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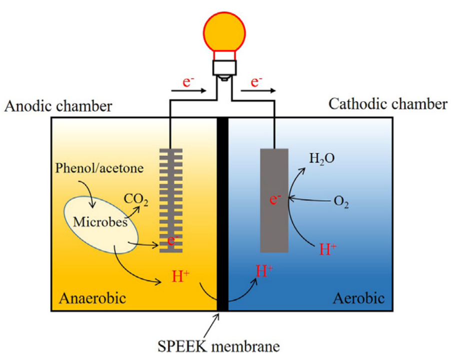
Electricity generation and removal performance of a microbial fuel cell using sulfonated poly (ether ether ketone) as proton exchange membrane to treat phenol/acetone wastewater

Hao Wu[a], Yu Fu[b], Chunyu Guo[a], Yanbo Li[a], Nanzhe Jiang[a], Chengri Yin[a,c,*]

[a] Key Laboratory of Natural Resources of Changbai Mountain and Functional Molecules, Ministry of Education, Department of Chemistry, Yanbian University, Yanji 133002, China
[b] College of Chemistry and Life Science, Anshan Normal University, Anshan 114007, China
[c] Analytical and Testing Center, Yanbian University, Yanji 133002, China

GRAPHICAL ABSTRACT



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ABSTRACT

The microbial fuel cell (MFC) has emerged as a promising technology for wastewater treatment and energy recovery, but the expensive cost of proton exchange membranes (PEMs) is a problem that need to be solved. In this study, a two-chamber MFC based on our self-made PEM sulfonated poly (ether ether ketone) membrane was set up to treat phenol/acetone wastewater and synchronously generate power. The maximum output voltage was 240–250 mV. Using phenol and acetone as substrates, the power generation time in an operation cycle was 289 h. The MFC exhibited good removal performance, with no phenol or acetone detected, respectively, when the phenol concentration was lower than 50 mg/L and the acetone concentration was lower than 100 mg/L. This study provides a cheap and eco-friendly way to treat phenol/acetone wastewater and generate useful energy by MFC technology.

1. Introduction

Phenol and acetone are important and essential raw materials in the pharmaceutical, chemical, petroleum and leather industries, and can be produced by an industrial synthesis via the cumene process from benzene and propylene, which has the advantages of being cheap and

* Corresponding author at: Key Laboratory of Natural Resources of Changbai Mountain and Functional Molecules, Ministry of Education, Department of Chemistry, Yanbian University, Yanji 133002, China.
E-mail address: cryin@ybu.edu.cn (C. Yin).

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effective. Every year, nearly 700 million ton of phenol is annually produced (Senthilvelan et al., 2013), and until 2010, the worldwide production capacity for acetone was estimated at 6.7 million tons per year. The excessive discharge of phenol/acetone wastewater (even at low concentrations) into aqueous ecosystems causes a series of environmental problems (Khaksar et al., 2017; Ren et al., 2017). Thus, to guarantee the safety and health of people, and the ecosystem, the efficient treatment of phenol/acetone wastewater is necessary.

Many methods have been successfully used in the treatment of phenol/acetone wastewater such as solvent extraction, adsorption, oxidation (including chemical, photo, electrochemical and other advanced oxidation processes), and biodegradation (Abbassian et al., 2014; Shi et al., 2017; Zhang et al., 2013). Among these methods, biodegradation offers the advantages of being economical, eco-friendly and producing no secondary by-products or pollutants, compared with other methods (Wu et al., 2016). However, traditional biodegradation processes have only focused on degrading organic pollutants and have ignored the extraction of chemical energy from the pollutants. As a result, most of this chemical energy is converted into biomass. The excessive biomass produced can often cause clogging of the various bioreactors and inhibition of mass transfer and biodegradation. Thus, resource utilization of waste is important.

Microbial fuel cells (MFCs) make it possible to deal with the two problems of water availability and energy shortage. MFCs use microbes to convert the chemical energy of pollutants in wastewater into electrical energy (Hernández-Fernández et al., 2015). In a MFC system, proton exchange membranes (PEMs) are an important factor that directly affect the final cost of the process and the performance both in pollutant removal and electricity generation. Due to the advantage of their high proton conductivities, perfluorinated ionomer membranes, such as Nafion membranes, have often been used as PEMs in MFC systems. However, Nafion membranes are very expensive (sometimes reaching 3000 USD per m^2 in China). In addition, the high degree of fluorination can cause potential environmental problems. Thus, a cheap and environmentally-friendly PEM alternative is required.

