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Low Cost Materials for the Air Cathodes in Single-Chamber Microbial Fuel Cells: A Mini Review

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Microbial fuel cells produce significant lower power output than conventional hydrogen and oxygen powered fuel cells. Thus, the utilization of fuel cell-grade materials is not practical in microbial fuel cells for recovering energy from wastewater. Air cathode single chamber microbial fuel cells are cost effective and relatively high power output. In addition, the cathode was the major contributor in the total cost of microbial fuel cells. In this paper, we reviewed the latest literatures that report metal-free oxygen reduction catalysts, binders and gas diffusion layers. Based on the recent progress, a perspective was given to design an effective air cathode in single chamber microbial fuel cells.

1. Introduction

There are many limitation of utilizing fossil fuels including geological distribution, limited reservation and CO_2 emittion. Renewable energy such as solar and wind power are of great importance for cutting down the reliability of fossil fuels. However, the electrical power from above energy sources is unstable and demand of large area of land. The organic matters in domestic, industrial and animal wastewater offer a great potential energy source (Logan and Rabaey, 2012). Microbial fuel cell (MFC) is a bioelectrochemical device that directly converts organic matter in wastewater into electricity using bacteria as biocatalyst. Compared to conventional hydrogen and oxygen fuel cells, MFCs utilize low cost biocatalyst rather than noble metal based catalysts and the anode fuel is the COD in domestic wastewater. At the early stage of MFC development, ferricyanide was commonly used for electron acceptor to sustain the overall reaction. For the future practical application of MFC, the use of ferricyanide is not suitable due to the regular replenishment and increases the operational cost (Lu et al., 2011). Oxygen is an electron acceptor that can be easily obtained from atmosphere. Therefore, air cathode MFCs have been proposed for large-scale applications, especially for wastewater treatment (Dong et al., 2012; Liu et al., 2004; Logan et al., 2007).
The air cathode consisted of a catalyst layer, a current collector and a gas diffusion layer (Cheng et al., 2006). At the early stage of MFC development, fuel cell grade catalysts (Pt based catalysts), Nafion and carbon papers were used (Oh et al., 2004). Rozendal et al estimated the capital costs of MFCs based on materials currently being used in the laboratory; they showed that the price of cathode materials (air–cathode) can account for the greatest percentage (47%) of MFC capital costs (Rozendal et al., 2008). Thus, reducing the cost of cathode materials is very critical for the practical application of MFCs. In addition, the power output of a typical single-chamber air cathode MFC is around 1.0 W m^{-2} which is about four orders of magnitudes lower than hydrogen and oxygen fueled PEMFC (typically 1.0 Wcm^{-2}). Therefore, it is not economical viable to use high cost materials and gain such low power output. In this review, particular attention was given to low cost materials for fabricating air cathodes for single-chamber microbial fuel cells.

2. Metal free catalysts for oxygen reduction reaction

Methods have been developed for preparing N doped carbon materials including chemical modified NCNTs or

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graphite oxide (GO), hard-templating synthesis, chemical vapor deposition (CVD) and so on (Liang et al., 2014; Nagaiah et al., 2010; Wen et al., 2014). All of these materials showed high electrocatalytic activity and stability. However, the debate lies in whether the active sites are created by pyridinic N (N bonded to two carbon atoms) or graphitic or quaternary N (N bonded to three carbon atoms) (Niwa et al., 2009; Xing et al., 2014; Zheng et al., 2013). The controversial results from the previous reports suggest that there is need to develop a method that is able to observe the active sites and elucidate the catalytic mechanism. Xing et al investigated the chemisorbed oxygen reduction intermediates can be detected on the nitrogen-doped multilayer graphene after ORR using X-ray photoelectron spectroscopy (XPS) (Xing et al., 2014). The results revealed that the carbon atom neighboring pyridinic nitrogen plays an important role in the ORR process and should be the main active sites (Xing et al., 2014). A series well-defined π conjugation and well-controlled doping of N carbon materials (highly oriented pyrolitic graphite, HOPG) were prepared by Guo et al using a newly designed edge-patterned surface (Guo et al., 2016). By combining characterization techniques which include scanning tunneling microscopy/spectroscopy (STM-STS), XPS, density functional theoretical (DFT) calculations and electrochemical tests, they suggested the exact reaction site of N doped carbon materials is pyridinic N. The pathway of ORR proceeds on the active site was also proposed (Guo et al., 2016).

Logan et al blended PTFE with high surface area, low cost ($2.6 kg^{-1}) and commercial available activated carbon (AC) and then cold-pressed onto a Ni mesh (Zhang et al., 2009). The power output of single chamber MFC using AC and PTFE is 1220 mWm^{-2} even higher than the MFC that uses conventional Pt/C and Nafion binder (1060 mWm^{-2}) (Zhang et al., 2009). It showed great potential of replacing Pt/C catalysts in air cathode single-chamber MFCs. Wang et al further investigated the electrocatalytic behavior of AC powder using rotating disk electrode (RDE) technique (Dong et al., 2012). The calculated electron transfer number of ORR was 3.0 which is lower than Pt/C (3.9). However, the low cost electrode using AC catalyst and PTFE binder ($30-60 m^{-2}) showed superior performance than Pt/C and Nafion ($1400 m^{-2}) (Dong et al., 2012). The micro-structure (three-phase-boundary) that creates the channels for ion, electron and gas transport is the key to optimize an electrode.

