



INTERMITTENT ENERGY HARVESTING IMPROVES SEDIMENT MICROBIAL FUEL CELL PERFORMANCE

Internship Report – Nova Innova – Feb-Aug 2022

Abstract

This study reports on the response of sediment microbial fuel cells to intermittent and continuous energy harvesting. Ceramic separators made from terracotta and terracotta with 20% bentonite were employed. The aim of this study was to investigate whether intermittent energy harvesting with a 20% duty cycle would provide higher power density than continuous loading. Results show that continuous energy harvesting provides more average power over a 1 hour period. Intermittent energy harvesting showed relatively more power, with the 20% duty cycle providing 32%-46% of the power of a continuous cycle. Current densities during intermittent cycles were 2.6 to 4 fold higher than during continuous operation. Performance tapered off over the course of the experiment for both strategies. Cells using separators with bentonite performed better during intermittent harvesting while terracotta cells outperformed during continuous operation. Cathode overpotentials were identified as the rate limiting factor during operation.

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LAY SUMMARY

In virtually every soil and sediment around the world live bacteria that are able to transfer the energy gained by breaking down food to their outside environment. These *exoelectrogenic* bacteria do this, simply said, to breathe. When we digest food, the process of digestion frees up electrons which are used to power our daily lives. But these electrons need to go somewhere, and that is why we breathe. Oxygen is very good at taking up these electrons and we then breathe them out as carbon dioxide. For exoelectrogens, this process is slightly different as they can only live in anaerobic environments – areas where no oxygen is present. The electrons gained when they break down food are moved to their outside environment where they use other elements like sulfur, nitrogen and metals as the terminal electron acceptor. Microbial fuel cell (MFC) technology harnesses this process by providing an electrode (the anode) on which these bacteria can live and ‘breathe’ and subsequently uses the electrical energy provided to generate power. In an MFC, the electrons do eventually react with oxygen on another electrode (the cathode), but this compartment is kept separate from the anode compartment where the bacteria reside. When electrons are released during oxidation of organic matter, so are protons. These also migrate to the cathode compartment to close the cycle. A separator is placed between the cathode and anode compartment to prevent oxygen from leaking into the anode and allow a path for protons to migrate. In the past these separators were often made from expensive cation exchange membranes, but in recent years it was found that cheap ceramic separators perform just as well if not better. Even though the currents of MFCs cells are small, they can be used for low-power electronics. The fuel cells are also good candidates for wastewater treatment as the bacteria degrade matter in the water efficiently when provided with a good environment to live in.

Nova Innova is a start-up company that integrates MFCs in designs. Previously they have created the Living Light, a plant MFC in which exoelectrogenic bacteria use the sugars released in the soil during plant photosynthesis to power a light. This research was done in the context of their POND project, which comprises a sediment MFC powering a set of water quality sensors and LED lights in a dome that translate the sensor information to color. Over the course of their research, the company has mostly focused on materials and fuel cell design. Ceramic separators were also a novelty in this company and the effects of different compositions of base material are actively being researched, which is why they were also a focus of this study. This work presents the first foray into variation of harvesting strategies for Nova Innova, with the aim of improving power generation. Most MFC research has focused on continuously drawing energy from the cells, often resulting in low power. In this work the energy was harvested intermittently with a 20% duty cycle, meaning 15 seconds of discharge and followed by a 60 second recovery period. On average this resulted in 32%-46% of the power of continuously harvesting while the cell was only on 20% of the time, making intermittent harvesting roughly twice as effective. Power spikes at the start of each discharge period suggest that power is being stored rapidly by the bacteria. The limiting factor in this experiment was the cathode compartment, most likely caused by inefficiencies in the cell's materials and design.

