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Power generation using an activated carbon fiber felt cathode in an upflow microbial fuel cell

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ABSTRACT

An activated carbon fiber felt (ACFF) cathode lacking metal catalysts is used in an upflow microbial fuel cell (UMFC). The maximum power density with the ACFF cathode is 315 mW m^{-2}, compared to lower values with cathodes made of plain carbon paper (67 mW m^{-2}), carbon felt (77 mW m^{-2}), or platinum-coated carbon paper (124 mW m^{-2}, 0.2 mg-Pt cm^{-2}). The addition of platinum to the ACFF cathode (0.2 mg-Pt cm^{-2}) increases the maximum power density to 391 mW m^{-2}. Power production is further increased to 784 mW m^{-2} by increasing the cathode surface area and shaping it into a tubular form. With ACFF cutting into granules, the maximum power is 481 mW m^{-2} (0.5 cm granules), and 667 mW m^{-2} (1.0 cm granules). These results show that ACFF cathodes lacking metal catalysts can be used to substantially increase power production in UMFC compared to traditional materials lacking a precious metal catalyst.

1. Introduction

A microbial fuel cell (MFC) is a promising new technology for generating electricity directly from biodegradable compounds using bacteria [1–7]. The MFC consists of an anode, a cathode, and sometimes a membrane or separator between the electrodes. Various types of materials have been used as the anode, including plain graphite [8], carbon cloth [9], graphite foam [10], graphite granules [11], reticulated vitreous carbon (RVC) [12], and high surface area graphite fiber brush electrodes [13,14]. Common separators between the electrodes include cation exchange membranes (CEMs) [15–18], anion exchange membranes (AEMs) [17] and ultrafiltration membranes [17]. A separator can help to reduce oxygen transfer into the anode chamber, but can also increase ohmic resistance and produce pH gradients, and reduce power production.

The cathode is usually a limiting factor in power production in an MFC [14,19–21]. Various electron accepters have been used as catholytes in two-chamber MFCs, although chemicals such as ferricyanide and permanganate [8,16,22] are not practical or sustainable for long term operation. Using oxygen as the electron accepter has many advantages, including a high thermodynamic redox potential, good self-sustaining operation, and availability [23]. However, it is difficult to obtain high cathodic potentials unless metal catalysts are used. Platinum catalyzes oxygen reduction in MFCs, even at loading rates as low as 0.1 mg cm^{-2} [24]. Alternatives to this precious metal catalyst include non-precious metals linked with organics, such as co-tetramethylphenylporphyrin (CoTMPP) and iron (II) phthalocyanine (FePc) [24,25]. These non-precious metal catalysts have excellent performance, although higher loading rates than those used for Pt are needed to achieve similar performance [26]. In addition, metal catalysts can be poisoned by certain chemical species in the water such as sulfides [26]. Biocathodes have been developed based on enrichment of bacteria for oxygen [27] or nitrate reduction [28,29], photosynthesis [30,31], or enzyme production [32]. It is possible to increase the cathode efficiency using materials with high surface areas [14] or using granular materials such as graphite [26]. For example, Freguia et al. [26] achieved a maximum power density of 21 W m^{-3} using a two-chamber MFC with dissolved oxygen as the electrode acceptor in the cathode chamber. For non-catalyst MFC systems, the biggest challenge is to find an effective method to reduce cathode polarization losses.

Activated carbon fiber felt (ACFF) is a high surface area material (900–2500 m^2 g^{-1}) that has been used as an anode [33], but it has not been previously examined for using as a cathode. In addition to high surface area, it has excellent adsorption capabilities and an open pore structure [34,35]. Furthermore, due to its wide application in wastewater treatment [35] and air purifi-

