

Effect of the Anode Structure on the Performance of Oily Sludge Sediment Microbial Fuel Cells

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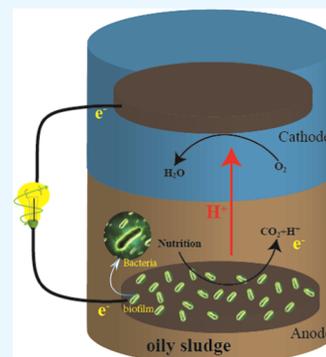
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ABSTRACT: The anode is considered to be a key factor to improve the single-chamber bioelectrochemical system's efficiency to degrade oily sludge in sediment while generating electricity. There are few studies on the effect of the anode structure on the performance of oily sludge MFCs systematically. In this paper, an oily sludge bioelectrical system was constructed using carbon felt and carbon plate as anode materials, adjusting the anode material arrangement as transverse and longitudinal, and using different anode materials from single to sextuple anodes. The results of this study showed that the rate of degradation of oily sludge was greater with carbon felt (17.04%) than with the carbon plate (13.11%), with transverse (23.61%) than with the longitudinal (19.82%) arrangement of anodes, and with sextuple anodes (33.72%) than with a single anode (25.26%) in the sediment microbial fuel cells (SMFCs). A similar trend was observed when the voltage, power density, and electromotive force (EMF) of SMFCs were estimated between the carbon felt and carbon plate, transverse and longitudinal arrangements, single and sextuple anodes. It is concluded that the proper adjustment of anode arrangements, using carbon felt as an anode material, and increasing the number of anodes to six may accelerate the rate of degradation of oily sludge in oily sludge sediment microbial fuel cells (SMFCs). Furthermore, the electricity generation performance was also improved.



1. INTRODUCTION

Oily sludge is a mixture of crude oil and water emulsion, which is produced during the process of crude oil exploration. Naturally, oily sludge is a hazardous environmental pollutant and may cause severe health risks in human beings and animals once exposed to such contamination.¹ The treatment methods of oily sludge include incineration, solvent extraction, biological treatment technology, etc.² For energy-positive waste treatment, novel microbial electrochemical systems are developed such as microbial fuel cells (MFCs), microbial electrolysis cells (MEC), microbial desalination cells (MDCs), microbial reverse electrodialysis cells (MRCs), etc.^{3–7}

MFCs are new bioelectrochemical systems in which a microorganism catalyzes and oxidizes the organic compounds and converts the chemical energy stored by the organic matter into electrical energy.^{8–11} Sediment microbial fuel cells (SMFCs) are special battery systems evolved from the traditional MFC structure. The anode is buried in the anaerobic bottom sludge, and the microorganisms in the bottom sludge are used as catalysts to degrade organic matter into CO₂ and H₂O and generate electrons. The cathode is suspended on the surface of the catholyte, and electrons flow to the cathode through an external circuit. O₂ is used as the electron acceptor to generate clean electricity.^{12–14} The main advantage of the SMFC is that it cannot be used only for waste or wastewater treatment, but its structure makes it particularly suitable for river, lake, or marine sediment restoration. In the early 1990s, MFCs have been used in wastewater treatment. In

recent years, the use of SMFCs to process oily sludge and simultaneous production capacity has become a hot topic for MFC research.¹⁵ Guo et al. showed that the oily sludge is suitable for SMFC operation, with a maximum output voltage of 299.13 mV.^{15,16} However, a key issue that currently limits the practical application of SMFCs is the low power generation efficiency. There are many limiting factors influencing internal resistance.^{17–20} The cathode, anode, pH, appropriate catalysts (activated carbon, cobalt-nitrogen framework), and external resistance are important limiting factors for the performance of the MFC.^{21–23} Among the many factors that affect its electricity production, the anode is considered to be a key factor because it affects the microorganism growth and metabolism of full stop. Anode performance can be enhanced using a better catalyst, anode structure, and anode configuration.^{24–29} It is well known that electrode materials have a great influence on the power generation and pollution control of MFCs. Research has shown that properly changing the number of anodes can improve the power generation capacity.^{30–32} Different arrangements (voltage series and

