Research progress of magnetic materials in microbial fuel cell applications

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Abstract. As a new type of environmental protection technology that can treat sewage and generate electricity, microbial fuel cells (MFC) have broad research prospects. In recent years, MFC has made great breakthroughs, but its high internal resistance and low power generation efficiency have prevented its development. Researchers began to study the influence of magnetic fields on the MFC power generation efficiency to improve the power generation efficiency, and the results are gratifying. This shows that magnetic materials may be the solution to push MFC to practical applications. This article briefly introduces the research progress of magnetic materials as the additional structure of MFC and MFCs electrode materials, and discusses the role and influence of magnetotactic bacteria in MFC.

1. Introduction
Microbial fuel cell (MFC) is a technology in which microorganisms degrade organic matter in wastewater through redox reactions and generate electricity [1]. The research of MFCs has developed rapidly in recent years. Researchers have done a lot of research on reactor structure [2, 3], electrode materials [4, 5], and screening of high-efficiency electricity-producing bacteria [6], but they rarely study the effect of magnetic field on the electricity-generating efficiency of MFC. Few articles have studied the role and influence of magnetotactic bacteria (MTB) in MFCs. In this paper, by reviewing the application research of predecessors of magnetic materials in MFC, it puts forward some own views and opinions, hoping to put forward new ideas for the development and research of MFCs.

2. Magnetic material

2.1 Overview of magnetic materials
People's early understanding of magnetic materials began with natural magnets (Fe3O4) about 5000 years ago. About 2,300 years ago, the appearance of "Sinan" indicated that mankind began to use magnetic materials. This is the earliest record of the application of magnetic materials in human history. Magnetic material refers to any substance that can show magnetism or can be induced by a magnetic field or can change the magnetization. According to the strength of magnetism, substances can be divided into diamagnetism, paramagnetism, ferromagnetism, antiferromagnetism, and ferrimagnetism. Ferromagnetic and ferrimagnetic materials are strong magnetic materials, and the rest are weak magnetic materials. The development of modern industry is inseparable from magnetic
materials, and magnetic materials are also widely used in various fields such as electronics, communications, machinery, optics, chemistry and biology.

2.2 Preparation method of magnetic material
As a novel functional material, magnetic material has a high adsorption effect on heavy metal ions and toxic organic matter\(^7\). People have begun to develop magnetic nanoparticles, a new type of magnetic functionalized material, since the emergence of nanomaterials in the 1980s. At present, the more common methods for preparing magnetic adsorption materials include precipitation method, high temperature hydrothermal method, solvothermal method, thermal decomposition method, microemulsion method and sol-gel method, etc\(^7\)\(^-\)\(^10\). Magnetic Fe\(_3\)O\(_4\) nanoparticles are often selected as one of the most commonly used materials\(^7\),\(^9\)-\(^11\), because they have the characteristics of high specific surface area, special magnetic structure, and good adsorption performance.

2.3 Application of magnetic materials in water treatment
There are many research scholars at home and abroad who have devoted themselves to the study of magnetic materials in wastewater treatment in recent years. Due to its unique physical structure and chemical properties, magnetic materials can be used as a new type of adsorption material, which has a good adsorption effect on heavy metal ions and organic pollutants\(^12\),\(^13\). Du Wenqi et al.\(^13\) combined the modified rice husk with magnetic materials to make a magnetic bio-carbon composite material (BC-Fe) and applied it to the treatment of heavy metal wastewater. The results show that the removal rates of Cd and Zn by BC-Fe are as high as 61.1% and 60.4%, respectively. The specific components of organic dye wastewater are complex, with high chroma, high organic content, strong toxicity, and difficult to biodegrade. Therefore, the treatment of this type of wastewater is also a hot spot for researchers\(^12\). Fan et al. developed a magnetic chitosan/GO composite material to adsorb methylene blue, and achieved a good removal effect\(^10\). Magnetic materials can easily separate the adsorption material from the water body, which is conducive to the reuse of materials and the centralized treatment of pollutants\(^14\). Refer to Table 1 for a detailed summary of magnetic materials in water treatment.

