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 orgraphite felt (GF) anodeand marine sediment, was performed. Graphite felt was used as a common cathode. SMFC was single chambered and did not use any redox mediator. The aimof this workwas 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 GF. The current density over voltage was found to be strongly correlated one another in most metal electrodesbutthe graphite felt electrode, in which relatively weaker electricitywas observed because of its bio-oriented mechanism. Metal corrosion reactionsand/or a complicated microbial electron transfer mechanismacting around the anodiccompartment may facilitate to generate electricity. We presume that more sophisticated selection of anodic material can lead to better performance in SMFC.

1. Introduction

Microbial fuel cell (MFC) systems have been of great interest as a potential applicant for future alternative energy production where employ microbes to generate electricity from biochemical reaction of organic and inorganic substances [1].These substances are converted into electricity in the anode of MFC by microbial metabolism (via the action of bacteria as catalysts) [2, 3]. The anodic part of SMFC is important in attempt 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 [4-6]; (B) study the effectiveness of redox mediators [7, 8]; (C) select more effective electrode materials [9-11]; (D) determine the most efficient anodic reactions, those producing the highest number of electrons per unit weight of the reactant [12-13]. Specific bacteria, including

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Geobacteraceae, were enriched on the anode where organic matter was oxidized and electrons were directly transferred to the anode under anaerobic conditions [14]. Bacteria capable of electricity generation from enriched domestic wastewater [15], ocean sediments [14], animal wastes [16], and anaerobic sewage sludge [17, 18]. 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) [19-21].

However, to increase the anode performance, different chemical and physical strategies have been followed. Park et al. [22] 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 polyanilins/Pt composites have also been shown to improve the current generation through assisting the direct oxidation of microbial metabolites [23-25]. Each of these compounds (mediators) has a different potential that possesses in the oxidation-reduction reaction, and their differences result in generate 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 [26].SMFC design must be made if SMFC systems are to be used for wastewater treatment. In a two chamber SMFC system, anode and cathode separated by proton exchange membrane. Proton exchange membranes (e.g. Nafion) are not suitable if SMFC systems are applied for wastewater treatment processing because of 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, metal 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. The challenges facing the community are many. Everyaspect 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. The specific objective in this study was to evaluate the possibility to choose a suitable anode material for field applications by using laboratory experiments as well as to increase power generation. Cost effective SMFC systems can offer a potentiallypromising remediation technology that also earns surplus energy.

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). 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.

2.2Establishing sediment microbial fuel cell(SMFC)

A one-chambered sediment cell was established to analyze the performance of different electrodes. The body of theSMFC wasa 500-mL Pyrex beaker. Aliquots of 50-g wet sediment and 120-mL of sea-water were loaded into the lower and upper part of sediment MFC, respectively. The SMFCwas monitored for 72 hours.During theruns, water loss due to evaporation was compensated daily by adding distilled water.

2.3Anode and cathode compartments

SMFC, equipping different metallic anodes, was used in respect of graphite felt as common cathode. The electrodes used in this experiment were Zinc (Zn), Aluminium (Al), Copper (Cu), Iron (Fe), and Graphite felt (GF). The dimension of the anode was 150×45×3 mm (length×width×thickness). The cathode (40×35×3 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 20Ω) and an insulated copper wire to an external load. All electrodes had a projected surface area of 0.002 m^2.

2.4Electricity Measurement

Redox potentials were measured using an Ag/AgCl reference electrode (9678BNWP, Thermo Orion, Beverly, MA, USA).The pH was measured using anautomatic 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) the current in electrochemical tests, R(Ω) t he external resistance, and A(m^2) the projected surface area of the studied electrode. Power density was calculated according to P (mWm^{-2})=10×iV (10 needed for the given units). Columbic efficiency was calculated as CE =C_p/C_{th}•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