INTRODUCTION

Microbial fuel cell (MFC) technology is an emerging field in which the electrochemical activity of bacteria is captured as electrical energy. The unique biology of species such as *Geobacter sulfurreducens* and *Shewanella oneidensis* allows them to reduce extracellular material in the anaerobic environments they inhabit (Bond & Lovley, 2003; Wu et al., 2013). While energy yields are low, often in the micro- to milliwatt scale, MFCs provide interesting solutions for both wastewater treatment and decentralized, local power generation for low-power applications (Liu et al., 2004; Walter et al., 2020). An MFC in its simplest form consists of an anode, a cathode and, possibly, a separator. Consumption of organic matter by

$$V_{op} = E_{thermo} - [(\eta_{act} + \eta_{ohmic} + \eta_{conc})_{cathode} + (\eta_{act} + \eta_{ohmic} + \eta_{conc})_{anode}] \quad (1)$$

electroactive bacteria (EAB) produces electrons and in the anaerobic environment where the anode is located, the anode serves as the terminal electron acceptor. Protons freed up by the degradation of substrates travel through the membrane or separator and react with the electrons and an oxidant (i.e. oxygen) in the cathode compartment, generating electrical energy over a connected load. The operating voltage of an MFC is the product of the operating voltage of its individual electrodes. While the voltage output of each electrode can be theoretically determined by the Nernst equation, the actual operation voltage is typically lower in practice due to overpotentials. The main sources of these overpotentials are activation losses, ohmic losses and mass transport losses. Actual operating voltage (V_{op}) can be calculated as follows:

where E_{thermo} represents the thermodynamically predicted operating voltage, η_{act} represents activation losses from reaction kinetics, η_{ohmic} are losses associated with electrical and ionic resistances and η_{conc} represent losses in reactant concentration from mass transport limitations (Rismani-Yazdi et al., 2008). In most MFCs V_{op} is governed by losses in the cathode compartment. Parasitic losses stem from substrate crossover between anode and cathode compartments, and

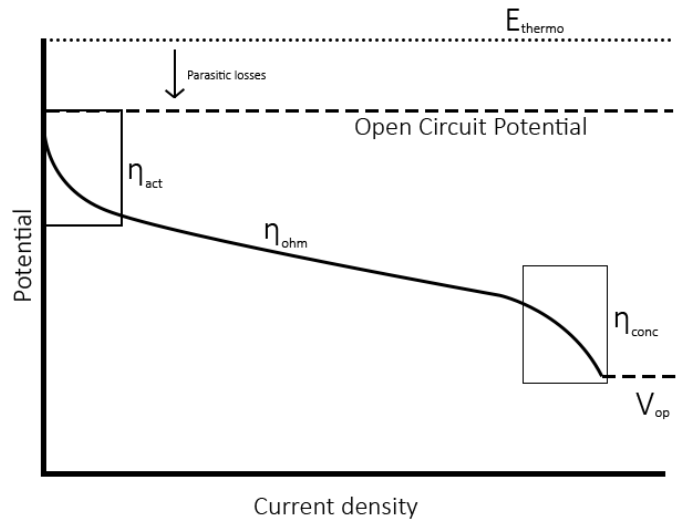


Figure 1 - Overpotentials as a function of current density. Adapted from Rismani-Yazdi et al.

unwanted side reactions that take away current from the desired reaction.

Separators perform two important tasks in the cell, being 1) allow the flow of ions between the anode and cathode compartment while maintaining concentration gradients and 2) prevent oxygen diffusion from the cathode compartment to the anode. In the search for durable and low maintenance MFC architectures, ceramic separators have proved to be a worthy alternative to the expensive ion exchange membranes (IEM) from conventional fuel cell technology. (Winfield et al., 2013, 2016; Yang et al., 2016). Conventional IEMs such as Nafion are prone to biofouling and inhibition of proton transport due to cations occupying functional sulfonate groups and require frequent replacement (Choi et al., 2011). Furthermore, while the relative preferential conductivity of protons compared to other cations is three- to sixfold higher than other cations, the concentration of protons is often 4 to 6 orders of magnitude smaller than cation concentrations in MFC anolyte solutions (Stenina et al., 2004). Ceramic separators make up this lack in selectivity with increased ion mobility. The porous structure facilitates cation transfer and also allows anion transfer, improving the pH balance in the cell (Harnisch & Schröder, 2009).