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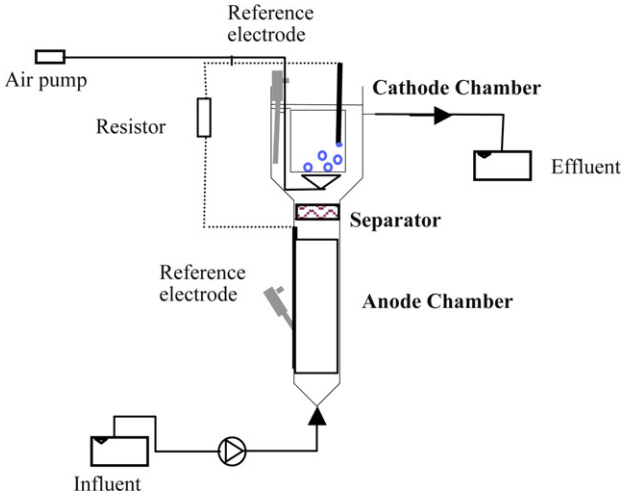

Fig. 1. Schematic construction of the upflow microbial fuel cell (UMFC).

cation, ACFF is relatively inexpensive. We examined here the use of ACFF in various configurations (tubular and granular) in an upflow MFC (UMFC), a type of reactor that has been previously examined due to its simplicity in construction and operation [11,12,36]. In this study, power production with plain and Pt-coated ACFF was compared to several commonly used materials, including plain carbon paper, plain carbon felt and Pt-carbon paper.

2. Methods and materials

2.1. MFC construction

The UMFC consisted of two cylindrical Plexiglas tubes having different diameters and heights (Fig. 1). The cathode chamber (9 cm diameter, 8 cm height, 540 mL total volume) was located on top of the anode chamber (4.5 cm diameter, 25 cm height, 430 mL total volume). These two chambers were separated by a PVC tube (12 cm^2 cross-sectional area, 1.5 cm height, Huawei Co., Beijing, China) filled with granular porous sponges (0.5 cm diameter, Huawei Co., Beijing, China). Nylon netting was used to contain the separator material (0.01 cm pore diameter, Huawei Co., Beijing, China), allowing fluid to pass from the anode to cathode chamber. Ag/AgCl reference electrodes (Luosu Co., Shanghai, China) were inserted into each chamber for electrode potential measurements.

2.2. Electrode materials

The anode was a single piece of carbon felt (5 cm thick, 26.5 cm × 16.5 cm in size, projected surface area of 317 cm^2, Beijing, China) that was drilled with ~100 holes (0.5 cm diameter) to increase surface area. The carbon felt was rolled into a tubular anode and placed vertically in the anode chamber so that the flow could pass around and through it. Carbon thread was used to connect the anode to the circuit.

The cathode was a single piece of ACFF (5 cm thick, Senxin Co., Liaoning province, China) with a geometric surface area of 72 cm^2 for both sides of the electrode (36 cm^2 each), except as noted. The specific surface area for this material was 1000 $m^2\ g^{-1}$ (provided by the manufacturer). The cathode was connected to the circuit by a graphite rod (1 cm diameter and 15 cm long, Sanye Co., Beijing, China). The ACFF was compared to three other materials of the same geometric size, consisting of: plain carbon paper (0.2 cm thick, 0.3 $m^2\ g^{-1}$ specific surface area, Lvneng Co., Beijing, China), Pt-coated carbon paper (0.2 mg-Pt cm^{-2}, prepared by the manufacturer), and carbon felt (5 cm thick, 67 $m^2\ g^{-1}$ specific surface area, Sanye Co., Beijing, China). In some experiments, a Pt-coated (0.2 mg cm^{-2}) ACFF cathode with the same geometric surface area of 72 cm^2 was used.

For Pt-containing cathodes, a commercial Pt catalyst (40 wt%Pt/C, J-M Co., Britain) was ultrasonically mixed with Nafion solution (5%, DuPont Co., USA) and isopropanol (analytically pure, Beijing, China) to form a paste. The paste was evenly applied to two sides of the cathodes (carbon paper or ACFF), and dried at 125 °C for 1 h under Argon gas.

Additional tests were performed to assess the power production of ACFF cathodes with higher surface areas and three different shapes. A large piece of ACFF (52.5 cm × 8.5 cm) was rolled into a tubular shape (as done with the anode) to produce a total pro-

Table 1
Summary of performance parameters of different cathode materials.

