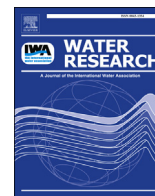




Contents lists available at ScienceDirect

Water Research

journal homepage: www.elsevier.com/locate/watres

Electro-osmotic-based catholyte production by Microbial Fuel Cells for carbon capture

Iwona Gajda ^{a, *}, John Greenman ^{a, b}, Chris Melhuish ^a, Carlo Santoro ^{c, d, e}, Baikun Li ^{c, d}, Pierangela Cristiani ^f, Ioannis Ieropoulos ^{a, b, *}

^a Bristol BioEnergy Centre, Bristol Robotics Laboratory, Block T, UWE, Coldharbour Lane, Bristol BS16 1QY, UK

^b Biological, Biomedical and Analytical Sciences, UWE, Coldharbour Lane, Bristol BS16 1QY, UK

^c Department of Civil and Environmental Engineering, University of Connecticut, 261 Glenbrook rd, Storrs, CT 06269, USA

^d Center for Clean Energy Engineering, University of Connecticut, 44 Weaver rd, Storrs, CT 06269, USA

^e Center for Micro-Engineered Materials (CMEM), Department of Chemical and Biological Engineering, University of New Mexico, Albuquerque, NM, USA

^f RSE – Ricerca sul Sistema Energetico S.p.A., Environment and Sustainable Development Dept., Via Rubattino 54, 20134 Milan, Italy

ARTICLE INFO

Article history:

Received 14 March 2015

Received in revised form

9 July 2015

Accepted 6 August 2015

Available online xxx

Keywords:

Microbial Fuel Cell (MFC)

Carbon veil cathodes

Electro-osmotic drag

Oxygen reduction reaction

Carbon capture

ABSTRACT

In Microbial Fuel Cells (MFCs), the recovery of water can be achieved with the help of both active (electro-osmosis), and passive (osmosis) transport pathways of electrolyte through the semi-permeable selective separator. The electrical current-dependent transport, results in cations and electro-osmotically dragged water molecules reaching the cathode. The present study reports on the production of catholyte on the surface of the cathode, which was achieved as a direct result of electricity generation using MFCs fed with wastewater, and employing Pt-free carbon based cathode electrodes. The highest pH levels (>13) of produced liquid were achieved by the MFCs with the activated carbon cathodes producing the highest power (309 μ W). Caustic catholyte formation is presented in the context of beneficial cathode flooding and transport mechanisms, in an attempt to understand the effects of active and passive diffusion. Active transport was dominant under closed circuit conditions and showed a linear correlation with power performance, whereas osmotic (passive) transport was governing the passive flux of liquid in open circuit conditions. Caustic catholyte was mineralised to a mixture of carbonate and bicarbonate salts (trona) thus demonstrating an active carbon capture mechanism as a result of the MFC energy-generating performance. Carbon capture would be valuable for establishing a carbon negative economy and environmental sustainability of the wastewater treatment process.

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1. Introduction

Addressing water scarcity and sanitation problems requires new methods of purifying water at lower costs and with less energy, whilst at the same time minimising the use of chemicals and their impact on the environment (Shannon et al., 2008). One method is water re-use and reclamation, where water is captured directly from industrial or municipal waste streams. Wastewater treatment is energy intensive, thus capturing part of this energy from waste and reclaiming the water would lower the total energy requirements. Novel technology platforms such as Microbial Fuel

Cells that use bacteria, capable of producing electric current thus recovering energy from wastewater, are particularly attractive. Research in the field of Bioelectrochemical Systems (BES) has focused on utilising compounds found in wastewater for the production of bioelectricity by Microbial Fuel Cells (MFCs), or biosynthesis of various compounds using Microbial Electrolysis Cells (MECs). On the one hand MFCs generate electricity, and on the other hand MECs require external electrical input to facilitate the electrolysis process for valuable product recovery such as hydrogen gas (Call and Logan, 2008), caustic soda (Pikaar et al., 2011; Rabaey et al., 2010) hydrogen peroxide (Rozendal et al., 2009) or acetate (Xafenias and Mapelli, 2014). This is an important area that is already attracting much attention. Regardless of the approach, cost-effective reactor designs and materials are urgently needed for field trials and large-scale implementation. To improve the slow kinetics of the electrochemical oxygen reduction reaction (ORR), various

* Corresponding authors. Bristol BioEnergy Centre, Bristol Robotics Laboratory, Block T, UWE, Coldharbour Lane, Bristol, BS16 1QY, UK.

E-mail addresses: iwona.gajda@brl.ac.uk (I. Gajda), ioannis.ieropoulos@brl.ac.uk (I. Ieropoulos).

catalysts are usually being employed (Wang et al., 2014). Platinum (Pt) for example, has been widely used in chemical fuel cells due to its high catalytic performance at low pH (Erable et al., 2009), however due to its high cost, in addition to relatively fast deactivation in the presence of pollutants such as sulphur, other alternatives are being explored. Some of these approaches include chemical (Haoran et al., 2014), enzymatic (Santoro et al., 2013a), microbial catalysts (Erable et al., 2012) or Non-Pt electrode modification to improve the performance (Ghasemi et al., 2011; Lefebvre et al., 2009; Santoro et al., 2012, 2013b). The electrochemistry of the ORR has been thoroughly studied in the development of chemical fuel cells (Gasteiger et al., 2005; Neyerlin et al., 2007). Complex mechanisms are dependent on the pH conditions, proceeding in the 4-electron or 2-electron (peroxide) pathways forming as a final product H_2O (in acidic conditions) or OH^- (in alkaline conditions), as illustrated in Table 1. Generally, the 4-electron pathway appears to be predominant on noble metal catalysts (Kinoshita, 1988), whilst the peroxide pathway is more common on carbon based electrodes (Kinoshita, 1988).