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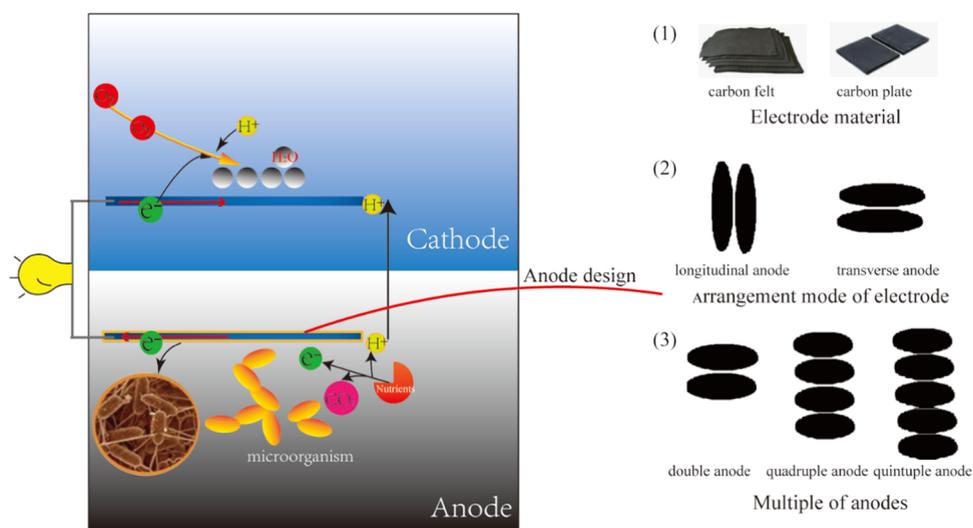


Figure 1. Schematic diagram of the biochemical system with oil sludge deposition.

parallel and biomass series and parallel) can increase the voltage of microbial fuel cells to different degrees.³³ However, the effects of the anode structure (electrode material, anode number, and arrangement) on the performance of oily sludge have not been systematically studied.

Therefore, to explore the influence of the anode structure on the performance of oily sludge SMFCs, in this paper, a series of SMFCs were constructed using oily sludge as the anode substrate, with different electrode materials (carbon felt and carbon plate), different anode arrangements (transverse and longitudinal arrangement), and different anode multiples (single anode, double anode, quadruple anode, quintuple anode, and sextuple anode), and their effects on SMFC power generation performance and oil sludge degradation were systematically studied, providing basic research for oily sludge performance improvement strategies.

2. EXPERIMENTAL SECTION

2.1. Construction and Operation of the Oily Sludge Bioelectrochemical System. The experiment uses a single-chamber bioelectrochemical system without a membrane, and the usable volume of the device is 2 L (Figure 1). The oily sludge is taken from Shengli Oilfield, and the oil content and water content are 40.71 and 10.70%, respectively. Oily sludge and nutrient medium ($\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ 250 mg/L, $(\text{NH}_4)_2\text{SO}_4$ 1000 mg/L, K_2HPO_4 10 000 mg/L, NaCl 5000 mg/L, MgCl_2 180 mg/L, NH_4Cl 500 mg/L) were mixed in 9:1 ratio and placed (1 L) on the bottom of the battery.³³ Both anode and cathode electrodes are made of pretreated carbon-based material (thickness: 10 mm, fixed carbon $\geq 98.5\%$, ash content $\leq 0.05\%$, tensile strength: 0.15 Mpa, purchased from Shanghai carbon factory store). The pretreatment methods of carbon-based materials are as follows: the electrode material was ultrasonically cleaned with absolute ethanol for half an hour to remove oil stains on the surface and then heated in a 30% hydrogen peroxide water bath for 1 h.^{15,16} When heating, it was turned over properly to further remove stains and then rinsed repeatedly with flowing pure water to remove the residues inside the material of hydrogen peroxide.³⁴ Then, it was soaked in an acid–base solution for 2 h, wherein the acid and base solutions were hydrochloric acid solution (1 mol/L) and sodium hydroxide solution (1 mol/L), respectively, then

soaked in deionized water for 2 h, and finally dried.³⁵ Reserve after drying.

