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ORIGINAL ARTICLE

Characteristics of Electricity Production by Metallic and Non-metallic Anodes Immersed in Mud Sediment Using Sediment Microbial Fuel Cell

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Abstract

Sediment microbial fuel cell (SMFC), equipped with Zn, Al, Cu, Fe or graphite felt (GF) anode and marine sediment, was performed. Graphite felt was used as a common cathode. SMFC was single chambered and did not use any redox mediator. The aim of this work was to find efficient anodic material. Oxidation reduction potential (ORP), cell voltage, current density, power density, pH and chemical oxygen demand (COD) were measured for SMFC's performance.. The order of maximum power density was 913 mWm^{-2} for Zn, 646 mWm^{-2} for Fe, 387.8 mWm^{-2} for Cu, 266 mWm^{-2} for Al, and 127 mWm^{-2} for graphite felt (GF). The current density over voltage was found to be strongly correlated with metal electrodes, but the graphite felt electrode, in which relatively weaker electricity was observed because of its bio-oriented mechanism. Metal corrosion reactions and/or a complicated microbial electron transfer mechanism acting around the anodic compartment may facilitate to generate electricity. We presume that more sophisticated selection of anodic material can lead to better performance in SMFC.

Key words : Sediment microbial fuel cell, Microbial corrosion, Oxidation reduction potential, Chemical Oxygen Demand

1. Introduction

Microbial fuel cell (MFC) systems have been of great interest as a potential applicant for future alternative energy production which employs microbes to generate electricity from biochemical reaction of organic and inorganic substances (Chaudhuri and Lovley, 2003). These substances are converted into electricity in the anode of MFC by microbial metabolism (via the action of bacteria as catalysts) (Rabaey and Verstraete, 2005; Rabaey et al., 2006). The anodic part of SMFC seems crucial in order to (A) determine the most efficient microorganisms, those that can offer the highest rate of oxidation or able to extract the highest number of electrons per mole of the substrate (Tsujimura et al., 2001; Kim et al., 2002; Choi et al., 2001), (B) study the effectiveness of redox mediators (Park and Zeikus, 2000; McKinlay and Zeikus, 2004), (C) select more effective electrode materials (Katz et al.,

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1999; Schroder et al., 2003; Gregory et al., 2004), and (D) determine the most efficient anodic reactions, those producing the highest number of electrons per unit weight of the reactant (Lee et al., 2002; Logan, 2004). Specific bacteria, including Geobacteraceae, were enriched on the anode where organic matter was oxidized and electrons were directly transferred under anaerobic conditions (Tender et al. 2002). Bacteria are capable of electricity generation from enriched domestic wastewater (Liu et al., 2004), ocean sediments (Tender et al., 2002), animal wastes (Min and Logan, 2004), and anaerobic sewage sludge (Kim et al., 2005; Rabaey et al., 2004). Several factors affect MFC performance including the microbial inoculum, chemical substrate (fuel), electrode materials, cell internal and external resistance, solution ionic strength, and electrode spacing, type of proton exchange material (and the absence of this material) (Liu and Logan, 2004; Cheng et al., 2006; Liu et al., 2005).

However, to increase the anode performance, different chemical and physical strategies have been followed. Park and Zeikus, (2003) incorporated Mn(IV) and Fe(III) and used covalently linked neutral red to mediate the electron transfer to the anode. Electro-catalytic materials such as polyaniline/Pt composites have also been shown to improve the current generate ion following through the direct oxidation of microbial metabolites (Videla, 1996; Froelich et al., 1979; Gil et al., 2003). Each of these compounds (mediators) has a different potential in the oxidation-reduction reaction, and their differences result in generation of electricity. The difference in the chance of contact between electron shuttles' and anode electrode can also cause the variation in electricity generation. In other views, the higher the concentrations of electron shuttles in the anode phase, the more the electricity generation (Logan et al., 2006). SMFC design must be made if SMFC systems are to be used for wastewater treatment. In a two chamber SMFC system, anode and cathode are separated by a proton exchange membrane. Proton exchange membranes (e.g. Nafion) are not suitable if SMFC systems are applied for wastewater treatment processing because of high cost. Finally, a general use of the SMFC might be the removal of organic matter from sediments or other polluted sites. As both the bio-electrochemical activity and microbial metal reducing activity appear to share the same or similar electron transport chains, electrochemical activity of microorganisms may play an important role in the biogeochemical cycling of carbon, nitrogen, and other organic contaminants. Indeed, the concept of mediator-less MFC studies provide new insights into the function of electrochemical activity bacteria directly associated with practical needs of environmental protection. In past years, environmental pollution control has mainly relied on how fast and feasible process could operate to treat environmental pollutants. very aspect of the SMFC operation can be regarded as sub-optimal at this point, including the anode, the cathode, the membrane, and SMFC design. While these challenges loom as great, the opportunities are equally great. Cost effective SMFC systems can offer a potentially promising remediation technology that might earns surplus energy. The specific objective in this study was to evaluate the possibility to choose a suitable anode material for field applications by using laboratory experiments to increase power generation.

