



# Maximising biohydrogen yields via continuous electrochemical hydrogen removal and carbon dioxide scrubbing



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## HIGHLIGHTS

- Electrochemical hydrogen removal increased hydrogen yield during anaerobic digestion.
- Increased yields are the result of reducing product inhibition and homoacetogenesis.
- Hydrogen removal process is potentially more compatible with high solids substrate digestion.
- Purified hydrogen is produced as an end product.

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## ABSTRACT

The use of electrochemical hydrogen removal (EHR) together with carbon dioxide removal (CDR) was demonstrated for the first time using a continuous hydrogen producing fermenter. CDR alone was found to increase hydrogen yields from 0.07 mol H<sub>2</sub> mol<sub>hexose</sub><sup>-1</sup> to 0.72 mol H<sub>2</sub> mol<sub>hexose</sub><sup>-1</sup>. When CDR was combined with EHR, hydrogen yields increased further to 1.79 mol H<sub>2</sub> mol<sub>hexose</sub><sup>-1</sup>. The pattern of carbohydrate utilisation and volatile fatty acid (VFA) production are consistent with the hypothesis that increased yields are the result of relieving end product inhibition and inhibition of microbial hydrogen consumption. In situ removal of hydrogen and carbon dioxide as demonstrated here not only increase hydrogen yield but also produces a relatively pure product gas and unlike other approaches can be used to enhance conventional, mesophilic, CSTR type fermentation of low grade/high solids biomass.

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## 1. Introduction

The increasing scarcity, security issues, and environmental concerns surrounding the use of fossil fuels has led to interest in clean, sustainable sources of energy such as biomass. Biomass can be converted directly into hydrogen, a flexible, energy dense and non-polluting fuel using a form on anaerobic digestion. Anaerobic digestion is a well-established process which has been proven to be viable at full scale when used to produce methane as fuel. Fermentative biohydrogen as part of a two stage biohydrogen/biomethane process has been shown to significantly enhance methane production and overall energy yield (Massanet-Nicolau et al. 2013, 2015). However, producing hydrogen is an attractive alternative since it can be produced via a broader range of sustainable routes, has no combustion by-products other than water vapour, and has a higher value as a chemical commodity.

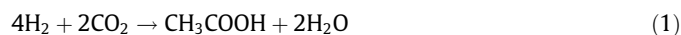
A considerable amount of research has been undertaken into producing hydrogen from biomass via anaerobic digestion and it has been demonstrated using a number of biomass types (Cheng et al., 2015; Han et al., 2015; Massanet-Nicolau et al., 2015; Massanet-Nicolau et al., 2013; Reilly et al., 2014). For this process to be viable at full scale however, yields of hydrogen must be maximised using processes applicable at full scale using a range of biomass types, including those containing high levels of solids, structurally complex carbohydrates and pre-existing microflora. In addition, the biohydrogen produced during digestion must also be separated from other gas produced such as carbon dioxide. The greater the purity of the hydrogen recovered the greater its utility and commercial value.

In this work the use of continuous in situ removal of hydrogen and carbon dioxide from the anaerobic digester is evaluated. Removal of these gases from the digester has the potential to increase hydrogen yields by lowering end product inhibition which limits microbial hydrogen production. Removing these gases in situ may also prevent hydrogen consumption via antagonistic

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microbial processes such as homoacetogenesis which produces acetic acid at the expense of hydrogen according to the following equation:



Removal of these gases during fermentation via sparging with external gases has been demonstrated previously with some success (Kim et al., 2006; Massanet-Nicolau et al., 2010; Mizuno et al., 2000), however this consumes a large amount of externally supplied gas and leads to a dilute end product, with hydrogen concentrations in the off gas frequently being lower than 5% (Kim et al., 2006). In order for a hydrogen digester system to be practical and economical at larger scales the gas used to remove hydrogen and carbon dioxide must be recycled and the hydrogen continually extracted and concentrated.

In this study a novel electrochemical method of hydrogen separation and purification is evaluated. The process, developed by Hydrogen Efficiency Technologies (HyET) in the Netherlands, utilises a proton exchange membrane together with a small electrical current, to separate hydrogen from a mixed gas stream (Fig. 1). Compared with other methods of purifying hydrogen from gas mixtures, the process is energy efficient with typical energy demands of 4 kWh/kg H<sub>2</sub>, (Hydrogen has a lower heating value of 120 MJ/kg) and product gas purity in excess of 99% (Bouwman, 2015; Casati et al., 2008).

