2013), bioelectrochemical biosensors for wastewater monitoring (Lovley and Nevin 2013),etc. However, application is limited as materials of MFC are expensive, in particularly the proton/cation exchange membrane.

Nafion is the most widely used proton exchange membrane (Daud, Kim *et al*. 2015) however it has serious drawback in its applications for its high cost of production and hence high market price which normally goes for about $55 per 100cm^2 (Tao, Sun *et al*. 2015). Researchers have recently developed alternative and cost effective material with the aim of increasing the economic feasibility of MFC by reducing the capital investment. Some of the membranes developed are Sulfonated poly-ether-ether-ketone (SPEEK) which shares a similar structure with Nafion, where ionic clusters co-exist with hydrophobic domains of the polymer backbones, Sulfonated polystyrene-ethylene-butylene-polystyrene(PSEBS)(Ayyaru, Letchoumanane et al. 2012) Ultrex CMI7000 made by membranes Inc. USA) and Hyflon Ion made by Solvay-solexis SPA. Ultrex CMI7000 is a strong, acid polymer membrane with a gel polystyrene backbone structure cross-linked with divinyl benzene containing a large amount of sulphonate groups(Ismail and Jaeel 2013) hyflon Ion is a copolymer of tetra fluoroethylene (TFE) and a short-side-side(SSC) perfluoroosulfonylfluoridevinyl ether(Arcella, Ghielmi *et al*. 2003). Porous separator materials that enable non-ion selective charge transfer have been used such as J-cloth(Fan, Hu *et al*. 2007) natural rubber(Winfield, Ieropoulos *et al*. 2013, Winfield, Chambers *et al*. 2014) earthen pot(Behera, Jana *et al*. 2010) and cellulose nanocrystals (CNC)(Rhim,

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Reddy *et al*. 2015). Another novel membrane that has been developed and exhibits voltage output is titanium dioxide (TiO2)-quaternised poly(vinyl alcohol) (QAPVA) hybrid anion membrane (Tao, Sun *et al*. 2015).

The food industry produces a significant amount of agricultural wastes, making it necessary to search for possible ways for their utilization, one of the major food crops is onions, it is estimated that more than 500,000 tonnes of onion waste are discarded every year within the European Union(Roldán, Sánchez-Moreno *et al*. 2008). Onion skin contains a high amount of bioaccessible and bioavailable compounds with well documented biological activity(Benítez, Mollá *et al*. 2011).One way could be to use these onion wastes as a natural source of high-value functional ingredients since onions are rich in several groups of compounds, which have perceived benefits to human health(Gawlik-Dziki, Kaszuba *et al*. 2015). Various research has noted that due to strong aroma of onion waste it is unsuitable for use in animal fodder, due to phytopathogens, it is not suitable for landfill disposal (Salak, Daneshvar *et al*. 2013) . Other research has found that the brown skin and top–bottom could be potentially used as functional ingredients rich in dietary fibre. Brown skin has a high concentration of quercetin aglycone and calcium (Benítez, Mollá *et al*. 2011) other research has shown onion skin as biosorbent which is effective in removal of methylene blue dye from contaminated waters (Saka and Sahin 2011) there has also been use of onion skins for development of sugars and quercetin by- products (Choi, Cho *et a*l. 2015) the onion skins have also recently been evaluated as raw materials for the enzymatic production of pectic oligosaccharides (Babbar, Baldassarre *et al.* 2016).This study aimed at assessing the feasibility of onion skins (*Allium cepa* L.) for application in the microbial fuel cells as a cost-effective membrane.

2. Materials and Methods

2.1. Chemicals and Materials

All chemicals with analytical grade were purchased from Sinopharm Chemical Reagent Co. Ltd (Shanghai, China) and directly used without further purification. Nafion proton exchange membrane (Nafion 117, thickness of 117 μm) was purchased from DuPont (). Red onion were brought from a local market, the fresh skin was collected and washed three times with distilled water. The washed onion skin was then dried under 60 °C for 24 hours and then cut to 3*4 cm (the same size of Nafion membrane used in this study).

2.2. Bacteria and Culture Conditions

Shewanella oneidensis MR-1 was routinely cultured in LB broth (peptone 10 g/L, yeast extract 5 g/L, NaCl 5 g/L, pH 7.0) with shaking 150 rpm at 30 °C as described elsewhere ().The bacteria cells were then harvested by centrifugation and re-suspended in mixed medium containing M9 salt medium (Tian et al. 2002), 5% LB broth and 10 mM sodium lactate to an optical density of 1.5 (OD600). The bacteria suspension was further bubbled with nitrogen and employed as anodic electrolyte.