While most research into MFCs in the past has focused on continuous energy harvesting (CEH), an increasing amount of literature suggests that intermittent energy harvesting (IEH) can increase yields substantially (Dewan et al., 2009; Ieropoulos et al., 2005; Ren, 2013; Walter et al., 2014). In IEH, a cell is subjected to a duty cycle: it is discharged for a short period of time and allowed to recharge. EAB have the ability to store charge in the biofilm by forming polymers, typically for long term storage, or by reducing cytochromes and flavins for short term storage (ter Heijne et al., 2021). This grants electroactive biofilms pseudocapacitive properties which can be harnessed with IEH. The charging of cytochromes and flavins can happen in a matter of milliseconds. The utilization of this pseudocapacitive nature of biofilms allows for bursts of current higher than in CEH, and subsequently higher power output. Furthermore, there is evidence that IEH can alter biofilm morphology, creating mushroom-like structures that facilitate nutrient transfer to and waste transfer from the biofilm. However, this effect was only observed when the biofilms subjected to an IEH regime starting from inoculation and after at least 10 days of operation (Zhang et al., 2018). The choice of separator also matters for IEH, as it was shown that ceramic separators were preferable to IEMs during intermittent operation (Walter et al., 2014).

This research was carried out for the benefit of the *POND* project by Nova Innova, a start-up company that combines research and design. *POND* is a water quality measuring device powered by an MFC situated in the sediment. It is designed to be low power and low maintenance. An added feature is a dome containing RGB LEDs, which can change color based on the sensor readings. The MFC does not power the electronics directly, as that would require a parallel charging stack which is vulnerable to effects such as voltage-reversal. Rather, *POND* uses a *multi-harvester* capable of hosting 4 MFCs. Each cell is individually connected to a DC/DC boost converter IC at a preset harvesting voltage. The IC steps up the voltage and stores the harvested energy in a 3.3V battery for later use. Charging efficiency is dependent on both voltage and current. Broadly speaking, the higher the current, the higher the efficiency. IEH provides higher current output and is therefore expected to charge a battery more

efficiently. Nova Innova only has previous experience with CEH and conventional IEMs. This research is their first exploration into the viability of ceramics and IEH for their prototypes.

MATERIAL AND METHODS

SEDIMENT

Sediment was collected from the camel pond in Blijdorp Zoo in Rotterdam in sealed buckets and stored at room temperature. MFC's were fed with new sediment monthly. No signs of decline due to nutrient deficiency were detected during experiments.

CERAMIC SEPARATORS

Materials used were terracotta (10 Giet, Witgert, DE) and terracotta mixed with 20% w/w bentonite. Clay slips were made using a mold and left to dry. After the drying period, the ceramic disks were fired at 1110 °C in a furnace. After firing, material was left to cool and weighed. Ceramic material was then submerged in boiling water for 2 hours and cooled for 2 hours in cold water and weighed again. Porosity was determined according to Eq. 2.

$$Porosity = \frac{(Boiled\ weight - Dry\ weight)}{(Dry\ weight)} * 100 \quad (2)$$

The porosity of the terracotta separators was determined to be 14.28% and separators with bentonite at 19.23%.

MFC CONFIGURATION

Figure 2 shows the MFC design. Electrodes consisted of two activated carbon felt (ACF) disks with a thickness of 5 mm and a diameter of 130 mm (projected surface area 132,7 cm²) for both the cathode and anode. Titanium wire was woven tightly through the electrodes in a triangular pattern to provide good electrical contact. Resistance measurements after weaving were performed with a multimeter by measuring from the end of the titanium wire to various points on the surface of the electrode to ensure a resistance between 3-6 Ω. The electrodes were encased in a 3D-printed housing (PETG, Prusa filaments, Czech Republic). On the

