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Capacitive Ferrosoferric Oxide as an Anode to Enhance the Electrical Energy Output and Storage of Microbial Fuel Cells

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Abstract: Microbial fuel cells (MFCs) are electrochemical electricity generation devices that use microorganisms to degrade organic matter to produce electrical energy. The anode of microbial fuel cells (MFCs) is the carrier to which electrogenic microorganisms attach. However, traditional anodes do not have a storage function, which limits the application scope of microbial fuel cells. Effectively storing and utilizing the energy generated by MFCs is an important focus of ongoing research and is also key to realizing their effective application. A carbon felt film (CF) was used as the substrate, and a carbon felt film/ferrosoferric oxide (CF/Fe_3O_4) electrode was prepared by a hydrothermal method. In the power density test, the MFC system constructed with the CF/Fe_3O_4 electrode as a capacitive biological anode had a maximum power density of 5.09 W/m^3, which was 4.43 times higher than that of the blank carbon felt film anode. In the charge–discharge test, the stored charge (Q_s) released by the CF/Fe_3O_4 electrode was 157.12 C/m^2 higher than that of the CF electrode after 45 min of charging–discharging. The modified ferrosoferric oxide anode was used in a microbial fuel cell to provide a scientific basis to construct microbial electrochemical systems with high power and energy storage functions.

Keywords: microbial fuel cell; ferroferric oxide; energy materials

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1. Introduction

Microbial fuel cells (MFCs) directly convert chemical energy stored in organic matter into electrical energy by using microorganisms as the anode catalyst. MFCs use waste biomass from a wide range of cheap sources or high-concentration organic wastewater and sludge as raw materials to realize waste treatment and high-efficiency electricity generation. They have diverse fuel sources, mild operating conditions, low costs, and do not produce pollution.

The anode is a key component of an MFC [1,2]. As the carrier for the attachment of electrogenerative microorganisms, it affects the number of attached electrogenerative microorganisms and the transfer of electrons from microorganisms to the anode. The surface of some anode materials can be modified to prepare highly active anode materials with a large surface area to promote the easy adhesion and growth of electrogenerative microorganisms. This can promote the easy transfer of electrons from microorganisms to the anode and provide low internal resistance and strong conductivity, which can improve the electricity generation ability of an MFC.

Most anode material modification studies have focused on carbon materials and their modification or the development of carbon composites [3]. There are many kinds of carbon materials, including graphite, carbon felt, carbon foam, carbon cloth, and mesh glass carbon (RVC). To improve the performance of the anode, researchers have attempted to improve the carbon material anode via anode modification. Ammonia treatment, chemical doping, iron oxide coatings, [4–6] and other methods have been used to modify anode materials. Carbon nanotubes (CNTs) have been widely used in MFCs because of their special tubular

structure, which allows their ends to infiltrate microbial species to promote electron transfer from electrogenerative bacteria to the anode [7]. To further improve the performance of CNTs in MFCs, they can be coated with polymers [8].

Due to the rapid development of supercapacitor materials in recent years, the application of capacitive materials in MFC systems has attracted attention. Lv [9] and Zou [10] used capacitive materials (metal oxides, conductive polymers, etc.) to modify an MFC anode. Studies have shown that the output power of an MFC is directly proportional to the capacitance of the electrode, and these same studies significantly improved the power of MFCs. The main metal oxides used in MFC anodes are Fe_3O_4 [11], TiO_2 [12], RuO_2 [13], etc. Among these, iron oxide is the most common. Due to its biocompatibility, low cost, and simple preparation process, iron oxide can promote the rapid transfer of redox electrons and has been widely used in MFCs. Park et al. [14] synthesized a Fe_3O_4/CNT nanocomposite and used it in dielectric-free microbial fuel cells. The effect of the Fe_3O_4 content was studied, and the optimal ratio was explored to obtain the optimal MFC performance. The maximum power density of 830 mW/m^2 was obtained by the Fe_3O_4/CNT composite anode with a ratio of 30 wt.%. Fu et al. [15] prepared an iron/iron oxide-modified BMFC anode by electrodeposition. The composite anode had a lower surface contact angle and higher wettability, which promoted the adhesion of bacteria on the anode surface. The open-circuit potential of the modified BMFC was about 1050 $\pm$ 50 mV, while that of the blank BMFC was only 700 $\pm$ 50 mV. The output power density of the modified BMFC was 17.4 times higher than that of the blank, and the anode exchange current density of the modified BMFC was 393 times higher than that of the blank. Ji et al. [16] modified ITO electrodes using α-Fe_2O_3 nanorods and chitosan (CS) via layer–layer self-assembly. Compared with the blank anode, the output power of the modified MFC was 320% higher, and the $(Fe_2O_3/CS)_4$/ITO-modified MFC had the highest output current. Pandit et al. [17] constructed a modified Fe_2O_3 anode, whose power density and Coulombic efficiency were increased by 40% and 33%, respectively. Cyclic voltammetry results showed that Fe_2O_3 modification improved the electrochemical activity of the electrode.