2. Materials and Methods

2.1 Sediment Collection

Estuarine sediment was used for microbial energy generation. The sediment was collected from the bottom (approximately 10 cm from the sediment - water interface) of Lake Sap-Kyo (N 36°52'9.3", E 126°50'29.12") in a southern province in South Korea. The sampling was performed using a Ponar

type grab sampler (2.4-L vol.). All samples including surface water were placed into clean polycarbonate jars (Nalgene, Fisher Scientific) with no headspace gas (i.e. no air) and transported to the laboratory in a cooler box with ice packs. All sediments were passed through a 2-mm sieve to remove plant debris, macro fauna, and other large terrestrial leaves and then homogenized by mixing with a stainless steel spatula prior to use. All samples were kept at 4°C before use.

2.2 Establishing sediment microbial fuel cell (SMFC)

A one-chambered sediment cell was established to analyze the performance of different electrodes. The body of the SMFC was a 500-mL Pyrex beaker. Aliquots of 150 mL wet sediment and 100-mL of sea-water were loaded into the lower and upper part of sediment MFC, respectively. The SMFC was monitored for 72 hours. During the runs, water loss due to evaporation was compensated daily by adding distilled water.

2.3 Anode and cathode compartments

SMFC, equipping different metallic anodes, was used with respect to graphite felt cathode (common for all anodes). The electrodes used in this experiment were Zinc (Zn), Aluminum (Al), Copper (Cu), Iron (Fe), and Graphite felt (GF). The dimension of the anode was 35×30×2 mm (length × width × thickness). The cathode (35×30×2 mm) was bare graphite felt and placed parallel to the anode and 4 cm above the sediment - water interface. Both anode and cathode were connected by a platinum wire (internal resistance 20 Ω) and an insulated copper wire to an external load. All electrodes had a projected surface area of 0.002 m^2.

2.4 Electricity Measurement

Redox potentials were measured using an Ag/AgCl reference electrode (9678BNWP, Thermo Orion, Beverly, MA, USA). The pH was measured using an automatic calibrated pH meter (Model pH 20, Hanna Instruments, USA). Cell voltage was recorded using a multi-meter and a data acquisition system (Model 2700, Keithley Instruments, Cleveland, OH, USA). Current density, i, was calculated as:

$$i=I/A=V/RA$$

where V (mV) is the voltage, I (mA) is the current in electrochemical tests, R(Ω) is the external resistance, and A(m^2) is the projected surface area of the studied electrode. Power density was calculated according to:

$$P\ (Wm^{-2}) = iV$$

where all in MKS units. Columbic efficiency was calculated as $CE = C_p/C_{th} \bullet 100\%$, where C_p is the total coulombs calculated by integrating the current over time, and C_{th} is the theoretical amount of coulombs available based on the Chemical oxidant demand (COD) removed in the SMFC. Current density was also calculated by dividing the current by the apparent surface area of anode.

