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Treatment of Sewage (Domestic Wastewater or Municipal Wastewater) and Electricity Production by Integrating Constructed Wetland with Microbial Fuel Cell

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http://dx.doi.org/10.5772/intechopen.75658

Abstract

Proper treatment of wastewater is important to human health and societal development, and the commonly applied wastewater treatment technologies based on aerobic treatment have a significant demand for energy. Thus, new treatment technologies with low energy consumption and possible recovery of valuable resources (e.g., energy and water) from wastewater become of strong interest. Among the newly developed concepts, microbial fuel cells (MFCs) integrated with constructed wetland appear to be very attractive because of direct electricity generation from organic compounds and treatment of wastewater. Constructed wetland coupled with microbial fuel cell (CW-MFC) is an emerging technology in recent years and has attracted a lot of attention from researchers in the fields of wastewater treatment and bioenergy production. CW-MFC is a promising technology in the fields of wastewater treatment and bioenergy. However, at current power levels, the advantage of combining the two is mainly because of the enhancement of wastewater treatment in anaerobic zones within the wetland. New operational strategies need to be explored to increase and utilize electricity output.

Keywords: microbial fuel cell, constructed wetland, electricity production, wastewater treatment, bioenergy

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1. Introduction

1.1. Water

Water is derived from the Anglo-Saxon and Low German word, *wæter* which is an odorless, flavorless, and colorless substance that is necessary to all living beings that we all are aware of. It is the main constituent of earth's streams, lakes, oceans, and the fluids of most living organisms [1]. Its chemical formula is $H₂O$, which means that each of its molecules contains one atom oxygen and two atoms hydrogen that are connected by a bond known as "covalent bond". Water is found in three different forms on earth, that is, solid, liquid, and gas [2]. These forms of water depend on the temperature. Water on our planet is present as a **solid form** in the ice-caped areas at the North and South Poles, **liquid** in rivers, streams, and oceans and is **gas** or vapor form in the atmosphere [3].

But the present scenario says that all the water resources that are present in the globe are under a major stress. Today, however, expansion of industries, agriculture, damming, urbanization, population, and pollution threaten these unique resources in many parts of the earth [4].

But the main concern now is providing safe drinking water to the more than 1 billion people who currently lack it; this is one of the utmost public health challenges facing governments today. In many developing countries, safe water, free of pathogens and other contaminants, is unavailable to much of the population, and water contamination remains a concern even for developed countries too with good water supplies and advanced treatment systems [5].

1.2. Distribution of water

The first living organisms undoubtedly arose in an aqueous environment and during the course of evolution it has been shaped by the properties of the aqueous medium in which life began [6]. Therefore, life on the earth that is originated from sea and water plays a pioneer role for evolutionary development of species, life forms, and relatively complex molecules [7]. It is an abundant essential resource on earth and covers 71% of the earth's surface. This earth's water consists of 3% freshwater of total water supply and is found as either surface water or groundwater, however, 97% as saltwater [8]. Therefore [9] has concluded that the human interference, inadequate supply, and inappropriate management are the major causes [10] leading to scarcity of resources that impedes sustainable development [11].

1.3. Wastewater

Wastewater or sewage is the byproduct of water. There are the household uses such as bathing, dishwashing, laundry, and, of course, flushing the toilet. Additionally, industries use water for many purposes including processing and cleaning or rinsing of parts. With rapid growth of cities, urbanization, and industrialization the quantity of gray/wastewater is increasing in the same proportion. As per Central Public Health and Environmental Engineering Organization (CPHEEO) estimates about 70–80% of total water supplied for domestic use gets generated as wastewater. Typically, 200–500 L of wastewater are generated for every person. In India, there are 234 Sewage Water Treatment plants (STPs). Most of these were developed under various river action plans (from 1978 to 1979 onward) and are located in (just 5% of) cities/towns along the banks of major rivers [12].

1.4. Energy cost of wastewater treatment

Energy use can account for as much as 10% of a local government's annual operating budget [13]. A considerable amount of this municipal energy use occurs at water and wastewater treatment services. With pumps, motors, and other equipment operating 24 h a day, 7 days a week, water and wastewater services can be among the largest consumers of energy in a community and thus among the largest contributors to the community's total GHG emissions. Nationally, the energy used by water and wastewater utilities accounts for 35% of typical U.S. municipal energy budgets [14]. Electricity use accounts for 25–40% of the operating budgets for wastewater utilities and approximately 80% of drinking water processing and distribution costs [14]. Drinking water and wastewater systems account for approximately 3–4% of energy use in the United States, resulting in the emissions of more than 45 million tons of GHGs annually [15].