Therefore, carbon materials and capacitive materials can give full play to their respective advantages and make up for their individual shortcomings. Considering that the electrode material is also attached to the biofilm, the biocompatibility of the electrode material must be also good. Therefore, Fe_3O_4 was synthesized by a one-step hydrothermal method on a carbon felt matrix, and the capacitive material Fe_3O_4 was used to modify the carbon fiber felt anode. This provided a large surface area, good conductivity, high specific capacitance, easy electrolyte diffusion, easy adhesion of microorganisms, and promoted the transfer of electrons from microorganisms to the electrode surface. These combined advantages improved the electricity generation and energy storage performance of the MFC. The results of this paper can also aid the development of anode materials for MFCs that are based on capacitive energy to promote wastewater recycling. A comparative study was conducted using the unmodified carbon felt film anode (CF). This paper can provide theoretical guidance for microbial fuel cells based on capacitive anode materials to promote wastewater energy recovery.

2. Experimental Methods

2.1. Carbon Felt Film Pretreatment

A carbon felt film with a thickness of 3 mm was cut into a size of 2 cm $\times$ 2 cm, and then the carbon felt film was soaked in an ethanol solution. Ultrasonication was carried out for 50 min to remove oil stains from the surface of the carbon felt film.

After the initial degreasing treatment, the carbon felt film was repeatedly rinsed with distilled water and then placed in a 10% hydrogen peroxide solution and heated in a constant-temperature water bath at 90 °C for 3 h.

The carbon felt film was removed from the water bath, rinsed repeatedly with distilled water, and then soaked in the 5 wt.% hydrochloric acid for 2 h to activate and increase the hydrophilicity of the carbon felt film.

The final processed carbon felt film was rinsed repeatedly with distilled water and placed in a drying oven at 60 °C.

2.2. Preparation of Carbon Felt Film/Ferric Oxide Electrode Material

A total of 20 mL distilled water was added to a 50 mL beaker and the carbon felt was added. Argon was added to remove oxygen for 30 min, and then 1.08 g $FeCl_3 \cdot 6H_2O$ and 0.556 g $FeSO_4 \cdot 7H_2O$ were added, stirred for 10 min (rotating speed 200 r/min), and the solution became orange-red. The water bath was heated to 60 °C, and then 2.5 mL of concentrated ammonia water was slowly added to the above solution (finished within 2 min). The pH changed to ~9, and the solution changed from red to black. Stirring was continued for 30 min, and the temperature rose to 80 °C. The solution was aged for 30 min, and it along with the carbon felt were poured into a hydrothermal kettle and placed in an oven. The hydrothermal reaction was finished and then cooled to room temperature.

2.3. Start-Up of Microbial Fuel Cells

A two-compartment glass cylinder made of Plexiglas was used as the MFC reactor (Plexiglass Manufacturing Co., Ltd., Harbin, China), in which the prepared electrode was used as the working electrode, and three graphite rods were fixed with polished titanium wire as the cathode. The anode chamber and cathode chamber were fixed with iron clips, and the two chambers were separated by a proton-exchange membrane. Anode chamber: 2.5 g/L sodium acetate was used as the carbon source for microbial degradation. Cathode chamber: 10 g/L potassium ferricyanide was used as an electron acceptor for the cathode. Microorganisms in the anode chamber were inoculated from activated sludge, and strains were successfully acclimated in other microbial reactors. The MFC was connected to 1000 ohm resistance and domesticated in an incubator at 30 °C. The output voltage and anode potential of the MFC were collected every 30 min by using a data recorder, and the operating cycle of the MFC was recorded. After the MFC operated stably for more than three cycles, electrochemical tests were carried out.