| Adsorbent                                                                 | pH   | Maximum adsorption capacity | References |
|---------------------------------------------------------------------------|------|----------------------------|------------|
| MgO-Fe composite material                                                 |      | 90%                        | \(^13\)    |
| Magnetic biochar composite material (BC-Fe)                               | Cd\(^2+\)/Zn\(^2+\) | 6.0                        | 8.6/21.3   | \(^13\) |
| Magnetic Ze-MOFs Nanocomposite                                            | Pb\(^2+\)/methylene blue | 7.0 | 102/128 | \(^14\) |
| Magnetic GO/Fe\(_3\)O\(_4\) composite material                           | Cu\(^2+\) | 5.3 | 18.26 | \(^9\) |
| Magnetic ferroferric oxide adsorption material                            | Cu\(^2+\) | 5.3 | 90% | \(^9\) |
| Amino and carboxyl functionalized                                         | Cd\(^2+\)/Pb\(^2+\) | 5.0 | 97.74%/91.44% | \(^15\) |
| Magnetic CuS/γ-Fe\(_2\)O\(_3\) composite material                        | Congo red | 96.51% |              | \(^16\) |
| Magnetic ZEOLITE/Chitosan Microspheres                                    | Phenol | 6 | 91.46% | \(^17\) |
| Magnetic Fe\(_3\)O\(_4\)/BiOBr composite material                        | Rodin Red | 99.6% |              | \(^18\) |
| Magnetic Magnetic Fe\(_3\)O\(_4\)/red mud nanoparticles                  | Ciprofloxacin | 111.11 |              | \(^19\) |
| Cerium/ferric oxide composite material                                    | Cr   | 2 | 98.93% | \(^20\) |
| Modified magnetic kaolin material                                         | Methylene blue | 50.2 |              | \(^21\) |

3. Microbial fuel cell

3.1. Introduction of microbial fuel cell
MFC began in 1839, derived from reports that Grove used gaseous fuels to generate electricity\(^22\). In 1911, Potter discovered that the catalysis of microorganisms can generate voltage in a fuel cell system, and microbial battery technology also began to develop. In 1990, Habberman and Pommer reported
for the first time an MFC that used municipal sewage for five years of continuous operation without any failure[23]. In 2004, Logan of the University of Pennsylvania in the United States applied single-chamber MFCs to wastewater treatment and generated electricity, and made great progress[24]. MFCs are devices that use microorganisms as catalysts to convert organic matter in wastewater into electricity[1, 25, 26]. The structure consists of a cathode zone and an anode zone with a proton exchange membrane in between. The microorganisms on the anode produce electrons and protons, and the electrons generate an external current through an external circuit[27]. Protons reach the cathode through the exchange membrane or other means, where the cathode undergoes a reduction reaction with oxidizing substances to complete the charge transfer between the batteries[28]. Studies have shown that MFCs have a better treatment effect when treating some wastewater with good biochemical properties, and output electrical energy of 146~371mW/m[29].

3.2. Application of magnetic substances in microbial fuel cells

At present, there are many ways to increase the power of MFCs. As shown in Table 2, a lot of research has been done on electrode materials, MFC frame design, etc., but there are few studies on the introduction of magnetic fields into MFCs. Magnetic fields have been used in sewage treatment for many years. Practice has proved that magnetic fields have effects on microbial metabolism, enzyme activity, and cell membrane permeability[30]. In recent years, researchers have continuously increased the research on the application of MFCs under magnetic fields, and magnetic materials have also shown good treatment effects. Liu et al. found that the electricity production of Shewanella MFC under a 100mT magnetic field increased by 20-27%[31] A magnetic field of appropriate strength can accelerate the start-up of anammox and MFCs in wastewater treatment[32].

| Table 2 | Several mechanisms to increase the output power of MFC |
| Mechanism | References |
| --- | --- |
| Structure design of MFC | [2] [3] [33][34] |
| Electrode material selection | [35, 36] [37] |
| Electrode spacing | [38, 39] |
| Fuel selection | [40] [41] [42] [41] |
| Introduction of magnetic field | [30, 31, 43, 44] |
| Improved cathode | [45] [46] |

