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Life Cycle Environmental Impact Comparison of Bioelectrochemical Systems for Wastewater Treatment

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Abstract

Bioelectrochemical systems (BESs) are developed to transform the energy harvested from the biomass into electricity. Different types of BESs including microbial fuel cell (MFC), microbial electrolysis cell (MEC), and microbial desalination cell (MDC) are under intensive research and development; however, their life cycle environmental impacts have not been systematically compared to identify the most environmentally friendly BES. To understand and eventually help reduce the environmental impacts of different BESs, life cycle assessment (LCA) models were developed in this study to assess and compare their potential environmental impacts. The results indicate that the MEC has better environmental performance than the MFC and MDC due to the large hydrogen peroxide production in the operation phase. The environmental performance of the MFC and MDC can be improved by the increase of power density, but their environmental impacts, at a relatively high power density that can be achieved by current technology, are still higher than the environmental impacts of the MEC at current power density. When the environmental impacts are benchmarked with those of the traditional wastewater treatment methods, the MEC has a better environmental performance, whereas the MFC and MDC have relatively large environmental impacts.

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Keywords: Life cycle assessment; bioelectrochemical systems; environmental impact comparison

1. Introduction

Biomass is the fourth largest energy source, and it is one of the most important renewable energy that can be transferred into different forms [1]. The bioenergy can be collected from biomass grown in nature or grown in purpose. The biomass has different forms, such as solid, liquid, or gaseous forms [2]. A bioelectrochemical system (BES) is such a system that captures the chemical energy stored in biomass, and transfers the energy into electricity or chemical products. Due to the increase of global population and the scarcity of freshwater, BESs can be one of the potential solutions to reclaim water with electricity generation. Depending on the operational modes and end products, BESs can be generally divided into five categories: microbial fuel cell (MFC), microbial electrolysis cell (MEC), microbial electrosynthesis (MES), microbial solar cell (MSC), and microbial desalination cell (MDC) [3-4]. From all BESs, the representative BESs are the MFC, MEC, and MDC [5]. Each BES has its own distinct characteristics and applications. For example, the MFC has been widely studied for its capacity to generate electricity by oxidizing the organics in wastewater, and its ability to operate at low loading rate [6-8]. The MEC uses external electricity to decrease the cathode potential, and in doing so, it produces hydrogen and other value-added chemicals [9, 10]. When compared with the MFC and MEC, the MDC has emerged more recently. It uses the potential

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difference between anode and cathode chambers to simultaneously treat wastewater and desalinate seawater [3, 11]. Additionally, all the BESs can utilize low-strength and diverse substrates, which is a desired characteristic because of the increasing demand of treating low-carbon footprint wastewater.

Although various advantages have been presented in BESs, the environmental concerns have been raised for different operation systems. It is doubtful whether the three representative BESs are all sustainable at the same level at current power density [12]. As a result, a comprehensive life cycle environmental comparison of the three BESs is imperative, and could be used to guide the sustainable development of the technology for future commercialization and deployment. Until now, several studies regarding the environmental impacts of different BESs have been conducted. Foley et al. (2010) conducted a life cycle assessment (LCA) research, from the raw material extraction to the operation phase, of a conventional anaerobic digestion (AD) system, a MFC, and a MEC with the assumption that the power density is 500 W/m_{anode}^3 . They concluded that the AD treatment and MEC had better environmental performances than the MFC in global warming potential (GWP) and ozone depletion potential (ODP), and the MEC had better environmental performance than the MFC in acidification potential (AP), ecotoxicity potential (ETP), eutrophication potential (EP), and respiratory inorganics (RI). They also found that the most energy-intensive process was the energy consumption in the operation stage. However, the environmental impact results were calculated based on a power density that cannot be achieved at current stage of development, which may overestimate the environmental performances of the MFC and MEC [13]. Pant et al. (2011) carried out a study to make suggestions on how to conduct an LCA on BESs, without providing any quantitative data analysis [14]. Schemfe et al. (2018) integrated dynamic simulation, techno-economic assessment, and LCA methods to assess a BES which could simultaneously treat wastewater and produce formic acid. The authors chose to use 1 kg of formic acid as the functional unit, which may not be comparable with other studies that have the functional unit of the unit volume wastewater treated. Furthermore, this study omitted the environmental impacts generated form the end-of-life (EoL) for comparison purpose [15]. The most recent study was conducted by Zhang et al. (2018), and they carried out a cradleto-grave LCA on the MDC and MFC [16]. They concluded that, at current power density, BESs have relatively high environmental impacts when compared with traditional wastewater treatment methods. The environmental impacts, however, could be largely reduced with the increasing power density. Although various studies have been conducted to assess the environmental impacts of different BESs, no study has systematically compared the environmental impacts of these three representative BESs from cradle-to-grave. The comparison among different types of BESs can provide a roadmap for the sustainable development of each type of BESs. As a result, this study, for the first time, systematically compares the environmental impacts of the three representative BESs, and provides a guidance for their further sustainable development and deployment.