2.4. Output Voltage of the Microbial Fuel Cell

A higher output voltage of an MFC indicates a longer duration and better performance. The output voltage refers to the potential difference between the anode and cathode. This was determined by connecting a load resistance to the MFC and collecting data to record the voltage of the MFC according to Ohm's law. The relationship between the output voltage and load resistance is shown in Formula (1):

$$U = R_{ex} \times I \quad (1)$$

In the formula:

- R_{ex}—Load resistance, unit: Ω
- I—Current, unit: A
- U— Output voltage, unit: V

2.5. Electrochemical Performance Test

To investigate the electrochemical performance of the prepared electrode, electrochemical tests were carried out. All tests were conducted using a traditional three-electrode system. The electrolyte was phosphate-buffered liquid, and the electrode materials were the working electrode, a metallic platinum electrode (opposite electrode), and a saturated calomel electrode (SCE) as the reference electrode. The test electrode data were recorded on a CHI760C workstation (Chenhua Company, Shanghai, China) and an SP-240 workstation (SP-240; Bio-Logic, Seyssinet-Pariset, France).

2.6. Scanning Electron Microscopy-Energy-Dispersive Spectroscopy (SEM-EDS)

An Inspect S50 scanning electron microscope (SEM) (Sirion200, FEI Ltd., Eindhoven, The Netherlands) was used to observe the microstructure and surface morphology of the samples. An energy-dispersive X-ray detector (EDS) (Sirion200, FEI Ltd., The Netherlands) was used for the quantitative and qualitative analysis of samples.

3. Results and Discussion

3.1. Physicochemical Characterization of the CF/Fe_3O_4 Electrode

Figure 1 shows the SEM images of the blank carbon felt electrode (CF) and ferric oxide electrode (CF/Fe_3O_4) under magnifications of 1000× and 10,000×. Figure 1a,b show the morphologies of the CF, which shows many interwoven carbon fibers and a smooth surface. In Figure 1c,d, the surface of the carbon fiber became rough after the deposition of ferrosoferric oxide. After the carbon fiber was coated with ferric oxide, its specific surface area was increased, which provided more sites for microbial metabolism.