2. Introduction of bioelectrochemical systems

2.1. Biochemical system

There is a growing demand for new energy sources due to the limited accessibility and pollution caused by the use of fossil fuels. At present, the annual energy demand is approximately 13 terawatts (TW) worldwide and it is estimated to reach around 23 TW by the year 2050 [16]. Bioelectrochemical systems (BESs) have considerably boomed over the past decade for their contribution as an emerging sustainable technology for concurrent electricity production and wastewater treatment [17]. Microbial fuel cells (MFCs) and microbial electrolysis cells (MECs) are two examples of a speedily developing biotechnology, generally known as bioelectrochemical systems (BES), that combine biological and electrochemical processes to generate electricity, hydrogen, or other useful chemicals. Moreover, BESs are also identified as efficient bioreactors for the treatment of recalcitrant pollutants and toxic wastewaters; the process is termed as bioelectrochemical treatment (BET) or microbial electroremediation [18].

2.2. Microbial fuel cell

Microbial fuel cell (MFC) technology is the symbol of the newest approach for generating electricity from biomass using microorganisms. While the first observation of electrical current generated by bacteria is generally credited to Potter in 1911 [19], very few practical advances were achieved in this field even 55 years later [20]. In the early 1990s, fuel cells became of more interest and work on MFCs began to increase [21]. A microbial fuel cell is a tool that converts chemical energy to electrical energy with the help of the catalytic reaction of bacteria [21–26].

A microbial fuel cell consists of anode and cathode sections, which are separated by a specific membrane. Microbes are present in the anode section and they oxidize fuel (electron donor) which finally generates electrons and protons. Electrons are transferred to the cathode area through the circuit and the protons through the specific membrane. Electrons and protons are consumed in the cathode compartment reducing oxygen to water.

2.3. Design of MFCs

A suitable design is foremost an important characteristic feature in MFCs and researchers have come up with several designs of MFCs over the past few years with better performance [27].

2.3.1. Single chamber microbial fuel cells

Single compartment MFC offers simpler design and cost savings. It typically consists of an anode chamber with a microfiltration membrane air-cathode. The cathode was exposed to air on one side and water on the other side (inside). There is no proton exchange membrane. The microfiltration membrane is applied directly onto the water-facing side of the cathode.

2.3.2. Dual chamber microbial fuel cells

Dual chamber MFC consists of an anaerobic anode chamber and an aerobic cathode chamber which are usually separated by a proton exchange membrane (PEM). Substrate is oxidized by bacteria generating electrons and protons at the anode chamber. The protons traveling through the PEM and the electrons traveling through the external circuit are combined with electron acceptors at the cathode chamber. The anode is inoculated with a mixed solution of anaerobic sludge and substrate like glucose. On the other hand, cathode is inoculated with aerobic sludge.

2.3.3. Air-chamber microbial fuel cells

Dual-chamber MFCs are mainly used in laboratory range and cannot be adapted for continuous treatment of organic matter due to the demand of oxygenated water. In a substitute design without aqueous cathode, cathodic electrode is bonded straight to proton exchange membrane so that air can be openly reduced [28–32]. The earliest air-cathode MFC architecture was designed and reported that an oxygen gas diffusion electrode could be used as a cathode in bioelectrochemical fuel cell [33]. But this air-cathode design has not drawn much attention in MFC research until Liu reported the air-cathode MFC could produce much greater power than typical aqueous-cathode ones.

The architecture of air-cathode MFCs is aimed to optimize some characteristics of two-chamber MFCs, such as low relative power output, high cost of cathode catalysts and membranes, and energy requirement for intensive air/oxygen sparging. Another advantage of the air-cathode over the two-chamber is the reduction of the high internal resistance of MFCs, which is a key factor to enhance electricity production.