Cathode material	ACFF	Carbon felt	Plain carbon paper	Pt-coated carbon paper	Tubular ACFF	ACFF granules (0.5 cm)	ACFF granules (1.0 cm)	Graphite granules Ref. [26]
Geometric surface area (m^2)	7.2×10^{-3}	7.2×10^{-3}	7.2×10^{-3}	7.2×10^{-3}	8.9×10^{-2}	[d]	[d]	[e]
Weight (g)	0.822	1.110	0.614	0.614	10.987	44.4	25.7	[e]
Specific surface area ($m^2\ g^{-1}$)[a]	1000	60.5	0.3	0.3	1000	1000	1000	[e]
Surface area (m^2)	822	67.3	0.184	0.184	1.10×10^4	4.44×10^4	2.57×10^4	1.26
Cathode chamber volume (CV) (m^3)	5.4×10^{-4}	5.4×10^{-4}	5.4×10^{-4}	5.4×10^{-4}	5.4×10^{-4}	5.4×10^{-4}	5.4×10^{-4}	3.5×10^{-4}
Surface area normalized by CV ($m^2\ m^{-3}$)	1.52×10^6	1.25×10^5	3.40×10^2	3.40×10^2	2.0×10^7	8.2×10^7	4.7×10^7	3.6×10^3
Power density at MPP[b] ($mW\ m^{-2}$)	315	77	67	124	784	481	667	[e]
Power to CV[c] at MPP[b] ($W\ m^{-3}$)	0.7	0.2	0.1	0.3	1.7	1.1	1.5	21
Current to cathode surface area at MPP[a] ($mA\ m^{-2}$)	1.67×10^{-3}	6.0×10^{-3}	1.5	4.69	[d]	[d]	[d]	17
Open circuit cathode potential (mV)	397	312	278	454	422	287	402	420–550[f]
Cathode resistance (Ω)	53	183	283	161	25	12	7	<9
Total resistance of reactor (Ω)	[d]	[d]	[d]	[d]	133	116	115	9[g]
Power to cathode weight at MPP[b] ($\times 10^{-5}\ W\ g^{-1}$)	[d]	[d]	[d]	[d]	8.6	1.3	3.1	[e]
Price of cathode material ($\$\ kg^{-1}$)	40	85	41	990	40	40	40	1.8[26]
Cathode material cost[h] per unit of energy ($\$\ W^{-1}$) at MPP	94	940	500	4100	520	3200	1400	79[26]

[a] Provided by manufacturer.
[b] Maximum power point.
[c] Cathode chamber volume.
[d] Not measured.
[e] Value not reported by the authors and could not be calculated from other data.
[f] pH = 6–9 in Fig. 1 of Ref. [26].
[g] Calculating the slope of polarization curves(pH = 6) in Fig. 1 of Ref. [26].
[h] The price is provided by manufacturer in 2009.

jected surface area of 446 cm^2 (2.0×10^7 m^2 m^{-3} based on the specific surface area of material per volume of the cathode chamber (Table 1)). This tubular ACFF was fixed to a single graphite rod (1 cm diameter and 15 cm long, Sanye Co., Beijing, China) placed vertically in the cathode chamber. ACFF was also tested in a granular form, which was obtained by cutting up pieces of felt into particles 0.5 cm × 0.5 cm (1948 cm^2 projected surface area) or 1 cm × 1 cm squares (1127 cm^2), each with a 0.5 cm thickness and weight specific surface area of 1000 m^2 g^{-1}. The filled volume (cathode volume) for each shape of the ACFF was 540 mL, corresponding to 8.2×10^7 m^2 m^{-3} (0.5 cm granules) and 4.7×10^7 m^2 m^{-3} (1.0 cm granules) based on the specific surface area of the material and the cathode chamber volume. The ACFF granules were packed into the cathode chamber, and were connected to the circuit using four graphite rods (diameter 1 cm and length 15 cm) inserted into the packed bed of granules.