A similarity between chemical and biological fuel cells is represented by the water fluxes taking place across the membrane. The water distribution in chemical fuel cells is influenced by the tendency of protonic currents to transport water molecules from the anode to the cathode via electro-osmotic drag (Larminie and Dicks, 2003). As a result, an excess of water could cause the decrease of the three phases interface (TPI), flooding the entire electrode and not allowing the oxygen in gas phase to reach the catalytic sites. Therefore the water content in Proton Exchange Membrane fuel cells, is strictly controlled and the water management is a significant aspect to be considered and one of the most important challenges (Larminie and Dicks, 2003; Park and Caton, 2008).

In wastewater-based biological fuel cells, studying water transport is important as electro-osmosis might function as an active filtration process and lead to water recovery from waste.

In open to air cathode MFCs, Kim et al. observed that anolyte loss varies with external resistance due to ionic flux driving the electro-osmotic transport of water and keeping the cathode hydrated (Kim et al., 2009). The transfer of ions through the cation-exchange membrane plays a major role in the charge-balancing ion flux from the anode into the cathodic compartment (Kim et al., 2009). The formation of OH^- at the cathode during the ORR, leads to a pH gradient between the anode and the cathode compartment (Zhao et al., 2006). Whilst the electro-osmotic drag represents an active transport of water from the anode to the cathode, a passive flux through forward osmosis occurs simultaneously, dragging water and other cations to the cathode (in addition to protons). The process of forward osmosis (FO) is natural, where the driving force is the high solute concentration solution (draw solution) that flows along one side of the membrane and low solute concentration (feed

solution) to the other. Water transport occurs passively, hence the FO process is less energy intensive than Reverse Osmosis (RO), despite the fact that it is slower (McCutcheon and Elimelech, 2006). In forward osmosis, the use of catholyte of high salt concentration is acting as a drag solution, passively extracting water from the low to the high concentration (Zhang et al., 2011).

Cathode flooding has been observed in MFCs (Kim et al., 2009; Zhuang et al., 2009) and it can serve as a method for extraction of water, salts and carbon capture (Gajda et al., 2014a, 2015), where the newly produced caustic catholyte is the important by-product of the MFC operation. Therefore, it is important to study the water transport phenomenon in light of the chemical carbon capture opportunities that the process could bring to the microbial fuel cell technology, and in general into a sustainable wastewater treatment processes.

Low-cost and mechanically robust air-cathodes that can achieve good performance, is urgently required for practical MFC applications. Activated carbon and Microporous Layer (MPL) coatings as well as the carbon fibres are cost effective cathode electrodes for MFCs with an added benefit of catholyte recovery (Gajda et al., 2014a,b). In this study, the main water fluxes occurring across the membrane that produced a net liquid catholyte in MFC, are described. This work aims to better understand water transport in a Microbial Fuel Cell system and present the bioelectrosynthesis of alkaline compounds directly onto the cathode surface as a non-limiting and highly beneficial recovery. It follows the previous work describing the production of catholyte on the surface of the cathode electrode, whilst generating electricity, using MFCs fed with wastewater and employing Pt-free carbon-based electrodes (Gajda et al., 2014a).

The current study is investigating the effect of electro-osmotic drag and osmotic pressure on the MFC water transport. Moreover, it is aiming to demonstrate the feasibility of MFC electricity production with simultaneous bioelectrosynthesis of caustic composition. This is particularly important for carbon capture and driving innovation in technologies for stabilising carbon dioxide levels in the atmosphere.

2. Materials and methods

2.1. MFC design and operation

Twelve MFC reactors comprised 25 mL anode and 25 mL cathode chambers were employed, as previously described (Gajda et al., 2013), separated by a CMI-7000 cation exchange membrane (Membranes International, USA). Anode electrodes were made of carbon fibre veil with a carbon loading of 20 g/m^2 (PRF Composite Materials, Dorset, UK) and had a total surface area of 270 cm^2 , folded into 3D rectangular cuboids (geometric surface area of 17 cm^3) in order to fit into the chamber and be fully immersed in

Table 1
Oxygen reduction reaction pathways (adapted from Kinoshita-Ref. (Kinoshita, 1988)).

Conditions	Pathway	Reaction	E_0 (vs. SHE) [V]
Acidic	4-electron pathway	$\text{O}_2 + 4\text{H}^+ + 4\text{e}^- \rightarrow 2\text{H}_2\text{O}$	1.229
	2-electron (peroxide pathway)	$\text{O}_2 + 2\text{H}^+ + 2\text{e}^- \rightarrow \text{H}_2\text{O}_2$	0.67
		Followed by reduction of peroxide: $\text{H}_2\text{O}_2 + 2\text{H}^+ + 2\text{e}^- \rightarrow 2\text{H}_2\text{O}$	1.77
		or decomposition: $2\text{H}_2\text{O}_2 \rightarrow 2\text{H}_2\text{O} + \text{O}_2$	
Alkaline	4-electron pathway	$\text{O}_2 + 2\text{H}_2\text{O} + 4\text{e}^- \rightarrow 4\text{OH}^-$	0.401
	2-electron (peroxide pathway)	$\text{O}_2 + \text{H}_2\text{O} + 2\text{e}^- \rightarrow \text{HO}_2^- + \text{OH}^-$	-0.065
		Followed by reduction of peroxide: $\text{HO}_2^- + \text{H}_2\text{O} + 2\text{e}^- \rightarrow 3\text{OH}^-$	0.867
		or decomposition: $2\text{HO}_2^- \rightarrow 2\text{OH}^- + \text{O}_2$	

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