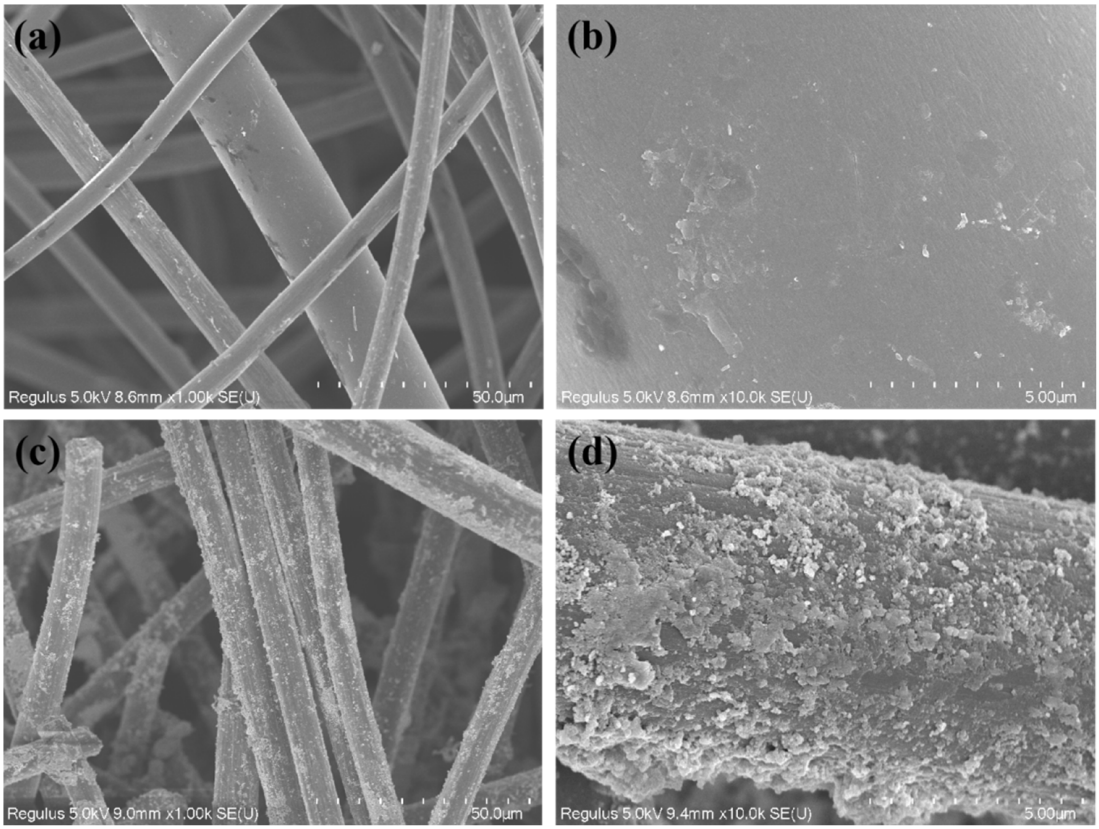


Figure 1. SEM images of the CF electrode (**a**,**b**) and CF/Fe_3O_4 electrode (**c**,**d**) prepared under optimal conditions at 1000× and 10,000× times magnification.

Figure 2a shows the EDS spectra of the CF electrode and CF/Fe_3O_4 electrode prepared under optimal conditions. In Figure 2a, carbon accounted for the highest mass percentage (96.89%), confirming the presence of the CF electrode. In Figure 2b, oxygen and iron accounted for mass percentages of 65.87% and 34.13%, respectively. This is consistent with the fact that ferric oxide is the main active substance and shows that ferric oxide was prepared on the carbon felt matrix.

