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# Performance of integrated bioelectrochemical membrane reactor: Energy recovery, pollutant removal and membrane fouling alleviation



Yue Dong<sup>a</sup>, Weihua He<sup>a,\*\*</sup>, Chao Li<sup>a</sup>, Dandan Liang<sup>a</sup>, Youpeng Qu<sup>b</sup>, Xiaoyu Han<sup>a</sup>, Yujie Feng<sup>a,\*</sup>

<sup>a</sup> State Key Laboratory of Urban Water Resource and Environment, Harbin Institute of Technology, No 73 Huanghe Road, Nangang District, Harbin, 150090, China <sup>b</sup> School of Life Science and Technology, Harbin Institute of Technology, No. 2 Yikuang Street, Nangang District, Harbin, 150080, China

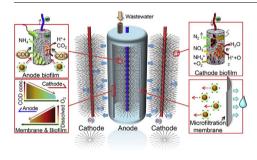
#### HIGHLIGHTS

- A novel bioelectrochemical membrane reactor (BEMR) was developed.
- Successful operation of the BEMR with microfiltration membrane as separator.
- The relationship between power out and aeration intensity was investigated.
- Oxygen utilization efficiency of biocathode was well addressed.
- The bioelectrochemical process was efficient for membrane fouling alleviation.

#### ARTICLE INFO

Keywords: Microbial electrochemical system Membrane bioreactor Biocathode Gas-water ratio Net energy generation

### G R A P H I C A L A B S T R A C T



#### ABSTRACT

A novel hybrid bioelectrochemical membrane reactor with integrated microfiltration membrane as the separator between electrodes is developed for domestic wastewater treatment. After accumulation of biofilm, the organic pollutants are mainly degraded in anodic compartment, and microfiltration membrane blocks the adverse leakage of dissolved oxygen from aerated cathodic compartment. The maximum system power output is restricted by gas-water ratio following a Monod-like relationship. Within the tested gas-water ratios ranging from 0.6 to 42.9, the half-saturation constant ( $K_Q$ ) is 5.9  $\pm$  0.9 with a theoretic maximum power density of 20.4  $\pm$  1.0 W m<sup>-3</sup>. Energy balance analysis indicates an appropriate gas-water ratio regulation (from 2.3 to 28.6) for cathodic compartment is necessary to obtain positive energy output for the system. A maximum net electricity output is 9.09  $\times$  10<sup>-3</sup> kWh m<sup>-3</sup> with gas-water ratio of 17.1. Notably, the system achieves the chemical oxygen demand removal of 98.3  $\pm$  0.3%, ammonia nitrogen removal of 99.6  $\pm$  0.1%, and total nitrogen removal of 80.0  $\pm$  0.9%. This work verifies an effective integration of microfiltration membrane into bioelectrochemical system as separator for high-quality effluent and provides an insight into the operation and regulation of biocathode system for effective electrical energy output.

#### 1. Introduction

In the past decades, microbial electrochemical system (MES) has been regarded as a promising technology that could accelerate the degradation and removal of organic pollutants in an energy-efficient mode [1]. The fundamental theory and system construction of MES have been well demonstrated, including microbial ecology, electrode material, and reactor configuration [2,3]. Currently, the application of MES is still limited in a scope of lab-scale reactors and the goal of scaling up MES for practical application in wastewater treatment is far

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<sup>\*</sup> Corresponding author.

<sup>\*\*</sup> Corresponding author.

E-mail addresses: ablesee@163.com (W. He), yujief@hit.edu.cn (Y. Feng).

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from success. One of the biggest challenges lays in cathode system [4]. Air-cathodes were widely used in a variety of pilot-scale systems, but they suffered from complicated structures, cumbersome fabrication process, and short service life [5,6]. A more feasible alternative might be biocathode due to its advantages in low cost, long service life, and small footprint [7].

In respect of biocathode, most of the previous studies have been focused on the working mechanism and function extension by cultivating special microorganisms. For example, the research of tapping external electrons as the energy source for exoelectrogens in the cathode biofilm was conducted to explore extracellular electron transfer mechanism [8,9]. A type of biocathode utilized nitrate or sulfate as major terminal electron acceptors to realize the removal of nitrogen and sulfur in wastewater [10,11], as well as the aerobic nitrifying biocathode [12]. Electrode materials were also intensively studied including stainless steel mesh coated CNT and graphene-impregnated carbon cloth [13,14]. These were all important studies contributing greatly to future application of MES. However, unfortunately, the available biocathode systems still presented several deficiencies needed to be overcome. Firstly, aeration in the cathodic compartment to supply oxygen as the terminal electron acceptor was not only essential to power generation and system performance but also considered as the primary system energy consumption source. Thus, proper aeration intensity needed to be confirmed urgently, when considering the balance between power output and aeration cost. However, none of these prior studies analyzed the quantitative parameters, such as the volumetric half-saturation or saturation aeration intensity, for the biocathode system. Secondly, it has been accepted that high chemical oxygen demand (COD) concentration in cathodic compartment had adverse effects on power and current generation [15]. For practical application, the COD variation in cathodic compartment might lead to a sudden change of system performance or even the collapse of the whole system. The current generation flow between the electrodes has also been proved to accelerate the COD and SCOD removal in air-cathode system [16,17]. However, the current assisted COD removal in the biocathode system was still unknown. So, the capability of separator in blocking COD penetration from anode and the ability of biocathode on COD removal should be well deliberated.

