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Municipal sludge-derived carbon anode with nitrogen- and oxygen-containing functional groups for high-performance microbial fuel cells

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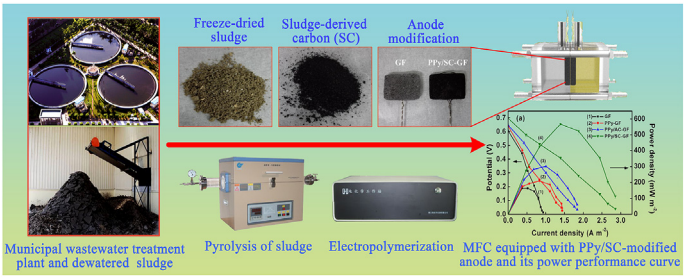
HIGHLIGHTS

- The municipal sludge derived-carbon (SC) is produced upon pyrolysis.
- The SC-modified anode is featured with increased surface area and surface N/C ratio.
- The SC-modified anode contains high amounts of O═C–O and O═C–NH_2 functional groups.
- The SC-modified anode significantly increased power performance of MFCs.

GRAPHICAL ABSTRACT



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ABSTRACT

The demand for efficient and cost-effective anode materials in microbial fuel cells (MFCs) provides the impetus to use carbon derived from solid waste to support bacterial growth and proliferation. Here we show that the municipal sludge-derived carbon (SC) with a porous structure and abundant surface functional groups is effective in improving performance of MFCs. The SC is coated on the 3-D graphite felt (GF) surface by pyrrole electropolymerization in order to increase the surface cites that are interacted with bacteria, resulting in the formation of PPy/SC-modified GF anode. The scanning electron microscopy analysis indicates that the PPy/SC-modified GF can substantially increase anode surface area. The X-ray photoelectron spectroscopy (XPS) results suggest that the PPy/SC-modified GF anode possesses higher surface N/C ratio and higher relative contents of O═C–NH_2 and O═C–O functional groups than other counterpart anodes. These characteristics are essential for increasing bacterial attachment to the anode surface, electron-transfer rate and thus anode performance and power performance. The maximum power density resulting from the PPy/SC-modified GF anode was 568.5 mW m^{-2} (13.6 W m^{-3}) increased by 1.9, 2.7 and 3.5 times as compared to the PPy/AC-modified GF anode, the PPy alone-modified GF anode and the unmodified GF anode, respectively.

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1. Introduction

Microbial fuel cells (MFCs) have attracted ever-growing and worldwide interests in sustainable power generation and wastewater treatment, due to their ability to extract energy from pollutants available in the wastewater by using bacteria as catalysts.

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The interaction between microbes and anode materials plays a fundamental role in anode performance and thus overall power output of MFCs. Substantial attention has been given to anode modifications that enhance surface area and functional groups of carbon-based anode materials, in order to improve bacterial adhesion and electron transfer from microbes to the anode [1,2]. The commonly used approach is surface functionalization via acid soaking [3–5], electrochemical oxidation [6–9] and heat ammonia treatment [10,11], as it increases nitrogen- and oxygen-functional groups that are beneficial to microbe-anode interaction. The increased interaction eventually results in a remarkable increase in power performance. For example, ammonia treatment enhanced the positively charged N-functional groups that favored the accumulation of microbes and increased the probability of facile and direct electron transfer from microbes to the anode, as reflected by 48% increment in the ammonia-treated anode compared to the untreated anode [11]. Electrochemical oxidation resulted in the generation of O-functional group such as carboxyl that facilitated the electronic coupling between the microbes and the anode, allowing the treated anode exhibited 40% increment in the current density [7]. Despite the efficiency in increasing power performance, these surface modification technologies are heavily relied on the use of exogenous chemicals and external energy, leading to additional costs.

The demand for effective and low-cost carbon-based anode materials abundant with surface functionalities encouraged some attempts to apply carbon derived from natural solid wastes in MFCs [12–15]. The municipal sludge, produced in huge quantity from wastewater treatment plants, is a hazardous solid waste but also a carbon-rich material. Recent studies have shown that the sludge-derived carbon (SC) produced upon pyrolysis is a versatile material used in a variety of applications such as adsorbents of a wide range of gaseous and liquid-phase pollutants [16–20], and catalysts for chemical/electrochemical reactions [17,21–23], owing to their well-defined porous structure and abundant surface functional groups. These inherent characteristics indicate that SC holds potential for application in MFCs as cost-effective anode materials.

