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The effect of chemical vapor deposition temperature on the performance of binder-free sewage sludge-derived anodes in microbial fuel cells

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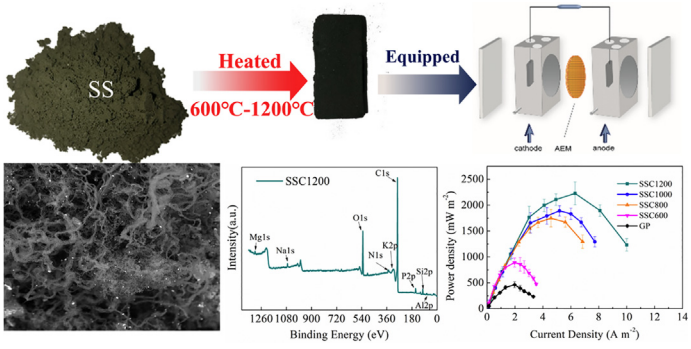
HIGHLIGHTS

- Sewage sludge was firstly converted into electrode without binder.
- An anode processed at 1200 °C achieved the highest power density (2228 mW cm^{-2}).
- High temperature processing increases the electrochemically active surface area.

GRAPHICAL ABSTRACT



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ABSTRACT

Conversion of sewage sludge (SS) into value-added material has garnered increasing attention due to its potential applications. In this study, we propose a new application of the sewage sludge-derived carbon (SSC) as an electrode without binder in microbial fuel cells (MFCs). SS was firstly converted into SSC monoliths by methane chemical vapor method at different temperature (600, 800, 1000 or 1200 °C). Scanning electron microscopy images showed that carbon micro-wires were present on the surfaces of the samples prepared at 1000 and 1200 °C. The results showed that it was beneficial for converting sludge into a highly conductive electrode and increasing carbon content of the electrode at higher temperatures, thereby improving the current generation. The conductivity results show that a higher temperature favors the conversion of sludge into a highly conductive electrode. The MFC using an SSC anode processed at 1200 °C generated the maximum power density of 2228 mW m^{-2} and the maximum current density of 14.2 A m^{-2}. This value was 5 times greater than that generated by an MFC equipped with a graphite anode. These results present a promising means of converting SS into electrode materials.

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1. Introduction

Microbial fuel cells (MFCs) have attracted increasing interest as a component of sustainable electricity generation and wastewater treatment processes, due to their ability to extract energy from contaminants using bacteria as catalysts (Logan, 2009; Kumar et al., 2013). In addition,

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Nomenclature

MFCs	microbial fuel cells
SS	sewage sludge
SSC	sewage sludge -derived carbon
GF	graphite felt
GP	graphite plate
CVD	chemical vapor deposition
CV	cyclic voltammetry
EIS	electrochemical impedance spectra
OCP	open circuit potential
XPS	X-ray photoelectron spectroscopy
SEM	scanning electron microscopy
XRD	X-ray diffraction
CLSM	confocal laser scanning microscope
FTIR	Fourier transform infrared
EDLC	electronic double layer capacitor

the potential of MFC applications in biosensors, desalination and hydrogen gas production attracted more and more attention (Reguera et al., 2005; Marsili et al., 2008; Zhang et al., 2015). However, scaling MFC for commercial application is still limited by the low power density and high cost. An ideal anode must be highly biocompatible and should environmentally friendly, as well as being inexpensive and durable (Wu et al., 2018; Hindatu et al., 2017). The most widely-used anode materials to date have been conventional materials, such as graphite rods, activated carbon, carbon cloth, carbon paper, and carbon felt, owing to their excellent electrochemical stability and commercial availability (Wu et al., 2018; Hindatu et al., 2017). Besides, several metallic materials have been used as anode electrode in MFCs, such as stainless steel, copper, nickel, silver, gold and titanium (Santoro et al., 2017). However, at present, practical applications of anode material, such as graphite felt, is limited by low performance (2 A m^{-2}) and high capital costs (25–75 k$ m^{-3}), which represent more than half the costs of the reactor (Santoro et al., 2017; Ma et al., 2016). Therefore, the future of MFCs technology used in wastewater treatment is the key to finding a low-cost and high-performance electrode material stead of conventional carbon materials.