2.3. Inoculation and operation

Anaerobic sludge was collected from a sludge digester at Gaobeidian Wastewater Treatment Plant in Beijing, China. The sludge was washed three times with distilled water. This seed sludge was cultivated in the anodic medium solution containing glucose (200 mg COD L^{-1}), NH_4Cl, and NaH_2PO_4 with a weight ratio of 100:5:1 (COD:N:P) for 1 week at 35 °C, and then the sludge suspension was added to the UMFC as the inoculum.

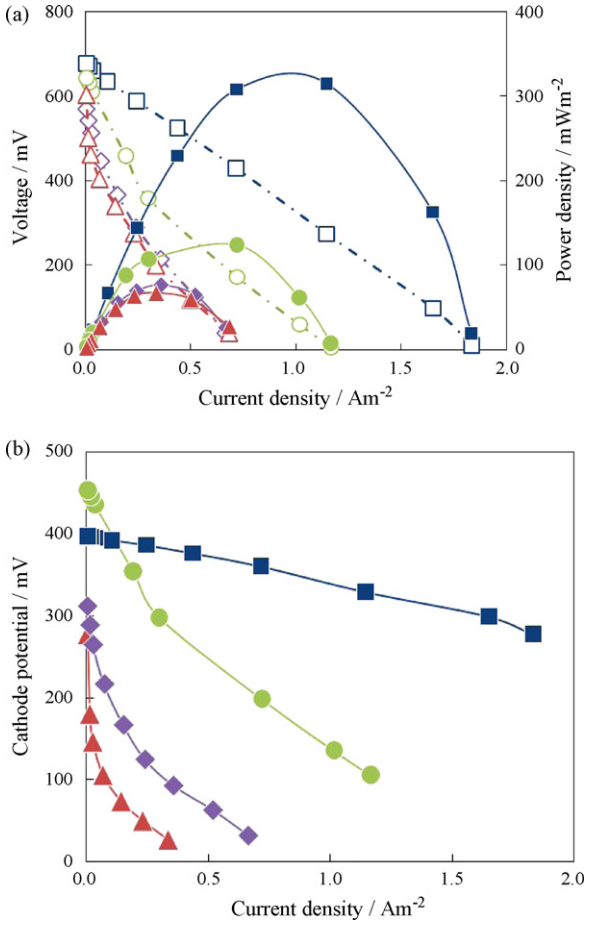

Fig. 2. (a) Polarization (dashed line) and power density (solid line) curves of MFC with non-catalyst carbon paper (triangles), carbon felt (diamonds), Pt-coated carbon paper (circles) and ACFF (squares). (b) Cathode polarization curves of non-catalyst carbon paper (triangles), carbon felt (diamonds), Pt-coated carbon paper (circles) and ACFF (squares).

The UMFC was fed a synthetic medium containing 187 mg L^{-1} (200 mg COD L^{-1}) glucose, 38.2 mg L^{-1} NH_4Cl, 7.7 mg L^{-1} NaH_2PO_4 and 986 mg L^{-1} NaCl (final solution conductivity of 2.3 mS cm^{-1}). In tests with different ACFF configurations, the NaCl concentration was increased to 4.1×10^3 mg L^{-1} to increase solution conductivity (6.6 mS cm^{-1}). No trace metals or buffers were added. Substrate was pumped into the bottom of anode chamber using a peristaltic pump (Lange Co., Hebei province, China) as shown in Fig. 1. The flow rate was fixed at 2.0 L d^{-1} to keep a constant HRT of 0.5 d. The cathode chamber was continuously aerated using an aeration stone placed in the bottom of cathode chamber, resulting in a dissolved oxygen concentration of 5–6 mg L^{-1}. All experiments were conducted in duplicate in a temperature-controlled room (35 °C).

2.4. Calculations and analyses

Cell voltages (V) across a resistor (R) and anode/cathode potentials versus Ag/AgCl reference electrode were measured using a data acquisition system (Reibohua Co., Beijing, China). The polarization and power density curves as a function of current were obtained by changing external circuit resistances. For each test, data for one resistor were collected after at least 2 h of stable voltage was maintained.