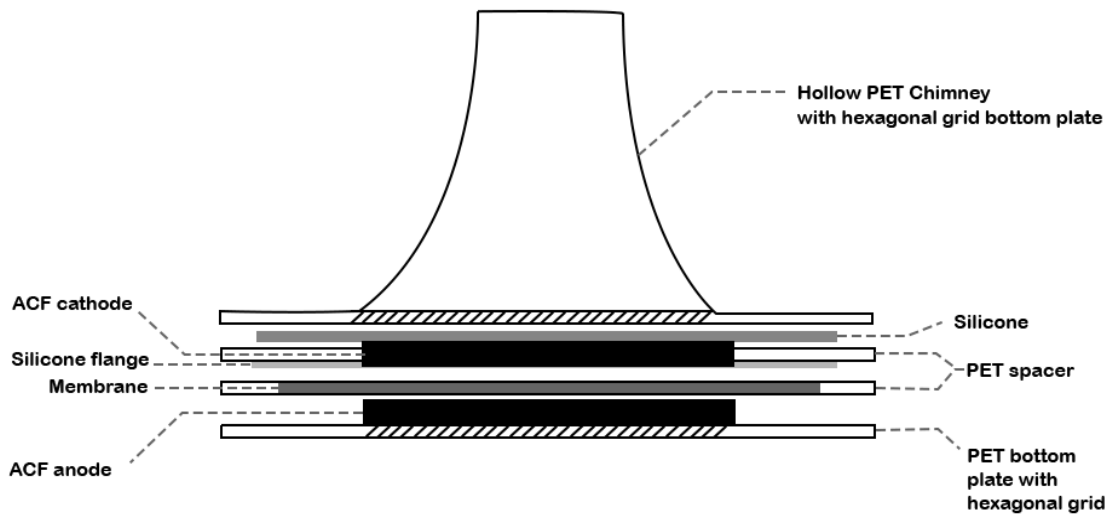


Figure 2 - Schematic diagram of MFC design.

cathode side, the cathode was separated from an air column in a central chimney by a 131 mm diameter O₂-permeable 100% silicone disc (Rubbermagazijn, the Netherlands). To maintain equal pressure on the entire sandwich, a hexagonal grid was used as the chimney base and the bottom plate. A 131 mm diameter 90% silicone ring between the membrane and cathode was used to prevent water leakage. The membrane/separator material varied between experiments but followed the same principal design. The anode was then placed against the separator and closed off with a bottom plate featuring the hexagonal grid to maintain pressure and allow nutrient exchange with the sediment. Tightening was done using 12 stainless steel bolts, washers and nuts and tightened crosswise using a power drill.

MFC OPERATION

MFC's were placed in plastic tubs containing mud with a layer of water on top. To provide heating, these tubs were placed in another plastic container filled with water and an electrical heater regulated at 25 ± 2°C. For the start-up phase the following protocol was developed. An electrical load of 1000 kΩ was connected between the anode and cathode. Cells were monitored and resistors were changed when the operating voltage reached 550 mV. Resistance was lowered stepwise from 1000 kΩ to 2.2 kΩ (table 2) when the operating voltage reached 550 mV again after lowering the resistance. After

lowering the resistance, the voltage was monitored. If the operating voltage dropped < 10% within 4 hours of connecting a lower resistance, the resistor was changed to the next step. If the operating voltage dropped less than 5% immediately after switching, the next resistor in line was also applied. The minimal value of 2.2 kΩ corresponds to the lowest value resistance value of our sensing equipment.

Table 1 – Resistor values used during MFC startup

Resistor values

1000 kΩ

100 kΩ

51 kΩ

20 kΩ

10 kΩ

5.1 kΩ

2 kΩ

DATA COLLECTION & HARVESTING

MFC's were connected to a proprietary datalogging system that measured voltage and temperature.

Data was measured and sent wirelessly every 5 seconds to a central server for display. Current and power were calculated according to Ohm's law using voltage and resistance. Dataloggers were also outfitted with an energy harvesting system (BQ25504, Texas Instruments, USA) capable of operating cells at a set voltage by regulating current with a MOSFET. In this mode, current was determined by measuring the voltage over a sense-resistance set at 2.2 k Ω . This was the de facto lowest possible resistance during harvesting. For the intermittent harvesting research, the sense resistance was brought down to 35 Ω to allow higher current and power as the 2.2k resistor was determined to be a limiting factor.

INTERMITTENT VS. CONTINUOUS OPERATION

Intermittent harvesting was performed using a Python script which could switch the harvesters on and off based on certain parameters like time. Various settings were tested internally (results not shown). Ultimately the method used in this study follows an adaptation of the approach used previously in a multi-anode setup (Gardel et al.,