3.1Redox potentials of SMFC

For each metallic anode has recorded oxidation reduction potential (ORP) of SMFC for both chambers (anode and cathode) with time. Four types(Zn, Al, Cu, Fe) of metals and graphite felt as non-metallic electrode were examined here. Anodic part was not modified any mediatorbut coverednaturally microbial environment condition.As recent studiesshow,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 [27, 28], making it possible to establish mediator-less MFCs. To date, many metal reducing bacteria, which are capable of the reduction of solid metal oxides ability including *Clostridium Butyricum*[29], *Geobactersulfurreducens*[30], *Rhodofoferaxferrireducens*[1] and *Shewanellaputerfaciens*[31]. Furthermore, in the SMFC system, growth and metabolism of *S. puterfaciens* were dependent on the presence of active anode-in essence, it served as the electron acceptor for growth and metabolism. Anodic potentials in our cellranged 184 to -155 mVfor Zn, 155 to -150 mV for Al;148 to -126 mV for Cu;161 to -42 mV for Fe and 178 to -5 mV for Graphite feltfollowed for 72 h, whereas the cathodic potentials showed a slow drift. Metallic anode has shown larger ORP change than that in non-metallic graphite electrode. The results in figure 1 show that as time progresses the potential of the anodetends to decrease.It might be due to corrosion decreases over time for changes in the properties of the electrodes' surfaces resulting from biofilm buildup and polarization effect. Generally biofilm deposit promotes corrosion in aerated waternot in anaerobic water.Oxidation reduction potential changes were higher for metallicanode than for non-metals because of variation in corrosion rate.Sufficient microbial growth guarantees formation of a certain thickness of biofilm. According to electromotive series Al is more reactive than Zn, and Cu metal is much lower than Fein potential value. **The potential of A**l is higher than that of Zn, which is attributed to itsmicrostructure as well as slightchanges with type,concentration, and temperaturearound the metal. In case of Fe and Cu, it might be due to overvoltage (producing hydrogen from H_2S) phenomena in Fe which is not absence inCu. 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 mV_{SCE}[19, 30, and 32].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[24].

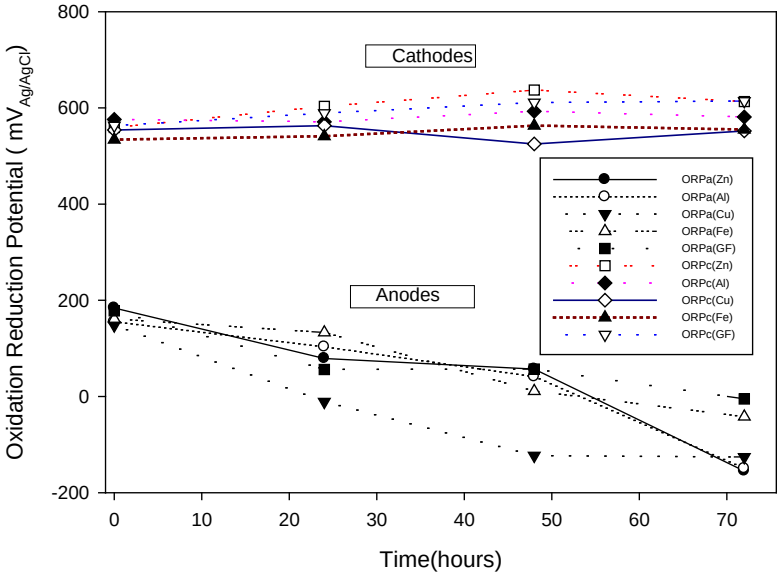

Figure 1. Oxidation reduction potential (ORP) variations in the cathode and anode (against Ag/AgCl)in our sediment microbial fuel cell.

3.2 Power Density of SMFC

From the measurements repeated several times, power density of SMFC using different metallic and non-metallic anode was depicted in figure 2.However, SMFCs were started with variety of anode distinctly and was run till 72 hours. We used Zn, Al, Cu, Fe, and graphite felt (GF) as anodic material. 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 (646 mWm^{-2}), GF (127 mWm^{-2}) varying with time. At beginning time of SMFC, the power density decreasesover time significantly (10 h) for all anodes but interestingly Zn anode was found to increase after that time. Increasing trends of power density of Zn anode have sustained till 45 hours after that drop. Power densities were shown to be different for different anodesprobably because of some various catalytic interactions on the electrode surface.Indeed,power densitiesof metallic anodeswere higher than non-metal graphite felt, because corrosion and microbial activity provide electrons to metal whereas only microbial activity provideselectrons to non-metal surface. Bacterial adhesion capacity to electrode might causevariation in power density. And the total area of the electrodes including the internal surface area [22], apparent surface area [32, 33], or projected area [19, 34] has been adopted computationof power density in MFCs. Schroder et al. [10] obtained a power density of 6.0 $Wm^{-2}$for 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 thenthe performance gradually decayed over time[14].