The anode resistance R_a, cathode resistance R_c and internal resistance R_{int} of the MFCs were calculated using the polarization slope method [37], applied to the linear portion of the current (I) versus voltage (V) plot. Power was calculated from voltage and current as $P = IV$, with the power density normalized to the cross-sectional area of the separator, and volumetric power density normalized using either the total volume of cathode chamber

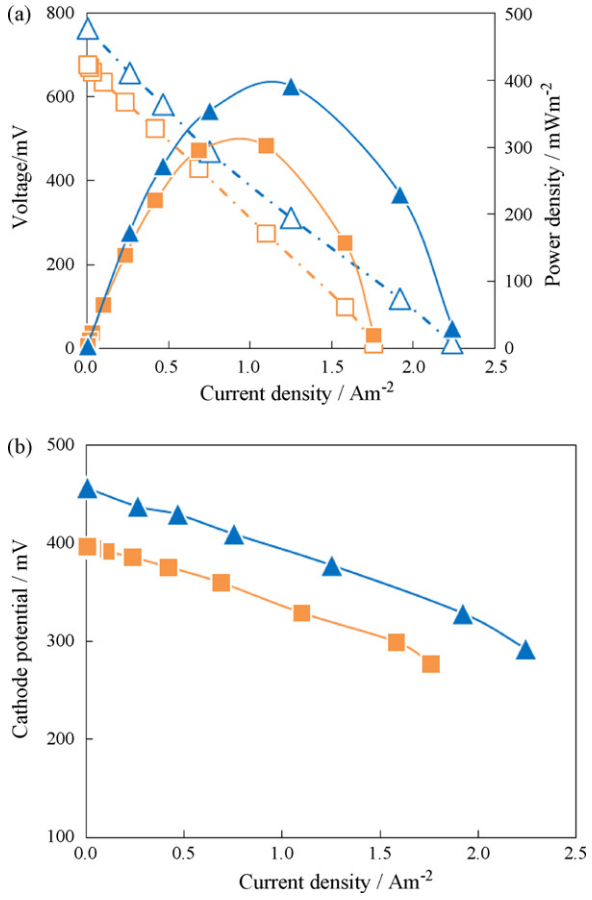

Fig. 3. (a) Polarization (dashed line) and power density (solid line) curves of MFC using non-catalyst (squares) and Pt-coated (triangles) ACFF.

