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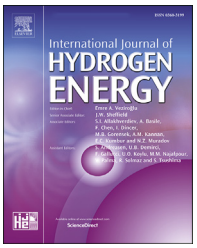

An effective method for hydrogen production in a single-chamber microbial electrolysis by negative pressure control

Huajun Feng [a,b], *Lijie Huang* [a,b], *Meizhen Wang* [a,b], *Yingfeng Xu* [a,b], *Dongsheng Shen* [a,b], *Na Li* [a,b], *Ting Chen* [a,b,*], *Kun Guo* [a,c,**]

[a] *School of Environmental Science and Engineering, Zhejiang Gongshang University, Hangzhou, 310012, PR China*
[b] *Zhejiang Provincial Key Laboratory of Solid Waste Treatment and Recycling, Hangzhou, 310012, PR China*
[c] *Center for Microbial Ecology and Technology, Ghent University, Coupure Links 653, Ghent, B-9000, Belgium*

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ABSTRACT

In this study, we construct a scalable tubular single-chamber microbial electrolysis cell that using negative pressure (40.52 kPa) to enhance the hydrogen production. The impact of negative pressure on current production, hydrogen recovery, and microbial community of microbial electrolysis cells are investigated. Negative pressure could effectively enhance the hydrogen recovery and inhibit the growth of methanogens. Consequently, the microbial electrolysis cell operated under negative pressure achieves a maximum hydrogen production rate of 7.72 ± 0.06 L L^{-1} d^{-1}, which is more than four times higher that of reactor running under normal pressure (1.51 ± 0.41 L L^{-1} d^{-1}). Energy quantification shows that the electrical energy recovery under negative pressure is 146.98%, which is much higher than 95.00% under normal pressure. Therefore, negative pressure control is as effective for increasing hydrogen production and energy recovery in the scalable MEC, and has a great practical application prospect. However, negative pressure cannot knick out methanogens. Once negative pressure is removed, methanogens will quickly take over and after that applying negative pressure again can only partly inhibit methane production.

Introduction

Microbial electrolysis cells (MECs) is an emerging technology for hydrogen production from organic waste [1,2]. Thus far, the normal hydrogen production rate (HPR) of MECs was around 2–7 $L/L_{reactor}/d$, which is similar to that of hydrogen production by dark fermentation [3]. However, the hydrogen yield of MEC was about one order of magnitude higher than that of dark fermentation because electricity producing bacteria (also known as exoelectrogens) could completely degrade organic matter into carbon dioxide [4]. Currently, the main bottlenecks for the practical application of MEC are its complex reactor configuration and high capital cost [5–7].

Removing the ion exchange membranes from MECs could not only simply the reactor design but also reduce the reactor internal resistance and capital cost [5,8]. Hence, numerous of membrane-less MECs from lab to pilot scales have been

* *Corresponding author*. School of Environmental Science and Engineering, Zhejiang Gongshang University, Hangzhou, 310012, PR China.
** *Corresponding author*. Center for Microbial Ecology and Technology, Coupure Links 653, Ghent, B-9000, Belgium.
E-mail addresses: chenting_2013@tsinghua.org.cn (T. Chen), kun.guo@ugent.be (K. Guo).

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designed for hydrogen production. However, it was found that the low hydrogen recovery was a big challenge for these reactors [9]. Without membrane as the barrier, the hydrogen gas produced by the cathode could be easily scavenged by methanogens, homoacetogens, and even exoelectrogens [10–12].

To inhibit hydrogen scavengers and improve hydrogen recovery of membrane-less MECs, a variety of approaches have been applied such as adding chemical inhibitors, changing operational parameters (e.g. pH, temperature, hydraulic retention time), optimizing reactor design, and actively recovering hydrogen by vacuum [13–17]. Among these methods, active H_2 harvesting by vacuum seems to be the most promising approach for methanogens inhibition. However, the experiment was done in a small scale reactor with gas diffusion cathode running in batch mode. Thus, it is still not sure whether this method could be applied to larger scale MECs with traditional submerged cathode and running in continuous mode. Moreover, the impact of vacuum pressure on the microbial community evolution in MECs was unknown yet.

