



Biohydrogen production from microalgal biomass: Energy requirement, CO₂ emissions and scale-up scenarios



Ana F. Ferreira^{a,*}, Joana Ortigueira^b, Luís Alves^b, Luísa Gouveia^b, Patrícia Moura^{b,c}, Carla Silva^a

^aIDMEC, Instituto Superior Técnico, Technical University of Lisbon, Av. Rovisco Pais 1, 1049-001 Lisbon, Portugal

^bLNEG – National Laboratory of Energy and Geology, Bioenergy Unit, Estrada do Paço do Lumiar 22, 1649-038 Lisbon, Portugal

^cInstituto Superior de Ciências da Saúde Egas Moniz, Quinta da Granja, Monte de Caparica, 2829-511 Caparica, Portugal

HIGHLIGHTS

- The H₂ yield was 7.3 g_{H2}/kg_{biomass} by *C. butyricum* from *S. obliquus* dried biomass.
- The H₂ production consumed 71–100 MJ/MJ_{H2} of energy and emitted 5–6 kg CO₂/MJ_{H2}.
- In a possible scale-up, the energy consumption may attain 6–8 MJ/MJ_{H2}.
- Scale-up is advantageous in terms of CO₂ emissions, reaching (–716) to (–613) g/MJ_{H2}.
- The best scenario would produce H₂ to supply 5.5% of a Lisbon urban taxi fleet.

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ABSTRACT

This paper presents a life cycle inventory of biohydrogen production by *Clostridium butyricum* through the fermentation of the whole *Scenedesmus obliquus* biomass. The main purpose of this work was to determine the energy consumption and CO₂ emissions during the production of hydrogen. This was accomplished through the fermentation of the microalgal biomass cultivated in an outdoor raceway pond and the preparation of the inoculum and culture media. The scale-up scenarios are discussed aiming for a potential application to a fuel cell hybrid taxi fleet.

The H₂ yield obtained was 7.3 g H₂/kg of *S. obliquus* dried biomass. The results show that the production of biohydrogen required 71–100 MJ/MJ_{H2} and emitted about 5–6 kg CO₂/MJ_{H2}. Other studies and production technologies were taken into account to discuss an eventual process scale-up. Increased production rates of microalgal biomass and biohydrogen are necessary for bioH₂ to become competitive with conventional production pathways.

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1. Introduction

The two main drivers of the production of biofuels are the depletion of fossil fuels, and environmental impacts of increasing fossil fuel consumption. Biofuels that can be readily produced without a large increase neither in arable land nor reduction in tropical rainforest areas are very important issues in the future. Microalgae may offer this opportunity. Its use as a feedstock for biofuels has led to much excitement and initiative within the energy industry (Darzins et al., 2010). Some microalgae species are extremely rich in lipids and sugars, making them suitable for biodiesel, bioethanol and bioH₂ production, respectively. Besides the fact that microalgae cultivation may avoid the need of arable land, there is also the possibility of using brackish, saline and wastewater for their growth. Additionally microalgal biomass could be harvested on a daily basis (John et al., 2011).

Microalgae are therefore a good source for liquid (e.g., biodiesel and bioethanol) and gaseous (e.g., biohydrogen and biogas) biofuels production. *Scenedesmus obliquus*, in particular, is a microalga with good biomass productivity rates, around 0.09 gL⁻¹ day⁻¹ (Gouveia and Oliveira, 2009), which has been proven to be very versatile as a raw material for biofuel production. This microalga contains approximately 12–14% (w/w) of oil and 10–17% (w/w) of sugars (Demirbas, 2009) and is therefore a good source for biodiesel (Gouveia and Oliveira, 2009; Mandal and Malick, 2009; Silva et al., 2009), bioethanol (Miranda et al., 2012ab) and bioH₂ production (Demirbas, 2009; Ferreira et al., 2013a). In a study conducted by Miranda et al. (2012b), *S. obliquus* biomass accumulated starch in a concentration of 30% (w/dw) starch (glucose equivalents).

In all bioconversion processes, the adequacy of the fermenting microorganisms to the substrate feedstock and the values of product yield are of primordial importance (Kotay and Das, 2008). Species of *Clostridium* are frequently found in hydrogen-producing consortia and are also very effective in producing H₂ from organic substrates, especially carbohydrates (Chong et al., 2009). The yields

* Corresponding author. Tel.: +351 218419554.

E-mail address: filipa.ferreira@ist.utl.pt (A.F. Ferreira).

reported for bioH₂ production by *Clostridium* sp. may range from 0.73 to 3.1 mol H₂/mol sugar (Chong et al., 2009; Junghare et al., 2012).

The production of liquid and gaseous biofuels consumes a lot of energy and emits CO₂ into the atmosphere, so it is important to consider both aspects in the evaluation of process effectiveness. LCA is a methodology used to quantify the energy consumption and CO₂ emissions of liquid and gaseous biofuel production. LCA is a tool that analyses a product during its lifetime from its production, to its utilisation and end-of-life, including its recycling process. It can also be used to compare different energy systems including vehicle technologies and production systems (e.g. biofuel production) (Ferreira et al., 2011). The findings of LCA clearly show the energy magnitude of each life cycle phase, providing the feedback required to focus future critical process stages, to make manufacturing processes more energy efficient and economically advantageous and profitable, at laboratorial scale and, in a near future, at industrial scale.