Another obstacle of MES for wider application was the low effluent quality, which has not been sufficiently addressed. In terms of highquality effluent, constructing hybrid systems with MES and membrane technology has been proved to be effective in improving effluent quality [18]. However, most of those systems simply employed membrane filtration as pre- [19] or post-treatment [20] processes. Only few studies effectively integrated membrane into MES construction, such as utilizing filter membrane as anode [21], cathode material [22], or separator [23]. Among these, the use of filter membrane as a separator could achieve high effluent quality and compact reactor construction. The feasibility of using filter membrane as a separator between anode and cathode has been investigated in MES equipped with abiotic chemical cathodes [24]. However, for biocathode, the issue of crossover leakage of oxygen and COD between anode and cathode chambers will inevitable arise in continuous mode. Using filter membrane as separator could utilize the biofilm attached on the membrane surface for COD degradation other than simple physical filtration process. Besides, the biofilm attached on filtration membrane could form dissolved oxygen (DO) and COD gradients. The DO gradient facilitated the availability of oxygen rejection performance between anaerobic anodic chamber and aerobic biocathodic chamber. The COD gradient could reduce the organic loading shock on autotrophic biocathode community. Moreover, the presence of DO and COD gradient within the biofilm on the microfiltration membrane was beneficial to the simultaneous enrichment of nitrifiers and denitrifiers [25]. This nitrification and denitrification processes would be in favor of nitrogen removal in the system.

In this study, a novel hybrid bioelectrochemical membrane reactor (BEMR) coupled with biocathode was constructed to treat low-strength

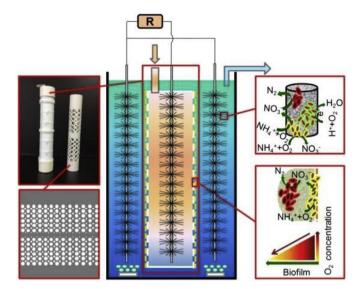


Fig. 1. Schematic of the bioelectrochemical membrane reactor (BEMR).

domestic wastewater. The microfiltration (MF) membrane was not only functioned as a filter layer but also integrated as the separator between anode and biocathode. The objectives of this research were: (1) to demonstrate the feasibility of MF-membrane as the separator in an aeration system with biocathode; (2) to establish the energy balance based on the dynamic change of membrane pressure and aeration intensity; (3) to assess the effect of aeration in the cathodic compartment on the power output and wastewater treatment; (4) to investigate the COD removal capacity and oxygen utilization efficiency of the biocathode.

#### 2. Materials and methods

#### 2.1. Reactor setup and operating conditions

The BEMR consisted of an anodic compartment and two biocathodes, with an effective volume of 700 mL (Fig. 1 and Fig. S1). The tubular anodic compartment was fabricated by inserting a carbon brush (4 cm diameter by 20 cm) into a perforated polyvinylchloride (PVC) tube, which had a total pore area of  $95.6 \text{ cm}^2$  (open area of 44.9%). The working volume of the anodic compartment was 210 mL. A piece of MFmembrane (pore size of 0.45 µm, Motian Inc., Tianjin, China) was used as the separator supported by the PVC tube from inside. To resist water pressure, a piece of nylon mesh (200 mesh, wire diameter 0.18 mm) covered outside of the MF-membrane. Two brush biocathodes (4 cm diameter by 20 cm) were parallel placed at both sides of the tubular anodic compartment with a distance of 0.2 cm. The two biocathodes were connected together in external circuit, and then connected with the anode through an external resistor of  $50\,\Omega$ . Two aerators were placed at the bottom of biocathode for oxygen dispersion, and the aeration rate was controlled by an air flowmeter (µFlow, bioprocess control, China).

The anode and biocathode were pre-cultured in another twochamber biocathode reactor and stably operated for 6 month before being installed into this BEMR (detailed information in SI). Domestic wastewater collected on HIT (Harbin Institute of Technology, China) campus was directly pumped into the anode from the top of anodic compartment. The effluent from the anodic compartment penetrated through the MF-membrane to the cathodic compartment, and finally discharged from the system. The influent flowrate was 0.7 mL min<sup>-1</sup>, producing a hydraulic retention time (HRT) for the anodic compartment of 5 h. The system was firstly operated under a low aeration intensity (Run 1, 1.6 mL min<sup>-1</sup>) with gas-water ratio (GWR) of 2.3 lasting 20 days for the enrichment of biofilm on MF-membrane to reject oxygen diffusion. Then the aeration rates varied from 0.4 to Download English Version:

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