Therefore, the objective of this study was to increase hydrogen production in a scalable MEC and to investigate the impact of negative pressure on the current production, hydrogen recovery, methanogenesis inhibition, and microbial community in a scalable single-chamber tubular MEC running at continuous mode.

Materials and methods

Reactor design and construction

The single-chamber tubular MEC consists of a glass vessel (working volume 0.5 L), a custom-made Perspex cap, a tubular anode (inner tube), and a tubular cathode (outer tube) (Fig. S1). The anode was made from a pieces of SS fiber felt (316 L SS, 100 mm filtering rate, Lier Filter Ltd, Xinxiang, China) which has a dimension of 40 cm × 8 cm × 1 mm (surface area 320 cm^2). The felt was first folded into a pleated configurations with 40 layers (1 cm per fold along the length) and then rolled into a tube with an inner diameter of 2.5 cm and outer diameter of 3.5 cm. Seven macro holes (2 mm diameter) were evenly drilled on each layer in order to improve mass transfer [18]. The SS felt tube was finally treated in a muffle furnace (600 °C, 5 min) as the procedure described before [19]. The cathode was a platinum-coated titanium mesh tube (surface area 175 cm^2, outer diameter 7 cm, height 8 cm, mesh thickness 1.2 mm, mesh hole size 3 mm × 6 mm, platinum coating thickness 1 μm). The Perspex cap and the glass vessel were clamped together with an O-ring in between for sealing. The five openings on the reactor cap were used for reference electrode, anode, cathode, gas collection, and gas pressure gauge, respectively. All reported potentials in this study are with respect to the Ag/AgCl (3 M KCl) reference electrode.

Startup and operation

The medium used in this study was a modified M9 medium with 2 g/L sodium acetate as the electron donor [20]. The medium was always sparged with nitrogen gas for 15 min before use. Three MECs were running in parallel. The inoculum of MEC1, MEC2 and MEC3 was effluent of an existing acetate-fed MEC running with BES (2-bromoethanesulphonate, BES) (Table 1). A magnetic stirrer was used to continuously mix the solutions at a speed of 350 rpm. The reactors were started up with a Biologic VSP potentiostat by setting the anode potential at −0.2 V (vs. Ag/AgCl) in batch mode. When the reactors were started up and their current started to drop for the first time, they were then switched to continuous mode with a medium feeding rate of 1.67 mL/min using a peristaltic pump (BT100-2J, Longer Pump Co.). To maintain the water level, another pump was operating at the exact same flow rate to pump water out of the reactor. The negative gas pressure was generated by the same peristaltic pump which was operated at its maximum flow rate to continuously draw gas out of the reactor (Fig. S2). The resulted negative pressure was stabilized around 40.52 kPa. During continuous mode, potentiostat channels were disconnected from the reactors and power supplies were used (KA3005P, Korad, China) to apply a voltage of 1 V to each reactor for the later operation. The current was then recorded by the software supplied by the power supply (KA3005P PC software). MEC2 was operated under negative pressure for the whole operational 30 days. MEC1 was operated under normal pressure (absolute pressure of 101.3 kPa) for the same time period. MEC3 was first operated under negative pressure for 10 days, and then 12 days under normal pressure, and finally back to negative pressure till the end of the experiment. All experiments were performed in a 30 °C temperature-controlled room.

Analyses

The gas produced from the reactor was collected and measured using a water displacement column (glass tube, internal diameter 26 mm, 500 mL) which was filled with pH 2 H_2SO_4. The top end of the column was closed by a rubber stopper so that gas samples can be taken by syringe and needle. The concentrations of H_2, N_2, O_2, CO_2 and CH_4 were determined by gas chromatography (GC7900, Techcomp, China). The GC was equipped with a thermal conductivity detector. The carrier gases were He for the N_2 channel and N_2 for all of the other gases.

In order to understand the evolution of microbial community under different operating conditions, the community composition of the inoculum and the anodic biofilm after 30 days of operation were analyzed by 16S rRNA gene sequencing. The samples' DNA were extracted using an extraction kit (V2.2, Biocolor, Shanghai, China) according to

Table 1 – Operational pressure of the reactors overtime.

Reactor	Pressure		
	0–10 day	11–22 day	23–30 day
MEC 1	Normal	Normal	Normal
MEC 2	Negative	Negative	Negative
MEC 3	Negative	Normal	Negative

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