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Review

Microbial electrohydrogenesis linked to dark fermentation as integrated application for enhanced biohydrogen production: A review on process characteristics, experiences and lessons

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ABSTRACT

Microbial electrohydrogenesis cells (MECs) are devices that have attracted significant attention from the scientific community to generate hydrogen gas electrochemically with the aid of exoelectrogen microorganisms. It has been demonstrated that MECs are capable to deal with the residual organic materials present in effluents generated along with dark fermentative hydrogen bioproduction (DF). Consequently, MECs stand as attractive post-treatment units to enhance the global H_2 yield as a part of a two-stage, integrated application (DF-MEC). In this review article, it is aimed (i) to assess results communicated in the relevant literature on cascade DF-MEC systems, (ii) describe the characteristics of each steps involved and (iii) discuss the experiences as well as the lessons in order to facilitate knowledge transfer and help the interested readers with the construction of more efficient coupled set-ups, leading eventually to the improvement of overall biohydrogen evolution performances.

1. Introduction

So far, practical scale biohydrogen generation has been realized via the so-called dark fermentation (DF) pathway. In comparison to other production methods i.e. photo-biological ones, DF demonstrates (i) fast gas formation rates, (ii) can be characterized with relatively facile reactor design requirements and (iii) gives possibility to valorize a broad range of organic matter, for instance biomass-derived solid resources and wastewaters (Bakonyi et al., 2017; Gómez et al., 2011; Kumar et al., 2015).

Though DF comes with a number of advantages as shortly highlighted above, certain existing issues related to this technology should be tackled. Among them, one of the most often referenced is the low achievable H_2 yield (expressing how much gas is generated per a certain amount of substrate added/utilized), which is attributed to the release of (dead-end) fermentation side-products, referred as soluble metabolites such as volatile fatty acids (VFAs) (Venkata Mohan et al., 2016). Ascribed to the undesired phenomena (where the chemical energy stored in the raw materials is not sufficiently converted to H_2), many ongoing research activities have aimed at finding solutions and earn improved system performance (Tapia-Venegas et al., 2015). As a result, various degrees of success could be attained by microbiology-assisted as well as process engineering approaches (Kumar et al., 2016; Sivagurunathan et al., 2016). Apart from these techniques that are normally employed to further develop single-stage DF, research directions have been devoted to the implementation of downstream-assisted, sequential applications. These are meant to treat and exploit the VFA-rich DF effluent and simultaneously, ensure the production of additional gaseous energy carriers for the realization of higher energy efficiency (Ghimire et al., 2015; Guwy et al., 2011, Lee et al., 2010).

Among these options, the most traditional one is anaerobic digestion where organic components in the DF spent liquor are further decomposed to get biogas (Nathao et al., 2013), which can be upgraded to biomethane (Ryckebosch et al., 2011). Furthermore, cascade processes involving photo-fermentative hydrogen gas evolution as an auxiliary stage were also suggested (Ren et al., 2011). In the recent years, other alternatives to complement one-step DF have attracted remarkable attention in the scientific community, especially those that rely on Bioelectrochemical Systems (BESs) (Wang and Ren, 2013). In this developing field, two particular systems, called Microbial Fuel Cells (MFC, Fig. 1A) and Microbial Electrohydrogenesis Cells (MEC, Fig. 1B) seem to be potentially applicable to manage (degrade and valorize) the residual DF liquid (Boboescu et al., 2016; Kadier et al., 2016; Lu and Ren, 2016; Venkata Mohan et al., 2014).

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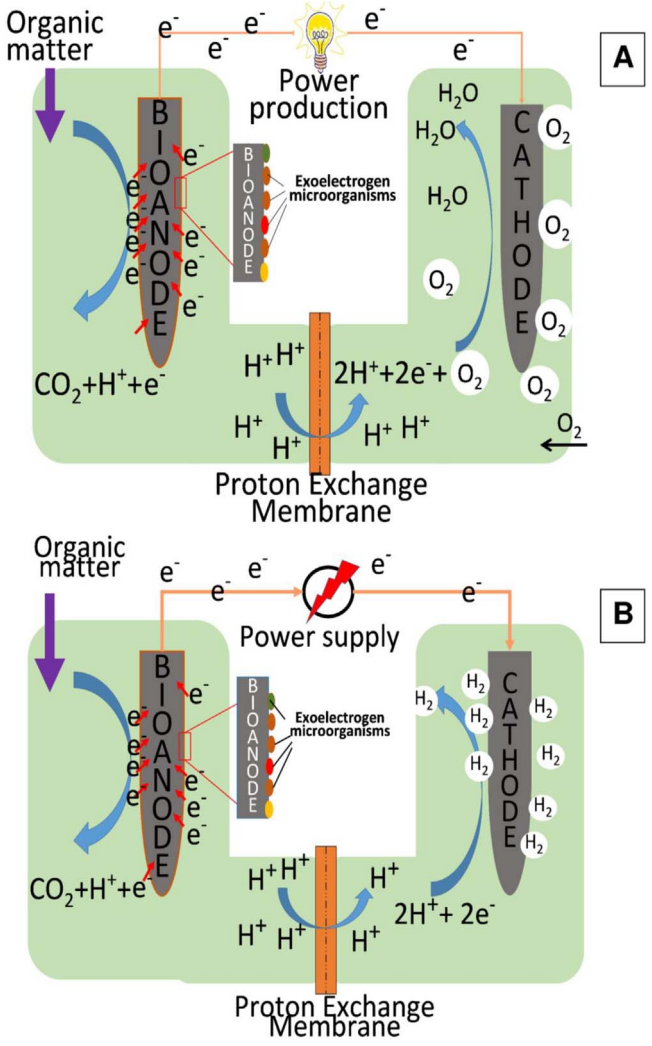

Fig. 1. Schematic illustration of microbial fuel cell (A) and microbial electrohydrogenesis cell (B).