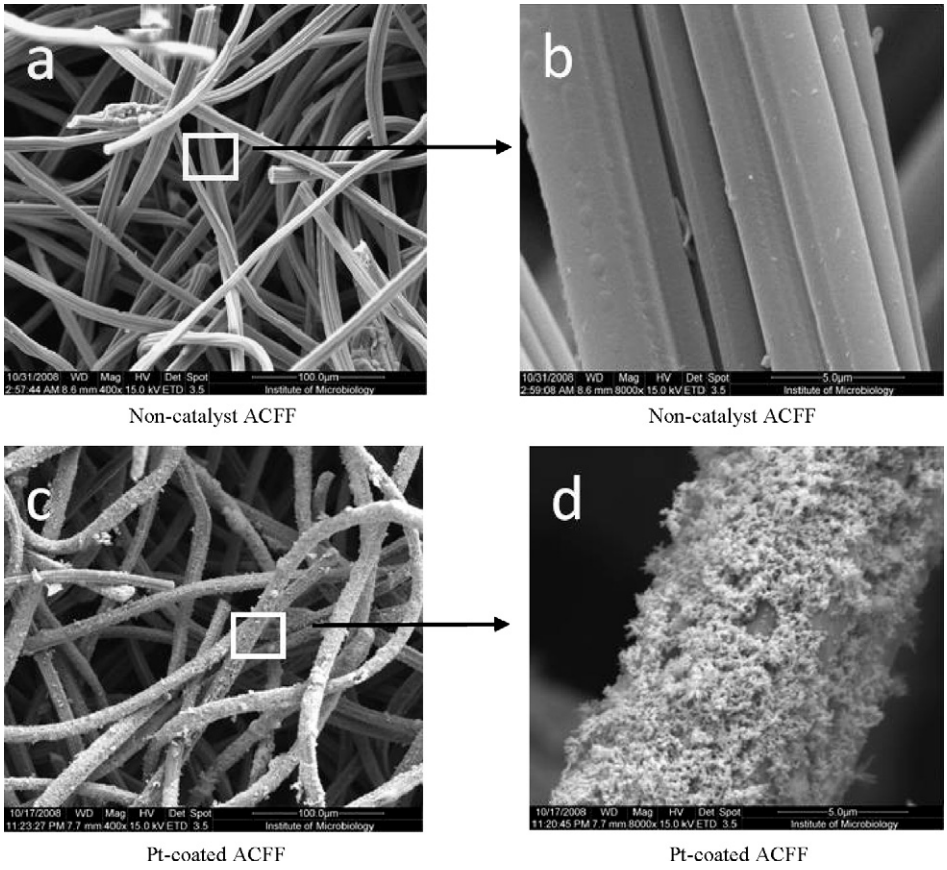

Fig. 4. Surface morphologies of non-catalyst (a and b) and Pt-coated ACFF (c and d) cathodes taken by scanning electron microscope (SEM).

(CV). Data were recorded on a personal computer at 10 min intervals.

Surface morphologies of plain and Pt-coated ACFF cathodes were observed using scanning electron microscopy (SEM) (FEI Co., Hongkong, China). The surface concentrations of metals on an ACFF cathode were determined with inductively coupled plasma-mass spectrometer (ICP-MS; VG Co., Britain).

3. Results

3.1. Comparison of ACFF with other cathode materials

The maximum power density obtained in tests with the plain ACFF cathode UMFC was 315 mW m^{-2} (0.7 W m^{-3} based on cathode volume) (Fig. 2a). This power density was 3.7 times larger than that with the same size carbon paper cathode (67 mW m^{-2}, 0.1 W m^{-3}) and 3.0 times greater than that with carbon felt (77 mW m^{-2}, 0.2 W m^{-3}). Adding a Pt catalyst to the carbon paper increased power density to 124 mW m^{-2} (0.3 W m^{-3}), but this was only 39% of that achieved with the ACFF in this cathode configuration. Cathode polarization data confirmed that the reason for the better performance with the ACFF material was cathode performance (Fig. 2b). The ACFF cathode had a higher potential at current densities >0.1 A m^{-2} (Fig. 2b). The carbon paper with Pt had the highest open circuit cathode potential OCP (454 mV), with the ACFF cathode slightly lower at an OCP of 397 mV. The OCPs for the other materials were much lower, 278 mV for the plain carbon paper and 312 mV for the carbon felt. Based on the slopes of the cathode polarization data, the resistances of the different materials were: 53 Ω for ACFF, 161 Ω for carbon paper with Pt, 183 Ω for the carbon felt, and 283 Ω for the carbon paper. It is therefore clear that improved performance can be credited to the lower cathodic resistance of the ACFF.

In order to determine if any trace metals on the ACFF cathode contributed to the cathodic reaction, the concentration of metals on the ACFF was examined. Fe had the highest surface concentration of 4×10^{-6} mg m^{-2}, followed by Mo (7×10^{-7} mg m^{-2}), Ni (5×10^{-7} mg m^{-2}), and V (4×10^{-7} mg m^{-2}). In order for a catalyst to be useful for oxygen reduction, the surface concentration needs to be at least 1000 mg m^{-2} [26]. Therefore, the surface concentration of metals on ACFF was not high enough to contribute to the oxygen reduction reaction. Based on SEM pictures (not shown), there was no biofilm formation on ACFF surface, indicating that the performance of the ACFF was not due to biocatalyzed oxygen reduction. These results demonstrate that the performance of ACFF cathode can be mainly attributed to its high specific surface area and carbon-based composition, not to the presence of metallic or biological catalysis.

