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Short communication

A novel stainless steel mesh/cobalt oxide hybrid electrode for efficient catalysis of oxygen reduction in a microbial fuel cell



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ABSTRACT

To explore efficient and cost-effective cathode material for microbial fuel cells (MFCs), the present study fabricates a new type of binder-free gas diffusion electrode made of cobalt oxide (Co₃O₄) micro-particles directly grown on stainless steel mesh (SSM) by using an ammonia-evaporation-induced method. In various electrochemical analyses and evaluations in batch-fed dual-chamber MFCs, the SSM/Co₃O₄ hybrid electrode demonstrates improved performances in terms of electrocatalytic activity, selectivity, durability and economics toward oxygen reduction reaction (ORR) in pH-neutral solution, in comparison with conventional carbon supported platinum catalyst. This study suggests a new strategy to fabricate a more effective electrode for ORR in MFCs, making it more technically and economically viable to produce electrical energy from organic materials for practical applications.

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1. Introduction

Recently, a microbial fuel cell (MFC) has drawn a dramatically increasing attention because it shows great promise for potential in recovering electrical energy from organic materials (Logan et al., 2006). Nevertheless, several limiting factors may present great challenge toward engineered and commercialized applications of MFC technology. In particular, the high power production cost is the key when measuring the balance between power recovery and energy input in an MFC system for wastewater treatment. A gas diffusion electrode (GDE) has been widely accepted as the cathode of MFC due to the availability of 21% gaseous oxygen in air used as a free electron acceptor. Many studies showed that the oxygen reduction reaction (ORR) at cathode dominated the MFC performance in terms of power density production and investment of materials (Birry et al., 2011; Harnisch et al., 2009; Rismani-Yazdi et al., 2008). Specifically, the cost of cathode material (*i.e.* support, catalyst, binder, PTFE, and carbon black) accounts for more than 60% of overall investment (You et al., 2011). Thus, if it were feasible to develop lower-cost cathode materials and structure without losing power density, one would expect remarkably improved MFC performances.

Stainless steel mesh (SSM) has been successfully used as electrically conductive, chemically stable and cost-effective support to make GDE for cathode of MFC applications (Wang et al., 2013). Initially, Zhang et al. (2010) and You et al. (2011) fabricated their SSM-based GDEs by directly coating Pt/C catalyst powders onto the prepared SSM electrode using a Nafion (5%) binder. Despite desirable power density produced and decreased cost, the Pt/C and Nafion binder are still quite expensive for scaling up the MFC system. Besides, the sulfonic ionomer may diminish interfacial proton transfer at the three-phase boundary layer, and thus prevent further increase of power production. To mitigate this limitation, Chen et al. (2012) created a method to grow a nitrogen-doped carbon nanotube on the SSM support; thereby the GDE was produced without the use of Nafion binder. Yet such synthetic process needs to be operated at high temperature, which not only accounts for intensive energy consumption but also appears quite difficult for tunable preparation of the electrocatalytic electrode. Some inexpensive metal oxides demonstrate the possibility of replacing Pt for increased ORR activity (Morris et al., 2007; Roche and Scott, 2009). For example, Co₃O₄ micro- or nano-particles have been reported which are capable of catalyzing ORR with activity comparable to that of Pt catalyst (Liang et al., 2011; Xu et al., 2012). Moreover, the oxidized SSM possesses abundant oxygen-containing groups on the surface, which provides a large number of active sites for grafting metal oxides to form active interface. This is expected to offer a new strategy to fabricate electrochemically active and cost-effective binder-free metal oxides/ SSM electrode for the possibly improved ORR process in MFC.

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In this study, we used a simple ammonia-evaporation-induced method to directly grow Co_3O_4 particles on the SSM at mild condition; thereby tunable preparation of a new kind of the so-called SSM/ Co_3O_4 hybrid electrode could be realized. First, the morphological observation of the electrode was performed by using scanning electron microscopy (SEM), followed by identification of crystalline structure of Co_3O_4 using X-ray diffraction (XRD) technology. Second, the catalytic activity and selectivity of the electrode toward ORR were examined using electrochemical methods at pH-neutral electrolyte. Last, the ORR performance of the electrode was evaluated in a batch-fed dual-chamber MFC reactor.

2. Materials and methods

2.1. Preparation and characterization

SSM supported cobalt oxide catalysts were prepared by a mild template-free method known as ammonia-evaporation-induced method (Marbán et al., 2008, 2010). In brief, 10 mmol $Co(NO_3)_2$ and 5 mmol NH₄NO₃ were dissolved in 35 mL of H₂O and 15 mL of 30 wt% ammonia solution. The homogeneous solution was magnetically stirred for half an hour in air. Then SSM washed previously in HNO₃ (4.0 M) at 60 °C for 4 h was introduced into the reaction solution. The vessel was heated in an oven at 90 °C for 18 h so that cobalt oxide could grow. After the synthesis, the SSM was taken out of the solution, thoroughly washed with deionized water and vacuum-dried at 60 °C. Finally, the samples were calcined in air at 300 °C for 2 h.

After cobalt oxide was grown on the SSM, a four-layer gas diffusion layer was prepared by brushing 60% Teflon[®] emulsion on the other side of the SSM (*i.e.* SSM/Co₃O₄ hybrid electrode). The Pt/C and Co₃O₄ particles based cathodes were prepared by mixing with hydrophilic Nafion ionomer (5%) and uniformly dispersed onto the

surface of SSM gas diffusion cathode, denoted as SSM–Nafion–Pt/C and SSM–Nafion– Co_3O_4 , respectively.

X-ray diffraction (XRD) analysis was conducted on an X-ray diffractometer (Bruke D8 Adv., Germany) using Cu K α radiation (λ =0.15406 nm) at a power of 40 keV × 30 mA. Scanning electron microscopy (SEM) images were obtained using a field emission scanning electron microscope (Guanta 200F, FEI, U.S.).

2.2. Electrochemical measurements

The electrochemical measurements were performed in a conventional three-electrode electrochemical cell containing 50 mM neutral phosphate buffer solution (PBS) saturated with gaseous oxygen at room temperature (25 °C). Line scan voltammetry (LSV) and electrochemistry impedance spectroscopy (EIS) measurements were performed on a dual-working-electrode electrochemical system (CHI750D, Chenhua Co. Ltd., China) using the saturated calomel electrode (SCE) as reference electrode and a platinum plate (1.0 cm²) as counter-electrode. The rotating disk electrode (RDE) measurement was performed on an RRDE-3A apparatus (BAS Inc., Japan) by stepwise variation of rotating speed (ω) from 400 rpm to 3600 rpm and the rotating-ring disk electrode (RRDE) measurement was tested at a rotating rate of 1600 rpm. The Pt ring electrode potential was set at 0.4 V for the oxidation of hydrogen peroxide ions.

Koutechy–Levich plots were derived from RDE data to calculate electron-transfer number (*n*) in the oxygen reduction process at different electrode potentials from the slope of fitting line $(i^{-1} vs \omega^{-1/2})$ according to the Koutechy–Levich equation (Gong et al., 2013; Kim et al., 2011). The percentage of hydrogen peroxide (H₂O₂) and the electron transfer number were calculated from the disk and ring potentials based on the RRDE data (Liang et al., 2012).

Fig. 1. SEM images of (A) pristine SSM, (B, C, D) the cobalt oxide growing on SSM for 6 h, 12 h and 18 h, respectively, (E) the magnified cobalt oxide particles, and (F) the XRD patterns of the as-prepared Co₃O₄ particles growing on SSM surface.

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