2.4. Substrates used in microbial fuel cells

In generating electricity in MFCs, substrate is regarded as one of the most important biological factors [34]. A huge variety of substrates can be used in MFCs for power generation. The substrates not only influence the MFC performance including the power density (PD) and Coulombic efficiency (CE) but also the integral composition of the microorganism's community in the anode [35]. During the development of this technology, low molecular weight substrates were employed, that is, carbohydrates such as glucose, fructose, xylose, sucrose, maltose, and trehalose [36–38], organic acids such as acetate, propionate, butyrate, lactate, succinate, and malate [39–42], alcohols such as ethanol and methanol [43], and inorganic compounds such as sulfate [44].

3. Constructed wetland

Constructed wetlands (CW) systems are entirely man-made wetlands for wastewater treatment, which relate various technological designs, using natural wetland processes, associated with wetland hydrology, soils, microorganisms, and plants. Thus, CWs are engineered systems that have been designed and constructed to utilize the natural processes involving wetland vegetation, soils, and their associated microbial assemblages to assist in treating wastewater. Synonymous terms to "constructed" include "man-made," "engineered," or "artificial" [45].

Constructed wetlands (CWs) have been used to treat wastewater ranging from domestic to industrial and from urban to agricultural along with treating stormwater runoff, leachates and mine drainage, and for sludge dewatering through a combination of physical, chemical, and biological processes. Constructed wetlands (CW) are a feasible alternative option for removing nutrients and other contaminants from wastewater and have been used to treat many different types of wastewater for decades. CW mimics the properties of a natural wetland, and filtration occurs as a result of physical, chemical, and biological processes that are similar to those that take place in a natural wetland. There are numerous types of CW, and they can be differentiated based on dominant vegetation type, hydrology (surface vs. subsurface water flow), and direction of flow.

3.1. Surface flow (free water surface)

A surface flow CW is comprised of a sealed packed basin or series of basins filled with 20–30 cm of gravel and with a water depth of 20–40 cm. Planted macrophytes emerge over the surface of the water but roots are in the soil. Effluent water is treated as it flows over the soil/ substrate. These systems effectively remove organic material through microbial degradation and settling and inorganic materials through settling alone [46]. They are efficient at removing nitrogen (N) through denitrification and ammonia volatilization but are unable to effectively remove phosphorus (P) as water does not tend to come in contact with soil particles which adsorb or precipitate phosphorus (P) [47] (**Figure 1**).

Figure 1. Diagram of a surface flow constructed wetland. Image by Dr. Sarah white [48].

3.2. Subsurface flow

Subsurface flow wetlands are made up of an impervious basin filled with a layer of gravel of size 10–20 mm [46]. Wetland plants are grown in the gravel layer and water flows through the gravel layer, around the plant roots. Subsurface flow systems are differentiated based on whether the main direction of flow is horizontal or vertical [48]. In a horizontal flow (HF) system, water enters through an inlet, flows slowly through the substrate, and exits through an outlet on the other side of the system. HF systems effectively remove organic material and suspended solids through anaerobic microbial and sedimentation, respectively. Nitrogen is removed mainly via denitrification, as ammonia volatilization may not take place due to lack of oxygen. Generally, because of the lack of ammonia volatilization occurring, total N removal by these systems is low [45] (**Figure 2**).

3.3. Hybrid constructed wetlands

Various types of CWs can be combined to achieve higher removal efficiency. The design consists of two stages, several parallel vertical flow (VF) beds followed by several horizontal flow (HF) beds in series (VSSF-HSSF system). The VSSF wetland is intended to remove organics and suspended solids and to promote nitrification, while in HSSF wetland denitrification and further removal of organics and suspended solids occur. Another configuration is a HSSF-VSSF system. A large HSSF bed is placed first to remove organics and suspended solids and to promote denitrification. An intermittently loaded small VF bed is used for additional removal of organics and suspended solids and for nitrification of ammonia into nitrate. To maximize removal of total N, however, the nitrified effluent from the VF bed must be recycled to a sedimentation tank [49] (**Figure 3**).

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Figure 2. Diagram of a subsurface flow constructed wetland. Image by Dr. Sarah white [48].

Figure 3. Diagram of a hybrid constructed wetland [50].

