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A novel tubular microbial electrolysis cell for high rate hydrogen production

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HIGHLIGHTS

- A double-chamber tubular MEC with low internal resistance (0.33 Ω) was designed.
- The pleated SS felt anode enabled a high current density and a low energy loss.
- A hydrogen production rate (7.1 L L^{-1} d^{-1}) was achieved with 1 V voltage.
- The reactor showed high hydrogen recovery/purity and excellent operational stability.

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ABSTRACT

Practical application of microbial electrolysis cells (MECs) for hydrogen production requires scalable reactors with low internal resistance, high current density, and high hydrogen recovery. This work reports a liter scale tubular MEC approaching these requirements. The tubular cell components (a platinum-coated titanium mesh cathode, an anion exchange membrane, and a pleated stainless steel felt anode) were arranged in a concentric configuration. The reactor had a low internal resistance (0.325 Ω, 19.5 mΩ m^2) due to the high conductivity of the electrodes, a compact reactor configuration, and proper mixing. With acetate as electron donor, the MEC achieved a volumetric current density of 654 ± 22 mA L^{-1} (projected current density, 1.09 ± 0.04 mA cm^{-2}) and a volumetric hydrogen production rate of 7.10 ± 0.01 L L^{-1} d^{-1} at an applied voltage of 1 V. The reactor also showed high hydrogen recovery (~100%), high hydrogen purity (>98%), and excellent operational stability during the 3 weeks of operation. These results demonstrated that high hydrogen production rate could be achieved on larger scale MEC and this tubular MEC holds great potential for scaling up.

1. Introduction

The microbial electrolysis cell (MEC) is an emerging technology for hydrogen production from biodegradable organic matter. In a MEC, exoelectrogenic bacteria oxidize organic matter into CO_2, protons, and electrons at the anode, and hydrogen evolution reaction takes place abiotically at the cathode [1]. The low operational potential of the microbial anode allows the MEC to start producing hydrogen by applying a substantially smaller cell voltage (>0.2 V in practice) than in traditional water electrolysis (>1.6 V) [2]. The gain of energetic efficiency induced by the use of a microbial anode is unfortunately tarnished by low current densities (i.e. H_2 production rate) in comparison with abiotic electrolysers. Since the first reports on MEC [3,4], numerous studies have investigated electrode materials and reactor configurations to reduce the capital cost and improve the performance of the system [5,6]. To the best of our knowledge, the maximum hydrogen production rates (HPRs) of small-scale (volume < 100 mL) and middle-sized (100 mL < volume < 1 L) MECs have reached 50 L L^{-1} d^{-1} [7] and 2.8 L L^{-1} d^{-1} [8], respectively, while for those larger than 1 L the HPRs have not exceeded 1 L L^{-1} d^{-1} [9]. The poor performance of larger MECs is largely attributed to the low electrode surface to reactor volume ratio, large internal resistances caused by the high resistance of carbon electrodes, and poor mass transfer in the larger reactor [10].

Based on the presence of a separating membrane, MEC reactors can be divided into double-chamber and single-chamber MECs. Most of the first designs included a membrane to favour high hydrogen recovery (i.e. faradaic efficiency) and hydrogen purity.

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However, these systems require a higher applied voltage to reach substantial current density because of their high internal resistance and pH difference across the membrane [3,11]. Removing the membrane could not only solve these issues but also simplify the reactor design and reduce the capital cost [12]. Hence, a variety of single-chamber MECs have been designed. However, a major drawback of single-chamber MECs is the hydrogen consumption by microorganisms such as methanogens [13,14], electrogens [15] and homoacetogens [16,17], especially for long-term operation. For example, methane rather than hydrogen dominated the gas production in a pilot single-chamber MEC [18]. Therefore, a membrane appears necessary to achieve a high production rate of pure hydrogen in MECs.

In order to reduce the internal resistance and the mass transfer limitations in a scalable double-chamber MECs, we have designed a novel tubular MEC reactor (total working volume 1 L). The tubular configuration allows to optimize the ratio of membrane and electrode surface area to reactor volume. In addition, a pleated and tubular stainless steel (SS) fiber felt was fabricated as the anode. We assumed this 3D-structured electrode would improve the volumetric current density of the MEC. Furthermore, it is also expected that the excellent conductivity of the SS felt could also reduce the ohmic resistance of the system. Our results demonstrated that these novel electrode and reactor design dramatically enhances the HPR and hydrogen recovery.