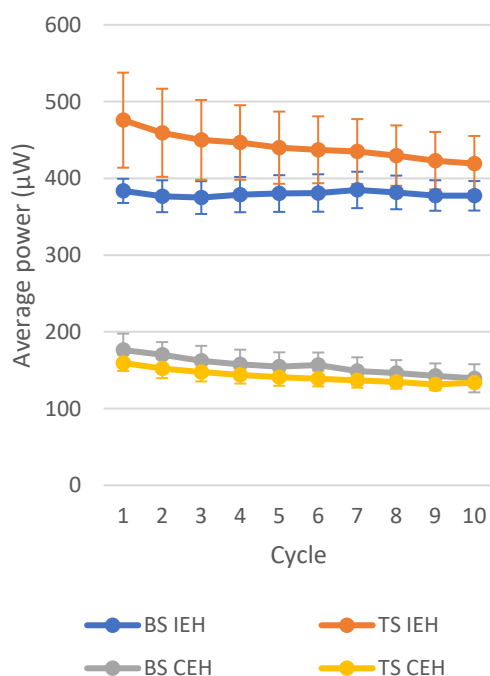
2012). This consisted of a 15 second ON period, after which the system switched the harvester for that cell off and moved to the next cell. The duty cycle was therefore determined by the total number of cells, in this case 5, resulting in a 20% duty cycle. Intermittent harvesting was performed at a voltage of 350 mV for 40 cycles (one ON period for each cell), followed by a 3 hour rest period under open cell conditions. Cells used were 2 cells containing a terracotta separator (TS) and 3 cells using a terracotta separator with added bentonite (BS). Afterwards the cell was set to harvest continuously at 350 mV for one hour, followed again by a 3 hour rest period after which the cycle restarted. This cycle was repeated 10 times. Anode and cathode voltage were measured against an Ag/AgCl reference electrode during operation.

RESULTS AND DISCUSSION

POWER AND CURRENT PRODUCTION

During intermittent operation, average power generation over 1 hour ranged from $176 \pm 21 \mu\text{W}$ to $139 \pm 21 \mu\text{W}$ after 10 cycles for BS, while TS ranged from $159 \pm 10 \mu\text{W}$ to $134 \pm 3 \mu\text{W}$ over the same

(A)



(B)

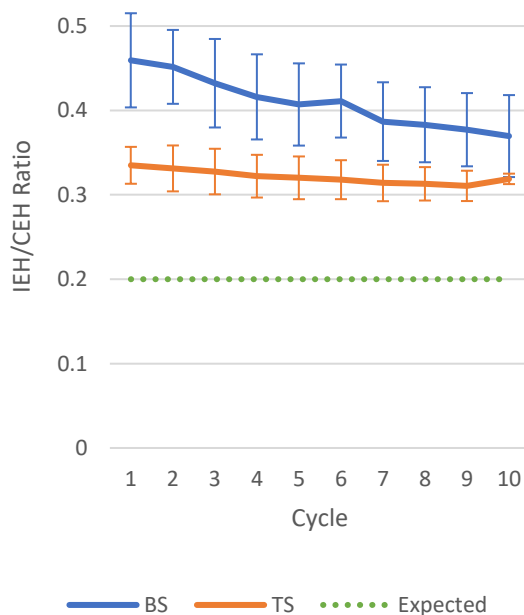


Figure 3 – (A) Average power generation during 1h of intermittent and continuous operation for MFCs with terracotta separators and terracotta + 20% bentonite separators over 10 IEH-CEH cycles. (B) Ratio of average yield from IEH to CEH during 1h of operation over 10 cycles and the expected ratio of 20% for a corresponding duty cycle. Error bars represent one standard deviation from the mean.