2.3. Membrane Characterization

Fourier transformation infrared (FT-IR) spectra of the samples obtained between 400 and 4000 cm^{-1} with AVATAR 360 FTIR (Madison, USA). The morphology of the prepared onion membrane was characterized with Thermal Field Emission SEM JSM-7001F (Joel, Japan).

2.4. MFC Set-Up and Analysis

A dual chamber MFC (inner size of 4 cm × 4 cm × 2 cm for both chamber) separated with onion skin membrane or Nafion 117 membrane was used in this experiment. Carbon cloth (2cm×3cm) was used for cathode and (1cm×2cm) for anode electrode. The anodic chamber was filled with the bacterial suspension electrolyte prepared above. The cathode chamber was filled with 0.05mol/L K3 [Fe (CN) 6] and KCl solution. The MFC operation started when a 2 kΩ external resistor was connected

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between the anode and the cathode. Operating temperature was maintained at 30⁰C. The voltage (V) across the external resistor (R) was monitored by a digital multimeter.(Zheng, Xu et al. 2015). The power output and polarization curves were obtained by varying the external resistor when the current output reached steady-state. The experiment was done in three triplicates, and the error bar for the power output in voltage was generated using Statistical software, notably SPSS.

3. Results and Discussion

3.1. Preparation and Characterization of Onion Skin Membrane

To apply the onion membrane for MFC utilization, fresh onion skin was peeled off and washed three times with DI water to remove possible contaminants. After that, the onion skin was dried at 60 °C for 24 hours and then cut to 4×3 cm (Figure 1). During the fabrication process, a major concern is the possible damage to the membrane. Optical microscopy was performed to check the membrane and make sure the intact membrane without damage and defect was selected for further characterization and application. The SEM images showed that the dried onion membrane has dense, compact, and smooth surface (Figure 2a). The surface of the onion skin is characterized as hydrophobic membrane .The dense structure and hydrophobic nature of the onion skin surface might be helpful to prevent diffusion of water or large molecules and avoid short-circuiting. From the SEM image from the cross-section of the onion skin membrane (Figure 2b), it is assembled layer-by-layer with the thickness of about 50-100 μm, which is comparable or even thinner than the commercially available Nafion membrane .Thinner membrane might result in faster proton diffusion and lower the internal resistance of the MFC, which might be beneficial to get higher output from MFC.

Figure Legends

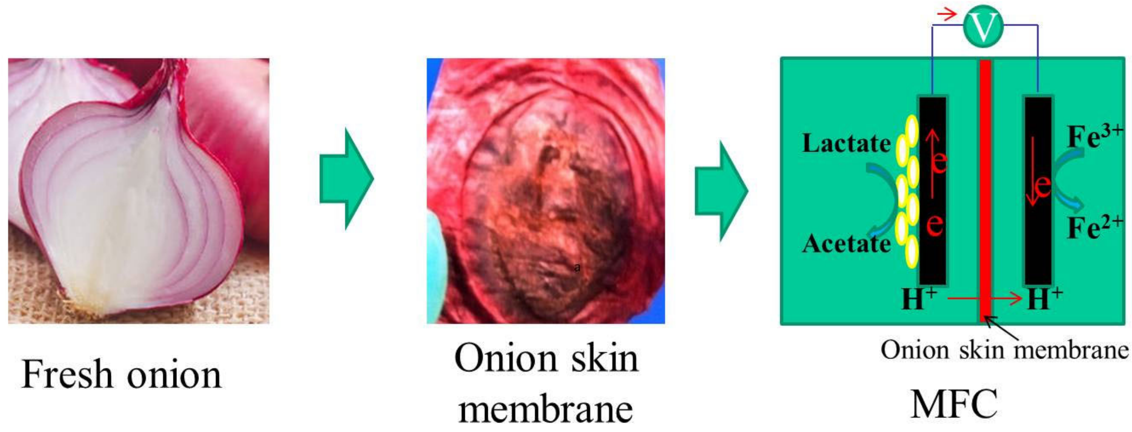

Figure 1. Photograph of onin skin, prepared onion skin membrane and the prototype of onion skin membrane equiped MFC.

