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Research progress of microbial fuel cell and constructed wetland coupling system

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Abstract: The coupling system of microbial fuel cell and constructed wetland has become a new production capacity and wastewater purification process in recent years. In this paper, the current research situation and system characteristics of MFC-CW coupling system are reviewed. On the basis of this, the composition factors (plant, microorganism and electrode materials) and the operating conditions (substrate concentration, dissolved oxygen, redox potential and HRT) of the current research are further analyzed and discussed, and finally MFC-CW coupling system is summarized and the key problems that have not been solved at present are put forward.

1. Introduction

Microbial fuel cell (MFC) is a new bioelectrochemical system which uses microorganism to degrade organic matter and convert the chemical energy of organic matter into electric energy[1]. The principle is similar to the fuel cell (FC), but can use more complex fuel than methanol or hydrogen. The traditional microbial fuel cell consists of four basic components: anode chamber, cathode chamber, proton exchange membrane and electrolyte. The basic principle of microbial fuel cell is to break the transfer direction of conventional electron transfer chain and draw the generated electrons to the outside world to obtain energy. On the other hand, the original redox reaction is extended to the outside environment outside the cell and the whole cell structure system. MFC generally consists of an anode chamber, a cathode chamber, separating layer and a circuit. The structure is showed in Figure 1.

![Microbial fuel cell diagram](image)

Figure 1. Microbial fuel cell

Microbial fuel cells have broad application prospects, but the low power of fuel cells has
constrained the further development of microbial fuel cells. In recent years, many studies on the coupling of MFC with other biological treatment technologies have emerged. For example, the combination of MFC with membrane biological treatment technology and activated sludge technology can improve sewage treatment capacity\cite{2,3}.

Constructed wetlands (CW) are a comprehensive ecosystem. It is a ground analogous to the marshland constructed and controlled by manpower. The sewage and sludge are controlled and allocated to the artificially constructed wetland. Sewage and sludge flow mainly in the process of using the soil. A combination of physical, chemical, and biological interactions of artificial media, plants, and microorganisms to treat sewage and sludge. At the same time, pollutants in sewage can promote the growth of plants and realize the resource and harmlessness of wastewater\cite{4,5}. Different types of CWs can be combined with other types to achieve the best treatment efficiency\cite{6,7}(Fig. 2). Its mechanism of action includes adsorption, retention, filtration, redox, precipitation, microbial decomposition, transformation, plant cover, residue accumulation, transpiration and nutrient absorption, and the role of various animals. In recent years, it has been widely used due to the low cost of installation, operation and maintenance of CW\cite{9}.
Both MFC and CW are biological systems for the treatment of organic compounds. Coupling the two can improve the efficiency of electricity production while achieving efficient pollutant treatment. In recent years, the coupling of microbial fuel cells with constructed wetlands has received increasing attention. By filling a carbonaceous filler with conductive properties, the constructed wetland can be evolved into a short-circuited microbial fuel cell [10-12]. At this time, the bottom of the artificial wetland bed (lower ORP) is used as the anode of the microbial fuel cell, where the organic matter in the water is degraded, and the electrons released by the degradation process are transported upward along the conductive filler to the biocathode of the surface of the bed (higher ORP). Although the research on constructed wetlands and microbial fuel cells is relatively deep, there is not much research on the coupled systems. Microbial treatment technology can be divided into three types of aerobic, anaerobic and facultative metabolism. CW-MFC technology has obvious advantages in traditional sewage biological treatment technology. (Table 1)

| Technology          | Microbe               | Condition | Capacity mode | Energy Utilization pattern | Sludge production | Scope of application          |
|---------------------|-----------------------|-----------|---------------|---------------------------|-------------------|-------------------------------|
| Aerobic treatment   | Aerobic microorganism | Medium    | Energy consumption | Complex                  | Many              | Medium and low concentration  |
| Anaerobic treatment | Methanogen            | Higher    | Biogas        | Complex                  | Less              | Medium and high concentration |
| CW—MFC              | Electrogenic bacteria | Temperature and pressure | Electric energy | Simple                  | Less              | Unlimited                     |

