INTERNATIONAL JOURNAL OF HYDROGEN ENERGY XXX (2014) 1–8

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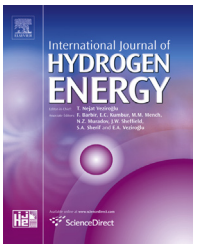

Simultaneous biohydrogen production and purification in a double-membrane bioreactor system

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ARTICLE INFO

Article history:
Received 17 October 2014
Received in revised form
20 November 2014
Accepted 1 December 2014
Available online xxx

Keywords:
Biohydrogen
Membrane bioreactor
Gas separation
Integrated system
PDMS

ABSTRACT

In this work the establishment of a double-membrane bioreactor was aimed. Initially, a continuous hydrogen fermenter was coupled with a commercial Kubota® microfiltration membrane module and the production performance of the cell-retentive design was evaluated under various hydraulic retention times. As a result, it has been observed that altering HRT influenced the rejection feature of the microfiltration module while had an inverse effect on hydrogen productivity and yield, since shortened HRTs were accompanied by gradually decreasing H_2 yields (HY) and progressively increasing volumetric H_2 production rates (HPR). The highest HY and HPR were achieved as 1.13 mol H_2/mol glucose and 0.24 mol H_2/L-d, respectively. Furthermore, a Permselect® (PDMS) gas separation membrane was installed to the anaerobic membrane bioreactor and its ability to separate hydrogen from the raw fermentation gaseous mixture was assessed. The highest purity hydrogen obtained in one-step purification by the PDMS module was 67.3 vol.%, which exceeds 30% enrichment efficiency considering 51.3 vol.% H_2 in the feed gas. Hence, it could be concluded that the poly(dimethyl siloxane) membrane has potential to attractively concentrate biohydrogen from fermenter off-gas and may be used for in-situ product recovery.

Introduction

The design and initiation of bioreactors is a crucial element of continuous dark fermentative hydrogen production [1,2]. Recently, fermenters attached with membranes have been demonstrated as highly attractive reactor configurations to achieve intensified microbiological hydrogen generation [3]. Membranes coupled to hydrogen forming reactors can potentially serve two-fold but equally important purposes. On one hand, pressure driven membrane processes such as microfiltration can be employed to enrich hydrogen

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http://dx.doi.org/10.1016/j.ijhydene.2014.12.002

generating whole cell biocatalysts and thus accomplish intensified H_2 formation as compared to traditional free-cell applications, e.g. CSTRs. On the other hand, membranes i.e. gas separation modules may provide a sufficient way to purify hydrogen in order to obtain concentrated biohydrogen applicable for fuel cells [4].

Conventional anaerobic hydrogen producing bioreactors using porous liquid filtration membranes provide the chance to maintain separate hydraulic- and solid retention times. This is an important trait since it has turned out that decoupled and altered sludge retention times (SRT) could be responsible for remarkable shifts in the hydrogen evolving microbial consortia and the related gas production values [5]. Furthermore, Lee et al. [6] have reported that attaching microfiltration membrane module to continuous hydrogen fermenter could improve the efficacy of the process. The results indicated notable enhancement in the volumetric production rates and hydrogen yields, which could be attributed to the enriched biomass of active whole cell biocatalysts. It has also appeared that peak values of H_2 productivity and yield took place under different reactor operation. In another relevant study [7] CSTR and MBR arrangements were compared for biotechnological hydrogen production, when operated with various organic loading intensities. The outcomes were somewhat beyond the preliminary expectations since the integrated membrane bioreactor operation did not provide advantages over the conventional suspended free-cell reactor in terms of hydrogen yield. Nevertheless, the performance of the anaerobic membrane bioreactor could apparently exceed that of its traditional counterpart in the view of H_2 evolution rate by approximately 50% under certain experimental sets. Additionally, Lee et al. [8] assessed the feasibility of MBR and CSTR configurations for biological hydrogen production. The final conclusion of the long-term, steady-state measurements was that the achievable H_2 yields in both reactor set-ups were quite comparable, while on the other hand, MBR took the advantage from the point of view of volumetric hydrogen generation rate, which was approximately 2.6 times higher than in the CSTR. The same group of scientists expanded their research on AnHPMBR (anaerobic hydrogen producing membrane bioreactor) under various solid retention times [9]. It was obvious from the results that the extremely high solid retention time as long as 90 days remarkably decreased both hydrogen productivity and yield. This behavior was associated with the increasing amount of extracellular polymeric substances (EPS) under longer SRT. It was elucidated that EPS, as secondary products of fermentation process – depending on their concentrations – are potential inhibitors of microbiological hydrogen formation. Recently, Kim et al. [10] comparatively assessed the H_2 production performances of anaerobic membrane bioreactor and completely stirred tank reactor. The critical evaluation of the tentative results obtained demonstrated that AnMBR design was far more viable to get better H_2 productivities and yields. In the optimized conditions of the MBR, the increase of H_2 yield was about 50% whilst hydrogen production rate has been more than doubled in comparison to the conventional continuous reactor.