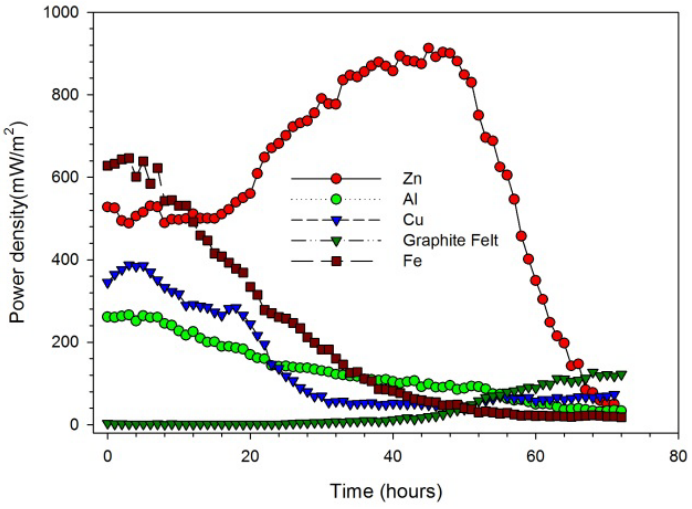

Figure 2. Power density of SMFC according to different anode with respect to time.

3.3 *Cell Voltage and Current density of SMFC*

Figure 3 presents the cell voltage and figure 4 represents the current density of SMFC with adding neither external substrates nor 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.Corrosion and microbial activity vary over time and condition of solution. The overall pattern of electricity generation was analogous to that observed by Holms et al. [35, 36], 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 [37] and Mathis et al. [38] 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 densityamong 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 trend 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 (Fig.3). According to anode, the observed maximum cell voltages were Zn(777 mV), Al(751 mV), Cu (645 mV), Fe (734 mV) andGF (499 mV) over different time.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 (E_{corr}) of one metal in the positive direction while another type can shift corrosion potential (E_{corr}) of some other metal in the negative direction [39,40].

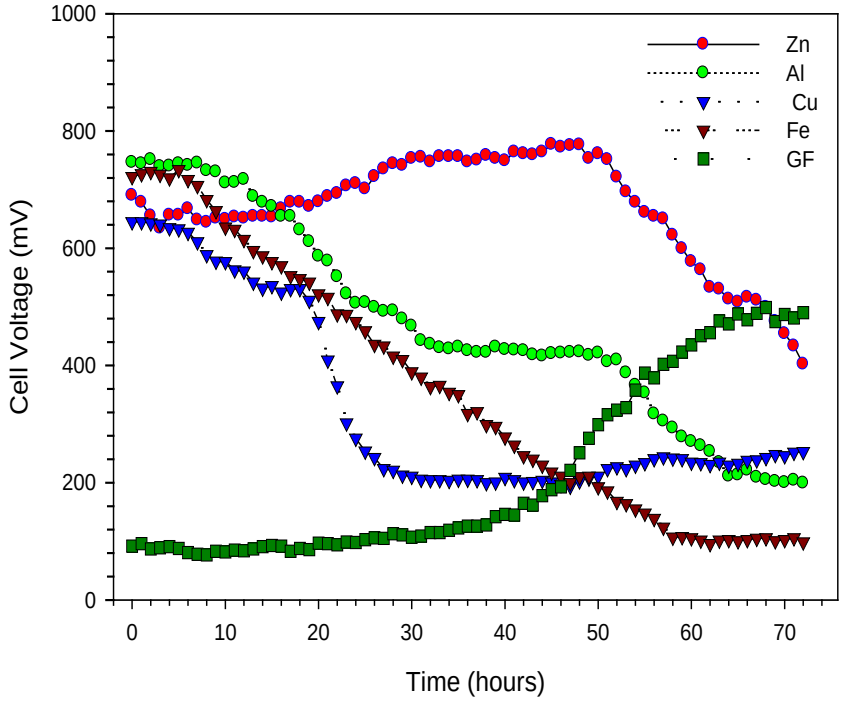

Figure 3. Cell voltages of SMFC with respect to time.