To date, there are no development studies about bioH₂ production scale-up from microalgal biomass. Then, the up-scaling of bioH₂ production should be addressed; and it will most likely depend on several variables such as microalgae culture conditions (e.g., light-source and regime, nutrient availability, temperature, agitation, pH), growth kinetics, microalgae downstream processing, operational parameters of the fermentation and electricity mix. The electricity mix could influence the scale-up process in terms of energy consumption and CO₂ emissions both in pilot and/or industrial plants. The use of more renewable energy makes the production energetically more efficient and sustainable. The studies presented in the literature on up-scaling processes of bioH₂ production using laboratorial data are limited. A pilot-scale study using a continuous anaerobic fermentation reactor using molasses as feedstock was reported in Show et al. (2012). In a development of the process to a larger scale application using Computational Fluid Dynamics (CFD) simulation, hydrogen production rates under sunlight illumination were estimated by Rosner and Wagner (2012). As a consequence, a more efficient scaled-up photobioreactor of 100 L was designed. This study compared different hydrogen production technologies and included the respective LCA analysis. The authors concluded that hydrogen production rates should be increased by a factor of 100, and that processes optimisation is still needed to get bioH₂ into competition with other hydrogen technologies production (Rosner and Wagner, 2012).

Recent studies cover bioH₂ production by cyanobacteria and microalgal biomass, along with energy and CO₂ emission analysis. Ferreira et al. (2012) focused on the photoautotrophic production of hydrogen by *Anabaena* sp. cultured in closed photobioreactors with artificial light, and subsequent dark fermentation of the spent biomass. In Ferreira et al. (2013a), hydrogen production by *Clostridium butyricum* from the sugars contained in *S. obliquus* hydrolysate was considered. The process was optimised by merging several process steps, improving the production efficiency and the values of life cycle inventory. Ferreira et al. (2013b) reported the hydrogen production by *Enterobacter aerogenes* from the *Nannochloropsis* sp. microalgal biomass leftover, after the oil and pigment extractions. However, none of these studies considered the possible scale-up of the biohydrogen production.

This paper presents the evaluation of the energy consumptions and CO₂ emissions during experimental bioH₂ production by *C. butyricum* through the fermentation of the whole *S. obliquus* biomass cultivated in an outdoor raceway pond. The first attempt of process scale-up was performed, by using scenarios development. The authors analysed two scenarios by identifying the process steps that produce higher emissions and have higher energy requirements: pilot, the optimised scenario and industrial, the best scenario.

2. Methods

2.1. Life cycle inventory of bioH₂ production from microalgal biomass fermentation

In this work a cradle-to-gate approach, which is an assessment of a partial product life cycle from resource extraction (cradle) to the factory gate, was regarded. All emissions are reported on a volume or mass basis (e.g., kg of CO₂).

The Life cycle inventory (LCI) is part of the Life cycle Assessment methodology (LCA) and its analysis allows a possible optimisation of the production system in order to decrease the energy requirement and CO₂ emissions during all stages. It is possible to identify the bottlenecks and suggest improvements on both parameters chain.

2.1.1. Characterisation of the boundaries, main processes, inputs and outputs

The evaluation of input/output analysis considered each step of the hydrogen production, starting from the microalga growth until hydrogen production through the dark fermentation process. The storage and distribution were excluded from the system boundary.

Fig. 1 shows the system boundary considered in hydrogen production and the corresponding inputs.

The main stages considered in the system boundary were the microalgal biomass production in an outdoor raceway pond, the preparation of the fermentative medium, which includes the preparation of the basal medium (BM1) (Moura et al., 2007), additional solutions, the preparation of the pre-inoculum, and the dark fermentation process. The sum of the impacts was allocated to the biohydrogen production.

The power generation mix of the country, where the biofuel is produced, was taken into account. Around 99.9% of the energy requirements of whole processes were fulfilled by electricity, in this case, from Portuguese electricity mix. However, to produce the electricity, the conversion of raw materials into energy is subjected to the conversion processes efficiency. Therefore, the electricity production is considered as a unit process of bioH₂ production and thus being part of the system boundary. In the present case, the electricity consumed in all hydrogen production stage was generated from the Portuguese electric mix which is composed of 56% of non-renewable and 44% of renewable energy, with 8% of energy losses in distribution (REN, 2012; Concawe, 2011). The electricity and CO₂ conversion factor to estimate energy consumption in each process is described by Ferreira et al., 2013a. The resulting energy consumption and CO₂ emissions per 1 MJ of electricity produced are 1.17 (0.94–1.31) MJ and 76.32 (68.18–81.70) g, respectively. The uncertainty of the Portuguese electricity generation mix considered weighted minimum and maximum deviation values for each energy source, based on the Concawe study (Concawe, 2011). Despite considering the National mix, marginal mix approaches are discussed and included in the result analysis.

The SimaPro 7.1 software (Pre-Consultants, 2007) was the source database used to estimate the energy consumption and CO₂ emissions for the input nutrients, nitrogen gas and deionized water in a quantity, basis, i.e., MJ/kg and g/kg. These were multiplied by experimental quantities to obtain none mass specific values. The electricity needs, estimated by using this database, were adapted for the average Portuguese electricity generation mix. This was done by multiplying the energy required in the processes by the Portuguese electricity energy factor of 1.17 MJ/MJ_{el} (and respective uncertainty). The remaining energy inputs, of the equipment used, were obtained from the rated power of the devices and working hours (Ferreira et al., 2012, 2013b). Namely, it corresponds to

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