3. Results and Discussion

3.1 Redox potentials of SMFC

Oxidation reduction potential (ORP) has recorded for both chamber (anode and cathode) of SMFC at different times (using Ag/AgCl reference electrode). The lab data were summarized for four types of metal (Zn, Al, Cu, Fe) and graphite felt as non-metallic electrode. Anodic part was not modified by any mediator and covering naturally microbial environment condition. Recently, a number of bacteria have been found to possess the ability to transfer electrons from oxidized fuel (substrate) to a working electrode without a mediator (Kim et al., (1999a; 1999b) making it possible to establish mediator-less MFCs. Many metal reducing bacteria, which are capable of reducing solid

metal oxides: they include Clostridium Butyricum (Park et al., 2001), Geobacter sulfurreducens (Bond and Lovely, 2003), Rhodofoferax ferrireducens (Chaudhuri and Lovley, 2003) and Shewanella puterfaciens (Kim et al., 2002). Furthermore, in the SMFC system, growth and metabolism of S. puterfaciens were necessarily dependent on the presence of active anode, which served as the electron acceptor for growth and metabolism. Anodic potentials in our sediment MFC varied for Zn (184 to -155 mV), Al (155 to -150 mV), Cu (148 to -126 mV), Fe (161 to -42 mV), and Graphite felt (178 to -5 mV) for a period of time (0-72 h) whereas the cathodic potentials showed a slow drift (Figure 1). Metallic anode has shown large ORP change than that in non-metallic anode graphite electrode. The results in figure 1 shows that as time progresses the potential of the anode tend to decrease significantly, which seems due to decrease in chemical activities of organic and/or inorganic components within the sediment as well as electrochemical changes on the electrode resulting from biofilm buildup (Liu and Logan, 2004). The microbial fuel cell potential reported in the literature varies widely depending on the type of anodic and cathodic efficiencies of the cell used. The anode reaction potentials reported in the literature vary between -300 and -500 mVSCE (Bond and Lovely, 2003; and Liu et al., 2005). The open circuit potential (OCP) of the plain graphite anode, iron containing anodes, and manganese containing electrodes are different from each other, indicating that different electron transfer reactions occur at each of these anode type (Froelich et al.,1979).

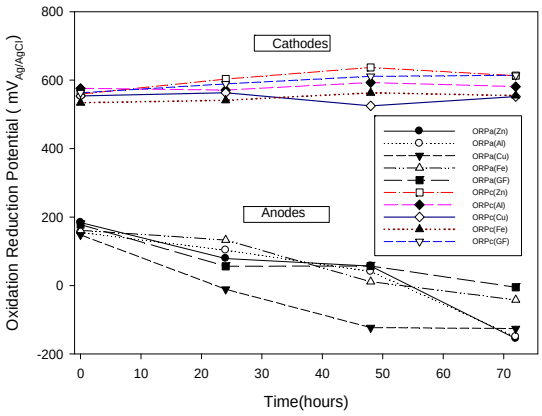

Fig. 1. Oxidation reduction potential (ORP) variations in the cathode and anode (against Ag/AgCl) in our sediment microbial fuel cell.

3.2 Power Density of SMFC

Power density of SMFC using different metallic and non-metallic anode was depicted in figure 2. SMFC was run for 72 hours for different anodic materials such as Zn, Al, Cu, Fe, and graphite felt (GF) which are actively conductive. Once operated with metallic and non-metallic anode, the deployed fuel cell has achieved its maximum power density of Zn (913 mWm^{-2}), Al (266 mWm^{-2}), Cu (387.8 mWm^{-2}), Fe (664 mWm^{-2}), GF (127 mWm^{-2}) varying with time. After starting SMFC, the power density tends to decrease over time significantly (10 h) for all anodes but interestingly Zn anode was found to be increase after that time. Increasing trends of power density of Zn anode have sustained till 45 hours after that drop. Power densities for all anodic material were shown to be different for different anodes probably because of some catalytic interactions on the electrode surface. And the total area of the electrodes including the internal surface area (Park and Zeikus, 2003), apparent surface area (Liu et al., 2005; Moon et al., 2005) or projected area (Liu and Logan, 2004; Logan et al., 2005) has been adopted for the computing of power density in MFCs. Schroder et al. (2003) obtained a power density of 6.0 Wm-2 polyaniline－modified platinum as an anode. From literature, it is reported that mediated anode produces higher power than non-mediated one. A similar fuel cell using a plain graphite anode could sustain maximum power of ~20 mW/m^2 (~66 mA/m^2)

at 0.30 V while AQDS modified graphite electrodes initially produced five-time greater power, and then gradually decay over time to the performance of the graphite anode (Tender et al., 2002).