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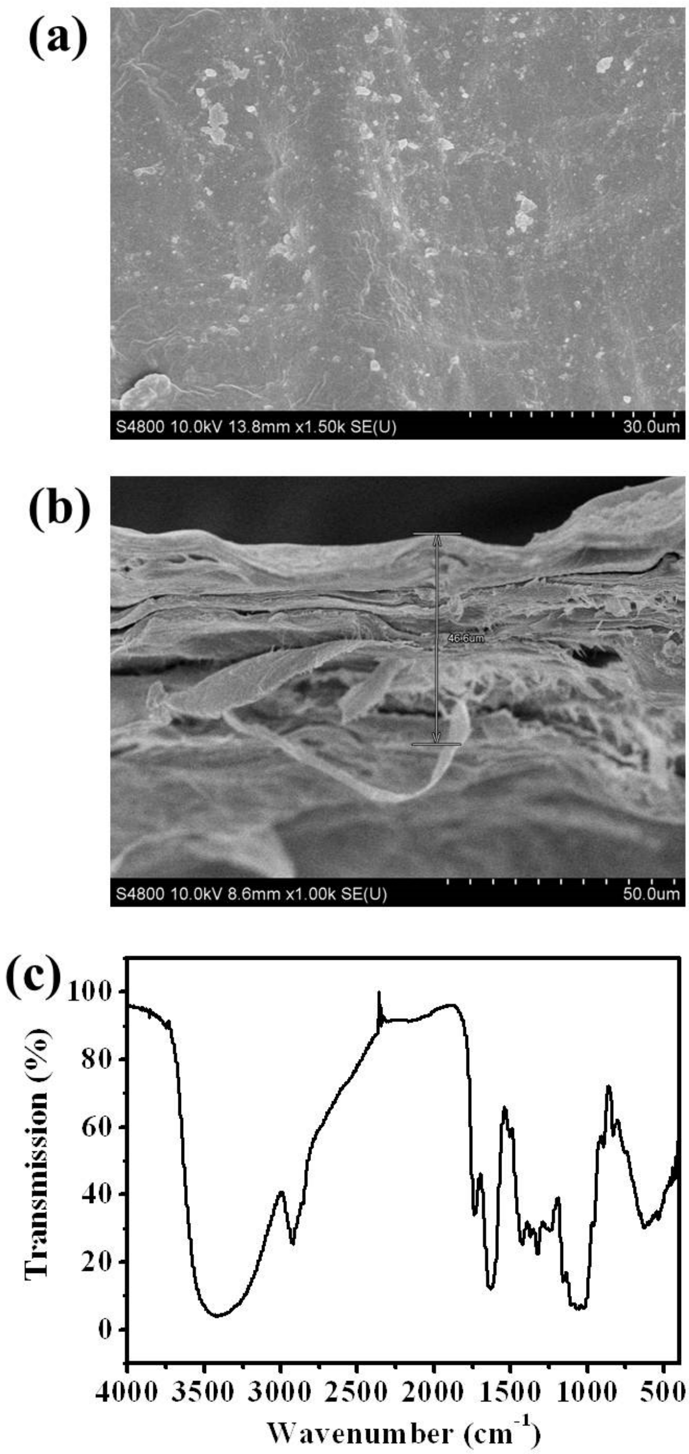

Figure 2. SEM images of the prepared onion skin membrane (a) top view and (b) view of cross section. (c) FTIR spectrum of the onion skin membrane.

Alliums species contains various sulphur compounds such as thiosulfinates compounds (Lanzotti, Bonanomi *et al*. 2013, Lanzotti, Scala *et al*. 2014) that are known to have high proton affinities might attribute to proton exchange (Block, Dane *et al*. 2010, Kubec, Cody *et al.* 2010). Thus, the functional groups of the membrane were characterized with FTIR. According to the FTIR spectrum (Figure 2c), the onion membrane showed various bands, the broad absorption band 3382 cm^2 is characteristic of the stretching vibration of hydrogen bonded hydroxyl groups, the bands at 2918 cm^2 which arise due to aliphatic C-H stretching in an aromatic methoxyl group. Other band around 1620 cm^2 were observed and are normally as a result of C=C vibrations in aromatic rings, C-O 1326 cm^2.

More impressively, the band with high adsorption at about 530 cm^{-1}, 630 cm^{-1} and 1100 cm^{-1} might attribute to the characteristic peaks of sulfo-group. Sulfo-group was characterized its high proton exchange ability and was usually used as the functional groups for proton exchange membrane . Thus, the existence of the sulfonyl groups might provide the onion membrane with proton exchange capability and thus might be applied for MFC.

3.2. Performance of MFC with Onion Skin Membrane

To test the possible application of onion skin membrane in MFC, *S. oneidensis* MR-1 was selected as the model exoeletrogen and a dual chamber MFC was used. As shown in Figure 3, both of the MFCs equipped with Nafion 117 or onion skin membrane showed increased voltage output with similar trends after the bacteria inoculation at 0 h. The voltage outputs increased to their maximum value at about 30 h after bacteria inoculation and then decreased to baseline level at about 42 h due to substrate depletion. More interestingly, the highest voltage output obtained by the MFC with onion skin membrane is about 0.21 V, which is about 60% higher than that obtained by the MFC with Nafion 117 (0.13 V). The results substantiated that the onion skin membrane could act as the membrane for MFC.