The anode is buried in the oily sludge, the cathode is floating on the catholyte surface and in contact with the air, the burial depth of the anode is about 6–7 cm, and the distance between the two electrodes is 12.5 cm. The anode and the cathode are connected to an external resistance (500 Ω) through a copper wire, and the two ends of the external resistance are connected to the data acquisition system, which is connected to a computer to export data, and the data acquisition device records every 10 min. The optimum external resistance is determined by a pre-experiment, and the optimum resistance value is when the external resistance value is similar to the internal resistance.²⁸ The anode design method is as follows (Figure 1): (1) pretreated carbon felt or carbon plate is used as electrodes; (2) the arrangement mode of the anode is set to longitudinal or transverse; and (3) the multiples of anodes are set to double, quadruple, quintuple, and sextuple anode, and the distance between adjacent anodes in the multiple anode setting is 2–3 cm. The SMFCs were kept at a constant temperature of $(30 \pm 2)^\circ\text{C}$, and the voltage was recorded, and the operation was stable. The influence of the carbon felt, carbon plate, electrode arrangement, and multiple anodes on the power generation performance of oily sludge SMFCs was observed. To ensure the accuracy of the experiment, three identical SMFCs were constructed in this experiment. All the data were expressed as the mean.

2.2. Electricity Production Performance Test. The output voltage of the oily sludge bioelectrochemical system is recorded in real time using a data collector and uploaded to the computer. When the SMFC performance became stable, the power density curve and polarization curve were characterized through steady-state discharge. The electromotive force (EMF) and the apparent internal resistance were calculated by fitting the polarization curve.²³ The power density calculation formula of the oily sludge bioelectrochemical system is as follows³⁶

$$P = UI/V \quad (1)$$

where P is the power density, mW/m^3 ; U is the voltage, mV ; I is the current, mA ; and V is the volume of the anode chamber, m^3 .

2.3. Determination of the Crude Oil Removal Rate.

Before and after the operation of the oily sludge bioelectrochemical system, the anode substrate was sampled; after it was naturally dried at room temperature, the crude oil was separated from the sample by the Soxhlet extraction method, and the weight was calculated after weighing.^{37–39} The oil mass fraction and crude oil removal rate are calculated as shown in the following formula. The treatment time of oily sludge SMFCs is 30 days.

$$R_1 = m_1/m_2 \quad (2)$$

$$R_d = (R_0 - R_t)/R_0 \quad (3)$$

where R_1 is the mass fraction of the oil, mg/kg; m_1 is the mass of the crude oil in the sample, mg; m_2 is the mass of the sample after dehydration, kg; R_d is the crude oil removal rate, %; R_0 is the mass fraction of the oil before treatment, mg/kg; R_t is the treatment after oil content, mg/kg.

3. RESULTS AND DISCUSSION

3.1. Influence of Electrode Materials on the Performance of Oily Sludge SMFCs. **3.1.1. Influence of Electrode Materials on the Degradation Performance of SMFCs.** The crude oil removal rate of oily sludge SMFCs with carbon felt (17.04%) is higher than that with a carbon plate electrode (13.11%) as shown in Figure 2. Using carbon felt as an

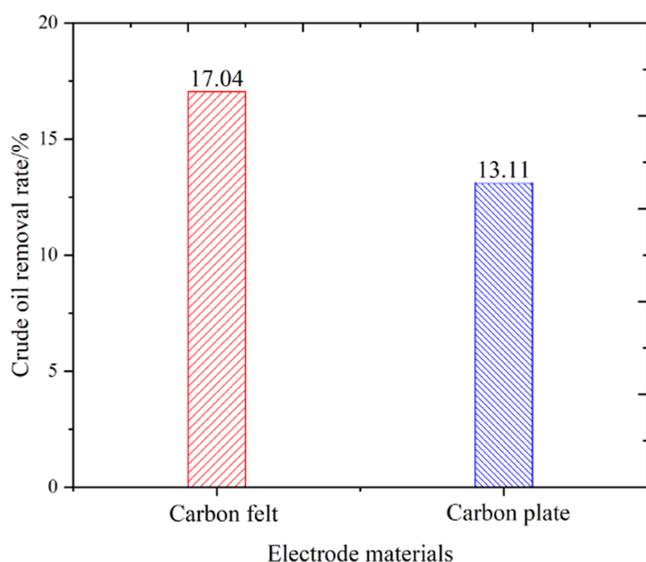


Figure 2. Different electrode materials' SMFC crude oil removal effect.

electrode material has a higher removal rate of crude oil, maybe because the carbon felt electrode has a larger specific surface area than the carbon plate electrode under the same projection area.⁴⁰ Carbon felt is conducive to the adhesion of electricity-producing bacteria, which improves the oily sludge degradation performance of SMFCs.⁴¹