2. MFC-CW research status

India's Yadav et al. [13] established the first MFC-CW system, using graphite as the anode and cathode electrode MFC embedded in the vertical flow CW of synthetic dye wastewater to study its decontamination and electricity production capacity. Then, Spain's Villasenor et al. [14] in the greenhouse use graphite as the cathode, anode electrode, and use gravel as the substrate, the electrode is placed below the surface of the wetland, the distance between the electrodes is 25cm, the effects and
electrical properties of wastewater treatment at different temperatures, dissolved oxygen (DO), redox potential (ORP) and so on were studied. Li Xue[15] et al. constructed a microbial fuel cell-synthetic wetland coupling system with reeds as a wetland plant, and produced electricity while purifying the sewage. The system can operate stably for a long time. With the influent COD and hydraulic retention time, the system The output voltage first increases and then decreases. In recent years, many countries have used MFC-CW system to treat a variety of wastewater to achieve better treatment results, achieved better power production efficiency, and have a good application prospect. In the current study, in order to increase the redox gradient, most MFC-CW systems use an upflow, deep buried anode and a cathode on the surface of the system or at the root of the plant. This arrangement and operation mode minimizes the dissolved oxygen concentration in the anode region and maximizes the dissolved oxygen concentration in the cathode region which conducive to the system's sewage purification effect and power generation performance [16].

3. MFC-CW operating performance and influencing factors
MFC-CW electricity production and sewage purification performance are closely related to organic load and sewage composition, hydraulic retention time, cathode dissolved oxygen concentration, electrode materials and wetland plants [17-21].

3.1 MFC-CW basic composition
3.1.1 Wetland plants
The cathode potentials are 299mv and 202mv, respectively, and anode potentials are -341mv and -288mv, [23] respectively. The presence of plants can increase the system's electricity production by 142% [17]. Under natural reoxygenation conditions, gas cathodes and beds at the gas-water interface The distance between the anodes of the bottom layer is large, and the internal resistance is increased. Under the action of plants, the oxygen secretion of the rhizosphere of the plants is shortened. The wetland plants are an important part of the constructed wetlands. In the MFC-CW system, plants can directly absorb pollutants such as nitrogen and phosphorus, and enrich toxic substances such as heavy metals. They can also transport oxygen to the root zone to provide the required oxygen for microbial growth and reproduction in the root zone, and enhance the metabolic rate of microorganisms. In addition, plants can maintain and enhance the hydraulic transport of the MFC-CW system [22]. In addition to the above effects, the influence of plants on the electrochemical characteristics of the system has also received extensive attention. Studies have shown that the presence or absence of plant MFC-CW pole spacing reduces internal resistance. The internal resistance of the presence and absence of plant systems is 217.7 Ω and 272.9 Ω, respectively [24]. Wetland plants are beneficial to the formation of bipolar functional microorganisms, and the microbial density around the electrodes is significantly increased, increasing the system's redox potential difference [25]. Under the action of plants, the electrode cell density increased by 58%, and the number of microorganisms increased from 5.13±0.86×107 cells/g to 8.66±1.01×107 cells/g [23]. When the plant is in the anode region, root exudates can be used as a source of organic matter [26]. Plant roots can secrete various organic matter (carbon source) for microbial growth, create conditions for the survival of microorganisms, improve the processing capacity of the system [27, 28], and improve the electrochemical conditions of the system.