3. Binders

3.1 Existing fuel cell binders

PTFE is a well-known binder for fuel cell electrodes which cost 500 times cheaper than Nafion. It is highly hydrophobic polymer with no ion exchange capacity. Cheng et al investigated the MFC performance using PTFE or Nafion as binder in the catalyst layer and results suggested that PTFE based electrode generates around 14 % less power density than the Nafion based electrode (Cheng et al., 2005). Feng et al proposed a novel strategy for the heat-treatment of PTFE and Nafion (Na^+) to form a hybrid binder which improves the MFC performance using plain PTFE as binder, and this study demonstrated that this novel method provide a balance between cost and performance (Wang et al., 2010). Recently, novel fabrication techniques for preparing PTFE based electrodes were employed and improve the mciro-structure of the electrode catalyst layer, hence improved the air cathode performance. Wang et al developed a novel rolling technique for preparing PTFE based air cathode (Dong et al., 2012; Dong et al., 2012; Dong et al., 2013). The resulted catalyst layer and diffusion layer (GDL) possesses a more porous and uniform distribution of pore size than the conventional brushing method (Dong et al., 2012; Dong et al., 2012; Dong et al., 2013). A PVDF membrane that produced using a phase inversion method was used as DL in air cathode MFCs (Yang et al., 2015). The resulted MFC performance was similar to that of PTFE and carbon black based MFC, however, the cost of PVDF membrane is only one tenth of the DL that made of PTFE and carbon black.

3.2 Synthesized polymer binders for MFCs

A series of polystyrene-b-poly(ethylene oxide) (PS-b-PEO) polymers were synthesized via anionic polymerization (Saito et al., 2011). The content of PEO moiety was varied to investigate the binder's hydrophilicity on the performance of air cathodes. It was observed that the cathode performance was improved by 15% using an optimized PS-b-PEO binder, compared to the non-ionic conductive hydrophobic binder (Polysulfone) (Saito et al., 2011). A counterpart polymer poly(dimethylsiloxane) (PDMS) was prepared for preventing the catalyst layer from water flooding due to the hydrophobicity of PDMS (Zhang et al., 2012). Additionally, PMDS and carbon black were mixed and brush onto the air-facing side to form gas diffusion layer which similar to the method for preparing PTFE GDL. The cell performance with PDMS binder was similar to that of Nafion, however, the cost of PDMS is quite low, which is about 0.23% of the cost of using a Nafion binder with these cathodes (Zhang et al., 2012). Yu et al investigated a quaternary DABCO polysulfone anion exchange polymer as the binder of air cathode (Yu et al., 2012). Cyclic voltammetry test exhibited a redox couple which indicated that quaternary ammonia has certain electro-activity towards ORR and electrochemical characterization results exhibited even better performance than commercial Nafion binder with a cost only 1 %

that of Nafion (Yu et al., 2012). Another quaternary ammonia polymer containing fluorinated moieties showed a similar cell performance and stability to the electrodes using Nafion binder, and better performance than non-fluorinated quaternary polysulfone which indicated that polymer with fluorinated moieties may decrease the biofilm penetration into the cathode structure (Chen et al., 2012). Wang et al modified activated carbon with quaternary ammonium epoxide for the air cathode (Wang et al., 2014). Their results suggested that the addition of quaternary ammonium epoxide improved the MFC performance significantly in the absence of phosphate buffer (simulated real wastewater). A carbon nanofiber electrode was coated with metal nanoparticles and a polymer that produced from cross-linking polyvinyl alcohol monomer and poly methyl vinyl ether-alt-maleic anhydride (PMVEMA) (Singh et al., 2016). The OCV and maximum power density of the air cathode MFCs reached 980 mV and 1270 mW m^{-2} (Singh et al., 2016). The excellent performance was attributed to the reduced ionic and electrical resistance using such method. Most recently, Enrofloxacin, a broad spectrum fluoroquinolone antibiotic, was incorporated into the catalyst layer of activated carbon air cathode and a significant improvement on the stability of power output was observed，compared to that of the controlled MFC (Liu et al., 2015).

5. Challenges and Perspectives

The long-term stability of MFC performance is a key factor for scaling up MFCs. PTFE or PVDF and activated carbon based air cathodes showed promising results, however, biofouling is the main contributor for the performance degradation after a long-term operation (Zhang et al., 2011). Additionally, the hydrophobicity of PTFE or PVDF will be changed after bacteria growth on the surface. Another challenge is aqueous environment that cause electrode flooding which reduce the three-phase-boundary. In conventional fuel cells (e.g. PEMFCs), the water management is of great importance to avoid water accumulation or starvation at the anode and cathode. At last, the ion transportation is also an important factor due to the low conductivity of real wastewater. So, it is essential to identify the rate-determination step for improving the air cathode performance and utilize nano-materials or modification with particular functional groups to achieve effective ion, gas and electrical transportation for long term operation. New techniques, for example, electrospinning can be used to fabricate high surface area catalyst layers in air cathodes and even an extra outer layer for protecting the catalyst layer from biofouling (e.g. Ag composite) and maintain physical integrity.

6. Conclusions

The cost of air cathode materials in single chamber MFCs has been reduced and the performance was similar to Pt and Nafion based MFCs. The development of fuel cells provides a wide spectrum of materials which need to be tested in MFCs. Meanwhile, it is rational to take all challenges into account while designing a highly efficient air cathode in MFCs. New methods and materials should be explored to fabricate air cathodes in single chamber MFCs.

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