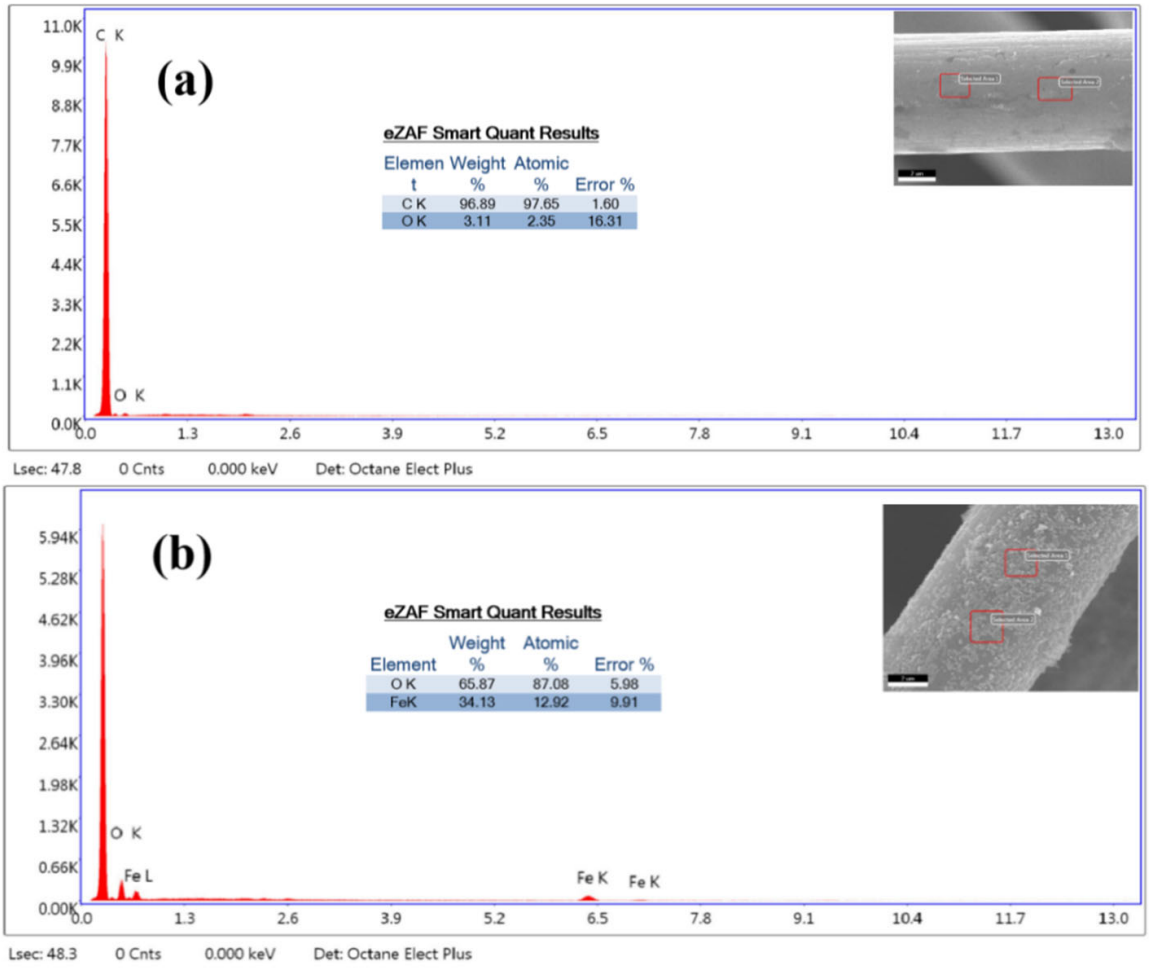


Figure 2. EDS spectra of CF (**a**) and CF/Fe_3O_4 (**b**) electrode prepared under optimal conditions.

3.2. Power Density Test and Polarization Curves of MFCs

CF/Fe_3O_4 and blank carbon felt film (CF) electrodes were used as MFC anodes to investigate their power generation and energy storage performance, in which CF was used as a comparison. Figure 3 shows the power density curves of the MFCs (a), polarization curves of the MFCs (b), and anode polarization curves (c) of MFCs with CF/Fe_3O_4 or CF as the anode. According to the results, the maximum power density of the MFC with the Fe_3O_4-modified anode reached 5.09 W/m^3, which was 4.42 times higher than that of the blank carbon felt (1.15 W/m^3). The output power of the MFC modified with Fe_3O_4 was greatly improved. Figure 3b shows the polarization curve of the MFC. When the voltage of the MFC was 0.3 V, the output current of the MFC with the CF anode was 1.3 mA/m^2, while the output current of the MFC equipped with the Fe_3O_4-modified anode was 2.2 mA/m^2. The results show that the polarization of the CF/Fe_3O_4-modified MFC cell had better polarization performance, with a trend consistent with the power density curve. The cathode condition was identical in all cells, showing that anode modification was responsible for the improved performance of the MFC. As shown in Figure 3c, the maximum open-circuit potential of the CF/Fe_3O_4 anode reached −0.3 V, while the CF electrode only reached −0.15 V. The CF/Fe_3O_4 anode also had a higher output current density under the same potential. These results indicate that the CF/Fe_3O_4 electrodes applied in an MFC can adsorb more microorganisms, increase the number of reactive sites, and thus increase the output voltage, reduce the cell polarization, and improve the power density of the MFC.