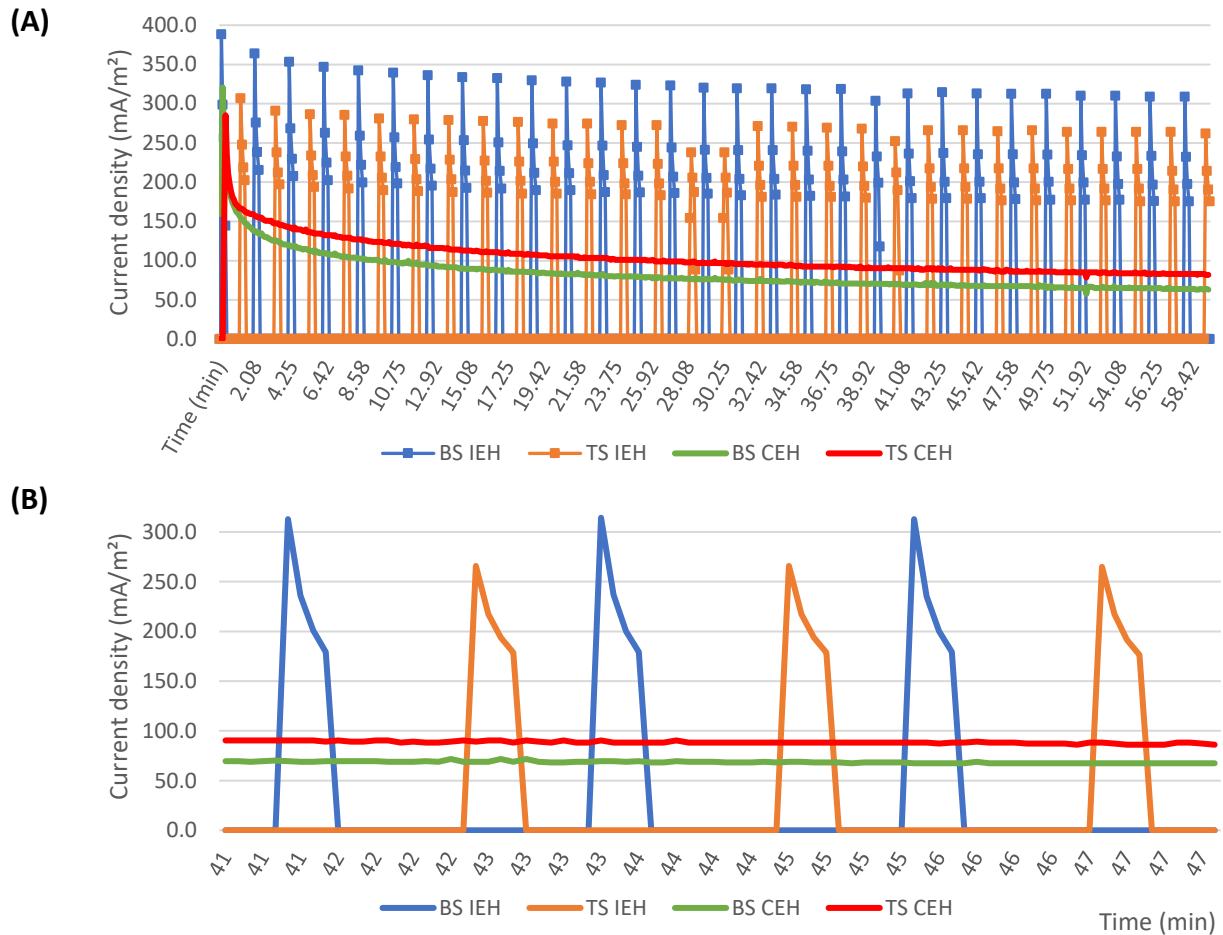


Figure 4 - Average current density for IEH and CEH over 1h (A) and an excerpt of the same graph (B).

period. Average power over one hour during continuous operation ranged from $384 \pm 16 \mu\text{W}$ to $377 \pm 19 \mu\text{W}$ for BS and $476 \pm 62 \mu\text{W}$ to $419 \pm 36 \mu\text{W}$ after 10 cycles (Fig. 3). In absolute terms, CEH yielded higher average power during one hour of operation. However, during intermittent operation cells were operated on a 20% duty cycle.

Assuming a linear relationship between duty cycle and yield, average power generation should be $77 \pm 3 \mu\text{W}$ to $75 \pm 4 \mu\text{W}$ for BS and $95 \pm 12 \mu\text{W}$ to $84 \pm 7 \mu\text{W}$ for TS over 10 cycles. When comparing the ratio of IEH/CEH, BS measured 0.459-0.369 for and TS was 0.335-0.319 from cycles 1 to 10. This is an efficiency improvement of 230-185% and 167-159% for IEH over CEH for BS and TS respectively. This increase can be accounted for by current density peaks at the start of each discharge cycle (Fig.4). During IEH, current density (Fig. 4) at each peak was about 4 times ($327.91 \pm 57.25 \text{ mA/m}^2$) higher for BS and 2.6 times higher for TS ($274.46 \pm 37.67 \text{ mA/m}^2$) than during CEH ($82.35 \pm 2.76 \text{ mA/m}^2$ and $104.23 \pm$

15.1 mA/m^2 respectively).

