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Self-sustained reduction of multiple metals in a microbial fuel cell–microbial electrolysis cell hybrid system

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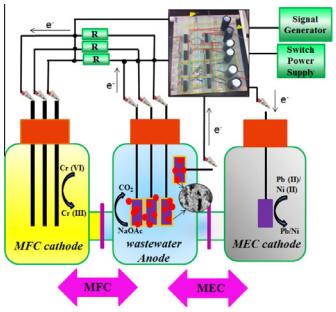
HIGHLIGHTS

- Self-sustained MFC–MEC hybrid systems simultaneously reduced multiple metals.
- The power density of MFCs was 189.4 mW m^{-2} and sufficient to support MECs.
- The reduction rates of Cr^{6+}, Pb^{2+}, and Ni^{2+} were 32.7, 32.7, and 8.9 mg L^{-1} d^{-1}.
- The energy recovery based on the ESC was ~470%, but overall recovery was ~1%.
- An electrochemical model was developed to predict metal recovery in the system.

GRAPHICAL ABSTRACT



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ABSTRACT

A self-sustained hybrid bioelectrochemical system consisting of microbial fuel cell (MFC) and microbial electrolysis cell (MEC) was developed to reduce multiple metals simultaneously by utilizing different reaction potentials. Three heavy metals representing spontaneous reaction (chromium, Cr) and unspontaneous reaction (lead, Pb and nickel, Ni) were selected in this batch-mode study. The maximum power density of the MFC achieved 189.4 mW m^{-2}, and the energy recovery relative to the energy storage circuit (ESC) was ~450%. At the initial concentration of 100 mg L^{-1}, the average reduction rate of Cr(VI) was 30.0 mg L^{-1} d^{-1}, Pb(II) 32.7 mg L^{-1} d^{-1}, and Ni(II) 8.9 mg L^{-1} d^{-1}. An electrochemical model was developed to predict the change of metal concentration over time. The power output of the MFC was sufficient to meet the requirement of the ESC and MEC, and the "self-sustained metal reduction" was achieved in this hybrid system.

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1. Introduction

Metal pollution caused by natural and human activities (e.g. electroplating, steel production, mining, refuse burning, transportation, and power generation) has posed severe environmental and health problems (Agarwal, 2009). Traditional physical, chemical, and biological processes (e.g. chemical precipitation, electrocoagulation, electrolysis, and bioadsorption) have been used to remove heavy metals from wastewater (Jüttner et al., 2000; Kurniawan et al., 2006; Wang and Chen, 2009; Dermentzis et al., 2011). However, most of these methods require high operational and maintenance costs, excessive chemical dosage, and toxic sludge treatment (Qin et al., 2012; Abourached et al., 2014).

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Nomenclature

Abbreviation	Description
A	projected area of anode in MFCs
BES	bioelectrochemical system
C_o	initial concentration of Cr(VI) in MFC unit
C_t	concentration of Cr(VI) after time (t) in MFC unit
C_{MEC}	concentration of metals reduced in MEC unit
COD	chemical oxygen demand
E_p	voltage produced from the ESC
EDX	energy-dispersive X-ray spectroscopy
ESC	energy storage circuit
F	Faraday's constant
I_{MFC}	total current generated from the three pairs of electrodes in MFC unit
I_{MEC}	current of MEC unit
M_{MFC}	molecular weight of Cr
M_{MEC}	molecular weight of metals in the MEC cathode chamber
MFC	microbial fuel cell
MEC	microbial electrolysis cell
n_M	moles of metals reduced in MEC
n_S	moles of substrate used in the anode
OCP	open circuit potential
Q_{exp}	total experimental charges accumulated during the MFC operation
Q_{the}	total theoretical charges accumulated during the MFC operation
r_{cat}	cathodic electron recovery
R_{ext}	external resistance over each pair of electrodes in MFC unit
R_{in}	internal resistance of MFC unit
R_e	external resistance of MEC unit
R_i	internal resistance of MEC unit
SEM	scanning electron microscope
V_A	volume of the anode chamber
V_{MFC}	liquid volume of the MFC cathode chamber
V_{MEC}	liquid volume of MEC cathode chamber
η_E	energy recovery of metal in the MEC cathode chamber based on ESC
η	overall energy recovery of metal in the MEC cathode chamber based on substrate consumption
COD	COD concentration change in the anode chamber
ΔG_M	energy content of metals based on the Gibbs free energy
ΔG_S	energy content of substrate in the anode based on the Gibbs free energy
Δt	time gap of the voltage measurement using the data logging system