The objective of this study is to evaluate and compare the environmental impacts of the three representative BESs, and guide their development during design-to-manufacturing cycle. In this study, the manufacturing data of the three BESs were obtained from our lab experiments. Sensitivity analysis was conducted to identify the environmental impact reduction from the increase of power density.

2. Materials and Methodology

LCA is a systematic tool to evaluate the environmental impacts of a product or a system from its life cycle, involving scope and goal definition, life cycle inventory (LCI), LCA, and interpretation. LCA models have been developed in this study to investigate the environmental impacts of three different types of BESs. The processes to treat wastewater is described in section 2.1. The goal and scope are defined in section 2.2, while the LCI analysis is in section 2.3.

2.1. Process description

The water treatment process is based on the description illustrated in the literature [13]. All of the three BESs undergo wastewater treatment, while the MDC also undergoes the saline water desalination process. The wastewater treatment experiences three distinct stages: pretreatment, treatment, and post-treatment. The wastewater is pretreated by being pumped through screens and undergoes pre-acidification for 12 hours in a pre-acidification tank. Citric acid or caustic soda is added to the wastewater to adjust the pH value at this point. In the MDC, the pretreatment of seawater is also required. Saline water is pretreated by the sand filters and cartridge filters. Solid wastes generated from the sand filters are disposed of through an underwater outlet. After filtering, antiscalant sodium tripolyphosphate is pumped into the saline water at 4 to 8 ppm doses to prevent clogging of the membranes. After pretreatment, the wastewater enters the BES. Detailed processes in the MFC, MEC, and MDC are illustrated below.

After pretreatment, the MFC uses bacteria to break down the organic substrates in the wastewater, with energy generation. Based on current experiment data, the power density of the MFC is set to be 10 W/m³anode, and the generated electricity is used directly on-site. The energy used to operate the MFC is 0.1 kW/m³anode. After pretreatment, the MEC also uses bacteria to treat organic substrates via catalytic breakdown. No electricity, however, is generated in the MEC operation due to the absence of oxygen in the cathode. Hydrogen peroxide is presumed to be generated in the cathode compartment in the MEC, with the addition of clean water and air. External voltage is required to apply to the MEC to operate the system. The hydrogen peroxide production rate is about 13.7 kg $H_2O_2/m^3 \cdot d$, while the operational energy of the MEC is 0.66 kW/ m³_{anode} [13]. The generated hydrogen peroxide is directly used on-site. After pretreatment, the MDC treats the wastes in the anode chamber, and desalinates saline water in the desalination chamber. The volume ratio between anode chamber and desalination chamber is 4:1. In the process, electricity is generated in the anode chamber, and the power

density is set to be 10 W/m_{anode}^3 according to current experimental data. The operational energy requirement of the MDC is 0.1 kW/m_{anode}. The manufacturing processes of the MFC and MDC were obtained from our lab experiments, and detailed description can be found in our previous publication [16]. The MEC is modified from the MFC, and it is assumed to have the same structure and material composition with the MFC.