The cells in this experiment alternated between CEH and IEH, and thus the two harvesting strategies interfered with each other. Using two sets of cells with each performing just one of the strategies might yield a better picture of performance over time. However, finding enough cells that were similar was challenging in the environment in which this research was performed. Different duty cycles may also yield different results, as it was noted by Zhang et al. that a 50% duty cycle with each phase lasting 10 seconds yielded the best result. With the hardware used in this experiment, such cycles were near impossible as signal transfer and transfer of results to the database coincided, resulting in errors and requiring a waiting time before sending the signals to prevent this, and thus lengthening either the charge or the discharge period. Decoupling the data transfer and signal transfer or hardware with an on-chip timing circuit would be optimal to achieve shorter periods.

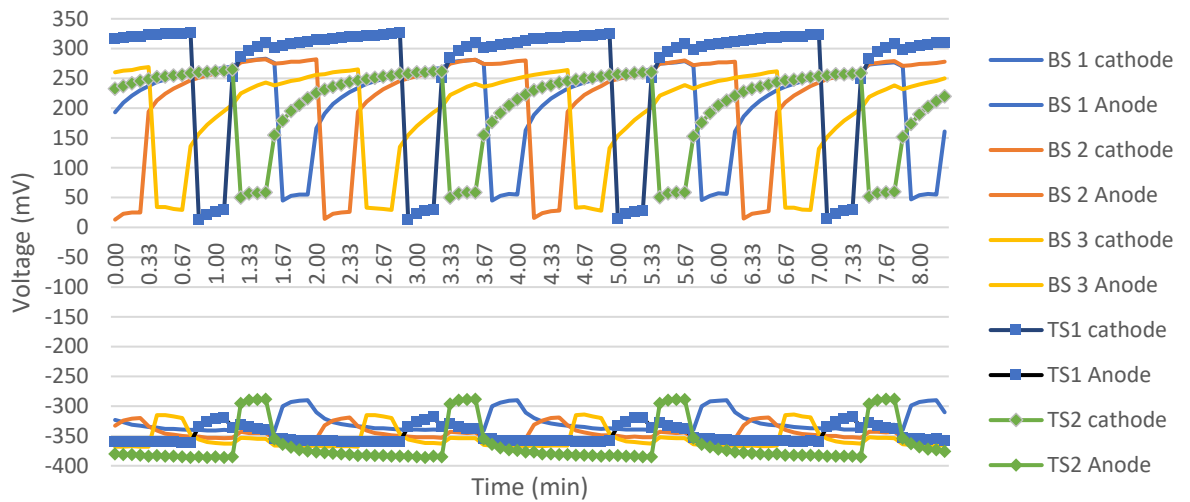
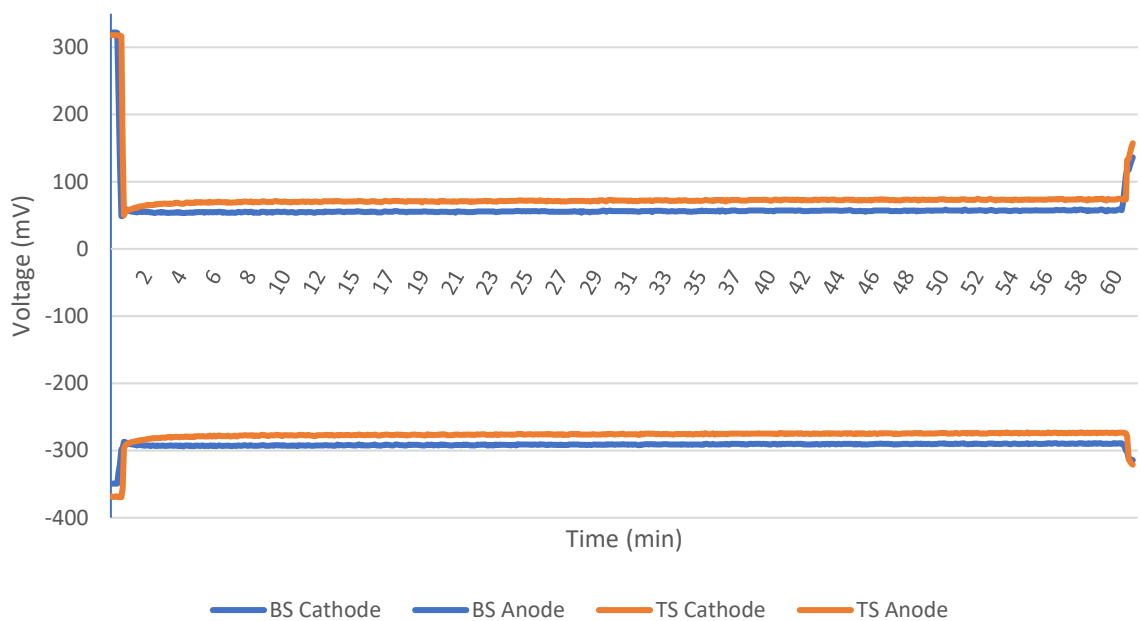
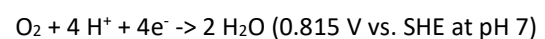
(A)**(B)**