Recently, bioelectrochemical systems (BES) have attracted global attention for converting the chemical energy stored in organic wastes to electricity (Rozendal et al., 2006; Pant et al., 2010). Two types of BES technologies have been extensively studied: microbial fuel cells (MFCs) for electricity generation, and microbial electrolysis cells (MECs) for metal reduction and hydrogen production (Logan et al., 2006; Wang et al., 2011; Qin et al., 2012; Santoro et al., 2013). MFC reactions occur spontaneously since the cathode potentials (e.g. oxygen, spontaneous-reacting metals (copper, mercury, chromium, and iron)) are positive and the anode potentials (e.g. organic substrates in anaerobic chambers) are negative (Logan et al., 2006; Wang et al., 2008; Heijne et al., 2010). Nevertheless, the gap between the anode and cathode potentials in MECs is too small to drive the electrons from anode to cathode, so that the MEC reaction requires external power supply. MECs have been applied to treat nonspontaneous-reacting metals (e.g. zinc, lead, nickel, and cadmium) (Modin et al., 2012; Qin et al., 2012). Using MFCs and MECs to reduce metals can save chemical dosage, avoid toxic chemical sludge generation, and more importantly, convert the chemical energy in heavy metals to electricity.

Due to the low voltage output of MFCs (voltage: <0.5–0.8 V), the electricity produced has not been used collectively. Many studied have focused on the enhancement of power generation in MFCs through novel electrode materials and configurations, and optimal operational conditions (Cheng et al., 2006; Santoro et al., 2011; Wei et al., 2011; Manickam et al., 2013). Some effort has been invested to utilize the low voltage output of MFCs as wastewater quality sensors, and harvest the power output of multiple MFCs to support the subsea devices (Karra et al., 2013; Liu et al., 2014). Recently, MFCs have been used as the power supply for MECs to produce hydrogen (Sun et al., 2008; Wang et al., 2011). But the direct connection of MFCs and MECs has the problem of voltage reversal for long-term operation due to the change of anode and cathode potentials. Charge pumps or DC/DC converters were used to increase voltage production, but these systems have low circuit efficiency (20–70%) (Donovan et al., 2008; Meehan et al., 2011). On the other hand, capacitor circuits have been found to have high circuit efficiency (45–95%) and high system recovery (Kim et al., 2011; Hatzell et al., 2013). Therefore, capacitor circuits could be used to integrate MFC and MEC units for reducing multiple metals simultaneously and saving the energy consumption in metal waste treatment.

The objective of this study was to achieve the self-sustained reduction of multiple metals in a novel MFC–MEC hybrid system by harvesting electricity generated from spontaneous-reacting metals in MFC unit through energy storage circuits (ESC) and powering the reduction of nonspontaneous-reacting metals in MEC unit. Three heavy metals representing spontaneous reaction metal (chromium, Cr) and unspontaneous reaction metal (lead, Pb; nickel, Ni) were examined individually in the MFC and MEC units. There were four tasks. First, the simultaneous reduction of metals occurring in the MFC and the MEC could change the pH and voltage output of the MFC. Thus, the MFC performance was examined over the period of metal reduction. Second, the power output of the MFC by reducing Cr(VI) was explored, and the reduction rate of Pb(II) and Ni(II) in the MEC was determined. Third, the energy recovery and coulombic efficiency of the MFC and MEC units in the hybrid system were determined to elucidate the correlation between metal reductions in the MFC and MEC units. Finally, a model was developed to predict the metal reductions in the MFC–MEC hybrid system at different concentrations over time.

2. Methods

2.1. The batch-mode MFC–MEC hybrid system setup

The batch-mode MFC–MEC hybrid system consisted of three plexiglass bottles (Fig. 1a). The anode chamber (total volume: 300 ml) was located in the middle of the hybrid system with one extension connecting to the cathode chamber (total volume: 300 ml) of the MFC and the other one connecting to the cathode chamber of the MEC (total volume: 140 ml). The chambers (bottles) were separated by a Nafion membrane (N117, DuPont Fuel Cells, DE). Plain carbon cloth (4×4 cm^2, Fuel Cell Earth LLC, MA) was used as the anode electrodes and MEC cathode electrode.

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