After treatment in the individual BES, the post-treatment of wastewater is carried out by using dissolved air filtration (DAF) method before further downstream treatment. To assist flocculation, polymer and ferric chloride are added to the DAF. Truck is used to transport biosolids between different processes. The influent concentration and removal efficiencies are set to approximate the real condition, which represents a typical domestic sewage treatment containing food wastes as the major organic compounds. Specifically, the chemical oxygen demand (COD) influent concentration is 500 mg COD/L, and the COD removal efficiency is 90%. The schematic of wastewater treatment process is illustrated in Fig. 1. In the MDC, the post-treatment of seawater is also required. After desalination, the seawater is post-treated by adding calcium hydroxide to adjust the pH value [17]. The influent saline water in the desalination chamber is presumed to be seawater, and the desalination efficiency in the MDC is set to be 90% [18].

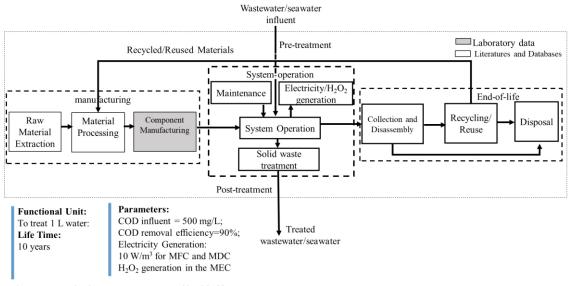


Fig. 1. Schematic of wastewater treatment process

2.2. Goal and scope definition

This LCA study aims to compare the environmental impacts of three typical BESs, and identify the potential methodologies to improve the process to treat wastewater and generate electricity or other useful chemical products. The environmental impacts of the three stand-alone systems were calculated and characterized with the ILCD (International Life Cycle Data System) method in GaBi 8.7 (www.gabisoftware.com). The functional unit of the three BESs is defined as 1 L of incoming wastewater. It is worth noting that in the MDC, the functional unit is defined as 0.8 L of incoming wastewater and 0.2 L of incoming sea water. In the MFC, the electricity generation is considered as a byproduct of wastewater treatment. In the MEC, the hydrogen peroxide generation is considered as a byproduct of wastewater treatment. In the MDC, the electricity generation is considered as a byproduct of wastewater treatment and seawater desalination. The overall environmental impacts were calculated by deducting the environmental credits from the life cycle impacts.

The system boundary covers the whole life cycle of three different BESs from cradle-to-grave, and it is illustrated in Fig. 2. Due to the complexity of BESs, its life cycle is divided into seven phases: raw material extraction, material processing, component manufacturing, pretreatment, operation, posttreatment, and EoL. Raw material extraction, material processing, and component manufacturing are collectively referred to as the system manufacturing. The operational phase consists of system operation, solid waste treatment, membrane maintenance, and electricity generation. Membrane maintenance is indispensable in the BES operation due to the biofouling and inorganic scaling problems [19-21], and the detailed maintenance process is described in the supporting information in our previous study [16]. The EoL phase consists of three different stages: BES collection and disassembly, recycling or reuse, and disposal. The BES collection and disassembly is modeled based on the regular battery collection and disassembly due to lack of data and similarities in functional electricity generation and system structure [22].



*note: transportation between processes are considered in this LCA

Fig.2. System boundary of the LCA on the BESs.

Additionally, recycling or reuse is presumed to recover valuable materials after disassembly. The detailed recycling and reuse ratios for different disassembled materials can be found in our previous study [16]. After reuse or recycling of valuable materials, the rest materials are presumed to be sent to landfill facilities. Land use and transportation between different phases are considered in this research.

2.3. Life cycle inventory

As shown in Fig.2, the LCI of the component manufacturing was obtained from our lab experiments, while energy and material consumptions in other phases were obtained from literature. The material and energy input inventories in the system manufacturing of the MFC and MDC are illustrated in our previous study [16]. The embodied mass and energy of the three different BESs were calculated through GaBi 8.7.

To treat 1 L of water, 4.95 kg of raw materials are used in the MFC, -1.25×10^5 kg of raw materials are involved in the MEC, and 4.63 kg of raw materials are acquired in the MDC. The large difference of the embodied mass consumed in the MEC, and MFC and MDC can be explained by the large amount of hydrogen peroxide produced in the operation phase in the MEC. According to the literature, about 13.7 kg/m³ of hydrogen peroxide are generated per day [13]. The large embodied mass saved by the hydrogen peroxide production is due to the relatively large amount of raw materials used to fabricate 1 kg of H₂O₂. Specifically, about 1.07×10^3 kg of raw materials are acquired to produce 1 kg of H₂O₂.