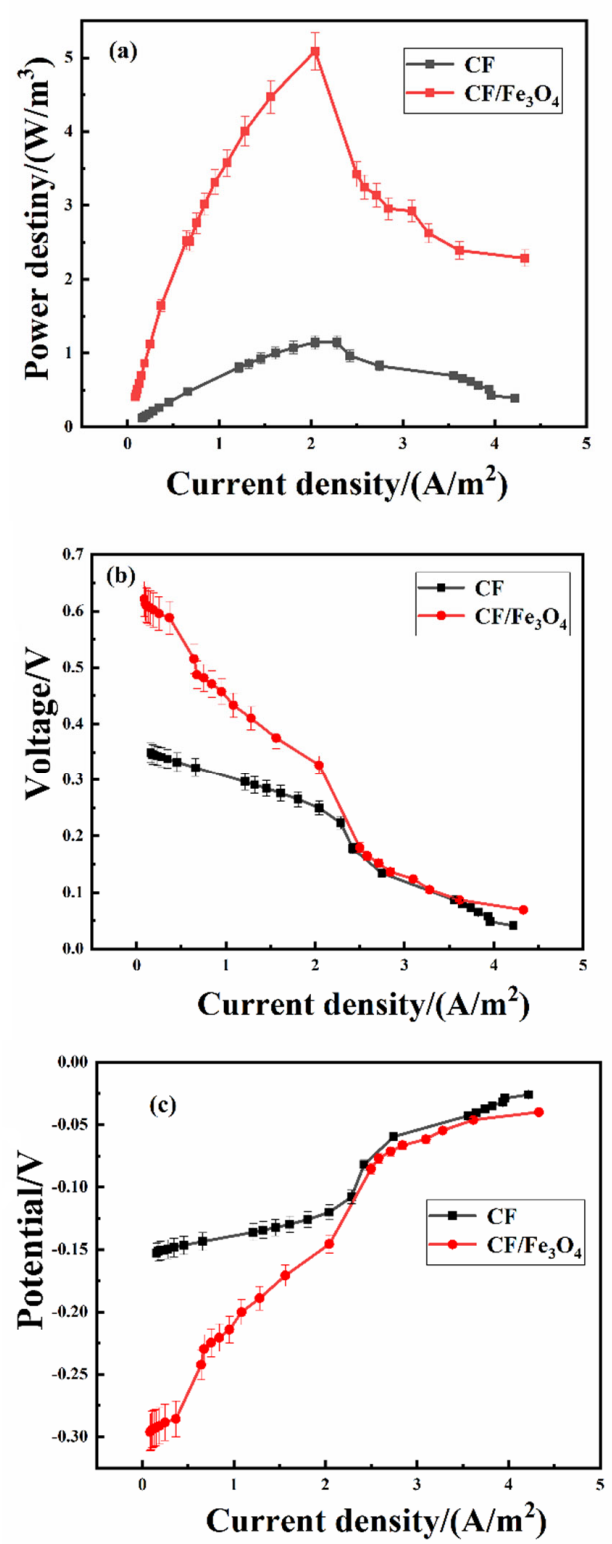


Figure 3. Performance of MFCs with two anodes: (**a**) power density curves; (**b**) polarization curves; (**c**) anodic polarization curves (the external resistance (R) from 100 to 9000 Ω).

3.3. Energy Storage Test of MFCs

Figure 4 shows the discharge curves of different electrodes at different charging times. Figure 3a–d, respectively, show the discharge curves of charging–discharging for 15 min (C15/D15), charging–discharging for 30 min (C30/D30), charging–discharging for 45 min (C45/D45), and charging–discharging for 60 min (C60/D60).