3.2. Performance of the ACFF with a Pt catalyst

Adding Pt catalyst to the ACFF material increased power by 24% to 392 mW m^{-2} (Fig. 3a). This increase was much smaller than that obtained when the Pt was added to the carbon paper (85%). As shown in Fig. 3b, Pt-coated ACFF resulted in an increase (40–70 mV) in the cathode potential produced for current densities of up to 1.1 A m^{-2}. The cathode resistance of the plain ACFF was 53 Ω, which was nearly the same as that of Pt–coated ACFF (55 Ω) due to the high specific surface area of ACFF. Thus, addition of the Pt catalyst did not have an appreciable affect on the resistance of the ACFF.

SEM analysis indicated that the Pt catalyst was evenly coated on the surface of the ACFF (Fig. 4). Pt particles reduce the activation energy of the cathodic reaction, which results in a higher cathode

potential than non-catalyst ACFF. Due to the high specific surface area of the ACFF (900–2500 $m^2\ g^{-1}$), the cathode resistance of plain ACFF was low. Therefore, when the Pt catalyst was added, the cathode resistance did not substantially change (53 Ω for plain ACFF, and 55 Ω for Pt-coated ACFF). Consequently, these results demonstrate that ACFF is an excellent cathode material primarily due to its high specific surface, and that Pt does not significantly improve performance.

3.3. Effect of ACFF configurations on UMFC performance

In order to further increase power production, tests were conducted using ACFF with larger total surface areas and three different shapes. Using a large piece of ACFF rolled into a tubular shape the maximum power density was increased to 784 mW m^{-2} (1.7 W m^{-3}). This increase in power density was due to the increase in the total surface area of the cathode ($2.0 \times 10^7\ m^2\ m^{-3}$ based on specific surface area per volume of the cathode chamber) compared to that obtained with the ACFF in the original configuration ($1.5 \times 10^6\ m^2\ m^{-3}$). As a result of this tubular configuration, the cathode resistance decreased to 25 Ω (Fig. 5b).

The larger ACFF granules (1 cm squares) produced a slightly lower maximum power density of 667 mW m^{-2} (1.5 W m^{-3}) than that obtained with the tubular ACFF cathode (Fig. 5). A decrease in the average size of the granules (0.5 cm squares) resulted in an even lower power density (481 mW m^{-2}, 1.1 W m^{-3}) (Fig. 5), even though the total surface area of these smaller granules ($4.4 \times 10^4\ m^2$) was larger than that of the 1.0 cm granules ($2.5 \times 10^4\ m^2$). Similar cathode potentials were obtained over the range of current densities for both the tubular ACFF and the 1.0 cm granules, but the 0.5 cm granules produced lower voltages (Fig. 5b). The resistances of the granules (5–15 Ω) were lower than those obtained for the tubular ACFF (25 Ω) based on cathode polarization data (Fig. 5b). Because of the higher power density, the use of the ACFF in tubular form is preferred to granules form for maximizing power in the UMFC.