Besides the traditional AnMBR arrangement relying on porous water filtration membrane modules, MBRs integrated with gas separation seem also attractive designs to enhance the feasibility of dark fermentative hydrogen production [4,11]. The concept of such systems is the in-situ recovery and purification of biologically formed hydrogen from the fermenter off-gas – containing notable amount of CO_2 beyond H_2 – in a way that it may be directly utilized in fuel cells. Moreover, the instant and continuous extraction of hydrogen helps to keep lowered hydrogen partial pressure in the reactor which has been proven advantageous for higher hydrogen production yields [11]. The separation of hydrogen from complex biological gas mixtures is a challenging task because several compounds, such as carbon dioxide, hydrogen sulfide, water vapor, etc. pose a threat to achieve the required enrichment efficiency [4]. Recently, our group has tested a range of gas purification membranes made of different materials such as polyimide, SAPO 34 and silicone for fermentative hydrogen concentration, using binary (H_2/CO_2) mixtures [11,12]. Regardless of the membrane module, it could be concluded that the composition of the feed gas is a key factor to be considered. However, in those previous experiments, the membranes were applied separately from the hydrogen fermenter and therefore, not much is known yet about their ability to deal with complex, raw fermentation gaseous mixtures containing the valuable component, biohydrogen.

In this investigation, a double-membrane bioreactor system was aimed to establish. Firstly, the performance of anaerobic hydrogen producing membrane bioreactor employing microfiltration liquid filtration membrane module was focused under different hydraulic retention times. Afterwards, this conventional AnHPMBR was installed with a PDMS membrane and the behavior of the module was evaluated with raw headspace gas mixture at certain operating circumstances. The novelty of the work is that this is the first time report when a single device – a double-membrane bioreactor – applying two kinds of membranes concerns both the upstream and downstream aspects of microbiological hydrogen generation and biohydrogen is directly enriched from untreated reactor off-gas during continuous operation.

Materials and methods

Hydrogen production measurements in AnMBR

A double-wall, laboratory scale device was used to construct the hydrogen producing anaerobic membrane bioreactor. The vessel of the reactor was made of borosilicate glass and had a nominal total volume of 3.5 L. To start-up the continuous reactor, 1.5 L of anaerobic digester sludge – receiving preliminary heat-treatment in water bath under the circumstances (75 °C, 30 min) found favorable in our previous paper [13] – was filled in. Afterwards, 0.5 L of feed solution comprising glucose and yeast extract (dissolved in dechlorinated tap water) was added to the pretreated sludge. The concentration of glucose and yeast extract in the feed solution was 40 g L^{-1} and 10 g L^{-1}, respectively. As the next step, pH of the broth was adjusted and automatically maintained at 5.5 ± 0.2 by means of 5 M sulfuric acid and NaOH solutions. After setting pH, the bioreactor was closed and purged for 20 min with 99.9% N_2 at a flow rate of approximately 5 L min^{-1}

Please cite this article in press as: Bakonyi P, et al., Simultaneous biohydrogen production and purification in a double-membrane bioreactor system, International Journal of Hydrogen Energy (2014), http://dx.doi.org/10.1016/j.ijhydene.2014.12.002