The aims of this study are to demonstrate the concept of using SC as a high-performance MFC anode material and elucidate what are important nitrogen- and oxygen-containing functional groups contributing to the increased MFC performance. To increase the surface cites that are interacted with bacteria, the SC is grafted to the 3-D graphite felt (GF) substrate by utilizing the electropolymerization approach with pyrrole as the monomer. The performance of the MFC equipped with polypyrrole (PPy)/SC-modified GF anode was evaluated and compared to its counterparts with different anodes. The characterizations on the surface morphology, surface composition and surface functional groups were performed with the scanning electron microscopy (SEM) and X-ray photoelectron spectroscopy (XPS) technologies.

2. Experimental

2.1. Preparation of carbon using municipal sludge as precursor

The dehydration municipal sludge was obtained from the Datansha Wastewater Treatment Plant, Guangzhou, China. Prior to pyrolysis, the sludge was further dehydrated in a freezing dryer for 48 h, ground and sieved to 100 mesh. To prepare SC, the dried sludge was heated in a tubular furnace at 800 °C for 2 h in a N_2 atmosphere, following the procedures described previously [22]. The resulting SC was directly used and characterized as described below.

2.2. Electro-synthesized PPy/SC composites as anode material

The original anode, the 3.0 × 2.0 × 0.5 cm piece of GF provided by Beijing Sanye Co. Ltd, China was cleaned by rinsing in 10% H_2O_2 at 90 °C for 3 h, subsequently rinsed with deionized water and dried at 60 °C. A titanium 1 mm-wire was stuck into the GF as a contact for circuit connection. The SC taken as prepared was suspended in deionized water at a concentration of 5 g L^{-1}. The suspension was vigorously stirred at ambient temperature for 10 min and ultrasonicated for 5 min. Subsequently, pyrrole monomer (0.2 M) was added to the suspension.

Pyrrole electropolymerization was executed in the solution containing no other supporting electrolyte except SC aiding as a weak electrolyte. The operation at ambient temperature was carried out in an electrochemical cell with three electrodes: a working electrode (GF), a reference (saturated calomel electrode, SCE) and a counter electrode (Pt mesh). The potentials reported throughout this paper were thus referred to SCE, if not otherwise specified. To control the working electrode current, a CHI 660C potentiostat (Shanghai CH Instrument Company, China) was used. Subsequent to the application of a potential of 1.0 V, the PPy/SC composites formed. The total passed charge density was observed as big as 5C cm^{-2} relative to the projected anode surface area. The composite sample was carefully rinsed by deionized water aiming to remove the absorbed inclusions with subsequent air-drying at room temperature. The anode modifications with PPy alone and PPy at the presence of commercial activated carbon (AC, Aladdin Industrial Inc. Shanghai, China) were also tested for comparison. The modified anodes were prepared under conditions identical to those described above.

2.3. Microbial fuel cell design and operation

The Perspex flames two-chamber MFCs were constructed as described earlier [24,25]. The MFCs were comprised of the modified graphite felt anode and a bare graphite felt cathode sized 3.0 × 2.0 × 0.5 cm placed into a separate chamber. The chambers with an effective volume of 25 cm^3 each were separated by a cation exchange membrane (Zhejiang Qianqiu Group Co., Ltd., China). *Shewanella oneidensis* MR-1 purchased from ATCC (700550) was used to inoculate the anode chamber. The anolyte comprised of 5.84 g L^{-1} NaCl, 0.10 g L^{-1} KCl, 0.25 g L^{-1} NH_4Cl, 10 mL of vitamin solution and 10 mL of mineral solution contained 20 mM lactate and 0.1 M phosphate buffer solution (PBS, pH 8.0). The cathode compartment was filled with a 0.1 M PBS solution at pH 7.0 containing 50 mM potassium hexacyanoferrate as the electron acceptor. All MFCs run at a constant 30 °C. A 32-channel voltage collection appliance (AD8223, China) with the external resistance of 500 Ω was used to record the cell potentials with 2 min increment.

To measure the power density and to reduce the transient power effect caused by the capacity of the anode material [26], the fed-batch cycle test was applied: the maximum sustainable potential over the cycle typically lasting for 4–12 h was recorded over a complete fed-batch cycle using a resistor. The length of the cycle depends on the external resistance and the anode material. Each resistance given in descending order starting from the open-circuit, followed by 2000, 1000, 500, 250, 100, 50 and 25 Ω was tested for three consecutive cycles confirming the potential response remaining constant along with different cycles. For the methods described above, the anode potential variations were obtained with a SCE electrode inserted into the anode compartment. The computer-interfaced desktop multimeter UT-805A (ShenZhen Uni-Trend Group Limited, China) was used for recording the cell and the anode potentials. Power (P) was calculated as a product of the cell

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