Sewage sludge, a main byproduct of the activated sludge process and >34 million tons per year in China, contains complex organic material viruses, bacteria and other microorganisms and has not been properly disposed (O'Kelly, 2005). Conventionally, the excess sewage sludge is treated by landfill, composting, and incineration (Jiang et al., 2011; Rozendal et al., 2008). While, all of these methods could be costly and unavoidably induce serious secondary pollutions, such as releasing a large number of pathogens, organic pollutants, dioxins and furans gases (Rozendal et al., 2008; Johnsson, 1994). Therefore, considering the dramatic increase of sewage sludge generated from wastewater treatment plants, continuing efforts are in urgent need on seeking for novel strategies to treat sewage sludge with low cost and alleviate the environmental problems. Note that SS is also a carbon-rich material well-suited for the production of black carbon, which can in turn be used for the preparation of electrodes for MFCs through thermal treatment (e.g., chemical precipitation) presents a potential economic opportunity (Zhang et al., 2012; Rinaldi et al., 2008). Previous studies have demonstrated that sludge-derived carbon anodes can enhance the power output of the MFC because of the improved adhesion of bacteria to the anode and the increased interfacial electron transfer efficiency (Ma et al., 2016; Yuan et al., 2015). As an example, sludge-derived carbon grafted to 3-D graphite felt (GF) was reported to increase the MFC power output by a factor of 3.5 compared with an unmodified GF anode (Ma et al., 2016). In addition, Yuan et al. demonstrated a maximum power density of 969 mW m^{-2} using an MFC anode made from an SS-derived carbon monolith mixed with 10% coconut shell powder (Yuan et al., 2015). These results have indicated that the use of sewage sludge derived carbon as electrode material is promising due to its low cost. However, their follow-up studies were limited by their relatively low performance. In addition, to the best of our knowledge, no effort has been exerted to symmetrically investigate the utilization of SS-derived as electrode materials without binder in a MFC, probably because of the poor electrical conductivity.

Previous investigations have demonstrated that cracking of carbon deposited on the electrode surface by methane chemical vapor deposition (CVD) can increase the anode surface area, enhancing the performance by an order of magnitude (Feng et al., 2016; Liang et al., 2017). The procedure used to prepare this type of anode by CVD is almost the same as that employed during the heat treatment of SS in a tubular furnace by Yuan et al. (2015). Based on it, it is very attractive if the carbonation of the sludge and the modification of the cracked carbon are accomplished simultaneously by adding a small amount of methane to the heating treatment, so as to obtain a kind of sewage sludge–derived carbon electrode with high surface area.

The purpose of this work was to develop a methane CVD method for converting SS into electrodes without binder used as the anode of MFCs. This work assessed the effects of the carbonization temperature on the properties of the SS-derived carbon (SSC), including specific surface area, conductivity and stability. The mechanism by which the performance of MFCs incorporating SSC is improved is also discussed.

2. Materials and methods

2.1. Preparation of the SSC monoliths

Briefly, SS with a water content of approximately 98% was collected from the Tianchuang Sewage Treatment Plant, Hangzhou, China. The SS was screened and filtered using a 40mesh sieve to remove large particles, then dried at 60 °C and further broken down using a blender (ZM200, RETSCH Co., Ltd., Germany). The SS about 4 g was subsequently packed into a mold (18 × 8 × 3 mm) and then pressed at 25 °C. And then placed in a quartz tube and heat-treated in a tubular furnace at varying temperatures for 120 min at a heating rate of 15 °C min^{-1} under a flow of CH_4 (150 mL min^{-1}) and N_2 (150 mL min^{-1}). The temperatures applied to the SS monoliths were 600, 800, 1000 and 1200 °C. Accordingly, the as-prepared SSC samples are referred to herein as SSC600, SSC800, SSC1000 and SSC1200.

2.2. MFC setup and operation

Each experimental trial was performed in a double-chamber reactor. The cathode (45 mL) and anode (45 mL) chambers were cylindrical, each with a cross-sectional area of 13 cm^{-2}. These two chambers were assembled on either side of a cation exchange membrane (CEM, DuPont, USA) and held securely in place by six clamping bolts. The anodes were SSC electrodes measuring 1.8 × 0.8 × 0.3 cm (length × width × thickness). For comparison purposes, a graphite plate (GP) electrode (18 × 8 × 3 mm) provided by Beijing Sanye Co. Ltd., China was also placed in MFC as an anode. The anode chamber and the cathode were run fed-batch mode. The anolyte consisted of CH_3COONa (1 g/L), M9 solution (NH_4Cl, 0.1 g/L; NaCl, 0.5 g/L; KH_2PO4, 4.4 g/L; K_2HPO_4, 3.4 g/L; $MgSO_4$, 0.1 g/L; $NaHCO_3$, 2 g/L) and trace elements ($FeSO_4$, 1.0 mg/L; $CuSO_4$, 0.02 mg/L; H_3BO_3, 0.014 mg/L; $MnSO_4$, 0.10 mg/L; $ZnSO_4$, 0.10 mg/L; Na_2MoO_4, 0.02 mg/L; $CoCl_2$, 0.02 mg/L) (Liang et al., 2016).The anolyte was flushed with high-purity nitrogen gas for 15 min and inoculated with (10 mL) fresh anodic effluent (OD600 was about 1) of an existing acetate-fed bio-electrochemical reactor that was rich in *Geobacter*. The bio-electrochemical reactor comprised of the graphite the anode and graphite felt cathode has been continuously operated for five months in the lab. Carbon felt (2 cm

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