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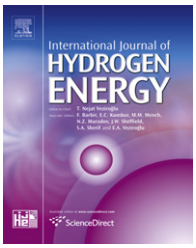

Characterization of energy losses in an upflow single-chamber microbial electrolysis cell

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ABSTRACT

We characterized electrode energy losses and ohmic energy loss in an upflow, single-chamber microbial electrolysis cell (MEC) with no metal catalyst on the cathode. The MEC produced 0.57 m^3-H_2/m^3-d at an applied voltage of ~1 V and achieved a cathodic conversion efficiency of 98% and a H_2 yield of 2.4 mol H_2/mol acetate. Eliminating the membrane lowered the ohmic energy loss to 0.005 V, and the pH energy loss became as small as 0.072 V. The lack of metal catalyst on the cathode led to a significant cathode energy loss of 0.56 V. The anode energy loss also was relatively large at 0.395 V, but this was artificial, due to the high positive anode potential, poised at +0.07 V (vs. the standard hydrogen electrode). The energy-conversion efficiency (ECE) was 75% in the single-chamber MEC when the energy input and outputs were compared directly as electrical energy. To achieve an energy benefit out of an MEC (i.e., an ECE >100%), the applied voltage must be less than 0.6 V with a cathodic conversion efficiency over 80%. An ECE of 180% could be achieved if the anode and cathode energy losses were reduced to 0.2 V each.

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1. Introduction

Although H_2 gas has many merits as an energy-carrier, it will not become a viable renewable option until it is produced from a non-fossil-fuel source and the cost of its production and delivery decreases substantially [1]. A microbial electrolysis cell (MEC) is a biomass-based approach that has the potential to meet these future energy requirements. It has advantages over dark-fermentative H_2 production due to its high H_2 yields (~9 mol H_2/mol glucose versus ~2 mol H_2/mol glucose for dark fermentation) [2–5]. One drawback of the MEC, however, is that it requires electrical energy input generated from fossil fuels for H_2 generation (i.e., an applied voltage), which can increase the H_2-production cost and lower the net energy output. A second drawback is that most MECs today include expensive metal catalysts on the cathode, typically platinum (Pt) [6–12]. The average cost of Pt is $38 per gram of Pt in 2009 [13]. For the MEC to become a practical H_2 producer, high H_2-production rates and yields must be attained with low applied voltage and without a high cost of precious catalysts. The first two features also are necessary for the MEC to produce a net energy benefit.

The applied voltage is one of the most significant factors controlling energy efficiency (i.e., the H_2-production cost) in an MEC. While the applied voltage does not affect the H_2 yield directly, a large applied voltage lowers the net energy value of the generated H_2. The applied voltage depends on the energy losses generated by MEC operation. Energy loss is the difference between the equilibrium electrical potential with no net current and the potential with a current. The energy losses increase with increasing current density, which normally is proportional to H_2-production rate in the MEC. Previous MEC studies reported a wide range of applied voltage (0.3–1.3 V) when utilizing acetate as the electron donor [6–12,14]; from

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this, the energy loss can be computed as being in the range of 0.16–1.16 V, based on −0.14 V as the standard potential for the overall reaction $CH_3COO^- + 3H_2O = CO_2 + HCO_3^- + 4H_2$ at pH 7. The wide range of applied voltages and energy losses occurs because each study had different current density, biofilm-anode composition and thickness, donor concentration, pH, electrode material, electrode distance, and membrane type.

Traditional energy losses in chemical fuel cells and electrolysis cells are divided into ohmic, activation, and concentration losses [15,16]. Ohmic loss is caused by electrical resistance to current in conductors (electrodes + wire) and ion transfers in the electrolyte (membrane + medium). The activation energy loss is the energy required for overcoming energy barriers across the electrode/electrolyte interface to generate net current, and it is characterized by the Butler–Volmer equation [16]. Concentration energy loss is due to concentration gradients between the bulk liquid and the electrode surface, which become significant at high current density in chemical fuel cells [15,16]. Because current density in a microbial fuel cell (MFC) or an MEC is orders of magnitude smaller than in chemical fuel cells/electrolysis cell, it may seem reasonable to assume that concentration losses in the MFC/MEC are negligible. However, concentration losses can become significant in the MFC/MEC, because concentration gradients develop for substrate in any type of biofilm system [17–19], including the biofilm anode in the MFC/MEC. Only if the biofilm thickness is thin enough can we assume no concentration gradients between bulk and electrode surface, which is called a fully penetrated biofilm [17,19]; activation and concentration energy losses are sometimes considered together as electrode energy losses [6].