3.1.2. Influence of Electrode Materials on the Electricity Production Performance of SMFCs. As shown in Figure 3, the voltage of the SMFC using a carbon felt electrode is higher than that with a carbon plate in the early stage of electricity generation. This is because although the carbon plate is resistant to corrosion, acid, and alkali, its purity and density are high and not conducive to the adhesion of electricity-

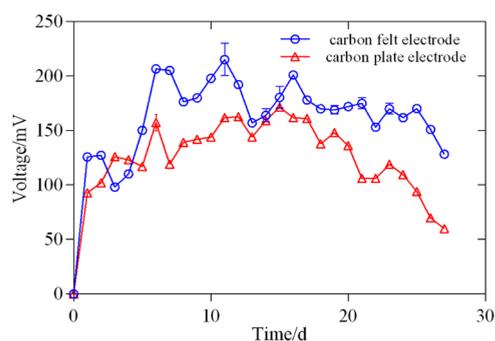


Figure 3. SMFC voltage–time curves of different anode materials.

generating bacteria.⁴² In contrast, carbon felt is disorderly arranged and has a loose texture and large surface pores, which are more conducive to the attachment of electricity-generating bacteria.⁴³ During the stable and declining periods of electricity production, the output voltage of the carbon felt electrode SMFC, up to 215.33 mV, is significantly higher than that of the carbon plate. It shows that compared to carbon plate electrodes, carbon felt electrodes are also more conducive to increasing the output voltage of oily sludge SMFCs.⁴³ Carbon felt is conducive to the adhesion of not only electricity-producing bacteria but also more petroleum-degrading bacteria, which improve the oily sludge degradation performance of SMFCs.⁴¹

It can be seen from Figure 4 that the maximum power density of the oily sludge SMFC using the carbon felt electrode is much higher than that with the carbon plate electrode, up to 81.49 mW/m³. Compared with the carbon plate electrode, the carbon felt electrode has better electricity generation performance. It can be seen from the SMFC polarization curves (Figure 4B) that the EMF of the carbon felt electrode SMFC (288.94 mV) is significantly higher than that of the carbon plate electrode SMFC (213.40 mV), and the apparent internal resistance (222.61 Ω) is much lower than that of the carbon plate electrode SMFC (310.54 Ω). This means that carbon felt has the lower mass transfer resistance, which can improve the electron transfer efficiency. It is because the fiber surface of carbon felt is relatively smooth, which is conducive to the diffusion of microbial metabolites.⁴⁴

3.2. Influence of the Anode Arrangement on the Performance of Oily Sludge SMFCs. **3.2.1. Influence of the Anode Arrangement on the Degradation Performance of Oily Sludge SMFCs.** The crude oil removal rate of the SMFC with horizontally arranged anodes (23.61%) is obviously higher than that of the SMFC with longitudinally arranged ones (19.82%), as shown in Figure 5. It may be because the horizontal arrangement of anodes is more conducive to the electrochemical activity of microorganisms,⁴⁵ and the better the electrochemical activity of the microorganisms, the more thoroughly the crude oil in the oily sludge is degraded. Thereby, the horizontal arrangement improves the crude oil removal rate of SMFCs.

3.2.2. Influence of the Anode Arrangement on the Electricity Generation Performance of SMFCs. In the SMFC voltage–time curve of different anode arrangements (Table 1), the output voltage of the SMFC with the transverse anode arrangement is higher than that with the longitudinal anode arrangement. It indicates that the power generation performance of the SMFC with the transverse anode