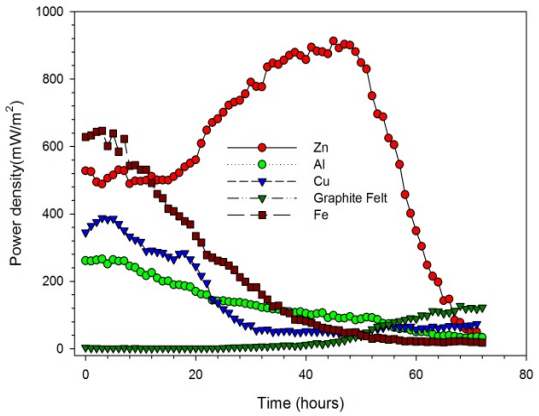

Fig. 2. Power density of SMFC according to different anode with respect to time.

3.3 Cell Voltage and Current density of SMFC

Figure 3 presents the cell voltage and figure 4 represents the current density of SMFC without external substrates and electron transport mediators over 72 hours of operation. Immediately after circuit connection (external circuit resistance fixed at 20 Ω) was established, the current density sharply increased. Maximum current density of Zn (1175 mA/m^2), Al (360 mA/m^2), Cu(610 mA/m^2), Fe (890 mA/m^2), and Graphite felt (255 mA/m^2) were observed at 45 h, 3 h, 5 h, 3h and 68 h, respectively. Current density and cell voltage are varying within a certain range over the next 72 hours. Figure 4 indicate that the majority of electrical charge was generated in the initial stage of the sediment MFC runs: approximately more than 50% of the total amount current density was delivered within the first 45 hours. The overall pattern of electricity generation was analogous to that observed by Holmes et al., (2004a, 2004b), who demonstrated microbially-mediated current production using sediment MFC systems: a rapid increase in current production within the first few days of reactor operation with no lag period, followed by a gradual decrease. Wei and Zhang, (2007) and Mathis et al. (2008) isolated the electricigenic (also referred to as anodophilic and electrochemically-active) bacteria from natural marine sediments and proved that the instant electricity generation in their MFC systems augmented with isolated cultures. Current density among the metallic anodes of Al, Cu and Fe showed a continuous decrease whereas Zn metal showed somewhat different phase. Zn anode showed rising to falling incidents and non-metallic anode graphite felt showed an elevated

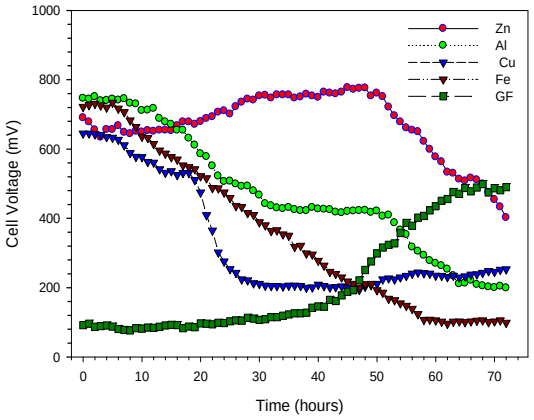

Fig. 3. Cell voltages of SMFC with respect to time.

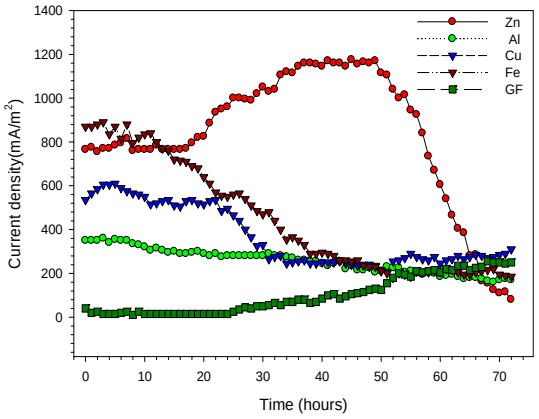

Fig.4. Current density vs time of SMFC operation (surface area 0.002 m^2 and internal resistance 20Ω).

tend till end time. These results suggest that catalytic or microbial activity on the surface of Zn or graphite felt was higher than Al, Cu and Fe anode. It is observed that voltage losses as like current density over time. According to anode, the observed maximum cell voltages were Zn (777 mV), Al (751 mV), Cu (645 mV), Fe (734 mV) and GF (499 mV) over different time (Fig.3). We have used same size of anode and cathode. Very little corrosion and microbial activity might be acted on graphite felt surface, which kept current density and cell voltage not that high with respect to other values. A number of different bacteria are able to reduce corrosion rates of different materials in several corrosive media. One type of bacteria can shift corrosion potential (Ecorr) of one metal in the positive direction while another type can shift corrosion potential (Ecorr) of some other metal in the negative direction (Kus et al., 2005; Mansfeld, 2007).