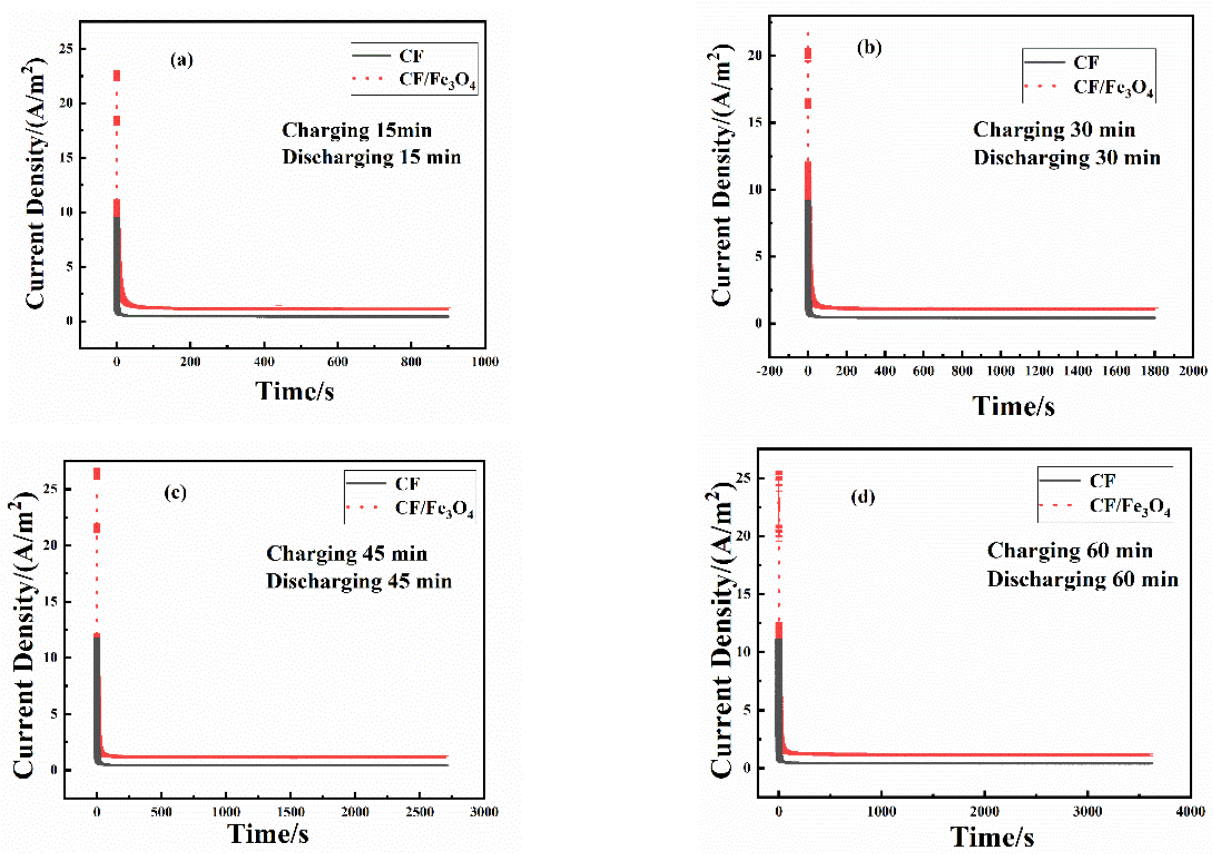


Figure 4. Discharge test of MFCs with two different anodes measured at −0.1 V (The MFCs was under the closed-circuit condition).

Figure 4 shows that both anodes had a peak current at the beginning of discharge, which was represented by a high current density (i_h). Upon increasing the discharge time, the peak current density began to decline and gradually became stable, as expressed by a stable current density (i_s). A higher peak current density indicates a greater charge storage capacity of the electrode [18–22]. Figure 4a–d shows that the peak current density and stable current density of the electrode modified with ferrosoferric oxide were both improved. In this paper, Q_s is the stored electricity, and Q_t is the total electricity generated. By comparing the charging–discharge curves of the two groups and combining them with Table 1, the peak current density and stable current density of the two electrodes increased over time from 0–45 min, reaching the maximum value at 45 min. The Q_t released by the CF/Fe_3O_4 anode reached 3313.85 C/m^2. The peak current density was 24.43 A/m^2, which was 2.79 times higher than that of the blank anode. A higher peak current density and total electric quantity indicate better electrical output performance of the MFC. Under the conditions of C45/D45, the stored energy Q_s released by the CF/Fe_3O_4 electrode was 157.12C/m^2 higher than that of the CF electrode. The results indicate that modifying the anode material with a capacitive ferrosoferric oxide significantly improved the electricity storage performance of the anode. Combined with the previous SEM results, the electrode surface of the modified ferric oxide was rougher, which provided more metabolic sites for microorganisms. Because ferrosoferric oxide is capacitive, during the charging process, the microbial charge was stored in the capacitive anode. During discharge, it instantly generated two parts of the charge: one was stored in the capacitive material, and some was instantaneous. Therefore, the anode modified with ferrosoferric oxide had a higher peak current density and storage capacity.

Table 1. The parameter of discharging curves with CF anode and CF/Fe_3O_4 anode.