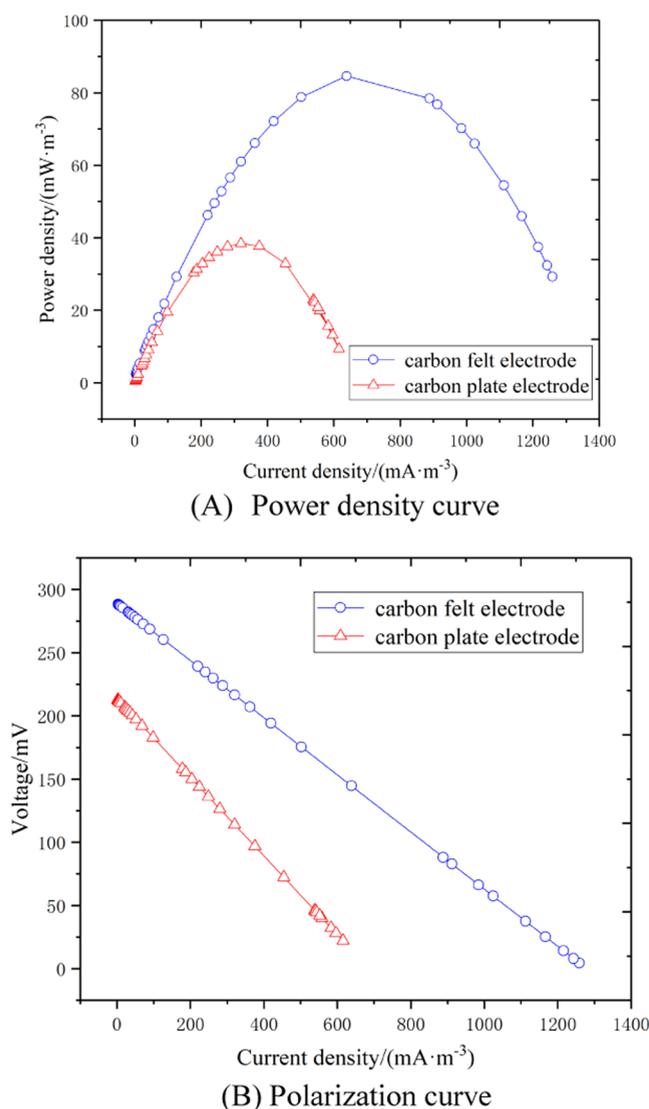


Figure 4. SMFC power density and polarization curves of different electrode materials.

arrangement is better than that of the SMFC with the longitudinal anode arrangement.

The power density curves of the SMFCs with different anode arrangements are shown in Figure 6A. The maximum power densities of the SMFCs with horizontally and longitudinally arranged anodes are 104.49 and 33.00 mW/m³, respectively. The EMF values of the SMFCs with horizontally and longitudinally arranged anodes are 429.29 mV and 324.46 mV, and the apparent internal resistance is 467.75 and 727.97 Ω , respectively. The maximum power density and EMF of the SMFC with horizontally arranged anodes are higher than those with longitudinally arranged anodes, and the apparent internal resistance is lower than that with longitudinally arranged anodes. It further shows that the power generation performance of the SMFC with horizontally arranged anodes is better than that with the vertically arranged anodes.

3.3. Influence of Multiple Anodes on the Performance of Oily Sludge SMFCs. 3.3.1. *Influence of Multiple Anodes on the Degradation Performance of Oily Sludge SMFCs.* After 30 days of oily sludge SMFC treatment, the effect of multiple anodes on the crude oil removal rate in the oily

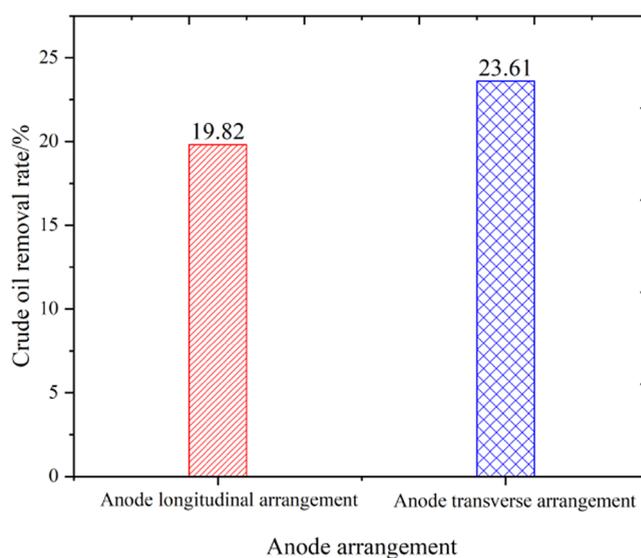


Figure 5. SMFC removal effect of crude oil with different anode arrangements.

