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Two stage bioethanol refining with multi litre stacked microbial fuel cell and microbial electrolysis cell

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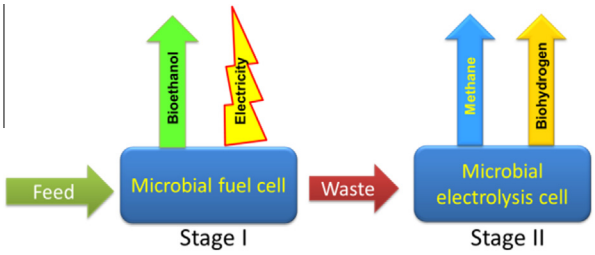
HIGHLIGHTS

- Ethanol was produced in a triple stack 10-L microbial fuel cell.
- With lower loads currents and potentials oscillated in a self-synchronized manner.
- Electrolysis conditions increased glucose conversion and ethanol productivity.
- Fermentation waste was methanized in a 33-L microbial electrolysis cell stack.

GRAPHICAL ABSTRACT



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ABSTRACT

Ethanol, electricity, hydrogen and methane were produced in a two stage bioethanol refinery setup based on a 10 L microbial fuel cell (MFC) and a 33 L microbial electrolysis cell (MEC). The MFC was a triple stack for ethanol and electricity co-generation. The stack configuration produced more ethanol with faster glucose consumption the higher the stack potential. Under electrolytic conditions ethanol productivity outperformed standard conditions and reached 96.3% of the theoretically best case. At lower external loads currents and working potentials oscillated in a self-synchronized manner over all three MFC units in the stack. In the second refining stage, fermentation waste was converted into methane, using the scale up MEC stack. The bioelectric methanisation reached 91% efficiency at room temperature with an applied voltage of 1.5 V using nickel cathodes. The two stage bioethanol refining process employing bioelectrochemical reactors produces more energy vectors than is possible with today's ethanol distilleries.

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1. Introduction

Bioethanol is an important bulk chemical and transportation fuel (Sanders et al., 2007) and its popularity is growing globally (Kim and Dale, 2004). In Brazil, one of the largest bioethanol producers in the world many cars have been running on pure ethanol for decades (Geller, 1985). In other countries such as: USA, Canada, Northern Europe and others, (European Biofuels Technology Platform) ethanol blended fuels have been introduced to reduce crude oil consumption (Niven, 2005). A total feed stock transformation into ethanol and other fuel vectors would improve the efficiency of today's bioethanol refineries. The emergence of bioelectric systems means renewable energy production from waste and dedicated crops can now be directed toward a total substrate conversion (Hernández-Fernández et al., 2015). However, this next

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generation of bioethanol refinery systems requires bioelectric reactors to be scale-up. The potential for upscaling was investigated by coproducing ethanol and electricity with a MFC and biodiesel from CO_2 exhaust that was passed into microalgae cultivation (Powell and Hill, 2009). The alternative to this method is to feed the CO_2 and fermentation waste into a bioelectric methanisator (MEC) to obtain high quantities of methane (Lovley and Nevin, 2013; Thygesen et al., 2010). This is of particular interest for an application as the methanisation of fermentation waste is as easy to realize as an ethanol production. Moreover, methane can be injected into existing gas pipelines for distribution.

A significant challenge of this approach is the scalability of microbial fuel cells (MFC) and microbial electrolysis cells (MEC) (Xiao et al., 2014; Janicek et al., 2014; Cusick et al., 2011). The reasons for this challenge are low microbial power and high internal resistances in scale-up bioelectric reactors. These limitations will be partly overcome with advanced electronics for power management, and electricity storage (Santoro et al., 2016).

Bioelectric ethanol refining appears to be therefore possible in a two stage bioethanol refining approach. In the first stage a Yeast-MFC produces ethanol and bioelectricity. In the second stage a microbial electrolysis cell converts fermentation waste and CO_2 into methane. This two stage biorefinery concept theoretically allows for a total feedstock conversion into energy vectors. The total methanisation process can be accomplished by injecting hydrogen from an external source or by an in-situ hydrogen evolution reaction in the MEC (Bassani et al., 2015). Recent research suggests an even more effective variant, called bioelectrosynthesis, where electrons pass directly into the methanogens to reduce CO_2 enzymatically (Cheng et al., 2009; Villano et al., 2010). This later mechanism also promises to reduce the danger of the explosion of hydrogen based methanisators, which is an important criteria for the safe use of such technologies in urban areas.

Many kinds of biomass are used for bioethanol production (Bonin and Lal, 2012). One is winery waste, which was used in this work. The liquid fraction is rich in sugar waste and often treated in municipal wastewater treatment plants (WWTP) and disturbs sewage works during grape harvest. Consequently, the WWTP used for this process are often larger than is needed for the rest of the year (Ioannou et al., 2015). As winery waste is particularly rich in glucose and fructose, a value added treatment strategy consisting of bioethanol production instead of complete digestion (Mateo and Maicas, 2015) is worth examining.

