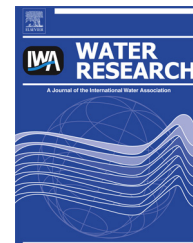


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Augmenting Microbial Fuel Cell power by coupling with Supported Liquid Membrane permeation for zinc recovery

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ABSTRACT

Simultaneous removal of organic and zinc contamination in parallel effluent streams using a Microbial Fuel Cell (MFC) would deliver a means of reducing environmental pollution whilst also recovering energy. A Microbial Fuel Cell system has been integrated with Supported Liquid Membrane (SLM) technology to simultaneously treat organic- and heavy metal containing wastewaters. The MFC anode was fed with synthetic wastewater containing 10 mM acetate, the MFC cathode chambers were fed with 400 mg L⁻¹ Zn²⁺ and this then acted as a feed phase for SLM extraction. The MFC/SLM combination produces a synergistic effect which enhances the power performance of the MFC significantly; 0.233 mW compared to 0.094 mW in the control. It is shown that the 165 ± 7 mV difference between the MFC/SLM system and the MFC control is attributable to the lower cathode pH in the integrated system experiment, the consequent lower activation overpotential and higher oxygen reduction potential. The change in the substrate removal efficiency and Coulombic Efficiency (CE) compared to controls is small. Apart from the electrolyte conductivity, the conductivities of the bipolar and liquid membrane were also found to increase during operation. The diffusion coefficient of Zn²⁺ through the liquid membrane in the MFC/SLM (4.26 × 10⁻¹⁰ m² s⁻¹) is comparable to the SLM control (5.41 × 10⁻¹⁰ m² s⁻¹). The system demonstrates that within 72 h, 93 ± 4% of the zinc ions are removed from the feed phase, hence the Zn²⁺ removal rate is not significantly affected and is comparable to the SLM control (96 ± 1%), while MFC power output is significantly increased.

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1. Introduction

The increasing value of metal resources and the need to remove heavy metal pollutants from water resources make

the recovery of metal ions from industrial wastewaters, acid mine drainage and natural leachates an important issue. Domestic and industrial wastewaters can also contain high levels of both organic matter and metal ions (Liang et al., 2010), which require suitable treatment to achieve regulatory

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levels of toxic metals and biological oxygen demand (BOD) prior to discharge to the receiving environment. Leaching from natural mineral deposits, historical industrial activity and acid mine drainage will also require treatment to protect or remediate the environment, but these sites may be remotely located without ready access to utilities. Current legislation such as the EU Water Framework Directive (Comission, 2012) restrict the discharge of toxic metals, nutrients and BOD in wastewater effluents and have led to the need for incentivisation and increasingly effective treatment processes capable of removing the contaminants (San Roman et al., 2010). Furthermore, the intrinsic value of some metals may warrant the deployment of energy efficient separation processes to recover ions of their salts from aqueous solutions. Political and economic factors surrounding the supply and procurement of metallic raw materials means that efficient metal recovery with low energy demands are key drivers with respect to the development of novel electrolytic recycling processes.

In a microbial fuel cell (MFC) system, electrical current is directly generated from bacteria which act as a catalyst in the anodic oxidation of biodegradable organic matter. The electrogenic metabolism also produces a protonic counter current, so maintaining charge balance in the cell. In air cathode MFC systems, oxygen acts as the electron acceptor at the cathode and combines with two protons to form water (Liu and Logan, 2004). Oxygen in air is widely considered to be the most plausible terminal electron acceptor for MFC in a deployable system at significant scale, because of the benign environmental consequences of its use compared with other oxidants. This is despite the high activation overpotentials that can act to significantly lower the cell potential. It is generally thought that the cathode is the current limiting electrode in existing systems (Fan et al., 2008; Rismani-Yazdi et al., 2008). Low concentrations of protons at the cathode may result from their limited flux from the anode and their consumption in the water generating oxygen reduction reaction (ORR). The restricted protonic flux is in part due to high levels of other cations such as Na^{2+} or K^{+} , which may be present in significantly higher concentrations (4–5 orders of magnitude) and are therefore more likely to be transferred through a cation exchange membrane (Rozendal et al., 2006). Lower pH values affect the ORR positively by reducing the cathodic overpotential; according to Nernst equation by 59 mV for each pH unit, resulting in higher cell potential and power output. This is in accordance with the findings from Zhuang et al. (2010) who operated a MFC with high anodic pH (10) and low cathodic pH (2) to achieve higher power densities and an open circuit potential (OCP) of 1.04 V with iron(II) phthalocyanine (FePc) as cathode catalyst or Erable et al. (2009) who observed the same effect with a low pH air cathode using Pt as the catalyst.