Table 1. Voltage of SMFCs with Transverse and Longitudinal Anode Arrangements in Different Times

arrangement	transverse	longitudinal
1	188.6 ± 0.2	181.1 ± 0.5
2	203.7 ± 0.4	171.1 ± 0.2
3	206.1 ± 0.6	179.8 ± 0.7
4	226 ± 0.3	171.6 ± 1.2
5	216.3 ± 1.0	173.2 ± 0.8
6	216.5 ± 0.9	186.8 ± 1.1
7	213.5 ± 0.8	177.8 ± 0.3
8	226.3 ± 0.4	191.9 ± 0.5
9	217.7 ± 0.2	180.1 ± 0.2
10	234.6 ± 0.3	208.2 ± 0.3
11	265.2 ± 1.2	197.8 ± 1.0
12	282.9 ± 0.2	191.4 ± 0.4
13	286 ± 0.5	194.3 ± 0.6
14	285.1 ± 0.5	198.6 ± 0.7
15	295.4 ± 0.7	217.3 ± 0.5
16	290.9 ± 1.5	231.5 ± 1.4
17	282.1 ± 0.6	265.8 ± 0.6
18	261.6 ± 0.8	261.4 ± 0.7
19	263.2 ± 1.0	249.7 ± 0.8
20	249.5 ± 1.7	250 ± 1.5
21	291.9 ± 1.8	269.2 ± 1.4

sludge SMFCs is shown in Figure 7. With the increase of the number of anodes, the crude oil removal rate of the anode substrate increases. With sextuple anodes, the oily sludge SMFC anode substrate has the highest crude oil removal rate (33.72%). Therefore, the degradation performance of the SMFC is best with sextuple anodes.

3.3.2. Influence of Multiple Anodes on the Power Generation Performance of Oily Sludge SMFCs. As shown in Table 2, with the increase of the number anodes, the output voltage increases, and the output voltage of the sextuple anode is the highest, up to 408.8 mV. It is because the anode of the SMFC is the hub for the accumulation of electricity-generating

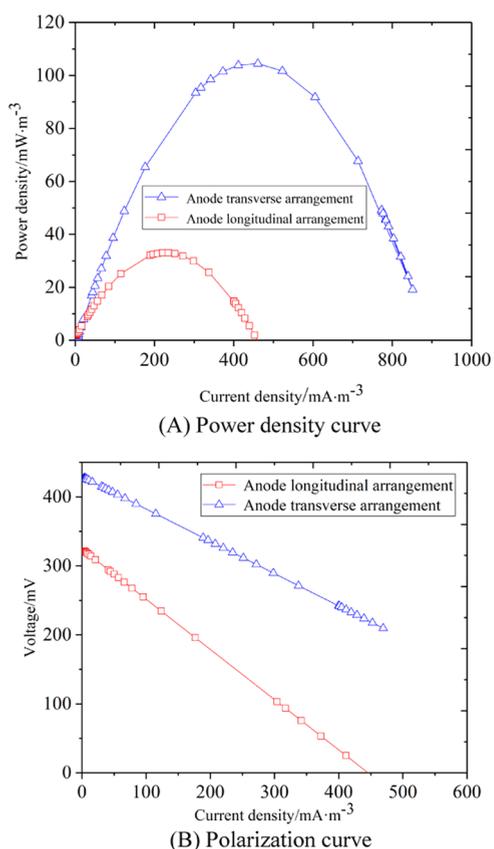


Figure 6. SMFC power density and polarization curves with different anode arrangements.

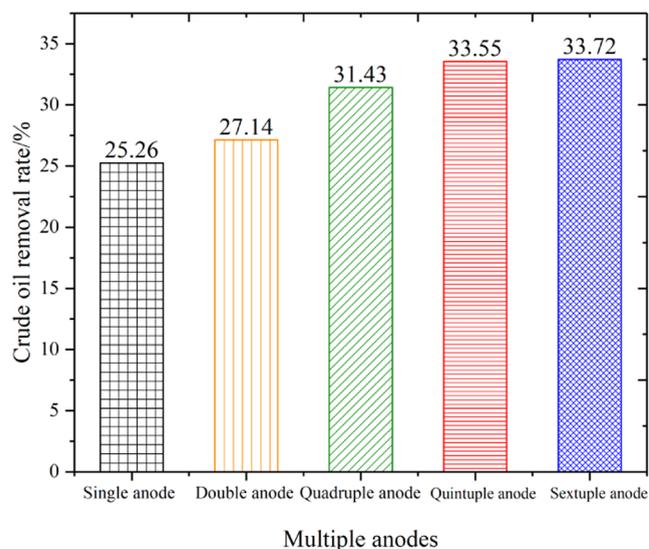


Figure 7. SMFC crude oil removal effect of multiple anodes.