Figure 5 - Anode and cathode potentials during IEH (A) and CEH (B).

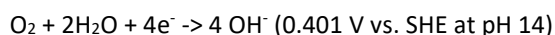
ELECTRODE POTENTIALS

Cathode potential was more affected during IEH discharge cycles, dropping by as much as 300 mV, while anode potential rose by a maximum, of 97 mV (Fig. 5). This indicates that this system is cathode limited. Carbon-based electrodes like ACF without catalysts exhibit relatively slow ORR rates (Gil et al., 2003; Šljukić et al., 2005), causing relatively high η_{act} . Oxygen depletion at the active sites due to poor solubility of oxygen and adsorption of reaction products to the active sites of the cathode prevents the adsorption of fresh O_2 to continue the reaction (Dange et al., 2022). The introduction of any kind of catalyst could reduce adsorption times and increase

the maximum current density. While some water is needed for reactant transport, this water in the cathode compartment also impedes O_2 mass transfer to the active sites. The addition of a water dispersal layer (e.g. PTFE, PVA) could facilitate higher current densities by removing water from the close cathode environment (Walter et al., 2018). It has also been reported that OH^- accumulation in the cathode causes overpotentials as the ORR E_{thermo} is dependent on pH (Motoyama et al., 2016; Yuan et al., 2013). At higher current densities especially, a lack of available protons causes the ORR to shift from its favorable reaction:



to its less favorable side:



Cation migration due to the electromotive force increases cathode pH while protons are used up in the ORR (Rozendal et al., 2006). This increases this effect as the working potential of the cathode falls by -59 mV per unit of pH following the Nernst equation. The pH of the electrode (micro)environment was not measured over the course of these experiments but could provide insight into the influence of proton depletion/OH⁻ accumulation in this system.

DIFFERENCE IN PERFORMANCE OF SEPARATORS

While BS cells outperformed TS cells during the IEH experiments, their performance was slightly worse during CEH. Overpotentials increase and shift in character with increasing current density (Fig. 1). Literature suggests that the addition of bentonite increases cation exchange capacity and current density, which was the main consideration when selecting this material (Ghadge & Ghangrekar, 2015). However, these findings suggest that this material is only preferable at higher currents. A possible explanation is that the increased cation

exchange capacity of BS allows for more ions to transfer to the cathode compartment. This higher cation concentration might increase η_{ohm} through increased ionic resistance. The porosity of BS was also higher than TS (19.23% vs. 14.28%), which could have a beneficial effect on mass transfer and thus better performance at higher current density. Thorough characterization of ceramic separators is required to determine with greater accuracy which of the factors (porosity, ion exchange capacity, diffusivity) yields the desired result. Because of the slight differences seen in this study, different energy harvesting strategies might call for different types of separators.

CONCLUSION

Intermittent energy harvesting is a promising harvesting strategy for sediment microbial fuel cells. With a duty cycle of 20%, cells produced 32%-46% of the power compared to a continuous

energy harvesting strategy. Cell performance during either harvesting strategy was dependent on separator material, with terracotta cells performing better during CEH and terracotta with 20% bentonite outperforming during IEH. Current densities during the initial discharge period were 4 times higher in BS and 2.6 times higher in TS during IEH than during CEH. Cathode overpotentials had the greatest influence on power density and steps should be taken to improve the current MFC design to alleviate these to achieve higher possible power density.

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