The mass embedded in the system manufacturing is further analyzed based on different components. 0.86 kg of raw materials are used to manufacture one functional unit of MFC, taking up 17.4% of total mass in the life cycle. The same amount of materials are consumed in the system manufacturing in the MEC, but only takes up a very small portion of embodied mass due to the dominant effect of the hydrogen peroxide. In the MFC and MEC system manufacturing, the cathode chamber takes up the majority embodied mass due to the material-intensive processes to manufacture carbon cloth used as the electron collector and polytetrafluoroethylene (PTFE) binder used to bind the powered activated carbon. In addition, 1.01 kg of raw materials are consumed to fabricate 1 functional unit of MDC, occupying 22.0% of the total embodied mass in the life cycle. In the MDC, the cathode and membrane chambers contribute to 44.0% and 43.0% of total mass, respectively. In the cathode chamber, the fabrication of PTFE is material-intensive. To manufacture 1 kg of PTFE, about 3.28E+4 kg of raw materials are consumed. The fabrication of the anion exchange membrane (AEM) and cation exchange membrane (CEM) is the major reason for the high embodied energy of the membrane chamber in the MDC.

The embodied energy to treat 1 L of water was also calculated based on the inventory inputs. About 9.72×10^{-2} MJ of energy is required in the MFC, -2.18×10^{3} MJ of energy is required in the MEC, and 8.32×10^{-2} MJ of energy is required in the MDC. The large difference of the embodied energy can be also explained by the large amount of hydrogen peroxide produced in the operation phase in the MEC. The embodied

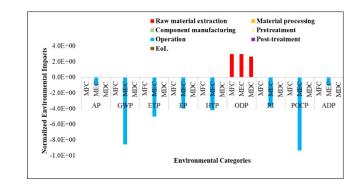
energy credits obtained from the operation phase completely override the embodied energy consumed by other phases in the MEC. The MDC has slightly less embodied energy than the MFC, and this can be explained by the different structures built to satisfy different functions of the two cells.

The energy embodied in the system manufacturing is also further analyzed based on different components. In the MFC and MEC, the same amount of energy 0.018 MJ is embedded in the system manufacturing, taking up 18.2% and 0.0% of total embodied energy in the MFC and MDC, respectively. The small percentage of the embodied energy for the MEC system manufacturing is due to the large embodied energy saved by the hydrogen peroxide production. In the MFC and MEC system manufacturing, the cathode chamber is energyintensive, which takes up 51.0% of embodied energy. The fabrication of PTFE binder is still the main reason for the high embodied energy of the cathode chamber. In the MDC, about 0.02 MJ of primary energy is consumed for the MDC, taking up 24.0% of total energy in the life cycle. In the MDC system manufacturing, the cathode and membrane chambers take up 39.0% and 42.2% of the total embodied energy, respectively. The high embodied energy of the cathode chamber is due to the fabrication of PTFE binder, while of the membrane chamber is mainly due to the manufacture of the AEM.

3. Results and Discussion

3.1. Life cycle assessment

Life cycle assessment comparison among the three representative BESs has been illustrated in Fig.3. As shown in Fig.3, the MEC has better environmental performance than the other two systems in all categories. The environmental benefits generated from the hydrogen peroxide production is far beyond other factors, such as external energy applied to the MEC. Except for ODP, the MEC has positive environmental impacts (negative absolute values). The MFC and MDC, however, have negative environmental impacts (positive absolute values) in all categories, which can be explained by the relatively low power densities generated at current stage. Only 10 W/m³_{anode} of power density is generated by oxidizing organic substrates in the anodic chamber in both the MFC and MDC. The change of environmental impacts with the increasing power density will be discussed in the sensitivity analysis.