Maximum power density output was examined with power density curves. It was observed an increase in power density with a decrease in applied external resistance until a peak obtained (Figure 4). In good agreement with the voltage output, the MFC with onion skin membrane also showed higher (84% higher) power density output than that equipped with Nafion membrane. The MFC with onion skin membrane s was impressive with a power density of 134±1.9 mW/m^2, while the MFC with Nafion 117 was 73±3 mW/m^2.

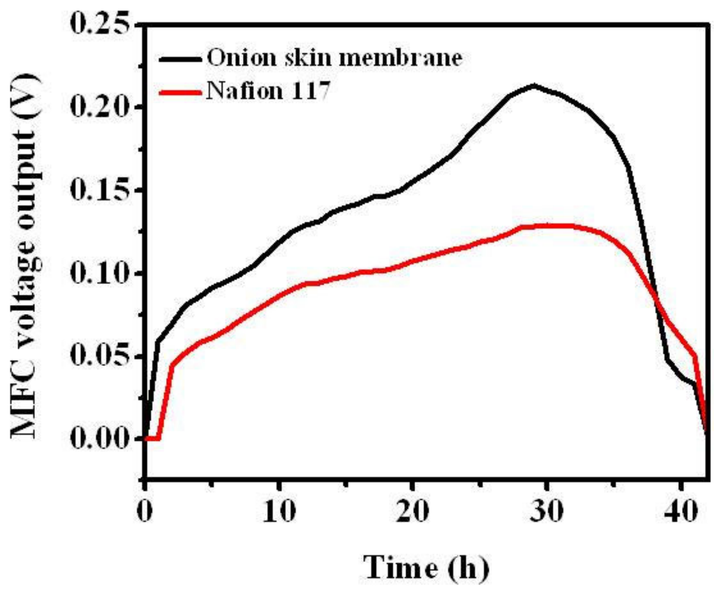

Figure 3. Voltage output of MFCs equiped with onion membrane or Nafion 117.

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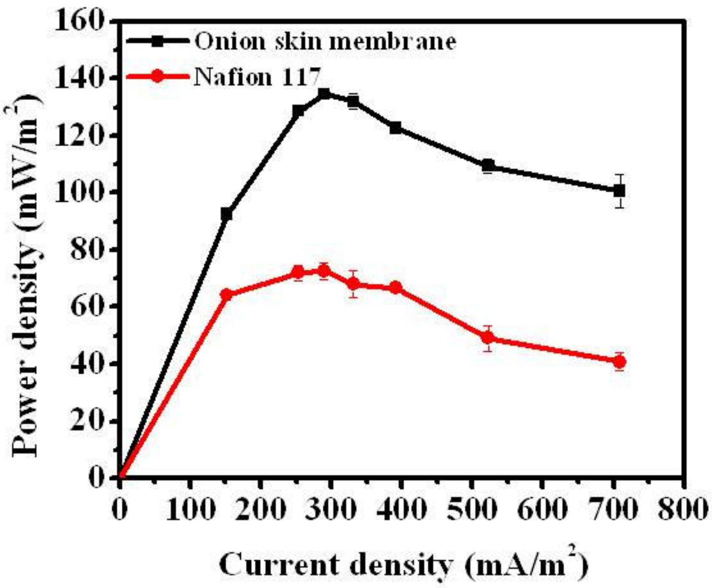

Figure 4. Power density output of MFCs equiped with onion membrane or Nafion 117.

As onion skin is biodegradable biomaterial, biodegradation generated pores might help proton transfer. However, long-term degradation might result in marco-size holes and hence short-circuiting. The long-term performance of the onion membrane was also tested. It was found that while the onion membrane showed superior to Nafion 117 in ~120 hours' operation (~3 cycles of MFC operation), the function of the membrane was gradually lost due to excessive deteroration after 120 h. However, due to the advantages of low cost and enviroment compability, it can be used for short-term, disposable devices such as biosensor applications derived from MFC.

4. Conclusions

This work demonstrated that it is possible to employ onion skin membrane with simple pretreatment as membrane of MFC. It was found that the performance of MFC with onion skin membrane was even higher than that with the commercial Nafion membrane. Although this bio-membrane could not be used for long-term operation, the low cost and good environment compability guaranteed it holds great promise in short-term, disposable applications.

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