Even if the anode's biofilm has no gradients in the concentrations of the donor substrate and protons, a dual-chamber MFC/MEC using a membrane to separate the anode from the cathode can present a unique concentration loss due to $[H^+]$ or $[OH^-]$ accumulation in a chamber, since they are net produced at half reactions on the electrodes [20–22]. The high concentrations of other ions in the liquid supplied to an MEC/MFC (e.g., Na^+), compared to $[H^+]$ or $[OH^-]$, means that charge neutrality can be achieved with little transport of H^+ or OH^- ions through membrane [6,20], and a strong pH gradient can develop across the membrane, causing a substantial concentration energy loss. Rozendal et al. [6] reported that the pH difference between the two chambers increased the concentration energy loss up to 0.38 V in a dual-chamber MEC. They also showed that this was the largest part of the total energy loss.

Of many possibilities to decrease energy losses, removing the membrane, which creates a single-chamber MEC, can be very efficient; however, the lack of membrane accentuates the need for rapid and efficient hydrogen recovery to counteract hydrogen scavenging by methanogens [14]. One potential benefit of a single-chamber MEC is that the concentration energy loss due to H^+ or OH^- accumulation should be negligible, because H^+ produced in the anodic reaction is neutralized directly by OH^- produced in a cathodic reaction alternately or by reacting with electrons at the cathode to form H_2 molecules. The other potential benefit of a single-chamber MEC can be a substantial reduction in ohmic energy loss, since the resistance to ion flow through the membrane can be the main ohmic energy loss [6–10,14,23]. Thus, a single-chamber MEC can provide a high H_2-production rate with smaller applied voltage as a consequence of energy loss attenuation. Previous studies using a single-chamber MEC reported 3.12 m^3 H_2/m^3 d (292 A/m^3) at an applied voltage of 0.8 V [7], 1.7 m^3 H_2/m^3-d (188 A/m^3) at an applied voltage of 0.6 V [24], and 0.65 m^3 H_2/m^3 d (39 A/m^3) at an applied voltage of 0.6 V [10]. The authors claimed that these high H_2-production rates were achieved with relatively low applied voltage, compared to dual-chamber studies. However, a study with a dual-chamber MEC [2] also achieved H_2-production rates and applied voltages similar to the single-chamber MECs. Thus, the validity of energy loss mitigation in single-chamber MECs requires more rigorous study.

The energy-conversion efficiency (ECE) is an essential criterion for MEC sustainability, but the definition of ECE is not agreed upon. Previous works defined ECE in MECs as the heat of combustion of captured H_2 divided by the input electrical energy [2,7,10,24]. ECEs were well over 100% (194–351%) using this definition. They also have added the energy value of the input substrate to the denominator. With this definition, ECE declined to 58–86% [2,7,24].

Another approach to computing ECE is to compare the input and output energy in the same form. The most logical way to make the energy inputs and outputs consistent is to use electrical energy for both. Since generation of electrical energy from output H_2 incurs losses, the numerator in this approach is smaller than that used with the previous approaches that led to ECE values greater than 100%. For example, typical efficiency of energy transformation from H_2 heat energy to electricity is ~55% in hydrogen fuel cells [15], while the efficiency is ~33% if the H_2 is combusted to produce electricity [25].

We performed this work to provide rigorous characterizations of energy loss and ECE in an upflow single-chamber MEC. In our previous study [14], we found that the upflow single-chamber MEC with the cathode placed on top of the MEC improved the cathodic conversion efficiency (CCE) approximately two-fold over a conventional MEC having the cathode alongside the anodes: a CCE of 98 ± 2% at the same time as the Coulombic efficiency was 60 ± 1%, and negligible CH_4 was generated. Despite having a metal-catalyst-free cathode, the upflow MEC produced 0.57 ± 0.02 m^3 H_2/m^3 d at an applied voltage of ~1 V. For this study, we first estimate the concentration energy loss from the maximum pH gradient that developed in the liquid contents of the MEC chamber. Second, we experimentally measured the cathode/the anode energy losses, and ohmic energy loss. We then determine which energy losses were mainly responsible for the applied voltage and if the concentration energy loss and ohmic energy loss were mitigated in the single-chamber MEC. Third, we define and compute the ECE in the upflow MEC, and this allows us to identify the factors most critical for obtaining the maximum energy benefit from an MEC.

2. Background on electrode energy loss

Electrode energy loss is the potential difference between the theoretical chemical potential for a reduction half-reaction of

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