microorganisms and electron transfer, which plays a vital role in improving the electricity-generating performance of the SMFC. Increasing the anode number will make more oily sludge participate in the battery reaction. In this process, more microorganisms are concentrated on the anode surface, producing more electrons and increasing the output voltage of the SMFC.⁴⁶ Increasing the anode number can increase the output voltage of the SMFC, but it does not mean that the output voltage will always increase with the anode number

increase. When the anode number increases excessively, the adhesion of electricity-generating microorganisms on the anode is insufficient, and the increase in the amount of adhesion is not large or small, resulting in an insignificant or low output voltage. It can be seen from the figure that when the anode is increased from quintuple and sextuple, the output voltage increase is not obvious, and the sextuple anode is the most suitable value for this experiment.⁴⁷

From the power density (Figure 8A) and polarization (Figure 8B) curves of the SMFC with different anode multiples, it can be seen that as the anode multiple increases, the power density and EMF of the SMFC gradually increase, and the apparent internal resistance gradually decreases. When the number of anodes is six, the power density and electromotive force are the largest, up to 145.32 mW/m³ and 516.39 mV, respectively, and the apparent internal resistance is the smallest, 417.08 Ω. The internal resistance is similar to the external resistance (500 Ω), indicating that the external resistance is reasonable.³³ Therefore, the power generation performance of the SMFC with sextuple anodes is the best. It shows that with appropriately increasing the number of anodes, the contact range of the anode oily sludge and the anode in the SMFCs increases. It helps more electricity-producing microorganisms adhere to the surface of the anode and reduce the mass transfer resistance between oily sludge and the anode.⁴⁸

It shows that when the crude oil removal rate of SMFCs is small, the output voltage and power density are also small. After adjusting the anode structure of oily sludge SMFCs, the oil removal rate increases, and the power production also increases gradually. The most proper adjustment of anode arrangements is using carbon felt as an anode material and increasing the number of anodes to six, and the highest crude oil removal rate reaches 33.72%; meanwhile, the output voltage of the oily sludge SMFCs is the highest, up to 408.82 mV, and the power density is also the highest (145.32 mW/m³).

The output voltage and the power density of oily sludge SMFCs increase with the crude oil removal rate, indicating that the electricity generation of SMFCs and the crude oil removal rate are in the same direction, that is, the degradation of crude oil by SMFCs can be efficiently converted into electricity. It is because the arrangement of the anodes is more conducive to the electrochemical activity of microorganisms; the better the electrochemical activity of the microorganisms, the more completely the crude oil in the oily sludge can be degraded, thereby increasing the crude oil removal rate of the SMFCs, and the better the power generation performance of the system.^{49–51}

4. CONCLUSIONS

Oily sludge SMFCs can effectively degrade crude oil and convert it into electricity. The anode structure will affect the oil removal and electricity generation performances of oily sludge SMFCs. The research results are as follows:

- The crude oil removal rate (17.04%) of the oily sludge SMFCs using carbon felt as the electrode material was better than that of the carbon plate and so were production voltage (215.33 mV), power density (81.49 mW/m³), and electromotive force (288.94 mV).
- The crude oil removal rate (23.61%) of the SMFCs with horizontally arranged anodes was better than that of the SMFCs with vertically arranged anodes and so were