The traditional BES architecture can be described by a two-chamber design, where the anode and the cathode compartments are spatially separated (in most cases by a thin membrane) and at the same time, the electrodes are connected through external wiring (Koók et al., 2016, 2017a,b; Kumar et al., 2017a). In fact, both in MFCs and MECs, electrochemically-active (so-called exoelectrogenic) microbes are found, favorably in a biofilm that is formed on the anode surface. They have the ability to assist the transformation of organic materials, for instance wastewaters including the effluent of the hydrogen producing bioreactors under anoxic conditions (Escapa et al., 2016; Oh and Logan, 2005; Pandey et al., 2016; Yang et al., 2015). In principle, one major difference between the two processes is in the products obtained as a result of the biological conversion: In MFCs, useful bioelectricity is directly obtained, whilst in MECs, H_2 gas is generated at the cathode (Chookaew et al., 2014; Du et al., 2007; Zhou et al., 2013).

Conceptually, MECs can be recognized as modified MFCs that require certain energy investment for the hydrogen formation to take place at the anaerobic cathode side (Logan et al., 2008). Subsequently, an external voltage is normally applied in order to drive this non-spontaneous reaction and induce the reduction of protons at the cathode surface (Pant et al., 2012). Practically speaking, at standard temperature and pressure conditions with acetate substrate (most generally used simple compound to test bioelectrochemical applications), the extra potential demand can be above 0.25 V due to internal system losses (Zhen et al., 2017). Nevertheless, this energy demand is considerably lower than in case of water electrolysis, where at least 1.2 V (Geelhoed et al., 2010) or even 1.8–2.0 V might be needed (Logan, 2008). The advantage of MECs built with two-compartments could be that the hydrogen evolved is less contaminated and hence, obtained with higher purity (Kumar et al., 2017). Consequently, in these cases, H_2 has to undergo only less-sophisticated separation, which claims definitive benefits from economical aspects. Additionally, it has turned out that the introduction of MEC technology to polish DF effluent could lead to impressive, 9.6 mol H_2/mol glucose hydrogen yield (Lalaurette et al., 2009), representing 80% of the theoretical maximum from this substance. Hence, dark fermentative biohydrogen production integrated with MEC has a lot of perspectives and not surprisingly, stands as a hot field for research.

In this paper, it is aimed to overview the existing relevant literature, where the concept of biohydrogen production in coupled DF-MEC systems is demonstrated. The scope will be on the evaluation of experimental results, experiences gained and lessons learned in order to provide an up to date insight to the interested readership and advance the international knowledge transfer of this area.

2. DF-MEC coupled systems

2.1. Description and characterization of DF as first stage

The major traits of DF (as 1st step of the cascade DF-MEC process) are summarized in Table 1, where it can be seen that it has been routinely investigated in batch studies. Nonetheless, bioreactors running as continuous systems were employed in some cases, as well. In the former operating mode, useful fundamental studies can be carried out, especially to (i) assess the hydrogen production potential of certain raw material(s) and substrates (Logan et al., 2002), (ii) describe the fermentation via revealing its kinetics for instance by relying on the Gompertz-model (Chen et al., 2006; Gadhamshetty et al., 2010), correlate the H_2 generation capacity with the soluble metabolic substance (SMS) pattern (Infantes et al., 2011) and qualities of the microbial population (O-Thong et al., 2009; Ren et al., 2008) and (iv) optimize the crucial operating parameters with respect to temperature, pH, etc. (Bakonyi et al., 2011, 2014a; Wang and Wan, 2009a). Afterwards, however, to investigate the DF on practical grounds and achieve mass production on bigger scale, continuous reactors should be started-up and established in steady-state to get feedback about scaled-up behavior and feasibility/robustness in a real i.e. non-sterile environment (Bakonyi et al., 2014b; Chen et al., 2008).

In concern with the starting materials employed in 1st stage DF, microbial conversions were performed to generate H_2 with various organic matter sources (Table 1) having significantly different characteristics from point of views such as complexity, origin, pretreatment applied or not, presence of nutrients and inhibitors, concentration, etc. These latter facts reasonably explain the varying attainable DF efficiencies (Guo et al., 2010; Shobana et al., 2017; Wong et al., 2014), first and foremost in relation with the widely-accepted, primary indicators namely the (volumetric) hydrogen production rate (HPR) (from a couple of 10 mL to 1–2 L H_2/L-d, Table 1) and hydrogen yield (HY) (Hallenbeck and Ghosh, 2009), though a direct and true comparison between some systems in terms of HY can cause difficult times due to (i) values reported with different units, i.e. L H_2/g COD vs. L H_2/g feedstock or substrate and (ii) lack of data to convert one to another (Table 1).

It is noteworthy that not only the feedstock properties, but the seed source, the structure and dynamics of the microbial community play a notable role on the success of DF via influencing SMS distribution and concomitantly, the quantity of H_2 evolved under actual environmental circumstances and bioreactor features (Bundhoo and Mohee, 2016; Kumar et al., 2012; Sivagurunathan et al., 2014; Wang and Wan, 2009b).

As for the most representative SMSs generated in course of DF, one