Many alternative PEMs, including sulfonated poly-(ether sulfone)s, poly-(ether ketone)s, poly-(phenylene)s, polyimidides and polybenzimidazoles have been studied in various fuel cells due to their low cost. However, they have problems with dimensional stability and mechanical strength due to the water uptake caused by their highly sulfonic natures (Han et al., 2014; Park et al., 2011). Sulfonated poly (ether ether ketone) (SPEEK) membranes are a cheap alternative and are widely used in polymer electrolyte membrane fuel cells (Kim et al., 2017). PEEK has many advantages that include excellent thermal stability and mechanical properties, low cost, and easy sulfonation. In addition, SPEEK is environmental friendly due to its lack of fluorine (Ayyaru and Dharmalingam, 2011). Increasing the sulfonation degree can help SPEEK achieve sufficient proton conductivity, however, it also creates the problems of poor mechanical properties and low dimensional stability (Han et al., 2014). Thus, an appropriate sulfonation degree is important for SPEEK membranes to be used in MFCs.

To date, SPEEK membranes have not been used in MFCs to treat phenol/acetone wastewater. In this study, a self-made SPEEK membrane was prepared and used as the PEM in a lab-scale MFC to treat phenol/acetone wastewater, while also analyzing the generation of electricity. This study will further decrease the cost of phenol/acetone wastewater treatment by MFCs.

2. Materials and methods

2.1. MFC set-up and operation

A two-chamber cube-type MFC was used in this study. The anode and cathode chambers (252 mL) were separated by a 16 cm^2 self-made SPEEK membrane with a 0.546 sulfonation degree (the preparation of SPEEK is described in the Supplementary data). A graphite brush was used as the anode and carbon felt was used as the cathode. The microorganism used in this study was the activated sludge collected from a sewage plant in Yanji, Jilin province, China. The activated sludge supernatant mixed with nutrient solution was inoculated into the anode chamber of the MFC. Copper wires were used to establish a contact between the electrodes, external resistance and data recorder. The anode chamber was operated with a peristaltic pump at an intermittent mode (the anolyte was replaced at each operation cycle). The operation time of the MFC can be divided into eight stages (shown in Table 1). During the start-up periods, activated sludge supernatant was added into the MFC anode chamber with an enrichment medium. The data from stage A to G were recorded for two cycles during each stage.

Table 1
MFC operation conditions.

Stage	Start-up	A	B	C	D	E	F	G
Glucose (g/L)	1	0.99	0.985	0.94	0.85	0.7	0.3	0
Phenol (mg/L)	–	5	5	20	50	100	200	500
Acetone (mg/L)	–	5	10	40	100	200	500	500

2.2. Analytical methods

The mechanical properties of the Nafion membrane and the prepared SPEEK membrane were determined by a Series IX Universal Testing System Model 4400 (INSTRON, MA, USA) with a testing speed of 50 mm/min. Each sample was tested at least three times and the final result was the average of the tested values. The ion exchange capacity (IEC) is the number of equivalents of bound dissociated groups in PEM related to the weight of dry matter (Bulejko and Stránská, 2018; Bulejko et al., 2016). The IEC of the membranes in this study was determined in the H^+ form by displacing H^+ with Na^+, and titrating the released the H+ with a NaOH solution using phenolphthalene as an indicator.

The phenol concentration was measured using a HP-1100 high performance liquid chromatography system (Agilent, CA, USA) with an ultraviolet detector. The acetone concentration was analyzed by a GC-2010 gas chromatography (Shimadzu, Tokyo, Japan) with a flame ionization detector. Based on the concentration change, the removal efficiency (RE) was calculated to evaluate the removal performance of the MFC.

Electrochemical data were measured using a UT52 multimeter (UNI-T, Dongguan, China). During the operating time, the external resistance was fixed at 500 Ω, and the output voltage was recorded with a 1 min interval. The power density (PD) and polarization curve of the MFC were plotted by changing the external resistance (50–9999 Ω). In addition, the current (I), power (P), current density (CD) and internal resistance of the MFC were calculated following a reported method (Wu and Lin, 2016).

3. Result and discussion

3.1. Characteristics of membrane analysis

The characteristics of the SPEEK membrane and Nafion 212 membranes are shown in Table 2. The IECs of the SPEEK membrane and

Table 2
Characteristics of SPEEK and Nafion 212 membranes.

Sample	IEC (meq/g)	Tensile strength (MPa)		Young's modulus (GPa)		Elongation at break (%)	
		Dry	Wet	Dry	Wet	Dry	Wet
SPEEK	1.527	81	21	3.0	0.4	48	85
Nafion 212	0.94	37	33	0.3	0.2	242	250