3.1.2 Electrode materials and fillers
Another factor influencing MFC-CW is the electrode material. The choice of electrode materials plays a crucial role in the development of MFC-CW. The ideal electrode material needs to have excellent electrical conductivity, high hardness and strength, electrochemical stability and biocompatibility, in addition to providing a strong attachment point for microorganisms. Carbon and graphite are the most commonly used electrode materials, both of which have high electrical conductivity, are not naturally oxidized, provide adsorption points for microorganisms, and promote microbial growth. Their overall performance is superior to that of nickel, stainless steel, brass and aluminum. Materials are therefore
the most commonly used electrode materials[29]. The main purpose of anode material research is to increase the specific surface area, catalytic activity and electron conversion efficiency of the electrode. The anode material of the microbial fuel cell is generally graphite, carbon paper, carbon cloth, platinum, platinum black, and reticulated platinum carbon. In addition to materials, the anode also needs to focus on the microorganisms attached to the anode. Currently known electrogenic microorganisms mainly include Shewanella, Geobacter, Pseudomonas, mud bacteria and the like. All materials used as anodes can be used as cathodes. In order to improve the catalytic efficiency, the cathode material often needs to support a catalyst such as platinum. Recently, the use of bacterial catalytic cathode reduction has become a new research direction for microbial fuel cells. Moon et al.[30] used graphite felt and platinum-coated graphite felt as cathode materials respectively, and the power density obtained by MFC coated with platinum graphite felt as cathode material was three times that of MFC with graphite felt as cathode material. The matrix material of MFC-CW can be selected from sand, ash, clay, quartz sand, ceramsite, granular activated carbon and granular graphite. These materials can increase the removal efficiency of organic compounds through the adsorption process. Pratiksha Srivastava et al.[20] used granular activated carbon and granular graphite as matrix materials respectively. The experimental results show that the MFC-CW system with granular activated carbon as the matrix material obtains higher power density.

3.1.3 Microorganisms
There are a large number of microorganisms in the MFC-CW system, including aerobic, anaerobic and facultative bacteria, which significantly affect the purification and power generation capacity of the system.

Electrogenic microorganisms are widely found in natural and polluted environments, and are often dominated by metal-derived reducing bacteria such as Geobacter, Shewanella, and Pseudomonas. The formation of the electronic circuit is conducive to the growth of electrogenic microorganisms, the number of which can reach 70% or even more than 90%. In the lower environment of ORP, anaerobic microorganisms metabolize organic matter to release electrons; in the higher environment of ORP, microbial consumption of organic matter consumes oxygen and is beneficial to the reduction and sedimentation of Cr(VI); secretions produced by microorganisms during metabolism For example, polysaccharides, glycoproteins, lipopolysaccharides and other reactive groups such as phenolic groups, phenolic hydroxyl groups and carbonyl groups have strong complexation and chelation of heavy metals [31]; some products produced by microorganisms during growth and metabolism have Conducive to the precipitation of heavy metals. Li Fengxiang et al. Microbial fuel cell electrode liquid contains electrogenic bacteria, electron donor and receptor, which are the material basis for microbial fuel cell to degrade organic pollutants and produce electricity. The analysis of electrode solution is of great significance for the improvement of MFC power generation performance and the removal rate of organic pollutants.

3.2 MFC-CW system features
MFC converts the energy available in biodegradable materials directly into electrical energy. To achieve this, it is only necessary to convert bacteria from natural electron-transporting receptors that utilize it, such as oxygen or nitrogen, to the use of insoluble receptors, such as the anode of MFC. This conversion can be achieved by using a membrane-linked component or a soluble electron shuttle. The electrons then flow through a resistor to the cathode where the electron acceptor is reduced. Compared with anaerobic digestion, MFC can generate electricity and generate carbon dioxide-based exhaust gas.

MFC has operational and functional advantages over other existing technologies that utilize organic production capacity. First, it converts the substrate directly into electrical energy, ensuring high energy conversion efficiency. Secondly, unlike all existing bioenergy treatments, MFC can operate effectively under normal temperature and even low temperature environment conditions. Third, MFC does not require exhaust gas treatment because the main component of the exhaust gas it produces is carbon
dioxide, which does not have reusable energy under normal conditions. Fourth, the MFC does not require energy input because the cathode gas can be passively supplemented only by ventilation. Fifth, in areas where there is a lack of power infrastructure, MFC has the potential to be widely used, while also expanding the diversity of fuels used to meet our energy needs.

3.3 MFC-CW operating conditions

3.3.1 Different substrate concentrations
When the substrate concentration is different, the MFC-CW system's power generation performance will have a certain impact. A typical microbial fuel cell consists of an anode and a cathode, and a piece of a proton exchange membrane. The microorganism decomposes the oxidized fuel at the anode and simultaneously generates electrons and protons. The electrons can reach the cathode via external electricity, while the protons pass through the proton exchange membrane to the cathode. At the cathode, electrons and protons are combined with oxygen to produce water. Li Xue[15] and others have shown that under the condition that HRT is 2d and the cathode is not aerated, the output voltage of MFC-CW system increases first and then decreases with the increase of influent COD. When the influent COD is The maximum voltage is 294mV at 200mg/L; the minimum voltage is 166mV when COD is 500mg/L. Therefore, MFC-CW has a higher power generation performance when the influent COD is lower.