Anodes	Parameters	C15/D15	C30/D30	C45/D45	C60/D60
CF anode	i_h (A/m^2)	8.72	8.37	10.75	10.16
	i_s (A/m^2)	0.42	0.42	0.42	0.41
	Q_s (C/m^2)	28.76	44.39	51.73	94.12
	Q_t (C/m^2)	406.76	800.39	1185.73	1570.12
CF/Fe_3O_4 anode	i_h (A/m^2)	20.97	21.69	24.43	22.82
	i_s (A/m^2)	1.11	1.06	1.15	1.15
	Q_s (C/m^2)	91.07	124.93	208.85	242.90
	Q_t (C/m^2)	1090.07	2032.93	3313.85	4382.90

Figure 5 shows changes in the anode potential over time when CF and CF/Fe_3O_4 anodes were charged for 15 min. At this time, the MFC circuit was in the open state, and the anode began to charge. Because electrons are negative, the anodic potential was negative. Over time, the potential of the two anodes gradually flattened out, and the blank electrode finally reached −0.29 V, and the CF/Fe_3O_4 electrode was about −0.21 V. The potential of the modified electrode declined relatively slowly because iron oxide has good capacitance, allowing it to store charges generated by microorganisms. Therefore, during charging, the capacitor-modified anode potential declined slowly, while the blank anode showed a low capacitance, so the potential declined rapidly [23–30].

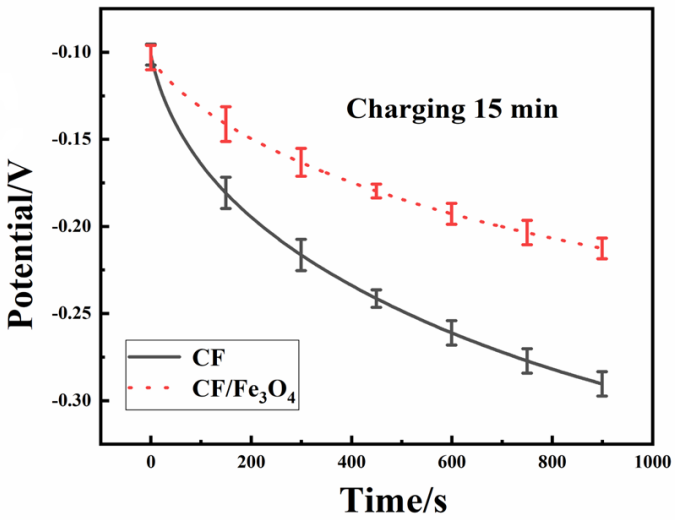


Figure 5. Time–potential test of MFCs with two anodes (OCV: 15 min. The MFCs were under open-circuit condition).

4. Conclusions

This paper studied the performance of a CF/Fe_3O_4 composite prepared by a simple one-step hydrothermal method. It was used as a modified anode in an MFC and compared with an unmodified CF anode. SEM observations indicated that the surface of the carbon fiber modified with ferrosoferric oxide was rougher, showing that the carbon fiber was coated with ferrosoferric oxide, which increased its specific surface area. The quantitative EDS results showed that oxygen and iron accounted for the main elements by mass, consistent with the fact that ferrosoferric oxide was the main active substance. The results showed that the maximum power density of the MFC of the iron oxide-modified anode was 5.09 W/m^3, which was 4.42 times higher than that of the MFC with a blank carbon felt anode (1.15 W/m^3). The energy storage research results showed that under the conditions of C45/D45, the stored energy Q_s released by the CF/Fe_3O_4 electrode was 157.12 C/m^2 higher than that of the CF electrode. Because Fe_3O_4 is cheap and uses a simple preparation

method, it provides an efficient, economical, and simple modification method to prepare Fe_3O_4 capacitive anodes to improve the performance of MFC electricity generation and energy storage. Therefore, the excellent result will facilitate the design of new kinds of anode materials to improve the energy output of MFCs by improving the anode capacitance.

Author Contributions: Y.W. (Writing—review & editing): Preparation, creation and/or presentation of the published work by those from the original research group, specifically critical review, commentary or revision–including pre- or post-publication stages. J.W. (Formal analysis): Application of statistical, mathematical, computational, or other formal techniques to analyze or synthesize study data. J.D. (Conceptualization): Ideas; formulation or evolution of overarching research goals and aims. Y.T. (Resources): Provision of study materials, reagents, materials, patients, laboratory samples, animals, instrumentation, computing resources, or other analysis tools. All authors have read and agreed to the published version of the manuscript.

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