3.4 pH and chemical oxygen demand (COD) of SMFC

In this work pH of SMFC changed over time in anode part and cathode part in reverse way (figure 5). When pH of anodic part were decreased, at the same time cathode pH increased. The pH variation ranges were 7.5 to 6.4 in anode Zn, 7.3 to 6.8 in Al, 6.6 to 6.4 in Cu, 6.7 to 6.2 in Fe, 7.5 to 6.7 in GF over starting time and ending time, respectively. But, in cathode part, pH changed very little (7.6~7.7). Acidic environment in anode part is an important issue because MFCs that are able to operate at low pH are technologically advantageous, as the proton transport rate from anode to cathode increases and the kinetic barrier for O2 reduction to H_2O at the cathode decreases, which leads to the higher current and power densities (Biffinger, 2008; Erable et al., 2009). In acidic medium acidophilic microorganisms that have colonized the anode and cathode surface can oxidize glucose and other organic compounds in the absence of redox mediators (Malki et al., 2008). It is well known that *Acidiphilium spp*. is able to use organic compounds such as glucose and glycerol as electron donors (Johnson and McGinness, 1991; Kusel et al., 1999; Malki et al., 2008). Chemical oxygen demand of SMFC was decreased over time in case of all anodes. But COD decreases mean sediment losses organic matter that might be essential for long time generation of electricity (Figure 6). Biomass production could account for additional COD removal, but it was not possible to establish a complete mass balance of COD in SMFC system. Not all the organic matter in a wastewater can be

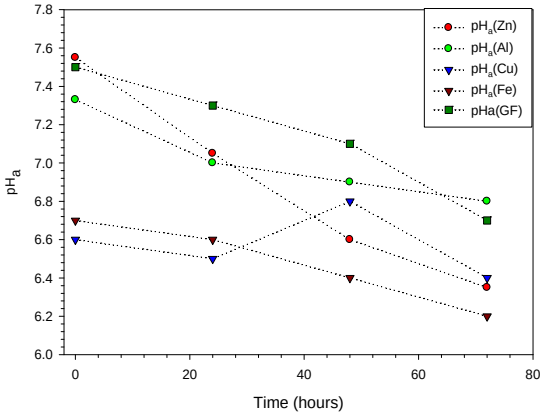

Fig. 5. pH changes in anode chamber over time of SMFC (pHa, anodic pH).

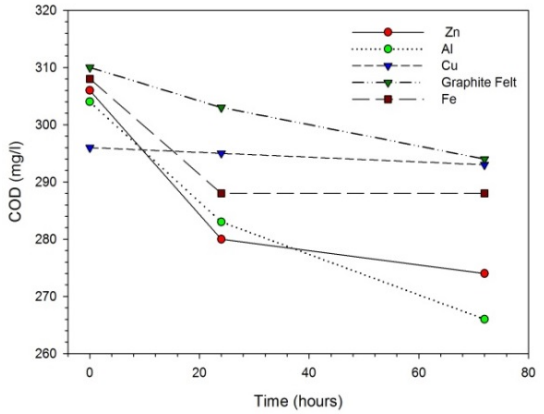

Fig. 6. Chemical oxygen demand (COD) in anode part decreasing with respect to time.

biologically degrade, so that COD removals are typically lower than those for biochemical oxygen demand (BOD) (Grady et al., 1999).

4. Conclusions

This research was aimed to evaluate the feasibility of electricity generation of SMFC using different metal and non-metal anodes. From our study we can assume that a mediator less single chamber sediment microbial fuel cell might produce higher power by changing anode material. As for power density Zn was excellent over other metals. On the basis of the results presented in figure 2, anode materials are in the order of Zn > Fe > Cu > Al > GF with respect to power density in the anode chamber. Some interfacial reactions or phenomena on metal or non-metal surface need to be investigated further as mentioned in Section 3. It is hoped that with sustain progress and continued research into cost effective materials and designs, and such systems could become commercially available in only a few years. Also time duration of the cell and limitation of current generation could be next issues.

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