3.3.2 Hydraulic retention time
Hydraulic retention time (HRT) is the most important design and influencing factor of the MFC-CW system. In the MFC-CW system, when the hydraulic retention time is extended, the removal rate of the pollutants and the output power of the system can be improved, that is, the power generation performance of the system is enhanced. Yang Guangwei et al[32] studied the HTR performance of the system at 6, 12, 18, 24, 48 h, respectively. It is concluded that with the extension of HRT, the time for the MFC-CW system to reach a stable output voltage gradually increases, and the internal resistance gradually increases. When it becomes larger, the power density gradually decreases, and the Coulomb efficiency gradually increases. The reason is analyzed. When the HRT is short, the shearing effect of water on the biofilm reduces the adhesion of non-conductive substances on the biofilm, increases the electron transfer rate, and makes the internal resistance smaller and the power density larger. With the extension of HRT, the matrix intercepts the organic matter in the sewage, the direct utilization of the dissolved organic matter in the sewage by the electrogenic bacteria, and the biodegradation of the organic matter trapped on the substrate by the microorganism on the substrate, so that the coulombic efficiency increases. At present, the HRT of the MFC-CW system is generally 2 to 3 days. Optimizing HRT to achieve optimal performance of the system's power generation performance is one of the directions that need to be studied in the future.

3.3.3 Cathode dissolved oxygen concentration
The concentration of dissolved oxygen in the cathode region is also one of the factors affecting the electrical performance of the MFC-CW system. The bubbles generated by the cathode aeration promote the movement of the packing and improve the oxygen transfer efficiency; the increase of the dissolved oxygen concentration of the cathode promotes the growth and reproduction of the aerobic bacteria degrading the organic matter, which is beneficial to the improvement of the sewage purification performance of the system. OonYoong-Ling et al[19] added an aeration device in the cathode region. The experimental results show that the power density of the MFC-CW system increases significantly during aeration, which indicates that the increase of dissolved oxygen concentration in the cathode region is beneficial to improve the power generation performance of the system.

3.3.4 Oxidation reduction potential
The oxidation reduction potential (ORP) gradient in the wetland system is an important theoretical basis for its coupling with MFC. Optimizing the oxidation-reduction potential gradient between the anode and the cathode is essential for improving the electricity production of MFC-CW. In theory, the greater the difference between the potential of the anode and the cathode, the greater the output voltage. Therefore, the electrode should be placed as far as possible in the position where the oxidation-reduction potential gradient is the largest \[33\]. Corbella et al.\[34\] explored the optimal operation and design conditions for the coupling of subsurface flow constructed wetlands with MFC, and found that the maximum ORP gradient is on the surface and bottom of the continuous operation CW system, followed by the surface and the middle of the continuous operation CW system. Part. The ORP in the middle part of the CW system with intermittent water inflows fluctuates greatly, and the ORP at the bottom and middle is higher than that of the CW system with continuous water inflow. The presence or absence of plants has a greater impact on the ORP in the middle of the system, and other parts of the ORP are largely unaffected. However, the ORP of each CW system is not the same thing. Therefore, before constructing the MFC-CW coupling system, there should be some knowledge of the ORP distribution inside the system in order to optimize the placement of the electrodes.

4. Summary

MFC-CW coupling system has received more and more attention as a new green environmental protection system. This system not only enhances the ability to remove pollutants, but also converts the chemical energy generated in the process of microbial metabolism into electrical energy, which improves the efficiency of electricity production, and has great room for development. However, this system has low productivity and high cost, and is practically applied. There is still a need for deeper exploration. However, there are still several important issues that have not yet been resolved: (1) The contribution of plant roots to electricity production is still unclear. Whether plant roots are used as electricity-generating anodes or cathodes is for further study; (2) looking for cheaper electrodes and matrix materials is to make MFC-CW system required for wide application.

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