| Fig.3. Normalized life cycle assessment comparisons among three different |
|--|
| BESs (AP×1E-1, GWP×10, ETP×1, EP×1E-4, HTP×1E-7, ODP×1E-8, |
| RI×1E-3, POCP×1E-2, ADP×1E-4). Acronyms: AP: acidification potential, |
| GWP: global warming potential, ETP: ecotoxicity potential, EP: |
| eutrophication potential, HTTP: human toxicity potential, ODP: ozone |
| depletion potential, RI: respiratory inorganics, POCP: photochemical ozone |
| creation potential, and ADP: abiotic depletion potential. |

Since the system manufacturing has relatively large environmental impacts in the life cycle, the environmental impacts of the system manufacturing in the three BESs are further analyzed here. In the MFC, the system manufacturing takes up a certain level of environmental impacts, ranging from 17.7% of EP to 99.7% of ODP. Although the system manufacturing only contributes to a very small percentage of environmental impacts in the MEC, the system manufacturing in the MEC contributes to the same absolute values of environmental impacts due to the same structure and composition with the MFC. In the MDC, the system manufacturing also contributes to a certain level of environmental impacts, ranging from 22.7% of GWP to 99.8% of ODP. The system manufacturing of the MDC has higher environmental impacts than that of the MFC and MEC. This can be explained by the large environmental impacts generated from producing the AEM, which is not included in the MFC and MEC.

The operation phase has large environmental impacts in almost all categories. In the MFC, the operation phase takes up more than 30% of environmental impacts in AP, GWP, EP, HTP, and RI, and this is mainly due to the energy consumption in the operation phase. The electricity usage takes up about 112.0% of embodied mass and 74.6% of embodied energy in the operation phase, and this can be explained by the pumping processes involved in the operation phase. In the MEC, the operation phase has dominant environmental impacts due to the large environmental credits obtained from hydrogen peroxide production. In the MDC, the operation phase occupies more than 30% of environmental impacts in AP, GWP, ET, and HTP, which can be also explained by the high energy consumption in the operation phase. The electricity usage occupies about 111.1% of embodied mass and 86.6% of embodied energy in the operation phase. The relatively high environmental impacts of the operation phase can be also explained by the relatively low power density generated at current stage of development.

Apart from the system manufacturing and operation phases, the pretreatment also contributes to relatively large environmental impacts. In the MFC, the pretreatment takes up 26.8% of AP, 35.6% of ETP, 22.8% of HTP, and 30.3% of RI. In the MDC, the pretreatment occupies 24.2% of AP, 31.7% of ETP, 22.8% of HTP, and 27.01% of RI. The relatively high environmental impacts in the pretreatment is mainly due to the electricity used to pump water.

3.2. Sensitivity analysis

Since the environmental impacts of the MFC and MDC are highly dependent on the power density, the sensitivity analysis has been conducted to investigate the change of environmental

impacts with the increase of power density. It is noticeable that in the MDC, the ion removed or transferred has a linear relationship with desalination capacity. Based on the charge balance, the power density is proportional to the quadratic of seawater flow rate. As a result, the increase of power density in the MDC also affects the desalination capacity, and this has been considered in the sensitivity analysis. According to Logan et al., the volumetric power density can vary from less than 0.1 W/m³_{reactor} to 1000 W/m³_{reactor}. As a result, a series of power densities have been selected to examine the environmental impact variations based on different power densities, and the selected power densities are 10 W/m³_{anode}, 50 W/m³_{anode}, 100 W/m_{anode}^3 , 200 W/m_{anode}^3 , 300 W/m_{anode}^3 , 400 W/m_{anode}^3 , and 500 W/m³_{anode}. Although the sensitivity analysis of the MDC and MFC with increasing power densities has been conducted in our previous research [16], the environmental variations have been recalculated based on the updated GaBi databases and ecoinvent database and a slight difference can be seen in the two papers. As shown in Fig.4, the environmental benefits generated from electricity generation begins to balance the environmental burdens at the power density of 300W/m³_{anode}. The overall environmental impacts of the MFC and MDC at the power density of 500 W/m³_{anode}, however, are still higher than the MEC at current power density. Detailed comparison among different BESs with different power densities can be found in section 4.