It is possible to reduce metal ions to their metallic form at the cathode of a modified MFC, generally described as a bio-electrochemical system (BES). When considering the standard redox potential for acetate ($\text{HCO}_3^-/\text{Ac}^-$); $E^0 = -0.29$ V (10 mM; pH = 7), which represents typical conditions in the experimental work presented herein, the reduction of the metal ions into their metal form will be thermodynamically restricted to Ag^+ (Tao et al., 2012) and Cu^{2+} (Heijne et al., 2010). Several

other metal ions have been reduced in the cathode chamber of BES/MFCs, including $\text{Fe}^{3+}/\text{Fe}^{2+}$ (Ter Heijne et al., 2006a), $\text{Hg}^{2+}/\text{Hg}^0$ (Wang et al., 2011), Cr^{6+} to Cr^{3+} (Wang et al., 2008a) and Cr^{6+} to Cr^{3+} in combination with vanadium V^{5+} (Zhang et al., 2012a). However, in these studies, whilst the metal ions were reduced, they were not reportedly and actively removed from the solute ion or wastewater. Recently, more than 95% of Cu, Zn and Pb in fly ash leachate were removed using a bio-electrochemical system (BES). Only Cu was reduced at the cathode of the system, while Zn and Pb were removed in a separate electrolysis cell (Tao et al., 2013). Another approach was reported by Xue et al. (2013), who operated a microbial fuel cell-zero valent iron (ZVI) hybrid process. Furthermore, an MFC's cathode was also used for cobalt leaching from LiCoO_2 (Liu et al., 2013), and acid mine drainage loaded with iron (Lefebvre et al., 2012).

Heavy metal ions such as Zn^{2+} , Ni^{2+} , Cr^{2+} , V^{5+} or Co^{2+} are widely present in industrial wastewaters from mining, electroplating, leaching and other hydrometallurgical processes, but cannot be directly recovered in MFCs by cathodic reduction due to their low standard reduction potentials relative to acetate or other economically available organic substrates. Zn^{2+} exhibits a standard redox potential of $E^0 = -0.76$ V and is one of the most commonly encountered heavy metal ions in industrial wastewater, as well as being the most abundant heavy metal found in surface and ground waters. Due to its acute toxicity and non-biodegradability, zinc-containing liquid and solid wastes are considered to be hazardous wastes (Hui et al., 2005; Alyüz and Veli, 2009), with a legal limit of 5 mg l^{-1} according to the World Health Organization. Zinc may be accumulated in the environment and can inhibit biological treatment processes.

Common techniques for Zn^{2+} removal from aqueous solutions include precipitation as hydroxide, sulfide or carbonate, removal via ion exchange (Alyüz and Veli, 2009), adsorption (Chen et al., 2011), evaporation or biosorption (Areco and dos Santos Afonso, 2010). Recently (Abourached et al.) have reported high Zn and Cd removal efficiencies in MFCs, mainly by biosorption and sulphides precipitation. However, a zinc concentration of 32 mg l^{-1} resulted in a significant MFC voltage drop of more than 70%. Common methods have not considered the recycling of zinc, and given that 30% of today's global zinc production already arises from recycling (Tsakiridis et al., 2010), this would be a desirable ambition. Zinc is recovered in part by hydrometallurgical electrolytic zinc recovery. In order to maintain a favourable hydrogen overpotential and obtain high purity levels and energy efficiencies during electrowinning by electrolysis, the recycling of zinc requires an electrolyte solution free of interfering ions. Zinc deposition is very sensitive to small quantities of particular impurities such as As, Sb, Ge, Fe, Cu, Co and Ni ions (Fosnacht and O'Keefe, 1983). An alternative and low energy technique for the selective Zn^{2+} removal from solutions is reactive liquid–liquid extraction (Noll et al., 2010). Supported Liquid Membrane (SLM) extraction allows the selective removal and concentration of metal ions from aqueous solutions against an outer concentration gradient (uphill) in the strip chamber. Supported Liquid Membrane Permeation (SLM) uses porous supports where the pores are impregnated with an extractant dissolved into a solvent phase (Lozano

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