3.2.1. The influence of magnets on the efficiency of MFC power generation

As a convenient and reusable substance, especially magnet, it provides a new research method to enhance the activity of microorganisms[47]. Previous studies have shown that an appropriate magnetic field can improve the microbial activity in the sewage treatment system and accelerate the degradation of pollutants. Li et al. demonstrated that magnetic fields can be used to increase the electricity production of single-chamber MFCs inoculated with Shewanella. However, the MFC in this pure breeding environment has poor ability to resist the external environment, and the sewage needs to be sterilized in advance[31]. Tao et al.[30] found that the magnetic field can also have a positive effect on the dual-chamber MFC with activated sludge, but the stimulation mechanism and scope of application are still unclear. Tong et al.[43] used a weak static magnetic field (MF) near the anode to enhance the MFC performance. Studies have shown that the application of MF can be used as a cost-effective and convenient strategy to improve the performance of MFC and has great potential. The power generation efficiency of MFCs with different structures is also different. Li et al. used three different structures of MFC reactors to test the MF effect under different working conditions. The structure types include single-chamber MFC, double-chamber MFC, and three-electrode MFC systems (Figure 1). Experiments have found that, depending on the reactor and the selected medium, the microbial fuel cell continuously produces greater power under MF irradiation, with a difference between 20 and 27%. After 170 hours of operation, the current of the magnetic microbial fuel cell was completely higher than that of the control group. However, the researchers found that the enhancement effect of the magnetic field on the MFC seems to be instant and reversible. Experiments were conducted with
different MFCs. When the MF is present, the voltage of the MFC rises immediately, and when the MF disappears, the voltage of the MFC drops significantly. This shows that after the magnet is suddenly removed, it will have a certain impact on the microorganisms that have adapted to the magnetic field. After the magnetic field is restored, the catalytic activity of the bacteria may be enhanced, but this will not permanently change the cell structure [31].

![Schematic diagram of MFC structure with three different structures](image)

(A) Schematic diagram of single-chamber magnetic MFC; (B) double-chamber magnetic MFC; (C) three-electrode system; (D) magnetic field (1) resistance; (2) magnet; (3) anode; (4) cathode; (5) PEM; (6) working electrode; (7) reference electrode; (8) counter electrode; (9) voltage regulator

3.2.2 Magnetic material as MFC electrode

There are few researches on the magnetic electrodes of MFC at present, and the influence of the magnetic field on the electrodes is still unclear. Choi et al. [44] evaluated the impact of permanent magnets on membraneless microbial fuel cells (MLMFC) and made a series of evaluations on the electrochemical performance of magnetic materials. The structure of the reactor is shown in Fig.2. The experimental results show that when a magnet is added to the cathode in the MLMFC, the current increase reaches the maximum (about 22.5 mA/m) when the device is aerated. After gas, the current generation rate is low (about 19.1 mA/m). The installation of the magnet on the cathode increases the current of the MLMFC by approximately 18%. When the experiment is not aerated, the current generated by the MLMFC with magnets will also be greatly increased (4.8 mA/m) compared with the control experiment (2.7 mA/m). The results show that, with or without aeration, the addition of magnets will increase the current of MLMFC. The increase in current is proportional to the concentration of dissolved oxygen in the cathode compartment of the MFC. Installing a magnet on the cathode makes more dissolved oxygen to enter the cathode, which provides a new environmentally friendly and energy-saving method for increasing dissolved oxygen in the water. Zhou et al. [48] put a rubidium-iron-boron magnet with MF intensity of 160mT into liquid nitrogen at 100°C, 200°C, and 400°C for demagnetization, and obtained magnets with MF intensity of 0, 20, 80mT, and used them as anodes to construct MFC to explore the influence of different magnetic field strengths on anode microorganisms. MFC-0 mT, MFC-20 mT, MFC-80 mT and MFC-160 mT represent MFCs with magnet anodes with SMF strengths of 0 mT, 20 mT, 80 mT and 160 mT, respectively. After 9 days of operation of the reactor, the influence of the anode magnet on the performance of the MFC became obvious. The output voltage of the MFC with the magnet as the anode was much higher than that of the control group. Among them, the average peak voltage of MFC-80 mT was the highest (350 mV), followed by MFC-20 mT (315mV), MFC-160mT (293mV), MFC-0mT (171mV). Moreover, the power density generated by the magnetic anode MFC is significantly greater than that of the non-magnetic anode MFC, and the measured power density of MFC-80mT is about twice higher than that of MFC-0mT. By controlling the MF, it is found that the 80mT magnet MFC has the performance as the best anode material. It was found that magnetic MFC has lower internal resistance compared to
non-magnetic MFC. However, there are lower activation resistance and diffusion resistance in the magnetic MFC. Compared with the diffusion resistance, the ohmic resistance and the activation resistance are relatively small, which indicates that the transmission limitation plays a key role in the performance of the MFC with the magnet as the anode. Compared with non-magnetic MFC-0 mT (835.3), magnetic MFC has much lower diffusion resistance: MFC-160 mT is about 585.9, MFC-80 mT is about 365.1, and MFC-20 mT is about 608.2. Therefore, SMF reduces the internal resistance of the MFC with a magnet as an anode mainly by reducing its diffusion resistance. At the same time, Zhou et al. [49] found that the higher the magnetic field strength, the better the COD removal rate, indicating that the magnetic field promotes the degradation of organic matter. In terms of the coulombic efficiency, experiments have found that although the magnetic field can increase the coulombic efficiency, the exogenous electricity-producing bacteria can more easily capture electrons on the substrate to generate electricity.