The objectives of this study were as follows; firstly to demonstrate the feasibility of integrating an electrochemical purification system process, together with continuous carbon dioxide removal (via chemical adsorption) into a continuously operated anaerobic digester producing hydrogen. Secondly, to examine the effect of such an integration on hydrogen yields for the first time. In addition, by examining the patterns of substrate utilisation and volatile fatty acid production the mechanisms for any changes in hydrogen output were evaluated.

## 2. Materials and methods

### 2.1. Digestion experiments

A single, multiphase digestion experiment spanning several hydraulic retention times (HRTs) was used in this study. In phase one of the study the digester was operated for 3 HRTs with no in situ removal of either carbon dioxide or hydrogen. In phase two of the study carbon dioxide removal was used during digestion

and in phase 3 both carbon dioxide removal and hydrogen removal were employed.

### 2.2. Hydrogen producing anaerobic digester

A continuously fed digester with a working volume of 4 L and a headspace of 0.5 L was used in this study (Fig. 2). The digester incorporates a gas mixing loop which transports gas from the headspace of the digester and bubbles it through the digester contents at its base. Incorporated into this loop are the apparatus for in situ removal of hydrogen and carbon dioxide, together with sensors for continuous measurement of gas composition (H<sub>2</sub>, CO<sub>2</sub> and CH<sub>4</sub>). Positive displacement flow metres are used at the exit to the digester and to the hydrogen separator, and a demand valve is used to add small amounts of nitrogen (<0.16 L h<sup>-1</sup>) in order to prevent negative pressure build-up as a result of sampling and feeding the digester. The digester also allows for the continuous monitoring and control of pH and temperature. Data from all sensors are recorded every second and logged using a data acquisition hardware and custom control and logging software.

### 2.3. Hydrogen and carbon dioxide removal

Hydrogen was removed using an electrochemical purification system based on a proton exchange membrane as shown in Fig. 1. The mixed gas from the digester flows through the anode chamber and with the aid of a supplied electrical voltage hydrogen is oxidised into protons which migrate through the membrane and recombine with supplied electrons at the cathode into concentrated molecular hydrogen. The process is similar in nature to a hydrogen fuel cell but operates in the absence of oxygen so hydrogen is formed at the cathode rather than water. Carbon dioxide was removed by bubbling the digester gas through 3 M sodium hydroxide, this was replenished when its pH dropped below 9.5–10 as shown by an indicator solution (thymolphthalein 0.2% w/v).

### 2.4. Digester operation and start-up

The hydrogen producing digester was started by filling it with 5% heat treated inoculum and 95% feedstock by volume. In order to build up levels of hydrogen producing microorganisms, the digester was initially operated in batch mode (with no additional feeding) until production of hydrogen occurred (6–12 h). Continuous feeding then commenced and the digester was operated for a

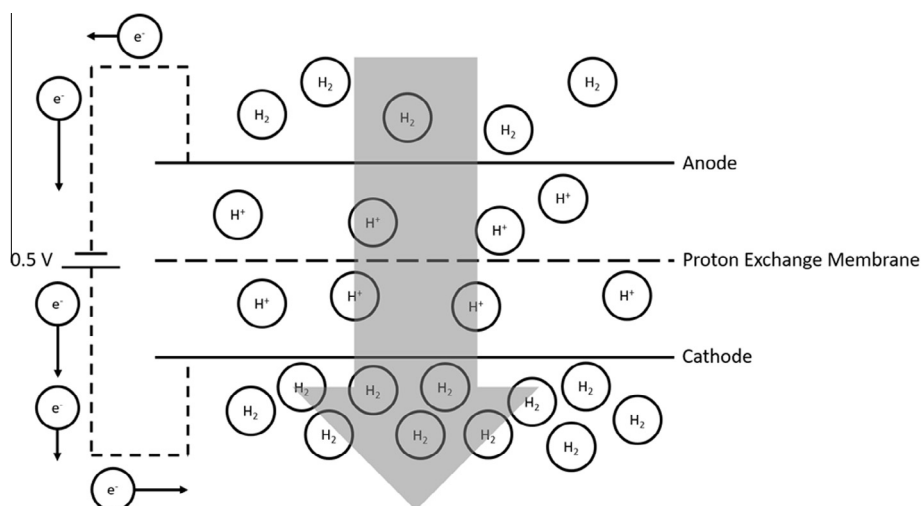


Fig. 1. Hydrogen separation using an applied current and a proton exchange membrane.

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