2. Materials and methods

2.1. Reactor design and construction

The MEC design can be seen in Fig. 1, photos of each reactor component are provided in the Supporting Information (Fig. S1). The MEC consisted of a jacketed glass vessel (inner diameter (ID) 9 cm, height 17 cm, working volume 1 L), a tubular anion exchange membrane (diameter 6 cm, height 15 cm, AMI-7001T, Membranes International, USA), and a custom-made Perspex reactor cap. The anode was made of 316L stainless steel fiber felt (SS felt, 40 cm × 15 cm × 1 mm, 100 μm filtering rate, Lier Filter Ltd, China). To fit in the reactor, the felt was folded into a pleated configuration with 40 layers (1 cm per fold along the length) and rolled into a tube (outer diameter (OD) 8.5 cm, ID 7.5 cm, height 15 cm). To further improve the mass transfer, 11 macro holes (5 mm diameter) were evenly drilled on each SS felt layer. The SS felt tube was finally treated in a Muffle furnace at 600 °C for 5 min under air as described before to improve its bio-compatibility toward electroactive biofilm [19]. The tubular membrane was plugged onto the central tube (OD 6 cm, ID 5 cm) of the reactor cap and the joint area (height 1 cm) was sealed by butyl tape. The other end of the tubular membrane was closed by a Perspex closing cap (height 1 cm, OD 6 cm), leaving the effective membrane height and surface area of 13 cm and 245 cm^2, respectively. The cathode was a platinum coated titanium mesh tube (OD 5 cm, height 15 cm, mesh thickness 1.2 mm, mesh hole size 3 mm × 6 mm, platinum coating thickness 1 μm). The reactor cap and the glass vessel were clamped together with a rubber O-ring in between as the sealing material. Four openings on the reactor cap were used for the reference electrode (RE, Ag/AgCl, 3M KCl), the pH probe, the anolyte inlet, and the anolyte outlet. All potentials reported are referred to the Ag/AgCl RE. The top opening of the cathode chamber was closed by a rubber stopper. A glass tube passing through the center of the stopper was used to collect the gas produced.

2.2. Startup and operation

The medium used in this study was a modified M9 medium which contains 6 g L^{-1} Na_2HPO_4, 3 g L^{-1} KH_2PO_4, 0.5 g L^{-1} NH_4Cl,

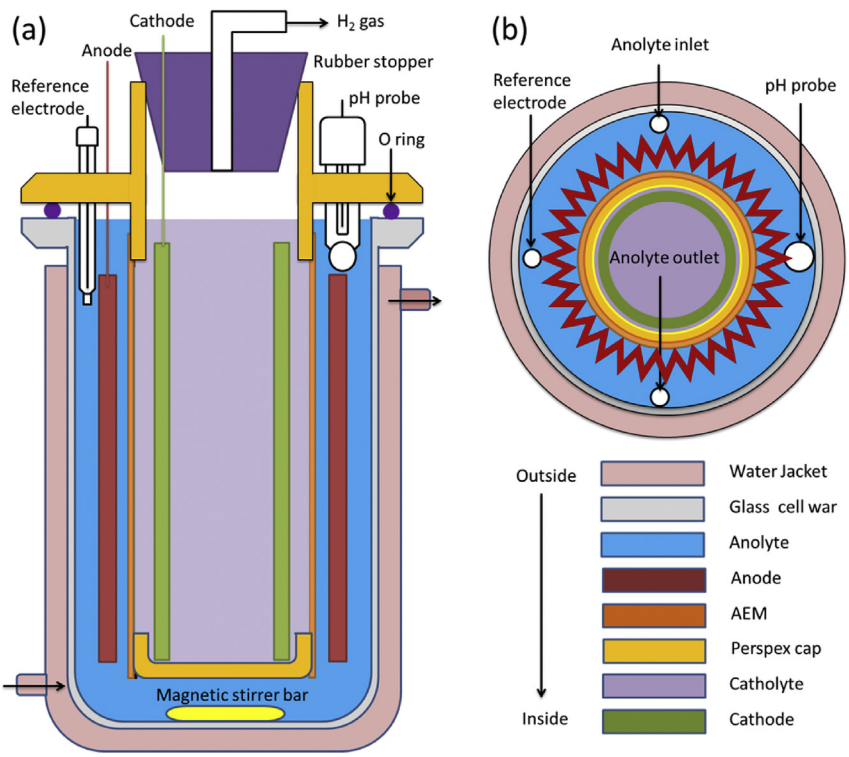

Fig. 1. Schematic sectional view (a) and top view (b) of the tubular MEC.

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