3.2.3. Magnetic materials to construct electroactive biofilms

The formation of biofilm is a complex process, including the adhesion of bacteria to the substrate surface and cells to cells, thereby forming a multilayer biofilm [50, 51]. It usually takes a long time to build a biofilm, usually ranging from a few days to a few months. Magnetic materials have been shown to be effective in capturing bacterial cells, and are commonly used to remove bacteria in various types of water. Zhou [50] demonstrated the use of magnetic materials to capture sulfur reducing bacteria to form biocomposite materials. An artificial electroactive biofilm is formed through the contact between the magnet and the electrode. In the construction of an electroactive biofilm with the help of an external magnet, the current was recorded and its electrocatalytic activity was evaluated. It was found that in the absence of an external magnet, almost no current was observed, but when the magnet was close to the working electrode, the current was generated immediately, which proved the feasibility of constructing electroactive biofilms in this way. This also indicates that bacterial cells can be attached to the electrode stably. At the same time, studies have also investigated the influence of magnetic fields on the bacterial communities of anode biofilms. Studies have shown that although the species richness index of non-magnetic MFC anode biofilms is higher than that of magnetic MFC, the magnetic field reduces the species richness, but had little impact on the species uniformity. Lee [52] immobilized glucose oxidase in nanostructured magnetic materials to make enzyme-based biocomposites, and constructed a magnetically switchable bioelectrocatalytic system by contacting a magnet with an electrode. Magnetic materials have been shown to be effective in capturing bacterial cells, and are commonly used to remove bacteria in various water.

3.2.4. The role and influence of magnetotactic bacteria in MFC

Magnetotactic bacteria (MTB) is a special kind of bacteria that can move along the lines of magnetic force. Its cells contain magnetosomes that are sensitive to magnetic fields. They play a guiding role and move with the help of their own flagella [53]. MTB was discovered by the American biologist Bracomor in 1975. Since then, people have carried out a lot of research on MTB and made great
progress\textsuperscript{[54]}. It is found that the MTB separated in the northern hemisphere will move to the geographic north pole, the MTB separated in the southern hemisphere will move to the geographic south pole, and near the equator, there are both the South pole and the North pole. Such tropism can help MTB escape the harmful oxygen environment, move down and stay in the optimal area suitable for its growth\textsuperscript{[55]}. In addition to growing in micro-oxygen water, MTB is also found in the soil. Fassbinder isolated MTB from the soil of a pasture in South Bavaria, and found that the MTB is diverse in shape, such as spherical, rod-shaped, arc-shaped, and spiral. The types of physiological metabolism are also diverse, including aerobic, microaerobic, facultative aerobic, anaerobic, etc\textsuperscript{[56]}. In terms of wastewater treatment, a certain amount of flocculant and MTB are added to the wastewater, and harmful impurities such as heavy metal ions in the wastewater are bound with MTB, and the steel wool filter layer of a high-gradient magnetic filter is used to remove the harmful magnetic fixation impurities, and enable bacteria, viruses and algae to be separated from the water\textsuperscript{[57]}. Compared with ordinary magnetic solidification technology, MTB has the advantages of strong fixing ability, easy separation of the carrier from the sludge, and recycling, which can greatly improve the treatment efficiency and save costs\textsuperscript{[57]}. Although MTB is widely used in wastewater treatment, research on the application of magnetotactic bacteria in MFC has not yet been found. If MTB is loaded on a new composite magnetic material and applied to MFC, the magnetic properties of MTB can be used to remove heavy metal ions from water and improve the power generation efficiency of MFC, which will provide a new direction for improving the power generation efficiency of MFC\textsuperscript{[58]}.