In this work ethanol and electricity were co-generated with a triple stack 10 L microbial fuel cell. In a second stage waste from ethanol fermentation was converted into methane using a 33 L stacked microbial electrolysis cell (Fig. 1). Smaller scale experiments were realized to examine glucose consumption, ethanol productivity and bioelectric effects more closely.

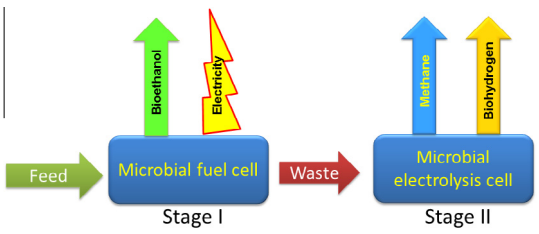

Fig. 1. Two stage bioethanol refining based on bioelectrical systems reactors with the aim of converting all feed stock into fuel vectors (electricity, EtOH, CH_4 and H_2). In the first stage ethanol and electricity are coproduced within a microbial fuel cell. In the second stage particulate and dissolved waste from ethanol fermentation, as well as evolving CO_2, are converted into methane and hydrogen using a microbial electrolysis cell.

2. Materials and methods

2.1. Material

Saccharomyces cerevisiae DSM 4266 was obtained from the German Collection of Microorganisms and Cell Cultures [Braunschweig, Germany]. Glucose, yeast extract, soy peptone and Methylene Blue were purchased from Fluka [Buchs Switzerland]. Sulphuric acid 98%, ethanol, n-butanol, and Kjeldahl tablets came from Merck [Darmstadt, Germany]. Sodium hydroxide was from Schweizerhall [Schweizerhall, Switzerland]. Two kinds of winery waste was prepared: (i) grape pomace (called bourbes in French), which is a Chasselas grape pulp (unfermented white wine); and (ii) lees (mostly dead yeast cells), which were obtained through the filtration of fermented must. Both wastes were prepared at the Oenology School of Changins [Nyon, Switzerland]. Electrode material came from reticulated vitreous carbon (RVC) tubes and plates, 100 ppi, which were prepared by the ERG aerospace corporation [Oakland CA, USA]. Nafion™-N117 proton exchange membranes (PEM), ordered from Ion Power [München, Germany], were reused and stored between experiments in phosphate buffer. Nickel steel plate cathodes came from BIBUS METALS AG, [Fehraltorf, Switzerland].

2.2. Construction of the MFC and MEC reactors

Three kinds of bioelectrochemical reactors were newly designed by Inventor CAD software (Autodesk GmbH, München, Germany) and constructed in-house with the poly-mechanics shop and a glass blower.

2.2.1. 10-L MFC triple stack

Three glass microbial fuel cells 3×3.2 L were based on commercial 1.5 L jacketed reactors with glass made T-tube extensions of similar size (Fig. S1, photo in the Supplemental materials). PEMs, Nafion-N117 of 38.5 cm^2, separated anode and cathode compartments. 3 Tube like RVC foam cylinders of 9.5 cm diameter, 38 cm length and an axial hole of 3.5×38 cm served as space filling anodes. Glass made L-shaped tubes served as cathode chambers containing 3 cathodes each made from platinum sputtered RVC slices ($0.5 \times 7 \times 7$ cm). The RVC cathodes were sputtered with elemental platinum in an SDS-60 Bal-Tec sputter chamber [Balzers, Liechtenstein] at 0.08 mbar/60 mA during 160 s. Air was purged through the aqueous catholyte.

2.2.2. 33-L MEC multi stack

This scale-up microbial electrolysis cell was constructed from PVC-U (polyvinylchloride-unplastified) ($50 \times 28 \times 30$ cm) with a total volume of 33.5 L, and that allowed for the addition of 11.5 L anolyte containing all electrodes (Fig. 2A, B). The 10 anodes (A) and 9 cathodes (C) were inserted in an alternating manner to establish a $[A(CA)_9]$ stack. In this case the anode cathode inter distance was 1.2 cm. The anodes were from RVC ($20 \times 1.8 \times 13$ cm) representing 2.9 m^2 of true surface per electrode (surface calculation based on published method by Friedrich et al., 2004). The cathodes were from nickel steel with $19 \times 13 \times 0.02$ cm^3 per electrode. Evolving biogas was guided to an external gas sampling port and passed further to a MilliGas counter (Ritter, Germany). In a second experiment 10 RVC anodes (A) were used and a single cathode cassette (C_{Naf}) that contained platinum sputtered RVC protected behind a Nafion-N117 membrane on both flat sides of the cassette (Fig. 2B) to form the following stack: $[A_5(C_{Naf})A_5]$.

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