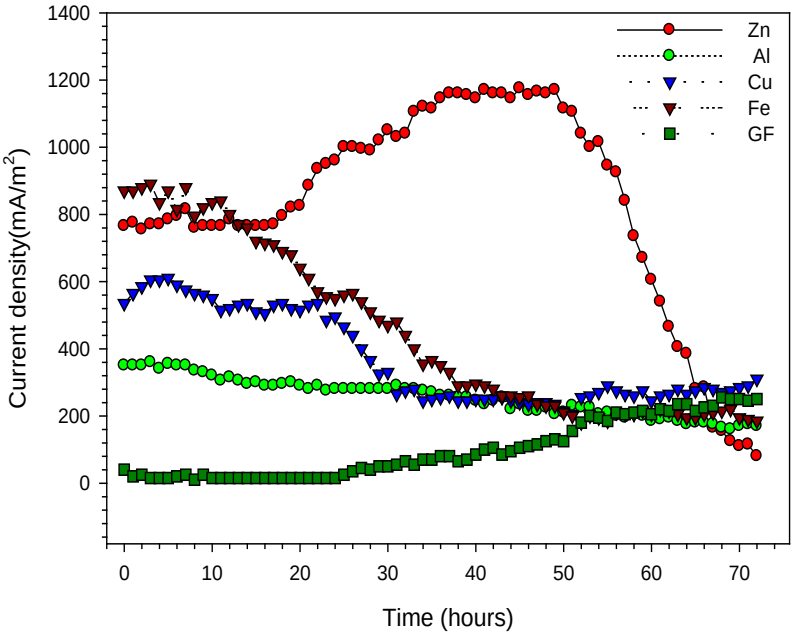

Figure 4. Current density vs time of SMFCoperation (surface area 0.002 m^2 and internal resistance 20Ω).

3.4pH and chemical oxygen demand (COD) of SMFC

pH of SMFCin this workhas changed over time at both 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 6.4in 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 GFover starting time and ending time, respectively. But, in cathode part, pH changed very little (7.6~7.7). pH reduction indicated that anodic environment is acidic and acidic media reduces the corrosion rate of metal. Metalsulfide or metal oxides are likely to reduce pH value. Acidic environment in anode part is an important issue because MFCs that are able to operate at low pH are technologically advantageous. The proton transport rate from anode to cathode is increases and the kinetic barrier for O_2 reduction to H_2O at the cathode decreases, which leads to the higher current and power densities[41, 42]. In acidic medium acidophilic microorganisms that have colonized the anode and cathode surface can oxidize glucose and otherorganic compounds in the absence of redox mediators [43]. It is well known that*Acidiphilium spp*. is able to use organic compounds such as glucose and glycerol as electron donors [44, 45, and 43]. Chemical oxygendemand of SMFC wasdecreased over time in case of all anodes.But losses of organic matter,shown in COD decreases

might be facilitative for long timeelectricity generation. 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 biologically degraded, so that COD removal is typically lower than that for biochemical oxygen demand (BOD) [46].

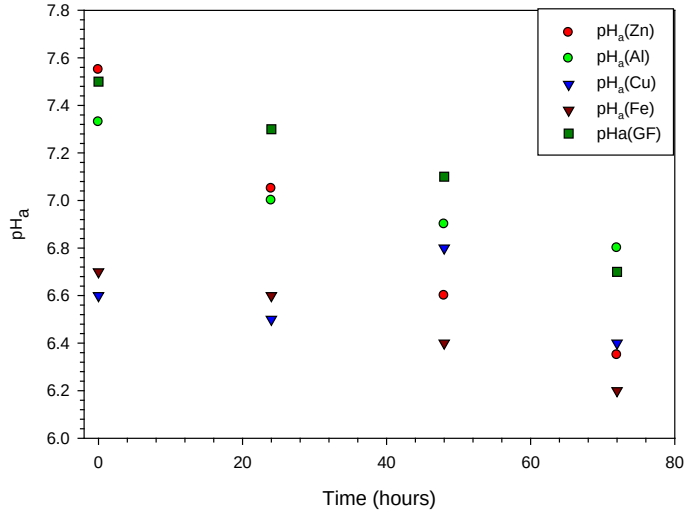

Figure 5.pHchanges in anode chamber over time of SMFC (pHa, anodic pH).

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 thata 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. Some interfacial reactions or phenomena on metal or non-metal surface need to be investigated further as mentioned in Section 3. Also time duration of the cell and limitation of current generation could be next issues.

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