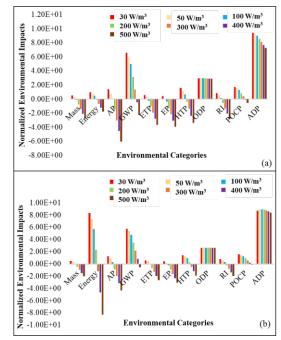


Fig.4. Sensitivity analysis: (a) Normalized environmental impacts for the comparison among different power densities in the MFC (Mass×10, Energy×1E-1, AP×1E-5, GWP×1E-3, ETP×1E-2, EP×1E-6, HTP×1E-10, ODP×1E-8, RI×1E-6, POCP×1E-5, ADP×1E-8).; (b) Normalized

environmental impacts for the comparison among different power densities in the MDC (Mass×10, Energy×1E-2, AP×1E-5, GWP×1E-3, ETP×1E-2, EP×1E-6, HTP×1E-10, ODP×1E-8, RI×1E-6, POCP×1E-5, ADP×1E-8).

4. Benchmarking with the conventional wastewater treatment

To evaluate the sustainability performances of different BESs, the greenhouse gas (GHG) emissions of the three representative BESs are benchmarked with the conventional wastewater treatment methods-AD treatment method. Two LCA studies have been selected because both of their functional units were the same as the functional unit selected for the BESs-the unit volume of incoming wastewater, and both of them assessed the environmental impacts from cradleto-grave [23, 24]. As shown in Table 1, the environmental performance of the MEC is much better than other options due to the large amount of hydrogen peroxide production. In addition, the environmental impacts of the MFC and MDC, at the current power density, are relatively high when compared with the AD treatment method. However, BESs have a large potential to become a complementary method for the traditional wastewater treatment methods with the increase of power density.

| Table 1.1 | Benc | hm | arking | of LC | A | results | s with | other BESs to treat 1 L of water. |
|-----------|------|----|--------|-------|---|---------|--------|-----------------------------------|
| | | | | | | | | |

| | Studied location | Description of the system | GHG emissions (kg/m ³ incoming wastewater) |
|----------|------------------|----------------------------------|---|
| Niero et | Denmark | Chemical | 0.195-0.213 |
| al. | | precipitation/Advanced | |
| (2014) | | biological treatment + activated | |
| | | sludge treatment + Agriculture | |
| | | sludge treatment | |
| Stokes & | California | Primary sedimentation + | 0.055 |
| Horwath | , US | Activated sludge + Secondary | |
| (2009) | | clarification + Agriculture | |
| | | sludge disposal | |
| This | US | Pretreatment + MFC + Post- | -2.28-6.58 |
| study | | treatment | |
| This | US | Pretreatment + MEC + post- | -8.58E4 |
| study | | treatment | |
| This | US | Pretreatment + MDC + post- | -0.53-5.72 |
| study | | treatment | |

5. Conclusion

To facilitate the sustainable development of BESs, the environmental impacts of three representative BESs have been compared with each other by using attributional LCA method. It has been found that the MEC has better environmental performance than the MFC and MDC, which is mainly due to the high production rate of hydrogen peroxide. In addition, the MDC has relatively high environmental impacts in the system manufacturing when compared with the other two BESs, which can be explained by the AEM used in the desalination chamber. Yet the environmental impact comparison results are based on the functional unit of 1 L of incoming water and the assumption that all BESs can achieve the same treated water quality. The results may still vary, especially considering the different water qualities that can be obtained by wastewater treatment and saline water desalination. As a result, further studies are needed for more accurate LCAs of BESs. Furthermore, the environmental impacts of the MFC and MDC can be dramatically decreased by the improvement of the power density. Their environmental impacts at a power density of 500

 W/m^3_{anode} , however, are still higher than those of the MEC at the current power density. When benchmarking the environmental impacts of BESs with the anaerobic digestion treatment method, the MEC has a better environmental performance, but the MFC and MDC have relatively large environmental impacts. In conclusion, the environmental impacts of the MFC and MDC have a large potential to be decreased with the increase of power density, and BESs can serve as a complementary method for wastewater treatment with the increase of power density.

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