Table 2. Voltage of Multianode SMFCs in Different Times

multianode voltage/mV time/day	sextuple	quintuple	quadruple	double	single
1	227.4 ± 0.4	221.9 ± 0.2	188.6 ± 0.2	177.4 ± 0.4	129.3 ± 0.2
2	235.5 ± 0.1	266.7 ± 0.1	203.7 ± 0.4	166.8 ± 0.5	140.9 ± 0.6
3	254.5 ± 0.5	252.2 ± 0.4	206.1 ± 0.4	189 ± 0.4	141.1 ± 0.4
4	303.5 ± 0.3	291.1 ± 0.4	226 ± 0.2	195.3 ± 0.6	155.2 ± 0.2
5	302.7 ± 0.2	308.3 ± 0.3	216.3 ± 0.4	173.7 ± 0.7	142.1 ± 0.5
6	318.7 ± 1.1	331.2 ± 1.2	216.5 ± 0.2	198.5 ± 0.5	172.1 ± 0.6
7	322.5 ± 1.2	309.1 ± 1.0	213.5 ± 0.4	194.9 ± 0.4	190.4 ± 0.4
8	318.5 ± 0.1	305.1 ± 0.2	226.3 ± 0.4	203.5 ± 0.4	171.7 ± 0.4
9	333.2 ± 0.2	302 ± 0.5	217.7 ± 0.3	208.3 ± 1.2	195.5 ± 1.0
10	308.4 ± 0.3	320.6 ± 0.4	234.6 ± 0.4	202.3 ± 0.2	179.2 ± 0.5
11	346.6 ± 0.4	349.6 ± 0.7	265.2 ± 0.8	202.2 ± 0.4	193.3 ± 0.9
12	373.1 ± 1.2	362.7 ± 0.5	282.9 ± 0.6	214.6 ± 0.1	197.2 ± 0.8
13	378.6 ± 0.4	367.5 ± 0.2	286 ± 0.3	222.2 ± 0.4	180 ± 0.7
14	386.1 ± 0.6	375.8 ± 0.4	285.1 ± 0.1	218 ± 0.3	179.8 ± 0.4
15	386.5 ± 0.2	376.4 ± 0.3	315.4 ± 1.1	223.6 ± 0.2	190.9 ± 0.5
16	408.8 ± 0.7	382.9 ± 0.4	320.9 ± 0.2	231 ± 0.3	198.1 ± 0.4
17	403.4 ± 0.9	400.9 ± 0.5	282.1 ± 0.8	225.3 ± 0.8	213.7 ± 0.3
18	394 ± 1.2	384 ± 1.1	261.6 ± 0.6	239.2 ± 0.9	225.2 ± 0.2
19	379.9 ± 1.3	381 ± 0.3	263.2 ± 0.5	233.9 ± 0.1	239.8 ± 0.5
20	395.3 ± 1.5	370.2 ± 0.2	249.5 ± 0.4	256.3 ± 0.2	230.9 ± 0.4
21	387.1 ± 0.4	385.8 ± 0.4	321.9 ± 0.2	267.8 ± 0.1	250.4 ± 0.3

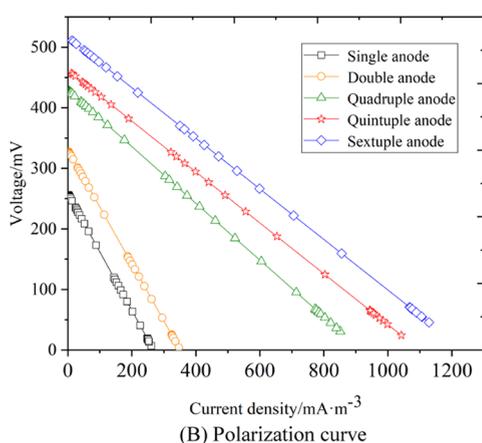
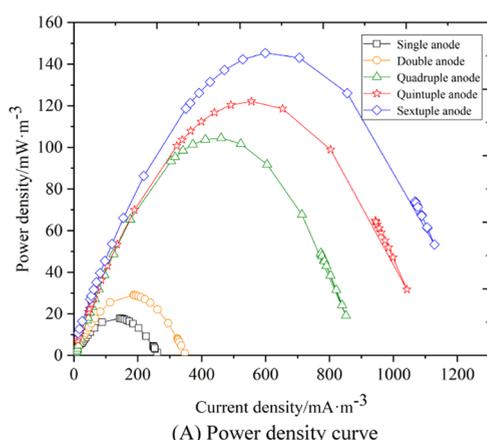


Figure 8. Power density and polarization curve of multianode SMFCs.

production voltage (290.40 mV), power density (104.49 mW/m³), electromotive force (429.29 mV).

(c) When using multiple anodes as electrodes, with the increase of the number of anodes, the voltage, power density, electromotive force, and crude oil removal of SMFCs first increase and then tend to be stable. The maximum voltage, power density, electromotive force, and crude oil removal rate were 408.82 mV, 145.32 mW/m³, 516.39 mV, and 33.72%, respectively.

It can be seen that the proper adjustment of the anode structure can well improve the oil removal and power generation performances of oily sludge deposition SMFCs, which will become an effective strategy to improve the performance of oily sludge SMFCs.

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Notes

The authors declare no competing financial interest.

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