4. Microbial fuel cell implemented in constructed wetland: Recently emerged technology

A microbial fuel cell coupled constructed wetland (CW-MFC) system is a latest mechanism that embeds the MFC into the constructed wetland (CW) to treat the wastewater and produce electricity. Research suggested that wetland plants promote the cathode performance of MFCs [51]. Technology combining CW systems with microbial fuel cells (CW-MFC) has

promise for both wastewater treatment and bio-electric production [52–54]. In this system approach, electricity is produced with biodegradable substances as bacteria oxidize organic or inorganic matter in wetland soils [54].

4.1. Architecture and operation of constructed wetland coupled with microbial fuel cells

In order to maximize the redox gradient (as it is an essential factor in producing an electrical current in MFCs), most CW-MFCs have been operated under upflow conditions with a buried anode and a cathode at the surface and/or in the plant rhizosphere. This arrangement minimizes the dissolved oxygen (DO) at the anode while ensuring maximum availability in the cathode region. Initially a glass wool separator was used by [55, 56] to provide a 'sharp' redox profile. However, the use of a separator where an upflow regime is being used with a buried anode and a cathode at the surface may not be necessary as this arrangement provides a sufficient redox profile for MFC integration [57–59]. Unfortunately, utilizing the natural redox gradient afforded by an upflow regime results in large electrode separation and contributes greatly to the ohmic resistance of the system [60–64]. Carbon and graphite are commonly used as electrode materials in MFC studies since they offer long-term sustainability owing to their high electrical conductivity, non-oxidative nature, and the fact that they offer a good medium for the attachment and growth of microbial communities.

4.2. Performance of constructed wetland coupled with microbial fuel cells

4.2.1. Functioning as constructed wetland regarding wastewater treatment

MFC integrated into CW is a possible and economical way to achieve the objectives of both wastewater treatment and electricity generation. The ability of CWs to treat wastewater is well established [65, 66] and, as such, integrating MFCs into CWs should not come at the price of reducing their effectiveness at removing contaminants from wastewater. Preliminary investigations indicated that CW-MFCs perform similarly to previous CW studies by removing 75% [55] and 76.5% [56] of COD. The inclusion of the MFC component has shown the ability to improve the COD removal efficiency in CWs. The inclusion of plant roots at the cathode of CW-MFCs slightly improves COD removal efficiencies in the wetland compared with non-planted and rhizosphere-anode CW-MFCs [58, 57]. The presence of the anode improved COD removal efficiencies by 12.65%. Thirty-three percent of COD was removed at the anode occupying 13.6% of the liquid volume in a CW-MFC operated by [53].

4.2.2. Functioning as MFC regarding electricity production

MFC converts the biodegradable compounds to generate electricity by utilizing bacteria. COD loading greatly affects the performance of CW-MFCs. A balance is necessary between providing sufficient organics for oxidation at the anode and limiting the amount of COD arriving at the cathode. In a vertical upflow CW-MFC designed by [67], an increasing trend in power densities was observed as influent COD was increased from 50 to 250 mg/L. However, further increases in concentration to 500 and 1000 mg/L resulted in average power densities of 33.7

and 21.33 mW/m², respectively, compared with 44.63 mW/m² for influent COD concentrations of 250 mg/L. Current densities were increased when the SSM was embedded in carbon cloth $(49.68 \pm 2.83 \text{ mA/m}^2)$ and granular activated charcoal (GAC) $(63.69 \pm 1.78 \text{ mA/m}^2)$. Both the carbon cloth and GAC increased the surface area of the electrode thereby facilitating bacterial growth and providing more reaction sites for the reduction of $O₂$. Other compounds in the wastewater will affect the ability of electrogenic bacteria to produce power. **Table 1** has shown the performance of CW-MFC by using different electrode materials and also of organic loads. Operating under batch mode, [55] noted that as dye concentration increased from 1000 to 1500 mg/L the average power density more than halved due to the toxic effect of the dye.

Table 1. Reported performance of CW-MFCs.

Similarly, [68] reported that as the proportion of ABRX3 dye (measured as COD) increased incrementally from 10 to 90% the maximum power density, obtained from the power density curves, fell from 0.455 to 0.138 W/m³. The reduction in electrical performance was primarily attributed to anodic polarization.

The CW-MFCs tested in this study were suitable for long-term stable operation and showed strong adaptability to different water qualities. HRT significantly influenced the decolorization process in the anode layer. The power density, the Coulombic efficiency, the open circuit voltage, the decolorization rate, and the COD removal rate increased initially and then decreased with the elongation of the HRT.