3.2.5. Cultivation and screening of magnetotactic bacteria

MTB can be obtained by culturing in a specific medium or by implanting magnetic particles in bacteria. But so far, there are few domestic relevant reports\textsuperscript{[59]}. Foreign researches on the cultivation of MTB have become mature, and most of the cultures are magnetospirillum\textsuperscript{[60]}. Magnetospirillum is a kind of freshwater bacteria. The ferromagnetic particles in the cell are Fe\textsubscript{3}O\textsubscript{4}, with a length of 3~5μm and a width of 0.5~0.7μm. The shape is shown in Figure 3. The particles are fine and uniform and the appearance characteristics vary depending on the species, which are mostly called chain arrangement in the bacteria. MTB is widely present in the micro-oxygen layer. Y. Matsunga isolated the spiral MTB in KOGANEI, Tokyo, and then cultured it in a fermenter\textsuperscript{[59]}. There is no uniform standard for MTB screening methods, and most of the current screening methods use directional magnetic field separation. Utilizing the characteristics of northward movement of MTB, a directional magnetic field is created so that MTB can move northward to achieve the purpose of separation and enrichment\textsuperscript{[60]}. Different types of MTB require different nutrients. Taking spiral MTB as an example, the growth medium includes the following ingredients (standard per liter): 0.68g KH\textsubscript{2}PO\textsubscript{4}, 0.12g sodium nitrate, 0.1g thiol Sodium acetate. At the same time, 10mL of Wolff vitamin solution, 5mL of Wolff mineral solution, and 2mL of iron quinic acid were added to the medium (each milliliter of water contained 0.27g of FeCl\textsubscript{3} and 0.19g of quinic acid). In addition, 5μg/mL tetracyclines should be added during use, and the pH value should be adjusted with NaOH aqueous solution to keep it around 6.75\textsuperscript{[59]}.

![Fig.3 Transmission electron micrographs of magnetotactic bacteria and magnetosomes\textsuperscript{[61]}](image)

4. Difficulties and challenges of MFC application

At present, due to the potential advantages of MFCs, it has a good development prospect, but it is still relatively far away to be used as the main power source in real life. The main reason is that the output
power of MFCs cannot meet actual requirements. The main factor that determines the output power density of MFCs is the related electron transfer process and the slow electron transfer rate of biological systems is the bottleneck of the development of MFCs. The main factors that affect the electron transfer rate are: the resistance of the external circuit, the transfer of electrons from the microorganism to the electrode, the oxidation of the substrate by the microorganism, and so on. The use of heavy metal nanoparticles and carbon nanotubes to modify the electrode surface is a good way to reduce the internal resistance of MFCs, but it is costly. It is expensive and not suitable for promotion and application. The appearance of magnetic materials can greatly improve the efficiency of electricity production. However, at present, the mechanism by which magnetic materials improve the power generation efficiency of MFCs is not clear, so a complete and reasonable method for improving the power generation efficiency of MFCs cannot be formulated, which limits the practical popularization and application of MFCs. There are also a few of researches on the correlation of MTB’s impact on MFCs. The correlation between MTB and MFCs’ power generation efficiency is still unknown, but this provides a new way for researchers studying MFCs.

5. Outlook
MFCs, as an innovative and environmentally friendly technology integrating wastewater treatment and power generation, has received rapid attention in recent years. Most of the current researches are mainly aimed at improving the power generation efficiency and sewage treatment performance of MFCs. The amount of researches related to magnetic materials as MFCs reactor structure, cathode and anode is increasing. Researches on magnetic materials and MFCs’ electricity production efficiency are also underway, but the influence of magnetotactic bacteria on the performance of MFCs has not yet been clarified. I hope that more scholars will participate in this researches in the future, in order to indicate new directions for the optimization of MFCs structure and electrode materials. I believe that MFCs technology will make breakthrough progress in the near future!

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