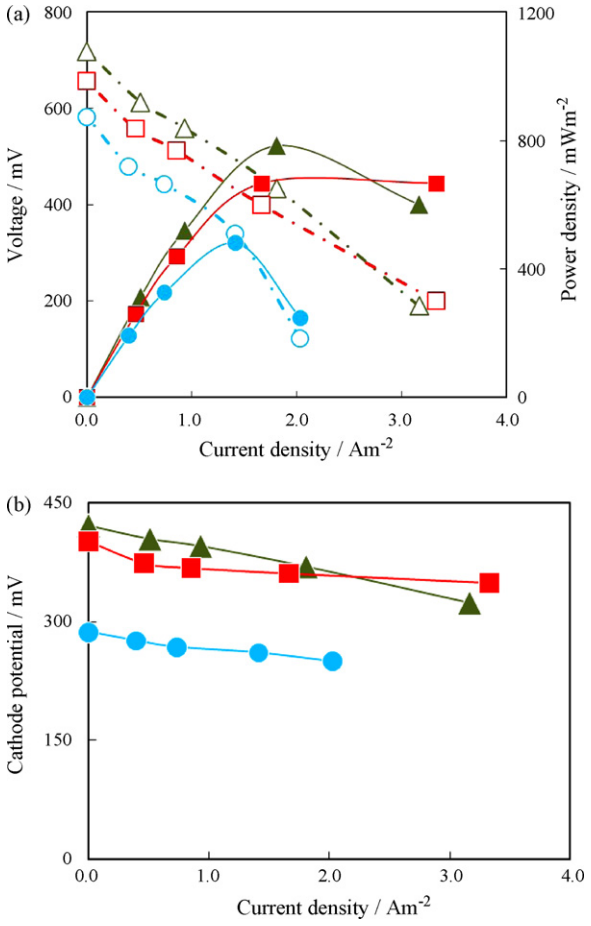

Fig. 5. (a) Polarization (dashed line) and power density (solid line) curves of MFC with three shapes of ACFF. Tubular type (triangles), 0.5 cm granules (circles) and 1.0 cm granules (squares). (b) Cathode polarization curves of three shapes of ACFF. Tubular shape (triangles), 0.5 cm granules (circles) and 1.0 cm granules (squares).

Power densities are usually normalized by the liquid or total volume of the cathode or reactor. In order to examine the most effective use of a material, we also considered power production based on mass of the electrode material. On this basis, the maximum power densities of the three different ACFF shaped materials were 8.6×10^{-5} W g^{-1} for the tubular ACFF, 3.1×10^{-5} W g^{-1} for the 1.0 cm granules, and 1.3×10^{-5} W g^{-1} for the 0.5 cm granules (Table 1). Thus, the tubular ACFF produced the highest power density per mass of cathode material used in this study.

4. Discussion

The use of an ACFF sheet rolled into a tubular shape produced a maximum power density of 784 mW m^{-2} (1.7 W m^{-3}), compared to 667 mW m^{-2} (1.5 W m^{-3}) for the granular ACFF. To our knowledge, this is the first time that the use of activated carbon has been examined as the cathode in an MFC. Although ACFF was previously used in an MFC with a sulfate-rich wastewater, it was examined as an anode material and not as the cathode [33]. The ACFF had a lower cathode resistance and higher cathode potentials than other cathode materials examined here, including Pt-coated carbon paper, plain carbon paper, and carbon felt (Table 1).

The reason for the high performance of the ACFF material was primarily its high specific surface area. On the basis of projected surface area, the plain ACFF produced 105 mW m^{-2} compared to 41 mW m^{-2} for the Pt-coated carbon paper (both with 36 cm^2 projected surface area). Cathodic overpotential decreases with the current density (mA m^{-2}) [26]. The very high surface area of the material resulted in a low current density per area thus producing a low cathodic overpotential compared to other cathode materials (Table 1). As a result, the ACFF cathode performed better than the other cathode materials, including carbon paper with a Pt catalyst. Graphite granules are another material with a high specific surface area that has previously been used in MFC tests as anodes and cathodes [26]. The ACFF used in this study has a higher volumetric specific surface area ($4.6 \times 10^7\ m^2\ m^{-3}$) and a comparable cathode resistance (7 Ω, 1.0 cm granules) to graphite granules ($6 \times 10^6\ m^2\ m^{-3}$; and <9 Ω based on the total reported reactor resistance of 9 Ω) [26] (Table 1). These data shows that the high specific surface area of these carbon materials largely compensates for inherent poorer performance on a surface area basis relative to precious metal catalyzed surfaces.

The overall volumetric power densities achieved here with an upflow design could likely be improved. The open circuit potential was 580 to ~660 mV, which is comparable to 420–550 mV found in other studies [26] (Table 1). However, the total reactor resistance of 115 Ω is large compared with other studies, such as the granular cathode MFC (9 Ω) [26]. Therefore, it will be necessary to either improve the UMFC design, for example by reducing the distance between the anode and cathode, or use ACFF in MFCs with different configurations. Further studies to examine the performance of the ACFF material in improved MFC designs, such as using air-breathing cathode MFCs, are in progress.

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