5. Comparison of CW-MFC with traditional biological treatment processes

Biological treatment is an important and integral part of any wastewater treatment plant that treats wastewater from either municipality or industry having soluble organic impurities or a mix of the two types of wastewater sources. The obvious economic advantage, both in terms of capital investment and operating costs, of biological treatment over other treatment processes like chemical oxidation, thermal oxidation, and so on, has cemented its place in any integrated wastewater treatment plant. Conventional activated sludge process (ASP) system is the most common and oldest bio-treatment process used to treat municipal and industrial wastewater, but the main problem with this is that it requires aeration, which uses a large amount of electrical energy. But on the other hand, MFC-centered hybrid technologies have attracted attention during the last few years due to their compatibility and dual advantages of energy recovery and wastewater treatment. In this system, oxygen is needed for the aerobic chamber but oxygen enters into that chamber through rhizosphere zone and no energy is required for this purpose but in fact electricity is harnessed during the system operation. **Table 2** lists the comparison of CW-MFC and conventional activated sludge process and highlights the benefits of CW-MFC which not only treating the wastewater but also we are harnessing electricity out of it.

Biological treatment system uses two processes, that is, aerobic and anaerobic. Aerobic, as the name suggests, means that the reactions takes place in the presence of air (oxygen) while anaerobic means in the absence of air (oxygen). These two terms are directly related to the type of bacteria or microorganisms that are involved in the degradation of impurities in a given wastewater and the operating conditions of the bioreactor. Therefore, aerobic treatment utilizes those microorganisms that use molecular/free oxygen to assimilate organic impurities, that is, convert them into carbon dioxide, water, and biomass. The anaerobic treatment processes, on the other hand, take place in the absence of air (and thus molecular/free oxygen) by those microorganisms (also called anaerobes) which do not require air (molecular/ free oxygen) to assimilate organic impurities. As this process takes place in the absence of air, it is relatively slow compared to aerobic, because to create an anaerobic environment is a very difficult task.

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Table 2. Differences between CW-MFC and biological treatment system.

If we talk about land requirement for the biological treatment system, lots of land are needed for different purposes like *Trickling filter, Rotating biological filter*, *Facultative (waste stabilization) ponds, Aerated lagoons, Activated sludge process, Anaerobic ponds, Septic tanks, Imhoff tanks,* and so on.

6. Conclusions and future perspectives

Wastewater treatment using conventional system like activated sludge technology is still energy and cost consuming, and chemicals have to be used in the treatment process. Constructed wetlands in combination with bioelectrochemical technology can provide an alternative. Wastewater treatment through CW-MFC is possible using a small-scale, constructed wetland-microbial fuel cell system. Good results were obtained with regard to organics removal, filtration of suspended solids, nutrient removal, and passive disinfection.

The CW-MFC is a promising technology in the fields of wastewater treatment and bioenergy. However, at current power levels, the biggest advantage of combining the two may come from the enhancement of wastewater treatment in anaerobic zones within the wetland. For the electrical output to be increased new operational strategies need to be explored to reduce the electrode spacing while maintaining the required redox conditions in the system. If the existing limitations of the combined system can be addressed, this prototype CW-MFC system can provide a real alternative for wastewater treatment, when built on a larger scale. Since the operational costs of a constructed wetland are very low, and given the MFC can produce electricity at a relatively low cost, this system could be competitive to existing water treatment plants.

MFCs implemented in CWs may increase not only CW treatment capacity but also would be of use as a biosensor tool to monitor treatment performance and operational conditions (such as influent organic matter concentration). Organic matter concentration is currently determined by means of analysis of the biochemical oxygen demand (BOD) after 5 days (BOD₅) or the chemical oxygen demand (COD). Despite the fact that these methods are universally used, BOD_5 has a limitation in terms of being time consuming, and is not suitable for online process monitoring.

COD is a faster procedure for assessing organic matter concentration in wastewater, yet it is quite costly and produces toxic reagents that might pose a threat to the environment. Overall, the synergy between CWs and MFCs has been so far mostly based on optimization for energy production. Besides the interest that an energy surplus can have in the context of CW technology, further research shall be focused on the optimization of both technologies to fully address other benefits of MFC implementation in CWs such as treatment efficiency improvement, process monitoring, and the reduction of clogging, or methane emissions.

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